Experimental and Theoretical Modeling of DNAPL Transport in Vertical Fractured Media

by

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ABSTRACT

In recent years, groundwater contamination by dense, non-aqueous phase liquids (DNAPLs) such as chlorinated solvents and polychlorinated biphenyls (PCBs) has become an important environmental concern in many industrialized areas. Accidental spills, poor storage facilities and inadequate disposal practices are factors contributing to the release of these chemicals into the subsurface environment. The detection and presence of DNAPLs at hazardous waste sites is likely to be a significant limiting factor in the site remediation process. This is especially true for the remediation of complex subsurface formations, such as fractured bedrock.

This work has developed a new model describing DNAPL infiltration into a water-saturated, vertical fracture idealized as a circular section capillary tube. A series of laboratory and geotechnical centrifuge experiments of DNAPL infiltration into vertical capillary tubes was performed to demonstrate the validity of the model, and examine the role played by capillary, gravity, viscous, and inertia forces during the infiltration process. The new model can be used to better understand the processes likely to influence DNAPL transport and remediation in real fracture systems.

Laboratory infiltration experiments showed that the common assumption of perfect wetting at the DNAPL/water interface is incorrect. Furthermore, it was shown that contact angle hysteresis and interface meniscus pinning influence the DNAPL pool height required to infiltrate a capillary tube and also contribute to its variability. The laboratory experiments also showed that during the infiltration process, the contact angle reduces with velocity leading to an increase in the capillary forces with velocity. A model of the infiltration kinetics incorporating a velocity-dependent dynamic contact angle and pinning effects was successful at predicting the experimental results.

Centrifuge infiltration experiments demonstrated the conditions required for the validity of the scaling laws used for centrifuge modeling. The experimental results suggest that the geotechnical centrifuge can be used to examine general DNAPL behavior in simple fracture systems. Nonetheless, the variability of the DNAPL infiltration pool height, the effects of inertia, and the dependence of capillary forces upon velocity are factors that also influence the centrifuge modeling process, and thus need to be accounted for in the interpretation of the centrifuge test data.

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To B... for putting up with me

And, of course, to the Geek Pack...
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<th>Description</th>
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<tbody>
<tr>
<td>1,1-DCA</td>
<td>1,1-Dichloroethane</td>
</tr>
<tr>
<td>1,1-DCE</td>
<td>1,1-Dichloroethene</td>
</tr>
<tr>
<td>1,2-DCA</td>
<td>1,2-Dichloroethane</td>
</tr>
<tr>
<td>1,2-DCE</td>
<td>Trans-1,2-Dichloroethene</td>
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<tr>
<td>2-CT</td>
<td>2-Chlorotoluene</td>
</tr>
<tr>
<td>4-CT</td>
<td>4-Chlorotoluene</td>
</tr>
<tr>
<td>ADC(s)</td>
<td>Analog-to-Digital (Conversion) Card(s)</td>
</tr>
<tr>
<td>CCD</td>
<td>Charge-Coupled Device (Camera)</td>
</tr>
<tr>
<td>CFC(s)</td>
<td>Chlorofluorocarbon(s)</td>
</tr>
<tr>
<td>CTET</td>
<td>Carbon Tetrachloride</td>
</tr>
<tr>
<td>D</td>
<td>Dry (Imbibition)</td>
</tr>
<tr>
<td>DCM</td>
<td>Methylene Chloride</td>
</tr>
<tr>
<td>DNAPL(s)</td>
<td>Dense Non-Aqueous Phase Liquid(s)</td>
</tr>
<tr>
<td>EPA</td>
<td>United States Environmental Protection Agency</td>
</tr>
<tr>
<td>GC/MS</td>
<td>Gas Chromatography/Mass Spectrometry</td>
</tr>
<tr>
<td>HDPE</td>
<td>High Density Polyethylene</td>
</tr>
<tr>
<td>LNAPL(s)</td>
<td>Light Non-Aqueous Phase Liquid(s)</td>
</tr>
<tr>
<td>MCL</td>
<td>Maximum Concentration Limit</td>
</tr>
<tr>
<td>MFDV</td>
<td>Model with Fluids of Differing Viscosities</td>
</tr>
<tr>
<td>MSDS</td>
<td>Material Safety Data Sheets</td>
</tr>
<tr>
<td>NAPL(s)</td>
<td>Non-Aqueous Phase Liquid(s)</td>
</tr>
<tr>
<td>NECER</td>
<td>Network of European Centrifuges for Environmental Research</td>
</tr>
<tr>
<td>NICCA</td>
<td>Negligible Inertia Constant Contact Angle (Model)</td>
</tr>
<tr>
<td>NIVCA</td>
<td>Negligible Inertia Variable Contact Angle (Model)</td>
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<tr>
<td>NPL</td>
<td>National Priority List</td>
</tr>
<tr>
<td>ODP</td>
<td>Ozone Depleting Potential</td>
</tr>
<tr>
<td>QUMPFS</td>
<td>Queen’s University Multi-Phase Flow Simulator</td>
</tr>
<tr>
<td>PAH(s)</td>
<td>Polycyclic Aromatic Hydrocarbon(s)</td>
</tr>
<tr>
<td>PCB(s)</td>
<td>Polychlorinated Biphenyl(s)</td>
</tr>
<tr>
<td>PCE</td>
<td>Tetrachloroethylene</td>
</tr>
<tr>
<td>POP(s)</td>
<td>Persistent Organic Pollutant(s)</td>
</tr>
<tr>
<td>PPR</td>
<td>Pulses per Revolution</td>
</tr>
<tr>
<td>PW</td>
<td>Pre-Wetted (Imbibition)</td>
</tr>
<tr>
<td>PWD</td>
<td>Prewet and Dried (Imbibition)</td>
</tr>
<tr>
<td>RPM</td>
<td>Revolutions Per Minute</td>
</tr>
<tr>
<td>TCA (or 1,1,1-TCA)</td>
<td>1,1,1-Trichloroethane</td>
</tr>
<tr>
<td>TCE</td>
<td>Trichloroethylene</td>
</tr>
<tr>
<td>TCM</td>
<td>Chloroform</td>
</tr>
<tr>
<td>UNEP</td>
<td>United Nations Environment Programme</td>
</tr>
</tbody>
</table>
Symbols—English

\( 1/H \)  
Drop shape factor, function of shape parameter

\( a_t \)  
Total gravitational acceleration at point \( M \) of centrifuge experimental package

\( b \)  
Dynamic contact angle-interface velocity relationship fitting parameter

\( b_c \)  
Height from the base of the centrifuge platform to the platform trunnion

\( b_j \)  
Dynamic contact angle-interface velocity relationship fitting parameter \((j = 1, 2)\)

\( c \)  
Initial penetration (Bosanquet) velocity

\( Ca \)  
Capillary number

\( C_{OS} \)  
Capacitance of frequency-to-voltage conversion circuit

\( C_{INT} \)  
Integration capacitance of frequency-to-voltage conversion circuit

\( \partial x \)  
Partial derivative operator of any variable \( x \)

\( d \)  
Capillary tube diameter

\( d_j \)  
Capillary tube dip in interfacial tension measurements \((j = 1, 2)\)

\( d_e \)  
Equatorial diameter of pendant drop

\( d_{ref} \)  
Length of reference to measure pendant drop diameters

\( d_s \)  
Diameter of selected plane of pendant drop

\( dx \)  
Differential operator of any variable \( x \)

\( dz_c/dt \)  
Interface displacement (model) velocity

\( dZ_c/dt \)  
Interface displacement prototype velocity

\( e \)  
Local fracture aperture

\( e_v \)  
Capillary tube entry drag force coefficient

\( E \)  
Local prototype fracture aperture

\( E_v \)  
Rate of work dissipated irreversibly by system

\( f \)  
Dynamic contact angle-capillary number function

\( f_1 \)  
Dynamic contact angle-capillary number function

\( f_{cd} \)  
Drag force at the entry to the capillary tube

\( f_{in} \)  
Frequency of centrifuge RPM input signal

\( f_p \)  
Magnitude of the pinning force associated with one pinning point

\( F^* \)  
Excess force term

\( g \)  
Earth's gravitational acceleration

\( h \)  
Reservoir DNAPL pool height (includes any pre-infiltration) or reservoir driving head

\( h_{wp} \)  
Distance from phreatic surface to top of DNAPL pool

\( h_i \)  
DNAPL infiltration (or critical) (model) pool height

\( h_j \)  
Heights of reference in viscosity measurements and heights of capillary rise in interfacial tension measurements \((j = 0, 1, 2)\)

\( h_e \)  
Capillary rise equilibrium height
$h_p$ DNAPL pool height contained in the reservoir tube
$h_{pi}$ Depth of DNAPL pre-infiltration into a capillary tube
$h_\mu$ Characteristic length dependent upon the viscosity contrast between DNAPL and water
$H_i$ DNAPL infiltration (or critical) prototype pool height
$i_{cs}$ Internal source current of frequency-to-voltage conversion circuit
$k$ Fracture intrinsic permeability
$k_w$ Hydraulic conductivity of water in a capillary tube
$k_{rj}$ Fracture relative permeability of fluid $j$
$k_\Delta$ (Model) equivalent conductivity of a fluid of density $\Delta \rho$ and viscosity equal to the viscosity of water
$k_{\Delta \Delta}$ Equivalent conductivity of a fluid of density $\Delta \rho$ and viscosity $\Delta \mu$
$K_\Delta$ Prototype equivalent conductivity of a fluid of density $\Delta \rho$ and viscosity equal to the viscosity of water
$K_{OC}$ Organic carbon-water partition coefficient
$K_{OW}$ Octanol-water partition coefficient
$KE$ Kinetic energy of system
$l$ Capillary tube flow (model) length
$l_e$ Transition length of capillary tube to parabolic profile
$l_j$ Length of capillary tube occupied by fluid $j$
$l_t$ Total length of capillary tube (includes pre-infiltration length)
$L$ Capillary tube flow prototype length
$m$ Hagenbach (entry drag force) correction
$m'$ Couette (entry drag force) correction coefficient
$m_e$ Dry mass of capillary tube
$m_f$ Mass of capillary tube filled with distilled water
$n$ Centrifuge g-level
$n_p$ Density of defects (pinning points) present on the capillary tube walls
$N$ Number of integration time constants of frequency-to-voltage conversion circuit
$o(x)$ Function of the order of $x$
$p$ Pressure
$p_{0j}$ Reference pressure of fluid $j$
$p_1$ Pressure at the capillary tube inlet cross-section
$p_c$ (Dynamic) capillary pressure at liquid/fluid interface
$p_e$ Fracture local entry pressure
$p_j$ Pressure of fluid $j$
$p_j^*$ Dimensionless pressure of fluid $j$
$P_e$ Fracture local prototype entry pressure
$P_j$ Prototype pressure of fluid $j$
$PE$ Potential energy of system
$q$ Turbulence drag coefficient
\( Q \)  
Volumetric flow of liquid through a capillary tube

\( r \)  
Radial coordinate of cylindrical (or spherical) coordinate system

\( r^* \)  
Principal radius of curvature at liquid/fluid interface

\( r'' \)  
Principal radius of curvature at liquid/fluid interface

\( r^* \)  
Dimensionless radial coordinate

\( r_0 \)  
Capillary tube radius

\( r_a \)  
Radius of curvature of pendant drop at its apex

\( r_C \)  
Length of centrifuge arm

\( r_r \)  
Reservoir tube radius

\( r_G \)  
Radial distance between centrifuge axis and center of gravity of experimental package/platform system

\( r_{mc} \)  
Mean radius of curvature at liquid/fluid interface

\( r_M \)  
Radial distance between centrifuge axis and given point \( M \) of the experimental package

\( R_{INT} \)  
Integration resistance of frequency-to-voltage conversion circuit

\( R_{P,j} \)  
Principal radius of curvature at a point \( P \) of a pendant drop \((j = 1, 2)\)

\( R_{V/C}(z_c) \)  
Ratio of NIVCA predicted velocity to NICCA predicted velocity at depth \( z_c \)

\( \text{Re} \)  
Reynolds number

\( S \)  
Shape parameter of pendant drop

\( t \)  
(Model) time

\( t^* \)  
Dimensionless time

\( t_0 \)  
Reference representative time

\( T \)  
Prototype time

\( t_{c0} \)  
Time of infiltration at which perfect wetting starts

\( t_j \)  
Time of reference in viscosity measurement \((j = 1, 2)\)

\( t_{OS} \)  
Integration time of current source of frequency-to-voltage conversion circuit

\( T_{nw/w} \)  
Tension term acting to reduce the infiltration rate

\( u \)  
Longitudinal fluid velocity

\( u_0 \)  
Interface displacement velocity

\( u_{0j} \)  
Longitudinal reference displacement velocity of fluid \( j \)

\( u_c \)  
Perfect wetting velocity

\( u_j \)  
Longitudinal velocity of fluid \( j \)

\( u_j^* \)  
Dimensionless longitudinal velocity of fluid \( j \)

\( u_r \)  
Centripetal fluid velocity

\( v_{0j} \)  
Radial reference displacement velocity of fluid \( j \)

\( v_{cr} \)  
Critical interface velocity for turbulence drag to trigger

\( v_j \)  
Radial velocity of fluid \( j \)

\( v_j^* \)  
Dimensionless radial velocity of fluid \( j \)

\( w \)  
Mass flow rate

\( V_{in} \)  
Centrifuge RPM input signal

\( V \)  
Centrifuge RPM DC output signal

\( V_{out} \)  
Centrifuge RPM DC output signal
$V_{PP}$  Peak-peak output ripple of frequency-to-voltage conversion circuit

$W$  Width of fracture of rectangular section

$W$  Rate of work done against the system surroundings

$x$  Horizontal longitudinal coordinate (or distance)

$y$  Distance from the base of the centrifuge platform to a given point $M$ of the experimental package

$y_G$  Distance from the base of the centrifuge platform to the center of gravity $G$ of the package/platform system

$z$  Vertical longitudinal (model) coordinate (or distance)

$z^*$  Dimensionless vertical longitudinal coordinate

$z_0$  Representative fractional length of capillary tube

$z_c$  Infiltration (model) length of displacing liquid in capillary tube

$z_{c0}$  Depth of infiltration at which perfect wetting starts

$Z$  Vertical longitudinal prototype coordinate (or distance)

$Z_c$  Infiltration prototype length of displacing liquid in capillary tube

**Symbols—Greek**

$\alpha$  Parameter describing the magnitude of inertia forces

$\alpha_n$  Parameter describing the magnitude of inertia forces at a centrifugal acceleration of $n$ times the Earth’s gravity

$\alpha_t$  Parameter describing the magnitude of inertia forces if pre-infiltration is observed

$\beta$  Ratio of capillary entry drag forces to viscous forces

$\beta_1$  Upper bound of ratio of capillary entry drag forces to viscous forces

$\beta_t$  Ratio of capillary entry drag forces to viscous forces if pre-infiltration is observed

$\beta_{t1}$  Upper bound of ratio of capillary entry drag forces to viscous forces if pre-infiltration is observed

$\gamma$  Retardation factor associated with dynamic contact angle dependence upon velocity

$\gamma_t$  Retardation factor associated with dynamic contact angle dependence upon velocity if pre-infiltration is observed

$\delta$  Ratio of capillary tube radius to fractional length

$\Delta$  Small change, uncertainty error or differencing operator

$\Delta h$  Difference between DNAPL (model) pool height and critical (or infiltration) (model) pool height

$\Delta h'$  Negative difference between DNAPL pool height and perfect wetting critical pool height

$\Delta H$  Difference between DNAPL prototype pool height and critical (or infiltration) prototype pool height
\( \Delta p \)  
Total driving pressure acting across a capillary tube

\( \Delta p_h \)  
Hydrostatic pressure drop across a capillary tube

\( \Delta \mu \)  
Viscosity contrast between DNAPL and water

\( \Delta \rho \)  
Density contrast between DNAPL and water

\( \varepsilon \)  
Coefficient equal to 2 for circular-section capillary tubes and 1 for rectangular-section capillary tubes

\( \zeta \)  
Length scale over which inertia forces are significant

\( \theta \)  
Contact angle at given interface

\( \theta_a \)  
Static advancing contact angle

\( \theta_d \)  
Dynamic (or moving) contact angle

\( \theta_{ij} \)  
Contact angle at the fluid \( i/j \) interface

\( \theta_r \)  
Static receding contact angle

\( \theta_s \)  
Static contact angle

\( \theta_w \)  
Contact angle at the microscopic length scale

\( \lambda \)  
Dimensionless reservoir inertia coefficient

\( \mu \)  
Dynamic viscosity of given fluid

\( \mu_j \)  
Dynamic viscosity of fluid \( j \)

\( \xi \)  
Meniscus slip coefficient

\( \rho \)  
Density of given fluid

\( \rho_j \)  
Density of fluid \( j \)

\( \sigma \)  
Interfacial (or surface) tension of given liquid/fluid pair

\( \sigma_{ij} \)  
Interfacial (or surface) tension between fluids \( i \) and \( j \)

\( \sigma_{G/S} \)  
Surface tension of the solid/gas interface

\( \sigma_{L/S} \)  
Surface tension of the solid/liquid interface

\( \tau \)  
Time scale for which inertia forces are significant

\( \tau_i \)  
Characteristic time corresponding to the order \( i \)

\( \phi \)  
Inclination angle of the capillary tube to the horizontal

\( \phi \)  
Meniscus slip correction function

\( \Phi \)  
Turbulence drag function

\( \psi \)  
Inclination of the centrifuge platform to the horizontal

\( \Psi \)  
Angular coordinate of cylindrical coordinate system

\( \omega \)  
Centrifuge rotational velocity

**Common subscripts**

\( i/j \)  
Interface between fluid \( i \) and fluid \( j \)

\( j \)  
Fluid \( j \), either wetting or non-wetting

\( nw \)  
Non-wetting fluid

\( w \)  
Wetting fluid
CHAPTER 1

INTRODUCTION

1.1 Overview

1.1.1 Groundwater Contamination by DNAPLs

The acronym NAPL(s) standing for non-aqueous phase liquid(s) did not appear until 1981 when scientists and lawyers dealing with a hazardous waste landfill in Niagara Falls, New York, came across a black mixture of chlorinated solvents and other halogenated hydrocarbons, immiscible with, and denser than, water [Foster et al., 1996; Pankow et al., 1996]. At this time, the term NAPL was adopted to differentiate the liquid from other contaminated material and from the groundwater. This is because the term “fluid immiscible with water,” which had commonly been used to refer to hydrophobic petroleum compounds, was thought to be misleading in a context of groundwater contamination, as it did not clearly indicate that the organic fluid had a low-solubility in water. In the case of groundwater contamination, this is significant because even low-solubility NAPLs can potentially create long-term drinking water problems. Thus, from the early eighties, the term non-aqueous phase liquid became the politically correct form of immiscible fluid previously used in the literature [Morel-Seytoux, 1969; Bear, 1972; Corey, 1986].

In more recent years, the United States Environmental Protection Agency (EPA) has recognized that groundwater contamination is an increasingly important environmental concern [Huling and Weaver, 1991]. Of particular interest, groundwater contamination by entrapped dense non-aqueous phase liquid(s) (DNAPL(s)) has become widespread in many industrialized nations. The term “dense non-aqueous phase liquid(s)” refers to low-aqueous solubility organic chemicals having densities higher than that of water. In contrast, light non-aqueous phase liquid(s) (LNAPL(s)) are low-aqueous solubility organic chemicals having densities lower than that of water. At hazardous waste sites in the United States, as illustrated in Table 1.1, nine out the twenty most frequently detected groundwater contaminants are aliphatic chlorinated hydrocarbons, which are among the most common DNAPLs [National Research Council, 1994]. The results of other studies differ slightly [Barbee, 1994], but still give evidence that chlorinated alkanes and alkenes are among the most detected organic pollutants in groundwater. DNAPL contamination of groundwater has also been reported in many other industrialized nations including Canada [Feenstra, 1992;
National Research Council, 1994], the United Kingdom, Italy, the Netherlands [Rivett et al., 1990; Steele et al., 1999], and Germany [Schwille, 1988].

Table 1.1. The 20 Most Frequently Detected Groundwater Contaminants

<table>
<thead>
<tr>
<th>Rank</th>
<th>Compound b</th>
<th>Common Source c</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Trichloroethylene (TCE)</td>
<td>Metal cleaning/degreasing</td>
</tr>
<tr>
<td>2</td>
<td>Lead</td>
<td>Gasoline (prior to 1975), mining, construction material (pipes)</td>
</tr>
<tr>
<td>3</td>
<td>Tetrachloroethylene (PCE)</td>
<td>Dry cleaning, chemical intermediate</td>
</tr>
<tr>
<td>4</td>
<td>Benzene</td>
<td>Gasoline, manufacturing</td>
</tr>
<tr>
<td>5</td>
<td>Toluene</td>
<td>Gasoline, manufacturing</td>
</tr>
<tr>
<td>6</td>
<td>Chromium</td>
<td>Metal plating</td>
</tr>
<tr>
<td>7</td>
<td>Methylene Chloride (DCM)</td>
<td>Aerosol, paint removal, manufacturing</td>
</tr>
<tr>
<td>8</td>
<td>Zinc</td>
<td>Manufacturing, mining</td>
</tr>
<tr>
<td>9</td>
<td>1,1,1-Trichloroethane (TCA)</td>
<td>Metal and plastic cleaning/degreasing</td>
</tr>
<tr>
<td>10</td>
<td>Arsenic</td>
<td>Mining, manufacturing</td>
</tr>
<tr>
<td>11</td>
<td>Chloroform (TCM)</td>
<td>Solvents, rubber manufacturing</td>
</tr>
<tr>
<td>12</td>
<td>1,1-Dichloroethene (1,1-DCA)</td>
<td>Degreasing, solvents, manufacturing</td>
</tr>
<tr>
<td>13</td>
<td>Trans-1,2-Dichloroethene (1,2-DCE)</td>
<td>Transformation product of TCA</td>
</tr>
<tr>
<td>14</td>
<td>Cadmium</td>
<td>Mining, plating</td>
</tr>
<tr>
<td>15</td>
<td>Manganese</td>
<td>Manufacturing, mining</td>
</tr>
<tr>
<td>16</td>
<td>Copper</td>
<td>Manufacturing, mining</td>
</tr>
<tr>
<td>17</td>
<td>1,1-Dichloroethene (1,1-DCE)</td>
<td>Manufacturing</td>
</tr>
<tr>
<td>18</td>
<td>Vinyl chloride</td>
<td>Plastic and record manufacturing</td>
</tr>
<tr>
<td>19</td>
<td>Barium</td>
<td>Manufacturing, energy production</td>
</tr>
<tr>
<td>20</td>
<td>1,2-Dichloroethene (1,2-DCA)</td>
<td>Solvent, manufacturing</td>
</tr>
</tbody>
</table>

a Ranking generated using groundwater data from the National Priority List of sites to be cleaned up under the Superfund Act [National Research Council, 1994].

b Compounds that are DNAPLs are denoted in italic.

c After National Research Council [1994] and Pankow et al. [1996].

1.1.2 Usage of Chlorinated Solvents and other DNAPLs

From 1986 to 1994, the United States production of 1,2-dichloroethane (1,2-DCA), a common solvent for fats, oils and waxes, as well as a compound used to manufacture vinyl chloride, increased from 5.9 million metric tons to 7.6 million metric tons. During the same period, the production of other chlorinated solvents, such as tetrachloroethylene (PCE), chloroform (TCM), methylene chloride (DCM), carbon tetrachloride (CTET) or 1,1,1-trichloroethane (1,1,1-TCA or more simply TCA) was of the order of two to three hundred thousand metric tons [Feenstra, 1994; USITC, 1995;
Pankow et al., 1996]. Uses of chlorinated solvents are extremely varied (see Table 1.1) and include manufacture of resin, rubber, fumigant, insecticide, perfume, dye and paint. Chlorinated solvents are also frequently used as a means of degreasing and cleaning metal, leather, wool, fabrics and plastics.

Many halogenated compounds, whether they contain fluorine, bromide or iodine, are also DNAPLs at typical ambient temperatures. While less common than chlorinated solvents (with the exception of fluorinated compounds), they can be used as aerosol propellants, fire extinguishing fluids, refrigerants, organic synthesis solvents, or cleaning solvents for the manufacture of electronics or precision equipment. Generally, they can substitute chlorinated solvents in a large number of manufacturing processes [Pankow et al., 1996].

DNAPLs also include polychlorinated biphenyl(s) (PCB(s)), such as Arochlor 1254, which were used for many applications until their production was banned in the United States in the mid-seventies. PCBs can be found in plasticizers and polyester resins, or used as insulator fluids for electrical equipment. Finally, DNAPLs also include a large number of pesticides, such as chlordane, dichlorvos or parathion, as well as creosote and coal tars commonly used as wood preservatives and roofing or road paving. Crude coal tars are typically a mixture of polycyclic aromatic hydrocarbon(s) (PAH(s)), and are formed as by-products of the destructive distillation of coal, coke-ovens being the major source. Source of production, physical properties and environmental fate of these compounds are reviewed in Montgomery [2000].

1.1.3 DNAPL Environmental Fate and Health Risks

The aqueous solubility of DNAPLs in water varies from a few milligrams to a few grams per liter depending on the compound [Demond and Lindner, 1993; Howard and Meylan, 1997]. However, no matter how low, DNAPL solubilities are often larger than regulated drinking water limits by several orders of magnitude [Cary et al., 1989]. For example, the solubilities of trichloroethylene (TCE) and PCE are 1.1 g/l and 0.2 g/l at 20°C, respectively. The maximum concentration limit (MCL) set by the EPA is 5 µg/l for both compounds. Therefore, the solubility/MCL ratio for TCE and PCE are equal to $2 \times 10^5$ and $4 \times 10^4$, respectively [Pankow et al., 1996]. Thus, not only do DNAPLs pose a serious contamination problem as a pure organic phase, they also are a reason for concern as a dissolved aqueous species, as they can be a source of groundwater contamination when present, even in small quantities, in the subsurface environment.

In practice, at waste disposal sites, DNAPLs are frequently mixtures of many organics chemicals, reflecting the disposal of various chemical products [Feenstra and Cherry, 1996]. The mixtures can consist of pure-phase DNAPLs, or mixtures of DNAPLs and common LNAPLs, such as benzene (see Table 1.1). Moreover, the mixtures may contain compounds that are miscible with water, such as acetone, thereby increasing the mobility and the aqueous solubility of the mixtures. While considerable
information exists for pure DNAPLs, there is little knowledge on DNAPL-based field or waste samples, whose properties can differ considerably from those of pure solvents [Steele et al., 1999]. As illustrated in Table 1.2, DNAPL mixtures found at disposal sites can have an extremely complex chemical composition and frequently contain compounds that cannot be identified using conventional analytical methods, such as gas chromatography/mass spectrometry (GC/MS) [Feenstra and Cherry, 1996].

Table 1.2. Example of Chemical Composition of a DNAPL Sample Recovered from a Landfill at a Chemical Manufacturing Facility

<table>
<thead>
<tr>
<th>Compound b</th>
<th>Weight [%]</th>
<th>Specific Gravity c</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chlorobenzene</td>
<td>4.6</td>
<td>1.107</td>
</tr>
<tr>
<td>Tetrachloroethylene</td>
<td>4.5</td>
<td>1.6230</td>
</tr>
<tr>
<td>Toluene</td>
<td>3.9</td>
<td>0.866</td>
</tr>
<tr>
<td>Trichlorofluorotoluenes</td>
<td>3.0</td>
<td></td>
</tr>
<tr>
<td>1,2-Dimethylbenzene (o-xylene)</td>
<td>2.5</td>
<td>0.8801</td>
</tr>
<tr>
<td>1,3-Dimethylbenzene (m-xylene)</td>
<td>2.4</td>
<td>0.8684 d</td>
</tr>
<tr>
<td>1,1,2,2-Tetrachloroethane</td>
<td>0.92</td>
<td>1.58658 e</td>
</tr>
<tr>
<td>Trichloroethylene</td>
<td>0.75</td>
<td>1.4649</td>
</tr>
<tr>
<td>Dichlorotoluene</td>
<td>5.1</td>
<td>1.26 f</td>
</tr>
<tr>
<td>1,1’-Thiobisdodecane</td>
<td>1.7</td>
<td></td>
</tr>
<tr>
<td>Pentachloroethane</td>
<td>0.87</td>
<td>1.6712 e</td>
</tr>
<tr>
<td>Methyl ester (p-chlorophenyl) phenyl acetic acid</td>
<td>0.62</td>
<td></td>
</tr>
<tr>
<td>Unresolved by GC/MS analysis</td>
<td>61.1</td>
<td></td>
</tr>
</tbody>
</table>

a After Feenstra and Cherry [1996].
b Mixture density = 1.5 kg/l.
c Values from Budavari [1996]. Specific gravity at 20°C referred to water at 4°C unless otherwise noted.
d Specific gravity at 15°C referred to water at 4°C.
e Specific gravity at 25°C referred to water at 4°C.
f Unspecified temperature.

Most DNAPLs are suspected or known carcinogens. PCBs are notorious for their ability to bio-accumulate and have been shown to be endocrine-disruptive substances, meaning that they are able to mimic hormones. The effect of PCBs on wildlife and human populations has been documented [Jacobson and Jacobson, 1996; Eckley, 2001]. For example, it is well known that pollutants such as PCBs produced in industrialized countries can travel large distances and enter the Arctic food chain. High PCB levels have been measured in the blood, fatty tissues, and breast milk of women living in Northern Canada [Myers, 2001]. PCBs as well as chlordane are two of the dirty dozen addressed by the Stockholm Convention on Persistent Organic Pollutants (POPs) signed in May 2001 under the coordination of the United Nations Environment
Programme (UNEP). Upon ratification and following implementation, this treaty would ban, phase out, or undertake efforts to limit the production of twelve POPs [Fisher, 1999; Eckley, 2001; Myers, 2001].

The Woburn tragedy is another example of a serious case of municipal drinking water contamination by chlorinated solvents, which was thought to have caused an elevated incidence of cases of childhood leukemia between the sixties and the eighties [Bureau of Environmental Health Assessment, 1997]. The groundwater contamination problem in Woburn prompted a major trial, which inspired Jonathan Harr’s novel A Civil Action [Harr, 1995] and subsequent feature film of the same name [Bair and Wood, 1999].

Some DNAPLs have been shown to be stratospheric ozone depleting substances [Molina and Rowland, 1974; Parson and Greene, 1995]. The ozone depleting DNAPLs include TCA, CTET, as well as the chlorofluorocarbons (CFCs) and halons that are denser than water and at a liquid state at ambient temperature, e.g. trichlorofluromethane (CFC-11), trichlorotrifluoroethane (CFC-113) [Jackson et al., 1992], and dibromotetrafluoroethane (halon-2402) [Lide, 1995; Montgomery, 2000]. The ozone depleting potential (ODP) is a numerical estimate of the total quantity destroyed by a given mass of the substance over its entire life. ODPs are measured relative to CFC-11, one of the most prevalent chlorofluorocarbon; that is, CFC-11’s ODP is set equal to 1.0. ODPs for TCA, CTET, CFC-113, and halon-2402 have been estimated to be respectively equal to 0.1, 1.1, 0.8, and 6.0 [EPA, 1995]. The Montreal Protocol (1987) and the subsequent London Amendment (1990) and Copenhagen Amendment (1992) have been designed to freeze, cut or eliminate the production of these compounds [Parson and Greene, 1995]. Consequently, the EPA has restricted their use to laboratory applications not to be resold or used in manufacturing [EPA, 1995].

1.1.4 The Challenges of Restoring DNAPL-Contaminated Groundwater

Poor past practices, accidental spills, inadequate underground or above-ground storage facilities, inadequate disposal practices, and damaged or decaying storage vessels or disposal ponds are factors that have contributed to the release of DNAPL compounds into the subsurface environment. For example, in the past, solvents were often disposed of onto waste ground in the belief that their high volatility would cause them to evaporate to the atmosphere and thus not infiltrate into the subsurface [Rivett et al., 1990]. DNAPL contamination can originate from both industrial sites and waste-disposal sites. Numerous case studies of sites contaminated by DNAPLs have been reported in the literature [e.g., Rivett et al., 1990; Jackson et al., 1992; Kueper et al., 1992; Feenstra, 1994; National Research Council, 1994; Foster et al., 1996].

Restoration of environmental systems contaminated with hazardous wastes is a large and costly problem. In the United States, as of October 2000, 1,450 highest priority hazardous waste sites were assigned to the national priority list (NPL) for
cleanup, with 59 additional sites proposed for inclusion [EPA, 2000]. These sites cover more than 3 million acres in total surface area [EPA, 1999]. However, it is believed that as many as 40,000 sites are potential candidates for the federal Superfund remediation program [EPA, 1999]. It is difficult to estimate costs, but it appears that the remedial cost at a Superfund site averages $9 million per site as of 1996, and that over the next 30 years, the United States could well spend between $480 billion and $1 trillion cleaning up sites [EPA, 1999]. For comparison, the 2001 gross national income of the United States was a little less than $10 trillion dollars [World Bank, 2002]. Given the expense, it is worth considering if the benefits of cleaning up these sites will seriously outweigh the costs.

Contamination of the subsurface environment by DNAPLs is an important subset of this overall restoration problem. DNAPLs are usually long-lived and extremely difficult to remove because of their ability to migrate deep into aquifer formations under gravity-driven flow, while leaving a residual trail of small blobs trapped by capillary forces (see Section 2.2.2). For that reason, DNAPLs are often long-term sources of groundwater pollution, for which restoration efforts could well be in perpetuity [Dekker and Abriola, 2000; Miller et al., 2000]. Traditional pump-and-treat remediation schemes based on direct pumping or pumping combined with water flushing have proven inefficient, costly and time-consuming for addressing DNAPL contamination.

The inadequacy of the pump-and-treat technology is often accompanied with insufficient site subsurface characterization, poor documentation of a DNAPL release, and the complexity of predicting DNAPL transport pathways [Huling and Weaver, 1991]. The EPA has concluded that 57% of all Superfund sites in the United States have a high or moderate potential for the presence of DNAPL, even though DNAPL was only observed at only 5% of them [Bedient et al., 1999]. In field investigation of sites where extensive contamination exists, pools of free-product solvent are found only rarely, even when their existence is not in doubt. Conventional site investigation techniques are not well-suited for pure-phase DNAPL detection, as they would otherwise require an unfeasibly large number of boreholes [Feenstra et al., 1996].

### 1.1.5 DNAPLs in Fractured Media Aquifers

In the United States, the National Research Council [National Research Council, 1994] has identified remediation of DNAPL-contaminated, fractured hydrogeological systems as an extreme technical challenge, and has stated that the clean-up of such sites to drinking water standards is currently unlikely. Yet, many public water supplies and private homes rely on fractured bedrock aquifers for their water. For this reason, the scientific and engineering community is showing more and more concern regarding the remediation of DNAPLs in fractured media [e.g., Wickramanayake and Hinchee, 1998; Pichtel, 2001].
Rock bodies are usually considered to be materials of low permeability. In the geologic environment, fractures (also called discontinuities) in rock masses interconnect and create conditions for fluid flow through discrete channels that in some fashion may form an interconnected system [Domenico and Schwartz, 1998; Ivanova, 1998].

There are two major types of fractures. A joint is a fracture along which no notable rock mass displacement has occurred [Domenico and Schwartz, 1998]. In contrast, a fault is a fracture where rock mass displacement has occurred parallel to the surface along which the break occurs [Stokes and Varnes, 1955; Domenico and Schwartz, 1998].

Ivanova [1998] has identified five major geologic settings for brittle fracturing. In the first category, fractures are generated by the folding of rock strata. Folding is either flexural (buckling or bending of the rock strata, see Figure 1.1.a and Figure 1.1.b) or shear folding (displacement oblique or perpendicular to bedding, see Figure
In the second category, fractures are related to crustal faults. Ivanova considers three major types of crustal faults. In a *strike-slip fault*, rock differential displacement is generally along the strike of the fault (see Figure 1.2.a). Faults with displacement along the line of maximum dip are called *dip-slip faults* and are either *normal faults* (downward movement of overhanging block, see Figure 1.2.b) or *thrust faults* (upward movement of overhanging block, see Figure 1.2.c) [Priest, 1993]. In the third category, fracturing occurs when the minimum principal stress in the rock mass is tensile (negative). Such fractures are referred to as *tensile joints* and are created by various geologic mechanisms such as crustal extension or folding. A typical tensile joint surface is shown in Figure 1.3. In the fourth category, tensile joints are created in response to rock contraction due to cooling, desiccation, chemical reaction or phase change. Finally, in the fifth category, fracture systems occur around central diapirc structures (rise of low density plastic material though an overlying rock of higher density) or collapse structures (downward caving-in of rock material into underlying voids or weaker materials) [Ivanova, 1998]. An example of such fracture system is shown in Figure 1.4.

![Figure 1.3. Typical irregular surface of a tensile joint and common associated morphological features [after Ivanova, 1998].](image)

From a hydrologic perspective, the most important characteristics of a set of fractures include its orientation, density, aperture opening, smoothness of walls and—possibly above all else—degree of connectivity [Domenico and Schwartz, 1998]. It is difficult to provide values of a typical density, degree of connectivity or aperture, as
these parameters vary by several orders of magnitude. A fracture aperture, for example, can be expected to vary between several micrometers to several centimeters. It is generally acknowledged that the geometry of complex fracture networks cannot be described deterministically; instead a stochastic modeling approach is often adopted where networks are generated with statistical fracture properties [e.g., Ivanova, 1998] that can be compared directly with those observed in the field [Steele et al., 1999]. Physical characteristics of fractures with respect to fracture flow have recently been reviewed in National Research Council [1996].

![Diagram of igneous intrusion and associated fracture system](image)

**Figure 1.4.** Example of igneous intrusion and associated fracture system [after Ivanova, 1998].

Once DNAPL enters a fracture system, the probability of encountering the non-aqueous phase liquid in a borehole where a sparse distribution of migration pathways exists will be very low [Kueper et al., 1992]. Thus, the site investigation of a contaminated fracture system will rarely delineate an accurate distribution of DNAPL within the fracture network, and possibly lead to the conclusion that DNAPL is not present at a site. With little information on the probable distribution of DNAPL at contaminated sites, and given the added complexity of characterizing a fracture network, the planning of an effective remediation strategy is rendered virtually impossible. For this reason, the development and/or validation of models for DNAPL invasion of fracture systems is a critical step towards improving DNAPL remediation performances in these aquifers.
1.2 Thesis Objectives and Outline

1.2.1 Scope of Thesis

The work described in this thesis is part of a larger project designed to ultimately predict the behavior of DNAPL in rough-walled fracture networks. The scope of the work presented in this thesis is three-fold:

1. To develop a theoretical model for predicting pure-phase DNAPL infiltration into vertical, initially water-saturated fractures. Specifically, the model developed in this research provides a criterion for which DNAPL infiltration takes place in the fracture, as well as a prediction of the DNAPL infiltration kinetics (i.e. a prediction of the rate of DNAPL infiltration into the fracture).

2. To validate the theoretical model experimentally by conducting a testing program on simplified fractures described by smooth-walled, vertical capillary tubes. Both the infiltration criterion and the infiltration kinetics are investigated experimentally. In the infiltration kinetics, the role played by gravity, viscous, inertia and capillary forces is examined. Based upon the experimental results, criteria are developed for which inertia forces can be neglected and/or capillary forces can be assumed to be independent of the infiltration velocity.

3. To assess the potential of the geotechnical centrifuge as an experimental tool to carry out physical modeling of DNAPL infiltration behavior in fracture systems. Again, simplified fractures described by smooth-walled, vertical capillary tubes are used to carry out the centrifuge experimental program.

The research presented here intends on examining, at a fundamental level, the balance of forces that are important in modeling the DNAPL/water flow. The information can then be used to construct more complex, numerical models enabling prediction of DNAPL behavior in more realistic systems, including the initial contamination in an interconnected fractured networks or the implementation of a DNAPL remediation scheme in a fractured system.

1.2.2 Organization of Thesis

In Chapter 2, basic concepts on DNAPL flow in water-saturated fractured media are first introduced, along with a review of previous experimental and numerical work relevant to the field. Next, the kinetics of infiltration into capillary tubes is reviewed. Models developed by prior investigators to describe this phenomenon are introduced, and their limits are discussed. Finally, the basic principles of centrifuge modeling are discussed along with a review of previous research on NAPL transport using the geotechnical centrifuge.

Chapter 3 presents a theoretical model used to predict the infiltration behavior of pure-phase DNAPL into a vertical fracture initially saturated with water under
hydrostatic conditions. Theoretical modeling of DNAPL infiltration into a fracture system first requires expressing the criterion for DNAPL infiltration, i.e. the condition for which DNAPL will proceed to penetrate into the fracture by displacing water. Modeling also requires giving a prediction of the kinetics of the DNAPL/water interface displacement once infiltration has begun, i.e. an expression of the interface location in the fracture as a function of time.

In Chapter 4, a reduced-scale physical model of DNAPL behavior in a single rough-walled vertical fracture is developed to obtain the scaling laws that allow translation from a reduced-scale centrifuge model to a full-scale prototype. Prediction of DNAPL behavior during a centrifuge experiment is achieved by extending the theoretical model developed in Chapter 3 at the Earth’s gravitational acceleration, g, to the case where the body force acting upon the fracture system is a centrifugal acceleration equivalent to \( n \) times the Earth’s gravity.

Chapter 5 focuses on the dense non-aqueous phase liquids (DNAPLs) used for the experimental program of this research. This chapter presents and discusses results of measurements of the viscosity and interfacial tension properties of the DNAPLs. Equipment cleaning and safety procedures used throughout the experimental program are also reviewed.

Chapter 6 presents an overview of the methods and equipment used for the experimental program of this research. The experimental methodology used for conducting DNAPL infiltration experiments at 1 g (laboratory experiments) and in the geotechnical centrifuge is presented. Specific emphasis is given on the centrifuge instrumentation and data acquisition system used as part of the centrifuge experimental program of this research.

Chapter 7 presents the experimental results of DNAPL infiltration experiments and their comparison to the predictions of the theoretical model and physical model developed in Chapter 3 and Chapter 4, respectively. The laboratory experiments (1-g experiments) and centrifuge experiment results are successively examined. Both the criterion for DNAPL infiltration and the DNAPL/water interface infiltration kinetics are discussed.

Finally, Chapter 8 summarizes the results of this study and proposes recommendations for future research to be carried out in the area of DNAPL/water flow in fractured media as well as centrifuge modeling of geoenvironmental processes.

1.3 References


EPA, Protection of stratospheric ozone: Administrative changes and amendment to transshipment provision in final rule to phase out ozone-depleting chemicals; Final rule and proposed rule, *Fed. Regist.*, 60(90), 24969-25009, 1995.


CHAPTER 2

BACKGROUND

2.1 Introduction

This chapter is divided into three main parts.

In Section 2.2, basic concepts relevant to DNAPL flow in water-saturated fractured media are introduced. Next, a conceptual model for DNAPL infiltration into the subsurface is presented with an emphasis on the infiltration into fracture systems. Finally, previous experimental and numerical work pertinent to two-phase flow in fractured media is discussed.

In Section 2.3, the kinetics of infiltration into capillary tubes is reviewed. Models developed by prior investigators to describe this phenomenon are introduced, and their limits are discussed. Experimental results that validate or invalidate the proposed models are also presented. Finally, the motion of contact lines and dynamic contact angle dependence upon interface meniscus velocity are discussed in depth, for both liquid/air systems and liquid/liquid systems.

In Section 2.4, the basic principles of centrifuge modeling are introduced along with specifics regarding the modeling of geoenvironmental processes. Prior investigation on the transport of NAPLs in porous media using centrifuge modeling is then presented and discussed.

A list of references is provided in Section 2.5.

2.2 DNAPLs in Fractured Media: Basic Concepts and Previous Research Work

2.2.1 Physical Concepts on Two-Phase Flow Through Fractured Media

2.2.1.1 Overview

Two-phase flow mechanics has been widely used to study flow in the unsaturated, or vadose zone, of the subsurface, where void spaces are partly filled by air and partly by groundwater [Bear and Verruijt, 1987]. Two-phase flow mechanics has also been
used in petroleum engineering to characterize the flow of water and oil in oil-bearing formations [Morel-Seytoux, 1969]. In recent years, two-phase flow mechanics has been applied to modeling the transport of organic contaminants in groundwater [Mackay et al., 1985; Hunt and Sitar, 1988].

This section examines the basic mechanics driving DNAPL and water flow through fractured media. Fractures have been predicted and shown to exhibit capillary behavior because of their rough-walled nature [Pruess and Tsang, 1990; Kueper and McWhorter, 1991; Reitsma and Kueper, 1994]. For that reason, two-phase flow concepts commonly used for describing the interaction mechanisms between aqueous, organic, solid and gas phases in porous media, such as wettability, interfacial tension and contact angle, can be used in the context of fractured media.

In the following sections, basic physical concepts behind two-phase flow are summarized with a specific emphasis on DNAPL/water flow in geologic media. The reader is referred to textbooks [e.g., Morel-Seytoux, 1969; Bear, 1979] for developments in two-phase flow mechanics not directly relevant to this thesis. An extensive review of NAPL behavior in the subsurface is given by Mercer and Cohen [1990].

2.2.1.2 Interfacial Tension and Capillary Pressure

When a liquid is in contact with another fluid, which can be a gas or another liquid barely miscible with the first liquid, a free interfacial energy arises from the difference between the inward attraction of the molecules in the interior of each phase and those at the surface of contact [Bear, 1979]. This force imbalance exerts a tension on the interface of the two fluids causing the interface to contract to as small an area as possible. This is somewhat similar to the behavior of a stretched membrane under tension, and in contact with two fluids on either side of the membrane.

The interfacial tension of a liquid/fluid pair (again, the fluid is either a gas or another liquid) is defined as the amount of work that must be performed in order to separate a unit of the fluid from the liquid. Thus, interfacial tension is expressed in units of force per distance, dynes/cm or N/m in SI units. The term surface tension is often substituted for interfacial tension when the liquid/fluid pair is constituted by a liquid and its own vapor. This convention is used throughout this thesis.

In practice, the greater the interfacial tension between two liquids; the less likely an emulsion will form (an emulsion is defined as a suspension of small globules of one liquid in a second liquid); emulsions will be more stable if formed; and the better the phase separation after mixing [Huling and Weaver, 1991]. Interfacial tension decreases with increasing temperatures, and maybe affected by pH, surfactants and gases in solution [Huling and Weaver, 1991]. Values of interfacial tension vary from zero for completely miscible liquids to 0.0728 N/m for the surface tension of water at 20°C [Munson et al., 1994]. Values of interfacial tension between water and most pure NAPLs range between 0.015 N/m and 0.050 N/m. The interfacial tension of many
chlorinated solvents ranges between 0.030 N/m and 0.040 N/m [Mercer and Cohen, 1990; Demond and Lindner, 1993]. As discussed in Section 1.1.3, field or waste DNAPL samples may have physical properties that differ largely from those of pure samples [Steele et al., 1999]. This is particularly true of DNAPL/water interfacial tension, whose magnitude could be lowered should a compound with affinity to water, such as acetone, be present in the DNAPL phase.

As a consequence of interfacial tension, when two immiscible fluids are in contact, a discontinuity in pressure exists across the interface separating them. The magnitude of the pressure difference depends on the curvature of the interface at the point where it is considered. Point in this context means the microscopic point within the macroscopic void space under consideration. The capillary pressure, commonly denoted $p_c$, is defined as the difference in pressure between a liquid and a fluid, where the pressures are taken in the two phases as the interface is approached from their respective sides.

**Figure 2.1.** Forces at a curved liquid/fluid interface [after Bear and Verruijt, 1987].

Figure 2.1 shows an infinitesimal element of a curved interface between two fluids, 1 and 2, where either both or only one of the two fluids is a liquid. Given the curvature, the pressure in fluid 1 is larger than that in fluid 2. By writing a balance of force components along the normal to the interface, with a constant interfacial tension $\sigma_{1/2}$, one obtains

$$p_c = p_1 - p_2 = \sigma_{1/2} \left( \frac{1}{r^n} + \frac{1}{r^m} \right) = \frac{2\sigma_{1/2}}{r_{mc}},$$

(2.1)
where $p_1$ and $p_2$ denote the pressures in fluid 1 and fluid 2, respectively, $r'$ and $r''$ denote the principal radii of curvature, and $r_{mc}$ is the mean radius of curvature defined by $2/r_{mc} = 1/r' + 1/r''$.

In a geologic medium, the radius $r_{mc}$ will be related to the dimensions of the void space where the fluids are in contact. In a porous medium, $r_{mc}$ will be of the order of the radius of a local pore throat. In a fractured medium, $r_{mc}$ will be of the order of the fracture aperture. Equation (2.1) is known as the Laplace formula for capillary pressure [Bear and Verruijt, 1987]. A detailed derivation of this equation can be found in Morel-Seytoux [1969].

2.2.1.3 Wettability and Contact Angle

Interfacial tension and capillary pressures are closely related to another physical property, wettability. Wettability in a two-fluid system is defined as the tendency of one fluid to spread preferentially over a solid surface in favor of the second fluid [Bedient et al., 1999]. For example, a drop of water on an untreated glass surface typically spreads on that surface and tends to stretch into a thin film. Under these circumstances, water is said to be wetting with respect to air. Conversely, a mercury drop rolls over an untreated glass surface but spreads very little. Thus, in this system, mercury is said to be non-wetting with respect to air. It is important to note that wettability is not a state variable for a given fluid, as the wettability of a fluid may change depending upon the system in which it finds itself. For example, in the illustrations given above, air was both wetting and non-wetting.

Wettability is depicted by the concept of contact angle. Considering a droplet of test liquid formed on a solid surface in the presence of a surrounding second fluid, as shown in Figure 2.2, the contact angle, $\theta$, is defined as the angle between the solid surface plane and the tangent plane at the point of contact of the interface with the solid surface. When $\theta = 0^\circ$, the wetting fluid is said to perfectly wet the solid surface.

![Figure 2.2. Definition of wettability on the basis of the contact angle [after Domenico and Schwartz, 1998].](image-url)
The contact angle is often measured through the water phase [Sahimi, 1995]. In the absence of water, it is measured through the denser phase [Bear and Verruijt, 1987]. Strictly speaking, if $\theta < 90^\circ$, the liquid contained in the droplet is defined as the wetting fluid, while the fluid surrounding the droplet is the non-wetting fluid. However, in practice, $\theta < 65^\circ$ for wetting droplets, while $105^\circ < \theta < 180^\circ$ for non-wetting droplets. In the intermediate case, $65^\circ < \theta < 105^\circ$, the solid surface has no strong preference for either fluid [Sahimi, 1995]. Mercer and Cohen [1990] provide results of contact angle experiments using several DNAPLs and various substrates. Note that hysteresis of contact angles as well as effects related to the contact angle of an interface meniscus in motion (or dynamic contact angle) are discussed in Section 2.3.5 and Section 2.3.6.

A direct consequence of the definition of wettability is that the capillary pressure, as defined by (2.1), is the difference between the non-wetting fluid pressure and the wetting fluid pressure. In other words, fluid 1 in Figure 2.1 is the non-wetting fluid, while fluid 2 is the wetting fluid.

Applying the concept of wettability to two-phase flow transport in specific field situations requires a comprehensive physical description of the solid surfaces. Thus, one must be able to describe the wettability properties of soil grains or fracture surfaces at a representative scale. However, in most practical cases, a generalization is made. NAPLs are most often a wetting fluid when combined with air, and water is almost always the wetting fluid with respect to NAPLs or air on rock-forming minerals [Bear, 1972; Domenico and Schwartz, 1998]. Exceptions exist, for example in the presence of organic matter (coal, peat, humus), graphite, and talc-like silicate, where water is not strongly wetting with respect to NAPLs [Mercer and Cohen, 1990]. While some researchers have documented that petroleum reservoirs, particularly limestone and dolomite, may be partially or preferentially wet by oil, most natural media are strongly water-wet if not contaminated by NAPL. However, prior exposure to the immiscible phase can drastically change aquifer wettability [Steele et al., 1999].

In two-phase flow displacement in porous or fractured media, the fluid drawn into a pore throat or fracture is the wetting fluid, while the fluid repelled by capillary forces is the non-wetting fluid. Hence, capillary pressure is a measure of the tendency of a porous or fractured medium to imbibe the wetting phase or to repel the non-wetting phase. For example, water will spontaneously rise in a dry mass of porous material. The corresponding suction pressure, expressed as a negative pressure head, will be related to the capillary pressure as defined by (2.1). Conversely, a fracture or a small soil pore initially saturated with water and exhibiting capillary properties will provide resistance to the infiltration of NAPL. The entry pressure required to force NAPL into the fracture or pore will be equal to the minimum capillary pressure which achieves interface displacement [Domenico and Schwartz, 1998]. It is common to define this pressure as

$$p_c = p_1 - p_2 = \frac{2\sigma_{12} \cos \theta}{r_{nc}},$$  \hspace{1cm} (2.2)
which is (2.1) integrated over the curved surface in the medium, taking into account the boundary conditions at the solid surface.

Capillary pressure given by (2.2) results in a capillary force, the absolute magnitude of which increases as $r_{mc}$ decreases. For a wetting fluid displacing a non-wetting fluid, this force acts to draw the wetting fluid into the medium. Sometimes this phenomenon is referred to as soil capillarity. For a non-wetting fluid displacing a wetting fluid, the capillary force acts as a resistance force, preventing the non-wetting fluid from easily infiltrating the medium. Sometimes this phenomenon is called capillary resistance.

The displacement of a non-wetting fluid by a wetting fluid is commonly referred to as imbibition. The reverse process, non-wetting fluid displacing a wetting fluid, is called drainage. Therefore, DNAPL infiltration into an initially water-saturated fracture is a drainage process.

2.2.1.4 Viscosity and Density

Density is the mass per unit volume of a substance. It can be presented in terms of specific gravity, which is the ratio of a substance’s density to that of a standard substance, usually water. Density is a function of temperature [Munson et al., 1994].

The dynamic viscosity, absolute viscosity, or simply viscosity is the internal friction within a fluid that causes it to resist flow due to molecular cohesion. Viscosity is expressed in SI units of N.s/m² or Pa.s. It decreases with temperature [Munson et al., 1994]. NAPL viscosity may vary with time, as the more volatile components of a NAPL mixture are lost through evaporation, resulting in an oily mixture constituted of heavier and more viscous components [Huling and Weaver, 1991]. The ratio of dynamic viscosity to density is called the kinematic viscosity. Mercer and Cohen [1990] provide tables of density and viscosity of common NAPLs.

Mixed DNAPLs at waste disposal sites have densities ranging between 1.05 kg/l to 1.3 kg/l, which tend to be lower than pure chlorinated solvents. This is due to the presence of non-chlorinated petroleum hydrocarbons, which are LNAPLs (see, for example, Table 1.2) [Feenstra and Cherry, 1996]. The larger the density of a DNAPL compound, the larger the density contrast with water and the greater the downward mobility (or mobility due to gravity) of this compound as a pure organic phase. Relative density contrasts between DNAPL and water of about 1% are known to influence fluid movement significantly in the subsurface environment [Mackay et al., 1985], suggesting that even DNAPLs of low-density with respect to water have a potential for downward mobility.

The viscosities of mixed DNAPLs vary between $10^{-2}$ N.s/m² and $10^{-1}$ N.s/m² [Feenstra and Cherry, 1996], but are usually smaller for pure DNAPL chemicals. This range of values can be compared to the viscosity of water, which is approximately $10^{-3}$ N.s/m² at 20°C [Munson et al., 1994]. Following a spill, a DNAPL compound of
low viscosity will penetrate more rapidly into the soil than a compound of higher viscosity.

Overall, the potential mobility of DNAPL in the subsurface will be greater for substances with higher density and lower viscosity. As illustrated in Figure 2.3, aliphatic chlorinated solvents have the lowest viscosities and highest densities. Thus, they are the most mobile in groundwater. In contrast, coal tar and creosote are only slightly denser than water (1.01 kg/l to 1.05 kg/l), and their viscosities are one to two orders of magnitude greater than that of water. These properties make coal tar and creosote the least mobile of the common DNAPLs as a pure phase.

![Figure 2.3. Relative mobility of selected DNAPLs as a function of density and viscosity [after Feenstra and Cherry, 1996].](image)

2.2.1.5 Partitioning of DNAPLs Into Other Phases

In Chapter 1, DNAPLs were defined as organic liquids of low-aqueous solubility and of density larger than water. In a context of groundwater contamination, DNAPL is a liquid chemical or a liquid mixture of organic chemicals having a density greater than 1.01 kg/l. To be classed as a DNAPL, an organic compound must have an aqueous solubility of less than 2% by weight and a vapor pressure of less than 0.4 atm (300 mm Hg) [Feenstra, 1994]. Therefore, DNAPLs may be present in the subsurface
in various physical states or phases as illustrated in Figure 2.4. Liquids of higher solubility or vapor pressure would not likely persist for a significant time in the subsurface as a separate immiscible phase [Feenstra, 1994]. Nonetheless, the vapor pressure is sufficiently high to result in the formation of significant plumes of chemical vapors surrounding areas of DNAPL in the vadose zone [Feenstra, 1994].

![Figure 2.4. Four phase schematic representation of a porous medium. DNAPLs may be present in any phase. The concentration in a given phase is controlled by the phase/DNAPL partition coefficient [adapted from Huling and Weaver, 1991].](image)

Generally, it is found that DNAPL liquid chemicals have aqueous solubilities of the order of milligrams to grams per liter of water, and vapor pressures in the range $10^{-8}$ atm to $10^{-1}$ atm. Because of their hydrophobicity and size, polycyclic aromatics, such as PCBs, have very low vapor pressures and aqueous solubilities. As illustrated in Figure 2.5, the larger the aqueous solubility of a given DNAPL, the greater its potential for mobility as a dissolved species.
Figure 2.5. Relative mobilities of selected DNAPLs compared with those of selected non-halogenated organic substances. CBz, chlorobenzene; Bz, benzene; Tol, toluene; Xyl, xylenes; Naph, naphtalene; Phen, phenantracene; B(a)P, benzo(a)pyrene [after Feenstra, 1994].

DNAPLs may also adsorb onto the soil gains of a porous medium or sorb to the walls of a fracture system, and therefore be transported more slowly than the groundwater [Mutch and Scott, 1994]. Specifically, sorption is due to the partitioning of DNAPL into the solid organic carbon present in the geological medium [Feenstra, 1994]. The affinity of an organic chemical for sorption on geological media is usually expressed by the organic carbon-water partition coefficient, denoted $K_{OC}$. The $K_{OC}$ value varies from one DNAPL to another. For aliphatic chlorinated solvents, it is comparable to the $K_{OC}$ value of benzene or toluene (see Figure 2.5). It is highest for polycyclic aromatics because of their affinity for organic carbon. The higher the $K_{OC}$ value, the greater the degree of sorption and the less mobile the contaminant in groundwater. Thus, as shown in Figure 2.5, mobility as a contaminant dissolved in groundwater is driven by sorption and solubility. The organic carbon-water partition coefficient can be correlated to $K_{OW}$, the octanol-water partition coefficient, which is commonly tabulated in the literature [Schwarzenbach et al., 1993; Hemond and Fechner, 1994].
Groundwater supplies in Europe and North America typically come from sand and gravel aquifers, and to some extent from fractured bedrock aquifers. Many large plumes of DNAPL contamination have been found to occur in these aquifers. These plumes can extend many kilometers from the industrial or waste disposal site where they originate, and very often result from the dissolution of DNAPL persistent sources located below the water table. This section describes a typical scenario for DNAPL subsurface contamination and gives an overview of the main factors driving groundwater contamination by DNAPLs.

**Figure 2.6.** Typical scenario of DNAPL accumulation and transport through the subsurface [after Levy et al., 1998].

Figure 2.6 presents a conceptual diagram for DNAPL behavior in the subsurface. A release of DNAPL has taken place at the top ground surface or right below this surface. Subsequently, the organic chemical has migrated vertically through the vadose zone under both the forces of gravity and soil capillarity (see definition in Section 2.2.1.3). Capillary forces are also responsible for the horizontal spreading of DNAPL in the vadose zone. However, for a typical silt or sand medium, the gravity forces are larger in magnitude than the capillary forces, and downward movement prevails depending upon the soil stratum. A point can be reached where DNAPL no longer holds together as a continuous phase, but rather is present as isolated blobs. These
blobs, which are held in the smaller pores where capillary forces can overcome a blob’s self-weight, are sometimes referred to as residual saturation [Huling and Weaver, 1991]. If sufficiently volatile, the DNAPL blobs may volatilize into the air pores of the vadose zone and migrate laterally as a gas phase. Thus, within a period of weeks to months, a DNAPL vapor plume can be formed by means of gas molecular diffusion [Feenstra et al., 1996].

In many instances, large quantities of DNAPL may be able to reach the water table before volatilizing, as illustrated in Figure 2.6. DNAPL blobs can also be subjected to leaching from the seasonal fluctuations of the water table.

The distribution of the DNAPL below the water table is affected by the geologic layering present [Feenstra et al., 1996]. A DNAPL moving downwards through a coarse-grained material would have to overcome a larger capillary resistance to migrate through a finer-grained layer. This capillary resistance is the entry pressure introduced in Section 2.2.1.3. Hence, low-permeability lenses of silt or clay material will act as capillary barriers to downward migration, and cause lateral spreading of the DNAPL. The DNAPL will accumulate on top the lens and spread laterally until it reaches the edge of the layer, or until the self-weight force due to DNAPL accumulation overcomes the capillary resistance of the lens. Thus, low-permeability lenses in the aquifer generate a branch-like DNAPL migration pattern as sketched in Figure 2.6. Such pattern has been investigated theoretically by de Neef and Molenaar [1997] and observed in laboratory simulations by Kueper et al. [1989].

The DNAPL mass offset due to horizontal spreading is not necessarily dictated by the direction of the groundwater flow, but rather by the pathways of least capillary resistance associated with heterogeneities of all kind occurring within the aquifer. This generates complex patterns of DNAPL distribution which are generally unpredictable in the saturated zone even if considerable information on the stratigraphy of the subsurface environment is available [Feenstra et al., 1996; Domenico and Schwartz, 1998].

Although low-permeability confining layers retard the vertical migration of DNAPLs, some laboratory studies have demonstrated that free-phase DNAPL is able to penetrate low-permeability clays, whether natural clay deposits or compacted clay liners [Barbee, 1994]. The chemical-clay interactions could cause shrinkage and cracking of clay and subsequent increases in its permeability. However, these findings remain controversial [Middleton and Cherry, 1996]. In particular, it is not clear whether or not these laboratory experiments are representative of real field conditions.

As DNAPL is flowing through the aquifer, small components of the trailing edge of the liquid phase will snap off, leaving blobs and ganglia of residual liquid. Residual DNAPL typically occupies from 1% to 25% of the pore space in unconsolidated geologic material [Mercer and Cohen, 1990; Kueper et al., 1993]. Residual DNAPL can be extremely difficult to mobilize by hydraulic means alone if interfacial tensions are high or if the subsurface medium is fine-grained.

Natural flow of ground water through the region contaminated by DNAPL gives rise to a dissolved-phase plume. Because of the relatively low solubility of the contaminants, and because of low natural gradient occurring in groundwater, it may take several decades to centuries before residual and pool zones of DNAPL are
depleted by natural dissolution [Kueper et al., 1993]. In a series of laboratory experiments in porous media, DNAPL concentrations approximately equal to aqueous solubility were measured at typical groundwater natural gradients [Schwille, 1988]. These results seem to contradict field measurements where DNAPL concentrations did not seem to exceed 10% of the aqueous solubility [Mackay et al., 1985]. The discrepancy has been attributed to diffusional limitations of dissolution in conjunction with heterogeneous field conditions, such as non-uniform groundwater flow and variable DNAPL distribution [Mercer and Cohen, 1990]. The mass rate of dissolution also tends to decline with time, suggesting that natural dissolution may take even longer than predicted [Feenstra et al., 1996].

In Figure 2.6, DNAPL, having migrated through the overburden, comes to rest above a fractured bedrock and forms a pool. Upon accumulation of DNAPL, the capillary resistance of the fracture, often called the fracture entry pressure, is exceeded and DNAPL starts to migrate through the fracture system. The pattern of DNAPL movement and ultimate distribution in fractured geologic media is controlled primarily by the orientation, spacing and connectivity of the fractures. Similar to porous media, migration takes place through pathways that offer the least capillary resistance, i.e. through the largest aperture fractures, and is uncoupled from the direction of the groundwater flow, whose magnitude tends to be small in this type of deposits [Kueper and McWhorter, 1996].

Once in a fracture network, DNAPL that remains under the form of a continuous phase can progressively infiltrate fractures of smaller aperture [Kueper and McWhorter, 1991; Kueper and McWhorter, 1996]. In this configuration, the equivalent DNAPL pool height at the DNAPL lower front is the sum of the pool height in the overburden overlying the fractures plus the height of DNAPL accumulated in fractures beneath the pool. Thus, larger entry pressures, corresponding to the capillary resistance of smaller fracture apertures, can be exceeded.

Visual inspection of bedrock core holes performed by Foster et al. [1996] at a waste disposal area (S-area site, Niagara Falls, N.Y.) showed evidence compatible with those predictions. When drilling was undertaken, DNAPL was found to travel more extensively in the horizontal direction as depth increased, suggesting that DNAPL distribution was described by an upside down Christmas tree pattern.

It is generally not possible to predict the maximum depth of DNAPL penetration at fractured sites even if detailed information is known about their geology. DNAPL is known to have penetrated to depths of several hundred meters. For some cases, it may even be greater than one thousand meters [Feenstra et al., 1996].

As in the case of porous media, once a DNAPL intrudes into and passes through a fracture network, it leaves behind a trail of residual DNAPL contamination. It is believed that residual contamination can be found (1) as blobs or ganglia held within fractures by capillary forces, (2) as small pools and puddles lying on horizontal fractures, or (3) as accumulations within dead-end joints [Clarke et al., 1994]. Again, these act as major sources for dissolved groundwater species. It may take decades to centuries before residual saturation is completely dissolved in the surrounding groundwater.
In some case scenarios, diffusion of contaminant mass from highly conductive fractures into the relatively immobile pore water of the rock matrix can take place [Slough et al., 1999a]. When such diffusion phenomena is important, deposits are referred to as fractured porous media. They include media such as un lithified clay-rich deposits and sedimentary rocks, which have a matrix storage capacity that exceeds the storage capacity of the adjacent fractures [Parker et al., 1996]. This diffusive mass transfer from the immiscible phase of the fractures to the dissolved and sorbed phases in the matrix blocks can result in complete disappearance of the immiscible phase after some time [Parker et al., 1997]. Thus, groundwater samples obtained from fractures may underestimate the extent of the contamination. Furthermore, remediation effectiveness in fractures is likely to be limited by reverse diffusion and desorption from the matrix [Parker et al., 1996].

![Diagram](image)

**Figure 2.7.** Height of DNAPL in a well: a. DNAPL in well corresponds to DNAPL in formation; b. DNAPL in well has entered from an upper elevation and does not correspond to DNAPL in formation at lower elevation; c. Borehole short-circuiting [after Kueper and McWhorter, 1996].

In Section 1.1.4, it has been pointed out that pools of free-product solvent are found only rarely in site investigation, even when their existence is not in doubt. In the event that DNAPL is observed in a borehole of a fractured medium, it can be difficult to infer exactly what the conditions are in the surrounding formation, as illustrated in Figure 2.7.a and Figure 2.7.b. Moreover, the site investigation can actually worsen the level of contamination by short-circuiting an existing barrier layer or two sets of disconnected fractures (see Figure 2.7.c) [Mercer and Cohen, 1990; Kueper and McWhorter, 1996].
Considerations of contamination of fractured media by DNAPLs are not limited to natural fractured bedrock underlying an overburden. Other scenarios may involve fractured clay natural deposits or liners as illustrated in Figure 2.8 and Figure 2.9. Figure 2.8 shows a clay-lined disposal pond containing DNAPL and water. The base of the clay liner contains a set of fractures resulting from poor construction or damage made by the free-phase organic solvent. DNAPL is able to migrate to the aquifer underlying the waste disposal pond.

In the scenario sketched in Figure 2.9, a fractured clay aquitard separates two aquifers. DNAPL is initially present in the upper aquifer but the entry pressure of the clay fractures is large enough that DNAPL is prevented from infiltrating into the lower aquifer.
fractures and subsequently contaminating the lower aquifer. An enhanced pump-and-treat remediation system designed to clean up the upper aquifer makes use of surfactants that lower the interfacial tension between DNAPL and water to remobilize blobs of DNAPL held by capillary forces. While this scheme benefits the upper aquifer, lowering interfacial tension also results in decreasing the entry pressure of the clay fractures. Hence, remobilized DNAPL is able to migrate to and infiltrate the lower aquifer.

2.2.3 Previous Related Work on DNAPL and Two-Phase Flow Transport in Fractured Media

2.2.3.1 Numerical Modeling

Modeling multiphase flow through fractured systems on the field scale requires estimates of average properties describing the ability of the fractures to transmit each phase (the phase relative permeability), as well as the ability of the surrounding matrix blocks to imbibe the wetting phase fluid and emit the non-wetting phase fluid (the matrix/fracture transfer) [Hughes and Blunt, 2001]. The complexity of modeling numerically the behavior of multiphase flow in fractured media has led a great deal of research and development in recent years. Kueper and McWhorter [1991] developed a two-phase model simulating DNAPL flow through orthogonal fracture systems. The model included neither DNAPL dissolution or aqueous phase transport, nor sorption onto the matrix. Conservation of mass and generalized Darcy’s law were written for each phase. Saturation constraints, capillary pressure definitions (see (2.1)) and constitutive relationships, i.e. capillary pressure-saturation and relative permeability-saturation relationships, were used to solve the problem entirely. The model was similar to that used to solve two-phase flow transport in porous media [for complete system of equations, see, for example, Kueper and Frind, 1991; de Neef and Molenaar, 1997; Dekker and Abriola, 2000].

Slough et al. [1999b] introduced CompFlow, a model that solved the coupled, non-linear compositional equations describing multiphase, multi-component flow and transport in discretely fractured rock, together with component partitioning between phases and interaction with the rock matrix. The model intended to investigate the transport of both the DNAPL pure phase and dissolved plume, while incorporating the effects of matrix diffusion, which was predicted to considerably affect DNAPL flow in the fractures [Parker et al., 1997; Slough et al., 1999a].

Reynolds and Kueper [2001] developed a multiphase flow simulator called QUMPFS (Queen’s University Multi-Phase Flow Simulator), similar to that of Slough et al. [1999b], to investigate the rate of DNAPL migration into fractured clay interbedded with sand lenses. The model incorporated advection, dispersive and diffusive
fluxes, equilibrium and non-equilibrium phase partitioning, and capillary hysteresis [Reynolds and Kueper, 2001].

The major issue associated with all of the models cited above is the need for constitutive relationships, which have yet to be proven reasonable when it comes to fracture flow [Keller et al., 2000]. For example, constitutive relationships developed for porous media [van Genuchten, 1980; Corey, 1986] are ill-suited for studying the flow in a single fracture, since it is impossible to define a capillary pressure-saturation relationship for various locations in a fracture based on its local aperture [Keller et al., 2000]. Constitutive relationships describing two-phase flow in a fracture network may be usable, but are almost impossible to define for field scenarios.

A second numerical approach consists in generating the capillary pressure-saturation and relative permeability-saturation relationships in a single fracture of variable aperture by considering fluid displacement in a fracture as a succession of capillary equilibrium states. This approach is more suitable for predicting the flow in a single fracture of an impermeable matrix [Keller et al., 2000] and is commonly referred to as percolation process [Hughes and Blunt, 2001]. For this type of modeling, viscosity effects are typically ignored. Using this approach, Pruess and Tsang [1990] simulated the percolation process on a fracture of lognormal aperture distribution. The researchers found that the relative permeability-saturation relationship was non-linear, and that interferences between fluid phases flowing in a fracture were important. Similar percolation approaches have recently been used by Keller et al. [2000], Zhou [2001], and Hughes and Blunt [2001] to simulate multiphase flow in a single fracture.

2.2.3.2 Laboratory and Field Testing

Many laboratory tests and some field experiments have been conducted to study the transport of DNAPLs in porous media [e.g., Kueper et al., 1993; Held and Illangasekare, 1995; Dawson and Roberts, 1997]. The number of studies of multiphase flow in fractures and fractured porous media is, on the other hand, relatively small, but has been increasing in recent years. It is acknowledged that, although fractured rocks consist of complex fracture networks, the study of multiphase flow in a single fracture is a prerequisite before reasonable conclusions can be drawn about flow in multiple fractures [Longino and Kueper, 1999b; Zhou, 2001]. Experiments involving DNAPLs are partly complicated by the fact that chlorinated solvents react with experimental material (see Section 5.4.1), such that transparent epoxy replicas of fractures [Gentier et al., 1989; Persoff and Pruess, 1995; Kneafsey and Pruess, 1998] may not be suitable for investigating DNAPL flow.

Schwille [1988] performed some of the first experimental work on tetrachloroethylene (PCE) transport in fractured media. Using vertical fracture models consisting of parallel glass plates of 0.1 mm and 0.2 mm aperture, the author observed very low retention capacities compared to porous media in both hydraulically smooth plates and plates roughened by sandblasting. Typical PCE infiltration patterns obtained
by Schwille are reproduced in Figure 2.10. More recent studies have shown the DNAPL retention capacity of a single fracture to vary between 3% and 27% of the fracture volume depending on its orientation [Longino and Kueper, 1999b]. This compares to a range of 1% to 50% reported for porous media [Mercer and Cohen, 1990; Kueper et al., 1993]. The difference is attributed to the topology of a rough-walled fracture, which is believed not to be as irregular as that in a porous medium. Hence, disconnected blobs and ganglia may not be as abundant in the former [Kueper and McWhorter, 1996].

Figure 2.10. Investigation of PCE infiltration into 0.2 mm aperture vertical fractures simulated by parallel glass plates [after Schwille, 1988]: a. Fracture simulated by two smooth glass plates; b. Fracture simulated by two glass plates roughen by sandblasting.

Natural fractures encountered in soils and rocks rarely resemble smooth parallel plates of constant aperture. Instead, their aperture field fluctuates, and the opposite faces of their surfaces may contact across extensive areas [Gentier et al., 1989]. Experimental studies have shown that two-phase fluid transport occurring in rough-walled fractures exhibit capillary behavior not unsimilar to that of porous media [Fourar et al., 1993; Reitsma and Kueper, 1994; Persoff and Pruess, 1995; Amundsen et al., 1999] and confirm prediction based on theoretical approach [Pruess and Tsang, 1990] (see Section 2.2.3.1).

Fourar et al. [1993] and Fourar and Bories [1995] conducted air-water flow experiments in horizontal fractures simulated by smooth, parallel glass plates as well as roughen glass plates where glass beads had been glued to the plate surfaces. The experimental setup used by the authors is illustrated in Figure 2.11.a. Simultaneous injection of water and air at relatively high flow velocities resulted in observed flow regimes similar to those observed in pipe flow [Wallis, 1969] (see Figure 2.11.b).
Fourar and Bories [1995] fitted the generalized Darcy’s law and obtained a linear relationship between the liquid phase relative permeability and liquid saturation. It should be noted, however, that the generalized Darcy’s law could not have been expected to be valid at the range of Reynolds number under investigation. Most of the confusion came from the assumption that Darcy’s law was valid so long as the flow was laminar. This is not true of course, since flow can be both laminar and controlled by inertia forces at intermediate Reynolds numbers [Munson et al., 1994; Longino and Kueper, 1999b]. Furthermore, Darcy’s law is only applicable if the fluid phases are continuous. Hence, it cannot be applied to bubble flow, one of the flow pattern observed by the authors (see Figure 2.11.b). Limits associated with Darcy’s law have been reviewed by Gamliel [2000].

Reitsma and Kueper [1994] measured the capillary pressure-saturation relationship between oil (LNAPL) and water in a horizontal fracture induced in a dolomitic limestone. The experimental setup used by the authors is illustrated in Figure 2.12.a. The fracture was initially saturated with water (wetting phase). One edge of the fracture was connected through a porous barrier to a burette used to measure the volume of wetting phase drained by the non-wetting phase following infiltration into the fracture (see Figure 2.12.a). The burette location was adjustable so as to control the pressure of the wetting phase and, consequently, the capillary pressure. The set of capillary pressure-saturation relationships that Reitsma and Kueper obtained was well

Figure 2.11. Investigation of air-water flow in a horizontal fracture simulated by parallel glass plates [after Fourar et al., 1993; Fourar and Bories, 1995]: a. Schematic illustration of experimental setup; b. Flow structures observed in a smooth horizontal simulated fracture as a function of the velocities of the fluids.
represented by a porous media capillary pressure function. An example of capillary pressure-saturation curve measured by the authors is shown in Figure 2.12.b.

Figure 2.12. Measurement of the capillary pressure-saturation relationship between oil (LNAPL) and water in a horizontal fracture [after Reitsma and Kueper, 1994]: a. Schematic illustration of experimental setup (the fracture is hidden by the solid plate); b. Capillary pressure-saturation curve obtained during one of the tests (the capillary pressure is expressed as an equivalent height of water).

Persoff and Pruess [1995] used transparent replicas of a natural rock fracture to observe gas-liquid flow patterns. Relative permeability-saturation relationships were
found to be non-linear, countering experimentally the conventional view that the fracture relative permeability of each phase can be approximated by its saturation.

Amundsen et al. [1999] also investigated gas-water flow in horizontal rough-walled, transparent fractures. The aperture field was measured using light absorption techniques. Displacement of the wetting fluid (water) by the non-wetting fluid (air) showed patterns (invasion bursts, fragmentation) that were in good qualitative and quantitative agreement with their percolation model simulation.

Other experimental work has specifically focused on DNAPL flow in vertical fractures so as to better understand the role played by gravity forces with respect to capillary forces. Kueper et al. [1995] and Chown et al. [1997] measured the upward water gradient required to arrest downward migration of DNAPL into a rough-walled, vertical fracture, as per the scenario shown in Figure 2.9. They measured arresting gradients across the fracture varying from 0.3 to 0.9 with strong hysteresis depending on whether or not the fracture was in drainage or imbibition conditions. In imbibition conditions, the measured gradients were found to be equal or less than those predicted by theory.

It is important to point out that, for their experiment, Kueper et al. [1995] and Chown et al. [1997] used fractures of approximately 200 mm in height, for which the applied gradient was dependent on the fracture entry pressure and local capillary forces. However, for a longer fracture, maybe more typical of field conditions, the arresting gradient is not affected by capillarity. The gradient can be estimated simply by $\Delta \rho / \rho_w$ where $\rho_w$ is the density of water and $\Delta \rho$ is the density contrast between DNAPL and water [Chown et al., 1997]. Typically, for DNAPLs, $\Delta \rho / \rho_w$ will be of the order of 0.1 to 0.6, which is an impractical gradient for field conditions.

Longino and Kueper [1996; 1999b; 1999a] investigated the retention capacity of PCE in a fracture of varying orientation. A significant reduction in residual saturation was observed when changing the orientation of the fracture plane from horizontal to vertical (see Figure 2.13.a). The retained residual was then flushed through water flooding, thereby leading to further reduction in PCE residual saturation. The ratio of residual saturation after water flooding to initial residual saturation was found to be well correlated with a combined capillary and Bond number consistent with that defined by Dawson and Roberts [1997] for porous media (see Figure 2.13.b). One of the challenges faced by Longino and Kueper [1996; 1999b; 1999a] was to properly estimate the residual saturation of PCE, because the fracture volume was not known and had to be estimated as the product of the hydraulic aperture by the projected fracture area.

Stephens et al. [1998] examined the infiltration of 1,1,1-trichloroethane (TCA) into a rough-walled, vertical fracture made out of a fractured masonry brick sandwiched between two sand layers. These authors did not observe DNAPL pooling in the sand layer above the fracture prior to its entry into the fracture, in contrast with the prediction of Kueper and McWhorter [1991] and Kueper et al. [1992] that DNAPL will accumulate on top of the fracture until its entry pressure is exceeded. Their experimental fracture, however, was not in hydrostatic conditions since the portion of sand immediately below the fracture was unsaturated.
Figure 2.13. Investigation of retention capacity of PCE in a fracture [after Longino and Kueper, 1999b]: a. PCE residual saturation $S^{*}_{NWr}$ as a function of the fracture plane orientation (PV refers to the fracture pore volume); b. Ratio of residual saturation after water flooding, $S_{NWr}$, to initial residual saturation of horizontal fracture, $S^{*}_{NWr}$, versus a combination of the capillary number, $N_C$, Bond number, $N_B$, and wetting phase relative permeability, $k_{rW}$.

Recent work by Jørgensen et al. [1998] has examined trichloethylene (TCE) flow through undisturbed columns of naturally fractured and bioporous clayey till (see experimental setup in Figure 2.14). The size of the specimens was 0.5 m in diameter and 0.5 m in height. Transport of TCE appeared to be confined to biopores and fractures, bypassing the low-permeability clayey matrix. The actual pathways within the undisturbed fractures could not be assessed given the likelihood of redistribution of TCE during dismantling of the column prior to visual inspection. TCE flow was quite high with a hydraulic gradient of 0.91 for one test and 1.18 for another.

O’Hara et al. [2000] performed similar work on a sample of fractured glaciolacustrine clay. The sample had the same dimensions as that used by Jørgensen et al. [1998] (see Figure 2.14). Visual inspection of the sample fractures after TCE flow revealed some matrix diffusion haloes only at a limited number of locations, indicating that TCE was flowing through preferential fracture pathways. The equivalent hydraulic aperture of the fractures where flow had taken place was found to be equal to 8 µm-11 µm, which was greater than the mean hydraulic aperture equal to 5 µm-6 µm, but less than the aperture of 17 µm back-calculated from the entry pressure. This difference was attributed to the fact that the entry pressure corresponds to the region where the fracture aperture is the largest. However, perfect wetting of water with respect to DNAPL ($\theta = 0^\circ$) was assumed for their computations where only partial wetting may have applied, which could also explain why the calculated aperture was larger than the measured apertures.

Experimental and numerical investigation into DNAPL transport in fractured media is currently extended to more complex systems. Theodoropoulou et al. [2001] have attempted to develop a non-Darcian model for fluids that are non-Newtonian (crude oil, asphalt, creosote, etc…) through experimental simulation in artificial
etched-glass fractures. Experimental work by Geller et al. [2000] has examined the potential for biodegradation of NAPLs in fractured rock in the vadose zone.

Figure 2.14. Flexible wall permeameter setup for investigating TCE flow through large undisturbed columns of fractured clay [after Jørgensen et al., 1998; O'Hara et al., 2000].

2.2.3.3 Conclusions

The development of models predicting DNAPL behavior and remediation in complex fracture network remains a challenge. The majority of efforts in these areas currently relies on acquiring accurate descriptions of individual fracture properties, such as mean aperture size distribution and fluid wettability, at a scale smaller than can typically be achieved in practice. Moreover, many of these models have yet to be validated in the field.

Laboratory experiments can provide the necessary input parameters as well as model validation. However, the challenge in these areas is to design an experiment that is representative of field conditions. The use of fracture samples in the laboratory is often accompanied by gradients, boundary conditions and Reynolds numbers that are different than those encountered in the field by several orders of magnitude. Therefore, these experiments may also yield mechanisms controlled by forces that would be irrelevant on the larger in situ scale. With respect to capillarity, it is often difficult to analyze wettability effects in real porous media due to the difficulty of controlling
surface states as well as complex geometries [Calvo et al., 1991]. This would be also true for fractures.

Experiments such as capillary tube flow experiments may provide detailed information on the interplay in a real fracture system between capillary, viscous, gravity and inertia forces, and thus constitute a good complement to real fracture experiments. Despite being considered overly simplistic and sometimes difficult to connect to real field conditions, capillary tube experiments have been used for decades in a large number of engineering disciplines, and have uncovered the basic mechanisms taking place at a simulated pore scale before being extended to more complex systems. Since this was the approach used in this research, previous work on capillary flow kinetics is examined in the next section with a specific emphasis on spontaneous capillary infiltration.

2.3 Kinetics of Flow in Capillary Tubes: Background and Previous Research Work

2.3.1 Introduction

The rise of liquids in capillary tubes has been observed in countless natural and physiological processes. The number of fields of study and application where this phenomenon is of interest is extremely large. It includes the paper and ink industry, the fiber industry, the ceramic industry, food engineering, chemical engineering, powder engineering, nuclear engineering, textile engineering, oil and gas recovery, agriculture, medicine, the pharmaceutical industry, civil engineering and environmental remediation. More generally, rise of liquids in capillary tubes is a topic of interest for any discipline dealing with porous media or materials.

This section focuses on the dynamic aspects of capillary infiltration. The dynamics of capillary infiltration, also called the kinetics of infiltration, aims at computing the rate of rise of infiltration of a liquid into a capillary tube and thus gaining knowledge of the liquid interface displacement as a function of time.

There have been a large number of publications related to capillary infiltration. The first paper published in a scientific journal presenting a complete equation of rate of infiltration in capillary tubes is attributable to Lucas [1918]. Perhaps because this paper was written in German, it is traditionally believed that Washburn [1921a; 1921b] was the first person to develop such a model. Hence, the differential equation linking the rate of capillary rise to the total rise in a capillary tube often carries Washburn’s name [Dullien, 1992; Adamson and Gast, 1997], although in some cases the equation is referred to as the Lucas-Washburn equation. This equation is the object of Section 2.3.2.

At the beginning of the twentieth century, the capillary rise problem was far from new. The recorded scientific observation of capillary rise seems to date as far back as
Leonardo da Vinci [Bikerman, 1958]. The inverse proportionality law between the radius of a circular capillary tube and the height of capillary rise was known already to H. Fabry (1676) and J. Jurin (1718) [Bikerman, 1958]. Furthermore, the capillary effect was explained by Laplace (1805) [Quéré, 1997].

Towards the end of the nineteenth century, a French physicist, Decharme [1872; 1873b; 1874b], performed a series of experimental investigations on the infiltration of various liquids into capillary tubes and obtained an extensive set of data. Decharme extended his experimental work to the rise of liquids into porous media such as paper strips, textiles and porous solids [Decharme, 1873a; 1874c; 1874a]. While Decharme attempted to develop an equation that could predict the kinetics of infiltration [Decharme, 1874a], his treatment of viscosity and surface tension was somewhat incorrect. Although Decharme’s equation was in good agreement with his experiments, it required the estimation of three empirical constants, which depended on the type of liquid and geometry of the system.

The mathematical approach to capillary rise kinetics can be traced back to the eighteenth century. On September 24, 1748, the mathematics and physics professor Georg Wolfgang Krafft writes to Leonhard Euler: The mechanical rule \( dc = p \, dt/m \) or \( c \, dc = p \, ds/m \) assumes that the mass \( m \) is constant during the entire motion; would it not be possible to create such a rule in which the mass, or rather the moving point, could be variable?... I think that it would then become possible to derive the measure of the rise of fluids into capillary tubes, for in this case the mass that rises increases all the time. Please, Sir, let me know your thoughts regarding this matter at some suitable opportunity [Kornev and Neimark, 2001]. While history has lost Euler’s reply to this letter, this is certainly a great example of the articulation of a problem associated with the motion of a system whose mass varies with time.

It is a common belief that the old problem of kinetics of capillary rise has been solved a long time ago. There has nevertheless been a large number of publications on the topic since Lucas [1918] and Washburn [1921a], including several papers in recent years. That the problem has not been entirely solved can be attributed to several factors, one of them precisely connected to that which Krafft pointed to in his letter. While it is of course perfectly possible to derive an equation for a system of variable mass, it is not clear what the velocity of the mass added to this system should be. More than two hundred and fifty years later, a complete solution to the rate of capillary rise problem remains to be found.

### 2.3.2 Lucas-Washburn Equation

The simplest equation describing the infiltration of a liquid into a capillary tube is described by Lucas [1918] and Washburn [1921a; 1921b], although forms of the equation were obtained by Bell and Cameron [1906] and West [1912]. Lucas and Washburn both started from the Hagen-Poiseuille’s law, which relates the steady-state flow of a liquid through a capillary tube, as [Munson et al., 1994]
where $Q$ is the volumetric flow of liquid, $r_0$ and $l$ are, respectively, the radius and the length of the capillary tube, $\mu$ is the dynamic viscosity of the liquid, and $\Delta p$ is the total driving pressure acting to force the liquid along the capillary. Based on (2.3), both Lucas and Washburn obtained

$$\frac{dz_c}{dt} = \frac{r_0^2}{8\mu} \frac{\Delta p(z_c)}{z_c},$$  \hspace{1cm} (2.4)$$

where $z_c$ is the penetrated length of liquid displacing air inside a capillary tube at time $t$, as illustrated in Figure 2.15. Recalling that the velocity profile of Hagen-Poiseuille’s flow is parabolic \cite{Munson1994}, $dz_c/\,dt$ corresponds to the average velocity across a section of the capillary tube. It is assumed that the viscosity of the displaced air (or gas phase) is negligible with respect to the viscosity of the liquid.

**Figure 2.15.** Schematic of infiltration into a capillary tube of radius $r_0$ with inclination $\phi$ to the horizontal.

Assuming that the liquid is wetting with respect to air, the total driving pressure is the sum of the capillary pressure and the hydrostatic pressure, as

$$\Delta p(z_c) = \rho g h - \rho g z_c \sin \phi + \frac{2\sigma \cos \theta}{r_0},$$  \hspace{1cm} (2.5)$$
where $g$ is the Earth’s gravity, $\rho$ and $\sigma$ are respectively the density and the surface tension of the liquid, $\theta$ is the contact angle between the liquid and air, $h$ is the existing reservoir head, assumed constant with time, and $\phi$ is the inclination angle of the capillary tube to the horizontal, as illustrated in Figure 2.15. It is assumed that both the reservoir and the capillary tube are open to the air so that the atmospheric pressure can be ignored in (2.5).

Thus, on the basis of experimental measurements, if the capillary radius $r_0$ is known, (2.4) can be used for the measurement, or estimation, of the surface tension or the contact angle between the liquid and the porous medium. Equation (2.4) can also be used to study the capillary infiltration into porous solids like powder beds in order to characterize their wettability [e.g., Washburn, 1921b; Peek and McLean, 1934; Levine et al., 1977; van Brakel and Heertjes, 1977; Siebold et al., 1997].

In the case where the capillary tube is horizontal, $\phi$ is equal to 0 and $\Delta p(z_c)$ is independent of $z_c$. Washburn [1921a] solved (2.4) for these conditions and obtained

$$z_c = \sqrt{\frac{r_0^2 t}{4 \mu}} \left( \frac{\rho g h + \frac{2 \sigma \cos \theta}{r_0}}{r_0} \right), \quad (2.6)$$

which shows that the infiltrated length of a horizontal capillary tube ($\phi = 0$) is proportional to the square root of time. This property was shown theoretically and verified experimentally with water, alcohol and benzene by Bell and Cameron [1906] for 0.5 mm radius capillary tubes. However, it should be noted from (2.6) that the square-root dependence does not result from the fact that the flow is driven by the interfacial tension difference. Rather, it is typical of the laminar viscous flow of a Newtonian liquid in a tube under the action of a constant pressure difference, where the length of the liquid is changing with time [Marmur, 1992].

In the case where the capillary tube is vertical and located above the water reservoir, $\phi$ is equal to $+90^\circ$ and $\Delta p(z_c)$ now depends on $z_c$. Both Lucas and Washburn solved (2.4) for these conditions and obtained

$$z_c + (h + h_e) \ln \left( 1 - \frac{z_c}{h + h_e} \right) = -\frac{r_0^2 \rho g}{8 \mu} t. \quad (2.7)$$

where $h_e$ is the well-known equilibrium height at $t \to \infty$, given by

$$h_e = \frac{2 \sigma \cos \theta}{\rho g r_0^2}. \quad (2.8)$$

When hydrostatic forces are negligible with respect to capillary forces, for example at the early stages of capillary rise, (2.6) and (2.7) both reduce to
Equations (2.6), (2.7) and (2.9) assume, of course, that the contact angle $\theta$ is independent of the velocity of infiltration, $dz_c/dt$. The validity of this assumption is discussed in Section 2.3.5.

Equations (2.6) and (2.9) have been shown to be well suited to describe infiltration into long horizontal capillary tubes of small radius. Washburn [1921a] obtained very good agreement for pre-wetted 0.95 m long capillary tubes of radius 0.15 mm and 0.37 mm. Rideal [1922] obtained similar results for a number of solvents using capillary tubes of length 1.2 m and radius 0.35 mm. Malik et al. [1979] independently confirmed (2.9) with water and alcohol solutions flowing into capillary tubes of radius 0.2 mm, 0.3 mm and 0.4 mm. Fisher and Lark [1979] confirmed the validity of (2.9) for water and cyclohexane flowing into horizontal capillary tubes of very small radius ranging from 0.2 $\mu$m to 15 $\mu$m. For water infiltrating capillary tubes of radius smaller than 0.3 $\mu$m, however, the authors noticed the presence of bubbles within the liquid, as well as flow rates smaller than expected. Peek and McLean [1934] obtained good agreement between data and prediction for water flowing into a pre-wetted horizontal capillary tube of radius 0.36 mm. However, penetration into an initially dry tube could only be predicted correctly if a value of surface tension for
water equal to 0.0385 N/m was taken, versus 0.0702 N/m for the pre-wetted case. In their derivation, Peek and McLean had ignored the effects of pre-wetting on the contact angle (see discussion of Section 2.3.5.6).

A linear relationship between $t$ and $z_c^2$ is also commonly observed at the intermediate stage of the imbibition into packed beds [Washburn, 1921b; Siebold et al., 1997], which may presumably be treated as a bundle of capillaries having a so-called effective radius, $r_0$. However, as pointed out by Zhmud et al. [2000], a $t$ proportional to $z_c^2$ is characteristic of diffusion-related processes, and not necessarily related to the validity of the Lucas-Washburn equation.

![Figure 2.17](image)

**Figure 2.17.** Rise of pentane into a 0.191 mm radius vertical capillary tube [after Siebold et al., 2000]. (1) Experimental points; (2) Predictions from Lucas-Washburn model (2.7) (with $h = 0$); (3) Predictions from (2.9) (with $\theta = 0$).

There exist examples of the successful application of (2.7) in vertical capillary tubes. Washburn [1921a] obtained reasonable agreement between (2.7) and an experiment of capillary rise into a 0.15 mm radius capillary tube. Equation (2.7) is also in very good agreement with data on rise of various solvents into fine vertical capillary tubes [Ligenza and Bernstein, 1951], where the radii were of the order of 20-40 µm, i.e. about ten times smaller than typical radii used for most experimental investigations. An example of this agreement is shown in Figure 2.16. Peek and McLean [1934] also obtained good agreement between data and predictions given by (2.7) for water flowing into a capillary tube of radius 0.25 mm with $\varphi = 9^\circ 22'$.

Despite the reported successes, high resolution observations in capillary tubes most often conclude that the Lucas-Washburn equation fails to describe the initial stage
of the wetting liquid infiltration. Early stages of rise have been observed to follow a linear law in time [Quéré, 1997]. At short-contact times, predicted velocities of infiltration are always much larger than those measured experimentally [LeGrand and Rense, 1945; Calderwood and Mardles, 1955; Jeje, 1979; Siebold et al., 2000]. An example is shown in Figure 2.17, where the rise is found to be slower than predicted by the Lucas-Washburn model. While some predicted velocities assume perfect wetting, i.e. \( \theta = 0 \), it is not obvious that an assumption of partial wetting would be sufficient to explain the discrepancies.

A straightforward explanation is that (2.4) yields an infinite initial infiltration velocity \( \frac{dz_c}{dt} \) for the initial condition \( z_c = 0 \) at \( t = 0 \). Aware of high initial penetration velocities, Washburn [1921a] argued that (2.4) held true whenever Hagen-Poiseuille’s law was applicable, i.e. once the initial period of turbulence had ceased. This corresponds to a Reynolds number, as defined by

\[
Re = \frac{2r_0 \rho \frac{dz_c}{dt}}{\mu}
\]

(2.10)

of less than 2000. Washburn concluded that the capillary tube length scale associated with turbulent effects was of the order of magnitude of millimeters or less for typical capillary tubes. Unfortunately this analysis failed to take inertia effects into consideration. The inertia effects are the forces associated with the change in momentum of the liquid. Inertia forces are still large in the laminar regime for Reynolds numbers of the order of 1 to 2000 [Munson et al., 1994]. Hence, the length scale of the region for which (2.4) is not valid is certainly larger than that calculated by Washburn. The use of Hagen-Poiseuille’s law to derive the Lucas-Washburn equation makes the implicit assumption that the flow is quasi-static. Therefore, (2.4) is inappropriate for conditions where rapid changes in infiltration length take place over time, such as those conditions at the early stages of the meniscus rise into a capillary tube.

2.3.3 Infiltration Model Incorporating Local Inertia Forces

2.3.3.1 Motion Equation Using Momentum Balance on Column of Liquid

Recognizing the limits of Lucas-Washburn equation, Rideal [1922], Bosanquet [1923], Pickett [1944] in his critique of Rense [1944], Levine and Neale [1974] and Quéré [1997] used the momentum conservation equation to include inertia forces in their derivation of kinetic models for liquid infiltration into capillary tubes.

In their model, Bosanquet, Pickett and Quéré considered the momentum difference between \( t \) and \( t + \Delta t \), corresponding to a liquid advance from \( z_c \) to \( z_c + \Delta z_c \) in
an initially air-filled capillary tube of radius \( r_0 \). Using this approach, they obtained the following expression

\[
\rho \frac{d}{dt}\left( z_c \frac{dz_c}{dt} \right) = \Delta p(z_c) - \frac{8\mu}{r_0^2} z_c \frac{dz_c}{dt}.
\]  
(2.11)

Rideal, as well as Levine and Neale, used a similar approach but obtained

\[
\rho z_c \frac{d^2 z_c}{dt^2} = \Delta p(z_c) - \frac{8\mu}{r_0^2} z_c \frac{dz_c}{dt},
\]  
(2.12)

which differs from (2.11) by a term \( \rho \left( \frac{dz_c}{dt} \right)^2 \). The solution to (2.12) is more complicated than the solution to (2.11) as it is expressed in the form of an infinite series [Rideal, 1922; Levine and Neale, 1974].

In fact, the Bosanquet model, given by (2.11), does not consider the same system between time \( t \) and time \( t + \Delta t \). This is because it does not account for the momentum of an element of length \( \Delta z_c \) located in the liquid reservoir feeding the capillary tube at time \( t \). In other words, the derivation implicitly assumes that this element has no momentum. If one now assumes that the element of liquid entering the capillary tube has the same velocity as the rest of the infiltrated liquid, i.e. \( \frac{dz_c}{dt} \), the expression given by (2.12) is obtained. This problem is similar to the classical mechanics problem of a variable mass system, like a rocket losing mass by burning fuel [see, for example, Benson, 1996]. Whether or not the element entering the capillary tube should have a velocity illustrates the difficulties of determining the entry flow conditions. This issue has been discussed in depth by Levine et al. [1976].

Under conditions where the total driving pressure \( \Delta p(z_c) \) can be assumed independent of \( z_c \)—for example, when the capillary tube is horizontal—and also assuming an initial condition \( z_c(\frac{dz_c}{dt}) = 0 \) at \( t = 0 \), (2.11) can be solved to give

\[
z_c = \sqrt{\frac{r_0^2 \Delta p}{4\mu}} \left[ t - \tau \left( 1 - e^{-\frac{t}{\tau}} \right) \right],
\]  
(2.13)

where \( \tau \) is the time scale for which inertia forces are significant and is given by

\[
\tau = \frac{\rho r_0^2}{8\mu}.
\]  
(2.14)

Note that for \( t >> \tau \), (2.13) is reduced to (2.6). Considering that the capillary tube is horizontal and assuming the reservoir head \( h \) is equal to zero, the time scale \( \tau \) corresponds to a length scale \( \zeta \) given by (2.6) as
Examining, for example, the infiltration of water in a 1 mm radius horizontal capillary tube initially filled with air, using the physical properties of water at 20°C as $\mu = 1.002 \times 10^{-3}$ Pa.s, $\sigma = 7.28 \times 10^{-2}$ N/m, $\rho = 998.2$ kg/m$^3$ [Munson et al., 1994], and assuming perfect wetting, (2.14) and (2.15) yield respectively $\tau \approx 0.125$ s and $\zeta \approx 67.3$ mm. For a 0.1 mm radius horizontal capillary tube, $\tau \approx 1.25$ ms and $\zeta \approx 2.13$ mm is obtained. Therefore, the effects of inertia appear to be significant over a length of ten to one hundred times the capillary tube radius.

Considering the case where the capillary tubes are vertical and taking $g = 9.807$ m/s$^2$ [Munson et al., 1994], (2.7) can be solved numerically to give $\zeta \approx 14.9$ mm (equal to $h_e$ as given by (2.8)) for the 1 mm radius capillary tube, and $\zeta \approx 2.12$ mm (much less than $h_e = 148.7$ mm) for the 0.1 mm radius capillary tubes. Therefore, for the 1 mm radius tube, inertia effects are important over the entire range of capillary rise, whereas for the 0.1 mm radius tube, they are only important for less than 2% of $h_e$.

Neglecting viscosity and hydrostatic forces at the early stages of the capillary infiltration, (2.11) is reduced to

$$
\rho \frac{dz_c}{dt} \left( c \frac{dz_c}{dt} \right) = \frac{2\sigma \cos \theta}{r_0}.
$$

(2.16)

Taking the initial condition $z_c = 0$ and finite infiltration velocity $dz_c/dt$, (2.16) yields an equation linking $z_c$ and $t$ linearly, as

$$
z_c = ct,
$$

(2.17)

where $c$ is the penetration velocity given by

$$
c = \sqrt{\frac{2\sigma \cos \theta}{\rho r_0}}.
$$

(2.18)

The velocity $c$ is sometimes referred to as the Bosanquet velocity [Kornev and Neimark, 2001]. A balance between inertia and capillary forces, the Bosanquet velocity is behind several other capillary-related problems, like the bursting velocity of a soap bubble of thickness $r_0$ [Quéré, 1997].

It would have seemed natural to take $z_c = 0$ and $dz_c/dt = 0$ at $t = 0$ as the initial conditions for (2.11). However, there is a singularity at $t = 0$ when a finite force is applied to an infinitesimal mass. The remedy is to assume a finite initial velocity as given by (2.18). A drawback is an infinite acceleration at $t = 0$ [Zhmud et al., 2000], which was solved by Szekely et al. [1971], as will be show in Section 2.3.4.2.
It is interesting to compute the Reynolds number corresponding to the Bosanquet velocity. Equations (2.10) and (2.18) yield

\[
\text{Re}(c) = \sqrt{\frac{8r_0 \rho \sigma \cos \theta}{\mu^2}}.
\]  

(2.19)

Note that the Reynolds number associated with the Bosanquet velocity decreases with the radius of the capillary tube. Consider, for example, the infiltration of water into an air-filled 1 mm radius capillary tube, using the physical properties of water at 20°C given above and again assuming perfect wetting. Under these conditions, one obtains $\text{Re}(c) \approx 760$, meaning that the flow associated with the capillary infiltration is laminar, as it is below 2000.

Experimental observations of infiltration of liquids into capillary tubes have shown an initial behavior as predicted by (2.17), i.e. a length of infiltration that increases linearly with time at the very early stages of the liquid displacement [Quéré, 1997; Siebold et al., 2000] (see also Figure 2.17). However, the observed velocity of infiltration was found to be typically less than the predicted velocity, $c$ [Jeje, 1979; Quéré, 1997; Kornev and Neimark, 2001].

Quéré attributed the discrepancy between the observed and predicted velocities to (i) a difference between the dynamic contact angle and the static contact angle, and (ii) the boundary between the capillary tube and its reservoir which is responsible for a partial energy loss. Problem (i) rises from peculiarities of the flow at the contact line between the liquid/air interface and the capillary wall, while problem (ii) is due to the pressure distribution caused by the flow from the reservoir into the capillary.

Another issue has been pointed out by Levine and Neale [1974]. The left-hand side term of (2.12)—as well as the left-hand side term of (2.11)—implies that the liquid moves through the capillary tube as a solid plug, suggesting an apparent constant velocity profile across a capillary tube section. This contradicts the right-hand side term of these equations, which contains a term corresponding to the Hagen-Poiseuille’s viscous dragging force, which is based on a steady flow parabolic velocity profile across a section of the capillary tube.

Another issue disregarded by Levine and Neale [1974], Quéré [1997] and others, is that the equations presented here make the implicit assumption that the flow lines are parallel to the walls of the capillary tube and the main direction of flow. Under these conditions, inertia forces are limited to those related to the change of liquid velocity at a given location, also called local acceleration. At the entrance to the capillary tube, however, the flow lines may not be parallel to the walls, such that inertia may also result from convective acceleration, and further reduce the rate of capillary imbibition. Because the vast majority of researchers have written a balance of forces directly for the column of liquid in motion, instead of using the Navier-Stokes equations [e.g., Munson et al., 1994], they have failed to incorporate this term into account [e.g., Rideal, 1922; Bosanquet, 1923; Pickett, 1944; Siegel, 1961; Szekely et al., 1971; Quéré, 1997; Zhmud et al., 2000]. This issue is further explored in Section 3.4.2 and Section 3.5.2.4.
2.3.3.2 Motion Equation Using Navier-Stokes Equation

An attempt to remove the inherent limitations associated with the Hagen-Poiseulile’s friction approximation in unsteady flow was made by Letelier et al. [1979] (see also [Moshinskii, 1997]). In their derivations, Letelier et al. kept the assumption of flow lines parallel to the walls of the capillary tubes. In other words, while the radial component of velocity and convective inertia were assumed zero, the longitudinal velocity profile was not assumed to be parabolic. Starting from the longitudinal component of the Navier-Stokes equations, Letelier et al. wrote the longitudinal velocity in the form of a series expansion, and obtained the following after reduction

\[
\frac{4}{3} \rho z_c \frac{d^2 z_c}{dt^2} = \Delta p(z_c) - \frac{8\mu}{r_0^2} z_c \frac{dz_c}{dt} + \frac{1}{144} \frac{\rho r_0^2}{\mu} z \frac{d^3 z_c}{dt^3} + o\left(\frac{d^4 z_c}{dt^4}\right),
\]

(2.20)

where \(o(\frac{d^j z_c}{dt^j})\) is a function of all the \(j^{th}\)-order derivatives of \(z_c\) with \(j \geq 4\). If one neglects in (2.20) all of the terms corresponding to the derivative of \(z_c\) of an order larger or equal to two, the Lucas-Washburn equation is obtained, i.e. (2.4). Alternatively, if one neglects in (2.20) all of the terms corresponding to the derivative of \(z_c\) of an order larger or equal to three, (2.12) is obtained, i.e. the Rideal equation, where the coefficient of the second-order term has been multiplied by 4/3.

Basically, the smaller the time, the more important the terms of higher order. Inspection of (2.20) shows that the terms of second order and higher are negligible with respect to the first-order term when the time is much larger than the characteristic time \(\tau_1\) given by

\[
\tau_1 = \frac{3}{4} \frac{\rho r_0^2}{8\mu},
\]

(2.21)

which is similar to (2.14). Similarly, the terms of third order and higher are negligible with respect to the second-order term when the time is much larger than the characteristic time \(\tau_2\) given by

\[
\tau_2 = \frac{1}{144} \frac{3}{4} \frac{\rho r_0^2}{8\mu}.
\]

(2.22)

While Letelier et al. [1979] did not attempt to compare (2.20) to experimental data, their model was later evaluated by Batten [1984]. Batten used data obtained by LeGrand and Rense [1945] who investigated the rise of water and ethanol in capillary tubes of radius ranging from 0.242 mm to 0.350 mm. Batten did not find that (2.20) could predict the capillary rise more accurately than the Bosanquet equation, i.e. (2.11). However, it should be pointed out that Batten neglected the terms of third order and higher in his comparison. Nonetheless, his disagreement between prediction and
measurement goes beyond the characteristic time $\tau_2$. Thus, it does not appear that the hypothesis of parabolic flow alone may significantly affect prediction on capillary infiltration.

### 2.3.4 Infiltration Model Incorporating Interaction Between Capillary Tube and Reservoir

#### 2.3.4.1 Hagenbach Correction

A first attempt to incorporate end-effect drag forces at the boundary between the capillary tube and the reservoir of liquid feeding it was made by Brittin [1946]. Brittin added one term to the right-hand side of (2.11) corresponding to the friction loss due to the cross-sectional contraction at the inlet of the capillary tube. Physically, the friction loss can be attributed to eddies forming on the side of the tube entrance [see, for example, Brun et al., 1968]. Assuming that the cross-sectional area of the capillary tube is negligible compared to the cross-sectional area of the reservoir, the contraction drag force $f_{cd}$ can be expressed as

$$f_{cd} = \frac{e_v}{2} \frac{\rho \pi r_0^2}{\frac{d}{dt}} \left( \frac{dz_c}{dt} \right)^2,$$

(2.23)

where $e_v$ is a dimensionless coefficient equal to 0.5 [McAdams, 1942]. In examining what they called the vena contracta drag force, Szekely et al. [1971] used a similar approach and took a coefficient $e_v$ equal to 0.45 [Bird et al., 1960]. More generally, $e_v$ is determined by how the capillary tube is connected to the reservoir. The coefficient will be less if the connection is well-rounded or trumpet-shaped. Conversely, $e_v$ will be larger if the capillary tube is reentrant [Munson et al., 1994]. In any case, (2.11) can be rewritten as

$$\rho \frac{d}{dt} \left( \frac{dz_c}{dt} \right) = \Delta p(z_c) - \frac{8\mu}{r_0^2} z_c \frac{dz_c}{dt} - \frac{e_v}{2} \rho \left( \frac{dz_c}{dt} \right)^2.$$

(2.24)

Using (2.24), Brittin [1946] obtained good agreement with one set of data presented by Rense [1944].

The approach used by Brittin [1946] and Szekely et al. [1971] was later criticized by Levine et al. [1976] who pointed out that the drag force as given by (2.23) was only valid for turbulent flow. In the case of laminar flow, there is a larger pressure drop in the inlet region. Computation of such a pressure drop has been extensively described and reviewed in the literature [see, for example, Tietjens and Prandtl, 1957; Goldstein, 1965].
Another pressure drop must be considered corresponding to the conversion of pressure energy to kinetic energy at the inlet of the capillary tube. As a first approximation, assuming that the flow in the reservoir is inviscid and that the velocity $dzC/dt$ of liquid in the capillary tube is a constant, the pressure $p1(zc)$ at the inlet cross section can be expressed using Bernouilli’s equation, as [Tietjens and Prandtl, 1957]

$$p1(zc) = \rho gh - \frac{1}{2} \rho \left( \frac{dzC}{dt} \right)^2.$$  

(2.25)

Under conditions of viscous flow, it is well known that the velocity at the entry to a circular pipe will be practically constant over a cross-section [Munson et al., 1994]. However, the velocity at the wall is zero so that a thin boundary layer is found around the walls of the pipe. A certain distance downstream of the entrance, the boundary layer grows to ensure mass conservation until the velocity profile becomes parabolic. The length $le$ of the transition zone is typically of the order of 0.07 $r0Re$ [Bird et al., 1960]. Others [White, 1974] suggest $le = 0.16 r0Re + 1.3 r0$, where the second term, independent of the Reynolds number, makes the formula valid for creeping flow, i.e. at a Reynolds number below 1, as computed elsewhere [Lew and Fung, 1969; Lew and Fung, 1970].

Because the kinetic energy associated with a constant velocity profile is twice as small as the kinetic energy associated with a parabolic velocity profile, there is also a complementary pressure loss associated with creating the parabolic velocity distribution [Goldstein, 1965]. The drag force corresponding to the pressure loss is expressed in a form similar to (2.23). Computing the kinetic energy difference using constant and parabolic velocity profiles yields $e_v = 1$ [Goldstein, 1965]. Most often, (2.23) and the second term of the left-hand side of (2.25) are combined together to form a pressure drop term equal to $(1 + e_v)(\rho/2)(dzC/dt)^2$. The coefficient $1 + e_v$ is replaced by a single coefficient, $m$, which is sometimes called the Hagenbach correction [Oka, 1960].

A value of $m = 2$ yields a pressure drop equal to $\rho (dzC/dt)^2$, which is precisely the term that differentiates the Rideal model (2.12) from the Bosanquet model (2.11). Thus, it can be concluded that the Bosanquet approach already includes the term associated with pressure drop, whereas the Rideal approach does not. This observation is consistent with the fact that elements of liquid $\Delta zC$ enter the capillary tube with a velocity $dzC/dt$ for Rideal, and with a velocity of zero for Bosanquet (see discussion in Section 2.3.3.1).

More refined approaches to obtain values for the Hagenbach correction, $m$, have been investigated in several papers [e.g., see reviews by Langhaar, 1942; Tietjens and Prandtl, 1957; Goldstein, 1965]. Theories from Boussinesq, Schiller, Atkinson-Goldstein and Langhaar have suggested values equal to 2.24, 2.16, 2.41 and 2.28, respectively. Riemann and Schiller have obtained average experimental values equal to 2.248 and 2.32, respectively [Langhaar, 1942]. It should be noted that all these theories assume that the flow from the reservoir to the capillary tube takes place through a well-rounded entrance. Hagen has suggested a value for $m$ equal to 2.7,
which seems rather large compared to the values suggested above [Tietjens and Prandtl, 1957]. However, in Hagen’s case, the tube entrance was not well rounded but squarely cut off. This caused a contraction of the jet with subsequent spreading again, leading to a larger pressure drop [Tietjens and Prandtl, 1957].

Overall, the pressure drop across the contraction from the reservoir to the capillary tube can be attributed to the combination of three effects: (i) conversion of pressure energy to kinetic energy in the reservoir near the inlet of the capillary tube, (ii) eddy dissipation at the contraction and (iii) conversion of pressure energy to kinetic energy due to a change in velocity profile throughout the transition zone. The value of \( m \) for a well-rounded entrance can be expected to range between 2.24 and 2.41.

Taking the pressure drop terms given by (2.23) and (2.25) into account, (2.5) and (2.12) yield

\[
\rho z_c \frac{d^2 z_c}{dt^2} + \frac{8\mu}{r_0^2} z_c \frac{dz_c}{dt} = \rho gh + \frac{2\sigma \cos \theta}{r_0} - \rho g z_c \sin \varphi - \frac{m}{2} \rho \left( \frac{dz_c}{dt} \right)^2. \tag{2.26}
\]

Equation (2.26) was first proposed by Siegel [1961]. Taking \( m = 2 \) in (2.26), yields an equation which is not substantially different from (2.11), whose solution for the horizontal case, (2.13), and early-stage rise solution, (2.17), have been shown to overpredict the initial velocity of rise (see Section 2.3.3.1). Nevertheless, even a value of \( m = 2.7 \) would not significantly affect this trend. Indeed, Siegel [1961] had to take \( m = 5 \) before he could observe some agreement between prediction and measurement on a micro-gravity rise in a 0.95 mm radius capillary tube.

Zhmud et al. [2000] suggested that there could be a turbulence drag slowing down the infiltration velocity, and added a term to the right-hand side of (2.26) equal to

\[
\Phi(z_c) = \begin{cases} 
0 & \text{if } \frac{dz_c}{dt} \leq v_{cr} \\
- \frac{q}{r_0^2} z_c \left( \frac{dz_c}{dt} \right)^2 & \text{if } \frac{dz_c}{dt} > v_{cr}
\end{cases}, \tag{2.27}
\]

where \( v_{cr} \) is the critical velocity for which the turbulence begins and \( q \) is a turbulence coefficient taken to be equal to 0.3 kg/m². While Zhmud et al. [2000] obtained a very good fit for the capillary rise of dodecane in a 0.1 mm radius capillary tube, the authors noted that the critical velocity \( v_{cr} \) corresponded to a Reynolds number of the order of 2. Therefore, according to the authors, \( \Phi(z_c) \) was not exactly a turbulent drag but rather some second order dissipation correction related to the actual flow pattern. Clearly, this approach is empirical.
2.3.4.2 Reservoir Inertia

In presenting (2.25), the velocity \( \frac{dzc}{dt} \) of liquid in the capillary tube was assumed to be a constant, which of course is not the case. The velocity changes are responsible for reservoir inertia, whose magnitude must be examined. Siegel [1961] adapted the analysis of Morse and Feshbach [1953] for the flow through an orifice. The authors showed that, for a flow rate of \( \pi r_0^2 \frac{dzc}{dt} \) through an orifice, a plug of effective mass \( \rho \lambda \pi r_0^3 \), with \( \lambda = \pi/2 \), had to be accelerated when initiating flow. Considering the inertia term of (2.26), Siegel [1961] pointed out that this was equivalent to increasing the liquid height in the capillary from \( z_c \) to \( z_c + \lambda r_0 \). Hence, starting from (2.26), Siegel obtained

\[
\rho \left( z_c + \lambda r_0 \right) \frac{d^2 z_c}{dt^2} + \frac{8 \mu}{r_o^2} z_c \frac{dz_c}{dt} = \rho g h + \frac{2 \sigma \cos \theta}{r_o} - \rho g z \sin \phi - \frac{m}{2} \rho \left( \frac{dz_c}{dt} \right)^2. \tag{2.28}
\]

The empirical choice of \( \lambda = \pi/4 \) appeared to be in good agreement with Siegel’s experimental data, although most of the agreement might be attributed to Siegel’s other correction on the liquid surface tension (see Section 2.3.5.6). As pointed out by Zhmud et al. [2000], the condition of initial velocity equal to 0 is compatible with (2.28) because the correction \( \lambda r_0 \) removes the singularity associated with (2.11) at \( t = 0 \).

While the approach of Siegel [1961] to reservoir inertia seems rather empirical, it was independently confirmed by Szekely et al. [1971] through an innovative method. These authors examined the case of liquid rise into a vertical capillary tube. Rather than using the momentum conservation equation, they used an expression for the macroscopic energy balance of the system [Bird et al., 1960]. Szekely et al. [1971] used the liquid contained within the capillary tube for their system, for which the energy balance is

\[
\frac{d}{dt} \left( KE + PE \right) = -\Delta \left[ \left( \frac{u^2}{2} + gz + \frac{p}{\rho} \right) w \right] - W - E_v, \tag{2.29}
\]

where \( KE \) and \( PE \) denote the total kinetic energy and potential energy within the system, respectively, \( u \) is the longitudinal liquid velocity, assumed constant throughout the system and equal to \( \frac{dzc}{dt} \), \( p \) is pressure, \( w \) is the mass flow rate—i.e. the product of density, longitudinal velocity and cross sectional area—\( W \) is the rate of work done against the surroundings of the system, and \( E_v \) is the rate of work dissipated irreversibly. The three terms contained in the differencing operator, \( \Delta \), refer, respectively, to the net input of kinetic energy, the net input of potential energy and the net input of pressure energy at the cross-sectional boundaries of the liquid contained in the tube. In the particular case of capillary rise, the input of energy comes through the boundary with the reservoir, but there is no output of energy since the upper boundary is rising along with the liquid. The term \( W \) corresponds to the rate of work due to
viscosity forces. The term $E_v$ corresponds to the rate of work due to the contraction drag force given by (2.23). After calculation of each term and reduction, Szekely et al. [1971] obtained

$$\rho z_c \frac{d^2 z_c}{dt^2} = \Delta p(z_c) - \frac{8 \mu}{r_0^2} z_c \frac{dz_c}{dt} - \frac{e_c}{2} \rho \left( \frac{dz_c}{dt} \right)^2,$$  \hspace{1cm} (2.30)

which has the form of the equation that Rideal [1922] obtained, i.e. (2.12), but contains an energy loss term similar to the term proposed by Brittin [1946] (see (2.23)). It should be pointed out that the analysis of Szekely et al. is consistent with Rideal’s as the authors assumed an input of kinetic energy equal to $(1/2) \rho \pi r_0^2 (dz_c/dt)^3$ for elements of liquid entering into the capillary tube.

Szekely et al. [1971] proposed a different treatment of the total driving pressure as given by (2.5) to take into account the pressure distribution caused by the flow from the reservoir into the capillary tube. They took a total driving pressure equal to

$$\Delta p(z_c) = -\rho g z_c + \frac{2 \sigma \cos \theta}{r_0} + p_1(z_c),$$ \hspace{1cm} (2.31)

where $p_1(z_c)$ is the pressure at the inlet cross section. Note that, under conditions where $p_1(z_c)$ is given by (2.25), (2.30) is identical to (2.26) for the vertical case, i.e. for $\varphi = +90^\circ$.

![Figure 2.18. Far and near fields of the capillary tube reservoir.](image-url)
Under the assumption that the capillary tube was brought into superficial contact with the reservoir—i.e. that the reservoir head, $h$, was equal to zero—Szekely et al. [1971] obtained an expression for the inlet pressure, $p_1(z_c)$, by applying the energy balance (2.29) to the reservoir. The reservoir can be seen as a hemispherical liquid body extending from the inlet of the capillary to infinity, as illustrated in Figure 2.18. In the far field of the capillary tube—i.e. for any hemisphere of radius $r \geq r_0$—the liquid centripetal velocity, $u_r$, can be obtained from mass conservation equation in spherical polar coordinates [Batchelor, 2000], as

$$u_r = \frac{1}{2} \left( \frac{r_0}{r} \right)^2 \frac{dz_c}{dt}. \quad (2.32)$$

Hence, the corresponding kinetic energy can be computed. This problem is well known as the flow towards a spherical sink or away from a point source, and has been studied for both viscous and inviscid flow [O’Neill and Chorlton, 1989; Papanastasiou et al., 2000; Kornev and Neimark, 2001].

In the near field, i.e. the liquid contained in the hemisphere of radius $r \leq r_0$ (see Figure 2.18), the velocity field is not known. For this region, Szekely et al. [1971] assumed it was equal to the capillary tube velocity, $dz_c/dt$, which is, in fact, an upper-bound value of the actual velocity. Having derived the reservoir total kinetic energy, they obtained the expression for $p_1(z)$, as

$$p_1(z_c) = -\frac{7}{6} \rho r_0^2 \frac{d^2 z_c}{dt^2}. \quad (2.33)$$

However, in their energy balance, it seems that Szekely et al. mistakenly neglected the outflow of kinetic energy, $(1/2)\rho \pi r_0^2 (dz_c/dt)^2$, from the reservoir to the capillary tube. Hence, taking this into account, the authors should have obtained

$$p_1(z_c) = -\frac{7}{6} \rho r_0^2 \frac{d^2 z_c}{dt^2} - \frac{1}{2} \rho \left( \frac{dz_c}{dt} \right)^2, \quad (2.34)$$

which is analogous to (2.25), but takes the reservoir inertia into account. Substituting (2.34) into (2.31) and (2.30) yields

$$\rho \left( \frac{z_c}{6} + \frac{7}{6} r_0 \right) \frac{d^2 z_c}{dt^2} + \frac{8 \mu}{r_0^2} \frac{dz_c}{dt} = \frac{2 \sigma \cos \theta}{r_0} - \rho g z_c - \frac{m}{2} \rho \left( \frac{dz_c}{dt} \right)^2, \quad (2.35)$$

which is similar to (2.28) [Siegel, 1961] with $\varphi = +\pi/2$, $h = 0$, and $\lambda = 7/6$. Huang et al. [2001] used a similar approach to that used by Szekely et al. [1971], but instead computed a value of 3/8 for $\lambda$. 

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It is important to note that Szekely et al. [1971] should have obtained the coefficient $e_v$ instead of $m = 1 + e_v$ in the last term of (2.35), based on their use of the incomplete (2.33). However, the authors ended up with a coefficient $2 + e_v$ as the probable result of a sign error in their energy balance. This error was reported by Levine et al. [1976] and Sorbie et al. [1995], although none of the authors above referred to the error associated with (2.33). Batten [1984] extended the energy balance equation to capillary infiltration through porous media and made the same sign error resulting in an equation similar to that of Szekely et al. [1971]. The error went unnoticed in several publications, [e.g., Batten, 1984; Dullien, 1992; Ichikawa and Satoda, 1994]. The probable cause is that Szekely et al. took $e_v$ equal to 0.45 as mentioned in Section 2.3.4.1. Hence, their value of $2 + e_v$ equal to 2.45 is on the same order of $m$, which ranges from 2.24 to 2.41 as discussed earlier.

![Figure 2.19. Rise velocity of water in a vertical 0.133 mm radius capillary tube [after Jeje, 1979]. A. Experimental data; B. Model of Szekely et al. [1971] given by (2.35) ($m = 2.45$); C. Lucas-Washburn model (2.7).](image)

To conclude the discussion on the effect of reservoir inertia on capillary tube infiltration, it should be emphasized that replacing $z_c$ by $z_c + \lambda r_0$ for the inertia term only affects the early stages of the infiltration, i.e. when $z_c$ is of the order of $r_0$, the radius of the capillary tube. Therefore, the reservoir inertia has little effect on the overall behavior of the infiltration, and cannot explain the large differences between prediction and observation reported in Section 2.3.3.1. This is well illustrated in Figure
2.19, which shows velocity data on the rise of water into a vertical capillary tube. Equation (2.35) does little to improve Lucas-Washburn prediction (see (2.7)) and still overestimates the initial velocity of rise.

2.3.4.3 Couette Correction

In their critique of the paper of Szekely et al. [1971], Levine et al. [1976] suggested replacing the Hagenbach correction, \( m \), by \( m' + m'/Re \) where \( Re \) is the Reynolds number given by (2.10) and \( m' \) is a constant. Modifying (2.23), this corresponds to a total drag force given by

\[
f_{cd} = \frac{1}{2} \left( m + \frac{m'}{Re} \right) \rho \pi r_0^2 \left( \frac{dz_c}{dt} \right)^2.
\]  

(2.36)

The term \( m'/Re \) is sometimes called the Couette correction [Oka, 1960] and can be of importance when measuring the viscosity of fluids using a capillary rheometer [Ferguson and Kembłoski, 1991]. At low Reynolds numbers, the additional pressure drop associated with viscous dissipation of energy at the end of the tube is equivalent to an increase in effective length of the capillary tube, as illustrated by Ferguson and Kembłoski [1991]. Using (2.10) and replacing (2.36) in (2.28) for the case where \( h = 0 \) and \( \phi = \pi/2 \), the following expression is obtained

\[
\rho \left( z_c + kr_0 \right) \frac{d^2z_c}{dt^2} + \frac{8\mu}{r_0^2} \left( \frac{z_c + \frac{m'}{32} r_0}{r_0} \right) \frac{dz_c}{dt} = \frac{2\sigma \cos\theta}{r_0} - \rho g z_c - \frac{m}{2} \rho \left( \frac{dz_c}{dt} \right)^2.
\]  

(2.37)

Therefore, the supplementary viscous drag is equivalent to an increase in the total length from \( z_c \) to \( z_c + m'r_0/32 \) over which viscous forces apply.

As pointed out by Sylvester and Rosen [1970], (2.36) is not obtained directly from a solution of the equations of laminar flow. Instead, it is the superposition of the Hagenbach correction obtained for the inertia-dominant flow and the Couette correction obtained for the viscous flow. The form of (2.36) can be deduced from a dimensional analysis as, for example, that proposed by Bond [1922].

In experimental studies on viscous flow through an orifice [Bond, 1921] and viscous flow through short tubes of varying lengths [Bond, 1922], Bond measured values for \( m' \) equal to 36.7 ± 0.6. These results were found to be in very good agreement with a theoretical study of viscous flow through an orifice by Roscoe [1949], who derived a value for \( m' \) equal to 37.7—more exactly, 12\( \pi \). Based on Bond’s
results, Weissberg [1962] suggested that this value was independent of the length of the capillary tube \( l \), and derived an upper bound value for \( m' \) equal to 43.6, using an infinite size reservoir. However, in an experimental study, Astarita and Greco [1968] obtained an estimate for \( m' \) equal to 795, and suggested that \( m' \) was very sensitive to the geometry of the contraction. Their regression method was later criticized by Sylvester and Rosen [1970] who obtained a value for \( m' \) equal to 295 ± 50. Sylvester and Rosen [1970] also suggested that the value of \( m' \) decreased with a decrease in the surface area ratio of the tubes, \( r_0^2/r_r^2 \) where \( r_r \) is the radius of the reservoir.

Assuming that Sylvester and Rosen are correct, and considering a capillary tube of radius very small compared to the dimensions of the reservoir, \( m' \) should be of the order of \( 10^3 \). Therefore, the increase in effective length of capillary tube should not exceed a few radii, as demonstrated by (2.37). Thus, similar to the reservoir inertia correction \( \lambda r_0 \), the Couette correction is limited to the early stages of the capillary imbibition, for which the Reynolds number is large and the inertia dominant. Therefore, it is unlikely that this correction significantly affects the theoretical predictions of capillary infiltration. Nonetheless, it may prove significant at a low initial velocity, for example in the case of water drainage, for which a non-wetting fluid displaces water contained in a capillary tube.

The results obtained by Levine et al. [1976] support this conclusion. These authors derived (2.37) using a different approach. They wrote the Navier-Stokes equation for the longitudinal component of the capillary tube and assumed a parabolic velocity profile. With the exception of the vena contracta energy loss term, they obtained an equation identical to the combination of (2.30) and (2.31) obtained by Szekely et al. [1971]. Like Szekely et al., Levine et al. [1976] needed to obtain an expression for the inlet pressure \( p_1(z_c) \). Rather than writing the energy conservation equation for the liquid contained in the reservoir, the authors wrote the radial component of the Navier-Stokes equations, and, using the reservoir radial velocity, as given by (2.32), derived the reservoir pressure far field. Recall that the far field is the reservoir region for which \( r \geq r_0 \) (see Figure 2.18). Next, Levine et al. needed to link the inlet pressure to the pressure far field, which they did by writing a momentum conservation equation for the near field system, i.e. the liquid contained in the hemisphere \( r \leq r_0 \) (again, see Figure 2.18). Because the velocity near field was unknown, they had to approximate the rate of change of total momentum in the system. By taking into account the viscous forces acting along the surface \( r = r_0 \), the authors obtained an expression that took into account both the reservoir inertia and the Couette correction. They ultimately obtained an expression identical to (2.37) with \( \lambda = 37/36 \), \( e_v = 2.33 \) and \( m' = 8 \). It is, however, believed that Levine et al. should not have neglected the convective inertia term in their derivation of the pressure far-field and should have instead obtained \( e_v = 2.58 \), a value consistent with those found in the literature (see Section 2.3.4.1). In any case, the value the authors obtained for \( m' \) shows that the Couette correction for the effective viscous length is of the order of less than \( r_0 \), and can be neglected for practical purposes.
2.3.4.4 Conclusion on the Effects of Reservoir/Capillary Tube Interactions

Drag forces at the entrance to the capillary tube can potentially act to reduce the rate of infiltration of a liquid into the tube. Three potential pressure losses have been examined. (i) The Hagenbach correction incorporates the effects of energy loss as a result of a sudden contraction at the tube entrance. The resulting pressure drop is proportional to \((dz_c/dt)^2\). (ii) Because the infiltration velocity varies with time, the reservoir inertia also contributes to the pressure drop, and is equivalent to increasing the infiltration length of the capillary tube over which local inertia forces apply by a length of the order of \(r_0\). (iii) An additional pressure drop is related to the viscous dissipation at the entry to the capillary tube, and may be significant at low Reynolds numbers. The so-called Couette correction is equivalent to increasing the infiltration length of the capillary tube over which viscous forces apply by a factor of the order of one to several \(r_0\).

Overall, the reservoir inertia and Couette corrections are only significant at the very early stages of the capillary infiltration (when \(z_c\) is of the order of \(r_0\)). Past the early stages, and depending on the interface velocity, the Hagenbach correction may still significantly reduce the infiltration rate and needs to be accounted for in the model prediction. All three corrections fail to explain the discrepancies between measurements and predictions reported in the literature, thereby suggesting that other effects contribute to the infiltration rate reduction.

2.3.5 Infiltration Model Incorporating Meniscus Effects

2.3.5.1 Overview

There are two problems associated with the flow in the vicinity of the advancing meniscus. One is the departure from the parabolic velocity profile characterizing the Hagen-Poiseuille’s flow, where the moving meniscus appears to contradict the well-known no-slip condition at the wall of the capillary tube. The other is the observed dependence of the contact angle upon velocity. As will be shown here, these two problems are intimately connected. More generally, these topics are part of the broad topic of liquid spreading on solid surfaces, which has been extensively reviewed [Dussan V., 1979; de Gennes, 1985; Blake, 1993; Kistler, 1993; Blake and Ruschak, 1997], and has attracted the attention of many investigators both from a theoretical and a practical point of view [Shikhmurzaev, 1997]. Many unresolved issues remain, related mostly to the sub-microscopic mechanisms by which a liquid displaces another fluid from a solid surface [Kistler, 1993; Blake and Ruschak, 1997].
2.3.5.2 Static Contact Angles and Contact Angle Hysteresis

As introduced in Section 2.2.1.3, under ideal conditions, a liquid at rest in a bath of fluid (gas or other liquid) intersects a solid at a unique angle of contact (see Figure 2.2). The static contact angle, denoted $\theta_s$ (by opposition with the moving or dynamic contact angle introduced below, in Section 2.3.5.3), is the angle at which the meniscus at rest intersects the solid at the junction of the three distinct phases, liquid, fluid and solid. The three-phase junction line is commonly referred to as the contact line or wetting line.

![Figure 2.20. Contact line and static contact angle of at a liquid/gas interface [after Dussan V., 1979].](image)

Considering a liquid/gas interface as illustrated in Figure 2.20, Young’s equation relates $\theta_s$ and the surface tension of the liquid $\sigma$ to the surface tensions of the solid/liquid and solid/gas interfaces, denoted $\sigma_{L/S}$ and $\sigma_{G/S}$, respectively, for conditions of thermodynamic equilibrium and a perfectly flat, homogeneous solid surface. Young’s equation is given by

$$\sigma \cos \theta_s = \sigma_{G/S} - \sigma_{L/S}. \quad (2.38)$$

Deceiving in its simplicity, (2.38) has eluded experimental verification due to an inability to measure $\sigma_{L/S}$ and $\sigma_{G/S}$ [Dussan V., 1979]. Furthermore, according to Young’s equation, the contact angle is a unique property of the solid/liquid/gas system under consideration. In practice, however, a range of contact angles is usually possible. This is also true of solid/liquid/liquid systems [Fermigier and Jenffer, 1991]. The so-called contact angle hysteresis is attributed to the surface roughness, the solid heterogeneity and the presence of impurities on the solid surface [de Gennes, 1985; Blake and Ruschak, 1997]. While (2.38) may still apply at the microscopic scale, microscopic surface roughness, impurities and heterogeneities make multiple apparent
equilibrium configurations possible, thereby leading to the observed hysteretic behavior at the macroscopic scale. For those reasons, it is found experimentally that a contact line is pinned onto the solid, i.e. immobilized at a particular location of the solid surface, not only for \( \theta = \theta_s \), but whenever \( \theta \) lies within a finite interval around \( \theta_s \) [de Gennes, 1985]

\[
\theta_r \leq \theta \leq \theta_a ,
\]  

(2.39)

where \( \theta_a \) and \( \theta_r \) are the so-called static advancing contact angle and static receding contact angle, respectively. As illustrated in Figure 2.21.a, the receding contact angle, \( \theta_r \), is the smallest contact angle achievable before the wetting line begins to move in the direction of the wetting phase. In contrast, as illustrated in Figure 2.21.b, the static advancing contact angle, \( \theta_a \), is the largest contact angle achievable before the wetting line begins to move in the direction of the non-wetting phase. A measurement is usually made by causing the wetting line to move and determine the contact angle once movement has ceased [Blake and Ruschak, 1997].

![Figure 2.21. Contact angle hysteresis: a. Static receding contact angle; b Static advancing contact angle.](image)

For surfaces that have not been specially prepared, the interval \( \theta_a - \theta_r \) may be 10° or more in the case of liquid/gas systems [de Gennes, 1985]. For liquid/liquid systems, the interval can be even larger. Fermigier and Jenffer [1991] observed glycerin/silicon oil interfaces in horizontal capillary tubes of radius equal to 1 mm. For this system, the authors measured that the interval \( \theta_a - \theta_r \) was of the order of 60°.
2.3.5.3 Dynamic Contact Angles

During capillary infiltration by a liquid displacing or displaced by another fluid, the contact line at the liquid/fluid interface is observed to move. At a macroscopic length scale, or even at the scale of a few microns, the interface intersects the solid surface at a measurable dynamic contact angle \( \theta_d \), or more exactly an apparent dynamic contact angle given the resolution of common measurement techniques [Ngan and Dussan V., 1982; Kistler, 1993]. The dynamic contact angle is sometimes called the moving contact angle [Dussan V., 1979]. Dynamic contact angles under different geometrical configurations are shown in Figure 2.22. As shown in the figure, a variety of methods exist for observing and measuring the relationship between dynamic contact angles and the variables that influence them. These methods include the spreading of drops on a solid surface [Bracke et al., 1989; Foister, 1990] (see Figure 2.22.a), the flow in capillary tubes of circular section [Hoffman, 1975; Berezkin and Churaev, 1982; Legait and Sourieau, 1985; Calvo et al., 1991; Fermigier and Jenffer, 1991] or rectangular section [Ngan and Dussan V., 1982] (see Figure 2.22.b), the wetting lines formed by a plunging tape or plate [Blake and Ruschak, 1979; Bracke et al., 1989; Blake et al., 1999] (see Figure 2.22.c), and the rotation of a horizontal cylinder in a pool of liquid [Blake, 1993] (see Figure 2.22.d).

Hoffman [1975] examined the shape of the interface of various non-volatile liquids displacing air (i.e. imbibition) in a 1 mm radius capillary tube under conditions of forced displacement. That is, the meniscus driving force is imposed externally and typically results in a meniscus motion at constant velocity, by contrast with spontaneous displacement where the wetting process is inherently transient. Hoffman [1975] found that, under conditions where the different liquids perfectly wet the solid surface (i.e. perfect wetting \( \theta_s = 0 \) is assumed for those liquids), the apparent dynamic contact angle measured through the liquids follows a universal curve and depends exclusively on the capillary number defined as

\[
Ca = \frac{\mu u_0}{\sigma},
\]

where \( u_0 \) is the interface displacement velocity. The universal curve is illustrated in Figure 2.23. As shown in the figure, Hoffman provided data for capillary numbers ranging from \( 4 \times 10^{-5} \) to 36, corresponding to the full range of dynamic contact angles, from a few degrees at the lowest capillary number, to 180° for the highest capillary numbers. Other forced displacement data obtained by Fermigier and Jenffer [1991] and Hansen and Toong [1971] using an experimental setup similar to Hoffman’s, as well as those obtained by Bracke et al. [1989] and Ngan and Dussan V. [1982] using alternative experimental techniques, also fall close to the universal curve, implying that dynamic contact angles are independent of the flow geometry and measurement method (see Figure 2.23). This is further corroborated by data obtained for the spontaneous drop spreading of non-volatile liquids on a horizontal smooth surface,
which are also in good agreement with forced displacement data, and again suggest that capillary and viscous forces are the only dominant factors [Chen, 1988].

Figure 2.22. Dynamic contact angles in different geometries used to study them: a. Spreading drops; b. Liquid/fluid displacement in capillary tubes or between flat plates; c. Steady immersion or withdrawal of fibers, plates, or tapes from a pool of liquid; d. Rotation of a horizontal cylinder in a pool of liquid [after Blake, 1993].

Nevertheless, small deviations from the universal curve appear to exist [Kistler, 1993], particularly at small capillary numbers as shown by Blake and Ruschak [1997]. Furthermore, despite reasonable agreement with Hoffman’s data [Hoffman, 1975], data gathered by Ngan and Dussan V. [1982] using parallel glass plates (microscope slide) of varying gap (0.1 mm, 0.7 mm and 1.2 mm) show an influence of the gap on the dynamic contact angle-capillary number relationship. That is, at a given capillary number, the largest gap between plates is associated with the largest dynamic contact angle, as illustrated in Figure 2.24. A difference of about 10°-15° is observed between the 0.1 mm gap plate and the 1.2 mm gap plate. These findings imply that the dynamic contact angle is not a unique material property, as it can be somewhat affected by the macroscopic flow environment [Ngan and Dussan V., 1982]. Drop size effects on the
contact angle for spontaneous spreading have also been identified [Dussan V., 1979]. In addition, Hansen and Toong [1971] have reported radius size effects for flow in capillary tubes, although the trend is opposed to that of Ngan and Dussan V. [1982].

![Figure 2.23](image.png)

**Figure 2.23.** Dynamic contact angle as a function of capillary number for non-volatile, perfectly wetting liquids displacing air in capillary tubes [Hoffman, 1975; Fermigier and Jenffer, 1991] and between glass plates [Ngan and Dussan V., 1982] [after Kistler, 1993].

Considering liquids that partially wetted the solid surface (i.e. $\theta_s \neq 0$), Hoffman [1975] noticed that the static contact angle of a liquid could be absorbed in a shift factor $f^{-1}(\theta_s)$ as

$$\theta_d = f[Ca + f^{-1}(\theta_s)],$$

(2.41)

such that data on liquids with $\theta_s \neq 0$ also fell on the universal curve $\theta_d = f(Ca)$ associated with liquids of perfect wetting ($\theta_s = 0$). The mathematical structure of this generalized correlation indicates that surface wettability is important at low $Ca$ but its influence weakens at higher displacement speeds [Kistler, 1993]. Several relationships linking $\theta_d$, $\theta_s$ and $Ca$ have been proposed since Hoffman’s work. These relationships are most often of the form $\cos \theta_s - \cos \theta_d = f_1(\cos \theta_s, Ca)$ where $f_1$ increases with $Ca$ [e.g., Jiang et al., 1979; Bracke et al., 1989].

In practice, scatter of data is observed to be more important for partial wetting systems ($\theta_s \neq 0$) than for perfect wetting systems ($\theta_s = 0$) [Kistler, 1993]. Scatter can partly be attributed to an incorrect value assumed for the static contact angle, $\theta_s$, and
corresponding shift factor $f^{-1}(\theta_s)$ [Kistler, 1993]. Indeed, because of the static contact angle hysteresis discussed in Section 2.3.5.2, the measured static contact angle may not be the true static contact angle of Young’s equation (see (2.38)), such that the factor $f^{-1}(\theta_s)$ is computed from an incorrect value of $\theta_s$ and shifts the data away from the universal curve.

**Figure 2.24.** Variation of the dynamic contact angle, $\theta_d$, with capillary number during the displacement of air by silicon oil in between plane, parallel glass surfaces of nominal separation 0.1 mm (\(\nabla\)), 0.7 mm (O) and 1.2 mm (\(\Delta\)), at 25.0 ± 0.2°C. The oil has a viscosity of 0.97 Pa.s and a surface tension of 0.0197 N/m [after Ngan and Dussan V., 1982].

Furthermore, for partial wetting systems, a number of material-specific mechanisms have been reported, including distinct low-speed and high-speed regimes, polar liquid/surface electrostatic interaction, and unsteady advance (stick-slip) of the meniscus—possibly due to surface heterogeneity, roughness or contamination [Kistler, 1993].
2.3.5.4 Theoretical Treatment of Dynamic Contact Lines

For a long time, researchers could not solve the paradox associated with the contact line motion over a solid surface of a liquid displacing—or displaced by—a fluid, and the well-known no-slip boundary condition, which suggested that no such motion could occur [Dussan V., 1979]. This was further complicated by the mathematical development of conventional continuum theory and hydrodynamic assumptions giving rise to unbounded viscous stresses and pressures near the contact line [Huh and Scriven, 1971]. Experimental observations have since demonstrated that the no-slip condition and the moving contact line were compatible from a kinematic point of view [Dussan V. and Davis, 1974; Dussan V., 1977]. As shown by these experiments, the rolling motion of one fluid displacing the other is somewhat similar to the movement of a ball rolling down a plane, and yields the existence of a separating stream surface and toroidal eddies within one of the moving fluids [Dussan V., 1977]. This is well illustrated in Figure 2.25 where a liquid/fluid interface is observed to move on a solid surface. Note that the frame of reference moves with the interface OC. The separating stream surface is the line OA.

![Figure 2.25](image)

**Figure 2.25.** Velocity field during the displacement of a liquid/fluid interface over a solid surface [after Dussan V., 1979]. The solid is located below DOB and the fluids, above. Surface DOC bounds the fluid which is rolling off the solid and surface COB bounds the other fluid. The fluid point at D moves to O in a finite interval of time without slipping, whereupon it leaves the surface and arrives at C at some later time. The no-slip boundary condition is obeyed; however, the fluid does not adhere to the surface of the solid.

Thus, the stress singularity in the region of the contact line is not directly connected to kinematic incompatibility of the moving contact line. Instead, it is the
consequence of a multivalued velocity field at the contact line [Dussan V., 1979; Blake and Ruschak, 1997]. This suggests that in the neighborhood of the contact line, between the distance of a few molecules up to a few micrometers, microphysical processes other than classical hydrodynamics control the interface displacement [Kistler, 1993; Chen et al., 1997]. Identifying the processes taking place within a so-called inner region is no trivial matter, for its length scale is often beyond the resolution of most experimental techniques. Hence, verification of refined theories is often limited [Kistler, 1993; Chen et al., 1997; Ramé, 1997].

Figure 2.26. Schematic of the three regions of expansion used for hydrodynamic modeling of the moving contact line [after Kistler, 1993].

Despite these difficulties, hydrodynamic models predicting macroscopic behavior have been developed [e.g., Voinov, 1976; Huh and Mason, 1977; Cox, 1986; Dussan V. et al., 1991]. An example of hydrodynamic model is illustrated schematically in Figure 2.26. Typically, an ad hoc slip is permitted over a microscopic distance from the wetting line (the inner region shown in Figure 2.26), and is small compared to the macroscopic length scale of the flow—the radius of the capillary tube, for example [Hocking, 1977; Huh and Mason, 1977; Cox, 1986]. Other models ignore the vicinity of the contact line and associated singularity by truncating the solution in the inner region [Voinov, 1976]. The contact angle at this microscopic length scale, \( \theta_w \), is often taken to be equal to the static contact angle or some submicroscopic contact angle, so that existence of an apparent dynamic contact angle is attributed to viscous bending of the interface over an intermediate region (see Figure 2.26) much smaller than the macroscopic length scale, yet larger than length scale of the inner region where the unsolved wetting physics dominate [Dussan V. et al., 1991; Kistler, 1993; Blake and Ruschak, 1997]. The interface shape in the intermediate region is independent of geometry and may be used to form a boundary condition for the outer region where the macroscopic geometry dominates [Ramé, 1997].
Hydrodynamic models with ad hoc boundary conditions are successful at predicting the universal curve proposed by Hoffman [1975], i.e. (2.41), but begin to deviate at larger capillary numbers where the dynamic contact angle approaches 180° [Cox, 1986; Fermigier and Jenffer, 1991; Kistler, 1993].

![Figure 2.27](image1)

**Figure 2.27.** Schematic of a precursor film of non-volatile liquid spreading ahead of a macroscopic front (not to scale) [after Kistler, 1993].

Another treatment of the inner region is based on the idea that a thin precursor film extends ahead of the contact line, as illustrated in Figure 2.27. Under the precursor film assumption, the contact line where the dynamic contact angle, \( \theta_d \), is measured is only apparent, since the true contact line is now located at the microscopic leading tip of the liquid/fluid interface (see Figure 2.27) [Kalliadasis and Chang, 1994; Smith, 1995]. The macroscopic front merely acts as a reservoir for the precursor film, and thus slides over a pre-wet surface [Kistler, 1993]. Precursor films have been reviewed in detail by de Gennes [1985].

![Figure 2.28](image2)

**Figure 2.28.** Adsorption/desorption model of molecular displacement within the three-phase region [after Blake, 1993].
Finally, an alternative to the hydrodynamic theory views dynamic wetting as a rate process composed by individual molecule displacements [Blake and Ruschak, 1997]. According to Blake and Haynes [1969], surface tension provides the primary driving force of the wetting line motion and results in a disturbance of the thermodynamic equilibrium expressed by Young’s equation (see (2.38)). As illustrated in Figure 2.28, molecules adsorbed at localized sites on the initial solid/fluid interface are progressively displaced by molecules of the advancing fluid, resulting in the motion of the contact line [Blake, 1993]. This theoretical treatment of dynamic wetting is often referred to as adsorption/desorption model [Blake, 1993].

The molecular kinetic theory does not represent Hoffman’s universal curve [Hoffman, 1975] as effectively as the hydrodynamic theory. Nonetheless, it appears to be well-suited for data obtained for low capillary numbers where, for some systems, hydrodynamic bending alone does not provide a well suited mechanism [Blake and Ruschak, 1997].

2.3.5.5 Treatment Specific to Spontaneous Infiltration Into a Capillary Tube: Removal of the No-Slip Boundary Condition Near the Meniscus

Levine et al. [1980] have theoretically examined the rise of a liquid in a vertical capillary tube and allowed slippage at the contact line. The authors based their calculation on a number of assumptions that should be mentioned here: (i) The meniscus retains a fixed shape, which is the sector of a sphere. Therefore, the apparent dynamic contact angle \( \theta_d \) remains constant throughout the liquid infiltration. (ii) The liquid displacement is dominated by viscous forces, and inertia effects can be neglected. In other words, the liquid has risen sufficiently for a Lucas-Washburn type of flow to hold true. (iii) The usual non-slip flow condition along the tube wall holds true everywhere except in the immediate vicinity of the three-phase contact line, where a non-zero slip velocity is assumed. The length scale of the region over which slip occurs is negligible compared to the radius of the capillary tube, but the corresponding shear force has to be accounted for in the overall force balance.

Starting from the Navier-Stokes equations, Levine et al. [1980] used a longitudinal velocity profile as the sum of the classic parabolic profile term and a correction term due to the meniscus effects. The correction term was found to be equal the infinite sum of Bessel functions of the first kind. To obtain the rate of rise of the liquid, the \( z \)-component of the Navier-Stokes equations was integrated over the volume of liquid contained in the capillary tube. The classic Lucas-Washburn equation as given in (2.4) was derived, but contained a corrective term corresponding to the shear force exerted on the wall of the tube, as

\[
\frac{dz_c}{dt} = \frac{\nu_0^2}{8 \mu} \frac{\Delta p(z_c)}{z_c + \phi(\xi, r_c, \theta_d) r_0}, \tag{2.42}
\]
where $\Delta p(z_c)$ is the driving pressure similar to that given by (2.5), and $\phi(\xi, r_0, \theta_d)$ is a dimensionless function of the capillary radius $r_0$, the dynamic contact angle $\theta_d$, and $\xi$ the slip coefficient in the vicinity of the meniscus, which has the dimensions of a length. Levine et al. related $\xi$ to the length scale of microscopic irregularities, typically $10^{-3}$ mm to $10^{-5}$ mm, but suggested it was also dependent on $\theta_d$. The function $\phi(\xi, r_0, \theta_d)$ is calculated through a numerical procedure involving the boundary conditions and roots of Bessel functions. The expression given by (2.42) is identical to that obtained by Huh and Mason [1977] in a similar configuration, with the restriction in the case examined by Huh and Mason that $\theta_d$ is close to 90°.

Thus, according to Levine et al. [1980], the presence of the meniscus is equivalent to an increase in effective length of liquid infiltration from $z_c$ to $z_c + \phi(\xi, r_0, \theta_d) r_0$. The function $\phi(\xi, r_0, \theta_d)$ typically increases with decreasing contact angles. For $\theta_d = \pi/2$, it is of the order of 1 to 4, depending on the ratio $\xi / r_0$. For $\theta_d = \pi/10$, it is of the order 2 to 25. As pointed out by the authors, the correction becomes unrealistically large at low dynamic contact angles. The function $\phi(\xi, r_0, \theta_d)$ also increases with increasing radius $r_0$.

Equation (2.42) clearly shows that the velocity of rise $dz_c/dt$ at a given height $z_c$ is smaller than that predicted by Lucas-Washburn equation, i.e. (2.4), at the same given height. Hence, this trend is in good agreement with the experimental observations by Jeje [1979] and others mentioned in Section 2.3.2 that the measured velocity is less than the velocity predicted by Lucas-Washburn at the initial stages of the rise (see Figure 2.19). However, in their derivation of (2.42), Levine et al. [1980] made the assumption that inertia effects were negligible when compared to viscous effects, which only holds true past a certain tube length. Levine et al. argued that the inertia effects were significant over a length of the order of one tube diameter. This argument is in contradiction with the observation made in Section 2.3.3.1 that the length scale over which inertia effects are significant is of the order of ten capillary tube radii or more. The other issue associated with the model proposed by Levine et al. [1980] is, of course, the assumption of a constant dynamic contact angle.

A simplifying approach was proposed by Batten [1984] who based his reasoning on experimental observations [LeGrand and Rense, 1945; Calderwood and Mardles, 1955] indicating that the meniscus retained its shape during capillary rise once the flow was fully developed. Batten concluded that the flow in the region of the meniscus had to be uniform, and that the associated kinetic energy per unit volume for a uniform profile was $(1/2)\rho(dz_c/dt)^2$, twice as less as the kinetic energy per unit volume associated with the parabolic velocity profile in the region below the meniscus. The author hypothesized that the corresponding energy difference of $(1/2)\rho(dz_c/dt)^2$ was dissipated by liquid circulation below the meniscus. As discussed in Section 2.3.5.4, such eddies have been proposed and illustrated by Dussan V. [1977], although her experimental observations referred to a liquid-liquid displacement. Batten [1984] concluded that one could account for the uniform flow at the interface (and thus removal of the no-slip condition) by replacing the parameter $m$ in (2.35) by $m + 1$. Clearly, this approach is empirical.
Dynamic contact angle changes have always been a point raised in conjunction with the development of capillary infiltration models. Even Washburn [1921a] brought up the possibility that \( \theta_d \) could depend on \( dz_c/dt \), but assumed that it was a constant for the sake of simplification.

Successful prediction of an infiltration rate that accounts for variability of \( \theta_d \) is typically derived from a semi-empirical treatment. The idea is to use a correlation between the dynamic contact angle and velocity developed from a series of experiments [e.g., Hoffman, 1975; Berezhki and Churaev, 1982; Bracke et al., 1989] to predict the kinetics of another series of capillary infiltration experiments. The correlations need not be obtained from spontaneous capillary infiltration experiments, but may also be inferred from forced infiltration into capillary tubes [Blake and Haynes, 1969; Hoffman, 1975] or alternative techniques [Bracke et al., 1989]. As discussed in Section 2.3.5.4, the correlations find a theoretical justification using the molecular kinetic or hydrodynamic theory, and may be used as a boundary condition to investigate the macroscopic flow.

Earliest attempts to incorporate a change in dynamic contact angle with infiltration velocity may be attributed to Siegel [1961]. The author did not modify the contact angle per se, but suggested that the term \( 2\sigma \cos \theta / r_0 \) corresponding to the driving surface tension force in (2.37) be multiplied by a coefficient \( 1 - b \, dz_c/dt \), where \( b \) was a numerical constant to be chosen. Note that, if \( b \) is positive, the surface tension pull decreases with increasing interface velocity, and thus is consistent with an increase in dynamic contact angle with velocity. Siegel [1961] obtained reasonable agreement between experiments and his theory using a unique value of \( b \) for a series of four tubes of radii ranging from 0.95 mm to 16.4 mm, although poor agreement was observed for the largest tube.

Martynov et al. [1983] examined the rise of water in 52-µm radius capillary tubes. For their prediction of infiltration kinetics at an interface velocity smaller than 0.3 mm/s, i.e. passed the early stages of rise, they fitted their experimental data using the Lucas-Washburn equation (2.7) modified to account for a dependence of \( \theta_d \) upon interface velocity, as

\[
\cos \theta_d = \cos \theta - b_1 \left[ 1 - \exp \left( -b_2 \frac{dz_c}{dt} \right) \right], \tag{2.43}
\]

where \( b_1 \) and \( b_2 \) are two fitting parameters.

Perhaps the most convincing study to date is that due to Joos et al. [1990] and Van Remoortere and Joos [1991; 1993a]. These authors used the correlation developed by Bracke et al. [1989] for plates and strips drawn into liquids (i.e. forced wetting) as well as drops spontaneously spreading onto a solid. The correlation that Bracke et al. developed is given by
\[
\cos \theta_d = \cos \theta_s - 2(1 + \cos \theta_s)Ca^{1/2},
\]

(2.44)

where \( Ca \) is the capillary number (see (2.40)). Unlike (2.43), correlation (2.44) does not use fitting parameters. Correlation (2.44) is in good agreement with Hoffman’s experimental data [Hoffman, 1975; Van Remoortere and Joos, 1991].

Joos et al. [1990] substituted (2.44) into the Lucas-Washburn equation (2.4) for the case of the vertical capillary tube \((h = 0 \text{ and } \varphi = +90^\circ \text{ in (2.5)})\) and computed a numerical solution predicting the interface rise \(z_c\) as a function of time. Note that, in their study, the static contact angle in (2.44) was taken to be equal to 0. Next, they measured the infiltration kinetics of silicon oils of varying viscosity, ranging from 0.34 Pa.s to 58.8 Pa.s, into vertical capillary tubes of radius equal to 0.25 mm. They also examined the rise of silicon oil of viscosity 12.25 Pa.s into capillary tubes of varying radius, ranging from 0.25 mm to 1 mm. It should be pointed out that, because of the large viscosity of those silicon oils, the rate of rise was quite small. A rise of around 10 mm took of the order of several hundred seconds.

Overall, Joos et al. [1990] found that the Lucas-Washburn equation (2.7) (i.e. the equation that assumed a constant dynamic contact angle) offered a poor approximation of the rise kinetics, most particularly for the largest capillary tubes. In contrast, the numerical solution derived by Joos et al. offered a good prediction of the measured kinetics. An example is shown in Figure 2.29. Similar findings were obtained for horizontal capillary tubes [Van Remoortere and Joos, 1991].

![Figure 2.29.](image)

**Figure 2.29.** Capillary rise of silicon oil of viscosity 12.25 Pa.s in a tube of radius 0.25 mm [after Joos et al., 1990]. 1. Lucas-Washburn model incorporating (2.44); 2. Lucas-Washburn model (2.7) with constant contact angle \( (\theta_d = \theta_s = 0) \).
In a later investigation, Van Remoortere and Joos [1993a] examined the rise of viscous paraffin oil on silane-coated as well as solid paraffin-coated glass capillary tubes of radius ranging from 0.15 mm to 0.375 mm. Chemical bonding occurred for silane coating, while for paraffin coating, a thin film of solid paraffin adhered physically to the glass surface [Van Remoortere and Joos, 1993a]. In either case, the static contact angle was larger than zero, such that only partial wetting conditions were achieved.

Examining the rise of paraffin oil into silane-coated capillary tubes, Van Remoortere and Joos [1993a] found that Lucas-Washburn equation (2.4), where the contact angle is allowed to vary as in (2.44), gave a very good prediction of the observed rise kinetics. Incidentally, the authors also found that the static contact angle, \( \theta_s \), decreased with radius, thereby suggesting that this angle was not the true thermodynamic equilibrium static contact angle described by Young’s equation, i.e. (2.38). Note that Van Remoortere and Joos back-calculated the static contact angle from the final equilibrium height (i.e. using (2.8)).

Rise in paraffin-oil coated capillary tubes yielded a different behavior. The paraffin oil first started rising at a rate of rise consistent with (2.44). However, the liquid went past the silane-coated tube equilibrium point and kept rising at a much lower rate to finally reach an equilibrium height where the back-calculated contact angle appeared independent of radius. In this second rise phase, use of (2.44) could not be made to successfully predict the rise kinetics. Van Remoortere and Joos [1993a] concluded that at large capillary numbers (the transition was estimated at 0.01) corresponding to the first stage of rise, the contact line could slip over the tube wall roughness. On the other hand, at low capillary numbers, a second regime was in place, such that the actual circumference of the contact line depended on the surface roughness and led to more energy dissipation.

Recently, Hamraoui et al. [2000] have examined the rise of water and ethanol into vertical capillary tubes of radius close to 0.3 mm. Assuming perfect wetting they used an expression for the dynamic contact angle close to that of Siegel [1961], i.e.

\[
\cos \theta_d = 1 - b \frac{dz}{dt}, \tag{2.45}
\]

where, again, \( b \) is a numerical constant. Based on the molecular kinetic approach of Blake and Haynes [1969] to contact line displacement (see Section 2.3.5.4), Hamraoui et al. [2000] interpreted \( b \) as a molecular parameters describing the surface roughness of the capillary tube walls.

For both water and ethanol, the combination of Lucas-Washburn equation (2.4) with a variable dynamic contact angle as per (2.45) yielded very good agreement of the observed rise kinetics, and significantly improved the prediction made by the Lucas-Washburn equation alone. Values of \( b \) were back-calculated. For water, it was found that the value of \( b \) was larger for initially dry tubes than for pre-wet tubes, a result
consistent with the idea that $b$ is a friction coefficient. No difference was observed for ethanol. The observation that the rate of rise is slower for initially dry tubes than for pre-wetted tubes has been reported before [Calderwood and Mardles, 1955; Mumley et al., 1986; Ichikawa and Satoda, 1994]. When a capillary tube is pre-wet with the rising liquid, the existing film of water contributes to decrease the value of the dynamic contact angle, and thus increase the magnitude of the driving capillary force.

It should be mentioned that there have also been attempts in the case of capillary rise into vertical tubes to create a dependence of dynamic contact angle upon time. Typically, the relationship takes the form [Newman, 1968; Batten, 1984]

$$\cos \theta_d = \cos \theta_s (1 - b e^{-b t}), \quad (2.46)$$

where $b_1$ and $b_2$ are fitting parameters. Good agreement with experiment is reported when using (2.46) [Newman, 1968; Batten, 1984]. Equation (2.46) makes sense intuitively, since the capillary rise starts off with a large interface velocity and thus a large associated dynamic contact angle, before reaching equilibrium at infinite time with an interface at static contact angle. Therefore, equation (2.46) is an indirect form of the dependence of dynamic contact angle upon interface velocity.

### 2.3.6 Liquid/Liquid Displacement

#### 2.3.6.1 Overview

While liquid/gas imbibition and wetting line displacement have been studied extensively, liquid/liquid displacement has only received little treatment. The reasons for this are hardly surprising. Liquid/liquid displacements are more complicated both from a theoretical and an experimental point of view. Furthermore, recent research in wetting phenomena has been undertaken by engineers and scientists mostly interested in surface coating processes, where air is displaced by a liquid deposited onto a solid surface [Kistler, 1993; Blake and Ruschak, 1997]. Nevertheless, liquid/liquid motion has received some attention mainly stimulated by the developing technologies of enhanced oil recovery [Mumley et al., 1986].

As will be shown here, the physics taking place at the vicinity of a liquid/liquid contact line are not well understood. Although studies tend to agree qualitatively, quantitative disagreement exists that can partly be attributed to differences in experimental conditions and the range of capillary numbers that were under investigation.
2.3.6.2 Early Studies on Forced Liquid/Liquid Displacement

First attempts to deal specifically with a liquid displacing another in a capillary tube can be attributed to Chittenden and Spinney [1966] and Blake et al. [1967] who independently examined the validity of Lucas-Washburn equation for a pair of liquids flowing into a horizontal capillary tube both under drainage and imbibition conditions. In the configuration of Chittenden and Spinney, as well as that of Blake et al., a capillary tube initially saturated with one of the liquids is connected between two reservoir tanks, as illustrated in Figure 2.30. Each tank is filled with one of the two liquids under investigation. The sum of the capillary pull (or resistance for drainage conditions) and hydrostatic head difference between the tanks drives the liquid/liquid interface displacement within the capillary tube, such that the liquid initially present in the capillary tube is forced away by the other.

Starting from (2.3), Chittenden and Spinney [1966] and Blake et al. [1967] showed that the steady-state interface velocity, \( u_0 \), was given by

\[
    u_0 = \frac{2r_0 \sigma \cos \theta_d + r_0^2 \Delta p_h}{8(\mu_{nw} l_{nw} + \mu_{w} l_w)},
\]

(2.47)

where \( r_0 \) is the capillary tube radius, \( \theta_d \) is the dynamic contact angle at the interface measured through the wetting fluid, \( l_{nw} \) and \( l_w \) are the lengths of the capillary tube occupied by the non-wetting and wetting fluids with viscosities \( \mu_{nw} \) and \( \mu_{w} \) respectively, and \( \Delta p_h \) is the hydrostatic pressure drop across the total length of the tube, i.e. \( l_{nw} + l_w \) (see Figure 2.30).

Under conditions where the denominator of the right-hand side term in (2.47) remains constant—which requires that \( \mu_{nw} = \mu_{w} \), otherwise a correction must be
made—(2.47) shows that the interface velocity is a linear function of the applied pressure $\Delta p_h$. This relies, of course, on the assumption that the contact angle remains a constant independent of interface velocity. If, on the other hand, non-linearity is observed, a dynamic contact angle can be back calculated from the values of $u_0$ and $\Delta p_h$. By varying $\Delta p_h$, different flow rates $u_0$ and associated dynamic contact angle $\theta_d$ can be achieved, so that a plot of $\theta_d$ versus $u_0$ can be obtained.

Chittenden and Spinney [1966] examined the displacement of water/cyclohexane in a 0.134 mm radius horizontal capillary tube at interface velocities ranging from 1 mm/s to 17 mm/s. Cyclohexane and water have similar viscosities at ambient temperatures [Lide, 1995], so that the denominator of the right-hand side of (2.47) could be assumed constant.

Chittenden and Spinney [1966] reported a slight dependence of dynamic contact angle upon velocity, consistent in trend with that of liquid/air displacements (see Section 2.3.5.3), both in the imbibition and drainage modes. Significant scatter was reported, but variations in dynamic contact angle over the velocity range did not appear to exceed $10^\circ$, except for the first imbibition run, described below, where variations in dynamic contact angles were of the order of $80^\circ$.

One of the important findings of Chittenden and Spinney’s study was that the dynamic contact angle-velocity curve was strongly affected by the conditions of preparation of the glass surface. Indeed, contact angles measured through water were found to be generally smaller if a water film had been initially deposited on the glass surface, intermediate if no pre-treatment had been made, and larger if a cyclohexane film had been initially deposited. In the latter case, Chittenden and Spinney [1966] had to distinguish the first imbibition run (or dry runs) where water had never wetted the capillary tube (and for which dynamic contact angles were the largest), from later imbibition re-runs (or pre-wetted runs) where the tube had a prior history of water wetting. For the same given interface velocity, Chittenden and Spinney observed differences in contact angle of the order of $40^\circ$ between water and cyclohexane pre-treatment, and of at least $40^\circ$ between water’s first imbibition and later imbibitions.

Blake et al. [1967] examined the displacement of a water/benzene system and also found that the results were dependent on the conditions of preparation of the glass surface. Their first system consisted of an uncoated horizontal capillary tube of radius 0.45 mm. For that system, benzene was the non-wetting fluid. The authors varied the hydrostatic pressure drop and measured the associated interface displacement velocity. Results of their measurements are plotted in Figure 2.31. As shown in the figure, the interface velocity was found to be linearly related to the applied pressure, suggesting, unlike Chittenden and Spinney [1966], that the contact angle was independent of the displacement rate in the range studied in both imbibition (water displacing benzene) and drainage (benzene displacing water). Blake et al. measured a coefficient of proportionality between $u_0$ and $\Delta p_h$ that was close to that calculated using (2.47). The authors did not distinguish between dry run or pre-wetted runs.

For the experiments of Blake et al. [1967] on the first (uncoated) system, displacement rates below about 0.3 mm/s could not be achieved in both drainage and imbibition because of irregular movement at the interface. After extrapolating the $u_0$-
Δ\(\phi\) regression lines below the range of rates studied, Blake et al. noticed strong hysteretic effects at zero-velocity. That is, the back-calculated static receding contact angle (i.e., drainage history) was found to be equal to 43° whereas the static advancing contact angle (i.e., imbibition history) was equal to 96° (see Figure 2.31). Direct meniscus observation showed that, indeed, the interface curvature changed direction as the direction of displacement was reversed. Note that the magnitude of the contact angle hysteresis observed by Blake et al. [1967] is in good agreement with that later reported by Fermigier and Jenffer [1991] for glycerin/silicon oil interfaces (see Section 2.3.5.2).

![Diagram](image.png)

**Figure 2.31.** Benzene/water displacements in 0.45 mm radius uncoated capillary tube. Upper curve: water displacing benzene (imbibition); lower curve: benzene displacing water (drainage) [after Blake et al., 1967].

For their second system, Blake et al. [1967] used a horizontal 0.2 mm radius capillary tube. Glass surface exposure to dimethyldichlorosilane vapor created a chemically bonded organophilic layer on the capillary tube walls, such that benzene was now the wetting fluid. In this second configuration, stable velocities of a few micrometers per second could successfully be achieved.

The displacement rate \(u_0\) was found to be linearly related to \(\Delta p_h\), as long as \(u_0\) exceeded 0.15 mm/s. Similar to the uncoated tube, extrapolation of the \(u_0-\Delta p_h\) regression lines showed strong hysteretic effects. For water displacing benzene, a static contact angle of 19° (measured through benzene) was back-calculated. For benzene displacing water at rates larger than 0.15 mm/s, a static contact angle of 81°
could be back-calculated. For rates below 0.15 mm/s, however, $\Delta p_k$ and $u_0$ were non-linearly related, implying a possible dependence of contact angle upon interface velocity for liquid/liquid displacement in the case where the wetting fluid displaced the non-wetting fluid.

As pointed out by Blake et al. [1967], these results were only preliminary. In a later study on a system identical to the second system described above, Blake and Haynes [1969] found a dependence of liquid/liquid dynamic contact angle upon velocity in both the imbibition and drainage cases over the velocity range $10^{-3}$ mm/s to 1 mm/s. Their results are reproduced in Figure 2.32. As illustrated in the figure, Blake and Haynes [1969] demonstrated that, in the case of water displacing benzene (i.e., a non-wetting fluid displacing a wetting fluid, or in the drainage mode), the contact angle $\theta_d$ measured through benzene decreased with interface velocity. Conversely, for benzene displacing water (i.e., a wetting fluid displacing a non-wetting fluid, or in the imbibition mode), the contact angle increased with velocity. Overall, a linear relationship between $\cos \theta_d$ and $\ln u_0$ was established which supported the authors qualitative predictions from molecular kinetic theory (see Section 2.3.5.4).

**Figure 2.32.** Dynamic contact angle dependence on interface velocity for benzene/water displacements in silane-coated (hydrophobic) 0.2 mm radius capillary tubes [after Blake and Haynes, 1969]. -●- Water displacing benzene (so-called tube A); ○ benzene displacing water (tube A); □ benzene displacing water (so-called tube B). Note that the original reference uses contact angles measured through water.

Legait and Sourieu [1985] examined the displacement of a water-glycerine/oil interface into horizontal capillary tubes of radii ranging from 15 µm to 50 µm both in drainage and imbibition (setup similar to that shown in Figure 2.30). The interface
velocities varied from $10^{-2}$ mm/s to $10^{-1}$ mm/s. The authors found that the dynamic contact angle measured through the glycerine solution was nearly constant in drainage, but increased with an increasing capillary number in imbibition. While the drainage results appeared to contradict those shown in Figure 2.32, Legait and Sourieau [1985] reported a static receding contact angle of 0° for the pair of liquids under consideration. In other words, in their experimental range of drainage velocity, perfect wetting must have taken place at all times. Unlike Blake et al. [1967], there was no noticeable hysteresis between the advancing and receding modes.

Legait and Sourieau [1985] also observed that, at an identical interface imbibition velocity, the largest dynamic contact angle was associated with the largest capillary tube radius, a result in agreement with Ngan et Dussan V. [1982] for liquid/gas displacement (see Section 2.3.5.3).

### 2.3.6.3 Spontaneous Liquid/Liquid Imbibition Into Vertical Capillary Tubes

Mumley et al. [1986] considered the spontaneous rise of glycerol-water mixtures displacing silicon oils or hydrocarbons in vertical capillary tubes of about 1 mm in radius [see also Mumley et al., 1984]. The conditions of rise were similar to those of a liquid displacing air. That is, a tube initially filled with non-wetting silicon oil was set in contact with the wetting glycerol-water solution. The rate of rise was large at first, then decreased until the meniscus reached an equilibrium value given by a form of (2.8) where $\rho$ is replaced by the density contrast between the two liquids. Typical times of rise to equilibrium were of the order of a few hundred seconds. Examples of such rises are shown in Figure 2.33.

Mumley et al. [1986] noticed that the rise kinetics and interface final equilibrium height depended on the conditions of preparation of the glass surface. Observations were consistent with those reported by Chittenden and Spinney [1966] for liquid/liquid displacements and Calderwood and Mardles [1955] for liquid/air displacements. Mumley et al. [1986] noticed that if the capillary tubes were initially pre-wetted (PW) with a film of glycerol-water solution prior to being inserted into the silicon oil, then the rate of rise was faster than in the case where the tubes were initially dry (D) (see Figure 2.33). Intermediate rates of rise were also reported for so-called prewet and dried (PWD) tubes where a very thin film of glycerol-water was deposited onto the tube prior to the experiment. Overall, the equilibrium heights were found to be equal in the PW and PWD cases despite their differences in rate of rise. The equilibrium height in the dry case was found to be less than those of the PW and PWD cases by about 20%.

Mumley et al. [1986] suggested that for PW conditions, and to some extent, for PWD conditions, the wetting film of glycerol-water contributed to lower the dynamic contact angle towards perfect wetting such that the driving capillary force was larger and contributed to faster rates of rise. Modifying the Lucas-Washburn equation (2.4) for two liquids, the authors obtained
where \( z_c(t) \) is the height of rise of liquid 1 (of viscosity \( \mu_1 \) and density \( \rho_1 \)) displacing liquid 2 (of viscosity \( \mu_2 \) and density \( \rho_2 \)) in a vertical capillary tube of length \( l \). End effects and local inertia were found to be negligible for the conditions of the experiments.

\[
\frac{8\mu_1}{r_0^2} \frac{dz_c}{dt} + \frac{8\mu_2}{r_0^2} (l - z_c) \frac{dz_c}{dt} + (\rho_1 - \rho_2) g z_c r_0 = \frac{2\sigma \cos \theta_d}{r_0}, \tag{2.48}
\]

Equation (2.48) is evaluated at \( \theta_d = 0 \).

Upon plotting selected results of PW rise kinetics, \( z_c(t) \), and comparing them to (2.48) evaluated at \( \theta_d = 0 \), the experimental data were found to tend towards the equation although the observed rate of rise was still slower than predicted by (2.48) (see Figure 2.33). By directly measuring the observed dynamic contact angle in the PW case, Mumley et al. [1986] found that the dynamic contact angle decreased from 40°-50° at very short times of rise, and then remained near 10° when the rise was 70% complete.

Next, data were fitted using (2.48) under the assumption of a dynamic contact angle-velocity function of the form

\[
\cos \theta_d = \cos \theta_s (1 - b_2 \text{Ca}^h), \tag{2.49}
\]
where \( b_1 \) and \( b_2 \) are two numerical constants. The capillary number \( Ca \) is given by (2.40) where \( u_0 \) is the interface velocity, \( \sigma \) is the liquid/liquid interfacial tension and \( \mu \) is the viscosity of the displacing fluid (i.e. \( \mu_2 \)).

By combining (2.49) and (2.48), and taking a value of \( b_1 \) equal to 0.5, Mumley et al. [1986] reported good agreement between experimental data and prediction. As per Hamraoui et al. [2000] (see (2.45) in Section 2.3.5.6), the value of \( b_2 \) was affected by the glass surface preparation (PW, PWD or D). The value of \( b_2 \) was also found to be dependent upon the viscosity ratio of the liquid/liquid pair. Note that \( b_1 = 0.5 \) is consistent with the correlation of Bracke et al. [1989] for liquid/gas imbibition (see (2.44)).

Mumley et al. [1986] concluded that the concept of a dynamic contact angle was somewhat misleading. Visual observation of moving menisci showed that the displacements were generally not smooth. For D tubes, the movement was often jerky and asymmetrical. Thus, according to the authors, the dynamic contact angle was more a conceptualization of hydrodynamic forces in macroscopic films near the interface than a true back-calculated geometric contact angle. The hydrodynamic forces were taken into account in a single excess-force term \( F^* \), a difference between the driving capillary force—right-hand side term of (2.48) computed from the final equilibrium position—and the resisting viscosity and gravity forces—left-hand side term of (2.48) computed from the kinetics data. The excess-force term \( F^* \) was found to be a function of the square root of the capillary number.

2.3.6.4 Recent Studies in Horizontal Capillary Tubes

The findings of Mumley et al. [1986] were later confirmed by Calvo et al. [1991] in their study on forced displacement of cyclohexane/water in horizontal 0.55 mm radius capillary tubes (water was the wetting fluid). Again, cyclohexane and water have similar viscosities at ambient temperatures [Calvo et al., 1991; Lide, 1995]. Surprisingly, the authors made no attempt to compare their study to that of Chittenden and Spinney [1966] (see Section 2.3.6.2) despite similarities in the experimental configuration (see Figure 2.30) and the liquids under investigation.

Aware of the conclusions of Mumley et al. [1986] on contact angles, Calvo et al. [1991] did not attempt to back-calculate or measure contact angles. Instead, they restricted their measurements to a dynamic capillary pressure given by

\[
p_c = \frac{2\sigma \cos \theta_d}{r_0}.
\]  

Using (2.50), the authors rewrote (2.47) as

\[
(\mu_{nw} l_{nw} + \mu_w l_w)u_0 = \frac{r_0^2}{8}(\Delta p_h + p_c).
\]
Calvo et al. [1991] considered two imbibition scenarios. In the first scenario, the capillary tube was initially saturated with cyclohexane, and water had never traveled through the tube. This scenario was similar to the dry (D) capillary rise described by Mumley et al. [1986] and the first imbibition run described by Chittenden and Spinney [1966]. In the second scenario, water had already traveled once through the tube, but had been slowly drained back to its initial position. According to Calvo et al., a water film was supposed to have persisted on the tube wall. This scenario was similar to the pre-wetted (PW) or possibly prewet-and-dried (PWD) capillary rise described by Mumley et al. [1986], and the pre-wetted imbibition runs described by Chittenden and Spinney [1966].

Figure 2.34. Variation of the dynamic capillary pressure with the logarithm of the capillary number Ca for forced dry imbibition of water displacing cyclohexane into a horizontal 0.55 mm radius capillary tube [after Calvo et al., 1991].

Calvo et al. [1991] first examined dry imbibition for capillary numbers varying from $5 \times 10^{-7}$ to $5 \times 10^{-4}$ corresponding to velocities $u_0$ varying from 0.0125 mm/s to 12.5 mm/s. As illustrated in Figure 2.34, they found that in the dry imbibition mode, capillary pressure decreased with capillary number from the static advancing capillary pressure. This suggests again that the dynamic contact angle increased with velocity (see (2.50)), a result qualitatively consistent with Mumley et al. [1986], Chittenden and Spinney [1966], as well as Blake and Haynes [1969] (see Figure 2.32). Comparison between Figure 2.32 and Figure 2.34 is difficult. Blake and Haynes [1969] reported contact angle data for a hydrophobic surface and did not provide capillary numbers.

Examining the upper range of capillary numbers reported in Figure 2.34, i.e. for Ca varying from $5 \times 10^{-5}$ to $5 \times 10^{-4}$, Calvo et al. [1991] observed a linear dependence of the capillary pressure upon capillary number. This is best shown in Figure 2.35.
The dynamic capillary pressure was found to become negative at large velocities, corresponding to an inversion of the meniscus curvature (i.e. $\theta_d > 90^\circ$ in (2.50)).

![Figure 2.35. Variations of the dynamic capillary pressure with capillary number for the motion of a water/cyclohexane interface meniscus in a horizontal 0.55 mm radius capillary tube [after Calvo et al., 1991]. -■- Imbibition in a D tube; -♦- Imbibition in a PW tube; -▲- Drainage. The solid lines are the linear regressions performed on the high velocity part of the curves.](image)

Examining imbibition in PW tubes, Calvo et al. [1991] measured dynamic capillary pressures that were systematically larger than those measured in dry imbibition, were less dependent upon velocity, and always remained positive (see Figure 2.35). As per Mumley et al. [1986], Calvo et al. also suggested that the presence of a water film left on the tube walls during the first cycle of imbibition and drainage ensured lower dynamic contact angles and thus lower capillary pressures. The weak dependence of dynamic capillary pressure upon capillary number is also consistent with the findings of Chittenden and Spinney [1966] for the pre-wetted imbibition runs (see Section 2.3.6.2).

For drainage (cyclohexane displacing water), Calvo et al. [1991] reported a linear increase of capillary pressure with capillary number (see Figure 2.35), again in qualitative agreement with Blake and Haynes [1967] (see Figure 2.32). Scatter was more significant in drainage than in dry imbibition.

Acknowledging the abrupt changes of capillary pressure at low velocity, Calvo et al. [1991] concluded that there existed a transition between two mechanisms of displacement. Depending on the velocity, the advancing fluid would be directly in contact with the solid wall (slow regime) or separated by a thin film of the displaced...
liquid (faster regime). This conclusion is identical to that drawn by Van Remoortere and Joos [1993a] for paraffin oil/air system (see Section 2.3.5.6).

For dry imbibition and drainage, Calvo et al. [1991] concluded that the variations of capillary pressure with interface velocity were much larger than predicted by liquid/liquid hydrodynamic theory [Cox, 1986], a conclusion reached earlier by Foister [1990] for the spontaneous imbibition of drops of liquid displacing another liquid and by Fermigier and Jenffer [1991] for the forced glycerol/silicon oil imbibition in 1 mm radius horizontal capillary tubes. The disagreement with hydrodynamic theory implied that mechanisms other than viscous bending were present. Calvo et al. [1991] suggested that the existence of so-called strong interface pinning effects might explain their observations. By pinning effects, the authors meant that the interface could be immobilized at a particular location of the capillary tube despite the existing driving pressure. The existence of such pinning effects was supposedly supported by the large observed hysteresis between static advancing and receding modes reported by these authors as well as others [Blake et al., 1967; Fermigier and Jenffer, 1991] (see also Section 2.3.5.2). Because the walls of the capillary tubes used in their study were smooth, Calvo et al. [1991] believed that pinning was most likely due to chemical impurities present at the wall and acting as pinning centers or pinning points—meaning that an impurity could hook (or anchor [de Gennes, 1985]) the interface meniscus and temporarily stop its motion. Similarly, Fermigier and Jenffer [1991] hypothesized that discrepancies between observations and predictions from the hydrodynamic theory could be attributed to glass surface heterogeneity, specific interaction mechanisms between the solid and the liquids, or perhaps a combination of both effects.

Finally, a last study by Van Remoortere and Joos [1993b] should be mentioned here. These authors examined the spontaneous motion of silicon oil displacing paraffin oil in horizontal capillary tubes of radius ranging from 0.125 mm to 0.26 mm. The silicon oil was the wetting fluid. Unlike prior studies [Chittenden and Spinney, 1966; Blake et al., 1967; Legait and Sourieau, 1985; Calvo et al., 1991] where the interface movement was forced between two reservoirs of liquids, Van Remoortere and Joos [1993b] considered the displacement of a finite-length column of silicon oil/paraffin oil, as illustrated in Figure 2.36. The column was moving solely under a capillary pull due to the surface tension forces at the front of the column (air/paraffin oil advancing interface) and interfacial tension at the center of the column (silicon oil/paraffin oil interface). The surface tension of the rear, receding meniscus (air/silicon oil) provided capillary resistance (see Figure 2.36). The capillary tube was initially dry and filled with air before the column was introduced.

Van Remoortere and Joos [1993b] modified (2.47) to account for the air/liquid interfaces. Assuming a given experimental configuration (fixed lengths of liquids and capillary tube radius), they showed that the interface displacement velocity was independent of time and given by the expression

\[
\frac{4(\mu_{nw}l_{nw} + \mu_{w}l_w)u_0}{r_0} = -\sigma_{w/air} \cos \theta_{w/air} + \sigma_{nw/w} \cos \theta_{nw/w} + \sigma_{nw/air} \cos \theta_{nw/air}, \tag{2.52}
\]

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where \( l_w \) and \( l_{nw} \) are the lengths of silicon oil and paraffin oil columns, respectively. The subscripts \( w/air \) refers to the air/silicon oil interface, \( nw/w \) to the silicon oil/paraffin oil interface and \( nw/air \) to the paraffin oil/air interface.

For the advancing paraffin oil/air interface, *Van Remoortere and Joos* [1993b] assumed that the dynamic contact angle \( \theta_{nw/air} \) was given by (2.44) (with perfect static wetting, i.e. \( \theta_s = 0 \) in (2.44)). For the receding silicon oil/air interface, they assumed perfect receding wetting, i.e. \( \theta_{w/air} = 0 \) at any velocity. The value of \( \sigma_{nw/w} \cos \theta_{nw/w} \) was unknown, but predicted to be small in magnitude when compared to the other two values since \( \sigma_{w/air} \) was equal to 0.0207 N/m, \( \sigma_{nw/air} \) to 0.0325 N/m and \( \sigma_{nw/w} \) to 0.0013 N/m.

![Diagram](image.png)

**Figure 2.36.** Spontaneous liquid/liquid displacement in a horizontal capillary tube: silicon oil (wetting liquid) displaces paraffin oil in air at atmospheric pressure [after *Van Remoortere and Joos, 1993b*].

After varying the capillary tube radius, the liquid column lengths and the viscosity of the silicon oil, *Van Remoortere and Joos* [1993b] found the variation of the column location with time to be typically linear, thereby suggesting that, indeed, the interface displacement velocity was a constant independent of time. The silicon oil/paraffin oil meniscus was observed to be in a driving configuration, i.e. \( \theta_{nw/w} \) measured through the wetting fluid was a little less than 90°. Nonetheless, upon using (2.52) to back calculate the value of \( \cos \theta_{nw/w} \), *Van Remoortere and Joos* found that \( \cos \theta_{nw/w} \) was negative and took most often values smaller than \(-1\). These findings implied that the model given by (2.52) was incorrect and that there existed a resisting force improperly accounted for. In place of (2.52), the authors suggested using

\[
\frac{4(\mu_{nw} l_{nw} + \mu_{w} l_{w})u_0}{r_0} = -\sigma_{w/air} \cos \theta_{w/air} - T_{nw/w} + \sigma_{nw/air} \cos \theta_{nw/air}, \tag{2.53}
\]

where \( T_{nw/w} \) is a positive tension. \( T_{nw/w} \) took different values depending on the experimental configuration and varied between 0.0021 N/m and 0.0106 N/m (most values in the range 0.0090 N/m-0.0106 N/m). In other words, the magnitude of \( T_{nw/w} \) was comparable to that of the driving surface tension force.
Van Remoortere and Joos [1993b] suggested that the existing interfacial tension resistance might reflect pinning of the liquid/liquid contact line on the heterogeneities of the solid substrate of either chemical or physical nature, an argument also proposed by Calvo et al. [1991]. The authors also concluded that enhanced dissipation may be occurring at the contact line because the actual circumference of the contact line was larger than the circumference of the tube and depended on the tube’s roughness. This explanation had already been proposed by the authors in an earlier study [Van Remoortere and Joos, 1993a] on the partial wetting of a paraffin oil/air system discussed in Section 2.3.5.6. The magnitude of capillary numbers were comparable in both studies.

2.3.6.5 Conclusion on Liquid/Liquid Displacements

To date, a complete description of the dynamic capillary pressure or contact angle dependence upon liquid/liquid interface displacement velocity or capillary number is not available in the literature. Nonetheless, a number of trends can be inferred from all the studies reported in Section 2.3.6.

The liquid/liquid static contact angle is found to exhibit a large hysteresis. In fact, variations of 60° have been reported between the static advancing contact angle and the static receding contact angle for liquid/liquid systems (see Section 2.3.6.2).

Figure 2.37. Schematic view of the interface contact structure (a) with a dry tube and (b) with a pre-wetted tube [after Calvo et al., 1991]: w, wetting liquid; nw, non-wetting liquid.
Evidence of a dynamic contact angle dependence upon velocity is found in both imbibition and drainage. For imbibition, the contact angle increases with capillary number. For drainage, the contact angle decreases with capillary number, but the dependence is typically less than that in imbibition. Abrupt changes of the contact angle at low capillary numbers (less than $10^{-4}$) have been reported, suggesting two different contact line displacement regimes and associated mechanisms depending on the magnitude of the capillary number.

In imbibition, the surface preparation strongly affects the displacement kinetics. The contact angles are smaller, and thus the driving capillary pressures are larger if the tube is pre-wetted by the wetting liquid. Most studies conclude that in the pre-wetted case, the existing film of wetting liquid contributes to decrease the value of the contact angle, as illustrated in Figure 2.37, and reduce its dependence upon velocity.

Large changes in the dynamic contact angle have been attributed to chemical impurities, surface roughness, solid heterogeneity or other specific solid/liquid interactions. Given the large hysteresis of liquid/liquid contact angles, stick-slip resulting from temporary line pinning is a plausible explanation for the rapid changes of dynamic contact angles [Kistler, 1993]. In any case, it appears that the mechanisms governing liquid/liquid displacement may differ from those governing liquid/gas displacements and may not be simply inferred from the latter.

### 2.3.7 Summary of Capillary Infiltration Kinetics

Studies of capillary displacement into capillary tubes where one liquid displaces another fluid (gas or another liquid) generally show that the Lucas-Washburn model given by (2.4) over-predicts the rate of infiltration at its early stages. The discrepancies appear to increase for larger radius capillary tubes and if capillary tubes are initially dry of the infiltrating liquid.

For the infiltration of liquids displacing air, a number of studies has attributed the observed discrepancies between the Lucas-Washburn prediction and experimental data to the fact that the effects of inertia, and the pressure drop at the entry to the capillary tube act to reduce the rate of infiltration. While the magnitude of these forces may be significant, and hence might need to be accounted for under a number of circumstances, the Lucas-Washburn prediction corrected to account for these effects still most often fails to predict the observed rate of infiltration. A possible explanation is that corrections that account for inertia are typically limited to the effects of local inertia while convective inertia, a non-linear term, is largely ignored. This issue is further addressed in Section 3.4.2 and Section 3.5.2.4.

Studies, which obtain good agreement between their model predictions and experimental data, usually make use of a dynamic contact angle dependent upon interface velocity to predict and verify the infiltration kinetics. However, these studies always require the estimation of some fitting parameters back-calculated from a first series of contact line displacement experiments in order to make further predictions.
Although these experimental studies have developed a theoretical treatment in qualitative, and sometimes quantitative, agreement with the data, a unified description of the exact physics driving contact line displacement cannot be found in the literature. Controversies especially remain for partial wetting liquid/gas systems ($\theta_s \neq 0$) and for liquid/liquid systems. The mechanisms driving the contact line motion are likely to be different depending upon the nature and wetting history of the solid surface, as well as the magnitude of the capillary number.

2.4 Geotechnical Centrifuge Modeling: Background and Previous Geoenvironmental-Related Work

2.4.1 Overview

During geotechnical centrifuge testing, a container housing a soil or rock experiment is spun around a central axis at a high rotational speed, thereby increasing the body forces acting on the medium and fluids within the container. At any given rotational speed, $\omega$, the container and its contents will experience a centrifugal acceleration, $r_G \omega^2$, where $r_G$ is the radial distance of the center of gravity of the container from the central axis. In the centrifuge modeling community, it is customary to describe this acceleration as the product of the Earth’s gravitational acceleration, $g$, and a scaling factor $n$—often termed the g-level [Taylor, 1995].

The geotechnical centrifuge can be used to perform reduced-scaled modeling of a full-scale problem, usually termed the prototype. The scale model of the prototype is constructed from in situ materials where all macroscopic dimensions, $z$, are reduced by the factor, $n$. Note, because in situ materials are used, microscopic dimensions, such as pore throat radii, are equivalent in the scale model and the prototype. The scale model is then subjected to a centrifugal acceleration equivalent to $ng$, as shown in Figure 2.38. Under these conditions, the product $\rho gz$, where $\rho$ is the in situ material density, will be the same at homologous points in the model and the prototype. Because the mechanical behavior of geologic materials is heavily dependent on the in situ stress levels and fluid pressures [Lambe and Whitman, 1969], geotechnical engineers have, for decades, exploited the centrifuge for reduced-scale modeling of soil-structure interaction problems [Fahey et al., 1990].

The ability to generate homologous stress and pressure conditions in a reduced-scale model of a prototype also has advantages when it comes to the experimental investigation of subsurface flow [see Culligan-Hensley and Savvidou, 1995]. For example, the equivalence of geologic stress levels is often required to obtain equivalence in material density, i.e. particle packing, between a model and its prototype [Berner, 1980]. Because particle packing effects microscopic dimensions such as pore throat radii, material density equivalence between a model and its prototype has to be
achieved to ensure that parameters such as fluid hydraulic conductivity and tortuosity are properly replicated in the model.

Figure 2.38. Principle of centrifuge modeling. Gravity effects in a prototype are identical to inertial effects in a centrifuge model [after Schofield, 1980].

In problems involving multiphase flow, similitude of the product $\Delta \rho g z$, where $\Delta \rho$ is the density contrast between the fluid phases, can also be preserved between a scale centrifuge model and its prototype. Hence, the body forces acting to drive multiphase flow can be correctly replicated at a reduced scale in the geotechnical centrifuge. Due to the equivalence of microscopic dimensions between a centrifuge model and its prototype, the capillary forces acting to retard or drive multiphase flow are also correctly replicated. Thus, the balance between body forces and capillary forces can be properly accounted for in a reduced-scale centrifuge model. For this reason, centrifuge testing allows the possibility of conducting studies of multiphase flow behavior in a scale model under well controlled conditions. The same cannot be said of multiphase flow behavior observed during reduced-scale laboratory experiments. This is because the body forces driving multiphase flow are not correctly reproduced in a scale experiment carried out under the Earth’s gravity. Hence, flow phenomena observed under these conditions may not be representative of the field.

The fundamentals of centrifuge modeling of subsurface flow, scaling laws and stress variation in centrifuge models have been reviewed elsewhere [e.g., Culligan-Hensley and Savvidou, 1995; Ratnam, 1996; Marulanda, 2001] and will not be discussed here in extensive detail. It is, however, important to emphasize that reproduced flow velocities are larger than those existing in the field for both centrifuge experiments and laboratory experiments. For centrifuge experiments, velocity is multiplied by a factor $n$ while the applied gradient is a constant. This is because the hydraulic conductivity is multiplied by $n$ under conditions where the material
properties are unchanged [Nimmo and Mello, 1991; Culligan et al., 1997; Culligan and Barry, 1998]. For laboratory experiments, while the hydraulic conductivity is unchanged, samples are often tested at higher hydraulic gradients than those in the field because of time constraints. Hence, the flow velocity is increased. In either case, the flow velocity must remain sufficiently small such that the Reynolds number in porous media, given by a form similar to (2.10), is less than 1 to 10 [de Marsily, 1986; Bear and Verruijt, 1987; Longino and Kueper, 1999b] to ensure viscous laminar flow with negligible inertia forces. If the velocities lie outside of this range, Darcy’s law will no longer be valid. Hence, the hydraulic conductivity or pore sizes may be underestimated.

Because it is not possible to reproduce the full complexity of field scale heterogeneity in a reduced-scale experiment, centrifuge modeling, as per any other experimental technique, will not generate data that lead on to direct predictions at a field site. Nonetheless, the use of the geotechnical centrifuge to identify macroscopic trends and phenomena at a field site can provide an insight that might not be gathered using alternate experimental techniques, and is helpful in providing data for the development and validation of numerical models describing geoenvironmental processes [Culligan-Hensley and Savvidou, 1995; Mitchell, 1998].

2.4.2 Previous Related Research Work

In spite of the apparent world wide interest in geotechnical centrifuge modeling, application towards modeling of geoenvironmental problems is still relatively new, and publications in this area have been few in numbers [Ratnam, 1996]. However, recent efforts to grasp environmental issues combined with networking efforts from the different geotechnical centrifuge research groups around the world have led to the undertaking of a number of projects [Kimura et al., 1998; Ng, 2001]. In particular, the European Commission has sponsored the Network of European Centrifuges for Environmental Research (NECER) project [Garnier et al., 2000a; Garnier et al., 2000b].

Examples of environmental applications of geotechnical centrifuge modeling include the combined heat and solute transport around a buried, heat generating waste source [Hensley and Savvidou, 1993], the evaluation of the long-term performance of clay-liners [Theriault and Mitchell, 1997], the unstable infiltration of fluids into porous media, also known as wetting front instability or fingering [Culligan et al., 1997; Griffioen and Barry, 1999; Culligan et al., 2002], the flow of fluids in unsaturated porous media [Nimmo, 1990; Barry et al., 2001], and gas migration under the remediation technology of air sparging [Marulanda et al., 2000; Marulanda, 2001].

Of particular interest are centrifuge modeling investigations of NAPL flow. In fact, small radius centrifuges have long been used by the petroleum industry to measure capillary pressure-saturation relationships of cored samples because of the steep hydraulic gradients that can be achieved by this technique [Chen, 1997]. Because the
saturation is radius dependent, and thus not uniform along the sample, the flow measurements need to be interpreted. A large number of interpretation methods exist with different degrees of approximation [Forbes, 1998].

The use of larger radius, geotechnical centrifuges for investigating NAPL behavior in the subsurface through physical modeling is more recent. Illangasekare et al. [1991] have examined the one-dimensional downward transport of Soltrol 220 (LNAPL) released at the top of columns of dry fine sand, partially saturated sand and partially saturated silt. For the dry fine sand, scaled data of the reduced-scale infiltration front as a function of time were found to be in agreement with data for the equivalent prototype (see Figure 2.39). Infiltration of LNAPL into partially saturated sand and partially saturated silt showed patterns similar to those for the dry fine sand, but comparison with an equivalent prototype was not made. Overall, those results demonstrated the feasibility of using the centrifuge modeling technique for studying processes of immiscible fluid flow through soils. For the dry fine sand, the suction head due to capillary forces was of significant magnitude compared to the LNAPL penetration depth and pool height, such that both capillary forces and self-weight forces had to be taken into account for predicting the infiltration kinetics.

![Figure 2.39](image)

**Figure 2.39.** Downward transport of Soltrol 220 released at the top of a column of dry fine sand: comparison of prototype (1 g) infiltration front and infiltration front from a scaled centrifuge test [after Illangasekare et al., 1991].

In later studies, Mitchell and Stratton [1994] and Knight and Mitchell [1996] examined the fate of silicon oil (LNAPL) released into a partially saturated medium sand at different gravitational levels. Knight and Mitchell [1996] compared the oil saturation two-dimensional plumes after a prototype time of two months for scaled data of two centrifuge tests run at 15 g and 30 g, respectively, and found very good
agreement between the plumes (see saturation profile shown in Figure 2.40). Radial migration of the 15 g scale plume reached the edges of the centrifuge box such that comparison with the 30 g plume was limited in this direction. Knight and Mitchell [1996] also noticed that a higher rate of silicon oil release was associated with a deeper penetration into the partially saturated fine sand, a feature that the authors could not predict numerically.

Figure 2.40. Silicon oil point source release on top of a column of medium sand partially saturated with water: oil saturation profile along the axis of symmetry of the sample 2 months after release (in prototype time) [after Knight and Mitchell, 1996].

The work of Knight and Mitchell [1996] was extended by Esposito et al. [1999]. The authors examined motor oil (LNAPL) movement into partially saturated sand compacted at two values of sand density. Good agreement was observed between scaled data of tests conducted at 20 g and 30 g. Furthermore, an increase in porosity (i.e. decrease in sand density) and/or decrease in LNAPL source discharge appeared to decrease the horizontal extent of the LNAPL plume content and the downward migration of the contamination. The effect of sand density on the shape of the LNAPL plume is illustrated in Figure 2.41.

In the studies of Knight and Mitchell [1996] and Esposito et al. [1999], numerical modeling and theoretical prediction was complicated by the presence of three phase: air, water and LNAPL. In addition, it was not clear how important the magnitude of capillary forces compared to self-weight forces was. In fact, it is possible that self-weight forces exclusively controlled vertical displacement, while the control of
capillary forces was limited to horizontal displacement. For the LNAPL infiltration
test into the dense sand reported by Esposito et al. [1999] (see Figure 2.41.a), the
horizontal migration appeared to be slightly more important at 20 g than that at 30 g,
thereby suggesting that perhaps capillary forces were not properly scaled. Obviously a
broader range of g-levels would be necessary before any conclusion can be drawn.

Figure 2.41. Motor oil (LNAPL) point source release on top of a column of sand
partially saturated with water [after Esposito et al., 1999]. Boundary lines of LNAPL
prototype plumes at termination (110 prototype days) of 20 g and 30 g tests: a. Dense
sand; b. Loose sand.

In their scaling analysis of multiphase flow in porous media, Petersen and Cooke
[1994] concluded that the geotechnical centrifuge was not a suitable tool for the
physical modeling of such process because of the existing conflict between similitude
at the microscopic and macroscopic scales. The issues at stake were later addressed by
Culligan and Barry [1998] who showed that similitude between a centrifuge model and
its prototype could be achieved if the controlling length of the problem was
macroscopic. In problems such as those investigated by Illangasekare et al. [1991] and
Knight and Mitchell [1996], the height of the LNAPL column above its front
constitutes a macroscopic length, such that it can be properly scaled in the centrifuge.
Conversely, in the problem of residual LNAPL entrapment in porous media [Ratnam,
1996; Ratnam et al., 1996b; Ratnam et al., 1996a], the appropriate controlling length is
the pore size, a microscopic length. Under these circumstances, the Bond number
[Morrow and McCaffery, 1978; Dawson and Roberts, 1997] does not scale properly
between model and prototype, and residual saturation is observed to decrease with g-
level [Ratnam et al., 1996b].
Figure 2.42. Butyl phthalate (DNAPL) point source release on top of a column of water-saturated silt-size/sand-size glass beads [after Pantazidou et al., 2000]. Relationship between prototype penetration depth of DNAPL front and prototype time: a. Scaled data of a series of centrifuge tests run on sand-size glass beads; b. Scaled data of two centrifuge tests run on glass beads of different sizes.
Studies of transport of DNAPLs into saturated granular media using centrifuge modeling have only been reported recently [Abu-Hassanein et al., 1998; Garnier et al., 2000b; Pantazidou et al., 2000]. Pantazidou et al. [2000] investigated the migration into water saturated silt-size and sand-size glass beads. The authors used two different DNAPLs: Freon 113, a DNAPL of high density and low viscosity with respect to those of water, and butyl phtalate, a DNAPL of low density and high viscosity. The displacement of Freon 113 was unstable with pronounced fingering. Butyl phtalate migration, on the other hand, was more stable with a smooth DNAPL front. Measured DNAPL entry pressures at the onset of infiltration into the sand were compared with those obtained at 1 g, and found to be in good agreement. Pantazidou et al. [2000] also examined the kinetics of the butyl phtalate infiltration. Modeling of models of DNAPL penetration depth was performed at 5 g and 15 g. As shown in Figure 2.42.a, the scaled penetration depths of the different experiments as a function of scaled time were found to be in good agreement, implying that both reduced-scale models represented the same equivalent prototype. Another series of tests showed good agreement of scaled DNAPL penetration data for two soils of different capillary pressure-saturation curves (see Figure 2.42.b), a result that was supported by numerical predictions [Pantazidou et al., 2000]. This suggests that beyond the early stages of infiltration into the granular medium, DNAPL vertical displacement may have been essentially driven by gravity forces with little influence of capillary forces.

There have been recent attempts to focus on defining the scaling laws of capillary rise under static and capillary conditions. Lord [1999] demonstrated the usefulness of the bundle of capillaries approach to examine flow in unsaturated soils. By using Hagen-Poiseuille’s law, the author showed that the flow time into capillary tubes centrifuged at a gravitational acceleration equal to n times the Earth’s gravity was reduced by a factor $n^2$ with respect to the flow time into capillary tubes run at 1 g. This result applied independently of the orientation of the capillary tube. Perfect wetting was assumed at all time. Nonetheless, the author did not attempt any experimental validation of his derivations.

Depountis et al. [2001] have attempted to examine the scaling laws of capillary rise in porous media [see also Garnier et al., 2000b]. Four different centrifuge facilities examined the rate of rise and equilibrium capillary height of water in poorly graded sand spun at different rotational accelerations ranging from 1 g to 40 g. The final capillary height, a macroscopic length, appeared to be scaled by a factor equal to the inverse of the g-level as predicted by theoretical analysis. Variability was important for some of the reported test series, and deviation was observed at higher gravitational fields. Results for the rate of rise were quite inconclusive, as can be seen in Figure 2.43. For some tests, the time to capillary equilibrium appeared to be well scaled by a factor $1/n^2$, where n is the g-level. Other test series showed very poor agreement, particularly at higher g-levels. Typically, a slower prototype rate of rise was observed at higher g-levels (see Figure 2.43). Depountis et al. [2001] attributed the observed differences to the effect of gravity on the shape of the air/water interface menisci, and inertia effects at higher g-levels. Further investigation was recommended using silt-size material in order to decrease the rate of rise and increase the equilibrium capillary height.
Figure 2.43. Scaled data of the capillary rise kinetics of water in columns of poorly graded sand spun at different g-levels, n (hc and t refer to the model capillary height and model time, respectively) [after Depountis et al., 2001]: a. Bochum Normsand; b Congleton sand.

In an unpublished study, Allersma [1996] examined capillary rise of water in sand columns under increasing gravity. After reaching equilibrium at 1 g, the height of the capillary fringe decreased during centrifuge spinning under the action of the increased g-level. At a g-level n = 9, the model capillary height was found to be more than one ninth of the 1-g capillary height. The difference was attributed to the wetting history of the sand and associated hysteresis. Parallels can be drawn with a capillary tube where the capillary height associated with a static advancing meniscus would be less than that of a static receding meniscus.
It is not obvious that contact angle hysteresis as well as the possible dependence of dynamic contact angle upon capillary number may be neglected when deriving the scaling laws linking a reduced-scale centrifuge model to its equivalent prototype. To date, there exists no study using the centrifuge that has demonstrated those effects or ruled out their influence.

2.5 References


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CHAPTER 3
THEORETICAL MODELING OF DNAPL INFILTRATION INTO A VERTICAL FRACTURE

3.1 Introduction

This chapter presents a series of theoretical model equations used to predict the infiltration behavior of pure-phase DNAPL into a vertical fracture initially saturated with water under hydrostatic conditions at the Earth’s gravitational acceleration. Theoretical modeling of DNAPL infiltration into any fracture system first requires expressing the criterion for DNAPL infiltration, i.e. the condition for which DNAPL will proceed to penetrate into the fracture by displacing water, or otherwise remain as a pool on top of the fracture. Modeling also requires giving a prediction of the kinetics of the DNAPL/water interface displacement once infiltration has begun, i.e. an expression of the interface location in the fracture as a function of time.

Section 3.2 presents an overview of the DNAPL infiltration problem and introduces an idealization of a rough-walled fracture system into smooth-walled fractures simulated by capillary tubes. The criterion for DNAPL infiltration is presented in Section 3.3 and is valid for both rough-walled and smooth-walled fractures.

The governing equations of the infiltration kinetics and some simplified solutions are respectively presented in Section 3.4 and Section 3.5, but are restricted to smooth-walled circular section capillary tubes. Section 3.5 first examines the solution where the dynamic contact angle, i.e. the contact angle of the moving interface, remains constant throughout the entire infiltration process. Next, the solution where the dynamic contact angle varies linearly with velocity is introduced and discussed. Finally, the solution where DNAPL and water have differing viscosities is presented.

Section 3.6 extends the solutions to smooth-walled rectangular section capillary tubes and rough-walled fractures where homogeneous hydraulic properties can be assigned. Although not presented in this thesis, the extension of the theoretical model to DNAPL infiltration in a fracture of any dip is straightforward.

A list of references made in this chapter is provided in Section 3.7.

The model equations obtained in this chapter are used to predict the outcome of series of laboratory DNAPL infiltration experiments in vertical capillary tubes that are described in Chapter 6. Comparison between the theoretical model predictions and experimental data is examined in Chapter 7.
In Chapter 4, the theoretical model is extended to the physical modeling of centrifuge experiments in order to derive the scaling laws connecting the centrifuge reduced-scale experiments to the behavior in the equivalent prototype.

3.2 Overview and Idealization of the Problem

Figure 3.1 illustrates, schematically, a pool of DNAPL on top of a vertical, water-saturated fracture system. Because, the fractures are rough-walled, their apertures are spatially variable at a small scale. At some larger scale, however, it is assumed that homogeneous hydraulic properties can be assigned to each fracture. These properties represent an average of the small scale properties [Chown et al., 1997].

The DNAPL pool is of height $h$ and no DNAPL has yet invaded the fracture system. As previously discussed in Section 2.2.1.2, a pressure discontinuity across the liquid interface results from the interfacial tension between the two fluid phases in the system. The interface is curved to maintain equilibrium of forces. Water preferentially wets the fracture plane such that there is an angle $\theta_s$ between the interface of the two fluids and the wall at their point of intersection. This angle referred to as the static contact angle (see Section 2.2.1.3 and Section 2.3.5.2).

If infiltration into the fracture system takes place, then DNAPL will start displacing water as it moves downwards. This is shown in Figure 3.2 for a single rough-walled vertical fracture. At time $t$, the DNAPL has penetrated at a depth $z_c(t)$ below the fracture entrance where the local fracture aperture is $e(z_c)$. The total length of the fracture is $l$. Note that Figure 3.2 makes the implicit assumption that the problem is two-dimensional, such that flow is macroscopically unidirectional.

When a planar fracture is assumed, the problem is identical in any plane parallel to the plane of the figure. In other words, considering the axis normal to the plane of Figure 3.2, the problem is independent of the associated transversal coordinates and can be solved using the remaining two rectangular coordinates.

Alternatively, if a circular, pinhole-like fracture is assumed, the problem is axisymmetric such that the fracture looks like a circular capillary tube of varying diameter. In this case, the problem can be solved using cylindrical polar coordinates, but remains two-dimensional.
Figure 3.1. DNAPL pool on top of a vertical fracture system.

Figure 3.2. Schematic of a single, vertical rough-walled fracture. DNAPL pool height is \( h \). Total fracture length is \( l \). At time \( t \), DNAPL has penetrated \( z_c(t) \) below the fracture entrance where the local fracture aperture is \( e(z_c) \). Fluid conditions in the fracture are hydrostatic.
The configuration shown in Figure 3.1 and Figure 3.2 can be idealized by modeling a rough-walled fracture as a smooth-walled vertical capillary tube of length $l$, as shown in Figure 3.3. This approach is often used in fracture flow to provide a theoretical justification to Darcy’s law [e.g., de Marsily, 1986; Bear, 1993]. For this idealization, two cases can be considered. First, the idealized fracture can be assumed circular in shape, which corresponds to a capillary tube of circular section having a diameter equal to the fracture maximum aperture $e$ and a radius $r_0$ equal to $e/2$. Alternatively, the idealized fracture can be assumed planar in shape, which corresponds to a capillary tube of rectangular section having an aperture $e$ and a width $w$ that can be assumed infinitely long with respect to $e$. These two types of section are shown in Figure 3.4. In either case, it is possible to identify a vertical axis of symmetry of the tube that is oriented downwards with an origin $O$ located at the center of the top section of the capillary tube, and with the coordinate $z$ describing distance along this axis (see Figure 3.3.b).

![Figure 3.3](image)

**Figure 3.3.** Schematics of DNAPL infiltration into an idealized fracture: a. Prior to infiltration; b. During infiltration.

In the idealized system shown in Figure 3.3, a reservoir tube, mounted on top of the capillary tube, models the DNAPL pool of height $h$ (see Figure 3.1 and Figure 3.2). The diameter of the reservoir tube is large enough with respect to the diameter (or aperture) of the capillary tube to allow the capillary effects associated with the reservoir tube to be neglected.

To mimic initial hydrostatic fluid conditions in the field, the reservoir tube/capillary tube system is submerged in water. A pressure discontinuity across the liquid interface results from interfacial tension between the two fluid phases in the
system. Again, there is a static contact angle of $\theta_s$ at the DNAPL/water interface and DNAPL is assumed to be the non-wetting fluid.

Clearly, the idealized system assumes that phenomena such as DNAPL mass transfer into the rock matrix itself can be neglected. Obviously, this will not be the case in some fracture systems [e.g., Parker et al., 1997] (see also Section 2.2.2). In the theoretical model presented in the following sections, any effects related to DNAPL dissolution into the water phase will be neglected. However, these effects will be present in the field and also the experiments presented in Chapter 6.

![Figure 3.4](image)

**Figure 3.4.** Capillary tube cross-section: a. Circular shape; b. Rectangular shape ($w \gg e$).

### 3.3 Infiltration Criterion

In the simplified configuration shown in Figure 3.3.a, the pressure discontinuity $p_e$ that is sustained between DNAPL and water at the entrance to a fracture idealized as a capillary tube is given by [Kueper and McWhorter, 1991]

$$p_e = \frac{2\varepsilon \sigma \cos \theta_s}{e}, \quad (3.1)$$

where $\sigma$ is the interfacial tension between water and DNAPL, and $\varepsilon$ is equal to 2 for circular-section capillary tubes (see Figure 3.4.a) and 1 for rectangular-section capillary tubes (see Figure 3.4.b). The pressure discontinuity is called the fracture entry pressure [Kueper and McWhorter, 1991].

Referring to Figure 3.2, in the more general case of rough-walled fractures, the pressure discontinuity, $p_e(z_c)$, at interface depth $z_c$, that can be sustained between DNAPL and water is given by [Longino and Kueper, 1999]

$$p_e(z_c) = \frac{2\varepsilon \sigma \cos \theta_s}{e(z_c)}, \quad (3.2)$$
where the parameter \( e(z_c) \) is defined as the local fracture aperture. The dimensionless parameter, \( e \), is similar to the parameter defined in (3.1), and equal to 1 for fractures of approximately planar cross-section, and 2 for fractures of approximately circular cross-section. Equation (3.2) makes the assumption that the interface is at rest and that there is no hysteresis associated with the contact angle (see discussion in Section 2.3.5.2).

In this work, \( p_e(z_c) \) will be referred to as the local entry pressure. For \( z_c = 0 \), \( p_e(0) \) is the fracture entry pressure, as defined by (3.1) for idealized systems.

DNAPL will infiltrate the fracture when the additional fluid pressure exerted by the presence of the DNAPL pool is equivalent to the fracture entry pressure. Therefore, the general criterion for DNAPL infiltration is

\[ \Delta \rho g h \geq p_e(0), \quad (3.3) \]

where \( \Delta \rho \) is the density contrast between DNAPL and water \( (\Delta \rho = \rho_{nw} - \rho_w) \).

Let \( h_i \) be defined as

\[ h_i = \frac{2 \varepsilon \sigma \cos \theta}{\Delta \rho g e(0)}. \quad (3.4) \]

If the DNAPL pool height, \( h \), exceeds the critical pool height, \( h_i \), then infiltration into the fracture takes place. The height \( h_i \) is sometimes referred to as the minimum pool height required for entry [Kueper and McWhorter, 1996]. Here, \( h_i \) will be termed the critical pool height or infiltration height.

### 3.4 Kinetics of Infiltration: Formulation of Governing Equations

#### 3.4.1 Statement of Problem

Throughout this section, it will be assumed that the DNAPL pool height, \( h \), exceeds the critical height, \( h_i \), and, consequently, that infiltration has taken place. The derivation that follows will be restricted to the case where the fracture is idealized as a capillary tube that is circular in shape with a radius \( r_0 = e/2 \) (see Figure 3.4.a). As will be shown in Section 3.6, the equations can be extended to the case of a capillary tube of rectangular section, and to a rough-walled fracture if homogeneous hydraulic properties can be assigned to the fracture.

The model will use a cylindrical coordinate system \( (r, \Psi, z) \) of origin \( O \) where, again, the vertical longitudinal axis \( (z) \) is oriented downwards and is the axis of symmetry of the capillary tube (see Figure 3.3.b). The radial distance \( r \) of any point is measured from the axis normal to \( (z) \) in a horizontal plane, and \( \Psi \) is the angle of
rotation about \((z)\). As discussed in Section 3.2, the problem is axisymmetric such that the flow conditions are independent of \(\Psi\). At each point inside the capillary tube, the radial and longitudinal fluid velocity components will be noted as \(v_j\) and \(u_j\), respectively. The subscript \(j\) corresponds to the fluid phase where the point is located, that is either \(nw\) in the DNAPL (non-wetting) phase or \(w\) in the water (wetting) phase.

As illustrated in Figure 3.3.b, \(z_c(t)\) is the distance from the center of the top section of the capillary tube (point \(O\)) to the tip of the DNAPL/water interface. Because the cross-section of the reservoir tube is large enough with respect to the cross-section of the capillary tube, it will be assumed that the reservoir pool height remains constant, and equal to \(h\), throughout the infiltration process. Note, however, that the equation could be extended to the case of a draining reservoir tube without excessive complication.

For incompressible fluids, the differential form of the equation of continuity can be written as [Munson et al., 1994]

\[
\frac{\partial u_j}{\partial z} + \frac{1}{r} \frac{\partial(r v_j)}{\partial r} = 0 \quad (j = nw, w), \tag{3.5}
\]

For Incompressible, Newtonian fluids under conditions where viscous forces are not negligible, the Navier-Stokes equations of motion are applicable. The Navier-Stokes equation for the longitudinal velocity \(u_j\) is given by [Munson et al., 1994]

\[
\frac{\partial u_j}{\partial t} + u_j \frac{\partial u_j}{\partial z} + v_j \frac{\partial u_j}{\partial r} = g - \frac{1}{\rho_j} \frac{\partial p_j}{\partial z} + \frac{\mu_j}{\rho_j} \left( \frac{\partial^2 u_j}{\partial r^2} + \frac{1}{r} \frac{\partial u_j}{\partial r} + \frac{\partial^2 u_j}{\partial z^2} \right) \tag{3.6}
\]

\((j = nw, w)\),

where \(p_j\), \(\rho_j\), and \(\mu_j\) are the pressure, density and dynamic viscosity of fluid \(j\), respectively. The Navier-Stokes equation for the radial velocity \(v_j\) is given by

\[
\frac{\partial v_j}{\partial t} + u_j \frac{\partial v_j}{\partial z} + v_j \frac{\partial v_j}{\partial r} = - \frac{1}{\rho_j} \frac{\partial p_j}{\partial r} + \frac{\mu_j}{\rho_j} \left( \frac{\partial^2 v_j}{\partial r^2} + \frac{1}{r} \frac{\partial v_j}{\partial r} - \frac{v_j}{r^2} + \frac{\partial^2 v_j}{\partial z^2} \right) \tag{3.7}
\]

\((j = nw, w)\).

Equations (3.5)-(3.7) represent a set of six governing equations corresponding to the six unknowns \(u_j\), \(v_j\) and \(p_j\) in the problem.
3.4.2 Dimensional Analysis of Governing Equations

A set of reference representative quantities for each variable \( u_j, v_j, p_j, r, z, \) and \( t \) is now introduced. Let \( u_0 \) be a reference longitudinal velocity, \( v_0 \) a reference radial velocity, \( p_0 \) a reference pressure, and \( t_0 \) a reference time. The radius, \( r_0 \), and a fractional length, \( z_0 \), of the capillary tube simulating the fracture are characteristic lengths of the problem and taken as reference lengths for \( r \) and \( z \), respectively. In practice, \( r_0 \) is of the order of one millimeter or less and \( z_0 \) is of the order of the measurement resolution of the system, which will be one centimeter or more (see Section 6.2.2). Hence, the ratio \( \delta = r_0/z_0 \) is very small with respect to unity.

Dimensionless variables \( u_j^*, v_j^*, p_j^*, r^*, z^*, \) and \( t^* \) are defined by

\[
\begin{align*}
  u_j^* &= u_j / u_0, \\
  v_j^* &= v_j / v_0, \\
  p_j^* &= p_j / p_0, \\
  r^* &= r / r_0, \\
  z^* &= z / z_0, \\
  t^* &= t / t_0
\end{align*}
\]  

(3.8)

The equation of continuity (3.5) is rewritten in terms of the dimensionless variables defined by (3.8), leading to

\[
\frac{\partial u_j^*}{\partial z^*} + \left( \frac{v_j^*}{u_j^*} \right) \frac{1}{\delta u_0} \frac{\partial (r^* v_j^*)}{\partial r^*} = 0 \quad (j = nw, w).
\]  

(3.9)

Since both terms in (3.9) must be of the same order of magnitude [Li and Lam, 1964]

\[
    v_{0,j} = \delta u_{0,j} \quad (j = nw, w).
\]  

(3.10)

Equation (3.10) shows that the radial velocity is negligible compared to the longitudinal velocity.

The Navier-Stokes equations (3.6) and (3.7) can also be rewritten in their dimensionless form. Making use of (3.8) and (3.10) leads to

\[
\begin{align*}
  \left( \frac{u_{0,j}}{t_0} \right) \frac{\partial u_j^*}{\partial t^*} + \left( \frac{u_{0,j}^2}{z_0} \right) & \left[ u_j^* \frac{\partial u_j^*}{\partial z^*} + v_j^* \frac{\partial u_j^*}{\partial r^*} \right] = \frac{\partial p_j^*}{\partial z^*} + \left( \frac{\mu}{\rho_j z_0} \right) \frac{\partial^2 u_j^*}{\partial r^*^2} + \left( \frac{1}{\rho_j t_0^2} \right) \left[ \left( \frac{\partial^2 u_j^*}{\partial r^*^2} + \frac{1}{r^*} \frac{\partial u_j^*}{\partial r^*} \right) + \delta \frac{\partial^2 u_j^*}{\partial z^*^2} \right] \\
  \end{align*}
\]  

(3.11)

for the longitudinal component of velocity, and
\[
\frac{\left( \delta u_{0j} \right)}{t_{0}} \frac{\partial v_{j}}{\partial t^*} + \left( \delta u_{0j}^2 \right) \left( u_{j}^* \frac{\partial v_{j}}{\partial z^*} + v_{j}^* \frac{\partial v_{j}}{\partial r^*} \right) = - \left( p_{0j} \right) \frac{\partial p_{j}}{\partial r^*} + \left( \delta \mu_{j} u_{0j} \right) \left[ \left( \frac{\partial^2 v_{j}}{\partial r^*^2} \right) + \frac{1}{r^*} \frac{\partial v_{j}}{\partial r^*} - \frac{v_{j}}{r^*} \left( \frac{\partial^2 v_{j}}{\partial r^*^2} \right) \right] + \delta^2 \frac{\partial^2 v_{j}}{\partial z^*^2} \right] \quad (j = nw, w),
\]

for the radial component of velocity.

Given that \( \delta << 1 \), a number of terms in (3.11) and (3.12) can be neglected, leading to the following simplifications for (3.11) and (3.12), respectively

\[
\frac{\left( u_{0j} \right)}{t_{0}} \frac{\partial u_{j}}{\partial t^*} + \left( u_{0j}^2 \right) \left( u_{j}^* \frac{\partial u_{j}}{\partial z^*} + v_{j}^* \frac{\partial u_{j}}{\partial r^*} \right) = \left( p_{0j} \right) \frac{\partial p_{j}}{\partial z^*} + \left( \mu_{j} u_{0j} \right) \left[ \frac{\partial^2 u_{j}}{\partial r^*^2} \right] + \frac{1}{r^*} \frac{\partial u_{j}}{\partial r^*} \quad (j = nw, w),
\]

and

\[
\frac{\left( p_{0j} \right)}{\delta \rho_{j} z_{0}} \frac{\partial p_{j}}{\partial r^*} = 0 \quad (j = nw, w).
\]

Equation (3.14) shows that if the condition \( \delta << 1 \) is true, then the pressure field is a constant throughout a given section of a capillary tube and is thus only a function of \( z \).

Referring to (3.13), the two terms on the left-hand side involve a time derivative \( \partial u_{j}^*/\partial t^* \) and two spatial derivatives \( \partial u_{j}^*/\partial r^* \) and \( \partial u_{j}^*/\partial z^* \), respectively. The time derivative term is the local acceleration or local inertia of the problem. It describes the change of flow conditions at a fixed point in the system and is zero if the flow is steady. The spatial derivative term is the convective acceleration or convective inertia of the problem [Munson et al., 1994]. It describes the change in flow conditions when a particle moves from one point to another in the system. It is not necessarily zero under steady conditions. However, it is zero if a particle moves from one point to another such that the flow conditions are identical at both points even if they simultaneously change with time. For example, Hagen-Poiseuille’s flow in a capillary tube has a parabolic velocity distribution. Under the configuration shown in Figure 3.5, particles move from position (1) to position (2) along a path that is parallel to the axis of the tube. The flow conditions between two points along this pathway are the same, and thus the convective inertia is zero. Because the capillary tube is curved past (2), the flow conditions change between (2), (3) and (4) even if the flow is steady \( \partial/\partial t = 0 \). Thus, the convective inertia in the region (2)-(4) differs from zero.
Figure 3.5. Hagen-Poiseuille’s flow conditions in a capillary tube with a bend [adapted from Munson et al., 1994]: 1 and 2, fully developed Hagen-Poiseuille’s flow prior to bend; 3, developing flow past bend; 4, fully developed Hagen-Poiseuille’s flow past bend.

Referring to the DNAPL infiltration problem, convective inertia is not a priori zero for this case. As illustrated in Figure 3.6, the flow conditions near the entrance to a fracture simulated by a capillary tube (1) can be different from the flow conditions further away from the entrance of the tube, where parabolic flow (3) can usually be assumed. Thus, the convective inertia might not be negligible at the entrance but would be further away. It is important to understand what further away from the entrance means in the context of the DNAPL infiltration problem.

Figure 3.6. Flow conditions in a capillary tube near its entrance [after Munson et al., 1994]: 1, entrance region flow; 2, developing flow; 3, fully developed Hagen-Poiseuille’s flow.

Examining (3.13), convective inertia is negligible with respect to the viscous forces—which are given by the last term of the right-hand side of (3.13)—if the following condition is true.
Simplifying (3.15) leads to

\[
\frac{\rho_j f_0^2 u_{0j}}{\mu_j z_0} \ll 1 \quad (j = nw, w). \tag{3.16}
\]

The left-hand side of (3.16) is similar to a Reynolds number, as defined by (2.10) (see Section 2.3.2). Equation (3.16) expresses the conditions under which convective inertia can be ignored in a solution of the DNAPL infiltration problem. Clearly, it will be necessary to check that this condition is true if convective inertia is assumed negligible at any point in an analysis.

### 3.4.3 Integral Form of Governing Equations

To solve the governing equations, the Kármán-Pohlhausen momentum integral technique is used [Li and Lam, 1964]. In this method, a reasonable shape for the velocity profile is assumed (here, a parabolic profile, see Section 3.4.4), and the laws of conservation of mass and momentum are satisfied as a whole. This approach is used by Levine et al. [1976] in their study of rise of the rate of penetration of a liquid in a vertical capillary tube (see Section 2.3.4.3).

The objective is to obtain an integral form of the governing equations for the DNAPL infiltration problem shown in Figure 3.3.b. To do so, the governing equations are integrated over a section of capillary tube. The result is then integrated over the length of the finger of DNAPL or the finger of water, depending upon which phase is under consideration.

On multiplying (3.6) by \( r \) and using (3.5), it may be verified that

\[
\frac{\partial (ru_j)}{\partial t} + \frac{\partial (ru_j^2)}{\partial z} + \frac{\partial (ru_j v_j)}{\partial r} = gr \frac{r}{\rho_j} \frac{\partial p_j}{\partial z} + \frac{\mu_j}{\rho_j} \left[ \frac{\partial}{\partial r} \left( r \frac{\partial u_j}{\partial r} \right) + r \frac{\partial^2 u_j}{\partial z^2} \right] \quad (j = nw, w). \tag{3.17}
\]

Integrating (3.17) with respect to \( r \) across a horizontal cross-section of the tube from 0 to \( r_0 \) and assuming the (no-slip) boundary condition

\[
u_j = v_j = 0 \quad \text{at} \ r = r_0 \quad (j = nw, w), \tag{3.18}
\]

gives
\[
\frac{\partial}{\partial t} \int_0^r r u_j dr + \frac{\partial}{\partial z} \int_0^r r u_j^2 dr = \frac{1}{2} g r_0 \rho_{j} - \frac{1}{\rho_{j}} \int_0^r r \frac{\partial p_j}{\partial z} dr + \frac{\mu_{j}}{\rho_{j}} r_0 \frac{\partial u_j}{\partial r} \bigg|_{r=r_0} + \frac{\mu_{j}}{\rho_{j}} \frac{\partial^2}{\partial z^2} \int_0^r r u_j dr \\
\hspace{2cm} (j = nw, w).
\] (3.19)

Multiplying (3.5) by \(r\), integrating with respect to \(r\) from 0 to \(r_0\), and making use of the boundary condition (3.18) leads to

\[
\frac{\partial}{\partial z} \left( \int_0^r r u_j dr \right) = 0 \hspace{1cm} (j = nw, w).
\] (3.20)

Equation (3.20) means that the volume flux across a section of the tube is independent of the longitudinal coordinate \(z\). The volume flux is equal to the volume increase of DNAPL in the capillary tube, i.e.

\[
2\pi \int_0^r r u_j dr = \pi r_0^2 \frac{dz_c}{dt} \hspace{1cm} (j = nw, w).
\] (3.21)

The velocity \(dz_c/dt\) is the displacement velocity of the DNAPL/water meniscus. Thus, using (3.20) and (3.21), (3.19) can be simplified to yield

\[
\frac{1}{2} r_0^2 \frac{d^2 z_c}{dt^2} + \frac{\partial}{\partial z} \int_0^r r u_j^2 dr = \frac{1}{2} g r_0 \rho_{j} - \frac{1}{\rho_{j}} \int_0^r r \frac{\partial p_j}{\partial z} dr + \frac{\mu_{j}}{\rho_{j}} r_0 \frac{\partial u_j}{\partial r} \bigg|_{r=r_0} \hspace{1cm} (j = nw, w).
\] (3.22)

Integrating (3.22) with respect to \(z\) from 0 to \(z_c(t)\) for the DNAPL phase leads to

\[
\frac{1}{2} r_0^2 z_c(t) \frac{d^2 z_c}{dt^2} + \left[ \int_0^r r u_m^2 dr \right]_{z=0}^{z=z_c(t)} = \frac{1}{2} g r_0^2 z_c(t) - \frac{1}{\rho_{nw}} \int_0^r r [p_{nw}(z_c, r, t) - p_{nw}(0, r, t)] dr + \frac{\mu_{nw}}{\rho_{nw}} r_0 \int_0^{z_c(t)} \frac{\partial u_m}{\partial r} \bigg|_{r=r_0} dz.
\] (3.23)

Integrating (3.22) with respect to \(z\) from \(z_c(t)\) to \(l\) for the water phase gives

\[
\frac{1}{2} r_0^2 [l - z_c(t)] \frac{d^2 z_c}{dt^2} + \left[ \int_0^r r u_m^2 dr \right]_{z=z_c(t)}^{z=l} = \frac{1}{2} g r_0^2 [l - z_c(t)] - \frac{1}{\rho_{w}} \int_0^r r [p_{w}(l, r, t) - p_{w}(z_c, r, t)] dr + \frac{\mu_{w}}{\rho_{w}} r_0 \int_{z_c(t)}^{l} \frac{\partial u_m}{\partial r} \bigg|_{r=r_0} dz.
\] (3.24)
Equations (3.21), (3.23) and (3.24) are the integral forms of four out of the six governing equations. The two remaining equations are the radial component of the momentum for each fluid phase, i.e. (3.7). So far, no assumption has been made regarding flow conditions, and the integral equations derived are not approximations. In the section that follows, some assumptions are introduced that lead to approximate solutions for (3.23) and (3.24).

### 3.4.4 Parabolic Flow Approximation

To simplify the relationships given by (3.23) and (3.24), it is assumed for each phase \( j \) that the longitudinal velocity has a parabolic profile (Hagen-Poiseuille’s flow), independent of \( z \) and given by

\[
\begin{align*}
    u_j(r,t) &= 2u_{0j}(t) \left(1 - \frac{r^2}{r_0^2}\right) \\
    &= \begin{cases} 
      0 & (j = nw, w). 
    \end{cases}
\end{align*}
\]

(3.25)

where \( u_{0j} \) is the average longitudinal velocity of fluid \( j \) through a section of capillary tube. As noted in Section 2.3.3.1, although many capillary flow models rely on this assumption, parabolic profiles do not exist in the region of entry into the capillary tubes and near the advancing meniscus.

Hagen-Poiseuille’s flow inherently assumes that the direction of flow is parallel to the axis of the tube. Indeed, upon replacing (3.25) in the continuity equation (3.5), it can be shown that the radial velocity \( v_j \) is zero. Because \( u_j \) is independent of \( z \) and \( v_j \) is zero, it can be concluded from (3.6) that the convective inertia is zero. However, recalling the discussion made in Section 3.4.2, it will be necessary to check that the condition given by (3.16) is correct in order to validate the approximation of a parabolic velocity profile.

Having shown that the radial velocity \( u_j \) is zero, (3.7) is reduced to

\[
\frac{\partial p_j}{\partial r} = 0 \quad (j = nw, w),
\]

(3.26)

which shows that \( p(z, r, t) \) is independent of \( r \).

The velocity profile (3.25) verifies the integral form of the mass conservation equation (3.20) and is therefore a suitable approximation. Using (3.21), it can be shown that

\[
    u_{0j}(t) = \frac{dz_c}{dt} \quad (j = nw, w).
\]

(3.27)

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Using (3.25), (3.26) and (3.27), the integral forms of the momentum equation (3.23) and (3.24) can respectively be simplified to

\[ p_{nw}(z_c, t) - p_{nw}(0, t) = \left( \rho_{nw}g - \frac{8\mu_{nw}}{r_0^2} \frac{dz_c}{dt} - \rho_{nw} \frac{d^2z_c}{dt^2} \right) z_c(t), \]  
\[ (3.28) \]

and

\[ p_w(l, t) - p_w(z_c, t) = \left( \rho_wg - \frac{8\mu_w}{r_0^2} \frac{dz_c}{dt} - \rho_{nw} \frac{d^2z_c}{dt^2} \right) [l - z_c(t)]. \]  
\[ (3.29) \]

By adding (3.28) and (3.29), the pressure discontinuity across the DNAPL/water interface, \( p_{nw}(z_c, t) - p_w(z_c, t) \), can be expressed as

\[ p_{nw}(z_c, t) - p_w(z_c, t) = -[\rho_wl + \Delta\rho z_c(t)] \left( \frac{d^2z_c}{dt^2} \right) \]  
\[ -\frac{8}{r_0^2}[\mu_wl + \Delta\mu z_c(t)] \frac{dz_c}{dt} + \rho_wgl + \Delta\rho g z_c(t) + p_{nw}(0, t) - p_w(l, t), \]  
\[ (3.30) \]

where \( \Delta\mu \) is the viscosity contrast between DNAPL and water (\( \Delta\mu = \mu_{nw} - \mu_w \)), and \( \Delta\rho \) is, again, the density contrast between DNAPL and water. Equation (3.30) is the equation predicting the interface displacement \( z_c(t) \) under the assumption of parabolic flow. The following section seeks a set of boundary and initial conditions to apply to (3.30).

### 3.4.5 Boundary and Initial Conditions

Prior to solving for \( z_c(t) \), three boundary conditions are needed, i.e. the values of the functions \( p_{nw}(z_c, t) - p_w(z_c, t) \), \( p_{nw}(0, t) \), as well as \( p_w(l, t) \). Section 2.3.4 examined prior research work on the entrance flow into capillary tubes and on the spontaneous penetration into capillary tubes. This section concluded that, at the entrance to the capillary tube, the total drag force was a combination of the Hagenbach and Couette corrections as well as the effect of the reservoir inertia. The total drag force can be written as (see Section 2.3.4)

\[ f_{cd} = \frac{1}{2} \left( \frac{m + m'}{\text{Re}} \right) \rho_0 r_0^2 \left( \frac{dz_c}{dt} \right)^2 + \lambda \rho_0 r_0^4 \frac{d^2z_c}{dt^2}, \]  
\[ (3.31) \]
where \( \frac{dz_c}{dt} \) is the velocity of rise of the meniscus, Re is the Reynolds number as defined by (2.10) (see Section 2.3.2), \( r_0 \) is the radius of the capillary tube, \( \rho \) is the density of the rising fluid, \( m \) is a number ranging from 2.24 to 2.41, \( m' \) is a number of the order of 100 and \( \lambda \) is a number of the order of 1 (see Section 2.3.4). Using (3.31) and (2.10), the pressure \( p_{nw}(0, t) \) can be expressed as

\[
p_{nw}(0, t) = \rho_w g h_{wp} + \rho_{nw} gh - \frac{1}{2} m \rho_{nw} \left( \frac{dz_c}{dt} \right)^2 - \frac{\mu_{nw} m'}{4 r_0} \frac{dz_c}{dt} - \lambda \rho r_0 \frac{d^2 z_c}{dt^2},
\]

(3.32)

where \( h_{wp} \) is the distance from the phreatic surface to the top of the DNAPL pool (see Figure 3.3.b).

Replacing (3.32) in (3.30) leads to

\[
p_{nw}(z_c, t) - p_{w}(z_c, t) = -\left[ \rho_w l + \Delta \rho z_c(t) + \lambda \rho r_0' \right] \left( \frac{d^2 z_c}{dt^2} \right)
\]

\[-\frac{8}{r_0^2} \left[ \mu_w l + \Delta \mu z_c(t) + \mu_{nw} m'r_0 / 32 \right] \frac{dz_c}{dt} + \rho_w gl \]

(3.33)

\[+\Delta \rho g z_c(t) + \rho_w g h_{wp} + \rho_{nw} gh - p_{w}(l, t) - \frac{1}{2} m \rho_{nw} \left( \frac{dz_c}{dt} \right)^2.
\]

Given the orders of magnitude of \( m' \) and \( \lambda \) as well as the fact that \( r_0 << l \), it can be seen from (3.33) that the Couette correction and the correction due to reservoir inertia are not significant compared, respectively, to the inertia forces and the viscous forces. Therefore, these corrections will be ignored in further development. Nonetheless, the last term of (3.33), i.e. the Hagenbach correction, may be significant.

Next, an expression for \( p_{w}(l, t) \) is needed. Typically, the head loss due to a sudden expansion of infinite size compared to the capillary tube entirely dissipates the kinetic energy of the jet exiting the capillary tube, that is \( \rho_w (dz_c/dt)^2/2 \) [Munson et al., 1994]. Thus, the pressure \( p_{w}(l, t) \) can be taken as the static pressure right outside the capillary tube in a region where the fluid is at rest. This assumption ignores the inertia effect, similar to the reservoir inertia effect, associated with the fact that the jet velocity \( dz_c/dt \) changes with time. The pressure \( p_{w}(l, t) \) is given by

\[
p_{w}(l, t) = \rho_w g (h_{wp} + h + l).
\]

(3.34)

Replacing (3.34) in (3.33) gives
Finally, the expression $p_{nw}(z_c, t) - p_w(z_c, t)$ can be linked to the dynamic contact angle at the DNAPL/water interface as [Blake and Haynes, 1969]

$$p_{nw}(z_c, t) - p_w(z_c, t) = \frac{2\sigma \cos \theta_d \left( \frac{dz_c}{dt} \right)}{r_0},$$  

(3.36)

where $\theta_d(dz_c/dt)$ is the dynamic contact angle. As seen in Section 2.3.6, the dynamic contact angle is a function of the interface velocity and can differ from the static contact angle $\theta_s$ introduced in Section 3.2. Replacing (3.36) in (3.35) gives

$$\left[ \rho_w l + \Delta \rho z_c(t) \right] \left( \frac{d^2 z_c}{dt^2} \right) + \frac{8}{r_0^2} \left[ \mu_w l + \Delta \mu z_c(t) \right] \frac{dz_c}{dt} = \Delta \rho g[z_c(t) + h] - \frac{1}{2} \frac{m \rho_{nw}}{r_0 \rho_0} \left( \frac{dz_c}{dt} \right)^2,$$

(3.37)

where

$$\Delta h(dz_c/ dt) = h - \frac{2\sigma \cos \theta_d (dz_c/ dt)}{\Delta \rho g r_0}.$$

(3.38)

Equation (3.37) must be solved with the initial conditions

$$z_c = 0, \quad \frac{dz_c}{dt} = 0 \quad \text{at} \quad t = 0.$$

(3.39)

The solution of the non-linear, second-order equation (3.37) with the initial conditions (3.39) predicts the DNAPL/water interface displacement during infiltration into a vertical capillary tube of circular section. Equation (3.37) can be extended to a capillary tube of any inclination with the horizontal, by simply modifying the gravity term. It is analogous to the equation proposed by Mumley et al. [1986] for their liquid/liquid rise in capillary tubes.

It is possible to derive (3.30) and (3.37) using the macroscopic energy balance method as given by Bird et al. [1960] in lieu of the Navier-Stokes equations. As pointed out in Section 2.3.4.2, this method has been used by several researchers to derive the rate of rise of liquids in capillary tubes and porous media [Szekely et al.,
Again, a velocity profile must be assumed in order to calculate the different fluxes and kinetic energy of the system. One energy balance equation can be derived for each of the two fluid phases contained in the capillary tube. The resulting two equations can be combined to yield (3.30) and (3.37). The drag force at the entry to the capillary tube can be incorporated in the energy dissipation term.

3.5 Kinetics of Infiltration: Simplified Solutions of Governing Equations

3.5.1 Overview and Complementary Assumptions

Because of its non-linearity, (3.37) cannot be solved analytically. However, it is possible to obtain an analytical solution to this equation if complementary assumptions are made. This section lists three possible assumptions.

A first assumption consists in neglecting the drag force at the entry to the capillary tube. A \( \beta \)-number, which is a ratio of drag forces to viscous forces, can be defined by

\[
\beta = \frac{m \rho_{nw} \frac{d z}{dt}}{8 \mu_w l}.
\] (3.40)

If the \( \beta \)-number is very small with respect to unity, then the drag forces can be neglected. Consider for example a 0.1 m long, 1.33 mm diameter capillary tube and infiltration of 4-chlorotoluene (4-CT) DNAPL of density 1070 kg/m\(^3\). Assuming \( m = 2.35 \) (which is the mid-point of the range proposed in Section 2.3.4.1) and given that \( \mu_w = 10^{-3} \) Pa.s at 20\(^\circ\)C, the \( \beta \)-value will be less than 0.1 as long as the interface velocity does not exceed 0.14 m/s. There may be instances where the drag forces cannot be neglected, particularly for larger diameter capillary tubes. Thus, for each infiltration experiment, it will be necessary to calculate the value of \( \beta \), and verify if the simplified equations apply in this case.

A second simplifying assumption consists in neglecting the density contrast \( \Delta \rho \) between DNAPL and water with respect to the density of water \( \rho_{nw} \), and their viscosity contrast \( \Delta \mu \) with respect to the viscosity of water \( \mu_w \). As will seen in Section 5.5, this assumption is valid for 4-CT whose density and viscosity are close to the density and viscosity of water, respectively. Note that an alternative is to average the density and the viscosity of the liquids.
The third assumption has to do with the contact angle. As noted in Section 2.3.6.5, there is no simple model that can fully predict the dependence of the liquid/liquid interface dynamic contact angle upon interface velocity. In order to get an analytical solution, a common approach used by researchers is to assume that the dynamic contact angle remains constant and equal to the static contact angle throughout the infiltration process.

A solution to the infiltration problem with all three assumptions is presented and discussed in Section 3.5.2. The effects of the dependence of contact angle upon interface velocity are examined using an alternative treatment described in Section 3.5.3. Finally, Section 3.5.4 presents an analytical solution to the infiltration problem when the DNAPL viscosity significantly differs from that of water.

### 3.5.2 Analytical Solution Based on Assumption of Constant Contact Angle

#### 3.5.2.1 Solution of Governing Equation and Discussion

Based on all the complementary assumptions made in Section 3.5.1, (3.37) can be written as a second order differential equation with constant coefficient, as

\[
\frac{d^2z_c}{dt^2} + \frac{g}{k_w} \frac{dz_c}{dt} = \frac{k_\Lambda}{k_w} \frac{g}{l} [z_c(t) + \Delta h],
\]

where

\[
k_w = \frac{\rho_w g r_0^2}{8 \mu_w},
\]

\[
k_\Lambda = \frac{\Delta \rho g r_0^2}{8 \mu_w},
\]

and, under the assumption of constant dynamic contact angle, equal to the static contact angle,

\[
\Delta h = h - h_i,
\]

where \(h_i\) is the critical height defined by (3.4). The parameter \(k_w\) has the dimensions of a velocity and corresponds to the hydraulic conductivity of water in a capillary tube of radius \(r_0\) \cite{deMarsily1986}. The parameter \(k_\Lambda\) has also the dimensions of a velocity,
and corresponds to the equivalent conductivity of a fluid of density $\Delta \rho$ and viscosity $\mu_w$.

Equation (3.41) can be solved analytically using the initial condition given by (3.39) and leading to the following expression of the interface location, $z_c$, as a function of time $t$:

$$z_c(t) = \Delta h \left[-1 + \frac{1}{2\sqrt{1+\alpha}} \left(-1 + \sqrt{1+\alpha}\right) \exp\left(-\frac{g}{2k_w} (1 + \sqrt{1+\alpha}) t\right) + (1 + \sqrt{1+\alpha}) \exp\left(\frac{g}{2k_w} (1 + \sqrt{1+\alpha}) t\right)\right],$$  

(3.45)

where $\alpha$ is defined by

$$\alpha = \frac{4k_s k_w}{g l} = \frac{\Delta \rho \mu_w g r_0^2}{16 \mu_w^2 l}. \quad (3.46)$$

It can be shown that if $\alpha$ is small, then the solution given by (3.45) reduces to

$$z_c(t) = \Delta h \left[-1 + \exp\left(\frac{k_s t}{l}\right)\right], \quad (3.47)$$

which is also the solution of (3.41) with no second order term, i.e.

$$\frac{dz_c}{dt} = \frac{k_s}{l} \left[z_c(t) + \Delta h\right]. \quad (3.48)$$

The first-order equation given by (3.48) is similar to (2.4) developed by both Lucas [1918] and Washburn [1921] (see Section 2.3.2). In contrast, (3.41) incorporates a second order term due to local inertia forces. This equation is similar to the model developed by Rideal [1922] (see (2.12) in Section 2.3.3.1). Therefore, the $\alpha$-number can be seen as a parameter describing the magnitude of local inertia forces associated with the system. The higher the magnitude of $\alpha$, the larger the local inertia forces. Conversely, local inertia can be ignored when $\alpha \ll 1$.

To derive the infiltration kinetics governing equation (3.41), its associated solution (3.45) and simplified solution (3.47), the magnitude of convective inertia forces has been ignored. As discussed in the dimensional analysis of Section 3.4.2, it can be expected that the assumption of flow parallel to the axis of the capillary tube will no longer hold true below a certain capillary tube length. Therefore, it is necessary to check under which condition infiltration kinetics solution (3.45) and simplified solution (3.47) are valid.
To ensure that convective inertia forces are negligible, condition (3.16) must be met. The condition requires a value of the reference longitudinal velocity, $u_0$, which cannot be determined without first examining the governing equations (3.45) and (3.47) in more details. As will be shown in Section 3.5.2.4, condition (3.16) reduces to the condition $\alpha \ll 1$ where $\alpha$ is given by (3.46). In other words, whenever local inertia forces cannot be neglected, convective inertia forces cannot be neglected either. This conclusion is important because it implies that the infiltration kinetics solution (3.45) is always restricted to the case where $\alpha \ll 1$ and thus always reduces to the simplified solution (3.47). Use of the full solution (3.45) where the condition $\alpha \ll 1$ is not met can never be applied to the DNAPL infiltration problem because it does not properly account for convective inertia forces. If, on the other hand, the condition $\alpha \ll 1$ is insured, then both local and convective inertia forces can be neglected with respect to viscous forces. Under these conditions, the simplified governing equation (3.48) and solution (3.47) are valid, and may be used for predicting the interface displacement. Experiments supporting these conclusions are presented in Chapter 7.

In the remainder of this thesis, (3.47) and (3.48) will be referred to as the negligible inertia constant contact angle model (NICCA model). Section 3.5.2.2 further examines the NICCA model. Section 3.5.2.3 focuses on the limit of the NICCA model for short tubes. Section 3.5.2.4 demonstrates as stated above that the negligible convective inertia assumption obeys the same condition $\alpha \ll 1$ as that of negligible local inertia. Finally, Section 3.5.2.5 examines the case where a DNAPL finger has pre-infiltrated the capillary tube, but is immobilized at a certain depth, such that the total height of DNAPL—reservoir pool plus finger length—is still less than the infiltration height.

### 3.5.2.2 The NICCA Model

As shown by (3.44), $\Delta h$ is the difference between the DNAPL pool height and the pool critical height for which infiltration takes place. In the configuration shown in Figure 3.3.a, infiltration takes place as soon as the critical height, $h_i$, is exceeded. Therefore, $\Delta h$ is very small in comparison to $h_i$. If $l$ and $z_c$ are of orders equal to, or larger than $h_i$, then (3.48) is reduced to

$$\frac{dz_c}{dt} = k_\Delta \frac{z_c(t)}{l}.$$  \hspace{1cm} (3.49)

Equation (3.49) shows that, for asymptotic behavior in the simplified fracture system, i.e. as $z_c(t)$ approaches $l$, the velocity of the interface is not controlled by the absolute position of the interface, but by its position relative to the end of the capillary tube. This fact is important and implies that the exit velocity is independent of the length of the tube, and equal to $k_\Delta$. In other words, beyond a certain depth, the interface displacement is controlled by gravity and viscous forces alone. Thus, if (3.48) is
correct, the interfacial tension forces, expressed in the term $\Delta h$, only influence the early stage of the interface displacement.

![Diagram](image)

**Figure 3.7.** Theoretical effect of $\Delta h$ on the infiltration of DNAPL in vertical capillary tubes: a. Interface depth versus time; b. Interface depth versus interface velocity.

As an illustration, consider a 300 mm long, 1.33 mm vertical capillary tube. The tube is invaded by a DNAPL, for which the corresponding critical height is 150 mm, and the equivalent conductivity, $k_\Delta$, is 40 mm/s. Calculation of the hydraulic conductivity $k_w$ yields 541 mm/s using (3.42). The $\alpha$-number is equal to 0.029 using (3.46), and the $\beta$-number evaluated at $dz/dt = k_\Delta$ is equal to $9.3 \times 10^{-3}$ using (3.40) with $m = 2.35$. Both numbers indicate that inertia and drag forces can be neglected. In Figure 3.7.a, the interface displacement versus time has been plotted for different values of $\Delta h$. As can be seen, all three displacement profiles have the same asymptotic behavior, and would almost match by simple horizontal translation if one were to consider only the interface displacement beyond a depth of approximately 70 mm. This can be better understood by looking at Figure 3.7.b, which shows a plot of the interface depth versus its velocity. There appears to be a small influence on the interface velocity for $\Delta h = 0.1 h_i$ because $\Delta h$ is large enough compared to the length scales of interest, i.e. $h_i$ and $l$. However, little difference in velocity profile can be seen between the cases $\Delta h = 0.01 h_i$ and $\Delta h = 0.001 h_i$ where $\Delta h$ is too small compared to $h_i$ and $l$ to have any incidence on the velocity field. Thus, past the early stages of infiltration where capillary forces dominate, the asymptotic displacement profile tends to be the same for all three tubes.
When conducting a spontaneous infiltration laboratory experiment, DNAPL may be added by increments in the reservoir until infiltration takes place (see Section 6.2.2). Therefore, $\Delta h$ is related to the volume of DNAPL introduced in a single increment, which is controlled, and therefore can be made very small. Nonetheless, experience has shown that the exact measurement of $\Delta h$ is virtually impossible without large uncertainties. Under these circumstances, it is better to compare the asymptotic behavior of measured DNAPL/water interface displacements with the theoretical model, and arrive at some conclusion about the system behavior in this manner. This point is further discussed in Section 7.2.2.

In the remainder of this thesis, a plot of the interface depth versus time, such as that shown in Figure 3.7.a, is referred to as infiltration profile. A plot of the interface depth versus interface velocity, such as that shown in Figure 3.7.b, is referred to as velocity field.

3.5.2.3 Limit of the NICCA Model for Short Tubes

Using (3.49), it was shown that the exit velocity is independent of the capillary tube length, and equal to $k_\Delta$. When the overall length of the capillary tube decreases, it clearly takes more acceleration to achieve the same exit velocity. Consequently, the shorter the tube, the larger the inertia force. This explains why the $\alpha$-number, as given by (3.46), is inversely proportional to $l$. Below a certain capillary tube length, both local and convective inertia forces are not negligible, and the approximation offered by (3.48) is therefore no longer valid.

To illustrate this point, consider the infiltration of DNAPL into four 2.70 mm diameter vertical capillary tubes of varying lengths such that the $\alpha$-number takes a different value for each tube and is equal to 0.01, 0.1, 1 and 2, respectively. The values of the hydraulic conductivity, $k_w$, and equivalent conductivity, $k_\Delta$, are equal to 2.23 m/s and 0.16 m/s, respectively. For the purpose of illustration, drag forces ($\beta << 1$) are ignored, as well as the effect of convective inertia forces, which, again, are not accounted for in (3.45). Thus, due to drag and convective inertia forces, it can be expected that the displacement of the interface for shorter tubes will be even slower than that predicted here.

Figure 3.8.a shows a plot of the relative depth $z_c(t)/l$ versus the normalized time $k_\Delta t/l$ using the solution given by the NICCA model, i.e. (3.47). Because $\Delta h$ is chosen such that $\Delta h/l$ remains constant, the normalized curve representing (3.47) (solid line) is independent of $l$ and identical for the four capillary tubes, as shown by

$$\frac{z_c(t)}{l} = \frac{\Delta h}{l} \left[ -1 + \exp \left( \frac{k_\Delta t}{l} \right) \right],$$

which is the normalized form of (3.47).
On the same graph, for each capillary tube, the relative depth \( z_c(t)/l \) is plotted versus the normalized time using (3.45). Again, the normalized time is defined by \( k_\Delta t/l \). Four distinct plots are obtained corresponding to the four capillary tubes (see Figure 3.8.a). As can be seen in the figure, the plot associated with the longest capillary tube (i.e. \( \alpha = 0.01 \)) falls on top of the NICCA model plot (solid line), thereby showing that there is virtually no difference between (3.47) and (3.45) if \( \alpha \) is equal to 0.01. In contrast, if \( \alpha \) is equal to 2, the magnitude of the local inertia forces is large and increases the total normalized time of infiltration into the capillary tube by about 40\%. Given that the effects of drag and convective inertia forces have not been taken into account, it can be expected that the total normalized time of infiltration would be larger if those effects were included.

![Graph showing relative depth versus normalized time and interface velocity](image)

**Figure 3.8.** Theoretical effect of local inertia forces on the infiltration of DNAPL into 2.70 mm diameter capillary tubes of varying length: a. Relative depth of interface versus normalized time; b. Relative depth of interface versus interface velocity.

Similar findings are obtained when examining velocity fields. Figure 3.8.b shows a plot of the relative depth \( z_c(t)/l \) versus the interface velocity \( dz_c/dt \) using (3.48) for the NICCA model, and a time derivative of (3.45) for the model incorporating the effects of local inertia. Again, all four capillary tubes have the same relative depth-velocity field if the NICCA model is used (solid line), but are distinct if local inertia is included.

As can be seen in Figure 3.8.b, the velocity field of the interface using the full solution (3.45) is not affected by local inertia for the longest capillary tube corresponding to \( \alpha \) equal to 0.01. Indeed, this velocity field (solid line) matches that of...
the NICCA model. For the shorter tubes, the magnitude of interface velocity compared to that of the NICCA model decreases, and is smaller by about 25% if $\alpha$ increases to 2. Figure 3.8.b clearly shows that local inertia forces affect interface velocity throughout the entire infiltration process. Again, drag and convective inertia forces further reduce the interface velocity of short tubes.

3.5.2.4 Condition for Neglecting Convective Inertia

The assertion presented in Section 3.5.2.1 that convective inertia can be neglected when local inertia forces can be neglected is now demonstrated. As discussed in the dimensional analysis of Section 3.4.2, it can be expected that the assumption of flow parallel to the axis of the capillary tube will no longer hold true below a certain capillary tube length. Therefore, it is necessary to check under which condition (3.41)-(3.48) are valid.

At interface depth $z_c$ sufficiently larger than $\Delta h$, the interface velocity is at most equal to $k_\Delta(z_c/l)$ (see (3.49)), since drag and inertia forces may only contribute to slow down the interface velocity. Hence, using condition (3.16)

$$\frac{\rho_\mu r_0^2}{\mu_\phi l} k_\Delta << 1,$$

where $k_\Delta(z_c/l)$ is taken as a reference velocity and $z_c$ as a reference length.

Replacing the equivalent conductivity $k_\Delta$ in (3.51) using (3.43) gives

$$\frac{\Delta \rho \mu r_0^4}{8 \mu_\phi^2 l} << 1.$$

Noting that the left-hand side of (3.52) is equal to $2\alpha$, it can be concluded that, for convective inertia forces to be negligible, $2\alpha$ must be much less than 1. Therefore, the following statement is always true: if local inertia forces are not negligible, convective inertia forces cannot be neglected either. This conclusion, again, shows that validity of the infiltration kinetics equation (3.41) and its solution (3.45) are restricted to the case $\alpha << 1$. If $\alpha$ is very small with respect to 1, both conditions of negligible local and convective inertia are verified. Under these conditions, (3.41) and (3.45) are simplified to the NICCA model, i.e. (3.48) and (3.47), respectively.

It is interesting to note that the use of condition (3.16) can be extended to other capillary infiltration problems. A number of researchers have examined the rise of liquids in vertical capillary tubes and restricted their model to a balance of local inertia, viscosity, gravity and capillarity (see Section 2.3.3.1). Thus, they have ignored the role played by convective inertia, and attributed the difference between prediction and observation to a change in contact angle. While this change can certainly explain part
of the difference, (3.16) can be used to show that convective inertia was not always negligible in their systems and therefore should not have been ignored. For example, Quéré [1997] studied the rise of ethanol (\( \rho = 780 \text{ kg/m}^3 \), \( \mu = 1.17 \text{ mPa.s} \)) in a vertical glass capillary tube of radius \( r_0 = 0.689 \text{ mm} \). Although his prediction for initial velocity of rise was 28 cm/s accounting for local inertia forces, and 21 cm/s after estimating the effects of a dynamic contact angle, Quéré only measured a velocity of \( 17 \pm 1 \text{ cm/s} \). Using (3.16), it can be shown that convective inertia forces were not negligible at this speed throughout the first few centimeters of the capillary tube and certainly acted to further reduce the interface velocity in conjunction with the dynamic contact angle. The effects of convective inertia forces are also significant on similar experiments run by Jeje [1979], Siebold et al. [2000], and Zhmud et al. [2000], although not accounted for by the authors. Under circumstances where the effects of convective inertia cannot be neglected, the Navier-Stokes equations (3.6) and (3.7) are needed for solving the problem. Non-linearity of the convective inertia terms limits any infiltration kinetics prediction to a numerical solution.

Finally, the conditions for which entry forces can be neglected are re-examined. Equation (3.40) (see Section 3.5.1) defines the \( \beta \)-number and gives the condition for which entry drag forces are negligible compared to viscous forces. Using (3.49), it has been shown that the maximum interface velocity in the capillary tube is \( k \Delta \) provided that \( \Delta h \) remains negligible. Under these conditions, an upper bound \( \beta_1 \) describing the ratio of entry forces to viscous forces is given by

\[
\beta_1 = \frac{m \Delta \rho g r_0^4}{128 \mu_w l}.
\] (3.53)

Given that \( m \) ranges from 2.24 to 2.41 (see Section 2.3.4.1) and that \( \rho_{ow} \) is close to \( \rho_w \), the \( \beta_1 \)-number is only a fraction of \( \alpha \). Therefore, the following statement is true: under conditions where inertia forces can be neglected, the entry drag forces can be neglected as well. Thus, the \( \alpha \)-number can be used as a criterion for all three forces: local inertia, convective inertia and tube entry drag.

3.5.2.5 Effect of a DNAPL Pre-Infiltration Finger on the NICCA Model

This section examines the case where a finger of DNAPL has pre-infiltrated a capillary tube, such that the DNAPL/interface has already penetrated down to a depth \( h_{pi} \), but has stopped at this depth. The scenario is illustrated in Figure 3.9. As will be shown in Section 6.2.2 and further discussed in Section 7.2.1.2, such pre-infiltration can take place in practice, and the depth of pre-infiltration, \( h_{pi} \), can be significant with respect to the length of the capillary tube.

Under static conditions, the scenario shown in Figure 3.9.a is equivalent to having a DNAPL pool of total height \( h = h_{pi} + h_p \), where \( h_p \) is the height of the pool in the
reservoir tube, on top of capillary tube of reduced length $l$ (the total length of the capillary tube is $l_t = l + h_{pi}$). As per (3.3), complete infiltration of the capillary tube takes place if the total pool height $h$ (reservoir pool and pre-infiltration finger) exceeds the critical height $h_i$ given by (3.4).

In Figure 3.9.b, the complete DNAPL infiltration process is taking place and the DNAPL/water interface is moving (dynamic conditions). At time $t$, the capillary tube has progressed down to a depth $z_c(t)$ below the depth of pre-infiltration. It is important to realize, however, that the scenario shown in Figure 3.9.b is not strictly equivalent to that shown in Figure 3.9.c. Indeed, in Figure 3.9.c, a pool of height $h = h_{pi} + h_p$ infiltrates a capillary tube of length $l$. Using the assumptions of the NICCA model, the scenario shown in Figure 3.9.c is predicted by (3.48) with $\Delta h = h_{pi} + h_p - h_i$.

![Figure 3.9](image)

Figure 3.9. Schematics of DNAPL infiltration into a capillary tube with existing pre-infiltration: a. Prior to the onset of infiltration with existing pre-infiltration down to depth $h_{pi}$ ($t<0$); b. During infiltration ($t\geq0$); c. Scenario that, although statically similar, is not dynamically equivalent to that shown on (b).

In Figure 3.9.b, the DNAPL finger of length $h_{pi}$ contributes to slow down the interface velocity, because it contributes to extra viscous forces (as well as inertia forces) that do not appear in the scenario illustrated in Figure 3.9.c. Hence, if pre-infiltration exists, it is incorrect to directly use the NICCA model with the height $h = h_{pi} + h_p$ and the reduced length of capillary tube $l$, as can be done for the scenario shown in Figure 3.9.c.

Equation (3.37) can be used to solve the new problem of infiltration kinetics. Indeed, referring to Figure 3.9.b, the total finger length at time $t$ is $h_{pi} + z_c(t)$. Under
this configuration, and using the notation shown in Figure 3.9, (3.37) can be rewritten as

\[
\left[ \rho_w(l + h_{p_i}) + \Delta \rho \left[ z_c(t) + h_{p_i} \right] \right] \left( \frac{d^2 z_c}{d t^2} \right) + \frac{8}{r_0^2} \left[ \mu_w(l + h_{p_i}) + \Delta \mu \left[ z_c(t) + h_{p_i} \right] \right] \frac{dz_c}{d t} \\
= \Delta \rho g \left[ z_c(t) + h_{p_i} \right] + h_p - \frac{2 \sigma \cos \theta_p (dz_c / d t)}{\Delta \rho g r_0} - \frac{1}{2} m \rho \left( \frac{dz_c}{d t} \right)^2. \tag{3.54}
\]

Making use of all the complementary assumptions made in Section 3.5.1, (3.54) is reduced to

\[
\left[ \rho_w(l + h_{p_i}) \right] \left( \frac{d^2 z_c}{d t^2} \right) + \frac{8}{r_0^2} \left[ \mu_w(l + h_{p_i}) \right] \frac{dz_c}{d t} \\
= \Delta \rho g \left[ z_c(t) + h_{p_i} \right] + h_p - h_i, \tag{3.55}
\]

where \(h_i\) is the critical height defined by (3.4). After simplification, (3.55) is reduced to

\[
\left( \frac{d^2 z_c}{d t^2} \right) + \frac{g}{k_w} \frac{dz_c}{d t} = \frac{k_\Delta}{k_w} \frac{g}{l + h_{p_i}} [z_c(t) + \Delta h], \tag{3.56}
\]

where \(k_w\) and \(k_\Delta\) are defined by (3.42) and (3.43), respectively, and \(\Delta h\) is given by

\[
\Delta h = h_p + h_{p_i} - h_i = h - h_i. \tag{3.57}
\]

Thus, (3.56) is identical to (3.41), except that the length \(l\) in (3.41) is replaced by the length \(l + h_{p_i}\) in (3.56). Therefore, for the scenario shown in Figure 3.9.b, the total length of the tube rather than the reduced length of tube \(l\) must be used.

Under conditions where \(\alpha_s\), defined by

\[
\alpha_s = \frac{4 k_s k_w}{g(l + h_{p_i})} = \frac{\Delta \rho \rho g r_0^4}{16 \mu_w^2 (l + h_{p_i})}, \tag{3.58}
\]

is very small with respect to unity, it can be shown that (3.56) reduces to

\[
\frac{dz_c}{d t} = \frac{k_\Delta}{l + h_{p_i}} [z_c(t) + \Delta h]. \tag{3.59}
\]

Note that (3.59) is equivalent to (3.48) except that the former equation contains an extra viscous force term equal to \((\pi r_0^2)(8/r_0^2)\mu_w h_p dz_c/dt\). This viscous term corresponds to
the viscous force associated with the DNAPL pre-infiltration finger—with the assumption that the viscosity of DNAPL is approximately equal to that of water.

The solution of (3.59) using initial condition (3.39) is given by

\[ z_c(t) = \Delta h \left[ -1 + \exp \left( \frac{k_A t}{l + h_{pi}} \right) \right]. \]  

(3.60)

Similar to the \( \alpha \)-number (3.46), \( \alpha_t \), given by (3.58), is a parameter describing the magnitude of local inertia forces associated with the pre-infiltrated system. Moreover, condition (3.16) can be used in this pre-infiltration scenario to demonstrate that \( \alpha_t \) also describes the magnitude of convective inertia forces with respect to viscous forces. The derivation is similar to that shown in Section 3.5.2.4 and shows that convective inertia is negligible provided that \( \alpha_t << 1 \). Thus, for a pre-infiltrated system, the larger the magnitude of \( \alpha_t \), the larger the inertia forces (both local and convective). Because (3.56) only incorporates the effects of local inertia, this equation cannot be used when \( \alpha_t >> 1 \), since convective inertia forces must be taken into account as well. Conversely, for a pre-infiltrated system, inertia forces can be ignored when \( \alpha_t << 1 \). Under this condition, (3.59) and its solution (3.60) can be used to predict the infiltration kinetics.

As discussed in Section 3.5.2.2, \( \Delta h \) is very small in comparison to \( h_i \). If \( l \) and \( z_c \) are of orders equal to, or larger than \( h_i \), then (3.59) is reduced to

\[ \frac{d z_c}{dt} = k_A \frac{z_c(t)}{l + h_{pi}}. \]  

(3.61)

Therefore, if pre-infiltration exists, the interface exit velocity is predicted to be less than that in the case of a capillary tube of same radius not subjected to pre-infiltration. Under pre-infiltration conditions, the exit velocity is dependent upon the length of the capillary tube and is predicted to be equal to \( k_A l/(l + h_{pi}) \).

Examining the ratio of entry drag forces to viscous forces, a number, \( \beta_t \), similar to the \( \beta \)-number (3.40) (see Section 3.5.1), can be defined as

\[ \beta_t = \frac{1}{2} \frac{m \rho_c}{9 \mu_c} \frac{d z_c}{dt}. \]  

(3.62)

An upper bond \( \beta_{t1} \) of \( \beta_t \) is given by

\[ \beta_{t1} = \frac{m \Delta \rho g r_0^4}{128 \mu_w (l + h_{pi})^2}. \]  

(3.63)
using an approach similar to that used in Section 3.5.2.4. It can be easily shown that $\beta_{t1}$ is smaller than $\alpha_t$, thereby demonstrating that under conditions where inertia forces can be neglected in a pre-infiltration configuration, the entry drag forces can be neglected as well.

3.5.3 Analytical Solution Based on Assumption of Variable Contact Angle

3.5.3.1 Overview

In Section 2.3.6, it was shown that the dynamic contact angle, defined as the contact angle of the moving interface, is generally a function of the interface velocity. In the case of drainage, i.e. when the non-wetting fluid displaces the wetting fluid, the contact angle tends to zero if the interface velocity is sufficiently large. Hence, referring to the DNAPL infiltration into fractures simulated by vertical capillary tubes, it can be expected that perfect wetting will take place beyond a certain displacement velocity, $dz_c/dt$.

An alternative to the constant contact angle assumption is to assume a linear dependence of the cosine of the dynamic contact angle upon $dz_c/dt$. This approach is consistent with that used by Siegel [1961] and Hamraoui et al. [2000] for their study of liquids displacing air in capillary tubes (see Section 2.3.5.6). Most particularly, the linear dependence of contact angle cosine upon velocity (specifically dynamic capillary pressure dependence upon capillary number) has been suggested by Calvo et al. [1991] in their study of cyclohexane (non-wetting liquid) displacing water (wetting liquid) in a 0.55 mm radius horizontal capillary tube (see Section 2.3.6.4).

Under this hypothesis, the cosine of the dynamic contact angle, $\cos \theta_d$, ramps from the cosine of the static contact angle, $\cos \theta_s$, at zero velocity, to a value of unity, corresponding to perfect wetting at a certain velocity $u_c$. This is illustrated in Figure 3.10 where the dynamic contact angle cosine is plotted versus the normalized velocity $(dz_c/dt)/u_c$. The corresponding relationship between the dynamic contact angle and the normalized velocity is also plotted on the same graph. Obviously, the model is not realistic in the vicinity of $(dz_c/dt)/u_c = 1$ where an unlikely slope discontinuity on both plots is observed. A second issue is that $\theta_s$ and $u_c$ have to be measured, or fitted experimentally from a first series of experiments, before any prediction can be made. Nevertheless, this approach provides a qualitative trend regarding what might happen at the early stages of DNAPL infiltration.
3.5.3.2 Solution of Governing Equation

Referring to Figure 3.10, the cosine of the dynamic contact angle can be defined as

\[
\cos \theta_d \left( \frac{dz_c}{dt} \right) = \begin{cases} 
\cos \theta_s + (1 - \cos \theta_s) \frac{dz_c}{dt} & \text{if } \frac{dz_c}{dt} \leq u_c, \\
1 & \text{if } \frac{dz_c}{dt} > u_c.
\end{cases}
\]  

(3.64)

Again, it is assumed that DNAPL and water have reasonably close viscosities, and that inertia forces (local and convective) as well as drag forces are negligible (i.e. \( \alpha \ll 1 \)). Furthermore, it is assumed that no pre-infiltration is present \((h_{pi} = 0)\). Equation (3.37) is simplified to
\begin{align}
\frac{8 \mu \cdot l \ dz_c}{r_0^2} \frac{dz_c}{dt} &= \Delta \rho g \left[ z_c(t) + h - \frac{2 \sigma \ \cos \theta_s (dz_c/dt)}{\Delta \rho \ g r_0} \right] , \tag{3.65}
\end{align}

where use of (3.38) has been made.

Replacing (3.64) in (3.65), gives for the case \( dz_c/dt \leq u_c \)

\begin{align}
\frac{dz_c}{dt} &= \frac{k_\Delta}{l} \left[ z_c(t) + h - \frac{2 \sigma \ \cos \theta_s}{\Delta \rho \ g r_0} \right] , \tag{3.66}
\end{align}

where \( k_\Delta \) is given by (3.43). Rearranging terms in (3.66) leads to

\begin{align}
\frac{dz_c}{dt} &= \frac{k_\Delta}{\gamma l} [z_c(t) + \Delta h] \quad \text{for} \ dz_c/dt \leq u_c , \tag{3.67}
\end{align}

where \( \Delta h \) is the constant defined by (3.44) and \( \gamma \) is a retardation factor larger than unity and defined by

\begin{align}
\gamma = 1 + \frac{h_i \ k_\Delta}{l \ u_c} \frac{1 - \cos \theta_s}{\cos \theta_s} . \tag{3.68}
\end{align}

Equation (3.67) can be solved and yield for \( z_c(t) \) a solution similar to (3.47) except that the time constant \( l/k_\Delta \) is now increased to \( l/\gamma \ k_\Delta \).

The case \( dz_c/dt > u_c \) is now examined. Replacing (3.64) in (3.65) gives

\begin{align}
\frac{dz_c}{dt} &= \frac{k_\Delta}{l} [z_c(t) + \Delta h'] \quad \text{for} \ dz_c/dt > u_c , \tag{3.69}
\end{align}

where \( \Delta h' \) is a negative constant defined by

\begin{align}
\Delta h' = h - \frac{2 \sigma}{\Delta \rho \ g r_0} . \tag{3.70}
\end{align}

Let \( t_{c,0} \) and \( z_{c,0} \) be, respectively, the time and depth for which \( dz_c/dt = u_c \). Both can be calculated using (3.67) and its solution. The condition \( z_c = z_{c,0} \) at \( t = t_{c,0} \) serves as the initial condition used to solve (3.69), whose solution is also similar to (3.47).

The system of infiltration kinetics equations (3.67)-(3.70) is referred to as the negligible inertia variable contact angle model (NIVCA model).
To illustrate the effect of the dynamic contact angle as defined by (3.64), consider the infiltration of 4-CT ($\rho_{nw} = 1070$ kg/m$^3$) in a 305 mm long, 1.33 mm diameter water-saturated vertical capillary tube. Calculation of the conductivities $k_w$ and $k_A$ yield 541 mm/s and 39 mm/s using (3.42) and (3.43), respectively. The $\alpha$-number is equal to 0.028 using (3.46), while the $\beta_1$-number is equal to $8.9 \times 10^{-3}$ using (3.53) with $m = 2.35$. Both numbers indicate that inertia and drag forces can be neglected. A fixed interfacial tension, $\sigma$, between DNAPL and water of 0.032 N/m is assumed such that the critical height, $h_i$, is 136 mm in the case of static perfect wetting ($\theta_s = 0^\circ$), but decreases with increasing static contact angle as per (3.4). The DNAPL height is 1% larger than the critical height ($h = 1.01 \, h_i$).

The effect of the static contact angle, $\theta_s$, on (3.67) and (3.69) is first examined. The perfect wetting velocity, $u_c$, is fixed and assumed to be equal to 2 mm/s, which is less than $k_s$ and therefore clearly influences the infiltration process. The value of the static contact angle is varied between 0$^\circ$ and 60$^\circ$, such that the reduced interfacial tension, $\sigma \cos \theta_s$, the critical height, $h_i$, and the retardation factor, $\gamma$, vary as well, while the length $\Delta h'$ given by (3.68) remains constant. Using the NIVCA model equations (3.67) and (3.69), the interface depth, $z_c$, is plotted versus time in Figure 3.11.a, and versus interface velocity, $dz_c/dt$, in Figure 3.11.b.

Referring to Figure 3.11.a, a larger static contact angle is associated with a slower infiltration process. If the static contact angle is equal to zero, then the dynamic contact angle, $\theta_d$, remains constant, as it has been assumed that $\theta_d$ decreases to zero with increasing velocity. Under this condition, (3.67) and (3.69) are reduced to (3.48), which corresponds to the NICCA model. On the other hand, if the static contact angle increases to 60$^\circ$, the retardation factor, $\gamma$, increases to 5.4 and acts to slow down the interface displacement. As shown in Figure 3.11.b, the delay in infiltration is caused by smaller interface velocities over a longer portion of the capillary tube. In other words, the capillary forces are significant over a larger portion of the capillary tube than in the case of the NICCA model. The discontinuity in velocity change at $dz_c/dt = u_c$, observed in Figure 3.11.b is a consequence of the slope discontinuity of the dynamic contact angle-interface velocity relationship (see Figure 3.10 and discussion of Section 3.5.3.1). Again, it would not be expected in a real infiltration problem.

The effect of the perfect wetting velocity, $u_c$, on (3.67) and (3.69) is now examined. The DNAPL/water interfacial tension is still equal to 0.032 N/m. The static contact angle, $\theta_s$, is fixed and assumed to be equal to 35$^\circ$, such that the critical height is equal to 112 mm (using (3.4)). The value of the perfect wetting velocity is varied from 0.1 mm/s to the infinite. Thus, the retardation factor $\gamma$ varies as well, but the length $\Delta h'$ remain constant. The interface depth, $z_c$, is plotted versus time in Figure 3.12.a, and versus interface velocity, $dz_c/dt$, in Figure 3.12.b.
Figure 3.11. Effect of variable contact angle on DNAPL infiltration kinetics for different static contact angles (the perfect wetting velocity, $u_c$, is fixed at 2 mm/s): a. NIVCA model infiltration profile; b. NIVCA model velocity field.

Figure 3.12. Effect of variable contact angle on DNAPL infiltration kinetics for different perfect wetting velocities (the static contact angle, $\theta_s$, is fixed at $35^\circ$): a. NIVCA model infiltration profile; b. NIVCA model velocity field (recall that $k_\Delta = 39$ mm/s).
Referring to Figure 3.12.a, a smaller perfect wetting velocity is associated with a slower infiltration process. As clearly shown in Figure 3.12.b, there are two cases to consider. In the first case, the perfect wetting velocity, $u_c$, is less than the equivalent conductivity, $k_\lambda$, equal here to 39 mm/s, such that the infiltration process is successively described by (3.67) and (3.69). The interface first moves slowly over a capillary region of length equal to $z_{c0}$. Past the capillary region, the interface proceeds to move faster. As shown in Figure 3.12.b, the length and velocity field $dz_c/dt$ of the capillary region is a function of $u_c$, whereas the velocity field of the lower region is not.

In the second case, the perfect wetting velocity, $u_c$, is more than the equivalent conductivity, $k_\lambda$. Because the interface velocity can never exceed the equivalent conductivity, the infiltration process is only described by (3.67). Under this scenario, the dynamic contact angle continuously decreases throughout the infiltration process, but never reaches perfect wetting. The capillary region extends down to the lower end of the capillary tube and the velocity field is much smoother. When the perfect wetting velocity is infinite, the contact angle remains equal to 35° throughout the entire infiltration process, which corresponds to the NICCA model, i.e. (3.48).

3.5.3.4 Effect of a DNAPL Pre-Infiltration Finger on the NIVCA Model

Similar to Section 3.5.2.5, this section examines the case where a finger of DNAPL has pre-infiltrated a capillary tube, such that the DNAPL/interface has already penetrated down to a depth $h_{pi}$, but has stopped at this depth (see Figure 3.9.a in static configuration and Figure 3.9.b in dynamic configuration). The total length of capillary tube is, again, $l + h_{pi}$.

Under the same assumptions as those used in Section 3.5.3.2 (DNAPL and water have reasonably close viscosities, inertia and drag forces are negligible), it can be shown that, if pre-infiltration is present down to depth $h_{pi}$, then the governing equation of the infiltration kinetics (3.65) can be replaced by

$$\frac{8\mu_w(1 + h_{pi})}{r_0^2} \frac{dz_c}{dt} = \Delta\rho g \left[ z_c(t) + h - \frac{2\sigma \cos \theta_d(dz_c/dt)}{\Delta\rho g r_0} \right], \quad (3.71)$$

with the function $\cos \theta_d(dz_c/dt)$ given by (3.64).

Again, note that (3.71) is equivalent to (3.65) except that the former equation contains an extra viscous force term equal to $(\pi r_0^2)^2(8/r_0^2)\mu_w h_{pi}dz_c/dt$. This viscous term corresponds to the viscous force associated with the DNAPL pre-infiltration finger—with the assumption that the viscosity of DNAPL is approximately equal to that of water. Thus, for (3.71), the length over which viscous forces apply is the total length of the capillary tube, i.e. $l + h_{pi}$. Substituting (3.64) in (3.71) and rearranging terms, as was done in Section 3.5.3.2 to derive (3.67)-(3.69), leads to
\[
\frac{dz_c}{dt} = \frac{k_\Delta}{\gamma_t(l + h_{pi})}[z_c(t) + \Delta h] \quad \text{for } \frac{dz_c}{dt} \leq u_c, \quad (3.72)
\]

where \(\Delta h\) is the constant defined by (3.44) and \(\gamma_t\) is a retardation factor larger than unity and defined by

\[
\gamma_t = 1 + \frac{h_{li}}{l + h_{pi}} \frac{k_\Delta}{u_c} \frac{1 - \cos \theta_s}{\cos \theta_s}, \quad (3.73)
\]

and

\[
\frac{dz_c}{dt} = \frac{k_\Delta}{l + h_{pi}}[z_c(t) + \Delta h'] \quad \text{for } \frac{dz_c}{dt} > u_c, \quad (3.74)
\]

where \(\Delta h'\) is the negative constant defined by (3.70).

As can be seen from the set (3.72)-(3.74), the governing equations are controlled by the total length of the capillary tube \(l_t = l + h_{pi}\). At a given flow length \(l\), the interface displacement velocity decreases with increasing pre-infiltration length \(h_{pi}\) (and thus increasing total capillary tube length \(l_t\)). This trend is similar to that of the NICCA model in the case where pre-infiltration exists.

3.5.4 Analytical Solution for Fluids of Differing Viscosities

In this section, (3.37) is solved under conditions of negligible inertia forces, and drag forces. However, it is now assumed that the viscosity contrast between DNAPL and water can be significant with respect to the viscosity of water such that the contrast cannot be neglected. Assuming that the contact angle remains constant throughout the infiltration process, (3.37) yields

\[
\frac{8}{r_0^2} [\mu_w l + \Delta \mu z_c(t)] \frac{dz_c}{dt} = \Delta \rho g [z_c(t) + \Delta h], \quad (3.75)
\]

where, again, \(\Delta h\) is defined by (3.44). If DNAPL pre-infiltration down to depth \(h_{pi}\) takes place prior to the onset of complete infiltration, then the left-hand side term of (3.75) contains an extra term, \(8 \mu_{w,h_{pi}}/r_0^2 \frac{dz_c}{dt}\) (see (3.54) and discussion of Section 3.5.2.5). Again, the total length of the capillary tube is \(l_t\), with \(l_t = l + h_{pi}\).

Rearranging terms in (3.75) leads to
where

\[ k_{\Delta\Delta} = \frac{\Delta \rho g r_0^2}{8 \Delta \mu}, \]  

(3.77)

\[ h_{\mu} = \frac{\mu_{nw} + \mu_{nw} h_{pi}}{\Delta \mu}. \]  

(3.78)

Similar to \( k_{\Delta} \), the parameter \( k_{\Delta\Delta} \) has the dimensions of a velocity, and corresponds to the equivalent conductivity of a fluid of density \( \Delta \rho \) and viscosity \( \Delta \mu \). The parameter \( h_{\mu} \) has the dimensions of a length. Both parameters are negative if the viscosity of DNAPL is smaller than that of water, i.e. smaller than 1 mPa.s at 20°C [Munson et al., 1994]. This can happen in practice. In Section 2.2.1.4, it was shown that typical DNAPL viscosities span over several orders of magnitude, from slightly less than the viscosity of water for aliphatic chlorinated compounds to several hundred mPa.s for some PCBs (see Figure 2.3).

Equation (3.76) can be solved analytically. Using the initial condition \( z_c = 0 \) at \( t = 0 \), (3.76) leads to

\[ t = \frac{z_c + h_{\mu} - \Delta h}{k_{\Delta\Delta}} \ln \left( 1 + \frac{z_c}{\Delta h} \right). \]  

(3.79)

To illustrate the effect of viscosity contrast on infiltration, consider the infiltration of a DNAPL of density 1070 kg/m\(^3\) in a 305 mm long, 1.33 mm diameter water-saturated vertical capillary tube. A critical height \( h_i \) of 136 mm is assumed. The DNAPL pool height is 1% larger than the critical height \( (h = 1.01 h_i) \) at the onset of infiltration. The effects of pre-infiltration are neglected \( (h_{pi} = 0) \). Figure 3.13 illustrates the case where the DNAPL viscosity is smaller than that of water \( (\Delta \mu < 0, k_{\Delta\Delta} < 0 \text{ and } h_{\mu} < 0) \) and varies from 0.3 mPa.s to 0.9 mPa.s. The interface depth, \( z_c \), is plotted versus time in Figure 3.13.a, and versus interface velocity, \( dz_c/dt \), in Figure 3.13.b. Figure 3.14 illustrates the case where the DNAPL viscosity is larger than that of water \( (\Delta \mu > 0, k_{\Delta\Delta} > 0 \text{ and } h_{\mu} > 0) \) and varies from 1.1 mPa.s to 100 mPa.s. Again, the interface depth, \( z_c \), is plotted versus time in Figure 3.14.a, and versus interface velocity, \( dz_c/dt \), in Figure 3.14.b. For comparison, the NICCA model (i.e. (3.48)) has been added in all figures and corresponds to the case where \( \mu_{nw} = \mu_w \).
Figure 3.13. Influence of viscosity on infiltration kinetics if DNAPL viscosity is smaller than that of water: a. Interface depth versus time for different DNAPL viscosities; b. Interface depth versus interface velocity for different DNAPL viscosities.

Figure 3.14. Influence of viscosity on infiltration kinetics if DNAPL viscosity is larger than that of water: a. Interface depth versus time for different DNAPL viscosities; b. Interface depth versus interface velocity for different DNAPL viscosities.
As can be seen in both Figure 3.13.a and Figure 3.14.a, the larger the DNAPL viscosity, the slower its infiltration. In particular, if the DNAPL viscosity is sufficiently large with respect to that of water, $h_v$ becomes small and the logarithmic term in (3.79) is negligible beyond a certain interface depth $z_c$, which explains the linear relationship between $z_c$ and $t$ observed in Figure 3.14.a for $\mu_{nw} = 100$ mPa.s. On the other hand, a small viscosity contrast ±0.1 mPa.s is sufficiently small such that the NICCA model can be used under this condition for predicting the displacement of the DNAPL/water interface. This is the case for 4-CT whose viscosity is around 0.9 mPa.s at 20°C (see Table 5.3 in Section 5.5.2). The effects of a small viscosity contrast on the accuracy of the NICCA prediction are further examined and discussed in Section 7.2.2.5.

As clearly shown in Figure 3.13.b and Figure 3.14.b, the interface velocity deviation from the NICCA model is larger in the lower end of the capillary tube than in its upper part. Indeed as the infiltration proceeds, the proportion of the capillary tube replaced by DNAPL increases so as to give the viscosity contrast more and more impact on the magnitude of the viscous forces. Of course, this model does not take into account potential instabilities that could result from a large absolute viscosity contrast $|\Delta \mu|$ and assumes that DNAPL completely displaces water such that both fluids move downwards. One could imagine, for example, that DNAPL could be so viscous that water would flow upwards as a result of DNAPL downward movement. Conversely, low-viscosity DNAPL could flow downward through a reduced section of capillary tube while water might remain immobile. Finally, it must be noted that inertia and entry drag effects may become significant if the DNAPL viscosity is small.

### 3.6 Extension of Solutions to Rectangular Section Capillary Tubes and Rough-Walled Fractures

In deriving the governing equations of DNAPL infiltration into vertical fractures, the fracture has been idealized as a smooth-walled circular section capillary tube. The analytical solution proposed in Section 3.5.2 is now extended to rectangular section capillary tubes (Section 3.6.1), and rough-walled fractures (Section 3.6.2). Again, the fracture is assumed vertical although the analysis is easily applicable to a fracture of any dip.

#### 3.6.1 Solutions for Rectangular Section Capillary Tubes

The governing equations of DNAPL infiltration into circular capillary tubes have been derived using cylindrical coordinates and an expression of the continuity and Navier-Stokes equations in those coordinates (see Section 3.4.1). A similar derivation
can be achieved if, instead of a circular tube, a rectangular section capillary tube of aperture \( e \) is considered, such as that shown in Figure 3.4.b. Again, a sharp interface is assumed to separate the DNAPL and water phases (see Figure 3.3.b). Rectangular coordinates and an expression of the continuity and Navier-Stokes equations in those coordinates can be used to predict the interface displacement within the capillary tube. The different steps leading to one of the analytical solutions are similar to those used in Section 3.4 and Section 3.5.

The solution is restricted here to the NICCA model assumptions. That is, the inertia forces and drag forces at the entry to the tube can be neglected, the density and viscosity contrasts are small with respect to the density and viscosity of water, and the dynamic contact angle is a constant equal to the static contact angle (see Section 3.5.1 and Section 3.5.2). In addition, it is assumed that no pre-infiltration has taken place, although the effects of pre-infiltration could be included using an approach similar to that shown in Section 3.5.2.5. Under these conditions, it can be shown that the interface displacement is also predicted using (3.47) and (3.48). The only difference lies in the definition of the parameter \( k_{\Delta} \), which is now the equivalent conductivity of a fluid of density \( \Delta \rho \) and viscosity \( \mu_w \) flowing in a rectangular capillary tube of aperture \( e \). The square section parameter \( k_w \) is given by

\[
k_{\Delta} = \frac{\Delta \rho g e^2}{12 \mu_w}.
\]  

Equations (3.47) and (3.48) combined with (3.80) can only be applied if local and convective inertia forces as well as drag forces can be neglected. Similarly to (3.46), this condition can be written as

\[
\alpha = \frac{4k_{\Delta}k_w}{gl} << 1
\]  

where \( k_w \) is defined by

\[
k_w = \frac{\rho_w g e^2}{12 \mu_w}.
\]  

The parameter \( k_w \) is the hydraulic conductivity of water in a rectangular capillary tube of aperture \( e \). Note that both \( k_w \) and \( k_{\Delta} \), as defined by (3.80), are consistent with the cubic law of the transmissivity of a fracture [de Marsily, 1986; Bear, 1993]. Combining (3.82) and (3.80) to (3.81) leads to

\[
\alpha = \frac{\Delta \rho \rho_w g e^4}{36 \mu_w^2} << 1.
\]
Equation (3.47) and (3.48) have been successfully used to predict the interface displacement in rectangular section capillary tubes [Adams, 2000]. Other predictions incorporating the dependence of contact angle upon interface velocity or a significant viscosity contrast can also be derived using techniques similar to those shown in Section 3.5.

3.6.2 Solutions for Rough-Walled Fractures

The model equations for rectangular section capillary tubes presented in Section 3.6.1 suggest—but do not prove—that under similar conditions of negligible inertia and drag forces, the model can be extended to rough-walled fractures. Consider a vertical rough-walled fracture such as that shown in Figure 3.2. While the fracture aperture is variable at a small scale, it is assumed that at some larger scale, homogeneous hydraulic properties to the fracture can be assigned such that it has an intrinsic permeability of $k$. For the particular case of a smooth-walled capillary tube, the permeability would be $r_0^2/8$ for a circular tube of radius $r_0$, and $e^2/12$ for a rectangular tube of aperture $e$ [de Marsily, 1986].

As illustrated in Figure 3.2, a sharp interface is assumed to separate the DNAPL and water phases. It is assumed that no pre-infiltration has initially taken place. Water flows downward and is entirely displaced by DNAPL. Under these conditions, by analogy with the smooth-walled fracture systems, (3.47) and (3.48) can be extended if $k_\Delta$ and $k_w$ are, respectively, given by

$$k_\Delta = \frac{\Delta \rho g k}{\mu_w}, \quad (3.84)$$

and

$$k_w = \frac{\rho_w g k}{\mu_w}. \quad (3.85)$$

Inertia and drag forces are only negligible if the following condition is true

$$\alpha = \frac{4k_\Delta k_w}{gl} = \frac{4\Delta \rho g \kappa k^2}{\mu_w^2 l} \ll 1. \quad (3.86)$$

Equations (3.47) and (3.48) can, in fact, be proven for rough-walled fractures using Darcy’s law for multiphase flow. The flux $u_j$ of fluid $j$ in the fracture is given by [de Marsily, 1986; Chown et al., 1997]
\[ u_j = -k_{rj} \frac{k_j}{\mu_j} \left( \frac{\partial p_j}{\partial z} - \rho_j g \right) \quad (j = nw, w), \] (3.87)

where \( k_{rj} \) is the fracture relative permeability of fluid \( j \), and is a function of the fluid phase saturation in the fracture [Longino and Kueper, 1999]. Under the assumption of complete displacement of water by DNAPL, the relative permeability of fluid \( j \) is equal to unity wherever fluid \( j \) is present.

By conservation of mass and assuming homogeneous properties can be defined, in particular that the variations in fracture aperture are small, the macroscopic flow velocity is the same throughout the fracture, independent of depth \( z \) and equal to the interface displacement velocity, \( dz_c/dt \), such that

\[ u_{nw} = u_w = \frac{dz_c}{dt}. \] (3.88)

Integrating (3.87) from 0 to \( z_c(t) \) for the DNAPL phase and making use of (3.88) leads to

\[ p_{mw}(z_c, t) - p_{mw}(0, t) = \left( \rho_{nw} g - \frac{\mu_{nw}}{k} \frac{dz_c}{dt} \right) z_c(t). \] (3.89)

Likewise, integrating (3.87) from \( z_c(t) \) to \( l \) for the water phase and after making use of (3.88) gives

\[ p_w(l, t) - p_w(z_c, t) = \left( \rho_w g - \frac{\mu_w}{k} \frac{dz_c}{dt} \right) [l - z_c(t)]. \] (3.90)

Adding (3.89) to (3.90), and rearranging terms leads to

\[ p_{mw}(z_c, t) - p_{mw}(z_c, t) = -\frac{1}{k} \left[ \mu_w l + \Delta \mu z_c(t) \right] \frac{dz_c}{dt} + \rho_{nw} g l + \Delta \rho g z_c(t) + p_{mw}(0, t) - p_w(l, t). \] (3.91)

Equation (3.91) is similar to (3.30) except that inertia terms are no longer present and that the fracture intrinsic permeability \( k \) has been substituted to \( r_0^2/8 \). Thus, under the assumption that inertia forces are negligible and that homogeneous properties exist for the fracture, Darcy’s law for multiphase flow can be used to predict the interface displacement within rough-walled fractures and match the extension of laws derived for smooth-walled fractures.
3.7 References


CHAPTER 4
CENTRIFUGE SCALING LAWS
FOR DNAPL INFILTRATION
INTO VERTICAL FRACTURES

4.1 Introduction

In Section 2.4.1, the centrifuge scaling laws were introduced and shown to connect the behavior observed in a 1/n-scale model experiment conducted at n times the Earth’s gravitational acceleration to the behavior in the equivalent prototype. If the product of depth multiplied by the equivalent gravitational acceleration is the same in the model and the corresponding prototype, the stress and fluid pressure distribution throughout the model will be identical with that throughout the prototype.

The objective of Chapter 4 is to develop a reduced-scale physical model of DNAPL behavior in a single rough-walled vertical fracture, such as that shown schematically in Figure 3.2, and obtain the corresponding scaling laws that allow translation from the reduced-scale centrifuge model to the full-scale prototype. Physical modeling of DNAPL behavior is achieved by extending the theoretical model developed in Chapter 3 at the Earth’s gravitational acceleration, g, to the case where the body force acting upon the fracture system is a centrifugal acceleration equivalent to n times the Earth’s gravity (i.e. ng).

As was done in Chapter 3, the criterion for DNAPL infiltration and the kinetics of infiltration into the fracture are successively examined. The most general case of a rough-walled fracture is considered, and the implications associated with neglecting inertia forces and drag forces at the entry to the capillary tube are discussed. Two methods of analysis are proposed and treated under separate headings, in Section 4.2 and Section 4.3, respectively. Finally, Section 4.4 discusses the limits associated with the physical modeling of velocity-dependent dynamic contact angles using the centrifuge. A list of references used in this chapter is provided in Section 4.5.

The scaling laws obtained in this chapter are used to predict the outcome of series of centrifuge DNAPL infiltration experiments in vertical capillary tubes that are described in Chapter 6. Comparison between the model predictions and experimental data is examined in Chapter 7.
4.2 Physical Modeling of Infiltration

Consider the model of DNAPL infiltration in a fracture of axis parallel to the direction of the centrifugal acceleration as shown in Figure 4.1.a. Noting that the centrifugal acceleration is equal to \( ng \), and making use of the infiltration criterion (3.4) (see Section 3.3), the infiltration into the capillary tube representing the fracture takes place for a critical model pool height \( h_i \) given by

\[
h_i = \frac{2\varepsilon \sigma \cos \theta}{\Delta \rho (ng) e(0)},
\]

where the parameters in (4.1) are identical to the parameters defined in Chapter 3.

At time \( t \), infiltration has taken place down to depth \( z_c(t) \) as illustrated in Figure 4.1.b. It is assumed that no pre-infiltration has initially taken place prior to the onset of infiltration \( (h_{pi} = 0) \). It is also assumed that the inertia forces and drag forces at the entry to the fracture can be neglected so that the parameter \( \alpha_n \), defined by

\[
\alpha_n = \frac{4\Delta \rho w(ng)k^2}{\mu^2 l},
\]

is negligible with respect to unity. The parameter \( \alpha_n \) is equivalent to the parameters \( \alpha \) defined by (3.86) (see Section 3.6.2) and with the Earth’s gravitational acceleration, \( g \), replaced by the centrifugal acceleration \( ng \).

The density and viscosity contrasts between DNAPL and water are assumed to be small with respect to the density and viscosity of water. It is also assumed that the dynamic contact angle remains constant throughout the DNAPL infiltration process and equal to \( \theta_s \). Using (3.84) (see Section 3.6.2) and (3.48) (see Section 3.5.2.1), the governing equation describing the interface displacement at an acceleration of \( ng \) is given by

\[
\frac{dz_c}{dt} = \frac{\Delta \rho (ng)k}{\mu^2 l} [z_c(t) + \Delta h],
\]

where \( \Delta h \) is again the difference between pool height above the fracture entrance, \( h \), and critical pool height, \( h_i \).

Let the parameters \( H_i, Z_c, \Delta H, L \) and \( T \) be, respectively, defined as

\[
H_i = nh_i, \quad Z_c = nz_c, \quad \Delta H = n\Delta h, \quad L = nl, \quad T = n^2 t.
\]

Substituting (4.4) into (4.1) and (4.3) leads to
\[ H_i = \frac{2\varepsilon \sigma \cos \theta}{\Delta \rho \varepsilon e(0)} \quad \text{(4.5)} \]

and

\[ \frac{dZ_c}{dT} = \frac{K_\Delta}{L} [Z_c(T) + \Delta H] \quad \text{(4.6)} \]

where

\[ K_\Delta = \frac{\Delta \rho g k}{\mu_w}. \quad \text{(4.7)} \]

Equations (4.5) and (4.6) describe, respectively, the DNAPL infiltration criterion and the kinematics of the DNAPL infiltration for a problem \( n \)-times larger than the experimental dimensions (see Figure 4.1.c). In addition, (4.7) is identical to (3.84) (see Section 3.6.2), and describes the equivalent conductivity of a fluid flowing in a fracture of intrinsic permeability \( k \), having the viscosity of water and a density equal to \( \Delta \rho \).

The set of equations (4.4) is, in fact, the series of established centrifuge scaling relationships for flow phenomena in geologic media [e.g., Culligan-Hensley and Savvidou, 1995]. These relationships are used to connect the behavior observed in a reduced-scale experiment conducted at \( ng \) to the behavior in the equivalent prototype, i.e. the full-scale field problem. For example, (4.4) indicates that a centrifuge experiment conducted at 10 \( g \) using a capillary tube of diameter 1.33 mm and length 120 mm, having a 15 mm DNAPL pool height on top of the tube entrance, would mimic conditions in a tubular fracture of length 1.2 m, having an average aperture size of 1.33 mm with a DNAPL pool height of 0.15 m on top of its entrance.

In establishing the scaling laws, it can be noted that the entrance fracture aperture, \( e(0) \), and its intrinsic permeability, \( k \), which is directly connected to the fracture aperture field [Bear, 1993], are not scaled by a factor \( n \). This is in keeping with the observations of Culligan and Barry [1998] who demonstrated that microscopic lengths, such as soil grain sizes and fracture apertures, had to be equivalent in the centrifuge model and the equivalent prototype for correct scaling of NAPL behavior in the subsurface.

It is also important to note that scaling from the centrifuge experiment to the prototype is limited to cases where \( \alpha_n \), as defined by (4.2) is small in the centrifuge model. In fact, running an experiment on a tube of length \( L = nl \) at the Earth’s gravity, \( g \), and running an experiment on a tube of length \( l \) at \( n \) times the Earth’s gravity, \( ng \), are not strictly equivalent, because the \( \alpha_n \) number is \( n^2 \) larger in the latter case. Therefore, it is possible for inertia and drag forces to become significant with respect to viscous forces in the centrifuge model, even if they are not significant in the prototype.
Figure 4.1. DNAPL infiltration into a vertical fracture: a. Centrifuge model prior to infiltration ($\omega$ is the centrifuge rotational velocity); b. Centrifuge model during infiltration; c. Equivalent prototype of model.
Similar to spontaneous infiltration laboratory (1-g) experiments (see discussion of Section 3.5.2.2), ΔH becomes negligible with respect to Zc once the DNAPL/water interface has reached a depth of the order of Hf. Under these conditions, and so long as αn remain small, the prototype exit velocity, (dZc/dT) calculated at L, is independent of the prototype length and equal to KΔ, given by (4.7).

As will be explained in Section 6.3, when conducting a centrifuge experiment under the equivalent of n times the Earth’s gravitational acceleration, a fixed volume of DNAPL (fixed height h) is pooled on top of the capillary tube, and the g-level, n, is increased until infiltration takes place (varying H). Therefore, ΔH is related to the rate of increase of g-level of the centrifuge, which is controlled, and therefore can be made very small. Nonetheless, and as for laboratory experiments (see Section 3.5.2.2), experience has shown that exact measurement of ΔH is virtually impossible without large uncertainties. Therefore, as in the case of 1-g experiments, the asymptotic behavior of measured DNAPL/water interface displacements will be compared with the theoretical model. This point is further discussed in Section 7.3.4.1.

4.3 Method of Partial Inspectional Analysis

An alternative to the approach used in Section 4.2, is to perform a partial inspectional analysis [Shook et al., 1998; Pantazidou et al., 2000; Barry et al., 2001] by examining the governing equations for DNAPL infiltration into a fracture, as derived in Chapter 3. DNAPL and water fluid properties in the centrifuge model are taken to be those in the prototype. The scaling laws derived from this analysis are summarized in Table 4.1.

4.3.1 Infiltration Criterion

Recall (3.2) (see Section 3.3), which relates the local entry pressure at the DNAPL/water interface to the local fracture aperture at the interface location (see Figure 3.2). For similitude of the local entry pressure between a centrifuge model and its prototype, the ratio of model entry pressure to prototype entry pressure must be equal to one. Therefore

\[
\frac{p_e(z_e)}{p_e(Z_e)} = \frac{\frac{2\pi \sigma \cos \theta_s}{E(Z_e)}}{\frac{2\pi \sigma \cos \theta_s}{E(z_e)}} = \frac{E(Z_e)}{E(z_e)}, \quad (4.8)
\]
where, with the convention adopted in Section 4.2, lower-case letters have been used when referring to the reduced-scale model, and upper-case letters when referring to the equivalent full-scale prototype. The interfacial tension, the static contact angle and the parameter $\varepsilon$ have been taken to be identical in both the model and the prototype.

Table 4.1. Centrifuge Scaling Relationships

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Prototype-Model Ratio$^a$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Equivalent gravitational acceleration, $g$</td>
<td>$1/n$</td>
</tr>
<tr>
<td>Macroscopic lengths, $h$, $h_i$, $z$, $z_c$, $l$</td>
<td>$n$</td>
</tr>
<tr>
<td>Microscopic length, $e$</td>
<td>1</td>
</tr>
<tr>
<td>Intrinsic permeability, $k$</td>
<td>1</td>
</tr>
<tr>
<td>Fluid pressures, $p_e$, $p_{nw}$, $p_w$</td>
<td>1</td>
</tr>
<tr>
<td>Fluid properties, $\rho_j$, $\mu_j$, $\sigma$</td>
<td>1</td>
</tr>
<tr>
<td>Static contact angle, $\theta_s$</td>
<td>1</td>
</tr>
<tr>
<td>Fluid velocity, $u_i$</td>
<td>$1/n$</td>
</tr>
<tr>
<td>Conductivities, $k_w$, $k_\Delta$</td>
<td>$1/n$</td>
</tr>
<tr>
<td>Time, $t$</td>
<td>$n^2$</td>
</tr>
</tbody>
</table>

$a$ $n$ is the scaling factor.

Equation (4.8) requires that the local fracture aperture in the centrifugal model is similar at homologous points to that in the prototype. This relationship is comparable to the requirements that microscopic dimensions must remain invariant between a centrifuge model and its prototype during modeling of porous media flow in order to ensure that effects of capillarity are properly duplicated in a centrifuge test [see Culligan and Barry, 1998].

Recalling (3.3) (see Section 3.3), DNAPL will infiltrate the fracture when the additional fluid pressure exerted by the presence of the DNAPL pool is equivalent to the fracture entry pressure. Therefore, the criterion for DNAPL infiltration is given by

$$h \geq h_i = \frac{p_e(0)}{\Delta \rho g},$$  \hspace{1cm} (4.9)

where $h_i$ is the minimum infiltration pool height or critical height.

From (4.8) and (4.9)

$$\frac{h_i}{H_i} = \frac{\Delta \rho ng}{P_e(0)} = \frac{1}{n},$$ \hspace{1cm} (4.10)
where it has been assumed that the density contrast is the same in the centrifuge model and the prototype.

From (4.10), it can be concluded that the DNAPL pool height at infiltration will be \( n \) times smaller in the centrifuge model than in the prototype. Thus, the critical height, \( h_c \), correctly scales as a macroscopic dimension of the problem.

### 4.3.2 DNAPL/Water Interface Displacement

Recall Darcy’s law for multiphase law (3.87) used in Section 3.6.2 for the prediction of interface displacement in rough fractures

\[
\frac{\mu_j}{k_{ij}} u_j = -\frac{\partial p_j}{\partial z} + \rho_j g \quad (j = nw, w). \tag{4.11}
\]

Again, under the assumption of complete displacement of water by DNAPL, \( k_{ij} \) is equal to unity wherever fluid \( j \) is present.

From (3.88) (see Section 3.6.2), the fluid velocity in both fluid phases is independent of the longitudinal coordinate, \( z \), and given by

\[
u_{nw} = u_w = \frac{dz}{dt}. \tag{4.12}
\]

For DNAPL/water displacement in the model fracture to correctly mimic the process of DNAPL/water displacement in the prototype, all of the terms in (4.11) must scale by the same factor. Incorporating (4.12) into (4.11) and examining the scaling of the last term of the right-hand side of (4.11) leads to

\[
\frac{\rho_j}{\rho_j} = \frac{n}{g} \quad (j = nw, w). \tag{4.13}
\]

This suggests that the same scaling factor, \( n \), should be used for each term in (4.11), i.e.

\[
\frac{\partial p_j}{\partial z} = \frac{n}{g} \frac{\partial p_j}{\partial z} \quad (j = nw, w), \tag{4.14}
\]
\[
\frac{\mu_j}{k_j} \frac{dz}{dt} = n \\
\frac{\mu_j}{k_j} \frac{dZ}{dT} = n \\
(j = nw, w), \quad (4.15)
\]

where the intrinsic permeability of the fracture is assumed to be similar in the centrifuge model and the prototype. This is possible since permeability is directly connected to fracture aperture [Bear, 1993], and microscopic dimensions remain invariant in the model and the prototype according to (4.8). Equation (4.14) can be verified if

\[
z = \left(\frac{1}{n}\right) Z, \quad (4.16)
\]

which, again, describes the centrifuge modeling requirements for the scaling of macroscopic length.

Equation (4.15) describes the scaling requirements for correct modeling of viscous pressure effects during a centrifuge test. For (4.15) to be valid

\[
t = \left(\frac{1}{n^2}\right) T. \quad (4.17)
\]

Darcy’s law given by (4.11) is a phenomenological law, only valid at the macroscopic scale, and cannot be directly derived from Navier-Stokes equations [de Marsily, 1986]. For example, the cubic law is derived from momentum equations for the flow in fractures with parallel walls, and extended to the flow in fractures where the walls are not parallel [Bear, 1993]. One of the main assumptions for using (4.11) relies on neglecting inertia forces. Inspecting the \(\alpha\)-number given by (3.86) (see Section 3.6.2) gives

\[
\alpha_n = \frac{\Delta \rho_w g n k_i^2}{\mu_i^2 I} = \frac{ng L}{g I} = n^2, \quad (4.18)
\]

where (4.16) is used for correct scaling of the tube length. Equation (4.18) shows that the magnitude of inertia forces associated with a system will be much higher in a centrifuge model test than the prototype. Upon modeling a DNAPL infiltration test using the centrifuge, it will be necessary to check that \(\alpha_n\) is negligible with respect to unity for its result to correctly reproduce the condition of an equivalent prototype. This effect is demonstrated experimentally and further discussed in Section 7.3.2.
Proper scaling of inertia forces requires that \( \alpha_n = \alpha \). Inspection of (4.18) shows that scaling of \( \alpha \) can be achieved if liquids \( n \)-times more viscous than water (\( n\mu_w \) instead of \( \mu_w \)) are used in the reduced-scale model. Re-inspecting (4.15) for correct modeling of viscous pressure effects during a centrifuge test now gives

\[
t = \left( \frac{1}{n} \right) T. \tag{4.19}
\]

Equation (4.19) describes the scaling law of time for proper scaling of inertia and viscous forces if liquids in the reduced-scale model are \( n \)-times more viscous than those in the prototype.

The existence of a conflict in porous media between inertia forces and viscous forces is well known by the centrifuge modeling community [Schofield, 1980]. For most porous media transport problem, it is assumed that the effects of inertia are negligible. Therefore, the time-scale factor is presumed to \( 1/n^2 \) as in (4.17).

### 4.4 Issues Related to Modeling of Dynamic Contact Angle

In Section 4.2 and Section 4.3, it was assumed that the dynamic contact angle, i.e. the contact angle of the moving interface, was a constant equal to the static contact angle throughout the infiltration process. Section 3.5.3 presented a theoretical prediction of the interface displacement where the dynamic contact angle was allowed to decrease with increasing velocity. The object of this section is to show that physical modeling of the DNAPL infiltration using the geotechnical centrifuge is complicated if the dynamic contact angle has a dependence upon velocity.

Consider a reduced-scale model of DNAPL infiltration into a vertical, circular-section capillary tube run at a gravitational acceleration equivalent to \( n \) times the Earth’s gravity. The dynamic contact angle depends on the interface velocity and is given by (3.64) (see Section 3.5.3.2). Using the scaling relationships listed in Table 4.1 and replacing them in (3.67) (see Section 3.5.3.2) leads to

\[
\frac{dZ_c}{dT} = \frac{K_L}{\left(1 + \frac{H}{L/n} \frac{K_L}{u_c} \frac{1 - \cos \theta_s}{\cos \theta_s} \right) L}\left[Z_c(T) + \Delta H\right], \tag{4.20}
\]

where use of (3.68) (see Section 3.5.3.2) has been made. For proper scaling of the DNAPL infiltration problem, the retardation factor, \( \gamma \), given by (3.68) must be equal in the reduced-scale model and in the full-scale prototype. From (4.20), it can be seen that this can only be achieved if the perfect wetting velocity, \( u_c \), is \( n \) times larger in the model than in the prototype—as is every parameter having dimensions of a velocity (see Table 4.1).
More generally, referring to the general governing equation (3.37) obtained in Section 3.4.5, scaling is complicated by the velocity-dependent term $\Delta h(dz_c/dt)$ given by (3.38) (see Section 3.4.5). Correct scaling of $\Delta h(dz_c/dt)$ requires that the cosine of the dynamic contact angle function for the model DNAPL be a function of interface velocity that increases $n$ times slower than that of the equivalent prototype. Assuming that the dynamic contact angle-interface velocity relationship is unique for a given DNAPL/water pair, this would mean selecting a different DNAPL for the reduced-scale experiment than that for the equivalent prototype such that the dynamic contact angle-interface velocity relationship is correctly scaled. A different DNAPL would be required for each targeted $g$-level. This obviously introduces a strong complication and shows a limitation associated with modeling dynamic capillary effects using the centrifuge. These issues are further examined and discussed in Section 7.3.5. In particular, it will be shown that scaling of centrifuge experiments is possible in the case where perfect wetting ($\theta_s = 0$) is achieved.

4.5 References


CHAPTER 5
DNAPLS AND THEIR PROPERTIES

5.1 Introduction

This chapter focuses on the dense non-aqueous phase liquids (DNAPLs) used for the experimental program of this research, namely 4-chlorotoluene and 1,1,1-trichloroethane. This chapter also describes experimental aspects related to the measurements of the viscosity and interfacial tension properties of these DNAPLs.

Section 5.2 examines the capillary tubes used for the measurements of the viscosity and interfacial tension properties of the DNAPLs, and later for the series of DNAPL infiltration experiments (see experimental methodology in Chapter 6).

Section 5.3 presents the DNAPL solvents used in the experimental program.

Section 5.4 focuses on the issue of safety and DNAPL solvent disposal, and presents the procedure used to clean equipment and glassware for both the properties measurements and the infiltration experiments.

Section 5.5 examines the viscosity property of DNAPLs.

Section 5.6 examines the interfacial tension property of DNAPLs. In particular, this section reports measurement of the interfacial tension using the pendant drop method and the combined capillary rise method.

A summary of the viscosity and interfacial tension properties of the DNAPLs is given in Section 5.7.

A list of references made in Chapter 5 is provided in Section 5.8.

5.2 Capillary Tubes

In order to simulate a range of fracture sizes for the DNAPL infiltration experiments, borosilicate glass capillary tubes of various diameters were used. These tubes were not only used for the laboratory and centrifuge infiltration experiments, but also for the viscosity and interfacial tension measurements described in this chapter. Thus, they will be discussed here.

The tubes were purchased from McMaster-Carr. They were furnished in a standard length of 305 mm (12 inches) and had nominal inner diameters of 0.6 mm, 1.2 mm, 2.2 mm and 2.7 mm, with outer diameters of 5 mm, 6 mm, 7 mm and 6 mm,
respectively. Each capillary tube that was used in the research was assigned a number in order to facilitate tracking and data collection during this work.

The average diameter of the capillary tubes used in the test series was computed by measuring the mass of water that each tube could store. In the past, this technique was often done using mercury threads [Padday, 1969]. To measure the stored mass of water, the dry mass of a tube was first determined with an electronic scale. Water was then pumped through the tube using a pipet filler. The capillary tube was then carefully disconnected from the filler and maintained in a horizontal position so that no water would drain out of the tube. Next, it was wiped with a paper towel in order to remove drops of water present on the outer wall of the tube. Its water-saturated mass was then determined with the electronic scale. Prior to the second mass measurement, care was taken to ensure that the tube was completely filled with water. The operation was repeated three times for each tube. The temperature of the water at the time of the operation was measured using a digital thermometer. The corresponding density of water at this temperature was obtained using correlation equations proposed by Wagner and Pruss [1993].

The average diameter \( d \) of a capillary tube was obtained using the simple formula

\[
d = \sqrt{\frac{4(m_f - m_e)}{\pi l \rho_w}},
\]

where \( m_f \) is the mass of the tube filled with distilled water, \( m_e \) is the dry mass of the tube, \( l \) is the length of the tube and \( \rho_w \) is the density of water. The relative error \( \Delta d/d \) due to uncertainty on the diameter was computed using (5.1) and given by

\[
\frac{\Delta d}{d} = \frac{1}{2} \frac{\Delta m_f}{m_f - m_e} + \frac{1}{2} \frac{\Delta m_e}{m_f - m_e} + \frac{1}{2} \frac{\Delta l}{l} + \frac{1}{2} \frac{\Delta \rho_w}{\rho_w},
\]

where \( \Delta m_f, \Delta m_e, \Delta l \) and \( \Delta \rho_w \) are the errors due to uncertainty on the mass of the tube filled with water, the dry mass of tube, the tube length and the water density, respectively.

The density of water at 20°C is approximately 0.998 g/cm³ [Lide, 1995]. For the range of water temperatures encountered in this work, the variation of water density with temperature [Lide, 1995], \( \Delta \rho_w \) did not exceed 0.001 g/cm³. Thus the relative error on density, \( \Delta \rho_w/\rho_w \), was less than 0.1%. The length of a capillary tube varied from 120 mm to 305 mm depending on how short the tube had been cut. The error on the measurement of length was less than 0.5 mm with a ruler graduated in millimeters. Thus the relative error on the tube length ranged from 0.2% to 0.4%. The errors on mass measurement \( \Delta m_f \) and \( \Delta m_e \) are related to the precision of the scale. This research used a scale with precision of one milligram. Hence, \( \Delta m_f \) and \( \Delta m_e \) were equal to 0.0005 g.

The mass of water, \( m_f - m_e \), that was contained in a tube varied with the diameter and length of tube. For example, for one of the 130 mm long, 0.6 mm diameter
capillary tubes, the stored water mass was of the order of 0.035 g. Thus, the relative error on mass, i.e. the sum of the first two terms on the right-hand side of (5.2), was of the order of 1.5%. Hence, most of the error due to uncertainty arose from the uncertainty associated with the measurement of mass. Under these conditions, it was not possible to determine the exact diameter of a 0.6 mm nominal diameter capillary tube beyond a two-digit precision.

For a 1.2 mm diameter capillary tube of similar length, the relative error on mass was closer to 0.35%, and the total relative error was less than 1%. Nonetheless, this also resulted in a precision of about one hundredth of a millimeter in the measurement of the tube diameter. For a 2.7 mm diameter capillary tube of similar length, the relative error on mass was about 0.07%. Again, with a total relative error close to 0.5%, a one-hundredth of a millimeter precision was achieved.

In practice, the overall error was larger than one-hundredth of a millimeter. Moisture present on the outer wall of a capillary tube sometimes increased the measured mass of water. Conversely, if the tube was not fully saturated with water and air was trapped within the tube, the measured mass of water was less than that of a fully saturated tube. Based on repeatability, the measurement of the mass of the tube filled with water gave an absolute mass error of 0.0015 g, i.e. three times that estimated above, whereas the measurement of the mass of the dry tube was quite repeatable (±0.0005 g).

Based on the measurement procedure, the nominal diameters supplied by the manufacturer, 0.6 mm, 1.2 mm, 2.2 mm, and 2.7 mm, were corrected to their measured diameters of 0.66 mm, 1.33 mm, 2.20 mm, 2.70 mm, respectively. In general, individual tubes did not have a diameter that differed from this value by more than 0.01 mm. In the remainder of this thesis, the measured diameter will be used when referring to a capillary tube.

5.3 Dense Non-Aqueous Phase Liquids

Two dense non-aqueous phase liquids (DNAPLs), 4-chlorotoluene and 1,1,1-trichloroethane were used in the research. These chemicals are described using separate headings below. Safety procedures and general recommendations for handling the chemicals are discussed in Section 5.4.1.

5.3.1 4-Chlorotoluene

For all centrifuge infiltration tests, and for the vast majority of the laboratory infiltration tests, 4-chlorotoluene (4-CT) was used for the DNAPL. This solvent was purchased from Aldrich Chemical Company, product number 11,192-9 [Aldrich, 2000]. It had a chemical purity of 98%, and contained no more than 2% of 2-chlorotoluene.
4-CT is listed in many organic chemical reference books or review papers on chlorinated solvents [Mercer and Cohen, 1990; Lide, 1995; Budavari, 1996] and is also known as p-chlorotoluene or 1-chloro-4-methylbenzene. It was selected because of its low density contrast with water, low toxicity compared to other DNAPLs and high flash point [Aldrich, 1997a]. Relevant properties of 4-CT are listed in Table 5.1.

With a measured solubility of 1.06 mg per liter of water at 20°C [Howard and Meylan, 1997], 4-CT can be considered virtually immiscible with water for the purposes of this research.

5.3.2 1,1,1-Trichloroethane

1,1,1-trichloroethane (1,1,1-TCA or more simply TCA) was used for a reduced number of laboratory infiltration tests. TCA is a common contaminant found at many hazardous waste sites, as discussed in Section 1.1. The solvent was also purchased from Aldrich, product number 40,287-7 [Aldrich, 2000]. It had a chemical purity of 99+% (ACS-reagent grade). Note that TCA is an ozone depleting substance, and thus falls into the Class I-controlled substance category [EPA, 1995]. Therefore, Aldrich asks its customers to sign a release form certifying that “the chemical purchased will only be used for laboratory applications and not be resold or used in manufacturing.”

TCA is widely referenced in the literature [Mercer and Cohen, 1990; Lide, 1995; Budavari, 1996; Pankow et al., 1996], and is sometimes referred to as methylchloroform [EPA, 1995] or chlorothene. TCA is reportedly more toxic than 4-CT, but this may simply be that the fate of TCA is particularly well documented [Montgomery, 2000] in comparison to 4-CT. In particular, TCA is listed as a possible mutagen [Aldrich, 1996].

With a solubility of 1.50 g per liter of water at 20°C [Howard and Meylan, 1997], TCA can also be considered virtually immiscible with water for the purposes of this research. Relevant properties of TCA are also listed in Table 5.1.

5.3.3 Sudan IV

Both 4-CT and 1,1,1-TCA are colorless. In order to increase their visibility and permit tracking of the liquids as they traveled through the fracture systems, the DNAPLs were dyed red using Sudan IV hydrophobic dye. Sudan IV has been commonly used in projects involving NAPL transport [Rimmer et al., 1996; Jørgensen et al., 1998; Longino and Kueper, 1999; Pantazidou et al., 2000]. Thanks to its bright color, it also allows quick detection of any contamination that may take place during the experimental procedure.

Sudan IV was purchased from Aldrich, product number 19,810-2 [Aldrich, 2000]. It is supplied in the form of a very fine red-brown powder with a dye content of 80%.
Because of its lipophilic nature, it is readily absorbed through skin, and dyes just about every organic material that it comes into contact with. In addition, it is listed as a possible mutagen \textit{[Aldrich, 1997b]}. It was the most hazardous compound dealt with in this experimental project.

Dyed 4-CT and TCA were prepared by adding 0.2 g of Sudan IV to one liter of solvent and then stirring the mixture. Dyed solvents were typically kept in cleaned and labeled one-liter amber glass bottles. Cleaning procedure of glassware is described in Section 5.4.2. Even with such low concentrations of dye, the DNAPLs had a bright red color.

<table>
<thead>
<tr>
<th>Table 5.1. Common Properties of 4-CT, TCA and Water</th>
</tr>
</thead>
<tbody>
<tr>
<td>4-CT</td>
</tr>
<tr>
<td>Density at 20°C [g/cm$^3$]$^a$</td>
</tr>
<tr>
<td>Solubility in Water [g/l]$^b$</td>
</tr>
<tr>
<td>Flash Point [°C]$^c$</td>
</tr>
</tbody>
</table>

$^a$ \textit{Lide} [1995].

$^b$ \textit{Howard} [1997].

$^c$ \textit{Aldrich} [1997a].

$^d$ \textit{Aldrich} [1996].

5.4 Safety, Disposal and Cleaning Procedures

5.4.1 General Safety Considerations for Handling 4-CT, TCA and Sudan IV

General chemical laboratory safety and hygiene precautions had to be kept when running the experiments. Material safety data sheets (MSDS) are available from the chemical manufacturers or various websites, such as the Safety and Information Resources website at http://hazard.com/msds/index.php. Other resources include the Department of Chemistry website at http://web.mit.edu/chemistry/www/.

It is important to emphasize on a number of safety precautions that were used during the experimental part of this research, including working with DNAPL solvents and disposing of liquids or solids contaminated with solvents. It is strongly recommended that a formal chemical safety and hygiene training be dispensed to any prospective student involved in similar work in the future. This section summarizes common practices used during experimental work and taught to students that were involved in the project. Unless specified, glassware, small equipment such as Teflon
seal tape or hypodermic syringes, and common solvents such as acetone or methanol were obtained from the MIT laboratory supply stockroom (MIT room 56-070) or ordered from VWR through the MIT stockroom.

Generally, materials referred to as plastics are attacked by 4-CT and TCA in addition to being dyed by Sudan IV. Thus, material such as Lexan polycarbonate, commonly used for the construction of clear centrifuge strong boxes, could not be used for this experimental program. The effect of 4-CT on sheets of Lexan on which screw holes had been machined was examined. Cracks between screw holes would typically form between holes and split the sheet into several pieces. Moreover, acetone, a solvent commonly used for cleaning up DNAPLs, also reacted with plastics. Thus, whenever clear material was necessary for visualizing the experiment, only glass could be used. If Lexan, or other materials such as Teflon seal tape, RTV silicone adhesive sealant or plastic syringes, were to be used, exposure to the chemicals was minimized, or the material was used only once and then disposed of as hazardous waste. Water containing dissolved DNAPL, although hazardous because of its health toxicity, did not constitute a reactive chemical.

In addition to be reactive with plastics, 4-CT and TCA are strong oxidizing agents [Aldrich, 1996; Aldrich, 1997a]. TCA was observed in the laboratory to react with small pieces of aluminum to form a brownish oxide. Again, exposure of aluminum parts to 4-CT or TCA was minimized. Stainless steel parts should be used if prolonged exposure to pure DNAPLs is required for experimental applications.

Whenever possible, experiments were conducted in one of the two fume hoods located in the Department of Civil and Environmental Engineering at MIT, room 1-335 or room 1-047. Such experiments include glassware cleaning and solvent preparation or disposal, interfacial tension measurements, and DNAPL infiltration experiments in capillary tubes of small length (typically less than 250 mm). DNAPL infiltration experiments in longer capillary tubes were conducted next to the fume hoods in either one of the rooms listed above. Centrifuge infiltration experiments were prepared in the fume hood of room 1-047 and then transported to the centrifuge laboratory. According to the MIT Industrial Hygiene Office, small amounts of 4-CT or TCA can be handled outside the fume hood without any health risk. Generally, quantities of DNAPL exceeding 200 ml were not left in open containers, whether inside or outside the fume hood.

In order to limit the consequences of a spill, jugs of solvent and solvent-containing glassware were generally kept in secondary containment. In addition, solvent absorbent was stored in the laboratory. During any experimental procedure, a laboratory coat, safety glasses and gloves were generally worn at all times. The Industrial Hygiene Office recommended using Viton gloves for handling 4-CT or TCA or possibly Silver Shield gloves for TCA. Given their high cost and with virtually no cut or abrasion resistance [VWR, 1999], Viton gloves were not considered adequate. Nor were Silver Shield gloves, which were not very flexible and made precision work difficult. Despite little chemical breakthrough resistance [VWR, 1999], latex gloves were selected. Typically, two pairs were worn, and the upper glove was replaced whenever contamination was observed on this glove. Chemical breakthrough to the second glove was never observed.
Equipment contaminated with DNAPL, such as glassware and reusable needles, was rinsed with acetone until traces of dye could not be seen, and then rinsed with plenty of water to wash the acetone. Only then could the glassware be handled without gloves. Typically, acetone and water were dispensed using a wash bottle. Rinsing was made above a large glass funnel connected to a 1-gallon waste glass jug. Dry, non-reusable material contaminated with DNAPL, such as gloves, Teflon tape, paper towels, syringes, disposable needles (with cap on), and the bench liner (bench protector), was collected in a polyethylene bag and labeled as toxic solid waste containing traces of solvent and Sudan IV dye.

During the project, four types of waste were generated. The first type was the solid waste described above. The second type was a mixture of 4-CT (or TCA), water, acetone, and sometimes methanol, generated by the experiments and during cleaning of the equipment. This mixture was always stored in glass jugs and labeled as toxic waste. The third type of waste, consisting of water containing dissolved DNAPL, was stored in high density polyethylene (HDPE) jugs. Note that neither Sudan IV, nor acetone, nor pure DNAPL phase was present in this waste, which was essentially generated in DNAPL infiltration laboratory or centrifuge experiments, and consisted of water pumped out of the tank tube (see Section 6.2.2) or centrifuge box containing the experimental setup (see Section 6.3.4). This waste was also labeled as toxic waste. The fourth type of waste was a mixture of water, acetone and methanol, and was generated during the cleaning of glassware that took place prior to any experiment (see Section 5.4.2). This waste was free of dye and DNAPL, stored in HDPE jugs, and labeled as toxic and ignitable waste.

Waste jugs were either purchased at the MIT stockroom or were recycled solvent bottles. Waste was labeled using the MIT Safety Office hazardous waste red tags and stored in the satellite accumulation area of MIT room 1-335 or room 1-047 with secondary containment. Once a container was full, the date was written on the tag. Containers must be removed from the satellite accumulation area within three days after the waste container is full and dated. Removal is free of charge and ordered from the MIT Environmental Health and Safety Team by filling a pick-up request at http://web.mit.edu/environment/wastepickup/index.html.

5.4.2 Cleaning Procedure

This section describes the procedure followed to clean glassware for storing solvents or prior to running experiments involving DNAPLs. Cleanliness of glassware is a very important step, and has been shown to influence the results of liquid/fluid displacement in capillary tubes [Blake et al., 1967; Jeje, 1979; Berezkin and Churaev, 1982; Ichikawa and Satoda, 1994].

Various methods have been cited in the capillary flow literature for achieving clean glass surfaces of capillary tubes. Ligenza and Bernstein [1951] cleaned their
tubes with a solution of nitric acid for 10 minutes, followed by 15 minutes of washing with distilled water and 10 minutes of drying by drawing air through the tube. Other researchers used similar approaches, but used chromic acid—a mixture of potassium dichromate and sulfuric acid—instead of nitric acid [Siegel, 1961; Ichikawa and Satoda, 1994; Hamraoui et al., 2000; Zhmud et al., 2000]. Van Remoote and Joos [1993] used chromic acid over a period of days. Fermigier and Jenffer [1991] used acetone and boiling deionized water prior to chromic acid. Jeje [1979] used several cycles of chromic acid, ultrasonic and detergent cleaning. Mumley et al. [1986] started with isopropyl alcohol and followed with an Alconox detergent solution, ultrasonic cleaning and a solution of nitric acid. Schäffer and Wong [2000] used a solution of hydrochloric acid and then boiled the tubes in deionized water for several hours, but suggested that organic contaminants may still be present. Blake et al. [1967] cleaned the tubes with a sequence of hot chromic acid, ethanol, nitric acid and distilled water. Finally, Calvo et al. [1991] used their tubes only once without preliminary cleaning procedure. The authors argued that the manufacturing process ensured a smooth and clean surface.

The chromic acid cleaning procedure described above is effective at removing organic contaminants [Coyne, 1997]. However, chromic acid constitutes a hazard due to its highly corrosive nature. It is also difficult to dispose of potassium dichromate-based waste. Moreover, many of the experiments reported here involved large equipment, such as the centrifuge box, or the reservoirs used to contain the long capillary tubes. Thus, the use of chromic acid would have been hazardous, if not futile, given the precautions that would have been subsequently involved to protect the cleaned surfaces.

The cleaning method for this research was based on the procedure used by Rashidnia et al. [1992] for their capillary rise experiments. Following an experiment, glassware was rinsed with acetone and water to remove the DNAPL, as described in Section 5.4.1. Afterwards, the glassware was allowed to stand overnight in a solution of Alconox. It was then rinsed with distilled water, acetone, methanol and distilled water in that order. Acetone and methanol are two non-polar solvents typically used to clean glassware containing organic residues, methanol being the more polar of the two [Roberts et al., 1994; Coyne, 1997]. Finally, the glassware was oven dried at 120°C for an hour. The use of compressed air for drying is time saving but unfortunately a poor practice, as compressed air may contain droplets of oil, water and particles of rust [Fieser and Williamson, 1987].

If the glassware was new or had never been used before for this research, the glassware was first rinsed with hydrochloric acid prior to the cleaning procedure described above. This was an effective way of removing contamination resulting from calcium or other alkali deposits [Coyne, 1997].
5.5 Viscosity Measurements

5.5.1 Background

Viscosity data for common organic compounds and correlations between viscosity and temperature are widely available in the literature [Viswanath and Natarajan, 1989; Lide, 1995; Yaws, 1995; Yaws et al., 1999]. Table 5.2 gives the dynamic viscosity of 4-CT, TCA and water for different temperatures. As shown in Section 3.5.4, the exact value of the viscosity of the DNAPL is not needed for the prediction of the infiltration kinetics if the viscosity of the DNAPL does not differ significantly from the viscosity of water. As shown in Table 5.2, neither the viscosity of 4-CT nor the viscosity of TCA differs from the viscosity of water by more than 13%.

Table 5.2. Dynamic Viscosity of 4-CT, TCA and Water

<table>
<thead>
<tr>
<th></th>
<th>4-CT</th>
<th>TCA</th>
<th>Water</th>
</tr>
</thead>
<tbody>
<tr>
<td>Viscosity at 25°C [mN.s/m²]</td>
<td>0.834ᵃ, 0.837ᵇ</td>
<td>0.810ᵃ, 0.793ᵇ</td>
<td>0.900ᵇ</td>
</tr>
<tr>
<td>Viscosity at 20°C [mN.s/m²]</td>
<td>0.8925ᶜ, 0.900ᵃ</td>
<td>0.867ᵃ</td>
<td>1.002ᵇ</td>
</tr>
<tr>
<td>Viscosity at 16.5°C [mN.s/m²]</td>
<td></td>
<td>1.109ᵇ</td>
<td></td>
</tr>
</tbody>
</table>


It is possible that the presence of Sudan IV in the solvents affects their viscosities. Viscosity data for Sudan IV-dyed 4-CT and TCA were not readily available in the literature. Thus, it was necessary to check whether the dye had any effect on the viscosity of a pure compound. For that purpose, a simple falling head technique was used. In this technique (see illustration in Figure 5.1), a long capillary tube was set up vertically in the fume hood on top of a beaker. The upper part of the capillary tube was connected to a long graduated reservoir tube. The liquid of viscosity to be measured was quickly poured into the reservoir tube. Flow was controlled by the capillary tube, which had a much smaller diameter than the reservoir tube. The effluent was collected into the beaker, but the lower end of the capillary tube remained open to the air, so that the head could be assumed constant at the exit of the tube. As the liquid was draining through the reservoir tube, the head of the liquid slowly decreased along with the velocity of flow. This technique is well known in soil mechanics as the falling head test, and is used for the measurement of the hydraulic conductivity of soil samples [Lambe and Whitman, 1969; de Marsily, 1986].

Assuming viscous flow and applying Hagen-Poiseuille’s law [Munson et al., 1994] (see (2.3) in Section 2.3.2) between the time \( t \) and the time \( t + dt \), it can be showed that the dynamic viscosity of the liquid of interest, \( \mu \), is given by
\[ \mu = \left( \frac{\rho g r_0^4}{8 l r_2^2} \right) \frac{t_2 - t_1}{\ln \left( \frac{h_1 + l}{h_2 + l} \right)}, \]  

(5.3)

where \( r_0 \) and \( l \) are the radius and length of the capillary tube, respectively, \( r_r \) is the radius of the reservoir tube, \( g \) is the Earth's gravitational acceleration, \( \rho \) is the density of the liquid, and \( h_1 \) and \( h_2 \) are the heights of liquid contained in the reservoir tube at time \( t_1 \) and \( t_2 \) respectively (see Figure 5.1).

**Figure 5.1.** Schematic illustration of experimental setup used for viscosity measurements.

### 5.5.2 Methodology, Results and Conclusions

For the viscosity measurements, a 126 mm long 0.66 mm diameter capillary tube was used. The capillary tube was connected to a 230 mm long, 7.5 mm diameter reservoir tube. The reservoir tube was graduated every milliliter, corresponding to a graduation every 22.5 mm. The viscosity of Sudan IV-dyed 4-CT was measured at 20°C. In addition, for comparison purposes, the viscosity of water at 16.5°C was also measured. Three separate runs were performed for each liquid. During each run,
liquid was added to the top of the reservoir tube. A stopwatch was started when the liquid level dropped down to the first graduation corresponding to the reference height \( h_1 \) and time \( t_1 = 0 \) in (5.3). The time was then recorded whenever the liquid level passed a graduation. Eight data points \((t_2, h_2)\) were obtained for each run using this technique. A typical time between two consecutive readings was of the order of ten to twenty seconds.

Viscosity values were calculated from (5.3) using the density properties given in Table 5.1 and a gravitational acceleration \( g = 9.807 \text{ m/s}^2 \) [Munson et al., 1994]. The average viscosity was obtained from the three sets of 8 data points. The viscosity did not appear to be a function of the liquid level in the reservoir tube. The results of these measurements are shown in Table 5.3.

| Table 5.3. Measured Viscosity of Water and Sudan IV-dyed 4-CT Using the Falling Head Technique |
|-----------------------------------------------|-------------------|
| Dyed 4-CT (20°C) | Water (16.5°C) |
| Average viscosity [mN.s/m²] | 0.918 | 1.136 |
| Standard deviation [mN.s/m²] | 0.004 | 0.015 |

Both values of viscosity reported in Table 5.3 are reasonably close to their anticipated value, although larger than the values reported in the literature (see Table 5.2) by about 2.5%. In Section 3.4.2, it was shown that if a liquid of density \( \rho \) and viscosity \( \mu \) flows at a representative velocity \( u_0 \) in a tube of radius \( r_0 \) and length \( l \) with \( r_0/l << 1 \), such that the following condition is true

\[
\frac{\rho r_0^2 u_0}{\mu l} << 1,
\]

then inertia forces can be neglected with respect to viscous forces. The left-hand side is the dimensionless ratio of inertia to viscous forces and is similar to the Reynolds number [Munson et al., 1994].

A representative velocity \( u_0 \) for the viscosity experiments reported here can be estimated from Hagen-Poiseuille’s law and is found to be of the order of 350 mm/s. Using this value of \( u_0 \), the left-hand side of (5.4), is approximately 0.3, suggesting that inertia forces are not completely negligible with respect to viscous forces. Because, the inertia forces may act to slow down the flow process, the measured viscosity may appear larger than expected. Nonetheless, the error of 2.5% between viscosity measurements and values reported in the literature is reduced in magnitude, and acceptable for the intended purpose of these experiments.

Overall, the viscosity measurement results support the conclusion that the Sudan IV dye does not have a large effect, if any, on the viscosity of the pure DNAPL compounds.
5.6 Interfacial Tension Measurements

5.6.1 Background

Interfacial tension and surface tension data can be found in the literature for a number of common organic compounds [Mercer and Cohen, 1990; Demond and Lindner, 1993; Lide, 1995]. Interfacial tension data are scarce in comparison to surface tension data. They are usually less reliable and are available only at a given temperature. Again, as presented in Section 2.2.1.2, the term surface tension refers to the interfacial tension of a liquid with its vapor, whereas interfacial tension refers to the interfacial tension between the liquid and water. Table 5.4 gives the values of interfacial and surface tensions of 4-CT and TCA found in the literature. Note that the interfacial tension of 4-CT with water could not be found.

It is possible that the presence of Sudan IV affects the value of the interfacial tension of TCA and 4-CT with water. Indeed, it is well known that dirt, or any other contamination, may significantly change the interfacial tension [Padday, 1969]. Interfacial tension data for Sudan IV-dyed 4-CT and TCA were not available in the literature. Thus, this property had to be measured independently in the laboratory.

Table 5.4. Surface and Interfacial Tensions of 4-CT and TCA

<table>
<thead>
<tr>
<th></th>
<th>4-CT</th>
<th>TCA</th>
</tr>
</thead>
<tbody>
<tr>
<td>Surface tension</td>
<td>34.93-0.1082</td>
<td>28.28-0.1242</td>
</tr>
<tr>
<td>Interfacial tension</td>
<td>45</td>
<td></td>
</tr>
</tbody>
</table>

a After Mercer and Cohen [1990].

b The surface tension is given as a set of values, $a-b$; surface tension $= a - bT$ where $T$ is the temperature in °C.

5.6.2 Methods of Measuring Interfacial (or Surface) Tension

Because the measurement of surface tensions, interfacial tensions and contact angles are of primary importance in a number of applications of surface science, several techniques have been developed for this purpose. Conventional methods of measuring interfacial tension include force methods such as the drop weight, Du Noüy ring or the Wilhelmy plate, shape methods, such as the sessile or pendant drop, and several other methods such as differential capillary rise, maximum bubble pressure, light scattering, and the spinning drop. These methods have been described in a number of reviews [Padday, 1969; Rusanov and Prokhorov, 1996; Adamson and Gast, 1997]. Methods potentially convenient for measuring the interfacial tension of DNAPL and water are briefly discussed here under separate headings.
5.6.2.1 Du Noüy Ring Method

Certain MIT laboratories in the departments of chemical and mechanical engineering own commercial tensiometers. Interfacial or surface tension can be measured with these devices using the Du Noüy ring method, which is among the most common methods. In the ring method, a liquid, for which surface tension is to be measured, is poured in a small container. A ring—usually made of platinum—is lowered horizontally into the liquid and then pulled out (see Figure 5.2). The force required to detach the ring from the liquid can be correlated to the surface tension of the liquid [Adamson and Gast, 1997]. For measuring interfacial tensions, the technique is identical, except that the ring is pulled from the wetting phase to the non-wetting phase. This means that the ring method is restricted to LNAPLs, unless the device is equipped such that the ring can be pushed downwards. No such device was available at MIT.

![Figure 5.2. Schematic illustration of the Du Noüy ring method [after Adamson and Gast, 1997].](image)

5.6.2.2 Spinning Drop Method

The spinning drop method was also investigated as a mean to obtain the interfacial tension of the liquids used in this research [Holguin, 1999; Levy et al., 2001]. Traditionally, the spinning drop technique enables measurement of ultra low interfacial tension, such as the interfacial tension of LNAPL/surfactant/water systems [Cayias et al., 1975; Cai and Mohanty, 1997]. This technique seemed like a relevant method if research work was to be extended to the interaction of DNAPL and water with surfactants or co-solvents.

In a spinning drop device, a sample tube filled with the denser liquid contains a drop of the lighter fluid (liquid or gas). The sample tube rotates horizontally around its axis (see Figure 5.3). In the presence of the centrifugal forces, the lighter fluid drop
elongates along the axis of the tube. If the rotational speed is high enough, the shape of the drop can be assumed to be an infinite cylinder whose diameter can be measured. Knowledge of this diameter, as well as the speed of rotation, allows derivation of the interfacial tension [Vonnegut, 1942; Torza, 1975; Manning and Scriven, 1977].

Figure 5.3. Schematic illustration of the spinning drop method: A, lighter fluid; B, denser liquid [after Adamson and Gast, 1997].

Conventional spinning drop tensiometry makes use of a cylindrical sample tube, for which the diameter of the drop is magnified by a factor, which has to be measured or estimated [Bock, 1984; Puig et al., 1992]. This factor is often assumed to be equal to the index of refraction of the denser, outer phase, usually water. While indexes of refraction for many liquids are readily available from the literature, there are instances where it is not known, for example, in a mixture of DNAPLs. The use of a square section sample tube, demonstrated by Levy et al. [2001], enables the measurement of interfacial tension without prior calibration or knowledge of any index of refraction. Nevertheless, when measuring the interfacial tension of a DNAPL/water system with this method, the glass surface has to be made hydrophobic as pointed out by Cai and Mohanty [1997]. A second drawback of this method is that the interfacial tension is proportional to the cube of the diameter of the drop, which typically is of the order of one to two millimeters for the drop of a water/NAPL system rotating at a few thousand RPMs. Thus an error due to uncertainty of one to two hundredths of a millimeter on the drop diameter translates into a 3% relative error on the interfacial tension. Under these conditions, careful design, particularly vibration protection, has to be achieved if accuracy in the measurement is desired [Seeto and Scriven, 1982].

5.6.2.3 Pendant Drop Method

Surface tension tends to make a liquid drop hanging from the tip of a small diameter tube take the form of a sphere. In the Earth’s gravitational field, however, the sphere is distorted into an elongated tear drop shape as illustrated in Figure 5.4. The distortion increases with the liquid density and decreases with its surface tension [Ambwani and Fort, 1979]. If the density of the liquid is known and the drop shape can be accurately captured, then the surface tension can be calculated. If the drop is hanging in a bath of liquid of lower density—or pulling upwards in a bath of liquid of
larger density—then the interfacial tension of the pair of liquids can be obtained from the shape of the drop and the density contrast between the two liquids. This method was successfully used for the first time by Andreas et al. [1938], who computed the interfacial tension of a pair of liquids from the equatorial diameter of the drop and a simple geometrical construction that allowed them to derive a table of shape factors (see Section 5.6.3.1).

Several important advantages compared to other measurement techniques are listed by Ambwani and Fort [1979] in their detailed review of the method. In particular, the pendant drop method is very versatile and requires only small samples of liquids. Because the method does not disturb the surface, aging effects can be investigated. Moreover, the shape of the drop is independent of the contact angle made by the drop with the tube from which it hangs, an important issue when it comes to the capillary rise technique (see Section 5.6.4.3). Finally, for the purposes of this research, the interfacial tension of DNAPL with water can simply be obtained from a small drop of DNAPL hanging in a bath of water.

As part of the experimental program of this research, the pendant drop method was used for measuring the interfacial tension of 4-CT with water. Theoretical developments, experimental methodology and results using the pendant drop technique are presented in Section 5.6.3.

![Schematic illustration of a pendant drop](after Ambwani and Fort, 1979).

Figure 5.4. Schematic illustration of a pendant drop [after Ambwani and Fort, 1979].

5.6.2.4 Capillary Rise and Combined Capillary Rise Methods

The capillary rise method is considered one of the most accurate methods of all to measure surface tension (as opposed to interfacial tension), partly because the theory
has been worked out with considerable exactitude, and partly because the experimental variables can be closely controlled [Padday, 1969; Adamson and Gast, 1997].

If a capillary tube is partially or completely wetted by a liquid contained in a beaker, the liquid rises up inside the tube to some equilibrium position as illustrated in Figure 5.5. The surface tension of the liquid can be obtained from the height of the meniscus inside the tube above the level of the free surface of the liquid. Because of its transparency and because it is wet by most liquids, a glass capillary tube is commonly used. The glass must be very clean. The tube must be precisely vertical, and have an accurately known and uniform radius [Adamson and Gast, 1997].

![Figure 5.5. Schematic illustration of the capillary rise method.](image)

The combined capillary rise method [Rashidnia et al., 1992] makes use of a capillary tube to measure the interfacial tension between two liquids. This method is not as common as the capillary rise method, perhaps because the former is associated with more uncertainty.

The combined capillary rise method to measure the interfacial tension between a NAPL and water is basically a two-step method. During the first step, measurement of the NAPL surface tension is performed using the conventional capillary rise method described above (see Figure 5.5). NAPL must be wetting with respect to air, which is usually the case (see Section 2.2.1.3).

Following the rise, the capillary tube containing a NAPL column is transferred to a second beaker, which contains water. Once dipped into the second beaker (see Figure 5.6), water rises in the capillary tube above the free surface of water. Such rise takes place since water is usually wetting with respect to NAPL (again, see Section 2.2.1.3). By measuring the height of water rise as well as the length of the NAPL column, the
interfacial tension between NAPL and water can be calculated. The surface tension of NAPL measured in step 1 is used for the calculation.

![Diagram of capillary rise method](image)

**Figure 5.6.** Schematic illustration of step two of the capillary rise method.

The combined capillary rise method has a number of advantages compared to other measurement techniques. In particular, the method is extremely versatile. It does not require any specific device but a calibrated glass capillary tube, which were commonly used for this research. Thus, as part of the experimental program, the combined capillary rise method was used for measuring the interfacial tensions of 4-CT and TCA with water. Theoretical developments, experimental methodology and results using this technique are presented in Section 5.6.4.

### 5.6.3 Interfacial Tension Measurements Using the Pendant Drop Method

#### 5.6.3.1 Theory

Referring to the pendant drop shown in Figure 5.7.a, the profile of the drop is given by [Patterson and Ross, 1979]

\[
\frac{2\sigma}{r_a^*} + \Delta \rho g r = \frac{1}{R_{P,1}} + \frac{1}{R_{P,2}},
\]

(5.5)
where $\sigma$ is the surface or interfacial tension, $g$ is the Earth’s gravitational acceleration, $\Delta\rho$ is the density contrast between the two phases, $R_{P,1}$ and $R_{P,2}$ are the principal radii of curvature at any point $P$ of the drop profile having coordinates $(x, z)$, and $r_a$ is the radius of curvature at the apex (point $A$ with $x = z = 0$ in Figure 5.7.a).

Both radii of curvature at $P$ can be expressed as a function of $x$, $z$ and $dz/dx$, thereby yielding a differential form of (5.5) that must be integrated numerically. The solution gives values of $z(x)$ as a function of $r_a$, $\sigma$, $g$ and $\Delta\rho$.

The mathematical difficulties associated with this numerical problem were first circumvented by Andreas et al. [1938] who defined a conveniently measurable shape dependant quantity, $S = d_i/d_e$. As shown in Figure 5.7.b, $d_e$ is the equatorial diameter of the drop and $d_i$ is the diameter measured at a distance $d_e$ from the bottom of the drop. The method is known as the method of the selected plane. After simplification, (5.5) can be rewritten as [Andreas et al., 1938]

$$\sigma = \frac{\Delta\rho g d_e^2}{H},$$

where $1/H$ is a function of the drop shape parameter $S$. Andreas et al. [1938] evaluated $1/H$ corresponding to a range of values of $S$ from 0.700 to 1.000 using a pendant drop of water. The tables were somewhat inaccurate because of the empirical nature of the method. Numerical integration procedures of (5.5) were later developed [Fordham,
Series of tables were developed giving $1/H$ for $S$ varying from 0.300 to 1.002. Complete tables have been reproduced in recent reviews [Rusanov and Prokhorov, 1996; Adamson and Gast, 1997]. For computer applications, equations that best fit the $1/H-S$ data have been developed [Misak, 1968] and are quite accurate for most calculations of surface or interfacial tension [Ambwani and Fort, 1979]. The method of the selected plane requires that the drop have an equator. Alternatives for drops of any shape are described in Rusanov and Prokhorov [1996].

As pointed out in later work [Patterson and Ross, 1979; Rotenberg et al., 1983], the major source of error in the method proposed by Andreas et al. [1938] is that the whole surface of the drop is reduced into the measurement of a few pre-selected critical points which are compatible with the use of the tables. Referring to Figure 5.7.b, an error in the measurement of $d_e$ causes an error in the opposite direction in $d_e$, so that the error is even more important in the shape parameter $S = d_e/d_e$ [Patterson and Ross, 1979]. For example, if $d_e$ is overestimated, the plane corresponding to $d_e$ is reported closer to the tip, such that $d_e$ and $S$ are underestimated. This in turn overestimates the interfacial tension since the drop is now perceived as rounder. Error estimation is further discussed in Section 5.6.3.4.

By using many more points on the experimentally observed profile, the influence of random errors can be minimized. One approach to obtain a measure of the interfacial tension is to use curve-fitting techniques and minimize the deviation between the theoretical profile given by (5.5) and the experimental points. The development of video techniques including picture digitalization has significantly facilitated this optimization method [Rusanov and Prokhorov, 1996]. Computer-aided image processing of drop profiles has been successfully used in a number of studies [Patterson and Ross, 1979; Anastasiadis et al., 1987; Song and Springer, 1996a; Zeppieri et al., 2001].

Although this work obtained digital images of pendant drops as described in Section 5.6.3.2, the study was limited to the method of the selected plane to obtain a measure of interfacial tension.

5.6.3.2 Experimental Methods

A diagram of the setup used to obtain pictures of pendant drops is shown in Figure 5.8.a. A photograph of the experimental setup is shown in Figure 5.8.b. The apparatus is set up in the fume hood. It consists essentially of a light source, a pendant drop cell and syringe assembly, a microscope lens mounted to a camera, and a computer for image acquisition. Although diffused light was used to illuminate the drop, a collimating lens located between the light source and the pendant drop cell is generally recommended to achieve parallelism of the passing light beams and improve the sharpness of the drop contour [Ambwani and Fort, 1979; Rusanov and Prokhorov, 1996]. Another important requirement is that the walls of the cell be made of an optically flat material and positioned perpendicular to the optical axis of the measuring
Indeed, similar to the spinning drop method (see Section 5.6.2.2), a cylindrical cell would magnify the drop by a factor related to the index of refraction of the outer liquid, thereby changing the values of the shape factor and interfacial tensions. The cell used here was built using a set of five rectangular glass micro slides of dimensions 25 mm by 75 mm (VWR) glued together using RTV silicone adhesive sealant. The fifth slide was used for the bottom face.

The pendant drop method was used to measure the interfacial tension of three different DNAPLs. The first DNAPL was Sudan IV-dyed 4-CT, which was used for most of the experimental program of this research. The second DNAPL was undyed 4-CT for the purpose of assessing the effect of the dye on the interfacial tension of 4-CT. Finally, the interfacial tension of ACS reagent-grade trichloroethylene (TCE) was measured. This solvent of high purity was purchased from J.T. Baker and used in prior experimental work [Sinfield, 1998]. TCE is a common DNAPL of density 1.4642 g/cm³ [Demond and Lindner, 1993] and interfacial tension with water 0.0393 N/m [Seo and McCray, 2002] (a value of 0.0345 N/m has also been reported [Seo and McCray, 2002]). All the measurements are reported at room temperature, i.e. 19 ± 1°C.

For each measurement, the cell was first filled with distilled water. Conditions of preparation varied and are further described in Section 5.6.3.3. A small amount of DNAPL was then drawn into a hypodermic syringe, and poured into a tube whose lower end was immersed in the cell (see Figure 5.8). Next, a pendant drop of DNAPL was formed at the end of the tube. The size of the pendant drop varied as a function of the DNAPL head inside the tube.

Tubes of varying inner diameters (ID) and outer diameters (OD) were used for comparison. The following tubes were used: a thin-walled brass tube of 3.16 mm OD (2.06 mm ID), a thin-walled aluminum tube of 5.01 mm OD (4.65 mm ID), and a thick-walled glass capillary tube of 6.02 mm OD (2.70 mm ID). The diameter of the tubes appeared to influence the size and shape of the drop but not the final estimated value of the interfacial tension. Note that neither brass, nor aluminum is a good material choice as a reaction between the metals and DNAPL might take place. Glass, stainless steel or even titanium would have been preferable [Rusanov and Prokhorov, 1996]. In addition, the choice of the tube diameter depends on the properties of the liquid/fluid pair under investigation. For example, in the case of the 4-CT/water pair, it was impossible to use small diameter tubes (i.e. small needle gauge) as the drop would stick to the outer walls of the tube and could not be detached to form a hanging drop.

Once a drop was formed, it was ensured that the total height of the drop was at least 1.2 ᴅₛ, so that the edge of the tube did not affect the drop geometry and the value of ᴅₛ (see Figure 5.7.b). Although no specific recommendation could be found in the literature, it seemed necessary to rule out the potential impact of boundary effects on the value of the interfacial tension.
Figure 5.8. Pendant drop experimental setup: a. Schematic diagram [adapted from Zeppieri et al., 2001]; b. Photograph of the apparatus; 1, camera head; 2, microscope lens; 3, syringe; 4, tube containing DNAPL; 5, visualization cell containing water and DNAPL; 6, diffused light source; 7, computer and image acquisition software.
Following its formation, drop images were acquired using a microscope zoom video lens mounted on a CCD camera. A *Magna Fire* CCD camera head, model S60800, was used. It was purchased from Optronics, Goleta, California, and used in prior work at MIT [Castenson, 2000]. The camera head contained an 8.7 mm × 6.9 mm active area sensor, also known as a 2/3 inch format sensor. Captured images were 1280 pixels wide by 1024 pixels high. The camera accommodated most industrial lenses through a standard threaded C-mount.

The microscope zoom video lens was a model VZM 450i purchased from Edmund Industrial Optics. Its primary magnification factor could be varied from 0.7× to 4.5×, corresponding to a horizontal field of view ranging from 1.9 mm to 12.5 mm for a 2/3 inch sensor [Edmund Industrial Optics, 2001]. In other words, one pixel was equivalent to 9.7 µm at the smaller magnification and 1.5 µm at the largest magnification.

A built-in iris enabled control of the amount of light passing through the lens, and hence the depth of field and time of exposure of images. Complete specifications of the lens as well as general design guidelines can be found in *Edmund Industrial Optics* [2001].

The lens iris aperture was set such that sufficient depth of field allowed the drop and tube from which it was hanging to appear focused. On the one hand, it was important to have enough contrast between the drop and the background so as to ensure correct edge detection. On the other, excess light or a large time of exposure would have led to *burning* of the edges of the drop and incorrect measurement of diameters. A comprehensive discussion on edge detection can be found in *Song and Springer* [1996b].

A capture board and a Windows compatible Magna Fire camera software package were used to acquire images of drops on a computer. Monochrome (grayscale) images were *snapped* at a time of exposure of 1.037 s. Each image file was in the bitmap format and took 1.25 MB of disk space. Examples of DNAPL drop images are shown in Figure 5.9. The camera was set so as to optimize the size of the drop in the sensor field of view, explaining why the drops appeared horizontal on the computer screen.

The Windows compatible image analysis software Image-Pro Plus, version 4.0, was used to measure the drop size and shape factor. A simple pixel counting image software such as Microsoft Photo Editor would also have worked. The number of pixels corresponding to the equatorial diameter, \(d_e\), was counted. The distance \(d_e\) was then reported along the axis of revolution of the drop and the diameter \(d_s\) was obtained (see Figure 5.7b). As mentioned earlier, the total height of the drop was at least 1.2 \(d_e\). After calculating the shape factor \(S = d_s/d_e\), the corresponding value of \(1/H\) in (5.6) was read from the tables of *Adamson and Gast* [1997]. Finally, the outer diameter of the tube from which the drop was hanging was measured in pixels. As per *Song and Springer* [1996b], this diameter was used as the calibration length for obtaining the value of \(d_e\) in millimeters.
Figure 5.9. Examples of DNAPL drops of different shapes hanging in baths of water: a. Undyed 4-CT; b. Sudan IV-dyed 4-CT; c. TCE.
In the present study, equatorial diameters were of the order of 800 pixels (see Figure 5.9.a) corresponding to approximately 8 mm for 4-CT. The value of the interfacial tension was calculated from (5.6) using the density properties given in Table 5.1 and a gravitational acceleration $g = 9.807 \text{ m/s}^2$ [Munson et al., 1994].

It is not clear from the literature how much time should be allowed after the formation of the drop in order to achieve equilibrium and to photograph the drop. The time to static equilibrium is obviously a function of the viscosity of the liquid/fluid pair under investigation. Zeppieri et al. [2001] noted that it was important to allow sufficient time to achieve stability at the interface, but did not specify how much time was necessary. Donahue and Bartell [1952] argued that interfacial tension values did not alter with time, but measurement of the surface tension of drops of varying age showed that it took about two minutes to achieve equilibrium. Consequently, these authors waited five to fifteen minutes prior to photographing drops. Conversely, Smith and Sorg [1941] measured surface tensions on ten-second old drops, arguing that much of the data in the literature was the result of old, possibly contaminated, surfaces associated with lower values of surface tension. Andreas et al. [1938] also derived their surface and interfacial tensions from ten-second old drops. Others made no mention of how time they waited before acquiring images [Patterson and Ross, 1979]. Without further information, it was decided to measure interfacial tension as a function of time from a few seconds after the drop formation up to 92 hours after formation of the drop. Because the camera did not move in between photographs, the reference length remained unchanged, which enabled examination of the relative change of the drop size without incorporating the uncertainty related to calibration (see Section 5.6.3.4).

5.6.3.3 Results and Interpretation of Interfacial Tension Measurements

In this section, results of the pendant drop measurements are presented and compared to observations from other studies on aging effect of surfaces and time dependence of interfacial tension [Ambwani and Fort, 1973; Sobol et al., 1976; Song and Springer, 1996b].

A series of twelve pendant drop tests was run on Sudan IV-dyed 4-CT/water systems, including three tests that ran for 20 hours, 26 hours and 92 hours, respectively. In addition, one test was run for 24 hours on an undyed 4-CT/water system, and two short tests were run on TCE/water systems.

Very good agreement was noted between measurements of the TCE/water interfacial tension of drops of young age with the measurement of 0.0393 N/m reported by Seo and McCray [2002]. Indeed, a value of 0.0392 N/m was measured for a 2 minute-old TCE drop. Another value of 0.0390 N/m was measured for a 5 minute-old drop.

Variations of interfacial tension as a function of time are shown in Figure 5.10, where a logarithmic scale has been used for time. As can be seen in Figure 5.10, a
A decrease of the interfacial tension was observed with time for all three DNAPLs. After several hours, even days, the interfacial tension was still decreasing.

A number of hypotheses that could explain the reduction in interfacial tension with time was examined. The different hypotheses are treated under separate headings below.

**Dissolution effects (mass transfer between phases)**

It was first believed that the observed reduction in interfacial tension was due to the dissolution of DNAPL in the water phase, and dissolution of water in the DNAPL phase. Therefore, it was decided to saturate each phase by using the following procedure prior to the measurements: (1) distilled water was mixed with DNAPL in a vial and shaken for one minute to form an emulsion and then laid to rest for an hour; (2) the DNAPL-saturated water phase was collected with a syringe and poured into the cell; and (3) the water-saturated DNAPL phase was used for the pendant drop. However, this prior preparation of the water and DNAPL phases did not seem to modify the overall behavior of the interfacial tension. Not only did the value of interfacial tension of the saturated system start at approximately the same value of 0.034 N/m (see Figure 5.10), but it also continued to decrease with time.

Another experiment confirmed the conclusion that mass transfer between phases was not responsible for the interfacial tension reduction with time. For two of the tests on Sudan IV-dyed 4-CT/water systems, liquid from 4-CT drops which had been hanging in the water cell for more than twenty hours was recovered and recycled. Although the interfacial tension of these drops had decreased below 0.029 N/m after twenty hours, their interfacial tension rose back up to a value of approximately 0.034 N/m once recycled to form a new hanging drop.

**Geometrical effects**

It was also hypothesized that geometrical effects could be responsible for an apparent reduction in interfacial tension with time. Indeed, over the course of several hours, water evaporated from the visualization cell. Because the water level dropped, the DNAPL level inside the tube also dropped to ensure the static balance of forces. A consequence was that the volume of the DNAPL drop increased and the drop elongated. However, referring to (5.6), if the equatorial diameter increases, the shape factor should also increase, so that $1/H$ decreases and the interfacial tension remains constant. In other words, if (5.6) is correct, a change in geometry should not modify the value of the interfacial tension.

Because the results presented in Figure 5.10 suggested otherwise, it was decided to examine data on drops of approximately the same equatorial diameter, but having different ages. When comparing fresh drops and aged drops of roughly identical size, it was found that interfacial tensions of the former kind were always larger than those of the latter. Moreover, by adding distilled water in the cell so as to decrease the volume of the DNAPL drop and decrease its size, no increase in interfacial tension was observed. Additionally, a quick decrease in the level of water in the cell did not
produce a sharp decrease in interfacial tension. All of these observations suggest that the decrease in interfacial tension is not geometry-related, but is indeed time-related.

![Graph showing interfacial tension as a function of time]

**Figure 5.10.** Variation as a function of time of the interfacial tension of Sudan IV-dyed 4-CT, undyed 4-CT and TCE with water.

Selective migration of contaminants (active surface phenomenon)

It was finally hypothesized that the decrease in interfacial tension with time was due to the active nature of the boundary surface. It is known that if one or more of the components of a fluid system migrates into (or out of) the interface under observation, a change in the interfacial tension with time can usually be detected [Andreas et al., 1938]. Data on the aging of aqueous solutions of sodium stearate in contact with air show a decrease in surface tension by approximately 50% within one hour of forming the drop [Andreas et al., 1938].

The surface-active agent was not found to be the Sudan IV dye. Although the presence of Sudan IV appeared to reduce the interfacial tension of 4-CT with water by about 0.003 N/m, the interfacial tension of undyed 4-CT also decreased with time, as shown in Figure 5.10.
The presence of up to 2% 2-chlorotoluene (2-CT) in the 4-CT solvent that was used (see Section 5.3.1) could not explain the active nature of the surface. While the redistribution of 2-CT throughout the drop with time could have modified the interfacial tension of the system, interfacial tension measurements on high purity TCE also showed a decrease in interfacial tension (see Figure 5.10).

**Discussion**

Unexpected aging effects of pendant drop systems have been reported before. Results appear to differ from one study to another without any foreseeable explanation. Song and Springer [1996b] have observed a decrease of the surface tension of water drops from 0.072 N/m down to 0.070 N/m over time periods of the order of five minutes. This observation is in apparent contradiction with Touhami et al. [1996] who saw no noticeable change over the same time period. More drastic changes in the surface tension of water observed by Sobol et al. [1976] have been attributed to a change in air content of the water near the boundary surface. Ambwani and Fort [1973] have also observed a sharp decrease in the interfacial tension between mercury and cyclohexane, namely from 0.37 N/m to 0.32 N/m, over a period of five hours if cyclohexane contained atmospheric air.

At present, the precise cause of the decrease with time of the interfacial tensions of 4-CT/water and TCE/water is not well understood. That there is an active surface phenomenon is likely. Whether it is dissolved air, or another impurity, remains uncertain and should be further investigated in the future. The fact that aged 4-CT drops collected at the bottom of the visualization cell exhibit high interfacial tension once the drops are recycled and reformed suggests that impurities migrating at the boundary surface during the aging process could be remobilized within the drop once it is recycled (by sudden shaking).

An important consequence of the observed aging effect is that the interfacial tension between 4-CT and water may change during the infiltration experiments. The time period over which water and 4-CT were in contact varied from one experiment to the other, depending on how long it took to set the test up and achieve infiltration, but generally did not exceed 24 hours. Therefore, it can already be foreseen that age difference between tests might create variability in the results of infiltration height.

Based on the results shown in Figure 5.10, the interfacial tension between water and Sudan IV-dyed 4-CT can be taken to be equal to 0.032 ± 0.004 N/m (±12.5% relative uncertainty), if aging effects are neglected.

5.6.3.4 Sources of Error and Suggested Improvements

An error analysis of the method of selected plane of the pendant drop can be found in the review of Ambwani and Fort [1979]. The relative error ∆σ/σ due to
uncertainty on the measurement of interfacial tension can be computed using (5.6) and is given by

\[ \frac{\Delta \sigma}{\sigma} = \frac{\Delta (1/H)}{1/H} + 2 \frac{\Delta d_e}{d_e}, \tag{5.7} \]

where \( \Delta d_e \) and \( \Delta (1/H) \) are the errors on the equatorial diameter and factor \( 1/H \), respectively. It has been assumed in (5.7) that relative errors on the gravitational acceleration \( g \) and the density contrast \( \Delta \rho \) were negligible with respect to the relative errors on the equatorial diameter \( d_e \) and the factor \( 1/H \).

The equatorial diameter in units of meters is obtained by measuring its length in pixels and using a known reference length. Thus

\[ \frac{\Delta d_e [m]}{d_e [m]} = \frac{\Delta d_e [\text{pixels}]}{d_e [\text{pixels}]} + \frac{\Delta d_{\text{ref}} [\text{pixels}]}{d_{\text{ref}} [\text{pixels}]} + \frac{\Delta d_{\text{ref}} [m]}{d_{\text{ref}} [m]}, \tag{5.8} \]

where \( d_{\text{ref}} \) is the length of reference, [m] refers to the real length in meters, [pixels] refers to the length in pixels on the image of interest, and the operator \( \Delta \), again, refers to the error due to uncertainty.

Assuming that the edge detection on any object on the photograph is located with an uncertainty of 2 pixels, \( \Delta d_{\text{ref}} [\text{pixels}] = \Delta d_e [\text{pixels}] = 2 \times 2 = 4 \) pixels. Taking the outer diameter of the glass capillary tube as the length of reference, \( d_{\text{ref}} [m] = (6.02 \pm 0.03) \times 10^{-3} \) m, where the error of \( 0.03 \times 10^{-3} \) m is the uncertainty of the tube outer diameter obtained when measured with a caliper. Using (5.8) and taking a typical 4-CT drop size \( d_e [\text{pixels}] \) equal to 800 pixels, and a length of reference \( d_{\text{ref}} [\text{pixels}] \) equal to 600 pixels, the relative error on the equatorial diameter \( \Delta d_e [m]/d_e [m] \) is approximately equal to 1.7%.

The bulk of the error on the interfacial tension comes, in fact, from the uncertainty on \( 1/H \). As explained earlier, the factor \( 1/H \) is read from tables [e.g., Adamson and Gast, 1997] as a function of the shape parameter \( S = d_s/d_e \). An error of 4 pixels on the equatorial diameter offsets the location of the plane where \( d_s \) is measured. Therefore, there is additional uncertainty on the diameter \( d_s \). As a first approximation, a one to one slope is assumed in the region of the drop where the drop’s boundary intersects the selected plane. Hence, an overestimation of +4 pixels on \( d_s \) leads to an underestimation of \( d_s \) by 8 pixels (4 pixels on either side of the selected plane). Adding an edge detection error of 2 pixels results in \( \Delta d_s = 12 \) pixels.

Examining the case where \( d_s \) is of the order of 600 pixels, the shape parameter is equal to 0.750. By summing the relative errors on \( d_s \) and \( d_e \), the relative uncertainty on the shape parameter \( \Delta S/S \) is found to be equal to \((12/600) + (4/800) = 0.025\), i.e. \( S = 0.750 \pm 0.01875 \). Under these conditions, and using the tables of Adamson and Gast [1997], it is found that \( 1/H \) varies from 0.62764 to 0.71722, i.e. \( 1/H = 0.67243 \pm 0.04479 \). Thus, the relative error \( \Delta (1/H)/(1/H) \) on the factor \( 1/H \) is approximately 6.7%. Using (5.7), it can be concluded that, if an edge detection error of
2 pixels is assumed, the total relative error due to uncertainty on the interfacial tension, \(\Delta\sigma/\sigma\), is of the order of 10%.

The error band (dashed lines) sketched in Figure 5.10 corresponds to a relative uncertainty of approximately 5%, less than that predicted by the analysis above, thereby suggesting that the edge detection error is less than 2 pixels. Note that, for a drop of 4-CT, one pixel roughly corresponds to 0.01 mm, since \(d_{\text{ref}}[\text{m}] = 6.02 \times 10^{-3} \text{ m}\) and \(d_{\text{ref}}[\text{pixels}] = 600\) pixels.

Some improvements are suggested if the pendant drop device is to be used to further investigate the interfacial tension of DNAPLs with water. As shown above, the main source of error comes indirectly from the edge detection. Therefore, in addition to using the entire drop profile to derive its interfacial tension, improved illumination conditions and drop edge detection should be investigated. These are discussed in detail by Song and Springer [1996a; 1996b] and Touhami et al. [1996]. Moreover, temperature control using a water or air thermostat, as well as vibration control, would certainly result in improved accuracy [again, see Song and Springer, 1996b]. Finally, contamination from the atmosphere may be an issue. Thus, the use of a closed visualization cell could result in an improvement of the measured results.

### 5.6.4 Interfacial Tension Measurements Using a Combined Capillary Rise Technique

#### 5.6.4.1 Theory

Referring to Figure 5.11.a, a capillary tube is dipped into a beaker of DNAPL to a depth \(d_0\), and the DNAPL rises in the capillary tube to a height \(h_0\) above the free surface of DNAPL. The capillary rise, \(h_0\), is given by [Adamson and Gast, 1997]

\[
h_0 = \frac{2\sigma_{\text{nw/air}} \cos \theta_{\text{nw/air}}}{\rho_{\text{nw}} g r_0},
\]

(5.9)

where \(\sigma_{\text{nw/air}}\) is the surface tension of DNAPL, \(\theta_{\text{nw/air}}\) is the static contact angle of DNAPL with air measured through DNAPL, \(\rho_{\text{nw}}\) is the DNAPL density, \(g\) is the Earth’s gravitational acceleration and \(r_0\) is the capillary tube radius. Note that the capillary rise is independent of the depth \(d_0\) at which the capillary tube is dipped.

It should be pointed out that (5.9) is not the exact solution of the capillary rise of DNAPL because the forces holding the volume of liquid above the plane tangent to the apex of the meniscus (see Figure 5.11.a) have been neglected [Padday, 1969]. The reader is referred to detailed discussions and existing corrections described in the reviews of capillary rise methods by Padday [1969] and Adamson and Gast [1997]. However, as a first approximation, the correction terms will be neglected.
Following the DNAPL rise, the capillary tube containing a DNAPL column of height $h_2$ is transferred to a second beaker, which contains water. This can be accomplished if drainage of DNAPL is prevented from the capillary tube. Under these circumstances, the height $h_2$ is roughly equal to $h_0 + d_0$. Once dipped in the second beaker at depth $d_1$ (see Figure 5.11.b), water (wetting liquid) rises in the capillary tube at a height $h_1$ above the free surface of water.

![Diagram of capillary rise](image)

**Figure 5.11.** Combined capillary rise method: a. DNAPL rise to measure DNAPL surface tension; b. Water rise in presence of a finger of DNAPL to measure DNAPL/water interfacial tension.

Balancing the tension forces at the DNAPL/air and DNAPL/water interfaces with the weights of the DNAPL and water columns (see Figure 5.11.b) gives

$$
\frac{2\sigma \cos \theta_s}{r_0} + \frac{2\sigma_{nw/air} \cos \theta_{nw/air}}{r_0} = \rho_w g h_1 + \rho_{nw} g h_2,
$$

(5.10)

where $\sigma$ is the interfacial tension between DNAPL and water, and $\theta_s$ is the static contact angle at the DNAPL/water interface measured through water. Again, the rise of water is independent of the depth $d_1$ at which the capillary tube is dipped.

Replacing (5.9) in (5.10) leads to

$$
\sigma \cos \theta_s = \frac{1}{2} g r_0 (\rho_w h_1 + \rho_{nw} h_2 - \rho_{nw} h_0).
$$

(5.11)
Equation (5.11) links the reduced interfacial tension $\sigma \cos \theta_s$ to the different heights of rise $h_0$, $h_1$, and $h_2$. Note, however, that the DNAPL rise $h_0$, is directly related to the surface tension of DNAPL as given by (5.9), while the water rise $h_1$ depends on the height of the DNAPL column $h_2$, so that $h_1$ and $h_2$ are linearly related. In particular, if the DNAPL column height is excessively large, then $h_1$ becomes negative in which case a water depression—instead of a rise—is observed in the capillary tube.

If perfect wetting can be assumed at the DNAPL/water interface, i.e. $\theta_s = 0$ (see Section 2.2.1.3), then the interfacial tension between DNAPL and water can be directly obtained from (5.11). If $\theta_s$ is less than 10°, the error in assuming it to be zero in (5.11) is less than 1.5%. This was the case in the study of Rashidnia et al. [1992]. This is also the assumption made here. If the contact angle is larger than 10°, $\theta_s$ can be calculated from the apex and radius of curvature of the meniscus assumed spherical in shape [Hoffman, 1975; Fermigier and Jenffer, 1991] or measured directly by drawing a tangent line on an enlarged photograph [Dussan V., 1979]. Section 5.6.4.3 further discusses the importance of the static contact angle in the series of combined capillary rise measurements reported in this thesis.

5.6.4.2 Experimental Methods

Two glass beakers and a 1.33 mm diameter capillary tube were used for the combined capillary rise measurement series. The diameter of the capillary tube was precisely measured using the technique described in Section 5.2. All of the glassware was cleaned using the method described in Section 5.4.2.

A typical experimental setup is shown in Figure 5.12. The experiments were set up in the fume hood. The capillary tube was held vertically using a clamping system. The beakers were set on a support jack system (not shown in the figure) such that the capillary tube and beakers could be moved relative to one another. The interfacial tension of Sudan IV-dyed 4-CT/water and Sudan IV-dyed TCA/water were measured. Density properties used in (5.11) are listed in Table 5.1. All the measurements are reported at room temperature, i.e. 19 ± 1°C.

As illustrated in Figure 5.11.a, the capillary tube was first dipped in the beaker containing DNAPL (not shown in Figure 5.12). The DNAPL rise to height $h_0$ was measured using an optical caliper (Titan Tool Supply Co., Buffalo, N.Y.) with a precision of 0.1 mm. Typical rises were of the order of 5 mm to 10 mm, depending on the DNAPL.

The capillary tube containing a column of DNAPL was then transferred to the beaker containing distilled water. The height of the column depended on how deep the tube had been dipped (depth $d_0$ in Figure 5.11.a) and usually varied between 5 mm and 20 mm. Drainage of DNAPL was prevented by either connecting the top of the capillary to a suction device [Rashidnia et al., 1992], or more simply by pressing a finger at the top of the capillary. The latter technique was found to be more precise. Upon being dipped in the beaker, rise—alternatively depression—of water took place.
in the capillary tube (see Figure 5.12). Again, the height \( h_1 \) and \( h_2 \) were measured with the optical caliper. The height \( h_1 \) was of the same order as \( h_0 \), but could take negative values.

\[
\text{Figure 5.12. Setup of a combined capillary rise experiment}
\]

\textit{Rashidnia et al.} [1992] ensured that the interfaces were at equilibrium location by applying a small suction to the top of the capillary tube followed by a release to atmospheric pressure. The operation was repeated a few times. Using this technique, they found the equilibrium locations to be stable and repeatable. Note that this technique is used to ensure that a static receding meniscus is used for the measurements, so as to achieve a contact angle of zero, or of magnitude as small as possible [\textit{Padday}, 1969; \textit{Adamson and Gast}, 1997]. Recall, as discussed in Section 2.3.5.2, that a static receding meniscus is achieved when, in the most recent history of the meniscus displacement, the non-wetting fluid displaced the wetting fluid prior to achieving static equilibrium (see Figure 5.13.a). Conversely, in the recent history of a static advancing meniscus, the wetting fluid displaced the non-wetting fluid (see Figure 5.13.b). It is known that the \textit{true} static contact angle, as given by Young’s equation [\textit{Dussan V.}, 1979] (see (2.38) in Section 2.3.5.2), is in fact a property difficult or impossible to measure because of its apparent variability [\textit{Blake and Ruschak}, 1997]. Most angles are either static receding contact angles or static advancing contact angles, with the true static contact angle lying between the two [\textit{Blake and Ruschak}, 1997]. This issue is further discussed in Section 5.6.4.3.
Figure 5.13. Contact angle hysteresis: (a) Static receding contact angle, $\theta_r$, following displacement of the wetting fluid by the non-wetting fluid; (b) Static advancing contact angle, $\theta_a$, following displacement of the non-wetting fluid by the wetting fluid.

Unless otherwise noted, surface and interfacial tensions were measured in the receding meniscus situation by moving the beaker relative to the capillary tube, such that the interfaces would first move upward then downward prior to equilibrium. This operation was repeated several times before measuring the heights $h_0$, $h_1$, and $h_2$.

Other experimental aspects of capillary rise are discussed by Padday [1969] and Adamson and Gast [1997]. In particular, there is an issue related to identifying the exact location of the free surface of water or DNAPL in the beakers. Indeed, if the liquids are observed through the glass wall of the beaker with an optical caliper, the exact location of the free surface of the liquids appears to be above its true location. Padday [1969] recommends observing a needle placed very near the surface. The needle and its reflection give a very accurate indication of the position of the free liquid level. Another solution is to fill up the beaker up to the very top such that the free surface appears right above the beaker, as shown in Figure 5.12.

5.6.4.3 Results and Interpretation

The surface and interfacial tensions of Sudan IV-dyed TCA and Sudan IV-dyed 4-CT were measured five and six times, respectively. Three different 1.33 mm diameter capillary tubes were used for the measurements. Results did not appear to depend on the capillary tube that was used. Surface and interfacial tension measurement results are summarized in Table 5.5.
Using literature data provided in Table 5.4, the value of the surface tensions of 4-CT and TCA at the temperature of the experiment can be calculated, yielding respectively 0.0329 N/m and 0.0259 N/m at 19°C, respectively. These values are larger than the measured values of surface tensions of the two DNAPLs, which were found to be equal to 0.0296 N/m and 0.0225 N/m, respectively (see Table 5.5). The difference of about 3.5 mN/m can probably be attributed to the presence of Sudan IV, although the presence of impurities or existence of a contact angle at the air/DNAPL interface cannot be excluded.

Considering the interfacial tension of TCA, the measured value of 0.0344 N/m is a lot less than the quoted literature value of 0.045 N/m (see Table 5.4). Large differences between the properties of undyed and Sudan IV-dyed DNAPLs have been reported before. Longino and Kueper [1999] have measured the interfacial tension of Sudan IV-dyed perchloroethylene (PCE) using a ring tensiometer. They obtained a value of 0.0279 N/m at 22°C. Nonetheless, undyed PCE interfacial tension values of 0.0475 N/m at 20°C [Demond and Lindner, 1993] and 0.0444 N/m at 25°C [Mercer and Cohen, 1990] have been reported. It is possible that additional impurities present in either the DNAPL phase or the water phase also contribute to lower experimental measurements of the interfacial tension.

Referring to the interfacial tension of 4-CT, the obtained value of 0.0319 N/m (see Table 5.5) is in very good agreement with the interfacial tension value obtained using the pendant drop method, i.e. 0.032 ± 0.004 N/m, and suggests that the assumption of small contact angle $\theta_s$ is valid.

<table>
<thead>
<tr>
<th>Table 5.5.</th>
<th>Surface and Interfacial Tension Measurements of Sudan IV-Dyed TCA and Sudan IV-Dyed 4-CT Using the Combined Capillary Rise Method</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Sudan IV-dyed TCA</td>
</tr>
<tr>
<td>Surface tension</td>
<td>Average value [mN/m]</td>
</tr>
<tr>
<td></td>
<td>Standard deviation [mN/m]</td>
</tr>
<tr>
<td></td>
<td>Maximum deviation $^a$ [mN/m]</td>
</tr>
<tr>
<td>Interfacial tension</td>
<td>Average value [mN/m]</td>
</tr>
<tr>
<td></td>
<td>Standard deviation [mN/m]</td>
</tr>
<tr>
<td></td>
<td>Maximum deviation $^a$ [mN/m]</td>
</tr>
<tr>
<td>Number of measurements</td>
<td>5</td>
</tr>
</tbody>
</table>

$^a$Maximum deviation between a data point and the average measured value for all data points.

Overall, it can be seen that the measurement of surface tension yields a smaller standard deviation and maximum deviation than the measurement of interfacial tension. This is, in part, because the interfacial tension calculation relies on three measurements of height—and their associated uncertainties—as given by (5.11), whereas the surface
tension is calculated from only one height measurement as given by (5.9). Thus, the latter can be expected to be more repeatable.

Part of the scatter of the interfacial tension data might also be attributed to the variability of the contact angle at the dyed DNAPL/water interface. To prove this point, a series of experiments was performed on Sudan IV-dyed 4-CT, as illustrated in Figure 5.14 and Figure 5.15. A 1.33 mm diameter capillary tube was used. The surface tension of dyed 4-CT was first measured on a static receding contact angle at the air/DNAPL interface (see Figure 5.14.a) using the procedure described in Section 5.6.4.2. A rise, $h_0$, equal to 8.8 mm was measured, which corresponded to a surface tension of 0.0307 N/m using (5.9) and assuming perfect wetting. The depth $d_0$ of dip of the tube was also measured and found to be equal to 2.5 mm. The capillary tube, as shown in Figure 5.14.a, was then moved downwards, as shown in Figure 5.14.b, such that the depth $d_0$ increased to 13.2 mm. The meniscus was observed to rise at the same rate that the tube was lowered. Although under these conditions the meniscus was a static advancing contact angle, the height of rise remained approximately constant and equal to 8.8 mm. The operation was repeated a second time. The tube was lowered to a depth $d_0$ of 17.3 mm. Again, the meniscus rose at the same rate the tube was lowered and the height $h_0$ did not change. Finally, the beaker was moved upwards then downwards without moving the capillary tube (see Figure 5.14.c). This made the interface rise and fall back in a situation of static receding contact angle. However, no significant change in height was measured, suggesting that the difference between the receding and advancing contact angles under this configuration could not be detected.

![Figure 5.14. Principle of air/DNAPL meniscus contact angle experiment: a. Capillary tube in its initial position (contact angle is static receding); b. Capillary tube lowered (contact angle is static advancing); c. Beaker successively raised and lowered (contact angle is first advancing then receding).](image)

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The operation described above was repeated for the measurement of interfacial tension (see Figure 5.15). A capillary tube containing a DNAPL column of height $h_2$ equal to 12.7 mm was dipped in water. Upon conditions of receding contact angles (see Figure 5.15.a), a water rise of height $h_1$ equal to 6.6 mm took place, corresponding to an interfacial tension of 0.0351 N/m using (5.11) and again assuming perfect wetting. A photograph of the interface under these conditions is shown in Figure 5.16.a, where it can be seen that the assumption of perfect wetting is consistent with the shape of the meniscus. From a depth $d_1$ of 17.0 mm, the capillary tube was lowered to a depth of 29.2 mm. Under this configuration, the DNAPL/water meniscus was in a situation of static advancing contact angle (see Figure 5.15.b). For this case, it was noticed that the columns of water and DNAPL inside the capillary tube did not rise at the same rate that the tube was lowered. Instead, the height of the column of water $h_1$ decreased to 0.8 mm, the height $h_2$ obviously remained constant, and the DNAPL/water interface stated deforming, as can be seen in Figure 5.16.b. It is obvious from the shape of the meniscus shown in this figure that the assumption of perfect wetting is no longer valid. Using (5.11), the reduced interfacial tension $\sigma \cos \theta$ was found to be equal to 0.0162 N/m, from which a contact angle of 62° could be back-calculated using the value of 0.0351 N/m for the interfacial tension (i.e. the static receding contact angle was assumed to be perfectly wetting).

![Figure 5.15](image.png)

**Figure 5.15.** Principle of DNAPL/water meniscus contact angle experiment: a. Capillary tube in its initial position (contact angle is static receding); b. Capillary tube lowered (contact angle is static advancing); c. Beaker successively raised and lowered (contact angle is first advancing then receding).
The operation was repeated a second time. The tube was lowered to a depth $d_1$ of 35.8 mm. The DNAPL/water interface went below the free level of water in the beaker such that $h_2$ was equal to $-2.0$ mm. This corresponded to a reduced interfacial tension of 0.0071 N/m and a contact angle of 78°. Upon moving the beaker upwards without changing the capillary tube location (see Figure 5.15.c), the column of liquid inside the capillary tube appeared pinned at first (see definition of pinned in Section 2.3.5.2), but then proceeded to rapidly move upwards. After lowering the beaker back to its last position, i.e. $d_1 = 35.8$ mm, the heights $h_1$ and $h_2$ returned to their initial values of 6.6 mm and 12.7 mm, respectively, showing a strong hysteretic effects at this location. This last configuration is shown in Figure 5.16.c, where the meniscus shape differs very little from the meniscus shown in Figure 5.16.a.

Two similar experiments were run. Using the same derivation as done above, static advancing contact angles of 56°, 70° and 80° were back-calculated on one run, again, assuming that the static receding contact angle was perfectly wetting. In another run, static advancing contact angles of 27°, 44° and 71° were back-calculated. These results suggest that the static advancing contact angle appears to have an extremely broad range of values.

![Figure 5.16. 4-CT/water meniscus at various stages of a combined rise experiment: a. As a static receding meniscus; b. As a static advancing meniscus; c. Again as a static receding meniscus.](image)

The reason why the DNAPL/water interface apparently remains pinned on the capillary tube wall and starts to deform remains uncertain (see discussion on pinning in Section 2.3.5.2 and Section 2.3.6.4). It appears that pinning effects may take place when the capillary tube is lowered to such a depth that the rising DNAPL/water meniscus rises to a level in the capillary tube where it has never been before. In that case, water attempts, and fails, to replace DNAPL along the wall of the capillary tube, such that the meniscus starts deforming to ensure force equilibrium. However, there are times when the meniscus can remain pinned even if it has moved above its prior
location before, suggesting that local defects in the capillary tube or small impurities may act as pinning points (again, see discussion on pinning in Section 2.3.5.2 and Section 2.3.6.4).

Two conclusions can be drawn from this series of experiments. The first conclusion is that variability of results in the combined capillary rise method are expected, such that, overall, it is not a reliable way of measuring the interfacial tension between two liquids. It is, however, a very good method for measuring the surface tension of a liquid. The second conclusion has a direct impact on the infiltration experiments described in Chapters 6. The experiments just discussed above have shown that unless the contact angle is in the configuration of a static receding contact angle, it can vary significantly from 0, and can take any value up to 80°. When pooling DNAPL on top of a water-saturated capillary tube and first forming the DNAPL/water interface (see Section 6.2.2), there is not a static receding meniscus. Indeed, while the meniscus exists, it has no history of motion. Thus, what determines the contact angle at this point in the infiltration experiment is probably related to the geometric conditions at the bore of the capillary tube. Therefore, under these conditions, it may differ from zero, and not be precisely predictable. This points is further discussed in Section 7.2.1.2.

5.7 Conclusion

Based on the interfacial tension and viscosity measurements reported in this chapter, the following properties are assumed in the remainder of this thesis:

*Viscosity*

The viscosities of Sudan IV-dyed 4-CT and Sudan IV-dyed TCA are taken to be close to the viscosity of water, i.e. equal to $10^{-3}$ Pa.s at 20°C. Because both the density and viscosity contrasts between 4-CT and water are small with respect to the viscosity and density of water, the NICCA and NIVCA models (see Section 3.5) are generally applicable for 4-CT infiltration. When the specific effects of viscosity contrast are examined, the viscosity of 4-CT is taken equal to $0.918 \times 10^{-3}$ Pa.s at 20°C (see Table 5.3).

*Interfacial tension*

The interfacial tension of Sudan IV-dyed 4-CT is taken to be equal to $0.032 \pm 0.004$ N/m (value obtained from the pendant drop method, see Section 5.6.3.3). The interfacial tension of Sudan IV-dyed TCA is taken to be equal to $0.0344 \pm 0.0029$ N/m (value obtained from the combined capillary rise method, see Section 5.6.4.3).
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CHAPTER 6

EXPERIMENTAL METHODOLOGY

6.1 Introduction

This chapter presents an overview of the methods and equipment used for the experimental program of this research. The chapter is divided into three main parts.

Section 6.2 presents the experimental methodology used for conducting DNAPL infiltration experiments at 1 g. These experiments are referred to as DNAPL infiltration laboratory experiments. Both spontaneous infiltration experiments and controlled-head infiltration experiments are described.

Section 6.3 presents the experimental methodology used for running DNAPL infiltration experiments on the geotechnical centrifuge. These experiments are referred to as DNAPL infiltration centrifuge experiments.

Finally, Section 6.4 presents the centrifuge instrumentation and data acquisition system used as part of the centrifuge experimental program of this research. References made in this chapter are listed in Section 6.5.

6.2 DNAPL Infiltration Laboratory Experiments

6.2.1 Overview

In order to investigate the kinetics of DNAPL infiltration into simulated vertical fractures, and to test the theoretical models of infiltration presented in Chapter 3, two types of infiltration laboratory experiments into capillary tubes were conducted. The first category of experiments is referred to as spontaneous DNAPL infiltration laboratory experiments. For this type of test, DNAPL was pooled on top of a water-saturated capillary tube until infiltration into the capillary tube took place. The second category of experiments is referred to as controlled-head DNAPL infiltration laboratory experiments. Under this configuration, a valve controlled the flow through the capillary tube, such that a DNAPL pool of given height could be set prior to starting the flow. These two setups are described under separate headings. Note that, in contrast to the laboratory experiments, the centrifuge experiments described in Section 6.3 are all spontaneous infiltration experiments.
6.2.2 Spontaneous DNAPL Infiltration Experiments

As mentioned in Section 5.2, in order to simulate a range of fracture sizes, glass capillary tubes of different diameters were used. Again, these capillary tubes are referred to by their actual diameter, i.e. 0.66 mm, 1.33 mm, 2.20 mm or 2.70 mm diameter capillary tubes.

A diagram of a typical experimental setup used for the laboratory tests is shown in Figure 6.1. To illustrate the discussion, photographs of various experimental setups and enlarged details of interest are presented in Figure 6.2, Figure 6.3, and Figure 6.4.

![Diagram of Spontaneous DNAPL Infiltration Experimental Setup](image)

**Figure 6.1.** Spontaneous DNAPL infiltration experimental setup.

The spontaneous infiltration tests used capillary tubes of varying length, between 60 mm and 1220 mm, which were graduated every 10 mm. Graduating these tubes by scoring interval marks with a small file was not a convenient technique, as it was observed that the tubes became extremely fragile and would snap wherever the tubes had been scored. Instead, a fine-pointed correction pen was used to draw marks on the tube. The marks would dissolve in acetone and had to be redrawn in between tests.
The capillary tubes were set up in a transparent tank that will be referred to as *glass tank* or *tank tube*. For tests performed on short-length capillary tubes (length of less than 250 mm), the transparent tank was a 60 mm inner diameter, 400 mm long glass cylinder, a piece of glassware widely available in the MIT geotechnical laboratory and typically used for hydrometer tests (see Figure 6.3.a). For tests on capillary tubes of intermediate length (between 250 mm and 1000 mm), the tank was a glass tube with diameter between 20 mm and 65 mm, whose lower end was properly sealed (see Figure 6.2.a and Figure 6.4.a). For tests on capillary tubes of very long length (more than 1000 mm), the tank was a transparent PVC 25 mm (1 inch) inner diameter and 32 mm (1-1/4 inch) outer diameter hose (VWR), properly sealed at the bottom and stretched vertically with a series of clamps. Obviously, there was a trade-off between the diameter of the tank tube and the amount of generated DNAPL-contaminated wastewater that needed to be disposed off (see Section 5.4.1).

![Figure 6.2. Experimental setup of a 305 mm long, 1.33 m diameter capillary tube: a. General view prior to inserting the capillary tube inside the tank tube; b. Detail of the capillary tube/reservoir tube system; c. Detail of the capillary tube/reservoir tube system during the onset of DNAPL infiltration.](image)

The most convenient way of sealing the lower end of a tank tube was to use a test tube of diameter equal to the tank tube diameter. The test tube was held up against the tank tube by means of rubber tubing (see, for example, the detail in Figure 6.4.d).
Alternatively, a rubber stopper with an appropriate bore could also be used (see Figure 6.2.a). The lower end of the test tube shown in Figure 6.4.d was covered by a second stopper to protect the tube in case of unexpected fall. A secondary container was immediately located underneath the experimental setup to minimize the consequences of a potential spill.

The reason for using a removable test tube was two-fold. First, it did not require any particular custom-made piece of glassware, such as a long tube with one closed end. Second, it was safer. Indeed, following an infiltration test, most of the pure-phase DNAPL would fall into the test tube. The bulk of the water, which did not contain pure-phase DNAPL, was pumped out of the tank tube. Afterwards, the test tube could simply be disconnected and handled independently without the risk and problems associated with handling pure phase DNAPL in a tube of length that could exceed one meter.

![Figure 6.3. Experimental setup of a 135 mm long, 2.70 mm diameter capillary tube inside a glass cylinder: a. General view; b. Detail of the capillary tube/reservoir tube system (the capillary tube is not graduated).](image)

The upper part of each capillary tube was connected to a glass reservoir tube having an internal diameter at least ten times larger than the internal diameter of the capillary tube. As its name suggests, the reservoir tube was used to build up the DNAPL pool prior to its infiltration into the capillary tube, as shown in Figure 6.2.c. The length of the reservoir tube depended on the estimated critical height of the DNAPL pool (see Section 3.3) and was largest for 0.66 mm diameter capillary tubes.
Figure 6.4. Experimental setup of a 1221 mm long, 0.66 m diameter capillary tube: a. General view of the setup; 1, setup supporting frame; 2, optical caliper; 3, tray containing DNAPL and small equipment; b. Optical caliper and Teflon connection between reservoir tube and capillary tube (no DNAPL in reservoir tube); c. Reservoir tube containing a small pool of DNAPL and apparatus supporting frame; d. DNAPL collection test tube; e. Teflon connection between two consecutive capillary tubes (the piece of newspaper behind the tank tube is there to improve the overall contrast for filming the infiltration experiment).
The connection between a capillary tube and a reservoir tube was made using chemically resistant Teflon seal tape (VWR) (see Figure 6.2.b and Figure 6.3.b). The width of the Teflon tape was approximately equal to 15 mm, thus blocking the view of the upper part of a capillary tube. This is well shown in Figure 6.2.c where the view of the DNAPL finger is partially obstructed by the Teflon tape collar. The location of the Teflon tape collar was varied between tests, from 5 mm to 60 mm below the top of the capillary tube, so as to observe the early stages of the DNAPL infiltration.

The Teflon tape had to be wrapped sufficiently tight so as to support the entire weight of the capillary tube. If the capillary tube length exceeded 600 mm, the connection was usually reinforced with duct tape applied on the outer walls of the capillary tubes (see Figure 6.4.b), which again limited the view of the very top of the capillary tube.

Because the capillary tubes were furnished in standard lengths of 305 mm (see Section 5.2), consecutively joined capillary tubes were used for tests performed on long capillary tubes. Connections between consecutive tubes were made by wrapping their ends with Teflon tape and inserting them in short tubes of diameter slightly larger than the outer diameter of the capillary tubes. Typically, this connection was also reinforced with duct tape, as shown in Figure 6.4.e, so as to prevent any gap from forming between the ends of the capillary tubes. While these connections did not appear to affect the onset of DNAPL infiltration, it seriously complicated the setup. On the other hand, a set of long capillary tubes would have probably been difficult to clean or handle, and would have required one tube for each length of interest.

Tubes of length less than 305 mm were obtained by simply cutting a capillary tube to the desired length. The simplest way to cut a tube was to score a small mark with a file where the cut needed to be made, and then snap the tube under running water.

Prior to a test, the different parts of the experimental setup were rinsed using the procedure described in Section 5.4.2. Next, the reservoir tube-capillary tube system was set up inside the tank tube using a set of clamps (see, for example, Figure 6.2.a and Figure 6.4.c). The tank tube was then filled with water. To avoid trapping air in either the capillary tube or the reservoir tube, de-aerated distilled water was used as the wetting fluid. Air bubbles could easily be trapped in the tank, especially below or within the Teflon seal of the reservoir tube. Using de-aerated water helped dissolve undesirable air bubbles if sufficient time was allowed between the setup and run of the experiment.

After saturating the experimental setup, DNAPL was introduced into the reservoir tube with a syringe mounted with a long needle. If the reservoir tube was very long, the needle itself was connected to a small diameter hose (see Figure 6.5.a). Care was taken to ensure that the first droplets of DNAPL would sit on top of the capillary tube so as to form a DNAPL/water meniscus at the entrance to the tube. As the height of the DNAPL pool increased, the needle was raised up, and kept in contact with the DNAPL phase, in order to avoid trapping of water droplets below the DNAPL pool. This ensured the formation of a continuous DNAPL phase, as shown in Figure 6.5.b. During this process, the height from the top of the capillary tube to the top of the DNAPL phase was measured with an optical caliper, as shown in Figure 6.4.a, to
accurately determine the pool height. This caliper was the same as the caliper used for
the combined capillary rise interfacial tension experiments (see Section 5.6.4.2). Because DNAPL sometimes partly wets the reservoir glass surface, it was not always
possible to obtain a completely flat surface at the top of the pool (see, for example,
Figure 6.5.c). When this occurred, the DNAPL pool height was taken as the average
height of DNAPL in the reservoir tube.

DNAPL was slowly added until the pool reached a critical level. At this point
infiltration took place. The pool height immediately before infiltration started was
recorded as the critical height $h_i$, as defined in Section 3.3.

During the tests, it was observed that, at the early stages of pooling, when the
pool height was still significantly less than $h_i$, a finger of DNAPL would pre-infiltrate

Figure 6.5. Built-up of a DNAPL pool on top of a 0.66 mm diameter capillary tube:
a. Early stage of built-up with use of a long needle and extension hose (upon contact
with the solvent the hose gets dyed red); b. Intermediate stage of built-up (note the
small water droplets trapped by DNAPL along the walls of the reservoir tube); c. Stage
preceding DNAPL infiltration (note the uneven top surface of the DNAPL pool).
the tube to a depth of approximately 1 mm to 3 mm. As the experiment progressed, and more DNAPL was added, it was observed that this pre-infiltration finger would sometimes increase in length. For example, the pre-infiltration finger shown in Figure 6.2.c is 25 mm long. Some experiments yielded pre-infiltration depths down to 60 mm. The depth of pre-infiltration was not always repeatable, even using the same tube and experimental setup. Pre-infiltration did not appear to be a function of the capillary tube diameter. It is believed that this phenomenon is intimately connected to the variability of the static contact angle as discussed at the end of Section 5.6.4.3. This topic is further investigated and discussed in Section 7.2.1.2.

For the majority of infiltration tests, the displacement of the DNAPL/water interface in the capillary tube was monitored with time using a video camera. Because of the large differences in internal diameter between the reservoir tube and the capillary tube, it was possible to assume that the displacement was taking place with no change in reservoir pool height, an assumption used in the development of the theoretical model (see Section 3.4.1 and later discussion in Section 7.2.2.5).

Filming and recording equipment used during the laboratory infiltration experiments is shown in Figure 6.6. For the most part, it is constituted of equipment used to monitor centrifuge experiments [Marulanda, 2001] (see also Section 6.3.3). Filming of tests was made using a hand-held 1/2 inch color CCD miniature camera, model GP-KS162, manufactured by Panasonic Industrial Camera Division. The camera was mounted with a 3 mm super-wide angle lens, model GP-LM3TA, made by the same manufacturer. A 12-volt DC input voltage provided by a variable DC supply (Hewlett-Packard) powered the camera control unit.

Although the super-wide angle lens distorted the images that were filmed, distortion was not an issue here, as the interval marks scored on the capillary tubes were practically on the same focal plane as the moving DNAPL/water interface. During the infiltration process, the camera was displaced at the same velocity as the DNAPL/water interface, in order to keep the moving meniscus at the center of the image. The field of vision of the camera for these images was of the order of 100 mm. In other words, any still frame of the infiltration experiment film showed a section of capillary tube of approximately 100 mm.

The image signal coming from the miniature camera was fed through a BNC connector to a Panasonic time/date generator unit (model WJ-810), which made it possible to superimpose time, date and a stopwatch to the images of the experiments. Tests were recorded using a videocassette recorder (VCR Quasar, model VH5251YW). The VCR was set in the SP fast recording mode and acquired 32 images per second. This corresponded to an image every three to four hundredths of a second, and was perfectly adequate at typical DNAPL infiltration speeds. During data reduction, the experiment tape could be played frame by frame on the VCR, thus enabling the location of the DNAPL/water interface within a given capillary tube to be tracked at a given time, since a time reading from the stopwatch was superimposed on each image. Labeling of the capillary tube tick marks, using a number of small labels as shown in Figure 6.2.c and Figure 6.4.e, facilitated tracking of the location of the interface while reviewing the infiltration images. Such labels were particularly helpful on long tubes. Monitoring and data reduction of the experiments were conducted using an RCA black
and white monitor. Four two-hour tapes of recordings were generated for both the laboratory experiments and centrifuge tests (see Section 6.3.4).

![Setup of filming and recording equipment used for the infiltration experiments.](image)

**Figure 6.6.** Setup of filming and recording equipment used for the infiltration experiments.

Following an infiltration test, the experimental apparatus was taken apart and cleaned. The liquids were disposed of following the procedure described in Section 5.4.1. For long tank tubes, water free of pure-phase DNAPL was first removed using a peristaltic pump purchased from Masterflex and described elsewhere [Ratnam, 1996].

### 6.2.3 Controlled-Head DNAPL Infiltration Experiments

In parallel with spontaneous laboratory DNAPL infiltration experiments, a number of controlled-head infiltration tests were run on 305 mm long, 1.33 mm diameter capillary tubes. A diagram of the experimental setup used for this type of laboratory test is shown in Figure 6.7. Figure 6.8 presents two photographs of the experimental setup.

The controlled-head infiltration test allowed control of the height of the DNAPL pool prior to its infiltration into the capillary tube. As seen in Section 6.2.2, for spontaneous infiltration tests, DNAPL was pooled into the reservoir until the pool height reached its critical height \( h_i \). In controlled-head infiltration tests, the pool height could be set to any target value larger than the critical height before the infiltration started.

There are many similarities between the experimental setup of a controlled-head infiltration test and that of a spontaneous infiltration test. In particular, the tubes were
also graduated every 10 mm and connected to a DNAPL reservoir tube by mean of a Teflon tape collar. In fact, the main difference between these experimental setups had to do with the tank tube, which, in the case of the controlled-head infiltration test setup, was a 200 mm high, 75 mm diameter Lexan container held by a clamp. A bore of diameter equal to the outer diameter of the reservoir tube was drilled at the bottom of the Lexan container, such that the upper end of the reservoir tube was inside the tank tube and in contact with water, whereas its lower end was below the tank tube and connected to the capillary tube (see Figure 6.7 and Figure 6.8.a). A coating of RTV silicon was applied around the reservoir tube for sealing of the tank and waterproofing.

![Figure 6.7.](image)

**Figure 6.7.** Schematic illustration of the controlled-head DNAPL infiltration experimental apparatus.

A Swagelock tee and fittings (VWR) were connected to the lower end of the capillary tube through a Teflon collar (see Figure 6.8.b). One of the tee connections was fed back to the reservoir by way of a valve and pipe. The other tee end was connected to a second valve, referred to as the drainage valve.
The system was first saturated with de-aerated distilled water. When the feedback valve was open and the drainage valve was closed, the two ends of the capillary tubes were looped through the feedback tube. Thus, the water pressure distribution was hydrostatic, and the configuration was identical to the configuration of a spontaneous infiltration test.

When the two valves were closed, no flow through the capillary tube was possible and a DNAPL pool of height larger than $h_i$ could be built inside the reservoir tube. Using a pool build-up procedure similar to that of spontaneous infiltration tests, the target DNAPL pool height was set and measured with the optical caliper. As soon as the feedback valve was opened, DNAPL infiltration took place in the capillary tube. The interface displacement was monitored using the miniature camera (see Figure 6.8.b) and the video equipment presented in Section 6.2.2. The feedback valve was closed before the DNAPL started to infiltrate the feedback tube. The drainage valve was then opened to clean up the system and collect the waste liquids. Final clean-up and disposal procedures were similar to those of spontaneous infiltration experiments.

Typical pool heights were of the order of 10% to 30% larger than the critical pool height, $h_i$, obtained from spontaneous infiltration tests. The pool height was varied to examine its effect on the velocity of the interface displacement, particularly at the early stages of infiltration. These effects are discussed in Section 7.2.3.

![Figure 6.8.](image)

Figure 6.8. Controlled-head DNAPL infiltration experimental apparatus: a. View of the upper part of the apparatus; b. View of the lower part of the apparatus including the tee fitting and feedback valve.
6.3 DNAPL Infiltration Centrifuge Experiments

6.3.1 Overview

To test the physical model of DNAPL infiltration developed in Chapter 4, a series of capillary tube experiments was conducted using the balanced-arm geotechnical centrifuge located in the Department of Civil and Environmental Engineering at MIT, room 1-079. Similar experimental work and setup procedures using capillary tubes in a centrifuge environment have been reported before [Adams, 2000]. Throughout the rest of this section, the procedures and equipment used for these tests will be described. This description includes a brief presentation of the MIT geotechnical centrifuge (Section 6.3.2) as well as the preparation (Section 6.3.3) and run (Section 6.3.4) of a centrifuge infiltration experiment. The test instrumentation and centrifuge data acquisition system is discussed in a separate section (Section 6.4).

6.3.2 MIT Balanced-Arm Geotechnical Centrifuge

The MIT balanced-arm geotechnical centrifuge is a Genisco Model 1231 G-Accelerator, purchased in 1985 from the Genisco Technology Corporation, now Trio Tech International, San Fernando, California. It has been extensively described in prior theses [Pahwa, 1987; Ratnam, 1996; Marulanda, 2001]. As illustrated in Figure 6.9.a, the geotechnical centrifuge has two identical swinging platform mounted at opposite ends of a rotary arm so as to be diametrically opposed to each other. The center of the arm is the axle of a driving electric motor, located below the centrifuge and rigidly fixed to the floor.

A test package containing the experimental setup is placed on the main platform. Balance of the centrifuge is ensured by placing a counterweight package on the secondary platform. As the arm rotates, each platform swings up about a trunnion, such that the body force acts perpendicular to the platforms. The arm, the driving shaft and the platforms are enclosed in a cylindrical shell made of a 6.35 mm (1/4 inch) boiler plate. The radius of the arm from the axis of the shaft to each trunnion is 1067 mm (42 inches). The bases of the two platforms are located 235 mm (9-1/4 inches) below the axis of each trunnion. The main platform base is covered by a 12.7 mm (1/2 inch) thick aluminum plate upon which an experimental setup can be bolted. Therefore, practically speaking, the base of the main platform is 222 mm (8-3/4 inches) below the axis of the trunnion. This value is important when estimating the value of the g-level during the DNAPL infiltration process, since the g-level is radius dependent.
Figure 6.9. The MIT balanced-arm geotechnical centrifuge: a. Schematic illustration [after Pahwa, 1987]; b. View of the beam and main platform through the hatch door.
With a load capacity of 13,610 g-kg (30,000 g-lb), a test package of 90.7 kg (200 lb) can be accelerated to 150 g, or a test package of 68.0 kg (150 lb) can be accelerated to 200 g. A centrifugal acceleration of 200 g corresponds to approximately 400 RPM for this centrifuge. The dimensions of a test package must be contained in a cube of side not exceeding 0.54 m.

6.3.3 Preparation of the Centrifuge Experimental Package

Again, in order to simulate a range of fracture sizes, glass capillary tubes of different diameters were used. Capillary tubes of length varying from 40 mm to 136 mm were prepared using the cleaning procedure described in Section 5.4.2, graduated every 10 mm and connected to reservoir tubes, as was described for laboratory experiments in Section 6.2.2.

Figure 6.10. Capillary tube holding rack supporting a set of three capillary tubes.

In order to maintain the capillary tube/reservoir tube systems vertical throughout the experiment, a small aluminum and steel rack was used. A photograph of the rack is shown in Figure 6.10. The rack had a length of 350 mm and a width of 40 mm, and included a 5 mm thick beam placed at height 120 mm that supported the capillary tubes. The frame and bolts were spray painted to prevent corrosion. Several bores of diameters equal to the outer diameters of the capillary tubes were drilled on the beam, such that up to eight capillary tubes could be tested in the course of one centrifuge experiment. An o-ring sandwiched between the Teflon collar and the beam (see Figure 6.10) provided an even support around the capillary tube.
An important limitation associated with this system is that it did not allow observation of the interface displacement through the upper part of the capillary tube. Indeed, the view of the first 20 mm of the capillary tube was blocked by the succession of the Teflon collar, o-ring and holding beam. This design should be improved if more tests are to be run in the future.

The rack and capillary tubes were placed in a glass-fronted centrifuge strong box of internal length 350 mm, width 75 mm and height 250 mm. The front panel of the box was made of a 12.7 mm (1/2 inch) glass plate. The back panel of the box was made of a 12.7 mm (1/2 inch) Lexan plate. The side and base of the box were made of 9.5 mm (3/8 in) aluminum plates. The box is shown in Figure 6.11.a.

Because, DNAPL reacts with Lexan and possibly with aluminum (see Section 5.4.1), exposure of the box to pure-phase DNAPL had to be minimized. Thus, a series of 70 mm diameter glass dishes were placed at the bottom of the centrifuge box underneath the capillary tubes, such that DNAPL could be collected in the dishes at the end of the infiltration process.

![Figure 6.11. Preparation of centrifuge test DNAPL pools: a. General view of preparation; b. Detail of DNAPL pools.](image)

The box was slowly filled with de-aerated, distilled water. Care was taken to ensure that the water level in the centrifuge strong box was above the upper ends of the reservoir tubes by at least 50 mm. Because the water level fluctuates during a centrifuge test, it could have otherwise gotten below the reservoir tube water level, changed the experimental conditions and thus compromised the test. The setup was allowed to equilibrate overnight to dissolve any air bubbles trapped in the system during the test preparation.

The DNAPL pool preparation was conducted in the fume hood of room 1-047. A fixed amount of DNAPL was introduced into each reservoir with a needle and syringe. The pool build-up procedure was similar to that of laboratory tests. DNAPL was initially placed at the bottom of the reservoir tube in direct contact with the top of the capillary tube so as to form a DNAPL/water meniscus at the entrance to the tube. As the height of the DNAPL pool increased, the tip of the needle was kept in contact with
the DNAPL phase to avoid trapping water droplets below the DNAPL pool. The pool height, defined as the distance from the top of the capillary tube to the top of the pool in each reservoir tube, was measured with the optical caliper, as shown in Figure 6.11.a. As can be seen in Figure 6.11.b, it was not always possible to obtain a completely flat surface at the top of the pool. When this occurred, the DNAPL pool height was taken as the average height of DNAPL in the reservoir tube. Typical pool heights ranged from 10 mm to 50 mm, and were less than critical heights so that infiltration, and even pre-infiltration (see Section 6.2.2), into the capillary tubes would not take place before testing in the centrifuge.

Following the pool preparation, the centrifuge strong box and its contents were transported to the centrifuge room. A diagram and a photograph of the centrifuge setup prior to starting the test are shown in Figure 6.12.a and Figure 6.12.b, respectively. The strong box was first bolted onto the centrifuge main platform. Because the box was not initially designed for centrifuge tests, and since the fluid pressure acting on the walls of the strong box could become substantial as the \( g \)-level increased, it was decided to reinforce the box by clamping two reinforcement bars at the bottom of the box (see Figure 6.12.b). The box was successfully centrifuged to an acceleration corresponding to a \( g \)-level of 15.8 with no incident. However, a new strong box should be used if more tests are to be run in the future.

A fluorescent light source was located behind the strong box, making it possible to observe the flow through the capillary tube while in flight. It was best to interpose a diffuser between the light source and the box, as the fluorescent light otherwise produced a non-uniform illumination making it difficult to observe the infiltration process.

At the time, these experiments were performed, the electrical connections needed for instrumentation, including light power, camera power and signals, were made through a set of 24 slip rings (see Figure 6.9.a). Electrical cables running from the slip ring connections to the platform were held in place using a series of cable ties and plastic mounts glued onto the centrifuge arm and platform, and occasionally completed with copious amounts of duct tape (see Figure 6.12.b). The main platform could be observed in the control room by means of a TV control camera, RCA model TC 2000, reinforced to operate in a centrifugal field [Ratnam, 1996]. The camera was exclusively used to ensure that no incident was occurring during the first few tests and was dismantled from the centrifuge beam at some point during the project.

The miniature camera used for DNAPL infiltration laboratory experiments (see Section 6.2.2) was set up on a pedestal bolted to the main platform. This camera faced the centrifuge strong box (see Figure 6.12) and was used to record the DNAPL infiltration experiments. The control unit of the camera was located on the centrifuge arm, next to the shaft so as to minimize the effects of centrifugal acceleration. The camera power supply was located in the centrifuge control room. The input (power) and output (image) signals were carried through the slip rings.
Figure 6.12. DNAPL infiltration centrifuge experimental setup: a. Schematic diagram; b. Photograph of the final setup and detail of a capillary tube (note: in the first few centrifuge tests, funnels as those visible inside the box were used as reservoir tubes).
The image signal coming from the miniature camera was fed to the time date generator unit (see Section 6.2.2). The stopwatch was manually synchronized to the computer time used for RPM data acquisition (see Section 6.4.3) with a precision of less than a second. All tests were recorded using the VCR set in the SP fast recording mode. An image every three to four hundredths of a second was perfectly adequate at low speeds of DNAPL infiltration, but became somewhat slow for the infiltration experiments performed on larger diameter tubes spun at higher g-levels. Again, monitoring and data reduction of the experiments were conducted using the black and white monitor used for laboratory experiments. Data reduction was performed manually by visualizing the experiment film frame by frame.

6.3.4 Centrifuge Control and Experimental Package Testing

The operation of the centrifuge was performed by a computer located in the control room. A multipurpose microprocessor controlled the centrifuge through a program written in Forth. A complete description of the controller can be found in the Genisco Operating Manual available in the centrifuge laboratory. The description here is limited to the features used, as well as the experience gained through the course of this centrifuge testing program.

All tests were run in the Program Mode, for which a series of one to fifteen operation segments containing three input parameters were entered. For each operation segment, it was necessary to indicate: (1) the target velocity in RPM (between 0 and 410 RPM) of a run set; (2) time in seconds (between 0 and 300 s) during which the target RPM was maintained; (3) time in seconds (between 0 and 300 s) during which the centrifuge ramped from the RPM of this run set to the target RPM of the run set that immediately followed. Obviously, the target RPM was not reached if the ramp time was less than the response time required for acceleration or deceleration of the centrifuge to this target RPM.

Once the program was entered and started, the centrifuge followed the specified series of run tests, which usually consisted of an acceleration phase, with possible RPM plateaus, and a deceleration phase. The keyboard spacebar acted as a break and was convenient to use once the DNAPL infiltration was complete, or to abort a test that would otherwise have followed the specified program until its end. However, under abort conditions, the deceleration was rapid and water tended to spill out of the experimental package during the process. The red emergency button located on the front panel of the control room table shut off the electrical system of the centrifuge but did not action its breaks. Therefore, the centrifuge free-spun for some time before it reached a full stop. This was not a recommended procedure for ending a test.

Experience showed that there were discrepancies between actual and programmed rotational velocities. Typically, the actual RPM was 5 RPM to 8 RPM larger than the target RPM [Marulanda, 2001]. The discrepancies were taken into account during the programming of the run sequences, but did not compromise the experiments, which
were not aimed at achieving a particular target RPM. It was also noticed that the rotation velocity tended to fluctuate by ±1 RPM during the onset of a velocity plateau. Fluctuations tended to stop once the centrifuge had been running for a while. Thus, a centrifuge test usually included a warm-up plateau at the beginning of the experiment, which consisted of a 180 s run at a speed of 30 RPM, corresponding to a $g$-level below typical $g$-levels at infiltration.

Figure 6.13. Experimental setup during rotation. The container swings up such that the combination of centrifugal force and Earth’s gravity acts normal to the platform.

During each test and after the warm-up plateau, the $g$-level was continuously increased at a rate of 0.1 RPM per second. This corresponded to a rate of the order of 0.02 $g$ per second, although this rate varied since the $g$-level is proportional to the square of the rotational velocity (see Appendix B, Section B.2). Upon accelerating, the centrifuge platform on which the experiment was set up rotated freely about the trunnion, such that the overall component of the Earth’s gravity and the centrifugal acceleration remained normal to the centrifuge platform. This is shown in Figure 6.13. At each increasing $g$-level, the DNAPL pool of the equivalent prototype increased until it reached the critical pool height required for infiltration. At this point, the infiltration process took place and was recorded with the miniature camera. The issue of pre-infiltration of DNAPL during centrifuge experiments is further discussed in Section 7.3.3.2.
RPM data were recorded during the test (see Section 6.4) and later used to back-calculate the g-level at the onset of infiltration, using the procedure described in Appendix B. Because the DNAPL travel time through the capillary tube was of the order of seconds, the g-level acting on the centrifuge model could be assumed constant throughout the infiltration process. In addition, because the internal diameter of the reservoir tube was significantly larger than that of the capillary tube, it was assumed that the pool height change was negligible throughout the infiltration process. This assumption was made in the model development presented in Chapter 3 (see Section 3.4.1).

Following a centrifuge infiltration test, the strong box was transported back to the fume hood of room 1-047. Water free of pure-phase DNAPL was first removed using the peristaltic pump (Masterflex). The various parts were removed and cleaned. The liquids were then disposed of following the procedure described in Section 5.4.1.

6.4 Centrifuge Instrumentation and Data Acquisition

6.4.1 Overview

The object of this section is to present the data acquisition system used for the centrifuge program as well as the tachometers used to measure the centrifuge RPM and relay a signal readable by the data acquisition system. The data acquisition hardware and software are described respectively in Section 6.4.2 and Section 6.4.3. The tachometers are presented in Section 6.4.4. Since the end of this research, many modifications have been made to the centrifuge equipment. In particular, the data acquisition system has been completely redesigned.

6.4.2 Data Acquisition System: Hardware

The data acquisition system used for the centrifuge test program consisted of a 486-microprocessor personal computer combined with a set of two data acquisition cards. The data acquisition cards were designed by Chauhan [1995] and extensively described in his thesis. Chauhan made use of a modification of the MIT-designed [Sheahan, 1991] analog-to-digital conversion card (ADC), which was built around Analog Devices high resolution, programmable, integrating AD1170 converter [Analog Devices, 1992]. The AD1170 offers independently programmable integration time (from 1 ms to 300 ms) and allows the user to specify the bit-resolution.

The ADC serves as a translator between the DC signal produced by the measurement devices—RPM signal in this case—and the computer. Up to sixteen DC voltages, each one of them referred to as data acquisition channel, can be measured
using one of these cards. The ADC can convert analog readings to a resolution ranging from 7 to 22 bits (1 in $2^{22}$) over a ±5-volt range. Usable resolution is typically limited to 18 bits due to measurement and calibration noise error [Da Re, 2000]. This corresponds to a maximum resolution of $10/(2^{18} - 1) = 0.038$ mV if a ±5-volt range is used. For this research, the 16-bit precision was quite sufficient. Indeed, the resolution of the tachometers was the limiting factor on the precision of the centrifuge test data (see Section 6.4.4).

Prior to digital conversion, the analog signals are fed through an impedance amplifier AD524 [Analog Devices, 1994] which converts the DC voltage (differential signal) into a ground referenced single-ended signal and feeds it to the AD1170 converter chip. The gain of the AD524 card can be set to a value of up to 1000 if the input differential signal is small in magnitude, thus reducing the relative magnitude of the digital noise associated with the ADC. The output signal of the AD524 has to be trimmed using a variable resistor so as to remove the output offset. In this research, this was done periodically by short-circuiting the input signal and adjusting the output signal to a zero voltage.

The main difference between the old [Sheahan, 1991] and the new [Chauhan, 1995] data acquisition cards is that the old card contains an AD524 amplifier for each of the eight data acquisition channels, whereas the new card only contains one amplifier for sixteen channels. Therefore, the gain cannot be varied from one channel to the other using the new card. This was not a problem for this work, since data acquisition was limited to one channel, the RPM signal, whose input voltage ranged between 0 and 5 volts, and for which a gain of 1 was set on the AD524 amplifier. Other projects [Marulanda et al., 2000] used a set of two data acquisition cards in order to be able to work with two different gains. For that reason, the data acquisition software of this project was written in collaboration with Marulanda to accommodate two ADCs.

6.4.3 Data Acquisition System: Software

The centrifuge data acquisition software is a modification of the data acquisition software initially written by Sjoblom [2000] and described in his thesis. Sjoblom wrote a data acquisition program for the AD1170 data acquisition card in QuickBasic and applied it for acquiring data from an electronic balance. A number of Sjoblom’s subroutines were recycled for this present work, including the following: (1) the subroutine that sets up the ADC; (2) the subroutine that reads the voltages off each channel; and (3) the subroutine that writes time and voltage readings to the output files. Plot subroutines were not included in order to save processing time.

New setup subroutines as well as an RPM calculation subroutine were written and added. The typical time span of a centrifuge test was of the order of several tens of minutes compared to several days for one of Sjoblom’s tests. Hence, the structure of
the time loop was also modified. A complete listing of the data acquisition code is included in Appendix A. An algorithm is presented in Figure 6.14.

As mentioned at the end of Section 6.4.2, the software accommodated two ADCs. A first subroutine asked the operator if a new parameter file had to be created or, alternatively, what parameter file had to be loaded (see Figure 6.14). The parameter file contains the following information: (1) the number of channels from 0 to 16 read from the card that has a gain larger than 1, called low-voltage channels; (2) the number of channels from 0 to 16 read from the card that has a gain of 1, called high-voltage channels; (3) the time interval between readings in seconds; (4) the integration time of the ADCs to be picked from 7 possible integration times ranging from 1 ms to 300 ms.

For the purpose of this experimental work, only the first high voltage channel, called channel 0, was used. Channel 0 was the channel connected to the tachometer (see Section 6.4.4), which read the centrifuge RPM.

The choice of the time interval between readings was obviously affected by the number of channels that were read by the cards plus the time it took to write the measured data to a file. For example, it was not possible to read eight channels every second if the integration time was 300 ms for each channel. On the other hand, a large integration time increased the length of the averaging window, and was best for eliminating any high frequency noise associated with the signal. Thus, for a given number of channels, there was a tradeoff between the time interval between readings and the integration time of the processor. For this experimental program, channel 0 was read every second and, for that reason, the maximum integration time of 300 ms could be used.

It is important to note that only the number of channels was specified in the parameter file, but not which channel number. For example, if three low-voltage channels were entered as a parameter, the data acquisition program would successively read channel 0, 1 and 2. Hence the corresponding transducer outputs had to be connected at these locations. A transducer connected to channel 5 would be read only if channels 0 through 5 were read.

The initial version of the data acquisition program contained two other pieces of information in the parameter file. First, the operator had to give the maximum RPM at which the centrifuge test was to be performed. This piece of information was necessary to use the centrifuge tachometer described in Section 6.4.4.2, and select one of the four calibration coefficients necessary to compute the centrifuge RPM. Upon using the second centrifuge tachometer, described in Section 6.4.4.3, the calibration coefficient was fixed independently of the maximum RPM used during the test. In the program listing given in Appendix A, the subroutine Switch and code lines that became unnecessary were kept as comments—the comment code lines start with a dash.

The second piece of information the user needed to provide was the height relative to the base of the platform at which the g-level was to be computed from the RPM. This enabled the program to compute and display both the RPM and the g-level during the centrifuge test. Although interesting, this feature only slowed down the speed of data acquisition to provide a g-level that could be calculated later by reducing the RPM data. Hence, it was decided to disable this feature. Again, the corresponding code lines were kept as comments in the listing.
Figure 6.14. Algorithm of the centrifuge data acquisition program.
Following the creation or loading of the parameter file, the two data acquisition cards were set up and ready to grab data with a 16-bit precision. The output data file and the computer screen display were prepared. As shown in Figure 6.14, during each time interval, a loop was run to grab voltage data from each of the channels that had been specified in the parameter file. The RPM was computed from the tachometer reading. RPM and voltage data were then displayed on the computer screen and written to the output data file along with the time that had elapsed since the beginning of the data acquisition subroutine. The program was stopped as soon as the operator pressed the escape key esc on the keyboard.

The output data file could be opened on any spreadsheet software, such as Microsoft Excel for data reduction.

6.4.4 Centrifuge DC Output Tachometer

6.4.4.1 Overview

To the author’s knowledge, there has been no prior attempt on the MIT geotechnical centrifuge to obtain an output data file containing time and RPM information. Recent work [Ratnam, 1996] conducted work at targeted rotation velocities and relied on RPM data displayed by the centrifuge controller monitor. In Ratnam’s study, a pressure transducer was used, but only hand readings were made from a voltmeter and no automated data acquisition was used.

This present study relied on determining at which g-level infiltration of DNAPL was taking place throughout a phase of slow but continuous RPM increase. Consequently, it was important to obtain an accurate record of RPM versus time.

The idea was to find a signal in the centrifuge control system from which a DC signal, comprised between 0 and 5 V and linearly related to the centrifuge RPM, could be generated. Two tachometer systems meeting these specifications were built. These systems are presented in Section 6.4.4.2 and Section 6.4.4.3, respectively. The first tachometer system was used for centrifuge tests 2 through 17. The second tachometer system was used for centrifuge tests 18, 19 and 20 (see Appendix C).

6.4.4.2 Tachometer From Direct DC Signal

When the centrifuge is spinning, the centrifuge controller sends a command signal to the motor drive of the centrifuge via the processor unit assembly located in the centrifuge control room. The centrifuge sends back a 600 pulse per revolution (PPR) encoder, which acts as a tachometer feedback for the centrifuge controller. The command signal is a DC voltage ranging linearly from 0, when the centrifuge is not
spinning, to approximately 27 V when the centrifuge spins at 400 RPM. Thus, a first simple approach to obtain a good estimate of the centrifuge velocity is to connect to the controller command signal, and use a series of cut-off resistors to reduce the DC voltage to a maximum of 5 V, which the data acquisition system can read.

The communication connections between the controller and the centrifuge are located on the back of the processor unit assembly stacked inside the control desk (see drawing number 1231-1092 of the Genisco centrifuge manual). Among the series of connections, a separate pair of connections, labeled 1 and 2, is located on the upper right corner of the back of the assembly. This pair sends the control command DC signal to the centrifuge via connection A and B (see wiring diagram on drawing number 1231-1086 of the Genisco centrifuge manual). The connections 1 and 2 were used as the input signal for the voltage cut-off circuit.

Many of the centrifuge tests did not exceed rotation velocities of the order of 100 RPM. Hence, it did not seem worthwhile to use a single pair of cut-off resistors for which the maximum circuit output voltage of 5 V would correspond to the maximum rotation velocity of 400 RPM. Instead, a series of cut-off resistors and a switch were used in order to maximize the output voltage for the velocity range of the test. A diagram of the circuit is shown in Figure 6.9. Four selections were possible on the switch, corresponding to maximum speeds of 75 RPM, 225 RPM, 350 RPM and 400 RPM, respectively. The voltage ranges for the controller DC signal, $V_{in}$, and their associated cut-off circuit output voltage ranges, $V_{out}$, are shown in Table 6.1.

![Figure 6.15. Centrifuge control signal cut-off circuit.](image-url)
When using this tachometer, the centrifuge data acquisition program asked the operator what maximum RPM the centrifuge was going to reach during the test. Based on the answer, the operator was invited to turn the switch to a given selection corresponding to the appropriate voltage cut-off. Then, the software picked the appropriate calibration factor to turn output voltage readings into centrifuge RPMs.

<table>
<thead>
<tr>
<th>RPM Range</th>
<th>$V_{in}$ Range [V]</th>
<th>Switch Selection</th>
<th>Cut-off Ratio</th>
<th>$V_{out}$ Range [V]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 to 75</td>
<td>0 to 5</td>
<td>1</td>
<td>0.92</td>
<td>0 to 4.6</td>
</tr>
<tr>
<td>0 to 225</td>
<td>0 to 15</td>
<td>2</td>
<td>0.34</td>
<td>0 to 5.1</td>
</tr>
<tr>
<td>0 to 350</td>
<td>0 to 23</td>
<td>3</td>
<td>0.21</td>
<td>0 to 4.9</td>
</tr>
<tr>
<td>0 to 400</td>
<td>0 to 27</td>
<td>4</td>
<td>0.13</td>
<td>0 to 3.5</td>
</tr>
</tbody>
</table>

 Calibration factors for each switch selection were obtained by running the centrifuge at a set of rotational speeds. Each rotational speed was maintained for 120 sec. The auxiliary 600 PPR encoder was used to obtain a direct measurement of the rotational velocity. The encoder BNC connector output is located on the back of the processor unit assembly and is labeled J3 (see drawings number 1221-1087 and 1221-1088 of the Genisco centrifuge manual). The encoder was connected to a digital oscilloscope (Gould type 1425), and its period, corresponding frequency and RPM were measured for each velocity plateau. For quality control, this RPM measurement was compared to the RPM displayed on the centrifuge controller screen. Output voltage readings were taken by the data acquisition system throughout the test. For illustration, the calibration cycle of switch selection 3 is shown in Figure 6.16.

 For each given rotational velocity, a time average of the measured voltage output was computed. Next, for each cycle, the RPM data were plotted versus the averaged output voltage data. A calibration factor for a given switch selection was obtained by performing a linear regression on the cycle of this switch selection. The plots and linear regressions obtained for each switch selection cycle are shown in Figure 6.17.

 The scatter associated with the output signal at a given rotational velocity was used to estimate the noise level. It was found that the scatter was typically of the order of ±1 mV using switch selection 4, ±2 mV using switch selection 3, ±3 mV using switch selection 2, and ±10 mV using switch selection 1. Thus, the noise level was roughly proportional to the cut-off ratio shown in Table 6.1, suggesting that noise was almost exclusively generated by the input signal. Therefore, relative errors on RPM measurements were higher at lower RPMs. For example, at 60 RPM, i.e. switch selection 1, the output voltage was 3.69 V. Hence, for this RPM, the noise level of ±10 mV gave a relative error of 0.27%.
Figure 6.16. Centrifuge RPM calibration cycle of switch selection 3.

Figure 6.17. Centrifuge tachometer calibration curves for switch selection 1 through 4.
More problematic about the overall performance of this tachometer was that the signal being read was not the actual RPM of the centrifuge. Rather, it was the command signal given by the controller to the centrifuge. Hence, it somewhat overestimated the actual RPM during an acceleration phase if the rate of acceleration and the response time of the centrifuge were large.

Another issue related to the device was that it tapped onto a command signal. Considering that the impedance of the cut-off circuit was much larger than the impedance of the circuit of the centrifuge that received the signal, little current was going through the cut-off circuit, and the run of the centrifuge was not affected. However, when no command signal was sent by the centrifuge control program, any small current coming from the data acquisition computer could control the centrifuge through the cut-off circuit. Such a scenario was observed. In fact, there were times at the end of a test when the centrifuge would keep running at 1 RPM or so, despite receiving no command from the control program. Under these circumstances, a control signal was sent by the data acquisition program. Because this raised safety concerns, it was decided to use the alternative approach described in Section 6.4.4.3.

6.4.4.3 Tachometer From Frequency to Voltage Conversion

This tachometer design made use of the auxiliary 600 PPR encoder located on the processor unit assembly (see Section 6.4.4.2). During each centrifuge revolution, a series of 600 pulses is sent through the J3 BNC connector. For a centrifuge rotational velocity between 0 and 400 RPM, this corresponds to a signal varying between 0 and 4 kHz. A frequency-to-voltage converter AD650 (Analog Devices) was used to transform the encoder into a DC voltage. The circuit was designed using the guidelines described in Analog Devices [1998], although a much clearer design note has been released since the end of this research centrifuge experimental program [Martin, 2000]. Basically, the AD650 integrates an internal current source over a period of time that is related to the period of the input signal—the 600 PPR encoder in this case. Thus, the output voltage is a function of the input signal frequency and the characteristics of two components to select: a capacitor, $C_{OS}$, and a resistor, $R_{INT}$. A third component, the capacitor $C_{INT}$, is determined by the mechanical response time of the device being measured—i.e. the centrifuge.

If $t_{OS}$ is the time during which the current source is integrated, then [Martin, 2000]

$$V_{out} = t_{OS} R_{INT} i_{cs} f_{in},$$

(6.1)

where $i_{cs}$ is the magnitude of the internal source current (equal to 1 mA), $V_{out}$ is the output voltage, and $f_{in}$ is the frequency of the input signal, $V_{in}$. The time $t_{OS}$ must be less than the minimum period of the signal to be measured, i.e. $1/(4000 \text{ Hz}) = 250 \mu\text{s}$ for the centrifuge.
The capacitance $C_{OS}$ and the integration time $t_{OS}$ are related by [Martin, 2000]

$$C_{OS} = t_{OS} - \frac{3 \times 10^{-7}}{6.8 \times 10^3} \text{ sec/F}. \quad (6.2)$$

Thus, taking a capacitance $C_{OS}$ of 4.7 nF and a resistance $R_{INT}$ constituted by one fixed resistance of 28 kΩ and one variable resistance of 10 kΩ, $t_{OS} \approx 32.3 \mu s$ according to (6.2). Using (6.1), it is found that the output voltage $V_{out}$ at $f_{in} = 4$ kHz varies between 3.61 V and 4.90 V depending on how the variable resistance is adjusted. Therefore, $R_{INT}$ can be trimmed so that the output voltage is set to 4 V at 4 kHz, giving a very simple calibration factor of 100 RPM per volt of output voltage.

It is next necessary to calculate the integration capacity, $C_{INT}$, which can be obtained using the equation [Martin, 2000]

$$R_{INT}C_{INT} = \frac{\text{Mechanical response time}}{N}, \quad (6.3)$$

where $R_{INT}C_{INT}$ can be seen as the time constant of the internal integrator circuit and $N$ is the number of time constants chosen to allow adequate settling of the integrated signal.

The exact mechanical response time of the centrifuge to a sudden change in rotational velocity command signal is not known, but is probably of the order of a few seconds. Nonetheless, the centrifuge data acquisition system takes data at a minimum period of one second, which can be taken as the minimal response time required for this design. Thus, assuming a response time of one second, taking $C_{INT}$ equal to 1 µF, and making use of (6.3), it can be shown that $N$ varies between 26 and 36 depending on $R_{INT}$. A minimum number of time constants $N$ of 11.0 gives a 16 bit precision [Martin, 2000]. Therefore, the choice of $C_{INT}$ is adequate, since it is beyond the precision of the data acquisition system.

It should be noted that the output signal, $V_{out}$, is not a continuous signal. In fact, $V_{out}$, as given by (6.1), is the average output value of a triangular signal of period $f_{in}$. A typical output signal is shown in Martin [2000]. The peak-peak output ripple, $V_{PP}$, is given by [Martin, 2000]

$$V_{PP} = R_{INT}C_{INT} \left( e^{i(f_{in}t_{OS}-R_{INT}C_{INT})} - e^{i(f_{in}t_{OS}R_{INT}C_{INT})} + e^{i(f_{in}t_{OS}R_{INT}C_{INT})} -1 \right) \frac{1}{1 - e^{i(f_{in}R_{INT}C_{INT})}}. \quad (6.4)$$

It can be shown that the output ripple, $V_{PP}$, decreases when the integration capacity, $C_{INT}$, increases. It is also largest at the minimum of the frequency range over which $f_{in}$ varies. Taking 30 RPM as the lowest centrifuge velocity of interest gives $f_{in} = 300$ Hz. Using $C_{INT} = 1$ µF and any value for $R_{INT}$ between 28 kΩ and 38 kΩ, gives $V_{PP} = 0.319$ V, to compare to the 0.3 V average output voltage if a calibration factor of 100 RPM per volt of output voltage is used. For $f_{in} = 4$ kHz, $V_{PP} = 0.281$ V,
which compares to the 4 V average output voltage. Thus, the magnitude of the ripple is not negligible with respect to the average output voltage. This could have been problematic if the tests had not used a long data acquisition integration time. The minimum frequency of the range of interest, $f_{in} = 300$ Hz, corresponds to the maximum period of 3.33 ms. Because the data-acquisition integration time was 300 ms, the output signal was averaged over 90 periods.

![Diagram of the frequency-to-voltage conversion circuit](image)

**Figure 6.18.** Frequency-to-voltage conversion circuit.

The circuit was built using design components having the values given above. A diagram of the circuit is shown in Figure 6.18 and is very similar to the diagram provided by *Analog Devices* [1998]. The capacitance $C_{OS}$ was found to be extremely sensitive to the circuit noise, and consequently was shielded from the rest of the circuit. The ±15V power supply and ground reference were provided by a constant-voltage DC power supply (Power General, model 326ACM) widely used in the MIT geotechnical laboratory.

The two variable resistors were trimmed so as to achieve a calibration factor of precisely 100 RPM per volt of output voltage. First, the input voltage connections were short-circuited, and the 20 kΩ-variable resistor connected between pins 13 and 14 was trimmed (see Figure 6.9) to achieve a 0 V data acquisition output voltage. Next, the centrifuge was run at a series of different rotational velocities, and the 10 kΩ variable resistor (see $R_{INT}$ component in Figure 6.18) was adjusted so as to achieve the desired calibration factor. In general, excellent agreement was found between the RPM displayed on the centrifuge controller screen and the RPM obtained from the data acquisition system, with no more than 1 RPM difference at 100 RPM. The tachometer
had to be periodically recalibrated. To date, it appears that the tachometer design would benefit from a better quality component for $C_{OS}$ with shorter connections to the AD650. It would also benefit from less peak-to-peak ripple by increasing the magnitude of $C_{INT}$ to 10 µF.

6.5 References


CHAPTER 7

TEST RESULTS AND DATA

7.1 Introduction

This chapter presents the experimental results of DNAPL infiltration experiments and compares them to the models developed in Chapter 3 and Chapter 4. The chapter is divided into two main parts. Section 7.2 is restricted to laboratory experiments, i.e. those performed at the Earth’s gravitational acceleration (1-g experiments). Results of measurements of infiltration heights are examined in Section 7.2.1. The kinetics of infiltration and comparison with the NICCA model (see Section 3.5.2) and NIVCA model (see Section 3.5.3) are presented in Section 7.2.2 and Section 7.2.3, respectively.

Section 7.3 focuses on the infiltration centrifuge experiments. A brief introduction is given in Section 7.3.1. Validation of centrifuge scaling laws through modeling-of-models is examined in Section 7.3.2. Section 7.3.3 presents the results of the prototype infiltration height measurements. In Section 7.3.4, the infiltration kinetics of the centrifuge experiments are examined and compared to the NICCA physical model developed in Chapter 4. A comparison of the experimental data with the NIVCA model is examined in Section 7.3.5. Finally, in Section 7.3.6, summary plots characterizing the infiltration kinetics of centrifuge experiments are presented and compared to the results of the laboratory (1-g) experiments.

A list of references made in this chapter is provided in Section 7.4.

General conclusions arising from the outcome of the work presented here will be drawn in Chapter 8 along with recommendations for further work.

7.2 DNAPL Infiltration Laboratory Experiments

7.2.1 Infiltration Heights: Results and Interpretation

7.2.1.1 Summary of Results

Table 7.1 summarizes the results of the measurements of DNAPL infiltration pool height for spontaneous infiltration laboratory experiments (i.e. at 1-g condition) run on 4-chlorotoluene (4-CT) and 1,1,1-trichloroethane (TCA). The experimental
methodology used for the tests is described in Section 6.2.2. Details on each experiment can be found in Appendix C, Section C.2 and Section C.3 for 4-CT tests and TCA tests, respectively. As mentioned in Section 6.2.2, for these tests, DNAPL was pooled slowly until infiltration took place. Therefore, it can be expected that the pool height at the onset of infiltration was close to the critical pool height, so that $h \approx h_i$ ($\Delta h \approx 0$) can be assumed. Recall that a small increment of pool height above $h_i$ is necessary for infiltration to take place (see discussion in Section 3.5.2.2).

Table 7.1. Summary of Infiltration Pool Heights for DNAPL Spontaneous Infiltration Laboratory Experiments

<table>
<thead>
<tr>
<th>DNAPL</th>
<th>Capillary Tube Diameter [mm]</th>
<th>Measured DNAPL Pool Height at Infiltration</th>
<th>Theoretical Infiltration Height [mm]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Average [mm]</td>
<td>SD [mm]$^b$</td>
<td>Max. Deviation [mm]$^c$</td>
</tr>
<tr>
<td>4-CT</td>
<td>2.70</td>
<td>57.6</td>
<td>11.3</td>
</tr>
<tr>
<td></td>
<td>2.20</td>
<td>67.0</td>
<td>3.8</td>
</tr>
<tr>
<td></td>
<td>1.33</td>
<td>137.4</td>
<td>10.3</td>
</tr>
<tr>
<td></td>
<td>0.66</td>
<td>263.9</td>
<td>38.9</td>
</tr>
<tr>
<td>TCA</td>
<td>2.20</td>
<td>10.1</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>1.33</td>
<td>24.1</td>
<td>1.1</td>
</tr>
<tr>
<td></td>
<td>0.66</td>
<td>53.3</td>
<td>0.65</td>
</tr>
</tbody>
</table>

\(^{a}\) The measured pool height at infiltration is the sum of the reservoir tube DNAPL height and the pre-infiltration depth at the onset of infiltration.

\(^{b}\) Standard deviation. The number in parentheses is the coefficient of variation, i.e. the ratio of standard deviation to average.

\(^{c}\) Maximum deviation between a measurement and the average for the series of tests. The number in parentheses is the maximum deviation relative to the average.

\(^{d}\) Theoretical infiltration height calculated using (3.4) and taking densities listed in Table 5.1 and a value of $\sigma$ equal to $0.032 \pm 0.004$ N/m for 4-CT and $0.0344 \pm 0.0029$ N/m for TCA. Perfect wetting ($\theta_s = 0$) is assumed.

As mentioned in Section 6.2.2, a DNAPL pre-infiltration finger present in the capillary tube was observed during each laboratory test. The pre-infiltration depth could be significant with respect to the DNAPL pool height present in the reservoir.
tube. Therefore, the pool height $h_i$ reported in Table 7.1 is the sum of the DNAPL height in the reservoir tube, $h_p$, and the pre-infiltration depth at the onset of infiltration, $h_{pi}$. The effects of pre-infiltration are further discussed in Section 7.2.1.2.

A comparison between measured and predicted values of $h_i$ using the infiltration criterion (3.4) derived in Section 3.3 is given in Figure 7.1, where the measured infiltration heights have been plotted versus the diameters of the capillary tubes used for the experiments. For 4-CT, a prediction zone delimited by the dashed lines is calculated using the upper and lower limits of the interfacial tension measured using the pendant drop method (see Section 5.6.3 and Section 5.7). For TCA, the prediction is based on the upper and lower limits of the interfacial tension measured using the combined capillary rise method (see Section 5.6.4 and Section 5.7). Perfect wetting ($\theta_s = 0$) is assumed for each prediction plot. The density properties used in (3.4) are listed in Table 5.1. The Earth’s gravitational acceleration, $g$, is taken as $9.807 \text{ m/s}^2$ [Munson et al., 1994].

![Figure 7.1](image.png)

**Figure 7.1.** Measured DNAPL pool height at the onset of infiltration for different capillary tube diameters. Discrete points represent experimental data for 56 spontaneous infiltration laboratory tests. Dashed lines represent predictions for different interfacial tension, $\sigma$, and assuming perfect wetting ($\theta_s = 0$).
7.2.1.2 Discussion

Figure 7.1 indicates relatively good agreement between the measured and predicted values of DNAPL pool height at infiltration. The average measured infiltration heights reported in Table 7.1 are generally less than those predicted by (3.4) for both 4-CT and TCA. Indeed, the average measured infiltration heights are approximately 15% to 20% smaller than those predicted, with the exception of the 4-CT infiltration test series using 1.33 mm diameter capillary tubes. For this test series, the average measured infiltration height is found to be 0.8% larger than that predicted by (3.4).

Despite the good agreement, a large scatter of results can be seen in Figure 7.1. For 4-CT, large relative deviations from the average can be noted. Those deviations vary from 10% to 40%, depending on the capillary tube diameter under consideration (see Table 7.1). The coefficients of variation of the measured infiltration heights vary from 5% to 20% depending on the capillary tube diameter.

As shown in Figure 7.1, the scatter in the data is not due to outliers. Rather, there is a continuous distribution of measured infiltration heights. Given the variability of the measurements, one significant digit (i.e. ±10%) is what can be counted on for predicting the infiltration height of a DNAPL in a capillary tube of a given diameter. While the results for TCA appear more repeatable than those of 4-CT, the difference can be attributed to the smaller number of tests performed on TCA.

There are several mechanisms believed to be responsible for the variability of results of 4-CT spontaneous infiltration. These factors are treated under separate headings below.

\textit{Variability of interfacial tension}

Variability of interfacial tension was observed during measurements using the pendant drop technique (see Section 5.6.3 and Figure 5.10), where it was shown that the interfacial tension decreased with time, particularly within the first hour of the life of the drop. Indeed, for the Sudan IV-dyed 4-CT/water system, a reduction of about 0.003 N/m (9%) between the interfacial tension of a 2 minute-old drop (about 0.034 N/m) and that of a one hour-old drop (about 0.031 N/m) can be estimated from a linear regression of the data presented in Figure 5.10.

The time of contact between water and DNAPL varied from one laboratory test to the other, from a pool build-up time of about 10 minutes to a period of several hours (a precise time log was not kept). Therefore, it is possible that the interfacial tension at the onset of infiltration may have varied from one test to the other. The infiltration height prediction zone shown in Figure 7.1, incorporates the effects of the variability of the interfacial tension. As can be seen in the figure, most data points fall within the prediction zone. Thus, the change over time of the interfacial tension constitutes one hypothesis explaining the scatter of results.

Variations in interfacial tension related to temperature might also affect the measurements of the infiltration height, and will be addressed here. The effects of
temperature on the interfacial tension of the DNAPLs were not investigated. Although correlations between temperature and DNAPL surface tensions (DNAPL/air) can be found in the literature [Mercer and Cohen, 1990], no such correlations exist for DNAPL/water interfacial tensions. Generally, the DNAPL surface tensions decrease linearly by a factor of $1 \times 10^{-4} \text{ N/m}$ to $1.4 \times 10^{-4} \text{ N/m}$ per °C [Mercer and Cohen, 1990]. Assuming a similar reduction for DNAPL/water interfacial tensions, a temperature increase of 5°C would be associated with an interfacial reduction of up to 0.0007 N/m, corresponding to a 2% relative reduction if a value of 0.032 N/m is assumed for the 4-CT/water interfacial tension. Thus, temperature effects appear to be less important than the effects of DNAPL/water interface aging.

**Pre-infiltration of DNAPL: Observations**

As discussed in Section 6.2.2, it was observed that, during the infiltration tests, at the early stages of pooling, when the pool height was still significantly less than the final infiltration height, a finger of DNAPL would pre-infiltrate the tube to a depth of several millimeters. Again, pre-infiltration means that a finger of DNAPL is observed to infiltrate the capillary tube but the downward movement of the finger is suddenly stopped and does not proceed unless more DNAPL is added to the reservoir pool. For 1.33 mm diameter capillary tubes, pre-infiltration was first observed for a reservoir pool height, $h_p$, ranging from 40 mm to 60 mm. For 2.70 mm diameter capillary tubes, pre-infiltration was first observed for a reservoir pool height, $h_p$, ranging from 10 mm to 20 mm. For 0.66 mm diameter capillary tubes, pre-infiltration was first observed for a reservoir pool height, $h_p$, of the order of 100 mm (note that this value is only based on the records of one spontaneous infiltration experiment).

As the test progressed and more DNAPL was added, the pre-infiltration length increased down to depths as large as 60 mm and could be become so significant that its contribution had to be taken into account when computing the pool height at the onset of infiltration. Eventually, the pre-infiltration finger no longer stopped, and complete infiltration of the tube would take place. No clear repeatability pattern on the pre-infiltration length could be inferred from the test series. In particular, the pre-infiltration could change for two consecutive infiltration tests run on the same capillary tube (see, for example, test series L8-1.33st-122mm in Section C.2.3).

It was also noted that DNAPL pre-infiltration fingers left overnight would generally creep by a few millimeters. This observation is consistent with the reduction in interfacial tension with time reported above, since a reduction of interfacial tension contributes to lower the critical height required for infiltration.

To illustrate the pre-infiltration phenomenon, photographs of DNAPL fingers during the build-up of a DNAPL pool are shown in Figure 7.2 for a 1.33 mm diameter capillary tube. Figure 7.2.a shows that pre-infiltration has taken place down to a depth of about 3 mm. At a later stage of the build-up shown in Figure 7.2.b, the pre-infiltration is down to approximately 20 mm. As illustrated in Figure 7.3, similar observations can be made for a 2.70 mm diameter capillary tube.

That pre-infiltration could be connected to an over-pressurization of the reservoir tube by the DNAPL syringe used for building up the pool has been ruled out.
Spontaneous infiltration test L14-1.33slt-305mm (see Section C.2.3) run on a large, 32.5 mm diameter reservoir such that the extra pressure generated by the syringe was negligible showed a pre-infiltration pattern identical to that of tests with reservoir tubes of smaller diameters.

![Figure 7.2](a) Early stage of pre-infiltration; b. Later stage of pre-infiltration.

**Figure 7.2.** Pre-infiltration into a 1.33 mm diameter capillary tube: a. Early stage of pre-infiltration; b. Later stage of pre-infiltration.

![Figure 7.3](a) Early stage of pre-infiltration; b. Later stage of pre-infiltration.

**Figure 7.3.** Pre-infiltration into a 2.70 mm diameter capillary tube: a. Early stage of pre-infiltration; b. Later stage of pre-infiltration.
Evidence of static contact angle different from zero

When the first drop of DNAPL is poured down the reservoir tube, the DNAPL/water interface is formed at the entrance to the capillary tube. The DNAPL/water interface meniscus has no history of motion within the capillary tube. Hence, the contact angle is not a receding contact angle (see Section 2.3.5.2 and the discussion of Section 5.6.4.3). The interface contact angle is affected by the conditions of formation of the interface at the bore of the capillary tube. Parameters including the shape and roughness of the bore, the location of the reservoir tube and the location of the Teflon tape connection between the reservoir tube and the capillary tube can affect the whereabouts of the DNAPL drop, such that the initial contact angle cannot be easily predicted. Nonetheless, visual observations suggest that it is significantly larger than 0—one can, for example, compare the interface menisci shown in Figure 7.2 with that shown in Figure 5.16.a where perfect wetting is approached. Clearly, perfect wetting cannot be expected at the pre-infiltration stage.

As shown in Figure 7.2 and Figure 7.3, the apparent static contact angles of the DNAPL/water interfaces at the base of the pre-infiltration fingers are not equal to 0. Instead, values of the apparent contact angles lie between 30° and 70°, and may be affected by the diameter of the capillary tube. Based on observations made in Section 5.6.4.3 and previous research work on liquid/liquid interfaces (see Section 2.3.5.2 and Section 2.3.6), the value of the static contact angle can be expected to vary over a range as broad as 60° [Blake et al., 1967; Fermigier and Jenffer, 1991].

Pre-infiltration as a consequence of contact angle feedback

A first hypothesis that could explain the observed pre-infiltration is a contact angle feedback mechanism, as illustrated in Figure 7.4. Because the contact angle is greater than 0, pre-infiltration takes place at a pool height less than the infiltration pool height if perfect wetting were assumed (see Table 7.1). During pre-infiltration, displacement of water by DNAPL takes place. As discussed in Section 2.3.6, under drainage conditions, the dynamic contact angle of the moving meniscus is expected to decrease (see Figure 7.4). As the contact angle decreases, the critical height required for DNAPL infiltration is increased (see (3.4) and Figure 7.4). The increase in critical height, in turn, acts to stabilize the interface meniscus. Because of the velocity decrease, the magnitude of the contact angle increases again (again, see Figure 7.4). Thus, a contact angle feedback-loop acts to slow down the interface displacement process. Note that changes in contact angle during pre-infiltration have not been observed experimentally. These changes are believed to be small (a few degrees), and would probably require an observation of the DNAPL/water meniscus at a smaller resolution than achieved in this experimental program. As will be shown later, a change in contact angle by a few degrees can affect the critical height by a magnitude consistent with the scatter reported in Table 7.1.
Pre-infiltration as a consequence of contact angle feedback and contact angle hysteresis

The contact angle feedback mechanism provides a satisfactory explanation as to why DNAPL pre-infiltration starts prior to the predicted infiltration height and is slowed down by the changes in dynamic contact angle. Nonetheless, the feedback mechanism alone does not explain why the pre-infiltration stops. Indeed, referring to Figure 7.4, once the interface pre-infiltrates by a short distance and is back at rest, the mechanism illustrated in the figure predicts that the static contact angle (i.e. the contact angle when the interface is at rest) will increase back to its initial value. However, if the contact angle were back to its initial value, the total pool height, now increased by a pre-infiltration finger, would exceed the initial critical pool height that produced pre-infiltration. Hence, the interface could not be at rest. Thus, if the feedback mechanism were the only mechanism present, the pre-infiltration would never stop.

**Figure 7.4.** Illustration of contact angle feedback mechanism: interface meniscus dynamic contact angle and critical height required for infiltration as a function of the DNAPL pre-infiltration finger displacement velocity.
A second mechanism, contact angle hysteresis (see Section 2.3.5.2), is believed to be present and responsible for stopping the meniscus displacement. During the pre-infiltration process, DNAPL displaces water, such that, as defined in Section 2.3.5.2 (see also Section 5.6.4.3), the contact angle is receding. During the receding displacement, the contact angle is believed to decrease irreversibly as a consequence of the contact angle hysteresis. Thus, once back at rest, the final static contact angle is smaller in magnitude than the initial static contact angle.

The combination of contact angle feedback and hysteresis mechanisms is illustrated in Figure 7.5. As shown in the figure, during the pre-infiltration process, the contact angle decreases due to the interface motion. This reduction in contact angle raises the magnitude of the critical infiltration height, which, in turn, reduces the interface velocity. The reduction in interface velocity acts to increase the contact angle. Because of the interface receding motion, the contact angle cannot increase back to its initial static value and is irreversibly decreased to a new static value. This new static contact angle is associated with a larger critical height required for infiltration (see (3.4) and Figure 7.5) larger than the sum of the DNAPL reservoir pool height and infiltration depth. Hence, the final critical height is compatible with the liquid column equilibrium.

**Figure 7.5.** Illustration of contact angle feedback and contact hysteresis mechanisms: interface meniscus dynamic contact angle and critical height required for infiltration as a function of the DNAPL pre-infiltration finger displacement velocity.
There is good evidence supporting the static contact angle hysteresis. Referring to the two photographs shown in Figure 7.3, the DNAPL/water interface meniscus shown in Figure 7.3.a appears slightly flatter than that shown in Figure 7.3.b, which is photographed after an increase in pre-infiltration depth. These photographs suggest that the larger the pre-infiltration depth, the smaller the static contact angle, as predicted by the mechanism shown in Figure 7.5.

The reduction in contact angle with pre-infiltration depth would imply that infiltration tests associated with larger pre-infiltration depths, \( h_{pi} \), prior to complete infiltration are associated with larger measured infiltration heights, \( h_i = h_{pi} + h_p \). This trend is well shown in Figure 7.6 where \( h_i \) is plotted versus \( h_{pi} \) for 4-CT spontaneous infiltration tests into 1.33 mm and 2.70 mm diameter capillary tubes. Clearly, the larger the pre-infiltration depth, the larger the measured infiltration height required for complete infiltration. Thus, the mechanism described in Figure 7.5 is well supported by the observed trend and suggests that the variability of the data reported in Table 7.1 can partly be attributed to the variability of the static contact angle prior to complete infiltration.

**Figure 7.6.** Influence of 4-Chlorotoluene finger pre-infiltration depth, \( h_{pi} \), on the total 4-Chlorotoluene pool height at the onset of infiltration, \( h_i \) (recall that \( h_i = h_{pi} + h_p \)).
Contact angle hysteresis and pinning effects

As discussed in Section 2.3.5.2 (see also Section 2.3.6.4), contact angle hysteresis is attributed to pinning effects. Pinning is defined as the immobilization of the interface meniscus at a particular location of the solid surface and attributed to the solid surface roughness, the solid heterogeneity or the presence of impurities at the solid surface.

The effects of pinning have been demonstrated in Section 5.6.4.3, during measurement of the DNAPL/water interfacial tension with the combined capillary rise method. Using this method, it was shown that the DNAPL/water interface could be immobilized at a particular location of a capillary tube and deform so as to insure static equilibrium of the DNAPL/water column rising inside the capillary tube.

Referring to the DNAPL spontaneous infiltration problem, it is hypothesized that, at small interface displacement velocities, such as those under consideration during pre-infiltration, small microscopic defects or impurities present on the capillary tube walls can act as pinning points and block the further downward movement of the interface. This hypothesis is well supported by the conclusions of Calvo et al. [1991] (see discussion in Section 2.3.6.4) for liquid/liquid displacements. These authors concluded that abrupt changes of dynamic capillary pressure and strong contact angle hysteresis between drainage and imbibition observed at capillary numbers below $10^{-5}$-$10^{-4}$ could be attributed to chemical impurities acting as pinning points (note that Calvo et al. used the term pinning centers).

For the 4-CT/water capillary tube system, a capillary number of $10^{-5}$ corresponds to an interface velocity of 0.32 mm/s, using the capillary number definition (2.40) (see Section 2.3.5.3) and taking the values of interfacial tension $\sigma = 0.032$ N/m, and 4-CT viscosity $\mu_{nw} = 10^{-3}$ Pa.s. A value of 0.32 mm/s is qualitatively consistent with the observed velocity of pre-infiltration.

As more drops of DNAPL are added into the reservoir tube, disturbance of the meniscus equilibrium takes place. For each increment of DNAPL pool height, it is hypothesized that the pool height and contact angle can temporarily be large enough to overcome the metastable equilibrium created by pinning forces, such that further downward movement is possible. Under these circumstances, the interface starts to move and the pre-infiltration depth increases. The interface displacement is then slowed down by the contact angle decrease associated with the meniscus motion (see above) and ultimately stopped by another pinning point.

Eventually, the combined height of reservoir pool and pre-infiltration becomes large enough to overcome any resisting force. Under these circumstances, the interface movement cannot be prevented, and complete infiltration of the capillary tube takes place.

The presence of pinning forces hypothesized for spontaneous infiltration experiments is identical in nature to those observed during combined capillary rise experiments (see discussion in Section 5.6.4.3). It is believed that the pre-infiltration length varies from one test to another because the interface can be in equilibrium under different static contact angles, and different pinning points may act to block the
interface displacement. This is thought to be the case whether the capillary tube is the same or not. Also, increments of DNAPL, consisting of one or several drops of DNAPL, were not precisely controlled, so that the history of the pool build-up was not the same for different tests on the same tube.

A new infiltration criterion

Because at the onset of pre-infiltration, the contact angle is not observed to be perfectly wetting ($\theta_s \neq 0$), it can be expected that the criterion (3.4) for infiltration is not valid, even though it provides a reasonable prediction of the infiltration height when $\theta_s = 0$ is assumed. This would suggest the existence of a so-called pinning force that contributes to the liquid column equilibrium. It is hypothesized that the magnitude of the pinning force depends on the density of pinning points (defects) present on the glass wall of the capillary tube. Thus, for complete infiltration of DNAPL to take place, the following condition is assumed

$$\pi r_0^2 \Delta \gamma gh \geq 2\pi r_0 \sigma \cos \theta_s + 2\pi n_p f_p,$$

where $h$ is the sum of the DNAPL pool height contained in the reservoir and the depth of pre-infiltration. The term $f_p$ is the pinning force immobilizing the interface in place and $n_p$ is the density of defects, i.e. the number of pinning points per unit length of capillary tube perimeter.

Let $h_i$ be defined by

$$h_i = \frac{2\sigma \cos \theta_s}{\Delta \gamma g r_0} + \frac{2n_p f_p}{\Delta \gamma g r_0}.$$  

(7.2)

If the DNAPL pool height, $h$, exceeds the critical pool height, $h_i$, then complete infiltration takes place.

Under similar conditions of manufacturing (if pinning points are small surface grooves or surface heterogeneities) as well as similar storage and cleaning procedures (if pinning points are impurities), $n_p$ can be expected to be independent of the capillary tube radius. The force $f_p$, on the other hand, may depend on several parameters, including the static contact angle $\theta_s$, the radius $r_0$ and the interface displacement velocity, $dz_c/dt$. Furthermore, it is possible that the pinning force is not entirely localized at the interface meniscus, but in its vicinity, or throughout the entire liquid/liquid column. Clearly, a precise characterization of the pinning force is beyond the scope of this thesis and would require an experimental program of its own.

Nevertheless, it is possible to obtain an estimate of the magnitude of $n_p f_p$ compatible with the experimental results of 4-CT infiltration. For this purpose, it is assumed, as a first approximation, that the static $n_p f_p$ is a constant independent of the static contact angle and capillary tube radius. Taking the 4-CT/water interfacial tension’s average value of 0.032 N/m and assuming that the static contact angle ranges
between 45° to 70°, the range of measured 4-CT infiltration heights $h_i$ is well predicted by a value of $n_{pf_p}$ equal to 0.015 N/m, as illustrated in Figure 7.7. In the figure, the dashed zone represents the contribution to the capillary resistance attributable to the pinning force, i.e. $2n_{pf_p}/(\Delta \rho g r_0)$. The dotted lines $\theta_i = 45°$ and $\theta_i = 70°$ correspond, respectively, to the maximum and minimum capillary resistance attributable to interfacial tension, i.e. $2\sigma \cos \theta_i/(\Delta \rho g r_0)$.

![Figure 7.7. Measured 4-CT pool height at the onset of infiltration for different capillary tube diameters. Discrete points represent experimental data for 48 4-CT spontaneous infiltration laboratory tests. Dashed/dotted lines represent predictions using a pinning force $n_{pf_p}$ equal to 0.015 N/m.](image)

The pinning force and contact angle values assumed above can satisfactorily predict the range of 4-CT infiltration heights associated with 1.33 mm and 0.66 mm diameter capillary tubes shown in Figure 7.7. It appears, however, that infiltration data on 2.70 mm and 2.20 mm diameter capillary tubes shown in the figure are not well predicted by the assumed values of $n_{pf_p}$ and $\theta_i$. There are several hypotheses that can explain these observations. First, it is possible that the range of static contact angles for those tubes is different from that of smaller diameter tubes, and most particularly that...
larger static contact angles can be achieved for these diameters. The increase of static contact angles with capillary tube radius has been reported before [Van Remoortere and Joos, 1993a]. Second, it is possible that the pinning force $n_{pfp}$ is affected by the magnitude of the static contact angle and the capillary tube radius, something not assumed in Figure 7.7.

![Diagram](image)

**Figure 7.8.** Measured 4-CT pool height at the onset of infiltration for different capillary tube diameters. Discrete points represent experimental data for 48 4-CT spontaneous infiltration laboratory tests. Dashed/dotted lines represent predictions using a pinning force $n_{pfp}$ equal to 0.010 N/m.

Most important, a constant pinning force of 0.015 N/m implies, using (7.2), that pre-infiltration cannot start below a minimum height of $2n_{pfp} (\Delta \rho g r_0)$—assuming that the 4-CT/water interface is completely flat ($\theta_s \approx 90^\circ$) at the beginning of 4-CT pooling. For 1.33 mm diameter capillary tubes, this means that the minimum DNAPL pool height for first pre-infiltration is equal to 64 mm (see Figure 7.7). This can be compared to the observed range of 40 mm to 60 mm reported at the beginning of this section. For 2.70 mm diameter capillary tubes, the calculated minimum height for first pre-infiltration is equal to 31 mm, which can be compared to the observed range of
10 mm to 20 mm also previously reported. Finally, for 0.66 mm diameter capillary
tubes, the calculated minimum height for first pre-infiltration is equal to 129 mm,
which can be compared to the observed value of 100 mm. Therefore, the experimental
data suggest that the magnitude of the pinning force might be lower than the estimated
0.015 N/m.

If the static contact angle is assumed to range between 30° to 60°, the range of
measured 4-CT infiltration heights $h_i$ is well predicted with a value of the pinning force
$n_{pfp}$ equal to 0.010 N/m. This is illustrated in Figure 7.8, where, again, the dashed zone
represents the contribution due the pinning force, and the dotted lines represent the
minimum and maximum contributions due to interfacial tension. For a pinning force,
$n_{pfp}$, of 0.010 N/m, the calculated minimum heights before pre-infiltration can occur in
0.66 mm, 1.33 mm and 2.70 mm diameter capillary tubes are equal to 86 mm, 43 mm
and 21 mm, respectively. Hence, the value of 0.010 N/m is consistent with infiltration
tests into 1.33 mm diameter capillary tubes, appears to be underestimated for 0.66 mm
diameter capillary tubes and overestimated for 2.70 mm diameter capillary tubes. A
pinning force value of 0.012 N/m would predict the observed pool height of 100 mm
for first pre-infiltration into 0.66 mm diameter capillary tubes, whereas a pinning force
value of 0.005 N/m would be consistent with the range of pool height 10 mm-20 mm
for first pre-infiltration into 2.70 mm diameter capillary tubes. These observations
suggest that the pinning force $n_{pfp}$ is most likely dependent upon the radius of the
capillary tube.

Figure 7.9 shows a new dashed zone corresponding to the radius dependent
pinning force consistent with the range of pool heights at first observed pre-infiltration
(a value $n_{pfp}$ of 0.07 N/m has been used for 2.20 mm diameter capillary tubes). Again,
the static contact angle is assumed to range between 30° to 60° and the interfacial
tension of 4-CT with water is taken equal to 0.032 N/m. Very good agreement is noted
between the prediction and the infiltration data except for the 2.70 mm diameter capillary
tubes. As suggested before, it is possible that the range of static contact angles is a function of the capillary tube diameter. It is also possible that the pinning
force, $n_{pfp}$, varies between tests. Overall, Figure 7.9 shows that the magnitude of the
pinning force, $n_{pfp}$, is significant compared to the magnitude of the interfacial tension
force $\sigma \cos \theta$. Both forces contribute to the capillary tube entry pressure in
proportions of about the same order of magnitude, such that the effects of the pinning
force can never be neglected when predicting the critical height at the onset of
complete infiltration.

Using (7.2), it can be shown that a reduction in static contact angle contributes to
a significant increase of the capillary height compatible with the changes reported in
Figure 7.6. For example, assuming a value of $\theta_s$ of 50° and a value of $n_{pfp}$ of
0.010 N/m, the 4-CT infiltration height of a 1.33 mm diameter capillary tube is found
to be equal to 130 mm. If the contact angle is decreased by 10°, the infiltration height
is increased to 147 mm.

Note that the variability of the measured pool height at first observed pre-
infiltration can be attributed to the variability of the static contact angle of the interface
meniscus when the interface is first formed at the entry to the capillary tube.
Considering, for example, 4-CT infiltration into a 1.33 mm diameter capillary tube; a 4-CT pool height of 40 mm for first pre-infiltration corresponds to a pinning force of 0.010 N/m and a static contact angle of 90°, whereas a 4-CT pool height of 60 mm for first pre-infiltration corresponds to a pinning force of 0.010 N/m and a static contact angle of 80°. Again, it is also possible that the pinning force is variable from one infiltration test to the other.

Figure 7.9. Measured 4-CT pool height at the onset of infiltration for different capillary tube diameters. Discrete points represent experimental data for 48 4-CT spontaneous infiltration laboratory tests. Dashed/dotted lines represent predictions using a radius dependent pinning force, \( n_{p,fp} \), ranging from 0.005 N/m to 0.012 N/m.

7.2.1.3 Conclusion

In conclusion, good agreement is noted between the measured infiltration heights and the predicted infiltration heights given by the criterion (3.4), where perfect wetting (\( \theta_s = 0 \)) is assumed, although it is generally noted that the criterion provides an upper bound of the measurement average (see Table 7.1). Overall, one significant digit is what can be counted on for predicting the infiltration height of a DNAPL in a capillary
tube of a given diameter. Nonetheless, observations of the DNAPL/water interface show that the static contact angle is not in a configuration of perfect wetting, thereby suggesting that infiltration criterion (3.4) is not strictly correct, and that there exists another force present that contributes to raise the value of the capillary tube entry pressure.

A new infiltration criterion, given by (7.2), is proposed that is in very good quantitative agreement with the experimental data. The magnitude of the so-called pinning force \( n_{pf} \) is found to be of similar order as the DNAPL/water interfacial tension, \( \sigma \cos \theta_s \), such that both interfacial tension and pinning contribute to the tube capillary resistance. The pinning force is found to decrease with increasing radius, although it must be pointed out that the pinning force back-calculated for 0.66 mm diameter capillary tubes is only based on one infiltration experiment. The variability of the measured infiltration height is attributed to two factors:

1. Variability of the interfacial tension, which may decrease significantly over time as a consequence of interface aging, and may act to reduce the infiltration height.
2. Variability of the static contact angle. The static contact angle is believed to be affected by the condition of deposition of the first DNAPL drop on top of the capillary tube, the glass wall surface of the capillary tube itself, and the amount of pre-infiltration into the capillary tube. It is found that the more pre-infiltration, the larger the measured critical height at the onset of infiltration, a trend consistent with a decrease in static contact angle.

It is not excluded that changes in magnitude of the pinning force between different infiltration tests may also contribute to the variability of results. This point will be discussed further in Section 7.2.3.6.

During the infiltration tests, pre-infiltration is observed at a pool height significantly smaller than the DNAPL pool height at the onset of complete infiltration. A mechanism combining a dynamic contact angle feedback loop and contact angle hysteresis is believed to be responsible for this observation. The feedback mechanism prevents further infiltration by decreasing the dynamic contact angle and stabilizing the interface. At small interface displacement velocities, however, pinning forces hold the meniscus in place and delay the onset of infiltration until further build-up of the DNAPL pool is achieved. Pinning points are believed to stem from individual defects (solid surface roughness), surface heterogeneity or impurities present on the walls of the capillary tubes (i.e. cleanliness of the tube). The estimated magnitude of the pinning force is consistent with the measured DNAPL pool height of first pre-infiltration. Note that variability of the pinning force between infiltration experiments is likely if the presence and density of pinning points is dependent upon the cleanliness of the capillary tube.

The infiltration height measurement results are extremely important, as they suggest that the criterion (3.4) commonly used in the literature is inadequate for predicting the DNAPL infiltration height into porous and fractured media, as pinning forces may significantly affect the magnitude of the pore/fracture capillary resistance. Furthermore, the conventional assumption of perfect wetting at the DNAPL/water interface is found to be incorrect. It is believed that the extension of liquid/gas
interface mechanisms to liquid/liquid interface mechanisms is not straightforward, as
new physics arise at the liquid/liquid interface.

To the author’s knowledge, there has been no prior study on DNAPL infiltration
experiments into vertical capillary tubes reported in the literature. Nor has there been
any downward liquid/liquid flow study. Hence, pre-infiltration and its explicative
mechanisms have never been reported before. The findings noted above therefore
constitute an important contribution to the field.

7.2.2 Kinetics of Infiltration: Results of DNAPL/Water Interface
Displacement Experiments and Comparison With the NICCA Model

7.2.2.1 Summary of 4-CT Spontaneous Infiltration Laboratory Experiments

As described in the experimental methodology (see Section 6.2.2), the
displacement of the DNAPL/water interface was recorded for a number of spontaneous
infiltration laboratory experiments reported in Table 7.1. Summaries of results for
2.70 mm, 1.33 mm and 0.66 mm diameter capillary tubes are provided in Table 7.2,
Table 7.3 and Table 7.4, respectively. Details on each series of experiments can be
found in Appendix C, Section C.2.1, Section C.2.3 and Section C.2.4, respectively.
The measured and corrected mid-depth and three-quarter depth velocities reported in
the three tables are defined and discussed in Section 7.2.2.3.

7.2.2.2 Infiltration Profiles

The observed profiles of DNAPL/water interface displacement versus time,
referred to as infiltration profiles, are shown in Figure 7.10, Figure 7.11 and Figure
7.12, for 2.70 mm, 1.33 mm and 0.66 mm diameter capillary tubes, respectively. As
already discussed in Section 7.2.1.2, a pre-infiltration finger was observed during the
tests. To account for this effect, the length of the pre-infiltration finger, \( h_{pi} \), was
subtracted from the total length of the tube \( l \). Therefore, \( z_c = 0 \) always corresponds to
the location of the interface at the onset of full infiltration, and \( l \) corresponds to the
remaining length of travel to the lower end of the tube. Values of \( l \) for the infiltration
tests under consideration are reported in Table 7.2, Table 7.3 and Table 7.4.
Table 7.2. Summary of Kinetics of 4-CT Spontaneous Infiltration Laboratory Experiments Into 2.70 mm Diameter Capillary Tubes of Varying Length

| Test Reference Number | Capillary Tube Total Length, \( l_t \) [mm] | Capillary Tube Reduced Length, \( l = l_t - h_{pi} \) [mm] \(^a\) | Height of DNAPL Pool in Reservoir Tube, \( h_p \) [mm] | Depth of DNAPL Pre-Infiltration, \( h_{pi} \) [mm] | Measured Infiltration Height, \( h_j = h_p + h_{pi} \) [mm] | \( \alpha_t \)-Number \(^b\) | Measured Mid-Depth Velocity (\( z_c = l/2 \)) [mm/s] | Corrected Mid-Depth Velocity (\( z_c = l/2 \)) [mm/s] \(^c\) | Measured 3/4-Depth Velocity (\( z_c = 3/4 \)) [mm/s] \(^d\) | Corrected 3/4-Depth Velocity (\( z_c = 3/4 \)) [mm/s] \(^d\) |
|-----------------------|---------------------------------|----------------|----------------|----------------|-----------------|-----------------|----------------|----------------|----------------|----------------|----------------|
| L9-2.70st-1222mm      | 1222                            | 1202           | 36.1           | 20             | 56.1            | 0.120           | 73.9           | 75.1           | 114.1          | 116.0          |
| L9-2.70st-915mm       | 915                             | 909            | 38.0           | 6              | 44.0            | 0.160           | 75.0           | 75.5           | 113.6          | 114.4          |
| L9-2.70st-610mm       | 610                             | 606            | 35.3           | 4              | 39.3            | 0.240           | 63.3           | 63.7           | 106.8          | 107.5          |
| L9-2.70st-305mm       | 305                             | 301            | 31.5           | 4              | 35.5            | 0.480           | 47.5           | 48.1           | 82.0           | 83.1           |
| L8a-2.70st-130mm      | 130                             | 103            | 40.9           | 27             | 67.9            | 1.125           | 21.1           | 26.6           | 41.3           | 52.2           |
| L8b-2.70st-130mm      | 130                             | 88             | 39.1           | 42             | 81.1            | 1.125           | 22.0           | 32.5           | 38.7           | 57.1           |

\(^a\) The length \( l \) is the reduced flow length of the capillary tube, i.e. after subtracting the pre-infiltration depth to the capillary tube total length, \( l_t \).

\(^b\) \( \alpha_t \)-Number calculated using (3.58).

\(^c\) The corrected mid-depth (or 3/4-depth) velocity is equal to the measured mid-depth (or 3/4-depth) velocity multiplied by the ratio \( l/l_t \).

\(^d\) Theoretical values based on (3.49). The theoretical exit velocity, \( k_\infty \), is equal to 160.9 mm/s for non pre-infiltrated 2.70 mm diam. capillary tubes.
Table 7.3. Summary of Kinetics of 4-CT Spontaneous Infiltration Laboratory Experiments Into 1.33 mm Diameter Capillary Tubes of Varying Length

<table>
<thead>
<tr>
<th>Test Reference Number</th>
<th>Capillary Tube Total Length, ( l_t ) [mm]</th>
<th>Capillary Tube Reduced Length, ( l = l_t - h_{pi} ) [mm]</th>
<th>Height of DNAPL Pool in Reservoir Tube, ( h_p ) [mm]</th>
<th>Depth of DNAPL Pre-Infiltration, ( h_{pi} ) [mm]</th>
<th>Measured Infiltration Height, ( h_i = h_p + h_{pi} ) [mm]</th>
<th>( \alpha_t ) Number</th>
<th>Measured Mid-Depth Velocity (( z_c = \frac{l}{2} )) [mm/s]</th>
<th>Corrected Mid-Depth Velocity (( z_c = \frac{l}{2} )) [mm/s]</th>
<th>Measured 3/4-Depth Velocity (( z_c = \frac{3l}{4} )) [mm/s]</th>
<th>Corrected 3/4-Depth Velocity (( z_c = \frac{3l}{4} )) [mm/s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>L10-1.33lct-918mm</td>
<td>918</td>
<td>912</td>
<td>131.6</td>
<td>6</td>
<td>137.6</td>
<td>9.38 \times 10^{-3}</td>
<td>18.5</td>
<td>18.6</td>
<td>29.4</td>
<td>29.6</td>
</tr>
<tr>
<td>L6-1.33st-610mm</td>
<td>610</td>
<td>610</td>
<td>130.0</td>
<td>Small (not measured)</td>
<td>130.0</td>
<td>1.41 \times 10^{-2}</td>
<td>17.1</td>
<td>17.1</td>
<td>27.6</td>
<td>27.6</td>
</tr>
<tr>
<td>L12-1.33st-305mm</td>
<td>305</td>
<td>265</td>
<td>80.0</td>
<td>40</td>
<td>120.0</td>
<td>2.82 \times 10^{-2}</td>
<td>13.5</td>
<td>15.6</td>
<td>22.4</td>
<td>25.8</td>
</tr>
<tr>
<td>L7b-1.33st-184mm</td>
<td>184</td>
<td>151</td>
<td>112.8</td>
<td>33</td>
<td>145.8</td>
<td>4.68 \times 10^{-2}</td>
<td>7.5</td>
<td>9.1</td>
<td>15.7</td>
<td>19.1</td>
</tr>
<tr>
<td>L7a-1.33st-184mm</td>
<td>184</td>
<td>147</td>
<td>112.0</td>
<td>37</td>
<td>149.0</td>
<td>4.68 \times 10^{-2}</td>
<td>9.5</td>
<td>11.9</td>
<td>17.1</td>
<td>21.4</td>
</tr>
<tr>
<td>L8f-1.33st-122mm</td>
<td>122</td>
<td>104</td>
<td>131.6</td>
<td>18</td>
<td>149.6</td>
<td>7.06 \times 10^{-2}</td>
<td>8.8</td>
<td>10.4</td>
<td>15.1</td>
<td>17.7</td>
</tr>
<tr>
<td>L8b-1.33st-122mm</td>
<td>122</td>
<td>72</td>
<td>105.2</td>
<td>50</td>
<td>155.2</td>
<td>7.06 \times 10^{-2}</td>
<td>2.5</td>
<td>4.2</td>
<td>6.8</td>
<td>11.5</td>
</tr>
</tbody>
</table>

\( ^a \)The length \( l \) is the reduced flow length of the capillary tube, i.e. after subtracting the pre-infiltration depth to the capillary tube total length, \( l_t \).

\( ^b \)\( \alpha_t \)-Number calculated using (3.58).

\( ^c \)The corrected mid-depth (or 3/4-depth) velocity is equal to the measured mid-depth (or 3/4-depth) velocity multiplied by the ratio \( l/l_t \).

\( ^d \)Theoretical values based on (3.49). The theoretical exit velocity, \( k_M \), is equal to 39.0 mm/s for non pre-infiltrated 1.33 mm diameter capillary tubes.
Table 7.4. Summary of Kinetics of 4-CT Spontaneous Infiltration Laboratory Experiment Into a 1201 mm Long, 0.66 mm Diameter Capillary Tube

<table>
<thead>
<tr>
<th>Test Reference Number</th>
<th>Capillary Tube Total Length, $l_t$ [mm]</th>
<th>Capillary Tube Reduced Length, $l = l_t - h_{pi}$ [mm]</th>
<th>Height of DNAPL Pool in Reservoir Tube, $h_p$ [mm]</th>
<th>Depth of DNAPL Pre-Infiltration, $h_{pi}$ [mm]</th>
<th>Measured Infiltration Height, $h_i = h_p + h_{pi}$ [mm]</th>
<th>$\alpha_t$-Number $^b$</th>
<th>Measured Mid-Depth Velocity ($z_c = l/2$) [mm/s]</th>
<th>Corrected 3/4-Depth Velocity ($z_c = 3l/4$) [mm/s]</th>
<th>Measured 3/4-Depth Velocity ($z_c = 3l/4$) [mm/s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>L11-0.66lct-1221mm</td>
<td>1221</td>
<td>1201</td>
<td>288</td>
<td>20</td>
<td>308</td>
<td>$4.28 \times 10^{-4}$</td>
<td>5.0</td>
<td>5.0</td>
<td>7.6</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Theory</td>
<td>274.7 ± 34.3</td>
<td>$4.8^d$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$7.2^d$</td>
</tr>
</tbody>
</table>

$^a$ The length $l$ is the reduced flow length of the capillary tube, i.e. after subtracting the pre-infiltration depth to the capillary tube total length, $l_t$.  

$^b$ $\alpha_t$-Number calculated using (3.58).  

$^c$ The corrected mid-depth (or 3/4-depth) velocity is equal to the measured mid-depth (or 3/4-depth) velocity multiplied by the ratio $l/l$.  

$^d$ Theoretical values based on (3.49). The theoretical exit velocity, $k_{3/4}$, is equal to 9.6 mm/s for non pre-infiltrated 0.66 mm diameter capillary tubes.
Figure 7.10. Interface displacement profiles of 2.70 mm diameter capillary tubes and comparison with the NICCA model predictions: a. Long tubes; b. Short tubes.
(a) Time, $t$ [s] vs. Interface Depth, $z_c$ [mm]

- NICCA model (3.60)
- $l = 912$ mm
- $l = 610$ mm

(b) Time, $t$ [s] vs. Interface Depth, $z_c$ [mm]

- NICCA model (3.60)
- $l = 265$ mm
- $l = 151$ mm
- $l = 147$ mm
Profiles predicted by the NICCA model are also plotted in Figure 7.10, Figure 7.11 and Figure 7.12 using the density properties listed in Table 5.1. The NICCA model equation taking into account the effects of pre-infiltration is given by (3.60) (see Section 3.5.2.5) and is applicable here since the density and viscosity contrast between 4-CT and water are small. The impact of inertia forces as well as the effects of 4-CT reservoir pool drainage are examined in the discussion (see Section 7.2.2.5).

It is important to note that capillary tubes of identical total length \( l \), but differing \( l \) (see, for example, tests series shown in Figure 7.10.b, Figure 7.11.b and Figure 7.11.c) are associated with the same NICCA prediction since (3.60) is dependent on the total length of the capillary tube, \( l_t = l + h_{pi} \). The only difference is the interval over which \( z_c \) varies, i.e. from 0 to \( l \). For example, referring to Figure 7.11.c, tests L8f-1.33st-122mm and L8b-1.33st-122mm are run on the same capillary tube of total length \( l_t = 122 \) mm. The reduced lengths for each test are \( l = 72 \) mm and \( l = 104 \) mm, respectively, because of differing pre-infiltration depths (see Table 7.3). For these tests, the NICCA model (3.60) predicts the same infiltration profile, with \( z_c \) varying from 0 to 72 mm in the former case (solid line in Figure 7.11.c) and 0 to 104 mm in the latter (solid line plus dotted line extension).
Figure 7.12. Interface displacement profile of a 1201 mm long, 0.66 mm diameter capillary tube (test L11-0.66lct-1221mm) and comparison with the NICCA model prediction (prediction assumes $\Delta h = 0.01 h_i$).

It was not possible to estimate $\Delta h = h - h_i$ with any degree of accuracy because the early stage of the spontaneous infiltration experiments typically began with a very slow interface displacement, and because of the variability of the measured infiltration height, $h_i$, between different infiltration tests. However, it was possible to accurately determine the time at which the DNAPL finger reached the lower end of the capillary tube. Thus, the exit time was taken as the point of coincidence between the NICCA model predictions and the measurements. Under these conditions and in order to compare the NICCA model with the experimental data, it must be shown that there exists a small, positive value of $\Delta h = h - h_i$, such that the interface displacement data match the prediction made using the model. Conversely, if there exists no satisfactory value of $\Delta h$, then the NICCA model is not applicable for the test under consideration. It is important to emphasize that the NICCA model’s main assumption is that the capillary resistance $h_i$ remains constant throughout the infiltration process, so that the magnitude of $\Delta h$ remains constant, and negligible in comparison to $z_c$. In light of the results presented in Section 7.2.1, the capillary resistance includes both the effects of pinning force and interfacial tension, i.e. $h_i$ is given by the criterion (7.2).
Results of 2.70 mm and 1.33 mm infiltration tests

To obtain the prediction for the 2.70 mm and 1.33 mm diameter capillary tubes shown in Figure 7.10 and Figure 7.11, an arbitrary value of $\Delta h = 0.01 h_i$ was assumed, where $h_i$ is the theoretical value reported in Table 7.2 and Table 7.3.

As can be seen in both figures, agreement between the NICCA model and the experimental data is good for long capillary tubes. For 2.70 mm diameter capillary tubes, as shown in Figure 7.10.a, agreement is very good at interface depths beyond 400 mm. For 1.33 mm diameter capillary tubes, as shown in Figure 7.11.a, agreement between the model and the experimental data is very good at depth beyond 250 mm.

For shorter tubes and at the early stages of infiltration of longer tubes, however, agreement between the NICCA model and the experimental data is not good. This is particularly obvious in Figure 7.10.b for 2.70 mm diameter capillary tubes, and in Figure 7.11.b and Figure 7.11.c for 1.33 mm diameter capillary tubes. In particular, the observed exit velocity for those tubes, which is given by the slope of the line tangent to the infiltration profile at the exit point, is much less than predicted. Interface velocities are further examined in Section 7.2.2.3.

Also shown in Figure 7.10 and Figure 7.11, is the fact that the onset of infiltration starts at negative times for some capillary tubes. Recall that the measured time when the DNAPL/water interface reaches the lower end of the capillary tube ($z_c = l$) is matched to the time predicted by the NICCA model. Hence, it can be concluded that the experimental spontaneous infiltration takes more time than predicted by the NICCA model, so that the time for DNAPL to exit the tubes as predicted by the NICCA model constitutes a lower bound.

It could be argued that the lack of agreement between the experimental data and the NICCA model prediction is related to picking an arbitrary value of $\Delta h$ equal to 1% of the critical height, $h_i$, and that a lower value of $\Delta h$ would yield better agreement, since the total infiltration time increases with decreasing $\Delta h$. However, recalling Figure 3.7 and the related discussion in Section 3.5.2.2, variations of $\Delta h$ are only significant in the upper part of the infiltration profile and have virtually no impact on the lower part of the profile, which is controlled by $k_\alpha$. Therefore, while a smaller value of $\Delta h$ may give more consistent total infiltration times, it would not affect the shape of the lower part of the infiltration profile for short tubes. Hence, observed profiles in short tubes would differ significantly from the NICCA model prediction even if $\Delta h$ were less than 0.01 $h_i$.

Results of 0.66 mm infiltration test

To obtain the NICCA prediction for the 1201 mm long, 0.66 mm diameter capillary tube infiltration test (test L11-0.66ltc-1221mm) shown in Figure 7.12, a value of $\Delta h = 0.01 h_i$ was assumed, where $h_i$ is the theoretical value reported in Table 7.4. As can be seen in the figure, agreement the NICCA model and the experimental data is very good with the exception of the early stages of the infiltration (down to depth $z_c = 300$ mm), where the experimental data plot above the prediction line (as opposed
to the infiltration tests into 2.70 mm and 1.33 mm diameter capillary tubes, where data plot below the prediction lines). This observation suggests that, for the 0.66 mm diameter capillary tube infiltration test, a larger value of $\Delta h$ would provide a more accurate prediction. This point is well illustrated in Figure 7.13 where a value of $\Delta h = 0.1 h_i$ has been used to obtain the NICCA model prediction of the same infiltration test. As shown in Figure 7.13, the agreement between the experimental data and the prediction is excellent. Nonetheless, given that the theoretical value of $h_i$ is equal to 275 mm, this means that the prediction is obtained using $\Delta h = 27.5$ mm. Clearly, this value of $\Delta h$ is not realistic, as the DNAPL pool height change between increments during the DNAPL pool build-up would never exceed 5 mm-10 mm or so. Hence, it cannot be expected that $\Delta h$ would be larger than a few millimeters, so that the NICCA prediction shown in Figure 7.13 may not be valid.

It will be shown in Section 7.2.2.5 that the difference observed between the experimental data and the NICCA model prediction for the 0.66 mm diameter capillary tube has to do with the assumption made by the NICCA model that the viscosity of 4-CT and that of water are equal. This is not strictly correct as the viscosity of 4-CT has been measured to be equal to 0.918 mPa.s (see Section 5.5.2).

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{interface_displacement_profile.png}
\caption{Interface displacement profile of a 1201 mm long, 0.66 mm diameter capillary tube (test L11-0.66lct-1221mm) and comparison with the NICCA model prediction (prediction assumes $\Delta h = 0.1 h_i$).}
\end{figure}
7.2.2.3 Velocity Fields

Overview

From the results presented in Section 7.2.2.1 and Section 7.2.2.2, it can be concluded that the infiltration profiles are in good agreement with the NICCA model prediction, i.e. (3.60), provided that the diameter of the capillary tube is sufficiently small and if its length sufficiently long. Another way to compare the experimental data and the NICCA model is to examine the interface displacement velocities of each infiltration test reported in Table 7.2, Table 7.3 and Table 7.4. For each test under consideration, the interface velocity experimental data can be plotted versus the interface depth (or relative depth). As defined in Section 3.5.2.2, these types of plot are referred to as velocity fields.

Recall the NICCA model governing equation (3.59) taking into account the effects of pre-infiltration (see Section 3.5.2.5)

\[
\frac{dz_c}{dt} = \frac{k}{l_c} \left[ z_c(t) + \Delta h \right],
\]

(7.3)

where \( l_t = l + h_{pi} \) is the total length of the capillary tube. Multiplying either side of (7.3) by \((l_t/l)\) yields

\[
\frac{l_t}{l} \frac{dz_c}{dt} = \frac{k}{l} \left[ z_c(t) + \Delta h \right].
\]

(7.4)

Again, the NICCA predicts that \( \Delta h \) is negligible with respect to \( z_c \) when \( z_c \) is of the order of \( h_{pi} \). Under these circumstances, (7.4) becomes

\[
\frac{l_t}{l} \frac{dz_c}{dt} = k \frac{z_c(t)}{l}.
\]

(7.5)

Therefore, if the relative depth of infiltration experimental data, \( z_c/l \), are plotted versus the corrected interface displacement velocity data, \((l_t/l)dz_c/dt\), the NICCA model (7.4) predicts that the experimental data should fall on a straight line. Moreover, so long as \( z_c \) is much larger than \( \Delta h \), i.e. past the first few millimeters of infiltration, all experimental corrected velocity data run on a capillary tube of a given diameter should fall on the same line of slope \( k\Delta \), regardless of the tube length.

In order to obtain smoothed interface velocity data from interface depth data as a function of time, a three-point average technique was used. Prior to estimating the slope (i.e. average velocity) \((z_{cj+1} - z_{cj})/(t_{j+1} - t_j)\) between two consecutive depth-time data points \((t_j, z_{cj})\) and \((t_{j+1}, z_{cj+1})\), each data point was adjusted by substituting to a given point \((t_j, z_{cj})\) its average \(((t_j - 1 + t_j + t_{j+1})/3, (z_{cj-1} + z_{cj} + z_{cj+1})/3)\) with the points \((t_{j-1}, z_{cj-1})\) and \((t_{j+1}, z_{cj+1})\) preceding and following the point, respectively. The
operation was repeated a second time if necessary. No averaging was performed if the infiltration data yielded smooth velocity data. An example of three-point averaging is shown in Figure 7.14 where the three-point averaging technique is shown to eliminate the noise particularly visible at the lower end of the capillary tube. The noise is a consequence of the limited resolution of the recording. Because only 32 images per second were acquired during the course of an infiltration test, the position of the interface was located with an associated uncertainty of the order of 1 to 2 hundredths of a second. This uncertainty affects the value of velocity and is larger with increasing magnitude of velocity, since the interface travel time through a fixed interval of length becomes smaller.

\[
(\frac{l}{l}) \frac{dz_c}{dt} \quad [\text{mm/s}]
\]

![Graph showing the effect of three-point averaging technique on the velocity field of the 1202 mm long, 2.70 mm diameter capillary tube infiltration experiment.](image)

**Figure 7.14.** Effect of three-point averaging technique on the velocity field of the 1202 mm long, 2.70 mm diameter capillary tube infiltration experiment.

The interface relative depth versus the corrected interface velocity for each test reported in Table 7.2, Table 7.3 and Table 7.4 are shown in Figure 7.15, Figure 7.16 and Figure 7.17, respectively. For each figure, the line predicted by the NICCA model (7.5) is also plotted for comparison. Note that prediction lines given by (7.5) do not require an estimate of \( \Delta h \). Furthermore, as previously shown in Figure 3.7.b, any
prediction line using (7.4) and a positive value of \( \Delta h \) would fall to the right of the line given by (7.5), such that the (7.5)-line constitutes the lowest possible velocity field predicted by the NICCA model.

Figure 7.15. Interface relative depth versus corrected interface velocity for 2.70 mm diameter capillary tubes of varying length, and comparison with the NICCA model prediction.

Results of 2.70 mm and 1.33 mm infiltration tests

Referring to the experimental data plotted in Figure 7.15 and Figure 7.16, it can be seen that the longest capillary tubes \((l \geq 265 \text{ mm})\) show a quasi-linear relationship between the interface relative depth and the corrected interface velocity. The experimental data of shorter tubes, on the other hand, appear to be non-linear, most particularly at the early stages of the infiltration (i.e. at short relative depth \(z_c/l\)). For a given relative depth, it can be also seen that shorter capillary tubes are associated with smaller corrected interface velocities. This is true for both 2.70 mm and 1.33 mm diameter capillary tubes.

When comparing the experimental data with the NICCA model (7.5), Figure 7.15 and Figure 7.16 show that, for both 2.70 mm and 1.33 mm diameter capillary tubes, the
longest capillary tubes approach the NICCA model prediction. This suggests that, for those tubes, the corrected interface velocity is only a function of the depth relative to the end of the capillary tube, \( z_c/l \), and can be predicted by the NICCA model expression, i.e. \( k_{\Lambda}(z/l) \), with rather good agreement. Nonetheless, all the reported velocity fields appear to fall below the line corresponding to (7.5) with the exception of the test run on the 912 mm long, 1.33 mm diameter capillary tube (see later discussion on viscosity in Section 7.2.2.5).

Thus, overall, the NICCA velocity field (7.5) constitutes an upper-bound of the observed velocity field. As the capillary tubes get shorter, the NICCA model overestimates—sometimes significantly—the measured velocity fields. The contrast is generally largest for the tubes of smallest length, and the velocity field for those tubes is generally non linear, a feature not predicted by the NICCA model. This is true for both diameters currently under discussion.

**Figure 7.16.** Interface relative depth versus corrected interface velocity for 1.33 mm diameter capillary tubes of varying length, and comparison with the NICCA model prediction.
It is important to emphasize, again, that if $\Delta h$ were positive and significant in magnitude with respect to $zc$, then the NICCA prediction line as per (7.4) would be shifted to the right of the line as per (7.5) that is shown in Figure 7.15 and Figure 7.16. Thus, there exists no positive value of $\Delta h$ that can possibly predict the observed velocity fields. That the experimental data plot on the left of the NICCA prediction line (7.5) suggests that $\Delta h$ could, in fact, be a negative number, whose relative importance increases as the length of a capillary tube decreases. If this hypothesis were correct, this would mean that once infiltration takes place, the critical height $h_i$ changes and becomes larger than the pool height at the onset of infiltration, $h$. This is further discussed in Section 7.2.2.5.

![Image of Figure 7.17](image)

**Figure 7.17.** Interface relative depth versus corrected interface velocity for a 1201 mm long, 0.66 mm diameter capillary tube, and comparison with the NICCA model prediction.

**Results of 0.66 mm infiltration test**

Referring now to the 0.66 mm diameter capillary tube velocity field plotted in Figure 7.17, the experimental data show a quasi-linear relationship between the relative depth and the corrected interface velocity.
Overall, good agreement is noted between the experimental data and the NICCA model prediction given by (7.5). It must be noted, however, that (7.5) appears to underestimate the measured velocity field. The difference is of the order of 1 mm/s to 2 mm/s at the most, and seems largest at the lower end of the capillary tube. Again, it will be shown in Section 7.2.2.5 that this observation is attributable to the viscosity difference between 4-CT and water.

![Graph showing the ratio of observed velocity to velocity predicted by the NICCA model as a function of the capillary tube length, $l$.](image)

**Figure 7.18.** Ratio of observed velocity to velocity predicted by the NICCA model as a function of the capillary tube length, $l$.

7.2.2.4 Summary Plot of Interface Velocities

To further investigate the results of the infiltration experiments, interface velocities of reference were chosen to enable comparison of the relative magnitude of observed velocity with the NICCA model predicted velocity. Although, the interface velocity at the exit of the capillary tube would be an adequate velocity of reference, a disadvantage associated with the three-point averaging technique is that, for every set
of $m$ data points, there are only $m - 2$ data points left, such that an estimate of velocity at the lower end would require extrapolation. So, instead of the exit velocity, the capillary tube mid-depth, $z_c/l = 1/2$, and the three-quarter depth, $z_c/l = 3/4$, are used as the locations for velocity comparisons. According to the NICCA model (7.5), the corrected mid-depth velocity and corrected three-quarter depth velocity are expected to be constants independent of length, equal to $k_A/2$, and $3k_A/4$, respectively.

![Graph](image.png)

**Figure 7.19.** Ratio of observed velocity to velocity predicted by the NICCA model as a function of the capillary tube length normalized by its diameter, $l/d$.

For each infiltration experiment, the mid-depth and three-quarter depth measured velocities and corrected velocities are calculated through interpolation between the two nearest data points. The obtained values are reported in Table 7.2, Table 7.3 and Table 7.4 for 2.70 mm, 1.33 mm and 0.66 mm diameter capillary tubes, respectively. A graph of the ratio of the observed velocities to the NICCA model predicted velocity versus the capillary tube length, $l$, is shown in Figure 7.18. Note that the ratio of the corrected velocity to the corrected velocity predicted by the NICCA model is equal to the ratio of the measured velocity to the velocity predicted by the NICCA model.
Overall, the graph shown in Figure 7.18 confirms that velocity fields asymptotically approach the NICCA model velocity fields as the capillary tube length increases (ratio of velocities equal to one in Figure 7.18). It appears that the normalized three-quarter depth velocities plot above the mid-depth velocities, which shows that the velocity field get closer to the NICCA model velocity field as the interface approaches the lower end of the tube. Finally, the normalized velocities associated with 2.70 mm diameter capillary tube reported in Figure 7.18.a plot below the normalized velocities associated with 1.33 mm diameter capillary tubes, indicating that, for similar capillary tube lengths, the tube of smaller diameter tends to have a velocity field closer to the NICCA model velocity field. These observations are confirmed by the 1201 mm long, 0.66 mm diameter capillary tube test, which can be completely predicted by the NICCA model. The fact that smaller diameter tubes yield experimental data that are closer in agreement to the NICCA model than larger diameter tubes led to Figure 7.19. Figure 7.19 plots the ratio of measured to predicted velocity versus the tube length normalized by its diameter, $l/d$. The result is a seemingly unique normalized curve implying that the ratio of measured to observed velocity is solely dependent upon the normalized length of the capillary tube. Passed a ratio $l/d$ of the order of 600, the NICCA model correctly predicts the observed profile.

7.2.2.5 Discussion

It appears from the results presented in Section 7.2.2.1 through Section 7.2.2.4 that agreement between experimental data and the NICCA model is only good asymptotically and if the capillary tubes are long enough. Under these circumstances, the capillary resistance, expressed by the critical height $h_i$, has little impact on the overall infiltration process. As predicted by the NICCA model, the velocity of the interface for these capillary tubes, expressed by (3.61) (see Section 3.5.2.5), is dependent on the gravity and viscosity forces alone. Furthermore, if the effects of pre-infiltration are small with respect to the total length of the capillary tube, the exit velocity (i.e., the velocity at the lower end of the capillary tube) is independent of the capillary tube length and predicted by $k_\Delta$ (see (3.43) in Section 3.5.2.1). Moreover, the interface velocity at depth $z_c$ is proportional to the depth relative to the end of the capillary tube, $z_c/l$. If, on the other hand, the effects on pre-infiltration are important, the exit velocity is equal to $k_\Delta l/l_i$, and is, thus, dependent upon the capillary tube length and depth of pre-infiltration.

Clearly, more infiltration tests in 0.66 mm diameter capillary tubes are needed to further characterize the curve obtained in Figure 7.19, as well as define a more precise value of the ratio $l/d$ below which the NICCA model is no longer valid.

For shorter capillary tubes as well as at the early stages of infiltration into the longer capillary tubes, agreement between the experimental data and the NICCA model prediction is not good.
In order to examine which physical mechanisms may be responsible for the observed differences between the experimental results and the NICCA model predictions, assumptions made in the model development are discussed below under separate headings.

*What is the impact of the viscosity difference between 4-CT and water?*

In Section 3.5.4, a series of model equations (3.76)-(3.79) was developed that took into account the viscosity contrast between DNAPL and water. It was found that under conditions where the viscosity contrast between DNAPL and water was very small with respect to the viscosity of water, then the model approached the NICCA model (see Figure 3.13 and Figure 3.14).

For two of the 4-CT spontaneous infiltration experiments reported in the above sections, namely test L10-1.33lct-918mm \((d = 1.33 \text{ mm} \text{ and } l = 912 \text{ mm})\) and test L11-0.66lct-1221mm \((d = 0.66 \text{ mm} \text{ and } l = 1201 \text{ mm})\), the measured interface velocity in the lower part of the capillary tube was observed to be larger than predicted by the NICCA model. An explanation for this follows.

![Figure 7.20](image)

*Figure 7.20.* Velocity field of 0.66 mm diameter capillary tube of length \(l = 1201 \text{ mm}\) and comparison with model predictions. The solid line is the prediction obtained from the MFDV (3.76) (with \(\Delta h = 0\)), whereas the dashed line is the NICCA model prediction obtained from (3.61).
As 4-CT infiltration into the capillary tube proceeds during the course of the experiment, water is replaced by 4-CT. Given that the viscosity of 4-CT is slightly less than that of water ($\mu_{nw} = 0.918$ mPa.s for 4-CT, see Table 5.3), it can be expected that the capillary tube viscous total resistance decreases as more 4-CT displaces water in the capillary tube. Strictly speaking, under conditions where no pre-infiltration takes place, it should be expected that the interface exit velocity is $\Delta \rho g r_0^2/(8\mu_{nw})$ instead of $k_\Delta = \Delta \rho g r_0^2/(8\mu_w)$ (see Section 3.5.2.2).

![Figure 7.21.](image)

**Figure 7.21.** Velocity field of 1.33 mm diameter capillary tube of length $l = 912$ mm and comparison with model predictions. The solid line is the prediction obtained from the MFDV (3.76) (with $\Delta h = 0$), whereas the dashed line is the NICCA model prediction obtained from (3.61).

The object of this section is to further refine the NICCA model to take into account the 4-CT/water viscosity contrast and compare the refined model’s velocity field prediction with that of the original NICCA model. For each capillary tube diameter under investigation, the infiltration experiment run in the tube of longest length is examined. In the remainder of this thesis, the refined model is referred to as the Model with Fluids of Differing Viscosities (MFDV).
Figure 7.20 shows the observed velocity field of test L11-0.66lct-1221mm ($d = 0.66$ mm and $l = 1201$ mm). The model predictions are also shown in the figure. The solid line is the prediction obtained from the MFDV (3.76), that is the velocity field prediction developed in Section 3.5.4, incorporating the viscosity of 4-CT (it is assumed here that $\Delta h = 0$). The dashed line is the NICCA model prediction obtained from (3.61) (see Section 3.5.2.5). Note in Figure 7.20 that the MFDV adds non-linearity to the predicted relationship between the interface velocity, $dz_c/dt$, and the interface depth, $z_c$.

![Graph showing velocity field comparison](image)

**Figure 7.22.** Velocity field of 2.70 mm diameter capillary tube of length $l = 1202$ mm and comparison with model predictions. The solid line is the prediction obtained from the MFDV (3.76) (with $\Delta h = 0$), whereas the dashed line is the NICCA model prediction obtained from (3.61).

Clearly, for this infiltration test, the velocity field is extremely well captured by the MFDV, most particularly in the lower region of the capillary tube. This suggests that the flow velocity was under-predicted by the NICCA model because the effects of viscosity changes were not properly taken into account by NICCA.
Figure 7.21 shows the observed velocity field of test L10-1.33lct-918mm \((d = 1.33 \text{ mm} \text{ and } l = 912 \text{ mm})\). Again, the model predictions are also shown in the figure. The solid line is the prediction obtained from (3.76) (with \(\Delta h = 0\)), and the dashed line is the NICCA model prediction obtained from (3.61).

### Table 7.5. Summary of Measured and Predicted Mid-Depth and Three-Quarter Depth Velocities for the Spontaneous Infiltration Experiments

<table>
<thead>
<tr>
<th>Test Reference Number</th>
<th>Capillary Tube Reduced Length, (l) [mm]</th>
<th>Measured Interface Velocity [mm/s] (^a)</th>
<th>NICCA Model Predicted Velocity [mm/s] (^b)</th>
<th>MFDV Predicted Velocity [mm/s] (^c)</th>
</tr>
</thead>
<tbody>
<tr>
<td>L9-2.70st-1222mm</td>
<td>1202</td>
<td>73.9</td>
<td>79.1</td>
<td>82.6</td>
</tr>
<tr>
<td>L9-2.70st-915mm</td>
<td>909</td>
<td>114.1</td>
<td>118.7</td>
<td>126.5</td>
</tr>
<tr>
<td>L9-2.70st-610mm</td>
<td>606</td>
<td>75.0</td>
<td>79.9</td>
<td>83.3</td>
</tr>
<tr>
<td>L9-2.70st-305mm</td>
<td>301</td>
<td>113.6</td>
<td>119.9</td>
<td>127.7</td>
</tr>
<tr>
<td>L9-2.70st-130mm</td>
<td>103</td>
<td>63.3</td>
<td>79.9</td>
<td>83.3</td>
</tr>
<tr>
<td>L8a-2.70st-130mm</td>
<td>41.3</td>
<td>106.8</td>
<td>119.9</td>
<td>127.7</td>
</tr>
<tr>
<td>L8b-2.70st-130mm</td>
<td>88</td>
<td>82.0</td>
<td>119.1</td>
<td>126.9</td>
</tr>
<tr>
<td>L10-1.33lct-918mm</td>
<td>912</td>
<td>21.1</td>
<td>63.7</td>
<td>67.0</td>
</tr>
<tr>
<td>L6-1.33st-610mm</td>
<td>103</td>
<td>41.3</td>
<td>95.6</td>
<td>102.3</td>
</tr>
<tr>
<td>L12-1.33st-305mm</td>
<td>88</td>
<td>22.0</td>
<td>54.4</td>
<td>57.6</td>
</tr>
<tr>
<td>L7b-1.33st-184mm</td>
<td>151</td>
<td>13.5</td>
<td>17.0</td>
<td>17.8</td>
</tr>
<tr>
<td>L7a-1.33st-184mm</td>
<td>147</td>
<td>7.5</td>
<td>16.0</td>
<td>16.8</td>
</tr>
<tr>
<td>L8f-1.33st-122mm</td>
<td>151</td>
<td>9.5</td>
<td>15.6</td>
<td>16.4</td>
</tr>
<tr>
<td>L8b-1.33st-122mm</td>
<td>147</td>
<td>17.1</td>
<td>23.4</td>
<td>25.0</td>
</tr>
<tr>
<td>L11-0.66lct-1221mm</td>
<td>1201</td>
<td>8.8</td>
<td>16.6</td>
<td>17.5</td>
</tr>
<tr>
<td></td>
<td>7.6</td>
<td>15.1</td>
<td>25.0</td>
<td>26.7</td>
</tr>
</tbody>
</table>

\(^a\) The measured velocities are also reported in Table 7.2, Table 7.3 and Table 7.4.

\(^b\) Velocity calculated using (3.61).

\(^c\) Velocity calculated using (3.76).
As shown in the figure, the prediction from the model (3.76) overestimates the observed velocity field throughout the entire infiltration process by about 2 mm/s, but, unlike the NICCA model, shows a constant offset with the observed velocity field throughout the entire infiltration process. Clearly, for this infiltration experiment, there exists a small extra resisting force improperly taken into account by the two models: it is this force that reduces the velocity of the interface.

Finally, Figure 7.22 shows the observed velocity field of test L9-2.70st-1222mm ($d = 2.70$ mm and $l = 1202$ mm). Again, the solid line is the prediction obtained from (3.76) (with $\Delta h = 0$), and the dashed line is the NICCA model prediction obtained from (3.61).

As shown in the figure, while the NICCA model prediction offers a reasonably good prediction, the prediction from the MFDV (3.76) overestimates the observed velocity field throughout the entire length of the capillary tube. In the upper half of the tube, there appears to be a constant offset of the order of 5 mm/s between the measured velocity field and that predicted by the MFDV. There seems to be a transition zone at a depth of 550 mm, corresponding to an interface velocity of 70 mm/s. Passed this zone, the difference between the measured field and the MFDV-predicted field increases to about 10 mm/s-20 mm/s. This difference is largest at the lower end of the tube.

These observations, again, suggest that for this infiltration experiment, as for that in the 1.33 mm diameter capillary tube, that there must be a small extra resisting force, not accounted for by either model, that contributes to reduce the velocity of the interface. Consequently, it can be concluded that, for this infiltration test, the extra resisting force and decrease of the resisting viscosity force during the infiltration process, which are both unaccounted for by the NICCA model, are comparable, so that, overall, the NICCA model prediction appears correct.

In light of these observations, it is important to ensure that the trends shown in Figure 7.19 are similar, if instead of the NICCA model, the MFDV (3.76) is used to predict the interface mid-depth and three-quarter depth velocities. Table 7.5 summarizes the mid-depth and three-quarter depth interface velocities measured for each spontaneous infiltration test reported in Table 7.2, Table 7.3 and Table 7.4. In addition, the table provides the interface velocities predicted by both the NICCA model and the MFDV. It can be seen that the values obtained using the MFDV are typically 5% larger than those obtained from the NICCA model.

Figure 7.23 shows a graph equivalent to that shown in Figure 7.19. For each infiltration test reported in Table 7.5, the measured velocity normalized by the velocity predicted the MFDV is plotted versus the capillary tube length normalized by its diameter. Clearly, the trend shown in Figure 7.23 is similar to that shown in Figure 7.19. In particular, it can be seen that, for a given infiltration depth, the normalized three-quarter depth velocities generally plot above the mid-depth velocities, which shows that the velocity field get closer to the MFDV velocity field as the interface approaches the lower end of the tube. Figure 7.23 also suggests that, for longer capillary tubes, the MFDV provides a correct interface velocity prediction.
Figure 7.23. Ratio of observed velocity to velocity predicted by the MFDV as a function of the capillary tube length normalized by its diameter, $l/d$.

Is the assumption of constant DNAPL reservoir head correct?

One seemingly obvious reason that could contribute to interface displacements slower than predicted by the NICCA model is that the DNAPL reservoir tube pool height, $h_p$, does not remain constant throughout the infiltration process, but is slowly drained into the capillary tube, so that the reservoir head is less than assumed by the NICCA model, which assumes the head does not change (see Section 3.4.1). This drainage effect could be especially true for capillary tubes of larger diameter. Nonetheless, the experiments were designed to keep this effect small. Indeed, the reservoir tube radius, $r_r$, was always selected such that the reservoir cross-sectional area, $\pi r_r^2$, was much larger than the capillary tube cross-sectional area, $\pi r_0^2$.

The object of this section is to further address the effects of the DNAPL pool drainage, and show that, under the condition where the ratio $r_0^2/r_r^2$ is negligible compared to unity, the drainage has little effect on the infiltration profile or the velocity field.
Recall the NICCA model governing equation (3.59) where the effects of pre-infiltration are accounted for (see Section 3.5.2.5)

$$\frac{dz_c}{dt} = \frac{k_\Delta}{l + h_{pi}} [z_c(t) + h_p + h_{pi} - h_i]. \quad (7.6)$$

It is assumed that the reservoir tube 4-CT pool height decreases as a consequence of draining into the capillary tube. Hence, at time $t$, mass conservation leads to

$$\pi r_r^2 h_p(0) - \pi r_r^2 h_p(t) = \pi r_0^2 z_c(t). \quad (7.7)$$

Substituting (7.7) into (7.6) gives

$$\frac{dz_c}{dt} = \frac{k_\Delta}{l + h_{pi}} \left[ z_c(t) \left(1 - \frac{r_0^2}{r_r^2}\right) + h_p(0) + h_{pi} - h_i \right]. \quad (7.8)$$
As shown by (7.8), the larger the ratio of tube radii, \( r_0/r_r \), the slower the interface displacement velocity. Conversely, if \( r_0/r_r \) approaches zero, so that \( r_0^2/r_r^2 \) is negligible compared to unity, (7.8) is reduced to (7.6). Equation (7.8) can be solved analytically. Taking the initial condition \( z_c = 0 \) at \( t = 0 \) leads to

\[
z_c(t) = \frac{\Delta h}{\left(1 - \frac{r_0^2}{r_r^2}\right)} \left[-1 + \exp \left[\frac{k_h}{l + h_{pi}} \left(1 - \frac{r_0^2}{r_r^2}\right) t\right]\right],
\]

(7.9)

where \( \Delta h = h_p(0) + h_{pi} - h_i \).

![Figure 7.25. Velocity field of 1.33 mm diameter capillary tube of length \( l = 151 \) mm and comparison with NICCA model predictions. The solid line is the prediction obtained from (3.61) (constant 4-CT pool head), whereas the dashed line is the prediction obtained from (7.8) and taking \( \Delta h = h_p(0) + h_{pi} - h_i = 0 \) (draining 4-CT pool).](image)

Consider, as an illustrative example, test L7b-1.33st-184mm (see Table 7.3 and Figure 7.11.b) run on a 1.33 mm diameter capillary tube (\( l = 151 \) mm) with a reservoir tube of radius \( r_r = 4 \) mm (see Section C.2.3). The infiltration profile and velocity field.
for this test are plotted in Figure 7.24 and Figure 7.25, respectively, along with the NICCA model prediction assuming a constant 4-CT pool head (i.e. (3.60) and (3.61)) and that assuming a draining 4-CT pool as modeled above (i.e. (7.8) and (7.9)). As can be seen in the figures, the effect of the pool head decrease has little impact on the NICCA model prediction, so that \( h_p \) can be regarded as constant in time for practical purposes. Similar results are obtained when considering the other tests reported in Table 7.2, Table 7.3 and Table 7.4. Therefore, the effects of pool draining alone cannot explain why the interface displacement is slower than predicted by the NICCA model.

Note that, strictly speaking, Figure 7.23 shown above should take into account the effects of reservoir drainage, in addition to those of viscosity change, in order to provide the most accurate velocity prediction. Indeed, as can be seen in Figure 7.25, the mid-depth and three-quarter depth velocities predicted by the NICCA model are slightly larger than those predicted by the NICCA model that includes the effects of reservoir drainage. Nonetheless, use of both viscosity change and reservoir drainage would yield little change in the trends shown in both Figure 7.19 and Figure 7.23. For this reason a plot including both of these effects will not presented here.

*Are inertia forces always negligible?*

It is important to discuss the role played by inertia forces. The \( \alpha_t \)-numbers listed in Table 7.2, Table 7.3 and Table 7.4 are calculated using the equation defining \( \alpha_t \), i.e. (3.58), derived in Section 3.5.2.5. The numbers reported in these tables show that inertia forces are not negligible for 2.70 mm diameter capillary tubes, while they can be considered negligible for 1.33 mm and 0.66 mm diameter capillary tubes. It is important to note that, while the \( \alpha_t \)-number can tell whether or not the magnitude of inertia forces is expected to be important, it does not provide a quantitative estimate of the inertia forces themselves or the velocity reduction associated with these inertia forces. In other words, based on the magnitude of \( \alpha_t \), one would expect a negligible effect of inertia forces on 1.33 mm and 0.66 mm diameter capillary tubes, and a measurable effect on 2.70 mm diameter capillary tubes.

Figure 7.26 shows a plot of the ratio of observed interface velocity to velocity predicted by the NICCA model versus the \( \alpha_t \)-number. Clearly, the fact that the NICCA model over-predicts velocity cannot be exclusively attributed to inertia forces because this trend is observed for both 1.33 mm and 2.70 mm diameter capillary tubes, where the \( \alpha_t \)-numbers characterizing the magnitude of the inertia forces are an order of magnitude apart.

Recall that the \( \beta_{rl} \)-number given by (3.63) is always a fraction of \( \alpha_t \) such that entry drag forces are always negligible if inertia forces are negligible (see Section 3.5.2.5).
\( \alpha_t \)-Number \( (\alpha_t = \Delta \rho \rho_w g r_0^4 / (16 \mu_w^2 l_t)) \)

**Figure 7.26.** Ratio of observed velocity to velocity predicted by the NICCA model as a function of the \( \alpha_t \)-number, which characterizes the magnitude of inertia forces relative to the viscous forces.

That inertia forces are negligible for 1.33 mm diameter capillary tubes and of measurable effect for 2.70 mm diameter capillary tubes suggests three possible hypotheses for the fact that the NICCA model over-predicts observed velocity data:

1. The curve shown in Figure 7.23 (or Figure 7.19) is not unique, and the apparently unique behavior is coincidental. For 2.70 mm diameter capillary tubes, local and convective inertia forces contribute to reduce the measured velocity. For 1.33 mm diameter capillary tubes, there exists another resisting force, ignored in the NICCA model, and which contributes to the observed velocity reduction with decreasing tube length.

2. Alternatively, the extra resisting force exists for both capillary tube diameters, but its magnitude is less for 2.70 mm diameter capillary tubes than for 1.33 mm diameter capillary tubes. Combination of the resisting inertia force and extra resisting force for the 2.70 mm diameter capillary tube appears to have the same magnitude as the extra resisting force of the 1.33 mm diameter capillary tube.
3. The curve shown in Figure 7.23 (or Figure 7.19) is unique, and inertia forces contribute little, if any, to the reduction in velocity, while another resisting force contributes to most of the velocity ratio reduction for both 1.33 mm and 2.70 mm diameter capillary tubes. This resisting force would then also exist for short 0.66 mm diameter capillary tubes.

Clearly, it is difficult to conclude at this point, which hypothesis is correct. Most likely, the effects of inertia are indeed present for 2.70 mm diameter capillary tubes, but they are not solely responsible for the observed velocity reduction. In other words, the second hypothesis is most likely correct. This point will be demonstrated in Section 7.2.3.4. Furthermore, in light of the centrifuge infiltration test results, it will be shown in Section 7.3.6.3 that the curve shown in Figure 7.23 is not unique.

*Is the modeling of capillary forces correct?*

Whether or not the magnitude of inertia forces is important, there appears to exist another resisting force, not accounted for by the NICCA model, that acts to slow down the DNAPL/water interface displacement. The most straightforward explanation for the disagreement between NICCA and the experimental data is that the effects of capillary forces are not properly accounted for in the NICCA model. Indeed, as pointed out in Section 7.2.2.2, the model’s main assumption is that the capillary resistance, described in the model by the critical height $h_c$, remains constant throughout the infiltration process, so that the magnitude of $\Delta h$ remains constant, and negligible in comparison to $z_c$. In other words, the NICCA model assumes that the magnitude of the resisting capillary force combining the effects of pinning, $n_{fp}$, and interfacial tension, $\sigma \cos \theta_s$ (see (7.2)), is a constant independent of interface velocity, such that the dynamic capillary force is always equal to the static capillary force. The two forces contributing to the capillary force are considered under two separate headings below.

*Interfacial tension forces*

As discussed in Section 2.3.5 and Section 2.3.6, under drainage conditions, the dynamic contact angle of the moving meniscus is expected to decrease with increasing interface velocity. Similar to the contact angle feedback mechanism shown in Figure 7.4 (see Section 7.2.1.2), as the contact angle decreases, the critical height (7.2) required for DNAPL infiltration is increased, and thus cannot be expected to remain constant as assumed by the NICCA model. In Section 7.2.1.2, it has been shown that the static contact angle at the start of the infiltration process ranges between 30° and 60°. Hence, it can be expected that the contact angle decrease even further and reach perfect wetting during the course of the infiltration. If the contact angle does, in fact, decrease with velocity, the resulting increase in critical height associated with this decrease would correspond to an increase in the capillary resistance that would act to slow down the interface displacement velocity.
Considering the NICCA model governing equation (3.59) (see Section 3.5.2.5, or (7.8) above), the increase in critical height \( h_i \) is qualitatively consistent with the observation that \( \Delta h \) can assume negative values, as reported in Section 7.2.2.3 during the examination of the velocity fields of infiltration tests into 2.70 mm and 1.33 mm diameter capillary tubes (see Figure 7.15 and Figure 7.16).

Under the hypothesis that the infiltration tension forces increase with velocity, the region where \( |\Delta h| \) is not negligible with respect to \( z_c \) can be referred to as the \textit{capillarity-controlled region}. For short capillary tubes, the length of the capillarity-controlled region is believed to be significant in comparison to \( l \). Hence, the observed exit velocity will still be controlled by capillary forces, and will be less than that predicted by the NICCA model, i.e., \( k_\alpha l / l_i \). For longer capillary tubes, on the other hand, the interface is believed to reach a depth \( z_c \), where the maximum absolute value of \( \Delta h \), equal to \( |h - h_d(\theta_s = 0)| \) becomes negligible when compared to \( z_c \). Past this depth, the effects of the capillary force become small, so that flow is controlled by gravity and viscosity alone, as discussed at the beginning of Section 7.2.2.5. For this condition, the observed exit velocity will approach \( k_\alpha l / l_i \). The lower region of the capillary tube where \( z_c >> |\Delta h| \) can be referred to as the \textit{viscosity-controlled region}.

While the mechanism of interfacial tension force increase is qualitatively consistent with the observed infiltration behavior, it remains to be shown that a decrease in contact angle with velocity is in quantitative agreement with the experimental data. This is the object of Section 7.2.3 where the NIVCA model developed in Section 3.5.3 is compared with the experimental results.

\textbf{Pinning forces}

Evidence that pinning forces are present when the DNAPL/water interface is not moving was given in Section 7.2.1. There is also an indication that pinning forces are still present when the interface is in motion. This indication is based on the fact that if, indeed, the pinning forces were simply localized forces and collapsed as soon as the interface was in motion, then a sudden decrease of the critical height, \( h_i \), would be measured, and positive values of \( \Delta h \) would be back-calculated from the experimental velocity field.

Most likely, and in view of prior research work on liquid/liquid displacements [e.g., Calvo et al., 1991; Van Remoortere and Joos, 1993b] (see Section 2.3.6.4), pinning forces are still present during the interface motion, most particularly at small capillary numbers. Indeed, at small displacement velocities, it can be hypothesized that DNAPL displaces water present on the wall of the capillary—although a thin water film might remain adsorbed onto the glass surface, as the glass is preferentially water wetting. During this slow drainage process, the moving DNAPL/water contact line—i.e., the contour of the interface meniscus that intersects the capillary tube wall—can be visualized as \textit{bumping into} every pinning point present on the glass surface. Hence, an overall resisting pinning force may still be present and contribute to a stick-slip mechanism, as discussed elsewhere [Kistler, 1993].
For larger capillary numbers, on the other hand, it can be hypothesized, that the interface displacement velocity is large enough that the DNAPL/water interface could be \textit{sliding on water}. That is, as a consequence of the rapid interface motion, water remains trapped underneath the DNAPL finger to form a film of thickness much less than the capillary tube diameter but significantly thicker than the characteristic dimensions of the pinning points, so that the DNAPL/water interface meniscus is no longer in contact with the capillary tube walls. Under these circumstances, it can be hypothesized that the pinning forces and, thus, the capillary resistance are decreased.

Clearly, it is not possible to conclude at this stage whether the mechanism proposed above is correct. Even if the pinning forces decrease with velocity, it may be difficult to infer this trend if there is a simultaneous increase of capillary resistance associated with the increase of interfacial tension forces due to the decrease in $\theta_d$.

\textit{Relevance of the NICCA model}

That the NICCA model over-predicts interface velocities below a ratio $l/d$ of about 600 does not mean that this model is not useful in practice. For some real fracture systems, it can be expected that the length of the fracture will be of the order of one meter or more, and that the aperture will not exceed one millimeter, such that $l/d$ will—largely—exceed 600. For those systems, the NICCA model applies and can provide a good estimate of the time to complete infiltration. Furthermore, if the viscosity contrast between DNAPL and water is large, the MFDV can be used to provide a more accurate prediction of the infiltration time. For fracture systems where $l/d$ is less than 600, the NICCA model provides a lower bond of the time required for complete infiltration. More generally, if other physical phenomena are present (see Section 2.2.1.5 and Section 2.2.2), such as diffusion/adsorption onto the solid matrix or other retardation mechanisms, natural attenuation phenomena through spontaneous biodegradation, or dissolution into the aqueous phase, the time to complete infiltration of the fracture system will be increased. Hence, again, the NICCA model provides a lower bond of the infiltration time, i.e. the smallest possible time to complete infiltration of the fracture system. Under conditions where the DNAPL pool volume is comparable, or smaller than, the fracture system volume, such that the pool is draining and its size significantly shrinking with time, the NICCA model can be modified to include the effects of head change using the approach described in (7.6)-(7.9).

In Section 7.2.3.5, the criterion $l/d > 600$ for which the NICCA model is applicable will be generalized. A theoretical criterion will be derived that is valid for any DNAPL/water system ($l/d > 600$ has only been verified for 4-CT as a DNAPL). This new criterion—function of the DNAPL properties—will provide a quick way of checking whether or not the NICCA model can be used for predicting the DNAPL infiltration kinetics.
7.2.3 Kinetics of Infiltration: Results of DNAPL/Water Interface Displacement Experiments and Comparison With the NIVCA Model

7.2.3.1 Introduction

In this section, the kinetics of the 4-CT infiltration experiments presented in Section 7.2.2 are reexamined and compared to the prediction of the NIVCA model previously introduced in Section 3.5.3. In Section 7.2.3.2, the NIVCA model is revisited to account for the new infiltration criterion developed in Section 7.2.1.2. In Section 7.2.3.3, results of the infiltration experiments are compared to the NIVCA model prediction and discussed. Section 7.2.3.4 reexamines the summary plots obtained in Section 7.2.2.4 and shows how these plots can be successfully predicted by the NIVCA model. A theoretical criterion for which the NIVCA model reduces to the NICCA model is derived in Section 7.2.3.5. Section 7.2.3.6 examines the experimental results in term of the dynamic infiltration height corresponding to the resisting force missing in the NICCA model, and verifies that the results are consistent with those obtained in Section 7.2.3.3. Conclusions are given in Section 7.2.3.7.

7.2.3.2 NIVCA Model Revisited

In Section 7.2.1.2, a new infiltration criterion (7.2) was established that incorporated the effects of interface pinning. This new criterion led to little change in the NICCA model because this model assumes that $h_i$ remains constant throughout the infiltration process. However, the new infiltration criterion does require reevaluation of the equations of the NIVCA model developed in Section 3.5.3. As will be shown below, this reevaluation will lead to a modified definition of the retardation factor, $\gamma_t$, which is affected by the existence of pinning forces.

Recall (3.71) (see Section 3.5.3.4), developed for the case where pre-infiltration exists down to a depth of $h_{pi}$

$$\frac{dz_c}{dt} = \frac{k_s}{l + h_{pi}} \left[ z_c(t) + h - \frac{2\sigma \cos \theta_d (dz_c / dt)}{\Delta \rho g r_0} \right], \quad (7.10)$$

where $l + h_{pi}$ is equal to the total length of the capillary tube, $l$, and the function $\cos \theta_d (dz_c / dt)$ is given by (3.64) (see Section 3.5.3.2), i.e.

$$\cos \theta_d (dz_c / dt) = \begin{cases} \cos \theta_s + (1 - \cos \theta_s) \frac{dz_c}{u_c} & \text{if } dz_c / dt \leq u_c, \\ 1 & \text{if } dz_c / dt > u_c. \end{cases} \quad (7.11)$$
In (7.10), the term $2\sigma \cos \theta_d (dz_c/\,dt)/(\Delta\rho g r_0)$ is proportional to the dynamic capillary pressure at the DNAPL/water interface (see (3.36) in Section 3.4.5). This term can be seen as a velocity dependent infiltration height $h_i (dz_c/\,dt)$, or dynamic infiltration height, as defined per analogy with the former infiltration criterion (3.4). Thus, (7.10) can be rewritten as

$$\frac{dz_c}{dt} = \frac{k_A}{l + h_{pi}} \left[ z_c(t) + h - h_i (dz_c/\,dt) \right]. \quad (7.12)$$

Using the new infiltration criterion (7.2), (7.12) can be rewritten as

$$\frac{dz_c}{dt} = \frac{k_A}{l + h_{pi}} \left[ z_c(t) + h - \frac{2\sigma \cos \theta_d (dz_c/\,dt) - 2n_{pf} f_p (dz_c/\,dt)}{\Delta\rho g r_0} \right], \quad (7.13)$$

where $n_{pf} (dz_c/\,dt)$ is the dynamic pinning force, and $\sigma \cos \theta_d$ the dynamic interfacial tension force ($\cos \theta_d$ is given by (7.11)).

It is assumed, as a first approximation, that $n_{pf}$ is a constant independent of velocity. Substituting (7.11) in (7.13) and rearranging terms, as was done in Section 3.5.3, leads to

$$\frac{dz_c}{dt} = \frac{k_A}{l + h_{pi}} \left[ z_c(t) + \Delta h \right] \quad \text{for} \quad dz_c/\,dt \leq u_c, \quad (7.14)$$

and

$$\frac{dz_c}{dt} = \gamma_i (l + h_{pi}) \left[ z_c(t) + \Delta h' \right] \quad \text{for} \quad dz_c/\,dt > u_c, \quad (7.15)$$

where $\Delta h$, $\Delta h'$ and $\gamma_i$ are defined by

$$\Delta h = h - h_i = h - \frac{2\sigma \cos \theta_d}{\Delta\rho g r_0} - \frac{2n_{pf} f_p}{\Delta\rho g r_0}, \quad (7.16)$$

$$\Delta h' = h - h_i (\theta_s = 0) = h - \frac{2\sigma}{\Delta\rho g r_0} - \frac{2n_{pf} f_p}{\Delta\rho g r_0}, \quad (7.17)$$

and

$$\gamma_i = 1 + \frac{2\sigma}{\Delta\rho g r_0} \frac{1 - \cos \theta_s}{l + h_{pi}} \frac{k_A}{u_c}, \quad (7.18)$$
respectively. Note that the retardation factor $\gamma_t$, given by (7.18), differs from that defined by (3.73), which does not take into account the presence of pinning forces.

7.2.3.3 Results of 4-CT Spontaneous and Controlled-Head Infiltration Experiments and Comparison with the NIVCA Model Prediction

Estimation of the NIVCA parameters

In the system of equations (7.14)-(7.18), there are three unknowns to be determined, namely, the static contact angle, $\theta_s$, the perfect wetting velocity, $u_c$, and the pinning force, $n_{pfp}$. As discussed in Section 7.2.1.2, the static contact angle, $\theta_s$, can be expected to vary between infiltration experiments, and is expected to decrease as the pre-infiltration height increases. The perfect wetting velocity, $u_c$, may also vary as a function of the static contact angle and the capillary tube diameter. It is assumed here that the perfect wetting velocity is unique, independent of the static contact angle or the capillary tube diameter. Finally, as discussed in Section 7.2.1.2, the magnitude of the pinning forces may vary between experiments, even for a given capillary tube diameter or a given capillary tube.

As a first approximation, the value of the dynamic pinning force, $n_{pfp}$, is taken to be a constant for each tube diameter. This constant is equal to the static pinning force back-calculated from the infiltration measurements (see Section 7.2.1.2). That is, for infiltration tests into 2.70 mm diameter capillary tubes, $n_{pfp} = 0.005 \text{ N/m}$. For spontaneous infiltration tests into 1.33 mm diameter capillary tubes, $n_{pfp} = 0.010 \text{ N/m}$. Finally, for the spontaneous infiltration test into a 0.66 mm diameter capillary tube, $n_{pfp} = 0.012 \text{ N/m}$.

Next, the perfect wetting velocity, $u_c$, needs to be estimated. The perfect wetting velocity is the velocity for which the contact angle reaches its minimum value of $0^\circ$. Under conditions where $dz_c/dt \geq u_c$, the difference $h - h(dz_c/dt)$ is constant, independent of the infiltration velocity. Referring to the velocity field of spontaneous infiltration tests into 1.33 mm diameter capillary tubes shown in Figure 7.16, $u_c$ can be seen as the value for which the plot of velocity versus interface relative depth becomes linear. An average value of $u_c = 3 \text{ mm/s}$ appears to be generally in good agreement with the experimental data, with the exception of the infiltration test L8f-1.33st-122mm ($l = 104 \text{ mm}$), for which $u_c$ is closer to 8 mm/s. The specific reason why this test yields a larger value for $u_c$ is, at present, not clearly understood, although the data in general for $l = 104 \text{ mm}$ are not well behaved. For the sake of uniformity, the value of $u_c = 3 \text{ mm/s}$ is also taken as the perfect wetting velocity of infiltration tests into other capillary tube diameters.

The last parameter to be determined from the experimental data is the static contact angle, $\theta_s$. Using the infiltration criterion (7.2) leads to
\[ \cos \theta_s = \frac{\Delta \rho g r_0 h_i - 2n_p f_p}{2\sigma}. \]  

(7.19)

Hence, the static contact angle can be obtained by substituting in (7.19) the value of the pinning force and the measured value of \( h_i \) for the test under consideration.

In what follows, the interfacial tension of 4-CT with water is taken equal to 0.032 N/m as reported in Chapter 5. The viscosity of both 4-CT and water is taken equal to \( 10^{-3} \) Pa.s, as the NIVCA model assumes that both liquids have equal viscosities. The impacts of this assumption are discussed below in light of the test results and model comparison.

**Table 7.6.** Summary of NIVCA Parameter Values for 4-CT Infiltration Laboratory Experiments Into 1.33 mm Diameter Capillary Tubes

<table>
<thead>
<tr>
<th>Test Reference Number</th>
<th>Capillary Tube Reduced Length, ( l ) [mm]</th>
<th>Measured DNAPL Infiltration Height, ( h_i ) [mm] (^a)</th>
<th>Depth of DNAPL Pre-Infiltration Height, ( h_{pi} ) [mm] (^a)</th>
<th>Back-Calculated Static Contact Angle, ( \theta_s ) (^b)</th>
<th>Retardation Factor, ( \gamma_t ) (^c)</th>
</tr>
</thead>
<tbody>
<tr>
<td>L10-1.33lct-918mm</td>
<td>912</td>
<td>137.6</td>
<td>6</td>
<td>45.8°</td>
<td>1.585</td>
</tr>
<tr>
<td>L6-1.33st-610mm</td>
<td>610</td>
<td>130.0</td>
<td>Small (not measured)</td>
<td>50.1°</td>
<td>2.043</td>
</tr>
<tr>
<td>L12-1.33st-305mm</td>
<td>265</td>
<td>120.0</td>
<td>40</td>
<td>55.4°</td>
<td>3.512</td>
</tr>
<tr>
<td>L7b-1.33st-184mm</td>
<td>151</td>
<td>145.8</td>
<td>33</td>
<td>40.8°</td>
<td>3.340</td>
</tr>
<tr>
<td>L7a-1.33st-184mm</td>
<td>147</td>
<td>149.0</td>
<td>37</td>
<td>38.7°</td>
<td>3.114</td>
</tr>
<tr>
<td>L8f-1.33st-122mm</td>
<td>104</td>
<td>149.6</td>
<td>37</td>
<td>38.3°</td>
<td>4.124</td>
</tr>
<tr>
<td>L8b-1.33st-122mm</td>
<td>72</td>
<td>155.2</td>
<td>50</td>
<td>34.3°</td>
<td>3.526</td>
</tr>
<tr>
<td>Lch-1.33st-305mm (^d)</td>
<td>305</td>
<td>136.3 (^e)</td>
<td>None</td>
<td>46.6°</td>
<td>2.817</td>
</tr>
</tbody>
</table>

\(^a\) These values are also reported in Table 7.3.

\(^b\) Computed from (7.19), using a pinning force value, \( n_{pf} \), equal to 0.010 N/m unless otherwise noted.

\(^c\) Computed from (7.18), using a perfect wetting velocity value, \( u_c \), equal to 3 mm/s.

\(^d\) Parameters for all six controlled-head infiltration tests.

\(^e\) This value is the theoretical value reported in Table 7.1 as the infiltration height were not measured during the controlled-head infiltration tests.

\(^f\) Computed using a pinning force value, \( n_{pf} \), equal to 0.015 N/m.
Infiltration tests into 1.33 mm diameter capillary tubes

The method described above is applied to the series of spontaneous infiltration tests into 1.33 mm diameter capillary tubes reported in Table 7.3 (see also experimental data in Section C.2.3). In addition, six controlled-head infiltration experiments are also examined. Experimental data for these tests are reported in Appendix C, Section C.4. All controlled-head tests were run in the same capillary tube of length 305 mm, for which $\alpha = 2.82 \times 10^{-2}$ (see (3.46) in Section 3.5.2.1), so that inertia forces were negligible. The experimental methodology is described in Section 6.2.3. Note that, for controlled-head infiltration tests, there is no DNAPL pre-infiltration as the flow valve is closed during the build-up of the DNAPL pool.

Table 7.6 summarizes the parameter values used for obtaining the prediction of the NIVCA model (7.14)-(7.18), i.e. the back-calculated static contact angle, $\theta_s$, and retardation factor, $\gamma_t$, assuming a pinning force, $n_{pf,p}$, of 0.010 N/m and a perfect wetting velocity, $u_c$, of 3 mm/s.

Spontaneous infiltration tests into 1.33 mm diameter capillary tubes

Figure 7.27 through Figure 7.33 show the infiltration profile and velocity field of each spontaneous infiltration test reported in Table 7.3 (and thus Table 7.6). In all the figures, the symbols represent the experimental data points. The solid lines are the NIVCA model predictions calculated using the equation series (7.14)-(7.18) and the parameter values given in Table 7.6. For comparison, the NICCA model predictions from the series of equations (3.59)-(3.61) (see Section 3.5.2.5) are also plotted (dotted lines).

Referring to the velocity fields shown in every (b)-figure, the experimental data points are obtained after using a three-point averaging technique identical to that described in Section 7.2.2.3. The NIVCA velocity field prediction is obtained from the two equations (7.14) and (7.15). For calculating the interface velocity, $dz_c/dt$, using (7.14), it is necessary to have an estimate of $\Delta h$ (see (7.16)). It was assumed that the spontaneous infiltration took place for $h \approx h_i$, such that $\Delta h \approx 0$. For calculating $dz_c/dt$ using (7.15), it is necessary to have an estimate of $\Delta h'$ (see (7.17)). In $\Delta h'$, under the assumption $h \approx h_i$, the value of $h$ was taken as the infiltration height, $h_i$, measured for the given infiltration test under consideration (see Table 7.6). For the NICCA velocity field prediction, (3.61) was used, that is the NICCA smallest velocity field (i.e. the field for which, again, $\Delta h = 0$).

Referring now to the infiltration profiles shown in every (a)-figure, the experimental data points are translated along the time axis to match the NIVCA model exit time, as was done in Section 7.2.2.2 for the NICCA model. The NIVCA model prediction is obtained after numerical integration of the two governing equations (7.14) and (7.15) to obtain the interface depth, $z_c$, as a function of time. For the NIVCA prediction, it was necessary to assume a small positive value of $\Delta h$. Indeed, unlike the velocity field prediction, the value of $\Delta h$ cannot be taken equal to 0. In all the infiltration profiles shown here, it was assumed that $\Delta h = 0.01 h_i$, where $h_i$ is the
infiltration height measured for the spontaneous test under consideration (see Table 7.6). Similar to the velocity fields, for the infiltration profile prediction, the value of $h$ in $\Delta h'$ was taken equal to the measured infiltration height, $h_i$.

The NICCA model prediction shown in the infiltration profiles is that obtained in Section 7.2.2.2 (i.e. with $\Delta h = 0.01 h_i$), and after translating along the time axis to match the exit time of the NIVCA model predicted infiltration profile. For this reason, the NICCA infiltration time does not start at $t = 0$.

Referring to Figure 7.27 through Figure 7.33, it can be seen that, overall, both the infiltration profile and velocity fields are well predicted by the NIVCA model, which obviously appears to be a significant improvement to the NICCA model. Nevertheless, there are two notable exceptions: test L12-1.33st-305mm ($l = 265$ mm) and test L8f-1.33st-122mm ($l = 104$ mm) shown in Figure 7.29 and Figure 7.32, respectively.

For test L8f-1.33st-122mm, the measured velocity field in the upper half of the capillary tube (see Figure 7.32.b) is larger than that predicted by the NIVCA model, while less that predicted by the NICCA model. It has already been noted above that the perfect wetting velocity for this test as estimated from Figure 7.16 appeared to be of the order of $8$ mm/s. This can also be seen in Figure 7.32.b. Furthermore, Figure 7.32.b suggests that the interface velocity may be larger than 0 at the early stages of infiltration (about 2 mm/s or so, if the measured velocity field is extrapolated), implying that the DNAPL pool may have been built up beyond the infiltration height $h_i$.

For test L12-1.33st-305mm (see Figure 7.29), the measured velocity field is observed to be larger than that predicted by the NIVCA model throughout the capillary tube, while less that predicted by the NICCA model. For this experiment, it is suspected that the 4-CT/water interfacial tension of the liquid pair may have been lower than that of other infiltration experiments, possibly due to 4-CT contamination prior to usage. Indeed, the infiltration height measured during this test was quite low, equal to 120.0 mm, despite 40 mm of pre-infiltration. Also, note how the data point corresponding to this test stands out as lying below the rest of the data on the plot of infiltration height versus pre-infiltration shown in Figure 7.6.

Referring to tests L10-1.33st-918mm and L6-1.33st-610mm infiltration tests shown in Figure 7.27 and Figure 7.28, respectively, the NIVCA model appears to provide a very good prediction of both the infiltration profile and velocity field, and improves the prediction made by the NICCA model in the upper half of the two capillary tubes. It can be seen, however, that the measured velocity field close to the exit of the capillary tube is more than that predicted by the NIVCA model. This disagreement is attributable to the viscosity difference between 4-CT and water. As already discussed in Section 7.2.2.5, during the course of the capillary tube infiltration process, 4-CT displaces water, such that the liquid column becomes less and less viscous. As the interface reaches the lower end of the capillary tube, 4-CT entirely replaces water in the capillary tube, and the resisting viscous force of the NIVCA model can be expected to over-estimate the real viscous resistance. For 4-CT infiltrating a 1.33 mm diameter capillary tube, $k_\Delta = \Delta \rho g r_0^2 / 8 \mu_{nw} = 39.0$ mm/s (see (3.43) in Section 3.5.2.1). Taking $\mu_{nw} = 0.918$ Pa.s, the quantity $\Delta \rho g r_0^2 / 8 \mu_{nw}$ is found to be equal to 42.5 mm/s which is about 9% larger than $k_\Delta$. Examining the NIVCA
model equation (7.15), the exit velocity, $dz_c/dt(z_c = l)$, is proportional to $k_\Delta$. If $k_\Delta$ is replaced by $\Delta \rho g r_0^2/8\mu_{nw}$, the calculated exit velocity is found to increase by 9%, a prediction in agreement with the observed exit velocities shown in Figure 7.27.b and Figure 7.28.b.

Referring now to tests L7b-1.33st-184mm and L7a-1.33st-184mm shown in Figure 7.30 and Figure 7.31, respectively, very good agreement is noted between the experimental data and the NIVCA model prediction for both the infiltration profile and the velocity field. Most particularly, it must be noted that the NIVCA model provides a very large improvement in comparison to the NICCA prediction. It should be expected, however, based on the viscosity decrease phenomenon outlined above, that the two measured velocity fields in the lower part of the capillary tubes be larger than that predicted by the NIVCA model (see Figure 7.30.b and Figure 7.31.b). Two hypotheses could explain the trend shown here:

1. Extra resistance attributable to further capillary resistance for this particular test series, for example, if the pinning force is larger in magnitude than the 0.010 N/m assumed here.
2. Existence of a secondary resisting effect, of magnitude significantly smaller than the capillary resistance modeled by the NIVCA model, which further contributes to reduce the interface velocity.

This issue is further discussed in Section 7.2.3.6 when examining the dynamic infiltration height.

Referring now to test L8b-1.33st-122mm ($l = 72$mm) shown in Figure 7.33, it can be seen that the NIVCA model predictions of the experimental infiltration profile and velocity field are much closer than that of the NICCA model. Nonetheless, the observed velocity field shown in Figure 7.33.b is less than predicted by the NIVCA model, suggesting again that there may exist an extra resisting force unaccounted for by the NIVCA model or that the capillary resistance is larger.

Despite the differences between predictions and observed data reported above, it is important to recognize that, overall, the NIVCA model is an improvement to the NICCA model. It should also be recognized that the NIVCA model provides a prediction of the infiltration profile and velocity field without any fitting parameter, unlike other models proposed in the literature (see Section 2.3.5 and Section 2.3.6). Based solely upon one new parameter—namely, the pinning force—that is estimated from the infiltration height measurements and unique for a given capillary tube diameter, the NIVCA model is able to provide a reasonable prediction of the DNAPL infiltration kinetics and quantitatively justify the velocity reduction associated with the increase of dynamic capillary resistance.

The results of spontaneous infiltration tests into 1.33 mm diameter capillary tubes will be further discussed in Section 7.2.3.4 and Section 7.2.3.6 in light of the results of the controlled-head infiltration tests and the spontaneous infiltration tests into 0.66 mm and 2.70 mm diameter tubes.
Figure 7.27. Spontaneous infiltration experiment into a 1.33 mm diameter capillary tube of length \( l = 912 \) mm. The solid lines are the predictions obtained from the NIVCA model (7.14)-(7.18), whereas the dotted lines are the prediction obtained from the NICCA model (3.59)-(3.61): a. Interface displacement profile; b. Velocity field.

Figure 7.28. Spontaneous infiltration experiment into a 1.33 mm diameter capillary tube of length \( l = 610 \) mm. The solid lines are the predictions obtained from the NIVCA model (7.14)-(7.18), whereas the dotted lines are the prediction obtained from the NICCA model (3.59)-(3.61): a. Interface displacement profile; b. Velocity field.
Figure 7.29. Spontaneous infiltration experiment into a 1.33 mm diameter capillary tube of length \( l = 265 \) mm. The solid lines are the predictions obtained from the NIVCA model (7.14)-(7.18), whereas the dotted lines are the prediction obtained from the NICCA model (3.59)-(3.61): a. Interface displacement profile; b. Velocity field.

Figure 7.30. Spontaneous infiltration experiment into a 1.33 mm diameter capillary tube of length \( l = 151 \) mm. The solid lines are the predictions obtained from the NIVCA model (7.14)-(7.18), whereas the dotted lines are the prediction obtained from the NICCA model (3.59)-(3.61): a. Interface displacement profile; b. Velocity field.
Figure 7.31. Spontaneous infiltration experiment into a 1.33 mm diameter capillary tube of length \( l = 147 \text{ mm} \). The solid lines are the predictions obtained from the NIVCA model (7.14)-(7.18), whereas the dotted lines are the prediction obtained from the NICCA model (3.59)-(3.61): a. Interface displacement profile; b. Velocity field.

Figure 7.32. Spontaneous infiltration experiment into a 1.33 mm diameter capillary tube of length \( l = 104 \text{ mm} \). The solid lines are the predictions obtained from the NIVCA model (7.14)-(7.18), whereas the dotted lines are the prediction obtained from the NICCA model (3.59)-(3.61): a. Interface displacement profile; b. Velocity field.
Figure 7.33. Spontaneous infiltration experiment into a 1.33 mm diameter capillary tube of length $l = 72$ mm. The solid lines are the predictions obtained from the NIVCA model (7.14)-(7.18), whereas the dotted lines are the prediction obtained from the NICCA model (3.59)-(3.61): a. Interface displacement profile; b. Velocity field.

**Controlled-head infiltration tests into 1.33 mm diameter capillary tubes**

Figure 7.34 shows the infiltration profile and velocity field of the six 4-CT controlled-head infiltration tests reported in Section C.4. The parameter, $h$, listed in the figure is the pool height set prior to turning the flow valve on, and is obviously larger than the infiltration height, $h_i$, for a 1.33 mm diameter capillary tube. Again, for the controlled-head infiltration tests into 1.33 mm diameter capillary tubes, it is assumed that $n_{dp}$ is equal to 0.010 N/m and $u_c$ is equal to 3 mm/s. The corresponding value of $\theta_s$ and $\gamma_t$ are reported in Table 7.6.

It is important to note that, for a controlled-head infiltration test, the value of the infiltration height, $h_i$, is unknown, since it is not measured. Hence, it is not possible to back-calculate the value of $\theta_s$ and $\gamma_t$ unless $h_i$ is estimated. Thus, it was assumed that the infiltration height for all the controlled-head infiltration experiments was equal to the theoretical infiltration height reported in Table 7.1, i.e. $h_i = 136.3$ mm.

For the infiltration profiles shown in Figure 7.34.a, the NIVCA model prediction is obtained after integration of the governing equations (7.14) and (7.15) to obtain $z_c(t)$. Again, for the NIVCA prediction, it is necessary to calculate the two heights $\Delta h$ and $\Delta h'$. Contrary to spontaneous infiltration tests, the value of the pool height, $h$, is known, while the value of the infiltration height, $h_i$, for a given infiltration test is unknown, as discussed above. Thus, while the value of $\Delta h'$ (see (7.17)) can directly be
calculated, the value of $\Delta h$ (see (7.16)) also requires an estimation of the infiltration height. Again, it was assumed that the infiltration height was equal to 136.3 mm.

The NICCA model prediction is also shown in Figure 7.34.a, for comparison with the NIVCA model. The NICCA model prediction is obtained using (3.47) (see Section 3.5.2.1). Again, $h_i$ is taken equal to 136.3 mm for calculating $\Delta h = h - h_i$ (see (3.44)). Similar to the spontaneous infiltration experiments, the NICCA model prediction is translated along the time axis to match the exit time of the infiltration profile predicted by the NIVCA model, and thus starts at a time larger than 0. The experimental data are also translated along the time axis to match the exit time of the NIVCA model prediction.

For the velocity fields shown in Figure 7.34.b, the experimental velocity field was obtained after using a three-point averaging technique similar to that described in Section 7.2.2.3. Velocity fields predicted by the NIVCA and NICCA models are also shown for comparison. For better readability of the figure, the minimum and maximum predicted velocity fields are the only fields plotted in the figure, and correspond to the velocity fields associated with the controlled-head infiltration tests having the smallest ($h = 150.9$ mm) and the largest ($h = 178.1$ mm) 4-CT pool, respectively. The two solid lines are the velocity fields predicted by the NIVCA governing equations (7.14) and (7.15) for the controlled-head infiltration tests $h = 150.9$ mm and $h = 178.1$ mm, where $h_i = 136.3$ mm is assumed. Two of the dotted lines are the velocity fields predicted by the NICCA governing equation (3.48) for the controlled-head infiltration tests $h = 150.9$ mm and $h = 178.1$ mm, where again $h_i = 136.3$ mm is assumed. The last dotted line is not a test prediction, and is only shown for comparison. This line corresponds to the minimum velocity field predicted by the NICCA model, i.e. that obtained using (3.49), for which $\Delta h = 0$.

Figure 7.34.a shows that the infiltration time increases with decreasing pool height. Moreover, as shown in Figure 7.34.b, the velocity field decreases with decreasing pool height. Both these trends are expected since the driving gravity force decreases with decreasing pool height. Furthermore, for all tests plotted in Figure 7.34.b, there appears to be a linear relationship between the interface velocity and the infiltration depth as suggested by both the NICCA and NIVCA model (in the perfect wetting region), with a slope equal to both those of the NICCA model and the perfect wetting portion of the NIVCA model (see (7.15)), i.e. a slope of $k\Delta t/l$.

The NIVCA model prediction of the infiltration kinetics is much closer to the experimental data than the NICCA model prediction. Still, at a given depth of infiltration, the velocity predicted by NIVCA is higher than that observed. Consequently, as illustrated in Figure 7.34.a, the total infiltration time appears to take longer than predicted by the NIVCA model.
Figure 7.34. Controlled-head infiltration experiments into a 1.33 mm diameter capillary tube of length $l = 305$ mm. The solid lines are the predictions obtained from the NIVCA model (7.14)-(7.18) with $\eta_{fp} = 0.010$ N/m and $u_c = 3$ mm/s, whereas the dotted lines are the prediction obtained from the NICCA model (3.47)-(3.49): a. Interface displacement profile; b. Velocity field.
Figure 7.35. Controlled-head infiltration experiments into a 1.33 mm diameter capillary tube of length \( l = 305 \) mm. The solid lines are the predictions obtained from the NIVCA model (7.14)-(7.18) with \( n_{\eta f_p} = 0.015 \) N/m and \( u_c = 3 \) mm/s, whereas the dotted lines are the prediction obtained from the NICCA model (3.47)-(3.49): a. Interface displacement profile; b. Velocity field.
The difference between the experimental measurements and the NIVCA model prediction cannot be attributed to the choice of \( h_i \). Indeed, the offset in Figure 7.34.b between the experimental data and the prediction band of the NICCA model (i.e. the region between the NIVCA prediction \( h = 150.9 \) mm and the prediction \( h = 178.1 \) mm) is dependent upon \( \Delta h' = h - 2\sigma / (\Delta \rho g r_0) - 2n_{pfp}/(\Delta \rho g r_0) \) (see (7.17)). This might suggest that for this set of controlled-head infiltration experiments, the pinning force is larger in magnitude than the assumed 0.010 N/m.

To examine the effects of the pinning force on the NIVCA model, a graph similar to that shown in Figure 7.34 is provided in Figure 7.35, except that the magnitude of the pinning force assumed for the NIVCA model prediction is changed. While the perfect wetting velocity, \( u_c \), remains equal to 3 mm/s, the pinning force, \( n_{pfp} \), for obtaining the new prediction is now taken equal to 0.015 N/m. The corresponding values of \( \theta_s \) and \( \gamma_t \) (under the same assumption that \( h_i = 136.3 \) mm) are shown in Table 7.6.

As can be seen in Figure 7.34, the NIVCA model prediction, with the new parameters, offers a much better prediction of the infiltration experiments. Yet, the value of the pinning force selected here lacks theoretical justification.

An alternative explanation to a higher pinning force is that there exists another resisting force not taken into account by the NIVCA model that may contribute to further velocity reduction. For example, for controlled-head infiltration tests, it is possible that a pressure redistribution in the liquid column operates when the flow valve is turned on, leading to an extra pressure drop in the system that is not taken into account by the NIVCA model.

Clearly, it is difficult to determine for these tests which mechanism is present. Obviously, further experimental work would be necessary to define this mechanism and draw concrete conclusions. Again, it must be emphasized that this does not rule out the usefulness of the NIVCA model. Part of the resistance to flow is likely attributable to the increase in capillary resistance as 4-CT displaces water. What remains uncertain is the magnitude of the pinning force, and whether or not other mechanisms are present. The controlled-head infiltration tests are further discussed in Section 7.2.3.6 where the dynamic infiltration height-interface velocity relationship is examined.

**Infiltration tests into 2.70 mm diameter capillary tubes**

The series of spontaneous infiltration tests into 2.70 mm diameter capillary tubes reported in Table 7.2 is now examined (see also experimental data in Section C.2.1) and compared to the NIVCA model predictions. Table 7.7 summarizes the parameter values used for obtaining the prediction of the NIVCA model (7.14)-(7.18), i.e. the back-calculated static contact angle, \( \theta_s \), and retardation factor, \( \gamma_t \), using a perfect wetting velocity, \( u_c \), of 3 mm/s and a pinning force, \( n_{pfp} \), of 0.005 N/m—obtained from the measurements of infiltration heights into 2.70 mm diameter tubes.

Figure 7.36 through Figure 7.40 show the infiltration profile and velocity field of each spontaneous infiltration test reported in Table 7.2 (and thus Table 7.7) with the
exception of L8b-2.70st-130mm where the pinning force, $n_{\text{pfp}}$, of 0.005 N/m is found to be incompatible with the large observed infiltration height of 81.1 mm. Indeed, if $n_{\text{pfp}}$ and $h_i$ values are used to estimate $\Delta h'$ (see (7.17)), this parameter takes a positive value, incompatible with the NIVCA model. This suggests that the pinning force, $n_{\text{pfp}}$, may be larger than the assumed 0.005 N/m for this particular test. This point is illustrated in Section 7.2.3.6.

### Table 7.7. Summary of NIVCA Parameter Values for 4-CT Spontaneous Infiltration Laboratory Experiments Into 2.70 mm Diameter Capillary Tubes

<table>
<thead>
<tr>
<th>Test Reference Number</th>
<th>Capillary Tube Reduced Length, $l$ [mm]</th>
<th>Measured DNAPL Infiltration Height, $h_i$ [mm]</th>
<th>Depth of DNAPL Pre-Infiltration Height, $h_{\text{pi}}$ [mm]</th>
<th>Back-Calculated Static Contact Angle, $\theta_s$</th>
<th>Retardation Factor, $\gamma_f$</th>
</tr>
</thead>
<tbody>
<tr>
<td>L9-2.70st-1222mm</td>
<td>1202</td>
<td>56.1</td>
<td>20</td>
<td>47.2°</td>
<td>1.945</td>
</tr>
<tr>
<td>L9-2.70st-915mm</td>
<td>909</td>
<td>44.0</td>
<td>6</td>
<td>60.1°</td>
<td>2.971</td>
</tr>
<tr>
<td>L9-2.70st-610mm</td>
<td>606</td>
<td>39.3</td>
<td>4</td>
<td>64.6°</td>
<td>4.391</td>
</tr>
<tr>
<td>L9-2.70st-305mm</td>
<td>301</td>
<td>35.5</td>
<td>4</td>
<td>68.1°</td>
<td>8.407</td>
</tr>
<tr>
<td>L8a-2.70st-130mm</td>
<td>103</td>
<td>67.9</td>
<td>27</td>
<td>31.2°</td>
<td>5.013</td>
</tr>
<tr>
<td>L8b-2.70st-130mm</td>
<td>88</td>
<td>81.1</td>
<td>42</td>
<td>N/A</td>
<td>N/A</td>
</tr>
</tbody>
</table>

*These values are also reported in Table 7.2.

*b Computed from (7.19), using a pinning force value, $n_{\text{pfp}}$, equal to 0.005 N/m.

*c Computed from (7.18), using a perfect wetting velocity value, $u_e$, equal to 3 mm/s.

In Figure 7.36 through Figure 7.40, the solid line is the NIVCA model prediction calculated using the equation series (7.14)-(7.18) and the parameter values given in Table 7.7. For comparison, the NICCA model prediction from the series of equations (3.59)-(3.61) (see Section 3.5.2.5) is also plotted (dotted line). The method used to obtain the velocity fields and infiltration profiles shown in the figures is identical to that described above for the spontaneous infiltration tests into the 1.33 mm diameter capillary tubes.

Referring to Figure 7.36 through Figure 7.40, it can be seen that overall, the NIVCA model largely improves the prediction made by the NICCA model and that good agreement can be noted between the experimental data and the NIVCA model prediction for both the infiltration profile and the velocity field. Nonetheless, for the infiltration experiment run on the tubes of shortest lengths (e.g., Figure 7.39 and Figure...
agreement is poor. In fact, it is observed that the disagreement between experimental data and NIVCA model increases as the length of the capillary tube decreases.

Referring to the velocity field and infiltration profile of test L9-2.70st-1222mm \((l = 1202 \text{ mm})\), shown in Figure 7.36, it can be seen that overall, the NIVCA model and the NICCA model do not give very different predictions. Overall, agreement between the experimental data and both predictions is good. Indeed, the experimental velocity field is close to the velocity field predicted by both models (see Figure 7.36.b), with the exception of the lower end of the capillary tube where the experimental velocity field is 10 mm/s to 15 mm/s less than that predicted by the models, corresponding to a relative difference of the order of 10%.

In Figure 7.36.b, the velocity field predicted by the MFDV is also plotted for comparison (dashed line). This prediction is identical to that shown in Figure 7.22. Again, the MFDV velocity field is obtained by using (3.76) (see Section 3.5.4), and taking \(h \approx h_i\), i.e. \(\Delta h \approx 0\), corresponding to the smallest velocity field predicted by the MFDV. As already discussed in Section 7.2.2.5, the experimental data fall below the MFDV prediction, especially in the lower half of the capillary tube, indicating that an extra resisting force may be present in addition to the capillary force. The extra-resisting force is most likely attributable to the effects of inertia, which are not incorporated in any of the models examined above (MFDV, NICCA or NIVCA). Indeed, for this infiltration test, the parameter \(\alpha_t\) representing the magnitude of inertia forces is equal to 0.120 (see Table 7.2), implying that the inertia forces cannot be completely neglected even if they are still somewhat small in comparison to the viscous forces.

Referring now to the infiltration tests L9-2.70st-915mm and L9-2.70st-610mm shown in Figure 7.37 and Figure 7.38, respectively, it can be seen that, unlike test L9-2.70st-1222mm, there exists a measurable difference between the NIVCA and NICCA model predictions. Overall, the NIVCA model predictions are in very good agreement with the experimental data, and offer a good improvement to the NICCA model predictions throughout the entire capillary tube. Nonetheless, the NIVCA model does not account for the changes in viscous force associated with the infiltration process. Hence, similar to the conclusion made above on test L9-2.70st-1222mm (see Figure 7.36.b), the experimental velocity fields of tests L9-2.70st-915mm and L9-2.70st-610mm in the lower part of the capillary tubes should be larger than those predicted by the NIVCA model (see Figure 7.37.b and Figure 7.38.b). Referring to the earlier discussion on the effects of viscous force changes in the spontaneous infiltration tests into 1.33 mm diameter tubes, it can be similarly proven for 2.70 mm diameter tubes that the measured exit velocities should be of the order of 9% larger than those predicted by the NIVCA model. Again, this suggests the presence of an extra resisting force. Given the values of the \(\alpha_t\) parameters, equal to 0.160 and 0.240 for \(l = 909 \text{ mm}\) and \(l = 606 \text{ mm}\), respectively (see Table 7.2), the missing resisting force is likely to be inertia.

As the capillary tube length decreases, the magnitude of the inertia forces increases, implying that the prediction offered by the NIVCA model should over-
estimate the experimental velocity profile. This trend is very well shown in Figure 7.39.b and Figure 7.40.b. In both figures, the presence of the pinning force and the increase of capillary resistance with velocity incorporated by the NIVCA model can partly explain why the experimental velocity profile is smaller than initially predicted by the NICCA model. Nonetheless, in both cases, the experimental velocity profile is less than the NIVCA velocity profile. The difference is largest for the shortest capillary tube (test with \( l = 103 \) mm shown in Figure 7.40.b) associated with the largest \( \alpha_t \)-number (\( \alpha_t = 1.125 \)). For test L9-2.70st-305mm (see Figure 7.39.b) where the difference between experimental velocity field and NIVCA velocity field is smaller than for test L8a-2.70st-130mm, the \( \alpha_t \)-number is equal to 0.480 (see Table 7.2). Clearly, these effects can be attributed to inertia forces.

Note that, for all spontaneous infiltration tests into 2.70 mm diameter tubes reported above, other effects could also explain why the experimental velocity field is smaller than that predicted by the NIVCA model. For example, the pinning force could be larger than the assumed 0.005 N/m. Alternatively, a resisting effect other than inertia, capillarity or viscosity could be present. Nonetheless, that the resistance increases with decreasing tube length strongly suggests that the force missing in the NIVCA model is inertia.

![Figure 7.36. Spontaneous infiltration experiment into a 2.70 mm diameter capillary tube of length \( l = 1202 \) mm. The solid lines are the predictions obtained from the NIVCA model (7.14)-(7.18). The dotted lines and dashed lines are the prediction obtained from the NICCA model (3.59)-(3.61) and MFVD (3.76), respectively (with \( \Delta h = 0 \)): a. Interface displacement profile; b. Velocity field.](image)
Figure 7.37. Spontaneous infiltration experiment into a 2.70 mm diameter capillary tube of length $l = 909$ mm. The solid lines are the predictions obtained from the NIVCA model (7.14)-(7.18), whereas the dotted lines are the prediction obtained from the NICCA model (3.59)-(3.61): a. Interface displacement profile; b. Velocity field.

Figure 7.38. Spontaneous infiltration experiment into a 2.70 mm diameter capillary tube of length $l = 606$ mm. The solid lines are the predictions obtained from the NIVCA model (7.14)-(7.18), whereas the dotted lines are the prediction obtained from the NICCA model (3.59)-(3.61): a. Interface displacement profile; b. Velocity field.
Figure 7.39. Spontaneous infiltration experiment into a 2.70 mm diameter capillary tube of length $l = 301$ mm. The solid lines are the predictions obtained from the NIVCA model (7.14)-(7.18), whereas the dotted lines are the prediction obtained from the NICCA model (3.59)-(3.61): a. Interface displacement profile; b. Velocity field.

Figure 7.40. Spontaneous infiltration experiment into a 2.70 mm diameter capillary tube of length $l = 103$ mm. The solid lines are the predictions obtained from the NIVCA model (7.14)-(7.18), whereas the dotted lines are the prediction obtained from the NICCA model (3.59)-(3.61): a. Interface displacement profile; b. Velocity field.
In conclusion, for spontaneous infiltration tests into 2.70 mm diameter capillary tubes, the presence of the pinning force and the decrease in contact angle act to increase the capillary resistance to the interface displacement. The pinning force and variable dynamic contact angle are both incorporated in the NIVCA model, which provides a good qualitative justification to capillary resistance and its effect on the interface velocity. Nevertheless, the NIVCA model cannot completely predict the experimental velocity field, most particularly for shorter capillary tubes, suggesting that another resisting mechanism is present. For 2.70 mm diameter capillary tubes, the magnitude of inertia forces characterized by the $\alpha_t$-number is significant in comparison to other resisting forces. Hence, it is believed that inertia forces may explain the observed difference between measurements and NIVCA prediction. This hypothesis is further justified by the fact that shorter capillary tubes for which the $\alpha_t$-number is larger are associated with the largest difference between measurement and prediction.

Infiltration tests into 0.66 mm diameter capillary tubes

The spontaneous infiltration test into the 1201 mm long, 0.66 mm diameter capillary tube (test L11-0.66lct-1221mm) reported in Table 7.4 is now examined (see also experimental data in Section C.2.4) and compared to the NIVCA model prediction. Table 7.8 provides the parameter values used for obtaining the prediction of the NIVCA model (7.14)-(7.18), i.e. the back-calculated static contact angle, $\theta_s$, and retardation factor, $\gamma_t$, using a pinning force, $n_pfp$, of 0.012 N/m and a perfect wetting velocity, $u_c$, of 3 mm/s.

Table 7.8. Summary of NIVCA Parameter Values for the 4-CT Infiltration Spontaneous Laboratory Experiment Into a 1201 mm Long, 0.66 mm Diameter Capillary Tube

<table>
<thead>
<tr>
<th>Test Reference Number</th>
<th>Capillary Tube Reduced Length, $l$ [mm]</th>
<th>Measured DNAPL Infiltration Height, $h_i$ [mm] a</th>
<th>Depth of DNAPL Pre-Infiltration Height, $h_{pi}$ [mm] a</th>
<th>Back-Calculated Static Contact Angle, $\theta_s$</th>
<th>Retardation Factor, $\gamma_t$ d</th>
</tr>
</thead>
<tbody>
<tr>
<td>L11-0.66lct-1221mm</td>
<td>1201</td>
<td>308</td>
<td>20</td>
<td>41.7° b</td>
<td>1.183</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>21.0° c</td>
<td>1.048</td>
</tr>
</tbody>
</table>

a These values are also reported in Table 7.4.
b Computed from (7.19), using a pinning force value, $n_pfp$, equal to 0.012 N/m.
c Computed from (7.19), using a pinning force value, $n_pfp$, equal to 0.006 N/m.
d Computed from (7.18), using a perfect wetting velocity value, $u_c$, equal to 3 mm/s.

Figure 7.41 shows the infiltration profile and velocity field of test L11-0.66lct-1221mm. The solid line is the NIVCA model prediction calculated using the equation series (7.14)-(7.18) and the parameter values given in Table 7.8. For comparison, the
NICCA model prediction from the series of equations (3.59)-(3.61) (see Section 3.5.2.5) is also plotted (dotted line). The method used to obtain the velocity field and infiltration profile shown in the figure is identical to that described earlier for the spontaneous infiltration tests into the 1.33 mm diameter capillary tubes. The velocity field predicted by the MFDV (3.76) (see Section 3.5.4) with $\Delta h = 0$ is also plotted in Figure 7.41.b (dashed line).

Overall, it can be concluded, that the NIVCA model does not offer a very good prediction of the interface displacement and velocity field. In fact, the NIVCA model largely under-estimates the measured velocities. In contrast, as already pointed out in Section 7.2.2.5, the MFDV offers a much better prediction of the observed velocity field.

![Figure 7.41](image)

**Figure 7.41.** Spontaneous infiltration experiment into a 0.66 mm diameter capillary tube of length $l = 1201$ mm. The solid lines are the predictions obtained from the NIVCA model (7.14)-(7.18) with $n_{fp} = 0.012$ N/m and $u_c = 3$ mm/s. The dotted lines and dashed lines are the prediction obtained from the NICCA model (3.59)-(3.61) and MFDV (3.76), respectively (with $\Delta h = 0$): a. Interface displacement profile; b. Velocity field.

One possible explanation is that the pinning force of 0.012 N/m used to obtain the NIVCA model prediction is higher than it should be. Recall that the value of 0.012 N/m was based on only one estimate of the DNAPL pool height to first pre-infiltration (see Section 7.2.1.2). A graph similar to that shown in Figure 7.41 is provided in Figure 7.42, except that the magnitude of the pinning force used for the NIVCA model prediction is changed. While the perfect wetting velocity, $u_c$, remains
equal to 3 mm/s, the pinning force $n_{\rho f_p}$ for obtaining the new prediction is now taken equal to 0.006 N/m. The corresponding values of $\theta_s$ and $\gamma_t$ are given in Table 7.8.

As shown in Figure 7.42, the prediction offered by the NICCA and the NIVCA models are not very different in that case. The reason has to do with the back-calculated retardation factor, $\gamma_t$, which become very close to unity (see Table 7.8). As discussed in Section 7.2.2.5, the difference between the experimental velocity field and the field predicted by both the NICCA and NIVCA models has to do with the viscous force reduction that takes place during the 4-CT infiltration process. For that reason, the MFDV ultimately provides the best prediction.

![Figure 7.42](image_url)

**Figure 7.42.** Spontaneous infiltration experiment into a 0.66 mm diameter capillary tube of length $l = 1201$ mm. The solid lines are the predictions obtained from the NIVCA model (7.14)-(7.18) with $n_{\rho f_p} = 0.006$ N/m and $u_c = 3$ mm/s. The dotted lines and dashed lines are the prediction obtained from the NICCA model (3.59)-(3.61) and MFVD (3.76), respectively (with $\Delta h = 0$): a. Interface displacement profile; b. Velocity field.

While a smaller pinning force could explain why the NIVCA model with $n_{\rho f_p} = 0.012$ N/m provides a poor agreement with the experimental data, it remains uncertain why 0.66 mm diameter capillary tubes should have a pinning force lower than that of 1.33 mm diameter capillary tubes, yet marginally larger than that of 2.70 mm diameter capillary tubes.

An alternative to the hypothesis of a reduced pinning force is that the perfect wetting velocity, $u_c$, is not equal to 3 mm/s for 0.66 mm diameter capillary tubes. If
indeed a larger value of the perfect wetting velocity were to be assumed, then the retardation factor would be reduced (see (7.18)).

Obviously, it is difficult to conclude solely on the results provided by one test. As already pointed out in Section 7.2.2.5, more spontaneous infiltration tests into 0.66 mm diameter capillary tubes are needed to further characterize the effects of tube diameter on DNAPL flow behavior.

7.2.3.4 Summary Plots Revisited

In Section 7.2.2.4, Figure 7.18 showed a summary graph where, for each spontaneous infiltration test reported in Table 7.2, Table 7.3 and Table 7.4, the ratio of the observed mid-depth and three-quarter depth velocities to the NICCA predicted velocity were plotted against the capillary tube length. Figure 7.19 showed a similar graph, except that the velocity ratio was plotted versus the capillary tube length normalized by its diameter, \( l/d \). This latter plot appeared to be falling on a unique curve, thereby suggesting that the velocity reduction might solely be a function of \( l/d \). This point was discussed in Section 7.2.2.5. A set of three possible hypotheses was proposed. The object of this section is to demonstrate that the second hypothesis is correct. That is, the resisting capillary force is present for both 1.33 mm and 2.70 mm diameter capillary tubes, but its magnitude is less for 2.70 mm diameter capillary tubes. In addition, for the 2.70 mm diameter tubes, the combined resisting inertia force and resisting capillary force appear to have the same magnitude as the resisting capillary force of the 1.33 mm diameter capillary tubes. Hence, the velocity reduction appears to be solely a function of \( l/d \), even if it is not.

A simple way of demonstrating that the second hypothesis is correct is to build a theoretical plot of the ratio of the NIVCA predicted velocity to the NICCA predicted velocity versus the capillary tube length, and to compare this theoretical plot to the summary plot shown in Figure 7.18.

Assuming that the NIVCA predicted mid-depth and three-quarter depth velocities are larger than the perfect wetting velocity, \( u_c \), of 3 mm/s, then the model equation (7.15) applies. The NIVCA mid-depth and three-quarter depth velocities are thus given by

\[
\frac{dz_c}{dt} = \frac{k_h}{l + h_{pi}} \left[ z_c(t) + \Delta h' \right], \tag{7.20}
\]

with \( z_c(t) \) equal to \( l/2 \) for the mid-depth velocity and \( 3l/4 \) for the three-quarter depth velocity. Note that, when examining Table 7.2 and Table 7.3, it can be seen that all the measured mid-depth and three-quarter depth velocities are larger than 3 mm/s (with the exception of the mid-depth velocity of test L8b-1.33st-122mm where the velocity is equal to 2.5 mm/s). Thus, for all the reported experiments except for one, the prediction given by (7.15) can be used. As a first approximation, it is also assumed that this prediction is valid for test L8b-1.33st-122mm.
The NICCA mid-depth and three-quarter depth velocities are given by (see (3.61) in Section 3.5.2.5)

\[
\frac{dz_c}{dt} = \frac{k_A}{l + h_{pi}} z_c(t),
\]

(7.21)

with, again, \( z_c(t) \) equal to \( l/2 \) for the mid-depth velocity and \( 3l/4 \) for the three-quarter depth velocity. Let \( R_{VC}(z_c) \) be the ratio of the NIVCA predicted velocity to the NICCA velocity at depth \( z_c \) (\( dz_c/dt > u_c \)). Using (7.20) and (7.21), \( R_{VC}(z_c) \) is given by

\[
R_{VC}(z_c) = 1 + \frac{\Delta h'}{z_c}.
\]

(7.22)

Using the definition of \( \Delta h' \) given by (7.17), (7.22) leads to

\[
R_{VC}(z_c) = 1 + \frac{h - \frac{2\sigma}{\Delta \rho g r_0} - \frac{2n_p f_p}{\Delta \rho g r_0}}{z_c}.
\]

(7.23)

Assuming that \( h \approx h_i \) and replacing infiltration criterion (7.2) into (7.23) gives

\[
R_{VC}(z_c) = 1 - \frac{2\sigma (1 - \cos \theta_s)}{\Delta \rho g r_0 z_c}.
\]

(7.24)

Figure 7.43 shows a plot of the ratios of the mid-depth and three-quarter depth measured velocities to the velocities predicted by the NICCA model versus the length of the 1.33 mm diameter capillary tubes. The data points correspond to each spontaneous infiltration test reported in Table 7.3, and are identical to the 1.33 mm diameter capillary tube data points plotted in the summary graph shown in Figure 7.18. Figure 7.44 is similar to Figure 7.43 except that the tests under consideration are the spontaneous infiltration tests performed into 2.70 mm diameter capillary tubes (see Table 7.2 as well as Figure 7.18).

In both Figure 7.43 and Figure 7.44, the predictions of the ratio of the NIVCA predicted velocity to the NICCA predicted velocity (7.24) are also plotted. For mid-depth velocity plots (Figure 7.43.a and Figure 7.44.a), the ratio \( R_{VC}(z_c) \) is evaluated at \( l/2 \) (i.e. \( R_{VC}(l/2) \)). For three-quarter depth velocity plots (Figure 7.43.b and Figure 7.44.b), the ratio \( R_{VC}(z_c) \) is evaluated at \( 3l/4 \) (i.e. \( R_{VC}(3l/4) \)).
7.44.b), the ratio $R_{VC}(z_c)$ is evaluated at $3l/4$ (i.e. $R_{VC}(3l/4)$). Each solid line shown in the figures corresponds to (7.24) evaluated for a particular value of the static contact angle, $\theta_s$. For example, the solid line “$\theta_s = 40^\circ$” shown in Figure 7.43.a corresponds to (7.24) evaluated as

$$R_{VC}(l/2) = 1 - \frac{2 \times 0.032 \times (1 - \cos 40^\circ)}{72 \times 9.807 \times (1.33 \times 10^{-3} / 2) \times l / 2}.$$  (7.25)

Figure 7.43 shows that the ratios $R_{VC}(l/2)$ and $R_{VC}(3l/4)$ predict the observed velocity ratios of spontaneous infiltration tests into 1.33 mm diameter capillary tubes if a static contact angle of the order of $40^\circ$ is assumed. This value is consistent with the range of static contact angles $34.3^\circ$-55.4$^\circ$ (average of $43^\circ$) reported in Table 7.6. On that basis, it can be argued that the 1.33 mm diameter capillary tube velocity reduction observed in Figure 7.18 is mostly attributable to the presence of the pinning force, as well as the increase in interfacial tension force due to the decrease of the dynamic contact angle with velocity.

On the other hand, Figure 7.44 shows that the ratios $R_{VC}(l/2)$ and $R_{VC}(3l/4)$ predict the observed ratios of spontaneous infiltration tests into 2.70 mm diameter capillary tubes if a static contact angle of the order of $70^\circ$ is assumed. This value is an upper-bound of the range of contact angles $31.2^\circ$-68.1$^\circ$ (average of $54^\circ$) reported in Table 7.7. In other words, if the velocity reduction were solely attributable to capillary resistance, a capillary resistance larger than that back-calculated from the infiltration data (under the form of an initial static contact angle larger than that back-calculated) would be necessary to give a correct NIVCA prediction. Under these circumstances, it can be argued that the 2.70 mm diameter capillary tube velocity reduction observed in Figure 7.18 is partly attributable to an increase in capillary resistance due to the decrease of the dynamic contact angle with velocity, as well as partly attributable to inertia effects which, similar to the capillary resistance, increase as the capillary tube length decreases. This is further corroborated by the $\alpha_t$-number, the magnitude of which is consistent with existing inertia effects.

The capillarity-inertia double mechanism is illustrated in Figure 7.45, which shows a plot of the ratios of measured velocities to NICCA predicted velocities identical to that shown in Figure 7.44 (2.70 mm diameter capillary tubes). The solid lines are the predictions of the ratios of the NIVCA predicted velocity to the NICCA predicted velocity (7.24), $R_{VC}(l/2)$ and $R_{VC}(3l/4)$, evaluated at the average back-calculated value of the static contact angle, $\theta_s = 54^\circ$. The dotted lines are hypothesized predictions that take into account the combined effects of the capillary force and inertia force. The inertia force increases in magnitude as the length of the capillary tube decreases. Data points do not exactly fall on the dotted line, as the static contact angle varies between different tests, such that the solid line corresponds only to an average prediction for the entire test series.
Figure 7.43. Ratio of observed velocity to velocity predicted by the NICCA model as a function of the length, $l$, of 1.33 mm diameter capillary tubes, and prediction from (7.24) for different values of $\theta_s$: a. Mid-depth velocity and prediction $R_{VC}(l/2)$; b. Three-quarter depth velocity and prediction $R_{VC}(3l/4)$.

Figure 7.44. Ratio of observed velocity to velocity predicted by the NICCA model as a function of the length, $l$, of 2.70 mm diameter capillary tubes, and prediction from (7.24) for different values of $\theta_s$: a. Mid-depth velocity and prediction $R_{VC}(l/2)$; b. Three-quarter depth velocity and prediction $R_{VC}(3l/4)$. 
Figure 7.45 and, more generally, the mechanisms proposed here for the spontaneous infiltration experiments into 2.70 mm diameter capillary tubes are based on the assumption that the dynamic pinning force is equal to 0.005 N/m—recall that this value is based on an estimate obtained from data on the infiltration height. Using this pinning force, an initial static contact angle, $\theta_s$, can be back-calculated (see (7.19)). Clearly, it could be argued that a larger pinning force could be used that would yield a larger initial static contact angle in overall agreement with the data reported in Figure 7.45, so that it could be concluded that inertia forces may also be unimportant for 2.70 mm diameter capillary tubes. Conversely, it could be speculated that the pinning force is less than assumed in the NIVCA model, and that the velocity reduction for these tubes is essentially due to the effects of inertia.

![Figure 7.45](image)

**Figure 7.45.** Ratio of observed velocity to velocity predicted by the NICCA model as a function of the length, $l$, of 2.70 mm diameter capillary tubes, prediction from (7.24) for $\theta_s = 54^\circ$ (solid lines), and hypothesized prediction that includes the combined effects of capillary resistance and inertia (dotted lines): a. Mid-depth velocity; b. Three-quarter depth velocity.

What is required to further confirm the mechanism hypothesized above is an independent measurement of the dynamic pinning force and/or static contact angle, as well as a quantitative description of the inertia-driven velocity reduction. Neither the measurement of the pinning force nor that of the contact angle would be easy to perform experimentally. Measurements of the geometric static contact angle need to be corrected for the radial magnification of the interface meniscus due to the capillary tube refraction [see, for example, Hoffman, 1975]. Furthermore, contact angle measurements would be complicated here by the presence of the tank tube containing...
the bulk of water (see, for example, Figure 6.2.c), which would also contribute to the magnification of the radial dimensions of the interface. Measurements of the dynamic pinning force could be achieved, for example, by examining the DNAPL/water interface displacement in horizontal capillary tubes under a configuration of forced wetting, where the interface velocity can be controlled and made constant [see, for example, Calvo et al., 1991] (see also Section 2.3.6).

As also noted, a quantitative description of the inertia-driven velocity reduction would be required. Although the $\alpha_t$-number determines whether or not the magnitude of inertia forces are important compared to the viscous forces, it does not provide any estimate of the velocity reduction corresponding to those effects. For example, it is possible that a $\alpha_t$-number of the order of 0.5—significant compared to one—contributes only a small fraction of the velocity reduction. Resolution of a governing equation that would include both the effects of local and convective inertia is thus indicated, but would nevertheless involve a complex numerical procedure along with experiments to validate any prediction. Note that the results of the spontaneous infiltration centrifuge experiments will provide further characterization of the velocity reduction associated with the $\alpha_t$-number (see Section 7.3.6.3).

### 7.2.3.5 Theoretical Criterion for Which the NIVCA Model Reduces to the NICCA Model

Using (7.24), it can be seen that the NIVCA model reduces to the NICCA model provided that $z_c \gg 2\sigma(1 - \cos \theta_s)/(\Delta \rho g r_0)$. Thus, the NICCA model can generally be used instead of the NIVCA model to predict the DNAPL infiltration kinetics when the following condition is achieved

$$l \gg \frac{2\sigma(1 - \cos \theta_s)}{\Delta \rho g r_0}.$$  \hspace{1cm} (7.26)

The length $2\sigma(1 - \cos \theta_s)/(\Delta \rho g r_0)$ constitutes the length scale of the region over which capillary forces are important in comparison to viscous forces. When (7.26) is true, the length scale of the region over which capillary forces have an effect on the infiltration kinetics is negligible in comparison to the length, $l$, of the capillary tube. Under these circumstances, the increase in capillary forces with increasing interface velocity has no incidence on the infiltration kinetics, and the NICCA model can be used instead of the NIVCA model to predict the infiltration kinetics.

In the absence of information on the static contact angle, an upper bound of the length scale $2\sigma(1 - \cos \theta_s)/(\Delta \rho g r_0)$ is $2\sigma/(\Delta \rho g r_0)$. Thus, if the following condition is true

$$l \gg \frac{2\sigma}{\Delta \rho g r_0},$$  \hspace{1cm} (7.27)
then the changes in capillary force with interface velocity have no effect on the DNAPL infiltration kinetics, and the NICCA model can be used to provide a prediction of the kinetics. This criterion is a more general expression of the empirical criterion \( l/d > 600 \) proposed in Section 7.2.2.4, which has been verified only in the case where 4-CT is the DNAPL and if the capillary tube diameter is in the range of diameters 0.66 mm to 2.70 mm.

Note that the condition \( l/d > 600 \) is an empirical criterion that also “checks” that for the experimental system, inertia forces are negligible with respect to viscous forces. In contrast, the criterion (7.27) solely checks that the effects of capillary force changes have no measurable effect on the infiltration kinetics. In addition to (7.27), for the NICCA model (or the NIVCA model) to be applicable, recall that the inertia forces and entry drag forces must be negligible in comparison to the viscous forces, i.e. the \( \alpha_t \)-number (see (3.58) in Section 3.5.2.5) must be negligible with respect to unity. Therefore, for the NICCA model (or the NIVCA model) to be applicable, the following condition must also be true

\[
\alpha_t = \frac{\Delta \rho g r_0^4}{16 \mu_t^2 l_t} << 1, \quad (7.28)
\]

which can be rewritten as

\[
l_t >> \frac{\Delta \rho g r_0^4}{16 \mu_t^2}. \quad (7.29)
\]

In conclusion, if (7.29) is true, then the effects of inertia and entry drag forces are not measurable, and the NIVCA model can be used to obtain a prediction of the DNAPL infiltration kinetics. In addition, if (7.27) is also true, the NICCA model can be used instead of the NIVCA model to obtain a prediction of the kinetics. These criteria can easily be extended to different fracture geometries (capillary tubes of rectangular section and rough-walled fractures).

7.2.3.6 Dynamic Infiltration Heights

**Background**

In Section 7.2.3.2, the concept of dynamic infiltration height, \( h_t(dz_t/dt) \), was introduced. The dynamic infiltration height—as opposed to the static infiltration height, \( h_s \), measured at the onset of infiltration—is back-calculated under conditions where the interface meniscus is in motion. As will be shown here, the difference between the dynamic and the static infiltration heights, \( h_t(dz_t/dt) - h_s \), can be seen as the resisting force that is missing in the NICCA model prediction. The object of this
section is to further examine the experimental results in terms of the dynamic infiltration height to better characterize the missing force and to verify whether the resisting force can be attributed to inertia effects or capillary type resistance.

If the NICCA model were correct, the infiltration height, \( h_i \), would remain constant throughout the infiltration process and would be equal to the gravity force (or buoyant weight) of the DNAPL pool, \( h \) (strictly speaking the DNAPL pool \( h \) would be slightly larger than \( h_i \) for infiltration to take place). Under these circumstances, the driving gravity force associated with the DNAPL finger of length \( z_c \) would exactly be equal to the viscous resistance of the DNAPL/water column. This condition is expressed by (3.61) in Section 3.5.2.5 (see also (7.21)).

In contrast, the NIVCA model assumes that the dynamic contact angle, \( \theta_{dc} \), decreases with interface velocity, such that the dynamic capillary force is larger than the static capillary force. Taking this proposed mechanism into account, the governing equations (7.14) and (7.15) establish the relationship between viscous, gravity and dynamic capillary forces under an assumed dynamic capillary force-velocity relationship (7.11).

Recall (7.12)

\[
\frac{dz_c}{dt} = \frac{k_\Delta}{l + h_{pi}} \left[ z_c(t) + h - h_i(\frac{dz_c}{dt}) \right],
\]

(7.30)

where \( h_i(\frac{dz_c}{dt}) \) is the dynamic infiltration height. Rearranging (7.30), it can be shown that

\[
h_i(\frac{dz_c}{dt}) = z_c(t) + h - \frac{l + h_{pi}}{k_\Delta} \frac{dz_c}{dt}.
\]

(7.31)

Equation (7.31) is equivalent to the momentum equation characterizing the balance of forces acting on the entire DNAPL/water column. Therefore, the dynamic infiltration height, \( h_i(\frac{dz_c}{dt}) \), can be seen as the force term that includes all the forces not otherwise taken into account in the right-hand side term of (7.31). The forces in the right-hand side term of (7.31) are: (1) the buoyant weight of the DNAPL/water column—this weight is shown as an equivalent height \( z_c(t) + h \)—and (2) the viscous force—corresponding to the equivalent height \( (l/k_\Delta)dz_c/dt \)—acting along the DNAPL/water column over the total length of the capillary tube (note that, strictly speaking, this is the viscous force of an equivalent column of water, as it is assumed that \( \mu_{nw} = \mu_{nw} \)). Thus, \( h_i(\frac{dz_c}{dt}) \) corresponds to the combination of the missing driving/resisting forces, including the resisting dynamic capillary force and, under some circumstances, the resisting inertia force. By plotting \( h_i(\frac{dz_c}{dt}) \) as a function of the interface displacement velocity, \( \frac{dz_c}{dt} \), it is possible to have a better idea of the nature of the resisting force that is unaccounted for. Note that the missing force—in units of force—can be obtained by multiplying the infiltration height difference, \( h_i(\frac{dz_c}{dt}) - h_i \), by the coefficient \( \pi \Delta \rho g r_0^2 \).
Assuming that the NIVCA model is correct, then perfect wetting is reached \((\theta_s = 0)\) when the interface velocity reaches the perfect wetting velocity, i.e. \(dz_c/dt = u_c\). Beyond the perfect wetting velocity, i.e. for \(dz_c/dt > u_c\), the dynamic infiltration height, \(h_i(dz_c/dt)\), is a constant equal to \(h_i(\theta_s = 0)\), given by

\[
h_i(\theta_s = 0) = \frac{2\sigma}{\Delta \rho g r_0} + \frac{2n_p f_p}{\Delta \rho g r_0}. \tag{7.32}
\]

If the NIVCA model is correct, then the function \(h_i(dz_c/dt)\) is expected to increase as \(dz_c/dt\) increases, and reach an asymptotic value given by \((7.32)\). Therefore, for each infiltration test, it is possible to compute the magnitude of the pinning force, \(n_p f_p\), from the asymptotic dynamic infiltration height, \(h_i(\theta_s = 0)\), and the DNAPL/water interfacial tension, \(\sigma\). Incidentally, the velocity for which the asymptotic value of \(h_i(\theta_s = 0)\) is reached is the value of the perfect wetting velocity, \(u_c\). The static contact angle, \(\theta_s\), can then be obtained using (7.19) as was done in Section 7.2.3.3.

If, on the other hand, the dynamic infiltration height does not reach an asymptotic value, or if the back-calculated pinning forces are inconsistent with those assumed for the NIVCA model prediction in Section 7.2.3.3, it can be argued that other mechanisms, not taken into account by the NIVCA model, are present. In any case, the dynamic infiltration height is a combination of all the other driving/resisting effects, so that it may not be possible to tell them apart. For example, even if the pinning force were to decrease with interface velocity, as suggested in Section 7.2.2.5, it would be difficult to identify this mechanism given that the interfacial tension force could be simultaneously increasing.

Use of (7.31) to obtain dynamic infiltration height-interface velocity relationship (infiltration tests into 1.33 mm diameter capillary tubes)

The method described above is applied to the series of spontaneous infiltration tests into 1.33 mm diameter capillary tubes reported in Table 7.3 as well as the series of controlled-head infiltration experiments into 1.33 mm diameter capillary tubes examined in Section 7.2.3.3.

Using the time-depth infiltration data for the experiments, the dynamic infiltration height, \(h_i(dz_c/dt)\) is computed using (7.31). For spontaneous infiltration tests, the value of the infiltration height, \(h_i\) (see Table 7.3), measured for the test is used for the value of \(h\), as it is considered that infiltration takes place for \(h \approx h_i\). For controlled-head infiltration tests, the value of the DNAPL pool height set prior to turning the flow valve on is used as the value of \(h\).

Figure 7.46 shows plots of the dynamic infiltration height as a function of the interface velocity for the seven spontaneous infiltration tests reported in Table 7.3 (see Figure 7.46.a) and the six controlled-head infiltration tests into a 1.33 mm diameter capillary tube (see Figure 7.46.b). A three point average technique similar to that described in Section 7.2.2.3 was used to obtain smoother plots.
Figure 7.46. Dynamic infiltration height versus interface velocity using (7.31) for 4-CT infiltration tests into 1.33 mm diameter capillary tubes: a. Spontaneous infiltration tests into tubes of varying length; b. Controlled-head infiltration tests into a 305 mm long capillary tube under 4-CT pools of varying height.
As can be seen in Figure 7.46.a, the dynamic infiltration height increases as the interface velocity increases from 0 to a velocity ranging from 1 mm/s to 8 mm/s depending on the infiltration test under consideration. Passed this minimum velocity, the dynamic infiltration height appears to remain constant, with the exception of the two infiltration tests run on the tubes of longer length, i.e. \( l = 912 \text{ mm} \) and \( l = 610 \text{ mm} \). The maximum dynamic infiltration height varies between 145 mm and 185 mm depending upon the test under consideration. The variability of the maximum dynamic infiltration height, however, is consistent with that of the measured infiltration height, which varies from 120 mm to 155.2 mm for this test series.

For the two tests run on the tubes of length \( l = 912 \text{ mm} \) and \( l = 610 \text{ mm} \), the dynamic infiltration height decreases at interface velocities larger than 20 mm/s and, in the case of the tube \( l = 912 \text{ mm} \), takes values lower than the critical height measured at the onset of infiltration, \( h_i \).

Referring now to Figure 7.46.b, the dynamic infiltration height for controlled-head infiltration tests appears to remain more or less constant, independent of the both the interface velocity and the 4-CT pool height, \( h \). The dynamic infiltration height for these tests is of the order of 195 mm.

The decreasing trend shown in Figure 7.46.a for the spontaneous infiltration tests on the tubes of length \( l = 912 \text{ mm} \) and \( l = 610 \text{ mm} \) is not believed to be related to a physical phenomenon per se, for example that the pinning force decreases with velocity. Rather, it is a consequence of the structure of (7.31), in which the equivalent conductivity, \( k_{\Delta} \), is imbedded. Indeed, the right-hand side (7.31) incorporates the difference \( z_c(t) - (l/k_{\Delta})dz_c/dt \). Specifically, this term refers to the difference between the measured velocity field and the velocity field predicted by the NICCA model. Hence, (7.31) is sensitive to any approximation made when developing the NICCA model (see viscosity discussion of Section 7.2.2.5). In particular, the NICCA model assumes that the viscosity contrast between 4-CT and water \( \Delta \mu = \mu_{nw} - \mu_w \) is negligible with respect to the viscosity of water. In Section 5.5.2, the viscosity of dyed 4-CT has been measured and found to be equal to 0.918 mPa.s (see Table 5.3). While the viscosity contrast can be assumed negligible when comparing the measured infiltration profile and, to some extent, for the viscosity fields with the NICCA model, the approximation becomes relatively large when it comes to plotting the difference between measurement and prediction, such as the graphs shown in Figure 7.46.

Modification of dynamic infiltration height equation (infiltration tests into 1.33 mm diameter capillary tubes)

Taking into account the effects of viscosity change associated with a DNAPL finger of length \( h_{pi} + z_c \) replacing water inside a capillary tube, it can be shown that the equivalent conductivity \( k_{\Delta} \), which is inversely proportional to \( \mu_w \) (see (3.43) in Section 3.5.2.1), must be divided by a dimensionless correction factor \( ((h_{pi} + z_c)\mu_{nw} + (l - z_c)\mu_w)/(\mu_{nw} + l)\mu_w) \). Thus, for viscosity change effects to be incorporated, (7.31) must be replaced by
\[ h_i \left( \frac{dz_c}{dt} \right) = z_c(t) + h - \frac{(h_{pi} + z_c)\mu_{nw} + (l - z_c)\mu_w}{\mu_k\lambda} \frac{dz_c}{dt} \quad (7.33) \]

Note that this expression can also be derived using the governing equation (3.76) (see Section 3.5.4) obtained when examining the infiltration of a DNAPL of viscosity differing from that of water.

Furthermore, if the effects of reservoir head reduction discussed in Section 7.2.2.5 are also included, then (7.33) must be replaced by

\[ h_i \left( \frac{dz_c}{dt} \right) = z_c(t) \left( 1 - \frac{r_0^2}{r_r^2} \right) + h - \frac{(h_{pi} + z_c)\mu_{nw} + (l - z_c)\mu_w}{\mu_k\lambda} \frac{dz_c}{dt} \quad (7.34) \]

where use of (7.8) has been made.

Figure 7.47 and Figure 7.48 show plots of the dynamic infiltration height as a function of the interface velocity for seven spontaneous infiltration tests reported in Table 7.3 (see Figure 7.47.a and Figure 7.48.a) and the six controlled-head infiltration tests into 1.33 mm diameter capillary tube (see Figure 7.47.b and Figure 7.48.b). The plots shown in Figure 7.47 are obtained using (7.33) (4-CT viscosity correction with \( \mu_{nw} = 0.918 \) mPa.s), whereas the plots shown in Figure 7.48 are obtained using (7.34) (4-CT viscosity correction and reservoir tube correction with \( r_r \) ranging from 3.75 mm to 4 mm depending on the test).

As can be seen in Figure 7.47, when using (7.33) for a given infiltration test, the calculated dynamic infiltration height is found to be larger than that calculated using (7.31) (see Figure 7.46). Referring now to Figure 7.48, when using (7.34), the calculated dynamic infiltration height for a given infiltration test is found to be smaller than that calculated using (7.33) (see Figure 7.47), but larger than that computed from (7.31) (see Figure 7.46). It can also be seen that, for a given infiltration test, the difference in dynamic infiltration height between figures increases as the velocity increases.

The trends described above are expected. Equation (7.33) incorporates the correction for the viscosity of 4-CT, which is somewhat smaller than that of water. As the infiltration proceeds, more 4-CT infiltrates the capillary tube and replaces water, so that, at a given flow velocity, the magnitude of the resisting viscous force of the 4-CT/water column of length, \( l_c \), is less than that of an equivalent water column associated with (7.31). The contrast between (7.33) and (7.31) is largest at large velocities, as the largest velocities correspond to the times where 4-CT has almost entirely drained water out of the capillary tube.

Equation (7.34) incorporates both the effects of viscosity contrast and reservoir tube drainage. The driving gravity force expressed by the term \( z_c(t)(1 - r_0^2/r_r^2) + h \) in (7.34) is less than that of (7.33), so that the dynamic infiltration height is found to be less in the former case. Again, the difference in dynamic infiltration height between models increases as the velocity increases because \( z_c(t) \) increases along with the interface velocity and contributes to increase the magnitude of the correction.
Results and discussion on the dynamic infiltration height-velocity graph of spontaneous infiltration tests into 1.33 mm diameter capillary tubes

Referring to Figure 7.48.a, it can be seen that the dynamic infiltration height appears to reach an asymptotic value for the three spontaneous infiltration tests into the three long 1.33 mm diameter capillary tubes (l = 912 mm, l = 610 mm and l = 265 mm), although a decreasing trend is still observed for the longest capillary tube. For tube l = 912 mm, the dynamic infiltration increases to about 175 mm at the interface velocity of 2 mm/s and slowly decreases to 140 mm/s as the interface velocity increases to 40 mm/s. For tube l = 610 mm, the dynamic infiltration height increases to 180 mm and remains more or less stable at this value from 2 mm/s to 38 mm/s. For tube l = 265 mm, the dynamic infiltration height increases from 120 mm to 145 mm between 0 mm/s and 5 mm/s, and remains at 145 mm throughout the rest of the infiltration process.

For the four short capillary tubes, it does not appear, however, that the dynamic infiltration height reaches an asymptotic value. Instead, the dynamic infiltration heights increase from their initial infiltration heights to maximum values of about 200 mm when reaching the lower end of the capillary tube (i.e. at maximum interface velocity).

With the exception of capillary tube l = 265 mm, the plot of dynamic infiltration height shown in Figure 7.48.a suggests that generally for a given infiltration velocity, the shorter the capillary tube, the larger the dynamic infiltration height, and thus the larger the resisting force associated with \( h_i \left( \frac{dz}{dt} \right) \).

In the same figure, the asymptotic line \( h_i(\theta_s = 0) = 178.9 \) mm corresponding to the pinning force, \( n_{pf_p} = 0.010 \) N/m, is plotted. As can be seen, this line is in good agreement with the asymptotic lines of tests l = 912 mm and l = 610 mm, which explain why the experimental data shown in Figure 7.27 and Figure 7.28 were well predicted by the NIVCA model. Conversely, the asymptotic line \( h_i(\theta_s = 0) = 178.9 \) mm is far above from the asymptotic line of test l = 265 mm, explaining why the NIVCA prediction was so poor for this test.

For the four tests into the short capillary tubes, the dynamic infiltration height plots above the asymptotic line corresponding to the pinning force \( n_{pf_p} = 0.010 \) N/m, suggesting, as pointed out in Section 7.2.3.3 that either (1) there is extra resistance due to further capillary resistance for these tests, in particular that the pinning force is larger than 0.010 N/m, or (2) there exists a secondary resisting force.

Hypothesis (1) can be justified by the observation that the four spontaneous infiltration experiments into short tubes are associated with larger infiltration heights, \( h_i \), than those of the three longer tubes (see Table 7.6). While the variability of the infiltration height was attributed to the variability of the contact angle (see Section 7.2.1.2), it is possible that this variability is also connected to the variability of the pinning force. Equation (7.2) suggests that the larger the pinning force, the larger the infiltration height at the onset of infiltration. Hence, it is possible that the larger dynamic infiltration heights observed for the four short tubes are attributable to larger pinning forces, which in turn raise the dynamic infiltration heights.
If, however, hypothesis (1) were correct, the dynamic infiltration height would be expected to reach an asymptotic value, even for shorter tubes, a trend not visible in Figure 7.48.a. One explanation for the actual trend that is observed is that the perfect wetting velocity is larger than 25 mm/s for these tubes, although $u_c = 25$ mm/s would appear very high given that longer tubes reach asymptotic behavior at approximately 2 mm/s. Another explanation is that increasing/decreasing trends may not be true trends. Instead, these trends may be attributable to the uncertainty of the geometric characteristic of the capillary tube or the uncertainty of the liquid properties. Indeed, it is important to emphasize that (7.34) is generally quite sensitive to the factor $[(h_{pl} + z_e)\mu_{nw} + (l - z_e)]/(\mu_{nw}k_\Delta)$ preceding $dz_i/dt$ in the right-hand side term of the equation. For example, considering test L10-1.33lct-918mm ($l = 912$ mm as shown in Figure 7.48), it can be shown that a reduction of the viscosity of 4-CT by 0.015 mPa.s from $\mu_{nw} = 0.918$ mPa.s to $\mu_{nw} = 0.903$ mPa.s (i.e. a relative reduction by 1.6%) results in an increase of the dynamic infiltration height by approximately 13 mm at the largest velocities (between 30 mm/s and 40 mm/s), which corresponds to a relative increase of about 8%. A similar increase would also be achieved if, instead, the capillary tube diameter in $k_\Delta$ was increased from 1.33 mm to 1.34 mm, corresponding to an increase of $k_\Delta$ from 39.0 mm/s to 39.6 mm/s. Thus, overall, hypothesis (1) can be justified, even if asymptotic behavior is not observed for the shorter tubes.

Like hypothesis (1), hypothesis (2) can be justified. It can be argued that for short tubes, inertia effects may contribute to the flow resistance, and that these effects, although scarcely visible in the infiltration profiles or the velocity fields, can be visible in dynamic infiltration height-interface velocity graphs—which, as discussed earlier, tend to be sensitive to any model approximation.

The $\alpha_r$-number for the four infiltration experiments into short tubes are equal to 0.047 and 0.071, respectively (see Table 7.3). Therefore, it can be argued that for these values, inertia effects may start to be noticeable. A strong argument in favor of hypothesis (2) is the fact that the dynamic infiltration height, and thus the resisting force is observed to increase with decreasing length, a trend consistent with inertia effects, since the $\alpha_r$-number increases with decreasing capillary tube length (see (3.58) in Section 3.5.2.5). As will be shown later, for spontaneous infiltration tests into 2.70 mm diameter capillary tubes—where the effects of inertia are known to be important—the dynamic infiltration height at a given interface velocity is also observed to increase with decreasing capillary tube length.

From the discussion above, the fact that the resisting force for 1.33 mm diameter capillary tubes tends to increase with decreasing tube length most likely supports the presence of inertia effects, and thus the second hypothesis. Arguing that the first hypothesis is correct would mean that the observed increasing trend in resistance force with decreasing tube length is a coincidence for this test series, and that shorter tubes coincidently happen to have a larger pinning force. Clearly, further testing in tubes of varying length could confirm whether or not this statement is correct.
Figure 7.47. Dynamic infiltration height versus interface velocity using (7.33) for 4-CT infiltration tests into 1.33 mm diameter capillary tubes: a. Spontaneous infiltration tests into tubes of varying length; b. Controlled-head infiltration tests into a 305 mm long capillary tube under 4-CT pools of varying height.
Figure 7.48. Dynamic infiltration height versus interface velocity using (7.34) for 4-CT infiltration tests into 1.33 mm diameter capillary tubes: a. Spontaneous infiltration tests into tubes of varying length; b. Controlled-head infiltration tests into a 305 mm long capillary tube under 4-CT pools of varying height.
Again, it must be emphasized that should there be inertia effects contributing to the interface velocity reduction of infiltration tests into 1.33 mm diameter capillary tubes of short length, their contribution would remain small in comparison to the capillary resistance—i.e. the pinning force and the velocity-dependent interfacial tension force—which, according to the NIVCA model, appears to contribute to most of the velocity reduction.

Results and discussion on the dynamic infiltration height-velocity graph of controlled-head infiltration tests into 1.33 mm diameter capillary tubes

Referring to the dynamic infiltration height-interface velocity graph of controlled-head infiltration experiments into 1.33 mm diameter capillary tubes shown in Figure 7.48.b, it can be seen that the dynamic infiltration height has a constant value of approximately 195 mm for all infiltration tests. In addition, there is no visible dynamic infiltration height increase at low velocity, suggesting that perfect wetting could be reached at interface velocities as low as 1 mm/s.

In the same figure, the asymptotic line (7.32) \( h_i(\theta_s = 0) = 178.9 \) mm corresponding to the pinning force, \( n_{pf_p} = 0.010 \) N/m, is plotted. As can be seen, this line is located below the measured asymptotic line of 195 mm, explaining the poor agreement between experimental data and NIVCA model prediction reported in Figure 7.34. Again, this suggests that either pinning forces are larger than anticipated, or that there exists other effects contributing to further flow resistance.

The asymptotic line \( h_i(\theta_s = 0) = 200.2 \) mm, corresponding to the pinning force, \( n_{pf_p} = 0.015 \) N/m, is also shown Figure 7.48.b. Obviously, this second line is in much better agreement with the measured asymptotic line, which explains why the NIVCA predictions shown in Figure 7.35 were in good agreement with the experimental data.

As pointed out in Section 7.2.3.3, it is not obvious that the difference between the dynamic infiltration height for this test series and that of spontaneous infiltration tests into long tubes is only attributable to a larger pinning force in the case of controlled-head infiltration tests. Other mechanisms may be present, possibly a pressure drop that takes place at the onset of infiltration when the flow valve is turned on. Inertia effects are unlikely, since the \( \alpha \)-number for these tests is equal to 0.028, and because the dynamic infiltration height does not seem to vary with velocity. Clearly, further testing is needed to better characterize the sum of resisting forces for this type of test. Note that it would also be desirable to find a method that would allow either measurement or back-calculation of the infiltration height \( h_i \).

Results and discussion on the dynamic infiltration height-velocity graph of spontaneous infiltration tests into 2.70 mm diameter capillary tubes

Figure 7.49 shows plots of the dynamic infiltration height as a function of the interface velocity for the six spontaneous infiltration tests reported in Table 7.2. The
plots are obtained using (7.31), i.e. with no correction due to viscosity change or reservoir tube drainage. A similar plot incorporating the effects of viscosity change (i.e. (7.33)) is shown in Figure 7.50. A plot incorporating both the effects of viscosity change and reservoir tube drainage (i.e. (7.34)) is shown in Figure 7.51.

Similar to infiltration tests into 1.33 mm diameter capillary tubes, the dynamic infiltration height of tests into 2.70 mm diameter capillary tubes takes larger values when the correction due to viscosity change is taken into account (see Figure 7.50) than when neither correction is included (see Figure 7.49). When both corrections are accounted for, the dynamic infiltration height is smaller that the dynamic infiltration height that solely includes the viscosity correction, and is larger than the dynamic infiltration height that includes no correction.

Referring to Figure 7.51 (both corrections), it can be seen that for all spontaneous infiltration tests, the dynamic infiltration height is observed to increase with interface velocity. The rate of increase is smallest for the long capillary tubes, which exhibit some sort of asymptotic behavior—excepting the test for which \(l = 1202\) mm. For a given interface velocity, the dynamic infiltration height is observed to increase with decreasing capillary tube length, \(l\).

For test \(l = 1202\) mm, the dynamic infiltration height appears to oscillate between 60 mm and 90 mm as the interface velocity increases from 10 mm/s to 90 mm/s. The dynamic infiltration height then increases quickly to 200 mm as the interface velocity increases to 140 mm/s. At present, the reason why the dynamic infiltration increases so much is not well understood. That this increase occurs at the location of a sudden change of capillary tube radius could constitute an explanation. As discussed in Section 6.2.2, long capillary tubes were obtained by connecting 305 mm long capillary tubes together. It is thus possible that a small radius change may have occurred between consecutive tubes, thereby contributing to a sudden change of the magnitude of \(k_\Delta\). Nonetheless, use of (7.34) shows that, for a velocity of about 140 mm/s, a change in diameter from 2.70 mm to 2.72 mm would only increase the dynamic infiltration height by about 15 mm. Therefore, it is possible that true resisting effects may be present here, possibly connected to inertia or entry drag forces.

For test \(l = 909\) mm, the dynamic infiltration height appears to oscillate between 70 mm and 90 mm as the interface velocity increases from 10 mm/s to 90 mm/s. At larger velocities, the dynamic infiltration height is observed to increase to about 140 mm. Again, this later increase may possibly be due to a sudden increase of a resisting force.

For test \(l = 606\) mm, the dynamic infiltration height appears to remain more or less constant and equal to 100 mm with a slightly increasing trend from 90 mm to 110 mm throughout the range of velocities.

The three short tubes, \(l = 301\) mm, \(l = 103\) mm and \(l = 88\) mm are observed to increase from about 80 mm to 120 mm throughout their respective velocity range. The rate of change increases with decreasing capillary tube length.
Figure 7.49. Dynamic infiltration height versus interface velocity using (7.31) for 4-CT spontaneous infiltration tests into 2.70 mm diameter capillary tubes of varying length.

Figure 7.50. Dynamic infiltration height versus interface velocity using (7.33) for 4-CT spontaneous infiltration tests into 2.70 mm diameter capillary tubes of varying length.
Figure 7.51. Dynamic infiltration height versus interface velocity using (7.34) for 4-CT spontaneous infiltration tests into 2.70 mm diameter capillary tubes of varying length.

In the same figure, the asymptotic line (7.32) \( h_\theta(\theta_s = 0) = 77.6 \text{ mm} \) corresponding to the pinning force, \( n_p f_p = 0.005 \text{ N/m} \), is plotted. As can be seen, this line is located below the dynamic infiltration height plots of all the infiltration tests—with the exception of a few data points—and is closest to the tests performed on the longest capillary tubes. This explains why the agreement between the experimental data and the NIVCA model is best for the long capillary tubes. Note also that the asymptotic line is below the measured infiltration height \( h_i \) of test \( l = 88 \text{ mm} \), which explains why no NIVCA prediction could be obtained for this test (see Section 7.2.3.3).

Clearly, it is not expected to observe in Figure 7.51 the changes in dynamic infiltration height attributable to capillary resistance. In the NIVCA model, perfect wetting has been hypothesized to be reached for \( u_c = 3 \text{ mm/s} \), so that contact angle changes are expected to be taking place at a velocity well below the velocity range of the figure. Capillary effects are believed to be present because there exists a difference of about 30 mm to 60 mm between the measured infiltration height (see Figure 7.51) and the dynamic infiltration height at the lowest measured velocities.

Furthermore, it is very likely that the increase of dynamic infiltration height with velocity observed in Figure 7.51 is attributable to inertia forces. This assumption is well supported by the fact that at a given infiltration velocity, the dynamic infiltration
height increases with decreasing capillary tube length, and that for this test series, the $\alpha_t$-number is significant compared to unity. This assumption is also supported by the fact that, at least for the short tubes, and possibly for all tubes, the dynamic infiltration height continuously increases with velocity without reaching an asymptotic behavior.

Results and discussion on the dynamic infiltration height-velocity graph of the spontaneous infiltration test into a 0.66 mm diameter capillary tube

Figure 7.52 shows three plots of the dynamic infiltration height as a function of the interface velocity for the spontaneous infiltration test into the 1201 mm long, 0.66 mm diameter capillary tube reported in Table 7.4. The plots are obtained using (7.31) (no correction), (7.33) (viscosity change correction) and (7.34) (viscosity change and reservoir tube drainage correction). Similar to 1.33 mm and 2.70 mm diameter capillary tubes, the plot of (7.34) that includes both corrections is located between (7.31) and (7.33).

Focusing specifically on the plot of (7.34), a weak increasing trend can be observed from about 275 mm to about 325 mm throughout the velocity range 0 mm/s to 7 mm/s. The dynamic infiltration then decreases to about 300 mm as the velocity increases to 10 mm/s. Large oscillations are observed throughout the velocity range. The dynamic infiltration height takes values as low as 240 mm and as large as 340 mm.

The asymptotic lines (7.32) $h_t(\theta_s = 0) = 326.2$ mm and $h_t(\theta_s = 0) = 377.7$ mm are plotted in the same figure. These lines correspond to the pinning forces, $n_{pf} = 0.006$ N/m and $n_{pf} = 0.012$ N/m, respectively. As can be seen, the line $h_t(\theta_s = 0) = 326.2$ mm is closest to the experimental dynamic infiltration height, which explains why the agreement between the experimental data and the NIVCA model is better if a pinning force of $n_{pf} = 0.006$ N/m is assumed in the model (see Figure 7.42 versus Figure 7.41).

The large oscillations reported above are not well understood. It is possible that these oscillations are the result of noise and/or local change of the capillary tube radius. It is also possible that the oscillations are true change in either the dynamic contact angle and/or the pinning force as the interface displacement proceeds in the capillary tube, and runs into pinning points present on the capillary tube walls. If this was the case, a stick-slip type of motion would result that would be associated with changes in the dynamic infiltration height. Overall, the increase in dynamic infiltration height is consistent with a decrease in contact angle. Given the $\alpha_t$-number of $4.3 \times 10^{-4}$, inertia forces are of no importance here.

Clearly, more tests are needed on 0.66 mm diameter capillary tubes to further characterize the dynamic infiltration-velocity relationship. It is believed that the range of interface velocities 0 mm/s-10 mm/s is of great interest, as most of the dynamic contact angle changes are expected to take place throughout this range. More infiltration tests into capillary tubes of 0.66 mm diameter would thus enable to better capture the capillary force changes with velocity while staying away from the range of velocities for which inertia effects are important.
7.2.3.7 Conclusion

A series of spontaneous infiltration experiments reported in Table 7.2, Table 7.3 and Table 7.4, as well as a series of controlled-head infiltration experiments into a 305 mm long, 1.33 mm diameter capillary tube were compared to the NIVCA model developed in Section 3.5.3 and revisited in Section 7.2.3.2. The NIVCA model was developed to account for the capillary resistance attributed to the pinning force and the decrease of the dynamic contact angle with interface velocity.

Pinning forces of 0.05 N/m, 0.010 N/m and 0.012 N/m were taken for 2.70 mm, 1.33 mm and 0.66 mm diameter capillary tubes, respectively. These pinning forces were obtained from the measurements of infiltration heights reported in Section 7.2.1.

For spontaneous infiltration tests into 1.33 mm diameter capillary tubes, it was found that the NIVCA model prediction was generally in good agreement with the experimental data. The pinning force of 0.010 N/m was consistent with most of the experimental results. Towards the lower end of long capillary tubes, the NIVCA model was observed to underestimate the velocity field. It was shown that the difference was compatible with the viscous force decrease that takes place as 4-CT replaces water in the capillary tube. For shorter capillary tubes, a small extra resisting force was present.
in addition to the capillary resistance. This extra resistance, believed to be attributable to inertia effects, may be detectable for an \( \alpha_t \)-number larger than 0.05. The presence of small inertia effects for shorter capillary tubes is well supported by the fact that the dynamic infiltration height increases with decreasing capillary tube length.

For controlled-head infiltration tests into 1.33 mm diameter capillary tubes, it was found that a pinning force of 0.015 N/m instead of 0.010 N/m was compatible with the experimental results. It cannot be concluded whether or not the extra resistance is entirely attributable to capillary resistance, as the experimental setup may contribute to further pressure drop unaccounted for in the NIVCA and NICCA models.

For spontaneous infiltration tests into 2.70 mm diameter capillary tubes, it was found that the NIVCA model prediction was generally over-predicting the experimental data. It is believed that, for these tubes, two resisting forces beside viscosity are present: (1) the capillary resistance, which contributes to an important reduction in interface velocity; (2) inertia forces which increase in magnitude as the capillary tube length decreases and contribute to part of the velocity reduction. The NIVCA model predictions are very consistent with this trend and show that the infiltration profile and velocity field predictions are closest for the longest capillary tubes.

For the spontaneous infiltration tests into the 1201 mm long, 0.66 mm diameter capillary tube, it was found that the NIVCA model prediction was under-predicting the measured velocity field. Good agreement was obtained if a pinning force of 0.006 N/m was assumed instead of 0.012 N/m. The reason why a smaller pinning force value is needed is not precisely understood. More infiltration tests into 0.66 mm diameter capillary tubes are recommended to properly measure the pinning force for these tubes and investigate if a velocity reduction takes place as the capillary tube length decreases.

In the NIVCA model, constant values of the pinning force for a given capillary tube diameter were assumed, based on which a value of the static contact angle at the onset of infiltration could be calculated for each infiltration test. It is however possible that the pinning force varies between tests. While a quantitative argument has been successfully brought up here to prove that capillary forces could contribute to the observed velocity reduction, there is a need for an independent measurement of the static contact angle and/or the pinning force. As pointed out in Section 7.2.3.4, this is not an easy task and would require an experimental program of its own. Furthermore, there is obviously a need to characterize the relative magnitudes of the capillary force and the inertia force when the latter force is of importance. While the effects of local inertia can easily be included in a governing equation (see for example (3.37) in Section 3.4.5), the effects of convective inertia would add non-linear terms that would complicate the equation resolution.

A criterion on the capillary tube length was developed in Section 7.2.3.5 that can be used to determine when the NIVCA model reduces to the NICCA model to give a prediction of the infiltration kinetics. Provided that \( l >> 2\sigma/(\Delta\rho_0 g r_0) \) (criterion (7.27)), the changes in capillary force with velocity have no measurable effect on the infiltration kinetics, and the NICCA model can be used to compute the prediction. This criterion is in addition to criterion (7.29) (\( \alpha_t << 1 \)), which ensures that inertia forces and entry drag forces are negligible with respect to viscous forces.
7.3 DNAPL Infiltration Centrifuge Experiments

7.3.1 Introduction

This part of Chapter 7 focuses on the centrifuge infiltration experiments. The methodology for this experimental program is described in Section 6.3.

An outline similar to that used by Levy et al. [2002] is used here to present the experimental results. Validation of centrifuge scaling laws through modeling-of-models is first examined in Section 7.3.2. Next, Section 7.3.3 presents the results of the prototype infiltration height measurements. In Section 7.3.4, the infiltration kinetics of the centrifuge experiments are examined and compared to the NICCA physical model developed in Chapter 4 as well as the MFDV. A comparison of the experimental data with the NIVCA model is then examined in Section 7.3.5. Finally, in Section 7.3.6, summary plots characterizing the infiltration kinetics of centrifuge experiments are presented and discussed. In that section, the centrifuge summary plots are also compared to the summary plots of laboratory (1-g) experiments that were obtained in Section 7.2.2.4 and Section 7.2.2.5.

7.3.2 Verification of Centrifuge Scaling Laws: Modeling of Models

7.3.2.1 Overview

Equations developed in Chapter 3 for describing DNAPL infiltration into capillary tubes and rough fractures at the Earth’s gravitational acceleration (laboratory tests) were extended in Chapter 4 to the case where the acceleration is several times the Earth’s gravity (centrifuge tests). Centrifuge scaling laws were developed that connected the behavior of the reduced-scale model to the full-scale prototype (see Table 4.1, as well as the set of equations (4.4) developed in Section 4.2).

To investigate the validity of these laws, scaled data from centrifuge tests conducted on circular cross-section capillary tubes of aperture 0.66 mm and 1.33 mm are compared here to experiments conducted on the full-scale systems, i.e. the equivalent prototypes. If the centrifuge scaling laws are correct, scaled data from centrifuge tests should match the data obtained from the equivalent prototype.

Two full-scale experiments reported in Section 7.2.2 were used to investigate the validity of scaling laws: one on a circular cross-section capillary tube of length 1201 mm and diameter 0.66 mm (see Table 7.4, test L11-0.66ct-1221mm); a second on a circular cross-section capillary tube of length 610 mm and diameter 1.33 mm (see Table 7.3, test L6-1.33st-610mm). Note that, for both of these laboratory tests, the depth of pre-infiltration is small in comparison to the total length of the capillary tube,
so that it is possible to ignore the effects of pre-infiltration when comparing the prototype experimental data to the scaled centrifuge data.

As was done in Chapter 4, lower case notations are adopted when referring to the model and upper case notations when referring to the prototype.

7.3.2.2 Scaled Tests on 0.66 mm Diameter Capillary Tubes

The full-scale (prototype) experiment was performed on the 1201 mm long, 0.66 mm diameter capillary tube. Recall that, for this experiment, the onset of infiltration was observed at a pool height of 308 mm (see Table 7.4 and discussion of Section 7.2.1.2).

Figure 7.53. Depth of prototype interface versus prototype time for 0.66 mm diameter capillary tube modeling-of-models.

Next, the DNAPL infiltration problem was reduced by a factor \( n = 10 \). Data for these tests are reported in Appendix C, Section C.5.4 (tests c19a-0.66ct-119mm and c19b-0.66ct-119mm). Circular capillary tubes of the same diameter, but having a
scaled length of 119 mm, were set up in the centrifuge strong box. 4-CT was pooled in the reservoirs above the capillary tubes at heights 25.9 mm and 27.2 mm. The scaled models were then centrifuged, and infiltration into the capillary tubes was observed at g-levels of 10.1 and 10.8, respectively. The equivalent prototype pool heights at infiltration were therefore 261 mm and 294 mm, which fall within the range of experimental values $H_i$ that were measured for 1-g 4-CT spontaneous infiltration experiments into 0.66 mm diameter tubes, i.e. $263.9 \pm 44.1$ mm (see Table 7.1). Variability associated with those results is further discussed in Section 7.3.3.2.

![Figure 7.54](image)

**Figure 7.54.** Depth of prototype interface versus prototype interface velocity for 0.66 mm diameter capillary tube modeling-of-models.

The prototype infiltration profile, i.e. the prototype depth of the DNAPL/water interface, $Z_c$, versus the prototype time, $T$, for the two centrifuge experiments and the full-scale experiment is graphed in Figure 7.53. The prototype velocity field, i.e. the prototype depth of the 4-CT/water interface, $Z_c$, versus the prototype interface velocity, $dZ_c/dT$, for the two centrifuge experiments and the full-scale experiment is graphed in Figure 7.54. Again, as shown in Table 4.1, depth is scaled by a factor $n$, whereas time and velocity are scaled by factors $n^2$ and $1/n$, respectively. Therefore, an interface displacement test taking place over a time period of a few seconds in the centrifuge at
g-level \( n = 10 \) corresponds to a time scale of the order of a few hundred seconds once it is scaled.

Referring to the infiltration profile in Figure 7.53, good agreement is noted between the scaled centrifuge data and the prototype data, especially in the lower region of the capillary tube. Again, similarly to the approach adopted when comparing experimental data to the NICCA and NIVCA models in Section 7.2, time does not correspond to absolute time. Instead, times for which the DNAPL/water interface reaches the lower end of the capillary tubes are taken as reference values and matched. The rest of the profile is translated accordingly. Another way of comparing scaled centrifuge experiments and prototype experiment is to examine their velocity fields, as shown in Figure 7.54, where a three-point average technique identical to that described in Section 7.2.2.3 was used. Again, overall, reasonable agreement between the scaled models and the equivalent prototype is observed. However, as can be seen in the figure, the interface of the full-scale experiment appears to move somewhat faster than the interface of scaled centrifuge experiments. Furthermore, the offset between full-scale and scaled centrifuge interface velocity data appears constant throughout the infiltration process. The overall difference does not exceed 1 mm/s. It will be shown in Section 7.3.5.2 that the offset between the full-scale experiment velocity field and the centrifuge experiment scaled velocity fields is related to the difference in the measured prototype infiltration height, \( H_i \), between the experiments.

7.3.2.3 Scaled Tests on 1.33 mm Diameter Capillary Tubes

To further investigate the validity of the scaling laws, modeling-of-models was conducted on 1.33 mm diameter capillary tubes. A full-scale test was performed on the 610 mm long, 1.33 mm diameter capillary tube. Recall that, for this experiment, infiltration into the capillary tube was observed at a 4-CT pool height of 130 mm (see Table 7.3 and discussion of Section 7.2.1.2).

For the model tests, two scale factors were adopted, namely \( n = 5 \) and \( n = 10 \). Data for these tests are reported in Appendix C, Section C.5.3 (tests c13-1.33st-120mm and c17-1.33st-60mm). Two 1.33 mm diameter capillary tubes, one 120 mm long and the other 60 mm long, were placed in the centrifuge strong box. 4-CT was pooled above the 120 mm long tube at a height of 30.0 mm, while a pool height of 15.0 mm was formed above the 60 mm long tube. The tubes were centrifuged and infiltration was observed at \( g \)-levels equal to 4.8 and 9.8, respectively. The equivalent prototype pool heights at infiltration were therefore 143 mm and 148 mm, respectively. These values fall within the range of experimental values \( H_i \) that were measured for 1-g 4-CT spontaneous infiltration experiments into 1.33 mm diameter tubes, i.e. 137.4 ± 18.5 mm (see Table 7.1).

The depth of the DNAPL/water interface versus time for the scale model tests and the prototype is given in Figure 7.55. The prototype velocity field for the two centrifuge experiments and the full-scale experiment is given in Figure 7.56. From
these figures, good agreement between the prototype data and scaled centrifuge data performed on the 120 mm long capillary tube can be observed. Note, however, that Figure 7.56 suggests that the scaled velocity field of the centrifuge infiltration test on the 120 mm long capillary tube is less than that of the full-scale experiment velocity field. The offset appears to increase very slightly throughout the infiltration process, but does not exceed 2 mm/s at the lowest depth where the scale centrifuge velocity is reported.

![Graph](image)

**Figure 7.55.** Depth of prototype interface versus prototype time for 1.33 mm diameter capillary tube modeling-of-models.

Examining now the centrifuge test performed on the shorter 60 mm long capillary tube, poor agreement between the prototype data and the scaled centrifuge data is observed for both the infiltration profile and velocity field. At a given prototype depth, the interface velocity of the full-scale problem is found to be about 50% larger than the scaled interface velocity of the centrifuge experiment (see Figure 7.56). It can also be noted that the velocity offset between the two experiments increases with prototype depth to values of the order of 15 mm/s-20 mm/s at the lower ends of the capillary tubes.
To investigate further the effect of the model length on the validity of the centrifuge scaling laws, another centrifuge test at a scale factor $n = 15$ was performed. A 1.33 mm diameter capillary tube of length 40 mm was placed in the centrifuge strong box (test c13-1.33st-40mm, see Section C.5.3). A 4-CT pool height of 9.9 mm was added to the reservoir tube. The capillary tube was centrifuged and infiltration was observed at 12.2 $g$. The corresponding prototype infiltration pool height is thus 121 mm, which is lower than anticipated, but yet falls within the range of experimental values $H_f$ that were measured for 1-$g$ 4-CT spontaneous infiltration experiments into 1.33 mm diameter tubes, i.e. $137.4 \pm 18.5$ mm. The infiltration profile and velocity field for this test are also plotted in Figure 7.55 and Figure 7.56. Note that there were only five data points for this test, resulting in only two data points after three-point averaging and velocity computation (see Figure 7.56). From both figures, disagreement between the scaled data and the prototype data is even more significant than that for the scaled data of the 60 mm long capillary tube, suggesting that the model length and diameter of the capillary tube strongly affect the effectiveness of modeling-of-models. As shown in Figure 7.56, at a given prototype depth, the scaled interface velocity
velocity of the centrifuge test in the 40 mm long capillary tube is about half the interface velocity of the full-scale experiment.

7.3.2.4 Discussion

An important assumption of the centrifuge scaling laws presented in Table 4.1 is that the effects of inertia are negligible during DNAPL transport in the fracture system. Recalling the discussion of Chapter 4, successful modeling of DNAPL infiltration using the geotechnical centrifuge relies on neglecting the effect of inertia forces. In Section 4.3.2, it was shown that the magnitude of inertia forces associated with a scaled system is larger in a centrifuge model than in the equivalent prototype, such that the $\alpha$-number is multiplied by a factor $n^2$ (see (4.18)). Thus, it is necessary to check whether the $\alpha_n$-number associated with a centrifuge experiment run at $n$ gravities is negligible with respect to unity, before the results of a centrifuge test can be assumed to correctly reproduce conditions in an equivalent prototype.

For the scaled tests on the 0.66 mm diameter capillary tubes, the value of $\alpha$ for the prototype is equal to $4.35 \times 10^{-4}$ (using (3.46) in Section 3.5.2.1). Thus, $\alpha_{10}$ is expected to be equal to $4.35 \times 10^{-2}$ for a centrifuge model test run at $n = 10$. Values equal to $4.42 \times 10^{-2}$ and $4.74 \times 10^{-2}$ were calculated for the two scale model experiments that underwent infiltration at 10.1 and 10.8, respectively. The good agreement between the scaled centrifuge test data and the prototype data (see Figure 7.15 and Figure 7.16) indicates that the scaling laws given in Table 4.1 are valid under these conditions.

For the scaled tests on the 1.33 mm diameter capillary tubes, the value of $\alpha$ for the prototype is equal to $1.41 \times 10^{-2}$ (again, using (3.46)), yielding $\alpha_5 = 0.353$, $\alpha_{10} = 1.41$ and $\alpha_{15} = 3.18$. Values equal to 0.342, 1.41 and 2.63 were obtained at 4.8 g, 9.8 g and 12.2 g, respectively. Clearly, the effects of inertia are not negligible for the 1.33 mm diameter capillary tubes, $l = 60$ mm and $l = 40$ mm, and likely contribute to the offset observed between the full-scale experiment and scale centrifuge experiment into the 120 mm long capillary tube (see discussion in Section 7.3.5.2).

Thus, the experimental results show that there is a limit to the centrifuge scaling factor $n$ that can be used to model DNAPL transport in capillary tubes and more generally in a fracture system. The limit is fixed by the magnitude of $\alpha_n$ (see (4.2) in Section 4.2) and how its magnitude compares to unity. However, that this limit exists does not negate the utility of centrifuge testing in this area. Rather, it sets the bounds for the range of scale factors that can be used in the modeling of a specific prototype. For example problems will not arise with modeling DNAPL behavior in a fracture of two meters in length with an average planar aperture of 0.2 mm until $n$ exceeds 160. Alternatively, for the same fracture aperture and a fracture length of 0.2 m, $n$ cannot exceed 50.
7.3.3 Prototype Infiltration Heights: Results and Interpretation

7.3.3.1 Summary of Results

Table 7.9 summarizes the results of the DNAPL prototype infiltration (or critical) pool height measurements of the infiltration centrifuge experiments run on 4-CT. For these tests, the \( g \)-level was slowly increased until infiltration took place, so that it can be expected that the prototype pool height at the onset of infiltration was close to the prototype critical height. That is, \( H \approx H_i \) (\( \Delta H \approx 0 \)) can be assumed. Again, the prototype infiltration height \( H_i \) was obtained by multiplying the \( g \)-level at the onset of infiltration, \( n_i \), by the model 4-CT pool height, \( h_i \), set for the experiment. Pre-infiltration for these tests is believed to be negligible (see discussion in Section 7.3.3.2). No test was run using TCA as the DNAPL because the density contrast between water and TCA was so large that the corresponding \( g \)-level at the onset of infiltration would have been extremely low. Details on each experiment can be found in Appendix C, Section C.5.

Use of the former infiltration criterion to obtain an infiltration height prediction

As in the case of 1-\( g \) tests (see Figure 7.1), comparison between measured and predicted values of \( H_i \) is first made using the infiltration criterion that does not include the effects of pinning, i.e. (4.5) derived in Section 4.2. In Figure 7.57, the measured prototype infiltration heights are plotted versus the diameters of the capillary tubes used for the experiments. The prediction zone delimited by the dashed lines is estimated with (4.5), assuming perfect wetting (\( \theta_s = 0 \)) and using the upper and lower limits of the interfacial tension measured using the pendant drop method (see Section 5.6.3 and Section 5.7). The density properties used in (4.5) are listed in Table 5.1.

Use of the new infiltration criterion to obtain an infiltration height prediction

As discussed in Section 7.2.1.2, observations of 4-CT/water interfaces in the laboratory experiments showed that the static contact angle was different from 0. This suggests that the criterion (4.5) is not correct and that there exists another force, the so-called pinning force, which contributes to raise the value of the capillary tube entry pressure. The new infiltration criterion accounting for the pinning force contribution is given by (7.2). Using this equation, it can be shown that the infiltration height, \( h_i \), of a capillary tube of radius \( r_0 \), simulating a fracture and subjected to a centrifugal acceleration equal to \( n \) times the Earth’s gravity, is given by

\[
h_i = \frac{2\sigma \cos \theta_s}{\Delta \rho (ng)r_0} + \frac{2n_p f_p}{\Delta \rho (ng)r_0}.
\] (7.35)
Making use of the scaling laws (4.4), see also Table 4.1, (7.35) can be rewritten as

$$H_i = \frac{2\sigma \cos\theta_s}{\Delta \rho g r_0} + \frac{2n_{p,fp}}{\Delta \rho g r_0},$$  

(7.36)

where, again, $H_i$ is the infiltration height of the full-scale (1-g) prototype that is equivalent to the reduced-scale centrifuge model.

**Table 7.9. Summary of 4-CT Infiltration Centrifuge Experiments**

<table>
<thead>
<tr>
<th>Capillary Tube Diameter [mm]</th>
<th>g-Level Range of Tests</th>
<th>Measured Prototype DNAPL Pool Height at Infiltration</th>
<th>Theoretical Infiltration Height</th>
<th>Infiltration Height of Laboratory (1-g) Tests</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Average [mm]</td>
<td>SD [mm]&lt;sup&gt;b&lt;/sup&gt;</td>
<td>Max. Deviat. [mm]&lt;sup&gt;c&lt;/sup&gt;</td>
<td>No. of Tests</td>
</tr>
<tr>
<td>2.70</td>
<td>2.0-6.7</td>
<td>59.4</td>
<td>9.5</td>
<td>19.4</td>
</tr>
<tr>
<td>2.20</td>
<td>2.7-7.9</td>
<td>76.9</td>
<td>4.5</td>
<td>8.1</td>
</tr>
<tr>
<td>1.33</td>
<td>2.8</td>
<td>137.3</td>
<td>20.0</td>
<td>38.8</td>
</tr>
<tr>
<td>0.66</td>
<td>5.4</td>
<td>290.1</td>
<td>41.7</td>
<td>81.8</td>
</tr>
</tbody>
</table>

<sup>a</sup> Prototype pool at infiltration calculated as the product of the model pool height set in the reservoir tube prior to the experiment and the g-level for which the onset of infiltration is observed.

<sup>b</sup> Standard deviation. The number in parentheses is the coefficient of variation, i.e. the ratio of standard deviation to average.

<sup>c</sup> Maximum deviation between a measurement and the average for the series of tests. The number in parentheses is the maximum deviation relative to the average.

<sup>d</sup> Theoretical infiltration height calculated using (4.5) and taking densities listed in Table 5.1 and a value of $\sigma$ equal to $0.032 \pm 0.004$ N/m. Perfect wetting ($\theta_s = 0$) is assumed.

<sup>e</sup> Values reported in Table 7.1, and given here as a set $a \pm b$. The number, $a$, corresponds to the average infiltration height for the test series. The number, $b$, corresponds to the maximum deviation between a measurement and the average $a$.

In Section 7.2.1.2, pinning forces estimated using the height of first observed pre-infiltration were found to be radius dependent. The pinning forces, $n_{p,fp}$, were estimated—from first observed pre-infiltration—to be equal to $0.005$ N/m, $0.010$ N/m and $0.012$ N/m for 2.70 mm, 1.33 mm and 0.66 mm diameter capillary tubes, respectively. To obtain the graph shown in Figure 7.9, (7.2) was evaluated by using the pinning force values reported above. In addition, the 4-CT/water interfacial tension
was taken equal to 0.032 N/m, and the static contact angle was assumed to range between 30° to 60°. These same parameters are used in (7.36) to obtain a prediction for the prototype infiltration height of centrifuge experiments.

Figure 7.57. Measured 4-CT prototype pool height at the onset of infiltration for different capillary tube diameters. Discrete points represent experimental data from 71 infiltration centrifuge tests. Dashed lines represent prediction using (4.5) for different reduced interfacial tension, $\sigma$, and assuming perfect wetting ($\theta_s = 0$).

Figure 7.58 shows a plot of the measured 4-CT prototype pool height at the onset of infiltration versus the different capillary tube diameters. The discrete points represent the experimental data of the 70 infiltration centrifuge tests reported in Table 7.9. The prediction shown in the figure is obtained using (7.36). That is, the dashed zone represents the contribution to the capillary resistance attributable to the radius-dependent pinning force, i.e. $2n_p f_p/(\Delta \rho g r_0)$. The dotted lines $\theta_s = 30°$ and $\theta_s = 60°$ correspond, respectively, to the maximum and minimum capillary resistance attributable to interfacial tension, i.e. $2\sigma \cos \theta_s/(\Delta \rho g r_0)$. 

$\sigma = 36 \text{ mN/m} \quad 32 \text{ mN/m} \quad 28 \text{ mN/m}$
Figure 7.58. Measured 4-CT prototype pool height at the onset of infiltration for different capillary tube diameters. Discrete points represent experimental data for 70 infiltration centrifuge tests. Dashed/dotted lines represent predictions using a radius dependent pinning force, $n_{p}f_{p}$, ranging from 0.005 N/m to 0.012 N/m.

7.3.3.2 Discussion

Analysis of centrifuge experimental results

As shown in Figure 7.57 and Figure 7.58, a large scatter of the prototype infiltration height measurements for the centrifuge test series can be observed. Indeed, maximum relative deviations from the average infiltration height range from 10.6% to 32.7% depending on the tube diameter (see Table 7.9). Similar to laboratory (1-g) spontaneous infiltration tests (see Figure 7.1 or Figure 7.9), the scatter in the data is not due to outliers. Rather, as shown in Figure 7.57 and Figure 7.58, there is a continuous distribution of measured infiltration heights. For comparison with the centrifuge experimental results, the last column of Table 7.9 shows infiltration height measurements of 4-CT laboratory (1-g) spontaneous infiltration experiments, under the form $a \pm b$. The number $a$ is the average infiltration height for the test series on a given diameter. The number $b$ is the maximum deviation between a measurement and the
average measurement $a$. As can be seen, the average prototype infiltration heights obtained from the centrifuge tests are consistent with those obtained from the 1-g infiltration tests. Indeed, depending on the capillary tube diameter, the relative difference between average centrifuge and laboratory results varies between 0.1% and 13%. This observation suggests that the infiltration height is properly scaled by the factor $n$ in the centrifuge experiments. Nonetheless, as shown in Table 7.9 (see also Table 7.1), the relative deviations from the average is one to two times larger for centrifuge tests than for laboratory tests. This suggests that the results of centrifuge tests are associated with more scatter than the laboratory tests. Factors responsible for the variability of the prototype infiltration height are examined later in this section.

**Comparison of experimental results with predictions**

Figure 7.57 indicates a reasonable agreement between the measured and predicted values. However, it must be noted that the scatter of the experimental results is broader than the prediction band obtained from the infiltration criterion (4.5). Overall, as a first approximation, the infiltration criterion (4.5) where perfect wetting is assumed can provide the first significant figure of the maximum DNAPL pool height that can be sustained by a vertical capillary tube prior to its infiltration by DNAPL.

As discussed in Section 7.2.1.2, the criterion (4.5) is not believed to be correct, as it does not account for pinning forces that increase the capillary tube entry pressure. In addition, the DNAPL/water interface contact angle is observed to be different from zero. As shown in Figure 7.58, the infiltration criterion (7.36) provides a very good prediction of the experimental values of the prototype infiltration heights if the static contact angle is assumed to range between 30° and 60° and if the pinning force is assumed to increase from 0.005 N/m to 0.012 N/m depending on the radius of the capillary tubes. Again, note that these parameters are identical to those used to predict laboratory experiments, thereby suggesting that the entry pressure of the capillary tube, $(2\sigma \cos \theta_s + 2n_{fP})/r_0$ is more or less identical for a given capillary tube diameter in centrifuge and in laboratory (1-g) experiments.

**Correlation between g-level and prototype infiltration height**

There has been no correlation detected between the prototype DNAPL pool height and the g-level, suggesting that the DNAPL pool height at infiltration is properly scaled, and that (7.36) is valid over the range of g-levels used for this experimental program. Figure 7.59 shows a plot of the 4-CT prototype infiltration height of the centrifuge experiments versus the g-level at the onset of infiltration. As can be seen in the figure, for all four capillary tube diameters under consideration, there does not appear to be an increase or decrease in prototype infiltration height as the infiltration g-level increases.
Correlation between capillary tube model length and prototype infiltration height

Generally, the observed scatter does not correlate with individual tubes of similar length. That is, when two tubes of identical diameter and similar length are considered, it does not appear that one tube is systematically associated with prototype infiltration heights larger than the other’s. Nonetheless, it appears that the length of the capillary tube affects the observed prototype pool height. In other words, shorter capillary tubes are typically associated with smaller prototype infiltration heights than those of longer tubes, as is described below.

Table 7.10 summarizes the test series performed on 1.33 mm diameter capillary tubes. In the table, the average prototype infiltration height, $H_i$ is computed for each series of tests run on a capillary tube of a given model length, $l$ (i.e. the true physical length of the capillary tube, as opposed to the prototype length, $L = nl$). Note that each given tube length corresponds to one tube piece. In Figure 7.60, the prototype infiltration height data of these tests are plotted versus the model lengths of the tubes. In the figure, the experimental data are divided into four sub-series, each sub-series corresponding to a range of g-levels.

Figure 7.59. 4-CT prototype infiltration pool height versus centrifuge g-level at the onset of infiltration for different capillary tube diameters.
Table 7.10. Effect of Tube Model Length on the Prototype Infiltration Height of 1.33 mm Diameter Capillary Diameter Tubes

<table>
<thead>
<tr>
<th>Capillary Tube Model Length, l [mm]</th>
<th>Number of Tests</th>
<th>Average Prototype Infiltration Height, $H_i \pm$ Stand. Dev. [mm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>135</td>
<td>11</td>
<td>147.4 ± 16.9</td>
</tr>
<tr>
<td>129</td>
<td>6</td>
<td>150.9 ± 13.4</td>
</tr>
<tr>
<td>120</td>
<td>4</td>
<td>130.6 ± 14.2</td>
</tr>
<tr>
<td>60</td>
<td>5</td>
<td>128.0 ± 12.0</td>
</tr>
<tr>
<td>40</td>
<td>5</td>
<td>110.6 ± 6.7</td>
</tr>
</tbody>
</table>

As can be seen in Table 7.10, the average prototype infiltration height generally decreases as the model length of the capillary tube decreases. In Figure 7.60, the prototype infiltration height appears to decrease as the model length of the capillary tube length decreases, although a large scatter exists for the longest tubes. There is no obvious trend in infiltration height associated with the $g$-level. That is, a range of measured infiltration heights does not necessarily correspond to a particular range of $g$-levels, although it would have been suitable to test short tubes over ranges of small $g$-levels.

The differences in prototype height is not believed to be attributed to individual tube diameters, because the 60 mm long capillary tube and 40 mm long capillary tube were cut from the same initial capillary tube and that it would take a very unlikely difference in diameter of 0.2 mm to achieve the difference in average infiltration height reported in Table 7.10 for these two tubes (the difference 0.2 mm is obtained after making use of the relationship $H_i$(tube 1)/$H_i$(tube 2) = $r_0$(tube 2)/$r_0$(tube 1) derived from (4.5)). Furthermore, a similar correlation between model length and prototype infiltration height is also observed for 0.66 mm diameter capillary tubes (see later discussion in Section 7.3.6.1).

At present, the effect of model length on prototype infiltration height reported for centrifuge experiments is not well understood. Nonetheless, a qualitative explanation can be provided that might explain this effect. Consider two tubes of differing lengths spun at the same centrifugal acceleration, and assume that the NICCA model is correct. At a given interface model depth, the tube of shorter length is associated with an acceleration larger than that of the longer tube, since the NICCA model predicts that, for a given $g$-level, $n$, the same model velocity is achieved at the lower end of both capillary tubes (see (4.3) in Section 4.2, and recall that the velocity is solely dependent upon the relative depth, $z_c/l$, so long as $\Delta h \approx 0$). Assuming that a pre-infiltration/contact angle feedback mechanism similar to that described in Section 7.2.1.2 is present, it can be expected that the shorter tube’s interface, which is experiencing more acceleration, has more incentive for overcoming the pinning force and feedback mechanism than the longer tube. Thus, the DNAPL in the shorter capillary tube should be expected to infiltrate at a smaller pool height than the DNAPL in the longer tube.
Nevertheless, the mechanism described above remains uncertain. If indeed such a mechanism were correct, it should be also true for laboratory (1-g) spontaneous infiltration experiments. No strong trend, however, has been detected for laboratory experiments (e.g. see Table 7.2 and Table 7.3), except for the test series L9 run on the same 2.70 mm diameter tube pieces (see Table 7.2 and description in Section C.2.1). It is possible that the presence of long pre-infiltration depths for laboratory tests may complicate the impact of length on the DNAPL infiltration height.

![Figure 7.60. DNAPL prototype infiltration height versus tube model length for 1.33 mm diameter capillary tubes.](image)

The possibility of a bias in the experimental setup leading to prototype infiltration heights that are larger for longer tubes than for shorter tubes cannot be entirely excluded. For example, one could expect that longer tubes accelerated in a centrifugal field become a lot heavier than shorter tubes, and thus are subjected to more downward motion than shorter tubes. A relative movement between the capillary tube and the Teflon tape collar or reservoir tube (see Figure 6.10-Figure 6.12) could result in a
model pool height different from that measured prior to centrifugation. Nonetheless, tape recordings do not show that capillary tubes are moving during testing. Nor is there evidence of tube movement relative to the reservoir tube once the centrifuge box is removed from the centrifuge platform.

The correlation between infiltration height and tube model length and its consequences on the infiltration kinetics are further examined in Section 7.3.6.

**Causes for variability of infiltration height measurements**

Similar to laboratory tests, the scatter of the prototype infiltration heights can be attributed to the combined effects of pre-infiltration, contact angle variability and presence of pinning forces. In the prediction (7.36) plotted in Figure 7.58, the scatter has been assumed to be entirely attributable to the variability of the contact angle, which ranges between 30° and 60°, whereas the pinning forces are assumed constant for a given diameter, and equal to their estimated values from laboratory tests. Nonetheless, as discussed in Section 7.2.1.2 for laboratory tests, it is also possible that part of the data scatter is attributable to variability of the pinning force.

Pre-infiltration was observed in some tubes for tests that were interrupted in the middle of the centrifugation process, but prior to complete tube infiltration (see details of tests c18c-0.66ct-119mm and test c21-0.66ct-100mm in Section C.5.4). Pre-infiltration was not visible in the recorded images of the centrifuge test, and thus could not be measured. Therefore, the prototype heights measured for the tests and reported in Table 7.9 and Figure 7.57-Figure 7.60 only account for the height of DNAPL inside the reservoir tube and probably underestimate the total prototype pool height. Pre-infiltration, however, is believed to be limited for the centrifuge tests. Unlike laboratory tests, where the DNAPL pool height increases by increments, the prototype pool height continuously increases during a centrifuge test, thereby leading to complete infiltration once attempts to pre-infiltrate begins. Furthermore, because the Teflon tape collar blocks clear visualization of the capillary tube along its first twenty millimeters, infiltration cannot be detected until the interface has reached this depth. Therefore, assuming that the interface displacement at the early stages of the infiltration is very slow, the true g-level at infiltration may in fact be less that the g-level at which infiltration is detected. Overall, it is believed that the total model height (which includes the pre-infiltration depth) is systematically underestimated while the g-level is systematically overestimated, such that both errors are compensated for in the final measurement of the prototype height. There is obviously room for improving the experimental setup. In particular, a technique for capturing closer-range images of the DNAPL/water interface, most particularly at its early stages, is desirable for better characterizing the interface behavior and the effects of pre-infiltration during centrifuge testing.

Finally, it must be pointed out that the DNAPL model pool heights of centrifuge infiltration experiments were set and measured with an optical caliper (see Section 6.3.3). This optical caliper was also used for measuring the pool heights of laboratory (1-g) infiltration experiments. The precision of the measurements was identical for both laboratory and centrifuge experiments, and varied between 0.5 mm and 1 mm
depending on the flatness and shape of the top surface of the DNAPL pool. Therefore, considering, for example, a centrifuge experiment run at a g-level equal to 10, the model pool height is expected to be approximately ten times smaller than the equivalent prototype pool height. Nevertheless, the model pool height is measured with the same precision as the pool height of the equivalent prototype 1-g experiment. Therefore, the relative precision is ten times larger for the centrifuge experiment than for the laboratory experiment. This is most certainly why the scatter of prototype infiltration height data reported in Table 7.9 is observed to be larger than that of laboratory experiments.

7.3.4 Kinetics of Infiltration: Results and Comparison with NICCA Model and MFDV

7.3.4.1 Summary of Results on 0.66 mm Diameter Capillary Tubes

Infiltration profiles

Modeling-of-models presented in Section 7.3.2 has suggested that the centrifuge scaling relationships described in Table 4.1 can be used to convert measurements of the infiltration kinetics made during a centrifuge test to measurements in the equivalent prototype, provided that $\alpha_n$ is negligible with respect to unity for the conditions of the test. In this section, experimental data obtained for infiltration centrifuge experiments performed on 0.66 mm diameter capillary tubes are presented. For all those experiments, $\alpha_n$ is negligible compared to unity, so that inertia effects are not believed to have any measurable effect. This point is further discussed in Section 7.3.4.3.

The displacement of the DNAPL/water interface was monitored for twelve of the centrifuge infiltration experiments on 0.66 mm diameter capillary tubes reported in Table 7.9. A summary of the results for these twelve tests is given in Table 7.11. Figure 7.61 shows two plots of the prototype interface depth as a function of the prototype time for the test series. For some of the early tests reported here, it was not possible to successfully obtain the entire infiltration profile from the test recording. Thus, the lower part of the infiltration profile of those tests is missing.

Profiles predicted by the scaled NICCA model (4.6) (see Section 4.2) for each equivalent prototype are also plotted for comparison. Similarly to the comparison made between the NICCA model and the experimental data of spontaneous infiltration laboratory tests (see Section 7.2.2.2), a value of $\Delta H = 0.01 H_t$ is assumed to obtain the prediction, where $H_t$ is the theoretical prototype infiltration height reported in Table 7.9. Note that the average measured prototype infiltration height for a given tube diameter, or alternatively the specific prototype infiltration height of the test being predicted, could have been used in (4.6), instead of the theoretical value of $H_t$, in order to calculate $\Delta H$. Nonetheless, this would have led to little change in the prediction, as
most of the effect of $\Delta H$ would have been a small translation of the prediction along the time axis (see discussion of Section 3.5.2.2 and Figure 3.7). Similar to laboratory (1-g) infiltration data, centrifuge experimental data are translated along the time axis to match the prediction with the exit point or lowest available data point.

Table 7.11. Summary of Kinetics of 4-CT Infiltration Centrifuge Experiments into 0.66 mm Diameter Capillary Tubes at Varying g-Levels

<table>
<thead>
<tr>
<th>Test Reference Number</th>
<th>g-Level at the Onset of Infiltration into the Cap. Tube</th>
<th>Cap. Tube Prototype (Model) Length, $l$ [mm]</th>
<th>Prototype Pool Height at Onset of Infiltration, $H_i$ [mm]</th>
<th>$\alpha_n$-Number$^a$</th>
<th>Prototype Mid-Depth Velocity ($Z_c = L/2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>c20a-0.66ct-119mm</td>
<td>13.9</td>
<td>1651</td>
<td>278.9</td>
<td>$6.09 \times 10^{-2}$</td>
<td>4.8</td>
</tr>
<tr>
<td>c19a-0.66ct-119mm</td>
<td>10.8</td>
<td>1285</td>
<td>293.8</td>
<td>$4.74 \times 10^{-2}$</td>
<td>4.2</td>
</tr>
<tr>
<td>c7-0.66ct-137mm</td>
<td>10.4</td>
<td>1426</td>
<td>324.7</td>
<td>$3.97 \times 10^{-2}$</td>
<td>5.1</td>
</tr>
<tr>
<td>c7-0.66f-126mm</td>
<td>10.1</td>
<td>1277</td>
<td>309.0</td>
<td>$4.20 \times 10^{-2}$</td>
<td>4.5</td>
</tr>
<tr>
<td>c19b-0.66ct-119mm</td>
<td>10.1</td>
<td>1198</td>
<td>260.7</td>
<td>$4.42 \times 10^{-2}$</td>
<td>3.9</td>
</tr>
<tr>
<td>c18a-0.66ct-119mm</td>
<td>9.2</td>
<td>1100</td>
<td>234.9</td>
<td>$4.06 \times 10^{-2}$</td>
<td>3.7</td>
</tr>
<tr>
<td>c18b-0.66ct-119mm</td>
<td>8.2</td>
<td>976</td>
<td>208.2</td>
<td>$3.60 \times 10^{-2}$</td>
<td>3.5</td>
</tr>
<tr>
<td>c10-0.66st-126mm</td>
<td>7.6</td>
<td>958</td>
<td>322.3</td>
<td>$3.15 \times 10^{-2}$</td>
<td>4.6</td>
</tr>
<tr>
<td>c11-0.66ct-137mm</td>
<td>7.5</td>
<td>1023</td>
<td>318.8</td>
<td>$2.85 \times 10^{-2}$</td>
<td>4.1</td>
</tr>
<tr>
<td>c9-0.66ct-137mm</td>
<td>6.8</td>
<td>930</td>
<td>339.3</td>
<td>$2.59 \times 10^{-2}$</td>
<td>4.6</td>
</tr>
<tr>
<td>c11-0.66st-126mm</td>
<td>5.7</td>
<td>714</td>
<td>317.7</td>
<td>$2.35 \times 10^{-2}$</td>
<td>5.2</td>
</tr>
<tr>
<td>c21-0.66ct-100mm</td>
<td>5.4</td>
<td>535</td>
<td>261.2</td>
<td>$2.80 \times 10^{-2}$</td>
<td>3.9</td>
</tr>
</tbody>
</table>

$^a$ $\alpha_n$-number calculated using (4.2) with $k = r_0^2/8$.

$^b$ Theoretical values based on (4.6) (with $\Delta H = 0$). The theoretical prototype exit velocity $K_\lambda$ (see (4.7) with $k = r_0^2/8$) is equal to 9.6 mm/s for 0.66 mm diameter capillary tubes.
Figure 7.61. Prototype DNAPL/water interface displacement as a function of prototype time for centrifuge experiments on 0.66 mm diameter capillary tubes, and associated NICCA model predictions (4.6): a. Small g-levels; b. Large g-levels.
Velocity fields

Figure 7.62 shows a plot of the interface relative depth \( \frac{Z_c}{L} \) versus the prototype interface velocity, \( \frac{dZ_c}{dT} \), for the tests series reported in Table 7.11. A three point averaging technique (see Section 7.2.2.3) is applied whenever necessary. The NICCA model prediction (4.6) is also reported for the equivalent prototype. Note that the line (4.6) shown in the figure is evaluated assuming \( \Delta H \approx 0 \), as was done for the velocity fields of 1-g spontaneous infiltration experiments (see Section 7.2.2.3). As discussed in Section 7.2.2.3, if the NICCA model is correct, all experimental data run on a capillary tube of a given diameter should fall on the same line of slope \( K_\Delta \), regardless of the tube prototype length, as predicted by (3.49).

For comparison, the MFDV is also examined and plotted in Figure 7.62. Again, in the MFDV, the viscous force is allowed to decrease as infiltration progresses in the tube, in order to take into account the effects of the viscous contrast between 4-CT and water (see Section 3.5.4). In Chapter 4, physical modeling of the infiltration kinetics using the centrifuge scaling laws was restricted to the NICCA model where the viscosity contrast was assumed to be negligible in comparison to the viscosity of water. Nonetheless, if an infiltration experiment in a centrifugal acceleration field of \( ng \) is considered, the scaling laws given in Table 4.1 can be substituted into the MFDV model equations (3.76)-(3.78) (see Section 3.5.4), using an approach similar to that used for the NICCA model (see Section 4.2). Following this method, (3.76)-(3.78) can be rewritten as

\[
\frac{dZ_c}{dT} = K_{\Delta\Delta} \frac{Z_c(T) + \Delta H}{Z_c(T) + H_{\mu}}, \tag{7.37}
\]

where

\[
K_{\Delta\Delta} = \frac{\Delta p g r_0^2}{8\Delta \mu}, \tag{7.38}
\]

\[
H_{\mu} = \frac{\mu_{nw} L}{\Delta \mu}. \tag{7.39}
\]

In (7.37), \( T, \frac{dZ_c}{dT} \) and \( Z_c(T) \) are the prototype time, prototype interface velocity and prototype interface depth, respectively. In (7.39), \( L = nl \) is the prototype length of the capillary tube. Note that the effects of pre-infiltration are neglected here. Note also that, in the case of the pair 4-CT/water, the viscosity contrast \( \Delta \mu = \mu_{nw} - \mu_n \) in (7.39) is a negative constant (\( \mu_{nw} < \mu_n \)).

Similar to the NICCA model, the prototype interface velocity of the MFDV can be shown to be solely dependent on the interface relative depth, \( \frac{Z_c}{L} = \frac{z_c}{l} \). Assuming that \( \Delta H \approx 0 \), and making use of the expression of \( K_\Delta \) (see (4.7)), (7.37) leads to
\[
\frac{dZ_c}{dT} = K_\Delta \frac{Z_c(T)/L}{1 + \frac{\Delta \mu}{\mu_w} [Z_c(T)/L]}.
\]  
\hspace{1cm} (7.40)

In particular, the MFDV (7.40) predicts that the prototype exit velocity, \(dZ_c/dT(Z_c = L)\), is independent of the prototype length of the capillary tube, and equal to \(K_\Delta/(1 + \Delta \mu/\mu_w) = 9.6 \text{ mm/s}/[1 + (0.912 - 1)] = 10.5 \text{ mm/s}\) in the case of 0.66 mm diameter capillary tubes.

The MFDV (7.40) is also plotted in Figure 7.62 (dashed line). As previously mentioned in Section 7.2.2.5 for 1-g tests, the interface velocity of the MFDV is not a linear function of the interface depth, as opposed to the NICCA model case where the relationship is linear.

Figure 7.62. Interface relative depth versus prototype interface velocity for centrifuge tests on 0.66 mm diameter capillary tubes and comparison with predictions of NICCA model (4.6) with \(\Delta H = 0\) (solid line) and MFDV (7.40) (dashed line).
7.3.4.2 Discussion of Results and Comparison with Models

Experimental data

Examining the velocity field data shown in Figure 7.62, it can be seen that, overall, the prototype velocity field is quite repeatable between centrifuge tests, even though these tests are run at different g-levels on capillary tubes of different model lengths. Specifically, for any given prototype interface depth, the prototype velocity is known with a precision of the order of ±1 mm/s. While this may seem rather large in comparison to the prototype velocities, which range from 2 mm/s to 10 mm/s, one must bear in mind that these results are obtained after scaling the experimental data using the scaling laws of Table 4.1. That is, the prototype velocity is obtained after dividing the measured model velocity by the g-level, $n$. Hence, the relative uncertainty on the prototype data is the sum of the relative uncertainty on the model velocity and the relative uncertainty on the value of $n$.

Generally, it is observed that the prototype interface velocity increases as the prototype depth increases. Figure 7.62 suggests that the increase is almost linear. However, a closer look at individual tests demonstrates otherwise. Figure 7.63 shows a plot of the velocity fields of three selected centrifuge tests, namely test c21-0.66ct-100mm ($n = 5.4$), test c19b-0.66ct-119mm ($n = 10.1$), and test c20a-0.66ct-119mm ($n = 13.9$) (see Table 7.11). As can be seen in the figure, the experimental velocity fields appear curved. That is, rather than being constant (linear relationship), the rate of increase of interface velocity with depth increases as the depth increases.

Comparison of experimental data with model prediction

Referring now to the infiltration profiles shown in Figure 7.61, agreement between the NICCA model and the experimental data is good. For some tests, however, the agreement is only good beyond a certain interface depth. This is the case whether the $g$-level (and associated capillary tube length) is small (Figure 7.61.a) or large (Figure 7.61.b).

Referring now to Figure 7.62, the experimental data are in reasonable agreement with the NICCA velocity field line (4.6), although this line generally lies to the right of the experimental velocity fields. At a given relative depth, the difference between prediction and observation is of the order of 1 mm/s. This suggests, as in the case of laboratory (1-$g$) experiments, that the displacement is somewhat slower than predicted, particularly at the early stages of the interface displacement.

Despite the offset between prediction and experimental data, the overall reasonable agreement between the two suggests that, for 0.66 mm diameter capillary tubes, the experimental prototype interface velocity is proportional to the relative depth, $Z_c/L$, as predicted by the NICCA model, and thus that the observed exit prototype velocity of the interface is close to $K_\Delta$, a constant given by (4.7) (with $k = r_0^2/8$), which depends on the capillary tube radius, but does not depend on the prototype length of the tube.
Upon comparing the experimental data with the MFDV (7.40), which takes into account the decrease of the viscous resisting force, it can be seen that the experimental velocity field is generally slower than the MFDV predicted field throughout the entire length of the capillary tube. One important feature shown in Figure 7.63 is that the non-linear, crescent-like shape of the experimental velocity field is qualitatively consistent with the MFDV prediction. That is, the increase of the rate of interface velocity change with depth can be attributed to the decrease of the viscous force in the capillary tube. It must be noted that the curvature of the experimental data does seem to vary depending on the test, and is also somewhat different from the curvature of the MFDV. However, again, the scaled experimental results are sensitive to the g-level measurement, \( n \), so that an error on the measurement of \( n \) is passed along to the prototype velocities, and thus may distort the curvature of the prototype velocity field. Furthermore, the g-level in the centrifugal field increases with capillary tube depth, which thus will increase the infiltration rate with depth (see discussion in Section B.3.2 on the g-level variations across a capillary tube).

![Prototype Interface Velocity, \( dZ_c/dT \) [mm/s]](image)

**Figure 7.63.** Prototype velocity fields of three centrifuge tests on 0.66 mm diameter capillary tubes, and evidence of non-linearity between measured interface velocity and interface depth.
7.3.4.3 Interpretation of Results and Rationale

Figure 7.62 showed that the experimental velocity fields were found to be generally smaller than those predicted by the NICCA model and the MFDV. As for laboratory tests, this implies the presence of a resisting force unaccounted for in both the NICCA model and the MFDV. This resisting force slows down the interface displacement velocities. Potential resisting effects neglected by the NICCA model are discussed below under separate headings.

**Effect of DNAPL drainage from reservoir tube**

The effect of reservoir drainage will not be examined here in detail (see discussion for 1-g tests in Section 7.2.2.5). Similar to laboratory experiments, the cross-section of the reservoir tube is much larger than that of the capillary tubes (see Section C.5.4), so that the effect of head reduction at the entrance to the capillary tube is believed to be unimportant.

**Effect of inertia forces**

Table 7.11 provides the $\alpha_n$–numbers (see (4.2) in Section 4.2) computed for each centrifuge experiment. Again, an $\alpha_n$–number very small with respect to unity indicates that the effects of inertia forces are not expected to be measurable and can be neglected for practical purposes. As can be seen in Table 7.11, the $\alpha_n$–numbers range between $2.35 \times 10^{-2}$ and $6.09 \times 10^{-2}$. In Section 7.2.3.7, it was concluded that the effects of inertia could be measurable for $\alpha_t$-numbers as small as 0.05. Thus, it is possible that, for some tests, inertia may have had some measurable effect on the experimental displacement profiles and velocity fields, although it is believed that the impact of the inertia forces at such low $\alpha_t$-numbers would only lead to a fraction of the velocity reduction reported here. The effect of inertia forces in the centrifuge tests is further addressed in Section 7.3.6, and shown to be unimportant for 0.66 mm diameter capillary tubes.

**Effect of capillary resistance**

Capillary resistance in centrifuge reduced-scale models behaves in a similar fashion to that of laboratory tests. In both cases, the capillary resistance, i.e. the combination of the interfacial tension forces and the pinning forces, increases from its static value because the dynamic contact angle decreases with increasing velocity, and thus contributes to raising the magnitude of the interfacial tension forces. The increase in the interfacial tension force, in combination with the pinning force, acts, in turn, to slow down the interface velocity.
In Section 4.4, it was pointed out that the dependence of dynamic contact angle upon velocity—in the context of centrifuge tests—led to complications. This was because correct scaling of capillary forces in the centrifuge required that the dynamic contact angle-interface velocity relationship be scaled by a factor \( n \). Focusing specifically on the NIVCA model infiltration kinetics, it was shown in Section 4.4 that, for the retardation factor \( \gamma \) (see definition Section 3.5.3.2) to be equal in the reduced-scale model and in the full-scale prototype, the perfect wetting velocity, \( u_c \) (for which \( \theta_d = 0 \)) had to be \( n \) times larger in the model than in the prototype (see scaled NIVCA model equation (4.20)).

On the one hand, the perfect wetting velocity, \( u_c \), and more generally the dynamic contact angle-interface velocity relationship, could be seen as a unique property for a given DNAPL/water pair. In particular, \( u_c \) and the contact angle-velocity relationship would be independent of the system geometry (e.g. tube diameter) and the effects of gravity (e.g. tube orientation or centrifugal field). Under this assumption, for any centrifuge test, the value of \( u_c \) would remain a constant equal to 3 mm/s, as estimated for 1-g infiltration tests (see Section 7.2.3.3), since the same 4-CT/water pair is used for these tests. Thus, the perfect wetting velocity would not scale by the factor \( n \). For example, at a \( g \)-level \( n = 10 \), perfect wetting would be reached at a model velocity of \( u_c = 3 \) mm/s, corresponding to a prototype velocity of 0.3 mm/s.

On the other hand, it is possible that \( u_c \), and more generally the dynamic contact angle-interface velocity relationship, are affected by the system geometry and the effects of gravity. Correct scaling of \( u_c \) would require, for example, that at a \( g \)-level equal to 10, the 4-CT/water pair achieves perfect wetting at a model velocity of 30 mm/s, so that the corresponding prototype velocity would be 3 mm/s.

Overall, it is possible that the 4-CT/water perfect wetting velocity is not a unique liquid pair property (recall discussion on contact angle-velocity relationship in Section 2.3.5.3), so that it may be affected by gravity. Nonetheless, it seems unlikely that \( u_c \) would be linearly related to the \( g \)-level. Most certainly, centrifugal acceleration has some effect, so that the magnitude of \( u_c \) varies, but remains of the order of 3 mm/s.

Considering the experimental data plotted in Figure 7.62, it can be seen that the smallest reported prototype velocity is of the order of 1.8 mm/s at \( n = 5.4 \). This corresponds to a model velocity of approximately 10 mm/s, a number larger than the estimated 4-CT/water perfect wetting velocity of \( u_c = 3 \) mm/s. Therefore, for all the experimental data plotted in Figure 7.62, it can be expected that the 4-CT/water interfaces are already in a configuration of perfect wetting. In other words, any change in contact angle as a function of velocity is believed to be limited to the very early stages of the tube infiltration, and not captured in Figure 7.62. Indeed, Figure 7.62 does not show the capillary resistance increase effect, thereby supporting the assumption that all reported experimental data are already in the perfect wetting mode. This will be well illustrated in the figures presented in Section 7.3.5.2.

That the experimental data fall in the perfect wetting regime is of importance, as it can be shown that these data can be scaled successfully using the scaling laws of Table 4.1 and also be predicted by the NIVCA model. This is the object of the next section.
7.3.5 Kinetics of Infiltration: Results and Comparison with the NIVCA Model

7.3.5.1 Proof that the Asymptotic Portion of the NIVCA Model Can Be Scaled Using the Centrifuge Scaling Laws

Recall the NIVCA model equation (7.15) in the case $\frac{dz_c}{dt} > u_c$ (i.e. perfect wetting)

$$\frac{dz_c}{dt} = \frac{k_\lambda}{l} \left[ z_c(t) + h - \frac{2\sigma}{\Delta \rho g r_0} - \frac{2n_p f_p}{\Delta \rho g r_0} \right], \quad (7.41)$$

where the pre-infiltration, $h_{pi}$, is assumed equal to 0. Consider now an infiltration experiment in a centrifugal acceleration field equal to $n$ times the Earth’s gravity. Equation (7.41) can be used to describe the interface motion, $z_c(t)$, in such a gravity field. This equation can be rewritten as

$$\frac{dz_c}{dt} = \frac{\Delta \rho (ng) r_0^2}{8\mu_w l} \left[ z_c(t) + h - \frac{2\sigma}{\Delta \rho (ng) r_0} - \frac{2n_p f_p}{\Delta \rho (ng) r_0} \right], \quad (7.42)$$

where use of the definition of $k_\lambda$ (see (3.43)) has been made. Multiplying either side of (7.42) by $1/n$, and making use of the scaling laws of Table 4.1, (7.42) leads to

$$\frac{dZ_c}{dT} = \frac{\Delta \rho g r_0^2}{8\mu_w L} \left[ Z_c(T) + H - \frac{2\sigma}{\Delta \rho g r_0} - \frac{2n_p f_p}{\Delta \rho g r_0} \right], \quad (7.43)$$

which can be rewritten as

$$\frac{dZ_c}{dT} = \frac{K_\Lambda}{L} \left[ Z_c(T) + \Delta H' \right], \quad (7.44)$$

where $K_\Lambda$ is defined as per (4.7), and $\Delta H'$ is given by

$$\Delta H' = H - \frac{2\sigma}{\Delta \rho g r_0} - \frac{2n_p f_p}{\Delta \rho g r_0}. \quad (7.45)$$

Equation (7.44) shows that, so long as the interface velocities are restricted to the portion $\frac{dz_c}{dt} > u_c$ (or $\frac{dZ_c}{dT} > u_c/n$), then the DNAPL infiltration velocities in the reduced-scale centrifuge model—described by (7.42)—are equivalent to those in the
full-scale prototype—described by (7.44). This is because the prototype velocity field line given by (7.44) is an asymptotic line independent of the contact angle.

Consider a series of centrifuge tests run at different g-levels, $n$, on capillary tubes of different lengths, $l$, such that the prototype length, $L = nl$, remains constant. If the NIVCA model is correct then, passed the perfect wetting velocity, all prototype velocity fields of this test series should fall on the line given by (7.44), independently of the magnitude of the g-level. In fact, the g-level only determines the value of the prototype perfect wetting velocity, and thus at which point the data should start falling on the asymptotic line (7.44).

### 7.3.5.2 Comparison of NIVCA Model Velocity Field Asymptotic Line With the Experimental Prototype Velocity Field

As concluded in Section 7.3.4.3, all the prototype experimental data reported in Figure 7.62 are in the perfect wetting domain ($dZ_c/dT > u_c/n$). Thus, if the NIVCA model is correct, (7.44) predicts that the prototype velocity-depth data of a given centrifuge test should fall on a unique line that is parallel to the NICCA model line, and has the same slope $K_s/L$. The distance between the NIVCA asymptotic line and the NICCA line is determined by the constant $\Delta H'$ given by (7.45). Note that unlike the NICCA model and the MFDV (see (7.40)), the prototype interface velocity predicted by the NIVCA model is not solely dependent on the interface relative depth $Z_c/L$. Again, this property is true for the NICCA model and the MFDV because $\Delta H$ is negligible with respect to $Z_c$, and taken equal to zero. In the case of the NIVCA model, the constant $\Delta H'$ is different from zero, and not negligible with respect to $Z_c$, so that $\Delta H'/L$ takes different values for different capillary tube prototype lengths. Thus, the NIVCA model prediction of the interface velocity, $dZ_c/dT$, as a function of the relative depth, $Z_c/L$, would not be a unique line if different capillary tube prototype lengths were considered. This conclusion is important, as it demonstrates that, if the NIVCA model is correct, the experimental velocity infiltration data shown in Figure 7.62 are not expected to fall on a unique line, but instead are dependent on the quantity $\Delta H'/L$, $\Delta H'$ being constant.

Figure 7.64.a though Figure 7.64.f show the velocity fields of a selection of six centrifuge experiments reported in Table 7.11 (for these six tests, reasonably long series of velocity data points are available). In the figures, the symbols represent the experimental data points, and are identical to those shown in Figure 7.62. The solid and dashed lines are the NIVCA model asymptotic predictions calculated using (7.44). For comparison, the NICCA model asymptotic predictions calculated using (4.6) are also plotted in the figures (dotted line). The vertical dashed/dotted line shown in all the figures corresponds to the prototype perfect velocity, $U_c = 3$ mm/s, that would be required for correct scaling of the centrifuge experiment with the equivalent full-scale prototype (see discussion in Section 7.3.4.3).
Prototype Interface Velocity, $dZ_c/dT$ [mm/s]

(a)

Prototype Interface Velocity, $dZ_c/dT$ [mm/s]

(b)
Prototype Interface Velocity, $dZ_c/dT$ [mm/s]

Depth of Prototype Interface, $Z_c$ [mm]

(c)

Prototype Interface Velocity, $dZ_c/dT$ [mm/s]

Depth of Prototype Interface, $Z_c$ [mm]

(d)
Figure 7.64. a. through f. Prototype velocity fields of 0.66 mm diameter centrifuge experiments at different g-levels, $n$. The solid and dashed lines are the asymptotic predictions obtained from the NIVCA model (7.44), whereas the dotted line are the predictions obtained from the NICCA model (4.6).
For the NICCA model predictions shown in Figure 7.64.a though Figure 7.64.f, it is again assumed that $\Delta H = 0$. For the NIVCA asymptotic predictions shown in the figures, it is necessary to calculate $\Delta H'$ as given by (7.45). In (7.45), it is assumed that $H \approx H_i$. The height $H_i$ used in the equation is the prototype infiltration height measured for the centrifuge experiment that is being predicted. Furthermore, two values of the pinning force are considered here. The solid line is obtained when the pinning force, $n_{\rho f_p}$, is taken equal to 0.012 N/m. This value corresponds to the estimated value back-calculated from first observed pre-infiltration of 1-g spontaneous infiltration tests into 0.66 mm diameter capillary tubes (see Section 7.2.1.2 and Section 7.2.3.3). In addition, a NIVCA model prediction using a value of $n_{\rho f_p}$ equal to 0.006 N/m is considered (dashed lines). This latter value corresponds to the pinning force that provided a good NIVCA model prediction of the infiltration tests into the 1201 mm long, 0.66 mm diameter capillary tube (see Figure 7.42 and discussion in Section 7.2.3.3). In both NIVCA model predictions, the interfacial tension of 4-CT/water, $\sigma$, is taken equal to 0.032 N/m.

As can be seen in Figure 7.64.a though Figure 7.64.f, the experimental prototype velocity fields typically lie in the band delimited by the NIVCA model asymptotic velocity field with $n_{\rho f_p} = 0.012$ N/m and the NIVCA velocity field with $n_{\rho f_p} = 0.006$ N/m. This suggests that the magnitude of the pinning force range between these two values. Note that there is no observed correlation between the g-level and the pinning force that provides the best fit. Non-linearity of the experimental data is observed in most cases, and some of the data points corresponding to the lower portion of the capillary tube often lie on the right of the NICCA line. As mentioned in Section 7.3.4.2e, this phenomenon is attributed to the viscosity contrast between 4-CT and water, and increase of g-level with capillary tube depth.

In conclusion, the NIVCA model asymptotic prediction shows that the increase in interfacial tension force and the presence of the pinning force contribute to a velocity reduction from the NICCA model prediction by a factor that is consistent with the experimental observations. It is believed that the pinning force range between 0.012 N/m and 0.006 N/m, depending upon the centrifuge test. It should be noted, however, that other factors beside the pinning force might influence the location of the line where the experimental prototype velocity field lie. These parameters are:

1. The interfacial tension, whose magnitude has been shown to change with time (see discussion in Section 7.2.1.2). Hence, 4-CT/water interfaces of different ages can be associated with different interfacial tensions, which in turn would yield different prototype infiltration heights and values of $\Delta H'$ (see (7.45)).
2. The g-level, which directly influences the magnitude of $H_i$. Thus, an error on the measurement of the g-level affects the value of $\Delta H'$. Recall that an error on the g-level also distorts the experimental prototype velocity field.
3. Presence of pre-infiltration. As previously noted, pre-infiltration is believed to be negligible for the centrifuge tests. However, if pre-infiltration were present, this would mean that a given prototype depth reported in Figure 7.64.a through Figure 7.64.f would be less that is reported in the figure. Consequently, the experimental velocity field would be translated upwards and approach the NICCA model.
Comment on modeling of models and NIVCA model

In Section 7.3.2, modeling of models was performed to verify the validity of the scaling laws of Table 4.1. For 0.66 mm diameter capillary tubes, the experimental results of laboratory test L11-0.66lct-1221mm ($l = 1201$ mm) were compared to the experimental results of centrifuge tests c19a-0.66ct-119mm ($n = 10.8$) and c19b-0.66ct-119mm ($n = 10.1$).

Earlier in this chapter, comparison between the NIVCA model and 1-g test L11-0.66lct-1221mm was made. The reader is referred to Section 7.2.3.3 and Figure 7.42 (see also Figure 7.41). Comparison between the NIVCA model and centrifuge tests c19a-0.66ct-119mm and c19b-0.66ct-119mm was later made in Figure 7.64.e and Figure 7.64.d, respectively.

For the laboratory test and the two centrifuge tests, comparison with the NIVCA model showed that a pinning force of approximately 0.006 N/m appeared to provide the most accurate NIVCA model prediction (see figures referred to above). Nonetheless, when examining the velocity field of the 0.66 mm diameter tube modeling-of-models in Figure 7.54, an approximately constant offset of 1 mm/s was observed between the prototype velocity field of the laboratory test and the velocity fields of the two centrifuge tests. This velocity offset can also be characterized as a depth offset. Typically, for any given prototype velocity, the laboratory test interface depth is located 20 mm-40 mm above the centrifuge test prototype interface depth where the same prototype velocity is achieved.

In light of the NIVCA model, it can be shown that the offset between laboratory and centrifuge tests is attributable to the differences in prototype infiltration heights. For the laboratory tests, the infiltration height is found to be equal to 308 mm. For the centrifuge tests, the infiltration heights are found to be equal to 261 mm and 294 mm, respectively. Thus, according to the NIVCA model, if similar pinning forces are considered in all three cases, $\Delta H'$ (see (7.45) is expected to be larger for the laboratory test, as it is associated with the larger infiltration height. This difference in $\Delta H'$ leads, in turn, to differences in interface velocities, as the laboratory tests with the larger $\Delta H'$ is expected to have a larger velocity at a given infiltration depth $Z_c$. Also, note that the depth offset of 20 mm-40 mm is consistent with the difference in prototype infiltration height between the laboratory and centrifuge tests, which are equal to 14 mm and 47 mm, respectively.

While a valid qualitative argument, one issue remains unresolved here and would certainly require further testing to be clarified. Clearly, the difference in prototype infiltration height between the two centrifuge tests (294 mm – 261 mm = 33 mm) is larger than the difference in infiltration height between the laboratory test and one of the centrifuge tests (308 mm – 294 mm = 14 mm). Yet, the two centrifuge tests prototype velocity fields are in very close agreement (see Figure 7.54). This could mean that: (i) small experimental errors are present in the reported prototype infiltration heights, for example on the measurement of the g-level and/or the infiltration heights, or (ii) that other resisting effects are present. In particular, inertia could start having an
effect at the $\alpha_n$ values reported for the two centrifuge tests (about 0.045, see Section 7.3.2.4).

In the case of the modeling-of-models on 1.33 mm diameter capillary tubes (see Section 7.3.2.3), the prototype velocity field of centrifuge test c13-1.33st-120mm ($n = 4.8$) is observed to be less than the velocity field of laboratory test L6-1.33st-610mm. Nonetheless, the infiltration height of the laboratory test is $H_i = 130$ mm, which is less than the prototype infiltration height of the centrifuge test, $H_i = 143$ mm. In light of the results on the modeling of models on 0.66 mm diameter tubes, the centrifuge test prototype velocity field should be larger than the velocity field of the laboratory test, because of the larger centrifuge test prototype infiltration height. It is believed, however, that the opposite is observed because of the presence of inertia effects for the centrifuge test. At $\alpha_n = 0.342$, the inertia forces associated with the centrifuge test are expected to have a measurable effect, and act to slow down the infiltration velocity despite the larger infiltration height. Again, inertia has no effect on the laboratory test.

7.3.6 Kinetics of Infiltration: Summary Plots of Interface Velocities

7.3.6.1 Centrifuge Infiltration Tests Into 0.66 mm Diameter Capillary Tubes

To further examine the influence of the resisting forces on the interface velocity of centrifuge infiltration tests into 0.66 mm diameter capillary tubes, an approach similar to that used for laboratory (1-$g$) infiltration tests is adopted (see summary plot of Section 7.2.2.4). The prototype interface velocity at mid-depth of the capillary tube is taken as the velocity of reference for each test (see Table 7.11). This velocity is normalized by the mid-depth prototype interface velocity predicted by the NICCA model under assumption of negligible $\Delta H$. That is, the NICCA predicted velocity is equal to $K_{\alpha} / 2$. For a given centrifuge test, the measured mid-depth prototype velocity is obtained by linear interpolation between the two data points that are closest to $Z_c/L = 1/2$. Linear extrapolation is performed in some cases. Note that the ratio of measured prototype velocity to NICCA prototype velocity is equal to the ratio of measured model velocity to NICCA model velocity.

In Figure 7.65, the normalized mid-depth velocity is plotted versus different parameters measured for each centrifuge test: the $\alpha_n$-number (Figure 7.65.a), the $g$-level (Figure 7.65.b), the capillary tube model length, $l$ (Figure 7.65.c), the capillary tube prototype length, $L = nl$ (Figure 7.65.d), the DNAPL prototype infiltration height, $H_i$ (Figure 7.65.e). Finally, in Figure 7.65.f, the DNAPL prototype infiltration height is plotted versus the capillary tube model length. All the parameters are reported in Table 7.11.
\( \alpha_n \)-Number, \( \alpha_n = \Delta \rho v (ng)_{04}^4/(16\mu_w^2 l) \)

g-Level, \( n \)

(a) 

(b) 

(c) 

(d) 

Capillary Tube Model Length, \( I \) [mm] 

Capillary Tube Prototype Length, \( L = nl \) [mm]
Figure 7.65. Ratio of observed velocity to velocity predicted by the NICCA model at the mid-depth ($Z_c/L = 1/2$) of 0.66 mm diameter capillary tubes: a. Versus $\alpha_n$-number; b. Versus $g$-level; c. Versus capillary tube model length; d. Versus capillary tube prototype length; e. Versus DNAPL prototype pool height at infiltration; f. DNAPL prototype pool height at infiltration versus capillary tube model length.
As shown in Figure 7.65.a, there appears to be no correlation between the $\alpha_n$-number and the normalized mid-depth velocity. This suggests that the inertia forces associated with the $\alpha_n$-numbers—ranging here from 0.023 to 0.061—do not have a measurable effect on the interface displacement velocities of centrifuge tests into 0.66 mm diameter capillary tubes. Recall, however, that inertia forces appeared to have an effect on the laboratory experiment infiltration velocities for $\alpha_t$-numbers as low as 0.05 (see Section 7.2.3.7). Thus, it is possible that inertia forces may have some effect on the centrifuge experiment displacement velocities reported here, but that no trend is visible in Figure 7.65.a because the range of $\alpha_n$ is too small.

Referring now to Figure 7.65.b, no strong correlation can be seen between the normalized velocity and the $g$-level, either. Recall that the larger the $g$-level, the larger the $\alpha_n$-number (see (4.2) in Section 4.2).

While no clear trend can be derived from Figure 7.65.d for the prototype length, $L$, a small increasing trend can be seen in Figure 7.65.c. This trend suggests that, for shorter tube lengths, $l$, the measured mid-depth velocity is over-predicted by the NICCA model, and that the difference between model and measurement increases as the capillary tube model length, $l$, decreases.

Perhaps the most obvious trend is that shown in Figure 7.65.e, where, clearly, an increase in infiltration height leads to an increase of the normalized mid-depth velocity. This supports the capillary resistance mechanism proposed by the NIVCA model. Indeed, the smaller the infiltration height $H_i$, the larger the absolute value of the constant $\Delta H'$ (see (7.45) and recall that $\Delta H'$ is negative), and thus the smaller the prototype interface velocity given by (7.44). The influence of $H_i$ is identical to that discussed in Section 7.3.5.2 when revisiting the modeling of models on 0.66 mm diameter capillary tubes.

In the mechanism proposed above, it is important to note that capillary forces are only indirectly responsible for the correlation between mid-depth velocity and infiltration height. Indeed, infiltration takes place when the driving buoyant pressure, characterized by $H$, reaches the entry pressure of the capillary tube characterized by $H_i$ (so that $H \approx H_i$). During the infiltration process, the entry pressure is increased as a consequence of the decrease of contact angle and associated increase of interfacial tension, so that the difference $H - H_i(dZ_c/dT)$ becomes negative and acts to reduce the magnitude of the infiltration velocity. Consequently, if the magnitude of the driving buoyant pressure at infiltration, $H$ (equal to infiltration height, $H_i$) is noted to be larger than average, then the velocity reduction is smaller than average, and vice versa. Thus, it is the buoyant pressure rather than the capillary force that is directly influencing the interface velocity.

The correlation existing between the tube model length and the normalized velocity shown in Figure 7.65.c is believed to be a consequence of the correlation between infiltration height, $H_i$, and normalized velocity. As discussed in Section 7.3.3.2, capillary tubes of shorter model lengths are typically associated with smaller infiltration heights than tubes of longer lengths. This trend is well shown in Figure 7.65.f. Thus, a shorter tube for which infiltration takes place at a smaller pool height will be associated with a smaller normalized velocity, as illustrated in Figure 7.65.c.
It should be noted that the prototype mid-depth interface velocity predicted by the NIVCA model is given by

$$\frac{dZ_c}{dT}(L/2) = \frac{K_s}{2} + \frac{K_n \Delta H'}{L}. \tag{7.46}$$

Therefore, the ratio of NIVCA mid-depth velocity to NICCA mid-depth velocity, $R_{V/I/C}(L/2)$ can be written as

$$R_{V/I/C}(L/2) = 1 + \frac{2\Delta H'}{L}. \tag{7.47}$$

Because $\Delta H'$ is negative, the ratio $R_{V/I/C}(L/2)$ should decrease with decreasing prototype length, $L$, provided that $\Delta H'$ remains a constant. Thus, as $L$ increases, the NIVCA mid-depth $dZ_c/dT(L/2)$ tends to the NICCA mid-depth velocity $K_s/2$. Such a mechanism is believed to be responsible for the velocity reduction associated with 1-g infiltration tests into 1.33 mm diameter capillary tubes (see discussion in Section 7.2.3.4 and Figure 7.43).

Nonetheless, there is no visible correlation between mid-depth normalized velocity and prototype length in Figure 7.65.d. This observation is in apparent contradiction with (7.46). It is believed that the range of prototype lengths, from 500 mm to about 1700 mm (i.e. three-fold increase versus eight-fold increase for laboratory tests) may be too narrow to show a clear trend. Furthermore, the dependence of the normalized velocity upon the infiltration height may largely overshadow its dependence upon prototype length. Finally, the potential dependence upon inertia forces of interface velocities of tests for which the $\alpha_n$-number approaches or exceeds 0.05 may also complicate the correlation between normalized velocity and prototype length.

### 7.3.6.2 Centrifuge Infiltration Tests Into 1.33 mm and 2.70 mm Diameter Capillary Tubes

To confirm the trends inferred from centrifuge infiltration experiments run on 0.66 mm diameter capillary tubes, results obtained for centrifuge 4-CT infiltration experiments into 1.33 mm and 2.70 mm diameter capillary tubes are now examined. Again, the prototype interface velocity at mid-depth of the capillary tube is adopted as the velocity of reference for each test. This velocity is normalized by the mid-depth prototype interface velocity predicted by the NICCA model under assumption of negligible $\Delta H$, i.e. this prototype velocity is equal to $K_s/2$. The observed mid-depth velocity for a given centrifuge test is obtained by linear interpolation between the two data points closest to $Z_c/L = 1/2$ following three-point averaging if needed.
The kinetics of twenty-nine centrifuge infiltration tests on 1.33 mm diameter capillary tubes and twelve tests on 2.70 mm diameter capillary tubes selected from the tests series summarized in Table 7.9 were examined. The results for 1.33 mm and 2.70 mm diameter tubes are graphed in Figure 7.66 and Figure 7.67, respectively. Experimental data for these centrifuge tests are reported in Appendix C, Section C.5.1 and Section C.5.3.

In Figure 7.66 and Figure 7.67, the normalized mid-depth velocity is plotted versus different parameters measured for each infiltration test: the $\alpha_n$-number (Figure 7.66.a and Figure 7.67.a), the $g$-level, $n$ (Figure 7.66.b and Figure 7.67.b), the capillary tube model length, $l$ (Figure 7.66.c and Figure 7.67.c), the capillary tube prototype length, $L = nl$ (Figure 7.66.d and Figure 7.67.d), and the DNAPL prototype infiltration height, $H_i$ (Figure 7.66.e and Figure 7.67.e). In Figure 7.66.f and Figure 7.67.f, the DNAPL prototype infiltration height, $H_i$, is plotted versus the capillary tube model length, $l$.

**Centrifuge tests into 1.33 mm diameter capillary tubes**

For 1.33 mm diameter capillary tubes, Figure 7.66.a shows that the normalized velocity decreases from 0.95 to about 0.35, as the $\alpha_n$-number increases from 0.2 to 2.6. Recall that over the range of $\alpha_n$-number reported here, inertia forces are expected to have a measurable effect on the interface displacement. Thus, the trend shown in Figure 7.66.a can be attributed to inertia forces, which act to slow down the interface velocity displacement. Consequently, the ratio observed velocity to NICCA predicted velocity decreases, as the magnitude of inertia forces increases.

Figure 7.66.b shows that the normalized velocity of centrifuge tests into 1.33 mm diameter capillary tube decreases as the centrifuge $g$-level, $n$, increases. This trend is consistent with that of Figure 7.66.a. As the $g$-level increases, so does the $\alpha_n$-number, and thus the magnitude of inertia forces.

Examining now Figure 7.66.c, the normalized velocity of 1.33 mm diameter capillary tubes is observed to increase as the model length of the capillary tube, $l$, increases. Referring now to Figure 7.66.e, it can be seen that the normalized velocity of 1.33 mm diameter capillary tubes increases with increasing DNAPL prototype infiltration height, $H_i$. Finally, referring to Figure 7.66.f, the DNAPL prototype infiltration pool height of 1.33 mm diameter capillary tubes appears to increase with capillary tube model length. This trend has already been discussed in Section 7.3.3.2 (Figure 7.66.f is identical to Figure 7.60).

Thus, all three trends reported above are similar to those reported in Section 7.3.6.1 for centrifuge tests into 0.66 mm diameter capillary tubes, where the indirect effects of capillary resistance created a dependence of the normalized velocity upon infiltration height (see (7.44)). In the case of 1.33 mm diameter capillary tubes, it is believed that the velocity reduction is caused by both the effects of inertia and the indirect effects of capillary resistance.
For 1.33 mm diameter capillary tubes, the normalized velocity decreases with both decreasing capillary tube model length and decreasing DNAPL prototype infiltration height. The decrease in model length is accompanied by an increase of the magnitude of inertia forces (increase of $\alpha_n$ as per (4.2), see Section 4.2), which acts to slow down the infiltration process. This explains the trend shown in Figure 7.66.c and subsequently Figure 7.66.a. Furthermore, a decrease in model length is associated with a smaller DNAPL prototype infiltration height as per Figure 7.66.e. A smaller prototype infiltration height is associated with a smaller displacement velocity, as suggested by the NIVCA model (7.44). Thus, along with inertia effects, indirect capillary effects are also responsible for further decrease of the normalized velocity with decreasing model length.

Note again that, for 0.66 mm diameter tubes, as shown in Figure 7.65, the velocity reduction associated with decreasing model length is only believed to be due to indirect capillary effects. A smaller model length is associated with a smaller infiltration height (see Figure 7.65.f), which in turn reduces the normalized velocity (see Figure 7.65.e). Therefore, there appears to be a decrease of normalized velocity with decreasing model length (see Figure 7.65.c). Yet, because the magnitude of inertia forces remains small in that case, there is no noticeable effect of the $\alpha_n$-number or g-level on the normalized velocity of 0.66 mm diameter capillary tubes (see Figure 7.65.a and Figure 7.65.b).

Figure 7.66.d shows that the normalized mid-depth velocity of centrifuge infiltration tests into 1.33 mm diameter tubes increases as the prototype model length, $L = nl$, increases, although the trend is not always consistent for prototype lengths exceeding 800 mm. There are several competing mechanisms here. For 1.33 mm diameter tubes, the normalized velocity decreases with increasing g-level, $n$, but increases with increasing model length, $l$. In this test series, the largest prototype lengths $L$ correspond to centrifuge tests run at the largest g-levels, where inertia forces become important and thus reduce the magnitude of the normalized velocity. This explains why the increasing trend levels off for the largest prototype lengths.

Centrifuge tests into 2.70 mm diameter capillary tubes

Figure 7.67.a through Figure 7.67.e suggest that the normalized mid-depth velocity of centrifuge tests into 2.70 mm diameter capillary tubes remains a constant equal to about 0.4. The normalized velocity is not influenced by the $\alpha_n$-number although this number increases almost four-fold from 2.1 to 7.6 (see Figure 7.67.a). Similarly, the normalized velocity is not influenced by any of the followings: (1) the g-level, $n$ (see Figure 7.67.b), (2) the model length, $l$ (see Figure 7.67.c), (3) the prototype length, $L$ (see Figure 7.67.d), and (4) the prototype infiltration height, $H_i$ (see Figure 7.67.e) despite a two-fold increase in infiltration height. Finally, there is no obvious correlation in Figure 7.67.f between prototype infiltration height, $H_i$, and capillary tube model length, $l$. Note, however, that observed correlations with tube model length cannot be very conclusive, as centrifuge tests were run on two tubes of very similar model lengths (130 mm and 136 mm).
\( \alpha_n \)-Number, \( \alpha_n = \Delta \rho \nu (ng) r_0^4/(16 \mu_w^2 l) \)

(a) 

(b) 

g-Level, \( n \) 

Capillary Tube Model Length, \( l \) [mm] 

(c) 

Capillary Tube Prototype Length, \( L = nl \) [mm] 

(d)
Figure 7.66. Ratio of observed velocity to velocity predicted by the NICCA model at the mid-depth \((Z_c/L = 1/2)\) of 1.33 mm diameter capillary tubes: a. Versus \(\alpha_n\)-number; b. Versus \(g\)-level; c. Versus capillary tube model length; d. Versus capillary tube prototype length; e. Versus DNAPL prototype pool height at infiltration; f. DNAPL prototype pool height at infiltration versus capillary tube model length.
\[ \alpha_n \text{-Number, } \alpha_n = \Delta \rho \mu_w (ng) r_0^4 / (16 \mu_w^2 l) \]

(a) Ratio of Observed Velocity to NICCA Predicted Velocity

(b) g-Level, \( n \)

(c) Capillary Tube Model Length, \( l \) [mm]

(d) Capillary Tube Prototype Length, \( L = nl \) [mm]
Figure 7.67. Ratio of observed velocity to velocity predicted by the NICCA model at the mid-depth ($Z_c/L = 1/2$) of 2.70 mm diameter capillary tubes: a. Versus $\alpha_n$-number; b. Versus $g$-level; c. Versus capillary tube model length; d. Versus capillary tube prototype length; e. Versus DNAPL prototype pool height at infiltration; f. DNAPL prototype pool height at infiltration versus capillary tube model length.
That the normalized mid-depth velocity is not influenced by the g-level, the $\alpha_n$-number and the infiltration height suggests that the test series belongs to a flow regime different from that of 1.33 mm and 0.66 mm diameter capillary tubes. It is believed that for the $\alpha_n$-numbers reported here, the interface displacement may be predominantly controlled by inertia forces. The specific reason why the normalized velocity remains a constant as the magnitude of inertia forces increase is not known.

Note that an interesting feature can be seen when examining Figure 7.66.a and Figure 7.67.a simultaneously. For 2.70 mm diameter tubes, the normalized velocity remains approximately equal to 0.4 over the range 2.1-7.6 of $\alpha_n$-numbers. For 1.33 mm diameter tubes, the normalized velocity over the range 2.0-2.6 of $\alpha_n$-numbers is also approximately equal to 0.4. This suggests that, for $\alpha_n$-numbers exceeding 2.0, the normalized velocity becomes mostly dependent on the $\alpha_n$-number.

Figure 7.68. Normalized mid-depth velocity of centrifuge infiltration tests into capillary tubes of different diameters versus $\alpha_n$-number: a. Linear scale of $\alpha_n$; b. Log scale of $\alpha_n$. 

Figure 7.68 shows a plot of the normalized mid-depth velocity versus the $\alpha_n$-number for all the centrifuge infiltration tests examined in Figure 7.65-Figure 7.67. A linear scale of $\alpha_n$ is used in Figure 7.68.a, whereas a log scale of $\alpha_n$ is used in Figure 7.68.b. Overall, as can be seen in Figure 7.68.b, the normalized mid-depth velocities appear to decrease linearly as the log($\alpha_n$) increases. Nonetheless, the data are somewhat scattered, most particularly at the $\alpha_n$-numbers below 0.2. As pointed out earlier, this latter region corresponds to an inertia transition zone where other effects are present. For these $\alpha_n$-numbers, the normalized velocity is primarily affected by the
infiltration height and the capillary tube model length (so-called indirect effects of capillary resistance). For intermediate \( \alpha_n \)-numbers, larger than 0.2 but less than 2, it can be expected that infiltration height and model length will still affect the normalized velocity, but that inertia effects will also be important. Finally, for \( \alpha_n \)-numbers larger than 2, it is believed that the normalized velocity is primarily controlled by inertia forces.

7.3.6.3 Comparison of Centrifuge Tests With Laboratory Tests

The centrifuge summary plots obtained in Section 7.3.6.1 and Section 7.3.6.2 are now compared to the summary plots obtained in Section 7.2.2.4 and Section 7.2.2.5 for laboratory (1-g) infiltration experiments.

*Inertia effects in centrifuge and laboratory experiments*

Figure 7.69 shows a plot of the normalized mid-depth velocity versus the \( \alpha_n \)-number for all the centrifuge infiltration tests examined in Figure 7.65-Figure 7.67 (and thus Figure 7.68). A log scale of \( \alpha_n \) is adopted. In addition, the normalized mid-depth velocities of the laboratory (1-g) spontaneous infiltration tests reported in Table 7.2, Table 7.3, Table 7.4 are plotted in the same figure versus their \( \alpha_t \)-numbers. Thus, Figure 7.69 is the combination of the plot shown in Figure 7.26 and that shown in Figure 7.68.b. Recall that, for laboratory tests, the \( \alpha_t \)-number characterizes the relative importance of inertia forces in comparison to viscosity forces, and accounts for the effects of pre-infiltration.

It is interesting to note that, in Figure 7.69, the \( \alpha_t \)-numbers of laboratory infiltration experiments into 1.33 mm diameter capillary tubes are generally of the same order of magnitude as the \( \alpha_n \)-numbers of centrifuge infiltration experiments into 0.66 mm diameter capillary tubes. The \( \alpha_t \)-numbers and \( \alpha_n \)-numbers corresponding to these tests range approximately from \( 10^{-2} \) to \( 10^{-1} \). Similarly, the \( \alpha_t \)-numbers of laboratory infiltration tests into 2.70 mm diameter capillary tubes are of the same order of magnitude as the \( \alpha_t \)-numbers of centrifuge tests into 1.33 mm diameter capillary tubes. The \( \alpha_t \)-numbers and \( \alpha_n \)-numbers corresponding to these tests range approximately from \( 10^{-1} \) to 1.

When \( \alpha_n \) and \( \alpha_t \) vary between \( 10^{-2} \) and \( 10^{-1} \), a very large range of normalized velocities is observed for any fixed value of \( \alpha_n \) (or \( \alpha_t \)). This suggests again that, in this region, the inertia forces have no detectable effect on the interface velocity, and that other resisting mechanisms are driving the flow.

When \( \alpha_n \) and \( \alpha_t \) increase from \( 10^{-1} \) to 1, the normalized velocity decreases. In this region, the scatter of the normalized velocities is smaller than in the region where \( \alpha_n \) (or \( \alpha_t \)) varies between \( 10^{-2} \) and \( 10^{-1} \). Overall, it is believed that, in the region where the \( \alpha_n \)-numbers (or \( \alpha_t \)-numbers) range between \( 10^{-1} \) and 1, inertia forces have an
effect on the interface velocity, but that other resisting mechanisms of similar magnitude are also controlling the flow.

As the $\alpha_n$-number (or $\alpha_t$-number) increases, it is expected that inertia forces take a predominant role in controlling the flow, so that new mechanisms arise that are not accounted for in the NICCA model, MFDV or NIVCA model.

\[
\alpha_n \text{-Number, } \alpha_n = \Delta \rho \rho_w (ng) r_0^4/(16 \mu_w^2 l)
\]

or \[
\alpha_t \text{-Number, } \alpha_t = \Delta \rho \rho_w g r_0^4/(16 \mu_w^2 l_t)
\]

**Figure 7.69.** Normalized mid-depth velocity of centrifuge and laboratory infiltration tests into capillary tubes of different diameters versus $\alpha_n$-number (or $\alpha_t$-number).

**Effects of capillary tube model length**

Figure 7.70 shows a plot of the interface normalized mid-depth velocity versus the capillary tube normalized model length, $l/d$, for all the centrifuge infiltration tests examined in Figure 7.65-Figure 7.67. A linear scale of $l/d$ is used in Figure 7.70.a, whereas a log scale is used in Figure 7.70.b. In addition, the normalized mid-depth velocities of the laboratory (1-g) spontaneous infiltration tests reported in Table 7.2, Table 7.3, Table 7.4 are plotted in the same figure versus the capillary tube normalized length, $l/d$. Thus, Figure 7.70 is the combination of the plots shown in Figure 7.65.c-Figure 7.67.c and that shown in Figure 7.19 (without the three-quarter depth velocity
data). Again, for laboratory tests, the length $l$ corresponds to the length of capillary tube over which the interface travels once actual infiltration takes place.

![Figure 7.70. Normalized mid-depth velocity of laboratory and centrifuge infiltration tests versus capillary tube model length normalized by its diameter, $l/d$: a. Linear scale of $l/d$; b. Log scale of $l/d$.](image)

Overall, Figure 7.70 shows that the normalized velocity increases as the normalized model length increases. While the centrifuge test data appear to fall somewhat on the laboratory test data (see Figure 7.70.b), it can hardly be argued that the normalized velocity plot is solely dependent upon the normalized model length of the capillary tube. This suggests again, as shown earlier in Section 7.2.3.4, that the normalized velocity-normalized model length plot is not a unique curve, and that other parameters are affecting the normalized mid-depth velocity. In particular, it must be noted in Figure 7.70.b that the centrifuge tests show that broad ranges of normalized velocities can be achieved over relatively narrow normalized length ranges. For example, in Figure 7.70.b, the normalized velocity of 0.66 mm diameter centrifuge tests (open triangles) ranges between 0.7 and 1.1, whereas their corresponding normalized lengths remain of the order of $2 \times 10^{-2}$. Similarly, the normalized velocity of some 1.33 mm diameter centrifuge tests (open square) range between 0.6 and 1.0, whereas their corresponding normalized lengths remain of the order of $10^{-2}$. As discussed earlier this effect is attributable to the prototype infiltration height $H_i$, which can influence the interface velocity through the indirect action of capillary forces.
Effects of capillary tube prototype length

Figure 7.71 shows a plot of the interface normalized mid-depth velocity versus the capillary tube normalized prototype length, \( L/d \), for all the centrifuge infiltration tests examined in Figure 7.65-Figure 7.67. A linear scale of \( L/d \) is used in Figure 7.71.a, whereas a log scale is used in Figure 7.71.b. In addition, the normalized mid-depth velocities of the laboratory (1-g) spontaneous infiltration tests reported in Table 7.2, Table 7.3, Table 7.4 are plotted in the same figure. Thus, Figure 7.70 is the combination of the plots shown in Figure 7.65.d-Figure 7.67.d and that shown in Figure 7.19 (without the three-quarter depth velocity data). Note that, for laboratory (1-g) tests, the prototype length is the same as the model length.

![Figure 7.71](image)

**Figure 7.71.** Normalized mid-depth velocity of laboratory and centrifuge infiltration tests versus capillary tube prototype length normalized by its diameter, \( L/d \): a. Linear scale of \( L/d \); b. Log scale of \( L/d \).

Similar to Figure 7.70, Figure 7.71 shows that the normalized velocity increases as the normalized prototype length increases. However, again, it cannot be argued that the normalized velocity plot is solely dependent upon the normalized prototype length of the capillary tube. In particular, complication arises because long prototype capillary tube lengths may result from large \( g \)-levels, where the effects of inertia are important. Conversely, for 1-g infiltration tests, the mid-depth normalized velocity approaches 1 with increasing capillary tube length. Thus, the relationship between the normalized velocity and the normalized prototype length, \( L/d \), is not nearly as good as
that between the normalized velocity and the normalized model length, $l/d$, because the effects of inertia may be present throughout the entire range of $L/d$.

*Three major mechanisms responsible for the observed summary plots*

In conclusion, based on the findings made in Section 7.2.3.4 and those made in Section 7.3.6.1 and Section 7.3.6.2, it is believed that the following three mechanisms are causing deviations from the NICCA model prediction:

1. **Resistance due to (local and convective) inertia effects.** Inertia effects are important if the capillary tube normalized model length is small, and decrease the normalized mid-depth velocity with decreasing tube normalized model length. The magnitude of inertia forces is also dependent upon the $g$-level ($\alpha_n$ is proportional to $n$), so that the sole normalized model length does not constitute a good criterion to characterize the magnitude of inertia forces. Instead, the $\alpha_n$-number (or $\alpha_r$-number) should be used, as shown in Figure 7.69, as the $\alpha_n$-number combines both the $g$-level and the tube normalized length. Note that the $\alpha_n$-number (or $\alpha_r$-number) is not inversely proportional to $l/d$ but $l/d^4$.

2. **Capillary resistance.** This capillary resistance was shown to be the major mechanism acting on the 1.33 mm diameter capillary tube laboratory spontaneous infiltration tests, and was also believed to have a partial effect on the 2.70 mm diameter tube laboratory spontaneous infiltration tests. The decrease of contact angle acts to raise the dynamic infiltration height to a value larger than the pool height at the onset of infiltration, such that the quantity $\Delta h'$ (as defined in the NIVCA model, see (7.17)) becomes a negative constant. This constant acts to decrease the interface velocity. The constant $\Delta h'$ is dependent on the capillary tube diameter (as well as a number of other parameters), but does not theoretically depend on the capillary tube length. Therefore, $\Delta h'$ remains constant for a given capillary tube diameter, provided that the static contact angle and pinning forces are constant. Ultimately, the normalized velocity increases with capillary tube length because, for longer capillary tubes, the length scale over which the effects of $\Delta h'$ are important becomes relatively small in comparison to the capillary tube length (see (7.15)). Thus, for long capillary tubes, the normalized velocity approaches 1, whereas for capillary tubes of length of the order of $\Delta h'$, the constant $\Delta h'$ largely reduces the maximum velocity that can be achieved during infiltration. Overall, the normalized velocity is expected to increase with increasing capillary tube length.

In the case of centrifuge tests, the trend discussed above may be complicated for some tests by the magnitude of inertia forces. Focusing exclusively on centrifuge infiltration tests into 0.66 mm diameter capillary tubes where inertia is unimportant, it would be expected from (7.44) and (7.47) that the normalized velocity increases with prototype length. However, this trend cannot be seen in Figure 7.65.d. There are two reasons that may explain this behavior. First, it is believed that the range of $L$ of the tests series (500 mm to 1700 mm, i.e three-fold increase) may not be large
enough to yield any significant trend. Achieving a larger range is unfortunately no trivial matter, as (i) increasing the model tube length is difficult given the centrifuge platform and strong box dimensions, (ii) increasing the $g$-level raises the $\alpha_n$-number, and (iii) decreasing the tube model length also raises the $\alpha_n$-number. Second, the prototype infiltration height appears to have the most influence on the normalized mid-depth velocity of these centrifuge tests (see mechanism 3 below), so that the large variability of $H_i$ may overshadow the trend of normalized velocity increase associated with increasing prototype length.

3. **Indirect effect of capillary resistance (influence of infiltration height on the interface velocity).** This mechanism was shown to be the major mechanism acting on the 0.66 mm diameter capillary tube centrifuge infiltration tests, and was also believed to have a partial effect on the 1.33 mm diameter tube centrifuge infiltration tests. The variability of $H_i$—associated with either the variability of the static contact angle, or the variability of the pinning force—is believed to have an impact on the normalized mid-depth velocity, and more generally on the interface velocity field. Indeed, as infiltration takes place, the dynamic infiltration height increases, so that the difference $\Delta H' = H - H_i(dZ/dT)$ becomes negative. The flow velocity is affected by the magnitude of $\Delta H'$ (see NIVCA model equations (7.44)-(7.45)), and thus is also affected by the initial infiltration height, $H_i$, as $H \approx H_i$.

The normalized mid-depth velocities of centrifuge tests are particularly affected by the prototype infiltration heights because the infiltration heights vary over an extremely broad range for each centrifuge test series. For example, the prototype infiltration heights of the 12 centrifuge infiltration tests into 0.66 mm diameter capillary tubes plotted in Figure 7.65.e vary from about 210 mm to about 340 mm. Similarly, the prototype infiltration heights of the 29 centrifuge infiltration tests into 1.33 mm diameter capillary tubes plotted in Figure 7.66.e vary from about 100 mm to almost 180 mm.

Furthermore, for centrifuge infiltration tests into 1.33 mm and 0.66 mm diameter capillary tubes, there exists a positive correlation between infiltration height and capillary tube model length, despite a large scatter of these data (see Figure 7.65.f and Figure 7.66.f). Consequently, the normalized mid-depth velocity increases with increasing capillary tube model length, through the dependence upon infiltration height.

It must be noted that the influence of infiltration height upon velocity was not detected for the laboratory (1-g) spontaneous infiltration tests. Nonetheless, it is likely that such trend would be detected if more infiltration tests were run. However, for laboratory tests, an infiltration test run consecutively on the same capillary tube can be associated with different pre-infiltration depths. Therefore, the final length $l$ may vary between tests, even if the capillary tube has the same overall length, $l_o$, so that mechanism 2 may also act to influence the normalized mid-depth velocity.

As an example, consider tests L8f-1.33st-122mm and L8a-1.33st-122mm run on the same 122 mm long, 1.33 mm diameter capillary tube (see Table 7.2). For test
L8f-1.33st-122mm, the pre-infiltration depth is 18 mm, so that the reduced length, $l$, is equal to 104 mm. For test L8a-1.33st-122mm, the pre-infiltration depth is 50 mm, so that the reduced length, $l$, is equal to 72 mm. Despite a larger infiltration height, $h_i = 155.2$ mm, the normalized mid-depth velocity of the test run in the shorter tube is equal to 0.22 versus 0.53 for the longer capillary tube, for which the infiltration height is $h_i = 149.6$ mm (see Table 7.2). Clearly, in this case, mechanism 2 described above is the dominant one (with perhaps an influence of mechanism 1, as concluded in Section 7.2.3.7).

Consider now tests L7b-1.33st-184mm and L7a-1.33st-184mm run on the same 184 mm long, 1.33 mm diameter capillary tube (see Table 7.2). For test L7b-1.33st-184mm, the pre-infiltration depth is 33 mm, so that the reduced length, $l$, is equal to 151 mm. For test L7a-1.33st-184mm, the pre-infiltration depth is 37 mm, so that the reduced length, $l$, is equal to 147 mm. In this case, both tubes have very similar lengths. Furthermore, the normalized mid-depth velocity of the shorter tube, which is infiltrated for $h_i = 149.0$ mm, is equal to 0.61. For the longer tube, which is infiltrated for a smaller pool height, $h_i = 145.8$ mm, the normalized mid-depth velocity is equal to 0.47. Clearly, the difference in normalized velocities is not compatible with mechanism 2. However, this difference may be qualitatively explained with mechanism 3, even though it must be acknowledged here that the difference in infiltration height between the two tests is so small, that it may not generate such a large difference between normalized velocities.

### 7.4 References


8.1 Research Summary

8.1.1 Background

This thesis examined the infiltration behavior of dense non-aqueous phase liquids (DNAPLs) in vertical, initially water-saturated, smooth-walled fractures modeled by circular-section capillary tubes of varying lengths and diameters. A few experimental studies on liquid/liquid displacements in horizontal tubes, as well as one study in vertical tubes, have been previously reported in the literature (see Section 2.3.6). Nonetheless, to the author’s knowledge, this is the first work reported on the downward displacement of a wetting liquid by a non-wetting liquid in vertical capillary tubes.

Infiltration experiments were carried out in the laboratory, as well as at a reduced scale in the geotechnical centrifuge. For the laboratory experiments, the DNAPL transport was subjected to the Earth’s gravitational acceleration, g. For this reason, these experiments are referred to as DNAPL infiltration laboratory (or 1-g) experiments. In contrast, the DNAPL transport in the reduced-scale centrifuge experiments was subjected to a combination of the Earth’s gravity and the centrifuge rotational acceleration, equivalent to several times the Earth’s gravitational acceleration. Such experiments are referred to as DNAPL infiltration centrifuge experiments.

In order to obtain information on the DNAPL infiltration behavior, a new general model and a series of sub-models were developed to provide a criterion for the onset of infiltration into a water-saturated vertical fracture, as well as a prediction of the kinetics of the DNAPL/water interface following infiltration. The model was also extended to the prediction of centrifuge infiltration experiments. Scaling laws were developed that link observations made in the reduced-scale centrifuge model of a fracture system to the full-scale equivalent system (prototype). The model and its sub-models were modified and refined in light of the experimental results.
8.1.2 Findings From Review of Prior Research Work

Prior to developing the DNAPL infiltration model, an extensive review of literature was performed. Prior research work in each of the three following categories was found to be pertinent to this study, and was examined in Chapter 2: (i) experimental and numerical studies modeling DNAPL (or two-phase) transport in the subsurface environment, and most specifically in fractured media, such as fractured rock; (ii) flow kinetics in capillary tubes, where a liquid replaces another fluid (gas or other liquid) under conditions of spontaneous or forced displacement; and (iii) centrifuge modeling of geoenvironmental processes, and most specifically of two-phase flow transport in porous media (there is no known study of flow in fractured media).

With respect to the existing research work on DNAPL transport in fractured media, it was found that existing numerical models rely on acquiring accurate descriptions of individual fracture properties, at a scale smaller than can typically be achieved in practice, and that many of these models have yet to be validated in the field. It was also found that, in the case of experimental studies on real fractures, the use of a small laboratory bench scale generally distorts the relative importance of gravity, capillarity, viscosity and inertia in comparison to the larger in situ scale, and yields mechanisms controlled by forces that may be irrelevant at the in situ scale. From these experimental studies, it also appeared that the complexity of the fracture systems made it difficult to analyze the role played by capillary forces.

With respect to the existing research work on the flow kinetics in capillary tubes, it was found that researchers generally agree that the classic Lucas-Washburn kinetics \([\text{Lucas}, 1918; \text{Washburn}, 1921]\) most often over-predicts the rate of infiltration of the displacing liquid. Some researchers attribute the observed discrepancies between experimental data and Lucas-Washburn predictions to the effects of inertia and the pressure drop existing at the entrance to a capillary tube. However, the corrected models proposed by these researchers also often over-predict the experimental rate of liquid/fluid infiltration. Note that, in some studies, it was found that convective inertia forces are ignored even if, clearly, these forces are not negligible with respect to the viscous forces.

Review of a number of papers also showed that a popular explanation of the discrepancies between the Lucas-Washburn model predictions and experimental data is the well-acknowledged dependence of the liquid/fluid interface dynamic contact angle upon velocity, a point ignored by the Lucas-Washburn kinetics. However, it was noted that studies that obtain successful experiment predictions using variable dynamic contact angles typically rely on fitting parameters back-calculated from a first series of infiltration tests. Complications arise for the particular case of liquid/liquid interface displacements, where some experimental studies have shown that the mechanisms governing liquid/liquid displacements appear to differ from those governing liquid/gas displacements and may not be simply inferred from the latter.

With respect to the existing research work on the modeling of two-phase flow transport using the geotechnical centrifuge, it was found that a number of studies are
successful at scaling the downward transport of LNAPL in a dry or partially saturated porous medium or downward transport of DNAPL in a water-saturated porous medium. Nonetheless, it is suspected that scaling is successful under these configurations because gravity forces are dominant in comparison to capillary forces, and/or because a reduced range of centrifuge acceleration levels is used to perform the modeling study. For the small number of studies where capillary forces are clearly as important as gravity forces, such as the case of capillary rise in sand columns, scaling of the kinetics of capillary rise is quite inconclusive. This research indicates that the dependence of the dynamic contact angle upon velocity complicates centrifuge modeling of any infiltration process where capillary forces are important.

8.1.3 Model Development

Extending the results of previous research work, a general model and series of sub-models were developed in Chapter 3. The model described DNAPL infiltration from a pure-phase solvent reservoir pool into a vertical, initially water-saturated fracture located underneath the DNAPL pool.

It was first re-demonstrated that, theoretically, the maximum pool height of DNAPL that can accumulate on top of the fracture prior to infiltration into the fracture is a function of the fracture’s aperture, the DNAPL/water interfacial tension, the density contrast between DNAPL and water, and the static contact angle at the DNAPL/water interface.

Next, a fracture idealized as a circular-section, vertical capillary tube was considered. The Navier-Stokes equations were used to obtain a description of the DNAPL infiltration kinetics in the capillary tube, i.e. a prediction of the DNAPL/water interface depth, $z_c$, as a function of time, $t$. These equations were integrated over the DNAPL/water column within the capillary tube. Using a parabolic flow approximation, a simplified non-linear second order differential equation predicting $z_c(t)$ was obtained. This simplified equation required that the convective inertia forces of the DNAPL/water column be negligible with respect to the viscous forces.

Next, series of complementary assumptions were made in order to obtain approximate analytical solutions of the governing equation in $z_c(t)$. Several series of assumptions were considered:

1. **Negligible Inertia Constant Contact Angle (NICCA) model.** In this series of assumptions, (i) the drag forces at the entry to the capillary tube and the local inertia forces were neglected in comparison to the viscous forces, (ii) the viscosity contrast between DNAPL and water was assumed to be negligible with respect to the viscosity of water, and (iii) the dynamic contact angle at the DNAPL/water interface was assumed to be a constant independent of velocity.

   A solution was obtained that predicted that, for DNAPL spontaneous infiltration in a capillary tube with no existing DNAPL pre-infiltration, the interface velocity
increases continuously throughout the infiltration process, and is proportional to the relative depth, \( z_c/l \), of the interface. Consequently, the interface velocity at the lower end of the capillary tube was theoretically independent of the capillary tube length, and equal to a constant solely dependent upon the density contrast between DNAPL and water, the viscosity of water (and DNAPL) and the capillary tube diameter.

A number, called the \( \alpha_t \)-number, was defined for any given infiltration experiment. This number was shown to be a function of various parameters of the DNAPL/water column system. It characterized the importance of local inertia forces with respect to the viscous forces in the system. The effects of local inertia forces were shown to be theoretically negligible provided that the \( \alpha_t \)-number is negligible with respect to unity, in which case it was hypothesized that the NICCA model is applicable. Furthermore, it was shown that the \( \alpha_t \)-number can also characterize the magnitude of convective inertia forces and entry drag forces in the system. Thus, for any system for which local inertia forces are negligible, so are convective inertia forces and entry drag forces.

2. Negligible Inertia Variable Contact Angle (NIVCA) model. In this series of assumptions, (i) the drag forces at the entry to the capillary tube and the local inertia forces were neglected in comparison to the viscous forces, (ii) the viscosity contrast between DNAPL and water was assumed to be negligible with respect to the viscosity of water, and (iii) the dynamic contact angle at the DNAPL/water interface was assumed to vary as a function of velocity. Specifically, it was assumed for (iii) that the cosine of the dynamic contact increased linearly from the cosine of the static contact angle to unity, as the interface velocity increased from 0 to the so-called perfect wetting velocity.

It was demonstrated that the NIVCA model predicts that the decrease in contact angle and associated increase of the capillary force contribute to displacement velocities that can be significantly smaller than those predicted by the NICCA model. The NIVCA solution that was obtained predicted that DNAPL infiltration is a two-stage process. During the first stage, as the interface velocity increases, the dynamic contact angle decreases to zero. Thus, the capillary resisting force increases from its initial static value, for which DNAPL infiltration has taken place, to its maximum perfect wetting value. During the second stage, the interface velocity keeps increasing, but the dynamic contact angle remains equal to zero and the capillary resistance remains equal to its maximum value. At both stages of the infiltration process, the NIVCA model predicts that the interface velocity is linearly related to the interface depth, although the respective velocity-depth slopes are not equal.

3. Model with Fluids of Differing Viscosities (MFDV). In this series of assumptions, (i) the drag forces at the entry to the capillary tube and the local inertia forces were neglected in comparison to the viscous forces, (ii) no assumption was made on the viscosity contrast between DNAPL and water, and (iii) the dynamic contact angle
at the DNAPL/water interface was assumed to be a constant independent of velocity.

It was shown that, unlike the NICCA model, the MFDV predicts that the interface velocity is not linearly related to the depth of the interface. Furthermore, for a DNAPL less viscous than water, the MFDV predicts that the infiltration process is faster than that predicted by the NICCA model (conversely, the MFDV infiltration is slower than the NICCA infiltration if the DNAPL is more viscous than water). It was also shown that under conditions where the DNAPL/water viscosity contrast is less than a tenth of the viscosity of water, the MFDV appears to be reasonably well approximated by the NICCA model.

The NICCA model, NIVCA model and MFDV were modified and refined in Chapter 7 in light of the laboratory infiltration test results. In particular, the models were modified to account for the effects of DNAPL pre-infiltration as well as the effects of a so-called pinning force that was found to contribute to the capillary resistance against interface displacement during the experiments (see Section 8.1.6).

Finally, in Chapter 3, the NICCA model was extended to smooth-walled rectangular section capillary tubes, as well as rough-walled fractures where homogeneous hydraulic properties can be defined. Although, these models were not investigated experimentally in this thesis, data from a series of experiments run on rectangular section capillary tubes are reported elsewhere [Adams, 2000].

In Chapter 4, scaling laws of DNAPL infiltration into a vertical, water-saturated, rough-walled fracture were developed that allowed translation of data from a reduced-scale centrifuge model to its equivalent full-scale prototype. These scaling laws were obtained by extending the NICCA model to the case where the body force acting upon the fracture system is a centrifugal acceleration equivalent to \( n \) times the Earth’s gravity. Recall that the scale of a centrifuge model is \( 1/n \) time the scale of its equivalent prototype.

A number called the \( \alpha_n \)-number was defined, which characterizes for a reduced-scale centrifuge experiment the importance of inertia forces with respect to the viscous forces. Similar to the \( \alpha_t \)-number, the effects of inertia forces were shown to be theoretically negligible—and thus the NICCA model theoretically applicable—provided that the \( \alpha_n \)-number is negligible with respect to unity. It was also demonstrated that the \( \alpha_n \)-number of a given reduced-scale centrifuge experiment is \( n^2 \) times larger than the \( \alpha_t \)-number of the equivalent full-scale prototype. Consequently, it was concluded that for proper scaling in the centrifuge of capillary forces (assumed velocity-independent), gravity forces and viscous forces, one has to ensure that inertia forces are negligible not solely in the full-scale experiment, but also in the reduced-scale centrifuge experiment where inertia forces are \( n^2 \) times larger.

For the particular case of circular-section capillary tubes, extension of the NIVCA model to centrifuge experiments was also examined. It was shown that scaling of velocity-dependent capillary forces is possible provided that the perfect wetting velocity is \( n \) times larger in the reduced-scale centrifuge experiment than in the full-scale equivalent prototype.
4-chlorotoluene (4-CT), a DNAPL of density 1.070 g/cm$^3$ at 20$^\circ$C, was used for both the laboratory and centrifuge experiments presented in this research. 4-CT was selected for its small density contrast with water, its low toxicity relative to other DNAPLs, and its high flash point. In addition, 1,1,1-trichloroethane (TCA) was also used for a limited number of laboratory infiltration experiments. TCA is a common DNAPL pollutant found in groundwater. Its density is equal to 1.339 g/cm$^3$ at 20$^\circ$C. For all infiltration experiments, both DNAPLs were dyed red using Sudan IV hydrophobic dye at a concentration of 0.2 g per liter of DNAPL. Important safety, disposal and cleaning procedures when handling 4-CT, TCA, Sudan IV and other chemicals were reviewed in Chapter 5, Section 5.4.

To check whether Sudan IV affected the viscosity of the DNAPLs, the viscosity of Sudan IV-dyed 4-CT was measured using a capillary tube falling head technique (see Section 5.5). The viscosity of Sudan IV-dyed 4-CT was found to be $0.918 \pm 0.004$ mPa.s/m$^2$ at 20$^\circ$C, a value in close agreement with the undyed 4-CT viscosity values reported in the literature, thereby suggesting that Sudan IV has little effect, if any, on the viscosity of 4-CT. It was also concluded that, as a first approximation, the viscosity of 4-CT can be assumed to be equal to that of water, i.e. 1 mPa.s/m$^2$ at 20$^\circ$C, an assumption made in the NICCA and NIVCA models. However, the impact of the actual viscosity contrast was examined when inspecting the experimental results (see Section 8.1.6.2 and Section 8.1.7.3).

The interfacial tensions of 4-CT and TCA with water were not available from the literature, and thus had to be determined experimentally. After reviewing existing interfacial tension measurement methods, it was decided to use the pendant drop method (see Section 5.6.3) and the combined capillary rise method (see Section 5.6.4). In the pendant drop method, the interfacial tension between DNAPL and water is obtained by measuring the dimensions and shape of a drop of DNAPL hanging in a bath of water from the tip of a small tube. In the combined capillary rise method, the interfacial tension between DNAPL and water is obtained by measuring the capillary rise of a DNAPL/water column above the free surface of water.

At a room temperature of $19 \pm 1^\circ$C, the interfacial tension of Sudan IV-dyed 4-CT with water was found to be equal to $0.032 \pm 0.004$ N/m using the pendant drop method, and $0.0319 \pm 0.0025$ N/m using the combined capillary rise method. The former value was used in the thesis. At the same room temperature, the interfacial tension of Sudan IV-dyed TCA with water was found to be equal to $0.0344 \pm 0.0021$ N/m using the combined capillary rise method. Measurement of the interfacial tension of undyed 4-CT with water using the pendant drop method suggested that Sudan IV reduced the interfacial tension of 4-CT/water by approximately 0.004 N/m.

The pendant drop method demonstrated that important aging effects were taking place at the DNAPL/water interface. That is, within the first hour of the life of a drop of 4-CT, a reduction of 0.004 N/m, from about 0.035 N/m to about 0.031 N/m, was observed between interfacial tension measurements. After a day, the interfacial tension
was observed to decrease even further to about 0.028 N/m. A linear decrease as a function of the log of time provided a reasonably good fit of the aging trend.

The DNAPL/water interfacial tensions were back-calculated from the largest possible DNAPL/water column capillary rises corresponding to a configuration of perfect wetting (or close to it). Nonetheless, the combined capillary rise method provided evidence of contact angle hysteresis existing at the DNAPL/water interface. Indeed, it was shown that equilibrium of a 4-CT/water column above the free surface of water can be achieved under a range of capillary rises above the free surface of water. Observations of 4-CT/water interfaces showed that the meniscus can deform, such that a smaller 4-CT/water column rise is associated with a flatter 4-CT/water interface meniscus, and thus a larger static contact angle. That different equilibrium configurations were possible for a DNAPL/water column suggested that a DNAPL/water interface can be pinned onto locations of a capillary tube wall, so that movement of the meniscus contact line is prevented but deformation of the meniscus is possible. These results were later helpful in understanding the process of DNAPL infiltration into capillary tubes (see Section 8.1.6).

8.1.5 Experimental Methodology

As previously stated, verification and validation of the models developed in Chapter 3 and Chapter 4 were performed using fractures idealized by vertical, initially water-saturated capillary tubes of various length and diameters. Four different tube diameters were used, equal to 0.66 mm, 1.33 mm, 2.20 mm and 2.70 mm, respectively. For laboratory infiltration experiments, the length of the tubes varied from 60 mm to 1220 mm. For centrifuge experiments, the length of the tubes varied from 40 mm to 136 mm. The adopted experimental methodology was described in Chapter 6.

Two types of laboratory infiltration experiments were conducted:

1. In so-called "spontaneous" infiltration laboratory experiments, DNAPL was added into a reservoir tube connected to the top of a vertical capillary tube, the system reservoir tube-capillary tube being initially water saturated and plunged into a bath of water. A pool of DNAPL was slowly built up, and reached a height for which the pool buoyant weight exceeded the entry pressure of the capillary tube. At this point, infiltration of the tube by DNAPL took place. The height for which infiltration took place, termed the infiltration (or critical) height, was measured. In addition, for a number of tests, the DNAPL/water interface downward displacement was videotaped to obtain information on the infiltration kinetics and compare it to the model predictions.

2. In so-called "controlled-head" infiltration laboratory experiments, DNAPL was similarly added to a reservoir tube connected to the top of a vertical, water-saturated capillary tube. However, in this case, the lower end of the capillary tube was connected to a shut-off valve that prevented any flow of DNAPL, even if the DNAPL pool height was larger than the above-mentioned critical height. Unlike
spontaneous infiltration experiments, DNAPL pools could be built up past the critical height, and then the valve could be turned on to examine the infiltration kinetics. This technique was used for a limited number of infiltration experiments on 305 mm long, 1.33 mm diameter capillary tubes (see discussion in Section 8.1.6.2).

Centrifuge infiltration experiments were similar in principle to laboratory spontaneous infiltration experiments. A centrifuge experiment constituted of a vertical capillary tube and a reservoir tube, which were both set up in a centrifuge strong box filled with water. A DNAPL pool of given height was built up in the reservoir tube. The system was then centrifuged at a slowly increasing g-level until DNAPL infiltration into the capillary tube took place. Again, the experiment was videotaped to measure the infiltration kinetics. Scaling laws were used to translate the centrifuge experimental results into those of the equivalent prototype. The scaled data were then compared to the model predictions.

The centrifuge infiltration experiments involved the design of a tachometer to measure the centrifuge RPM, as well as a data acquisition system in collaboration with Sjoblom [2000] and Marulanda [2001] to record the RPM data as a function of time. The centrifuge instrumentation and data acquisition were presented in Section 6.4.

8.1.6 DNAPL Infiltration Laboratory Experiments

8.1.6.1 Infiltration Height Measurements and Model Predictions

Infiltration heights were measured for 48 laboratory spontaneous infiltration experiments using 4-CT as the DNAPL, and for 8 experiments using TCA as the DNAPL. Good agreement was noted between the measured infiltration heights and the predicted infiltration height, $h_i$, given by the infiltration criterion (3.4) (see derivation in Section 3.3), where perfect wetting ($\theta_s = 0$) is assumed. It was generally noted that the criterion provides an upper bound of the average infiltration height for a given DNAPL and capillary tube diameter.

Generally, a large scatter of infiltration height measurements was noted. Relative standard deviations of up to 20% were measured for a given DNAPL and tube diameter. Overall, it is believed that predictions to within one significant digit only can be counted on for the infiltration height of a DNAPL in a capillary tube using (3.4).

Although perfect wetting was assumed in (3.4), observations of the DNAPL/water interface showed that the static contact angle was not in a configuration of perfect wetting, thereby suggesting that infiltration criterion (3.4) is not strictly correct, and that there exists another force present that contributes to raise the value of the capillary tube entry pressure.

Furthermore, in all tests, DNAPL pre-infiltration was observed prior to the actual onset of DNAPL infiltration into a capillary tube. Recall that (actual) infiltration
means that a DNAPL finger penetrates the capillary tube without being stopped. In contrast, pre-infiltration means that the DNAPL finger penetrates the capillary tube but is later immobilized at a so-called pre-infiltration depth. The reservoir pool heights for which pre-infiltration was first observed were always much smaller than the final pool heights measured at the actual onset of DNAPL infiltration. In some cases, the DNAPL pre-infiltration depth could be significant compared to the DNAPL reservoir pool height. Consequently, each DNAPL infiltration pool height reported in the experimental results was the sum of the DNAPL reservoir pool height and the pre-infiltration depth.

In light of these findings, a new infiltration criterion that included pinning effects, given by (7.2) (see Section 7.2.1.2), was proposed that was in very good quantitative agreement with the experimental data on 4-CT. The magnitude of the so-called pinning force, \( n_{pfp} \), was found to be of similar order as the DNAPL/water interfacial tension, \( \sigma \cos \theta_s \), so that both interfacial tension and pinning contribute to the tube capillary resistance. The pinning forces were found to decrease with increasing radius. They were estimated to be equal to 0.005 N/m, 0.010 N/m and 0.012 N/m for 2.70 mm, 1.33 mm and 0.66 mm diameter capillary tubes, respectively, although the pinning force for 0.66 mm diameter tubes was estimated from only one infiltration test. The static contact angle for the test series was estimated to vary between 30° and 60° depending on the test. The variability of the measured infiltration height was attributed to two factors:

1. Variability of the interfacial tension, which can decrease significantly over time as a consequence of interface aging.
2. Variability of the static contact angle. The static contact angle is believed to be affected by the condition of deposition of the first DNAPL drop on top of the capillary tube, the make-up glass wall surface of the capillary tube itself, and the amount of pre-infiltration into the capillary tube. It was found that the more pre-infiltration, the larger the measured critical height at the onset of infiltration, a trend consistent with a decrease in static contact angle.

That changes in magnitude of the pinning force between different infiltration tests may also contribute to the variability of results was not excluded as a possibility.

That pre-infiltration was observed at a pool height significantly smaller than the actual DNAPL infiltration pool height was attributed to a mechanism combining a dynamic contact angle feedback loop and contact angle hysteresis. Indeed, it was hypothesized that pre-infiltration takes place at an early stage of the pool built-up because the initial static contact angle is approaching 90°. Next, the feedback mechanism prevents further infiltration of the pre-infiltrating finger by decreasing the dynamic contact angle and stabilizing the interface. At small interface displacement velocities, pinning forces hold the meniscus in place and delay the onset of infiltration until further build-up of the DNAPL pool can be achieved. The pinning points are believed to be individual defects (solid surface roughness), surface heterogeneity or impurities present on the walls of the capillary tubes (i.e. cleanliness of the tube). The magnitude of the pinning force for each capillary tube diameter was estimated from the DNAPL pool height at first observed pre-infiltration.
The infiltration height measurement results suggested that the criterion (3.4) commonly used in the literature is inadequate for predicting the DNAPL infiltration height into porous and fractured media, as pinning forces can significantly affect the magnitude of the pore/fracture capillary resistance. Moreover, the conventional assumption of perfect wetting at the DNAPL/water interface was found to be incorrect.

To the author’s knowledge, pre-infiltration and its explicative mechanisms have never been reported before. For this reason, these findings constitute an important contribution to the field.

8.1.6.2 Infiltration Kinetics and Model Prediction

The kinetics of several 4-CT infiltration laboratory tests was studied and compared to the NICCA, MFDV and NIVCA model predictions. All three models were revisited to account for pre-infiltration effects and the new infiltration criterion (7.2) that includes the pinning resistance.

For this study, a total of fourteen spontaneous infiltration tests were examined: six infiltrations tests into 2.70 mm diameter capillary tubes of length ranging from 88 mm to 1202 mm, seven infiltration tests into 1.33 mm diameter capillary tubes of length ranging from 72 mm to 912 mm and one infiltration test into a 1201 mm long, 0.66 mm diameter capillary tube. The length reported here corresponds to the actual flow length, i.e. the total length of the capillary tube piece minus the pre-infiltration depth achieved prior to the actual infiltration. In addition, for the comparison with the NIVCA model, six 4-CT controlled-head infiltration tests into a 305 mm long, 1.33 mm diameter capillary tube were examined.

NICCA model and MFDV

Comparison of the experimental results of spontaneous infiltration tests with the NICCA model showed that agreement between the experimental data and the NICCA model was good asymptotically, and if the capillary tubes were long enough. Indeed, passed a certain depth, the interface displacement was observed, as predicted by the NICCA model, to be exclusively controlled by gravity and viscous forces. For long capillary tubes, it was also shown that the interface velocity was proportional to the interface relative depth, i.e. the interface depth divided by the tube flow length. Furthermore, the velocities measured at the lower end of the capillary tube, or exit velocities, were shown to be independent of the capillary tube length, and well predicted by the NICCA model. Overall, the NICCA model was found to give good predictions provided that the ratio of tube flow length to diameter exceeded 600.

For the experiments performed on the longest 1.33 mm and 0.66 mm diameter capillary tubes, the measured exit velocities were found to be larger by about 10% than predicted by the NICCA model. Furthermore, for these experiments, the plot of
velocity as a function of depth was observed to be slightly non linear. Comparison of
the experimental data with the MFDV demonstrated that these observations were
consistent with the viscous force decrease associated with the 4-CT infiltration.
Indeed, as infiltration proceeds, 4-CT replaces water in the capillary tube. Because 4-
CT is a liquid of viscosity slightly less than that of water, the overall viscous force
decreases, and thus the observed velocity is slightly more than predicted by the NICCA
model. A non-linear relationship between interface depth and interface velocity is also
a feature predicted by the MFDV.

For short capillary tubes, as well as the early stages of infiltration into the longer
capillary tubes, agreement between the experimental data and the NICCA model
prediction was not found to be good. Specifically, it was observed that for any given
depth the measured interface velocity was always less than the NICCA predicted
velocity. Taking the mid-depth of the capillary tube as a reference depth, velocities
were measured that were between 20% and 50% of that predicted by the NICCA
model. The difference between model and measurement increased with decreasing
capillary tube length.

It was shown that for 2.70 mm diameter capillary tubes, the $\alpha_t$-number was not
always negligible with respect to unity, so that inertia forces may, at least partly,
contribute to the observed velocity reduction. In order to achieve exit velocities
identical to those of longer capillary tubes, the DNAPL/water column is submitted to a
larger acceleration, and thus larger inertia effects. This observation is consistent with
the fact that the difference between NICCA predicted velocity and measured velocity
increased with decreasing length.

However, for the experiments performed in 1.33 mm diameter capillary tubes,
disagreement between the experimental data and the NICCA model was observed even
though the $\alpha_t$-number was negligible with respect to unity. This suggests that a
resisting force unaccounted for by the NICCA model contributes to provide extra
resistance to the interface displacement. It is hypothesized that the extra resistance is
attributable to an increase of the capillary force with interface velocity. This increase is
believed to stem from the decrease in dynamic contact angle taking place as the
interface velocity increases. Because the NICCA model assumes that the dynamic
contact angle remains constant, the capillary force also remains constant, and thus the
NICCA model predicts velocities that exceed the actual velocities.

As a consequence of the capillary force increase with interface velocity, it is
believed that there exists a region in the upper part of the capillary tube over which
capillary effects are still important. For long tubes, the length-scale of the region is
comparatively small, and thus the NICCA model can still apply. However, the NICCA
model no longer works when the capillarity-controlled region length scale is significant
in comparison to the tube length.

As previously mentioned, that the NICCA model over-predicts interface
velocities below a ratio of flow length to diameter of about 600 does not mean that this
model is not useful in practice. For some real fracture systems, it can be expected that
the length of the fracture will be of the order of one meter or more, and that the
aperture will not exceed one millimeter, such that the ratio will largely exceed 600. For
those systems, the NICCA model can provide a good estimate of the time to complete infiltration. Furthermore, if the viscosity contrast between DNAPL and water is large, the MFDV can be used to provide a more accurate prediction of the infiltration time. For fracture systems where \( l/d \) is less than 600, the NICCA model provides a lower bound of the time required for complete infiltration. More generally, if other physical phenomena are present, such as diffusion/adsorption onto the solid matrix, or natural attenuation phenomena, the time to complete infiltration of the fracture system will be increased. Hence, again, the NICCA model provides a lower bound of the infiltration time, i.e. the smallest possible time to complete infiltration of the fracture system.

**NIVCA model**

In order to verify the capillary force increase hypothesis, the series of spontaneous infiltration tests reported above was compared to the NIVCA model. As previously mentioned, the NIVCA model was also compared to six controlled-head infiltration tests into a 1.33 mm diameter capillary tube.

For the NIVCA model prediction, the pinning forces were taken constant throughout the infiltration process and equal to the values estimated from the infiltration height measurements, i.e. 0.005 N/m, 0.010 N/m and 0.012 N/m for 2.70 mm, 1.33 mm and 0.66 mm diameter capillary tubes, respectively. Based on the experimental data, it was estimated that the DNAPL/water interface achieved perfect wetting when its velocity exceeded 3 mm/s. Based on these values, and on the value of the infiltration height measured for a given infiltration test, it was possible to back-calculated the static contact angle for each test, and generate a NIVCA model prediction.

For spontaneous infiltration tests into 1.33 mm diameter capillary tubes, it was found that the NIVCA model prediction was generally in good agreement with the experimental data. The pinning force of 0.010 N/m was consistent with most of the experimental results. Towards the lower end of long capillary tubes, the NIVCA model was observed to underestimate the velocity field. It was shown that the difference was compatible with the viscous force decrease that was taking place as 4-CT replaced water in the capillary tube. For shorter capillary tubes, a small extra resisting force was present in addition to the capillary resistance. This extra resistance, believed to be attributable to inertia effects, was found to be detectable for an \( \alpha \)-number larger than 0.05. The presence of small inertia effects for shorter capillary tubes was well supported by the fact that the back-calculated dynamic infiltration height increased with decreasing capillary tube length.

For controlled-head infiltration tests into 1.33 mm diameter capillary tubes, it was found that a pinning force of 0.015 N/m, instead of 0.010 N/m, was compatible with the experimental results. It could not be concluded whether or not the extra resistance was entirely attributable to capillary resistance, as the experimental setup might contribute to further pressure drop unaccounted for in the NIVCA and NICCA models.

For spontaneous infiltration tests into 2.70 mm diameter capillary tubes, it was found that the NIVCA model prediction generally over-predicted the experimental
data. It is believed that, for these tubes, two resisting forces beside viscosity are present: (1) the capillary forces (accounted for in the NIVCA model), which contribute to a large proportion of the resistance to flow; (2) inertia forces (unaccounted for in the NIVCA model), which increase in magnitude as the capillary tube length decreases, and contribute to part of the flow resistance along with capillary forces. The NIVCA model predictions were consistent with this trend and showed that the infiltration profile and velocity predictions are closest for the longest capillary tubes.

For the spontaneous infiltration tests into the 1201 mm long, 0.66 mm diameter capillary tube, it was found that the NIVCA model prediction was under-predicting the measured velocity field. Good agreement was obtained if a pinning force of 0.006 N/m was assumed instead of 0.012 N/m. The reason why a smaller pinning force value was needed is not precisely understood. The pinning force value of 0.006 N/m was later found consistent with the results of centrifuge infiltration tests into 0.66 mm diameter capillary tubes (see Section 8.1.7.3).

Overall, the NIVCA model was able to provide quantitative evidence that capillary forces can contribute to experimental velocities lower than predicted by the NICCA model. The experiments demonstrated that there exists a region in the upper region of the capillary tube over which capillary forces can influence the interface displacement. The length scale of this region was shown to be of the order of $|\Delta h'|$ corresponding to the difference between the maximum dynamic infiltration height and the infiltration height when the actual infiltration starts (see (7.17) in Section 7.2.3.2). For capillary tubes of length much larger than $|\Delta h'|$, the region of the capillary tube where capillary forces control the flow is essentially negligible in length, and the NICCA model prediction (or MFDV) can be used instead of the NIVCA model prediction. For capillary tubes of length of the order of $|\Delta h'|$, the NIVCA model prediction must be used. Consequently, criterion (7.27) ($l \gg 2\sigma/|\Delta \rho \rho_0 g r_0$), see Section 7.2.3.5) can be used to assess whether or not the NICCA model can be used instead of the NIVCA model to predict the infiltration kinetics of DNAPL. Note that (7.27) is in addition to condition (7.29) ($\alpha_t < 1$), which must also be checked to ensure that inertia and entry drag forces are negligible with respect to the viscous forces. Criteria (7.27) and (7.29) generalize the empirical criterion $l/d > 600$ obtained from the experimental results and their comparison with the NICCA model.

In the NIVCA model, constant values of the pinning force for a given capillary tube diameter were chosen, based on which a value of the static contact angle at the onset of infiltration could be calculated for each infiltration test. However, the fact that the pinning force might vary between tests on the same capillary tube diameter cannot be excluded. Clearly, there is a need for an independent measurement of the static contact angle and/or the pinning force. This point is further brought up in the recommendations for future research (see Section 8.3.2.2).
8.1.7 DNAPL Infiltration Centrifuge Experiments

8.1.7.1 Modeling-of-Models

The 4-CT infiltration laboratory experiment performed using the 1201 mm long, 0.66 mm diameter capillary tube and that performed using the 610 mm long, 1.33 mm diameter tube were reproduced as a series of equivalent reduced-scale centrifuge tests. For the 0.66 mm diameter capillary tube, a scale factor $n = 10$ was adopted. That is, the system dimensions were reduced to a tenth of their original scale, and centrifuged at an acceleration equivalent to 10 times the Earth’s gravity. For 1.33 mm diameter capillary tubes, scale factors, $n$, equal to 5, 10 and 15 were adopted, respectively.

For the 0.66 mm diameter capillary tubes, it was found that the reduced-scale centrifuge infiltration experiments are successful at reproducing the infiltration of the full-scale (1-g) experiment. For the 1.33 mm diameter capillary tubes, it was found that the scale factor $n = 5$ is successful at mimicking the full-scale experiment, but that larger scale factors yield an equivalent prototype kinetics with velocities that are 25%-50% slower than those of the full-scale model. These results were shown to be very consistent with the $\alpha_n$-number back-calculated for each of the centrifuge experiments. Inertia forces were found to have little impact on the reduced-scale centrifuge experiments on the 0.66 mm diameter capillary tubes, while they were predicted to have some effect on the reduced-scale centrifuge experiments on the 1.33 mm diameter capillary tubes, especially at the scale factors 10 and 15. Consequently, under the control of inertia, the equivalent prototype infiltrations corresponding to these tests were found to be slower than that of the full-scale prototype where the effects of inertia were unimportant.

Overall, these experimental results demonstrated that there exists a limit to the centrifuge scaling factor that can be used to model DNAPL transport in capillary tubes, and more generally in a fracture system. The $\alpha_n$-number sets the bounds for the range of scale factors that can be used in the modeling of a specific prototype. Note that a similar number can be defined for any two-phase flow process (in porous or fractured media) modeled using the geotechnical centrifuge.

8.1.7.2 Prototype Infiltration Height Measurements and Model Prediction

The results of 71 4-CT infiltration centrifuge tests into 2.70 mm, 2.20 mm, 1.33 mm and 0.66 mm diameter capillary tubes were examined. Similar to laboratory experiments, a large scatter of prototype infiltration height measurements was noted. Relative standard deviations of up to 16% were measured for a given tube diameter.

Very good agreement was noted between the measured prototype infiltration heights and the predicted prototype infiltration height, $H_i$, given by the infiltration criterion (7.36) (see Section 7.3.3.1), where the pinning forces and static contact angle
range are taken equal to the values obtained from the laboratory experiments. This suggests that the capillary tube entry pressure is more or less identical for a given capillary tube diameter in both the centrifuge and laboratory (1-g) experiments.

The causes for variability of the prototype infiltration height for centrifuge experiments are believed to be similar to those of laboratory experiments (i.e. variability of the static contact angle, interfacial tension, and possibly pinning force). Furthermore, uncertainty associated with the g-level measured at infiltration and the model DNAPL pool height—set prior to the experiment—may contribute to more uncertainty than in the case of laboratory tests.

While no correlation was detected between g-level and prototype infiltration height, the experimental results showed that for both 1.33 mm and 0.66 mm diameter capillary tubes, capillary tubes of longer model length were associated with slightly larger prototype infiltration heights. At present, the precise reason why this was the case is not clearly understood. No systematic length effect has been detected for laboratory tests, suggesting that an unknown experimental bias may be present in the case of centrifuge experiments. On the other hand, it is possible that larger accelerations associated with shorter tubes may easily overcome the pinning force and contact angle feedback mechanism of pre-infiltration, and thus lead to tube infiltration for smaller DNAPL pool heights.

8.1.7.3 Prototype Infiltration Kinetics and Model Prediction

The kinetics of a selection of 12 4-CT infiltration centrifuge tests was examined and compared to the NICCA, MFDV and NIVCA models. These 12 tests were all infiltration tests into 0.66 mm diameter tubes, for which $\alpha_n \ll 1$, and thus for which the effects of inertia were believed to be unimportant. For this series of tests, scaling laws were used to convert the experimental data of the reduced-scale model to those of the equivalent full-scale prototype.

In addition, to better understand the effects of inertia and capillary forces on the reduced-scale model, the mid-depth velocity, i.e. the prototype interface velocity at the mid-depth of a given capillary tube, was compared to that predicted by the NICCA model for a selection of 29 infiltration centrifuge tests into 1.33 mm diameter tubes and 12 into 2.70 mm diameter capillary tubes.

Comparison with NICCA, MFDV and NIVCA models

For 0.66 mm diameter capillary tubes centrifuge tests, it was found that, although in overall good agreement with the NICCA model, the measured prototype infiltration kinetics tends to be slower than that predicted by NICCA. For some tests, it was noted that the plot of velocity versus depth was non-linear—in contradiction with the NICCA model prediction—and that the measured exit velocity was sometimes larger than predicted. These observations were shown to be qualitatively consistent with the
MFDV prediction. That is, the non-linearity and larger exit velocity can be attributed to the decrease of the viscous force associated with the 4-CT infiltration. Furthermore, non-linearity could also be created by the increase in g-level along the capillary tube, since the g-level at any point depends on the centrifuge radius of rotation at this point.

Because the effects of inertia were believed to be unimportant for the centrifuge infiltration tests into 0.66 mm diameter tubes, the difference between the NICCA-predicted velocities and those measured from the experimental data was attributed to the increase in contact angle taking place at the interface. It was demonstrated that, so long as the experimental data lie in the perfect wetting region, scaling laws could be used to obtain a NIVCA model prediction of the equivalent prototype. The experimental results were then compared to the NIVCA model prediction and shown to be very well predicted by the model when using a pinning force of magnitude comprised between 0.006 N/m and 0.012 N/m.

Influence of capillarity and inertia

To further examine the role played by capillary and inertia forces on the infiltration velocity of centrifuge tests, the experimental prototype mid-depth velocities of centrifuge infiltration tests into 0.66 mm, 1.33 mm and 2.70 mm diameter capillary tubes was compared to those predicted by the NICCA model.

For 0.66 mm diameter capillary tubes, no correlation was detected between the mid-depth velocity and the \( \alpha_n \)-number, thereby confirming that inertia forces are not important for these tests. For 1.33 mm diameter capillary tubes, a large reduction in mid-depth prototype velocity was associated with the \( \alpha_n \)-number, which for these tests varied between 0.2 and 2.6. These findings confirmed the impact of inertia forces. For 2.70 mm diameter capillary tubes, the \( \alpha_n \)-number varied between 2.1 and 7.6. The measured velocities were found to remain constant at less than half the prototype velocity predicted by the NICCA model.

The measured prototype infiltration height was found to have an impact on both the infiltration tests into 0.66 mm diameter tubes and those into 1.33 mm diameter capillary tubes. For these tests series, it was found that, generally, the larger the measured prototype infiltration height, the larger the prototype mid-depth velocity. It was demonstrated that these results were consistent with the NIVCA model. Indeed, a large value \( H_i \) raises the value of \( \Delta H' \) (see (7.45) in Section 7.3.5.1), which, in turn, increases the value of the interface velocity. Such a feature would not be predicted by the NICCA model. Note that the influence of \( H_i \) on the interface velocity creates, in turn, a correlation between the capillary tube model length and the interface velocity, because of the observation that \( H_i \) is influenced by the tube model length (see Section 8.1.7.2).

Clearly, the variability of the infiltration height and its impact on interface displacement, the potential effect of inertia forces, and the dependence of dynamic contact angle upon interface velocity are factors that influence the centrifuge modeling process. If not negligible, these factors need to be accounted for in the interpretation of the centrifuge test data. Nonetheless, the validity of the scaling laws for the DNAPL
infiltration centrifuge experiments has been successfully proven, thereby suggesting that the geotechnical centrifuge can be used to examine general DNAPL behavior in simple fracture systems. Suggested improvements to the current centrifuge experimental setup as well as recommendations for future research are proposed in Section 8.3.3.

8.2 Conclusions

In Section 1.2.1, the objectives of the research work presented in this thesis were introduced. The scope of this project was three-fold:

1. To develop a theoretical model for predicting pure-phase DNAPL infiltration into vertical, initially water-saturated fractures.
2. To validate the theoretical model experimentally by conducting a testing program on simplified fractures described by smooth-walled, vertical capillary tubes.
3. To assess the potential of the geotechnical centrifuge as an experimental tool to carry out physical modeling of DNAPL infiltration behavior in fracture systems.

With respect to the first and second objectives, a model of DNAPL infiltration was developed that provided both a criterion for DNAPL infiltration and a prediction of the infiltration kinetics. The model was then evaluated by performing DNAPL infiltration experiments in vertical, circular section capillary tubes of different diameters. Based on the experimental results, the model was modified and refined. The following conclusions can be drawn:

1. The DNAPL infiltration criterion based solely upon (1) the interfacial tension capillary resistance and (2) the assumption of contact angle perfect wetting, both commonly used in the literature for predicting DNAPL pool heights prior to infiltration in fracture systems, is a misleading criterion. It was demonstrated that contact angle hysteresis and interface meniscus pinning can influence the pool height required to infiltrate a fracture modeled by a capillary tube, and also contribute to its variability. A new infiltration criterion was developed that incorporates the effects of contact angle hysteresis and those of the pinning force. The observed pre-infiltration phenomenon and infiltration height variability are well justified using this new infiltration criterion.

2. During the DNAPL infiltration process, the DNAPL/water interface contact angle reduces with velocity, leading to an increase of capillary forces with velocity along a so-called capillarity-controlled region. This increase in capillary forces may significantly impact the DNAPL infiltration kinetics if the capillary tube length is comparable to the scale-length of the capillarity-controlled region. A series of sub-models (NICCA, NIVCA and MFDV) of the DNAPL infiltration kinetics were developed that successfully predicted the infiltration experiments into fractures modeled by capillary tubes. The sub-models incorporated dynamic pinning effects and, for the NIVCA sub-model, a velocity-dependent dynamic contact angle.
3. The infiltration experiments showed the range of applicability of the sub-models: (1) Provided that the local and convective inertia forces, as well as the entry drag forces, have no measurable effect (corresponding to $\alpha_t << 1$, see (7.28)-(7.29) in Section 7.2.3.5), the sub-models are generally applicable (for this experimental program, measurable inertia effects were detected for $\alpha_t > 0.05$). (2) Provided that the length of the capillary tube largely exceeds the capillarity-controlled region length scale (see (7.26)-(7.27) in Section 7.2.3.5), the increase of capillary forces with velocity has little impact on the infiltration kinetics, and the NICCA model and/or the MFDV, which assume that the capillary forces remain constant, can be used to predict the infiltration kinetics.

With respect to the third objective of this research, results of centrifuge experiments of DNAPL infiltration in vertical capillary tubes suggest that the geotechnical centrifuge can be used under certain conditions to examine DNAPL infiltration behavior in simple fracture systems. The following conclusions can be drawn:

1. Scaling laws used to translate the experimental data of infiltration height from the scaled model to its equivalent full-scale prototype are applicable. The variability of the DNAPL prototype infiltration height of centrifuge experiments is consistent with that of laboratory experiments.
2. Scaling laws used to translate the experimental data of the infiltration kinetics from the scaled model to its equivalent prototype are applicable provided that the inertia forces are negligible in the reduced-scale model (and thus in the equivalent prototype), and that the DNAPL/water interface contact angle is in a perfect wetting regime, i.e. in the region of the capillary tube where capillary forces are independent of the interface velocity.

8.3 Suggested Improvements and Recommendations for Future Research

8.3.1 Interfacial Tension Measurements Using the Pendant Drop Method

In Section 5.6.3.3, it was concluded that the precise cause of the decrease with time of the interfacial tensions of 4-CT with water, as well as that of TCE with water, was not well understood. That there was an active surface phenomenon was thought to be likely. Nonetheless, it was not clear if this phenomenon could be attributed to the presence of dissolved air or any other impurity. It is thus recommended that the pendant drop method be further investigated in tandem with DNAPL infiltration studies. Suggested improvement on the drop edge detection, drop illumination and temperature control were listed in Section 5.6.3.4.
8.3.2 DNAPL Infiltration Laboratory Experiments

8.3.2.1 Equipment Improvement

Better control of the DNAPL pool built-up

One of the difficulties associated with the spontaneous infiltration tests is the proper control of the DNAPL pool built-up. During the experimental program reported in this thesis, the pool was slowly built up using a syringe by deposing drops of DNAPL in the reservoir tube on top of the capillary tube (see Section 6.2.2 and Figure 6.5). Overall, the process was tedious. The size of the DNAPL drops was poorly controlled, because the syringe was operated manually. The needle and syringe were often taken off the reservoir tube to be replaced and/or refilled with DNAPL, thereby disturbing the pool level. A finer control of the pool built-up process would enable studying the changes in pre-infiltration depth as a function of the DNAPL pool, and would likely contribute to less variability in the measurements of infiltration height. Varying the total time of DNAPL pool build-up with proper control of the built-up process would also enable studying the effect of interface aging and interfacial tension reduction on the DNAPL infiltration pool height.

One way of improving the pool built-up process is to permanently connect a needle and syringe to the DNAPL pool. The diameter of the reservoir tube could be made sufficiently large so that the presence of the needle and syringe reservoir does not disturb or influence the DNAPL pool head. The piston pushing DNAPL in the syringe could be controlled using a screw mechanism, which for every turn would push the piston and deliver a precise amount of DNAPL to the pool.

Improved connection between reservoir tube and capillary tube

For all laboratory infiltration experiments, as well as for centrifuge experiments, the connection between a capillary tube and a reservoir tube was made using chemically resistant Teflon seal tape (see Figure 6.2 and Section 6.2.2). The width of the Teflon tape was approximately equal to 15 mm, thereby partly blocking the view of the upper part of a capillary tube. The location of the Teflon tape collar was varied between tests, from 5 mm to 60 mm below the top of the capillary tube, so as to observe different parts of the early stages of the DNAPL infiltration.

Making tube connections using Teflon tape was quick and easy. The tubes could be disconnected at the end of a test for cleaning and later reusing. Different capillary and reservoir tubes could be connected together. Had an epoxy been used instead to make the tube connections, the process would have taken time and would have been permanent. Furthermore, the epoxy could have accidentally obstructed the capillary tube bore, and reacted—or at least been permanently stained—with DNAPL.
Nonetheless, for laboratory spontaneous infiltration tests, the presence of the Teflon tape partly blocked the view of the pre-infiltration finger and made it impossible to completely observe the displacements of the DNAPL/water interface. In some cases, the actual infiltration took place when the interface was still hidden by the Teflon tape. Thus, for these tests, it was impossible to obtain a precise measurement of the infiltration pool height.

Clearly, it would be useful to redesign the connection between the capillary and the reservoir tubes. For example, for their study on horizontal capillary tubes, Legait and Souriau [1985] made used of fused connections between two capillary tube pieces of different diameters. A fused connection between a capillary tube and its reservoir would need to be custom-made, and would require the services of a scientific laboratory glass blowing company (e.g. G. Finkenbeiner Inc., Waltham). Furthermore, similar to epoxy connections, the connection would be permanent, and would require several custom-made pieces if several tube diameters were to be investigated. Nonetheless, such system would enable studying DNAPL pre-infiltration and/or the early stages of DNAPL infiltration without the hassle of losing data points at the upper end of the capillary tube.

8.3.2.2 Suggestions for Further Research

Further infiltration testing into 0.66 mm diameter capillary tubes

As concluded in Section 7.2.2.5 and Section 7.2.3.6, more spontaneous infiltration experiments are recommended in 0.66 mm diameter capillary tubes in order to obtain a better estimate of the pinning force for this diameter, and to examine the effects of the capillary tube length on the infiltration profile. Comparison with the NICCA, MFDV and NIVCA models was obtained for only one test in a 0.66 mm diameter capillary tube of length 1202 mm, i.e. a very long length in comparison to tests performed in other capillary tube diameters. Thus, more tests are needed at a diameter of 0.66 mm to further corroborate the results of infiltration experiments into 1.33 mm and 2.70 mm diameter capillary tubes.

Tests in small tube diameters such as 0.66 mm are believed to be of great interest to further investigate the effects of the capillary forces. It is hypothesized that, for 4-CT, and possibly for other DNAPLs, most of the dynamic contact angle changes are taking place throughout the range of interface velocities 0 mm/s-10 mm/s, corresponding to the range of velocities observed in 0.66 mm diameter tubes. Complementary investigation of 4-CT infiltration experiments into 0.66 mm diameter capillary tubes would thus enable to better define the capillary force changes with velocity while staying away from the range of velocities for which inertia effects become influential.
Further controlled-head infiltration testing

As discussed in Section 7.2.3.3, the results of controlled-head infiltration tests into 1.33 mm diameter capillary tubes were quite inconclusive. This is because the presence of an extra pressure drop, unaccounted for in the model predictions, could not be entirely excluded. Controlled-head infiltration tests enable setting of the DNAPL pool height prior to the beginning of the experiment and prevent the effects of pre-infiltration. However, a drawback is that the DNAPL infiltration height for a given test is not measured and thus is not precisely known.

Further controlled-head infiltration testing in 1.33 mm diameter capillary tubes (or smaller diameter) would show whether the extra resisting force measured during the experimental program of this thesis can be attributed to the experimental setup (experimental bias), or if there exists a specific pressure drop associated with controlled-head infiltration tests.

Further investigation into dynamic contact angle-interface velocity relationship and pinning forces

In Section 7.2.3.4, it was concluded that the infiltration height measurement provided information on the magnitude of the static contact angle and of the pinning force, but that an independent measurement of the two was missing. Furthermore, this experimental program estimated that perfect wetting of a 4-CT/water interface was reached for an interface velocity of 3 mm/s, but did not have measurements of the dynamic contact angle to support this assumption. Thus, an experimental program specifically focusing upon the measurements of the pinning force and the changes in dynamic contact angles would be desirable to provide a better understanding of the changes in capillary resistance taking place during the infiltration process.

Measurements of dynamic contact angles would need to be corrected for the radial magnification of the interface meniscus due to the capillary tube refraction [see, for example, Hoffman, 1975]. If an experimental setup similar to that used for spontaneous infiltration tests was used, contact angle measurements would be complicated by the presence of the tank tube containing the bulk of water (see, for example, Figure 6.2.c), which would also contribute to the magnification of the radial dimensions of the interface. Thus, a simpler experimental setup is recommended (refer to Figure 2.30 for example). Guidelines for such experimental program are listed below:

1. Investigation of the DNAPL/water interface displacement in horizontal capillary tubes under a configuration of forced wetting and using an experimental setup similar to that shown in Figure 2.30 [see also, for example, Chittenden and Spinney, 1966; Blake et al., 1967, Fermigier, 1991; Calvo et al., 1991]. For small diameter horizontal tubes, the effects of gravity can generally be ignored. Under conditions of forced wetting in horizontal tubes, the interface velocity can be controlled and made constant.
2. Direct measurement of the dynamic contact angle at the DNAPL/water interface. From independent knowledge of the pressure drop across the capillary tube and the dynamic contact angle, it is then possible to obtain a measurement of the pinning force. These measurements are repeated at different interface displacement rates to establish a dynamic contact angle-interface velocity relationship, and to verify if the pinning force is independent of velocity.

3. The dynamic contact angle can be measured using an optical technique similar to that used by Hoffman [1975] or Fermigier and Jenffer [1991] where the capillary tube moves and the interface remains essentially stationary. Alternatively, a system can be designed where a traveling microscopic camera mounted on a rail parallel to the capillary tube follows the interface displacement. The displacement of the camera can be automated using a feedback loop driven by an infrared detector, which is mounted next to the camera and senses the motion of the interface (color change between 4-CT and water). Such an automated system would be particularly useful if, instead of having a constant rate of displacement, the interface velocity was varied during the infiltration process by modifying the pressure drop across the capillary tube. Indeed, the traveling camera could record the instantaneous changes of the meniscus shape, so that the images could provide a complete dynamic contact angle/velocity curve from a single infiltration experiment. This system could also be used for controlled-head infiltration tests (in vertical tubes) where the velocity changes throughout the infiltration process. Under this configuration, it would be possible to investigate the effect of gravity on the meniscus shape and the dynamic contact angle.

4. The forced displacement experiments can be repeated for different tube diameters in order to see if the tube diameter has an effect on the dynamic contact angle-interface velocity relationship, and on the magnitude of the pinning force.

Numerical solution of the interface displacement governing equation when the effects of inertia are not negligible

As noted in Section 7.2.3.4, although the \( \alpha_t \)-number determines whether or not the magnitude of inertia forces are important compared to the viscous forces, it does not provide any estimate of the observed velocity reduction associated with the inertia effects. For example, it is possible that a \( \alpha_t \)-number of the order of 0.5—significant compared to unity—contributes only a small fraction of the observed interface velocity reduction in comparison to the reduction attributable to capillary effects. The numerical solution of a governing equation that would include the effects of inertia (local and convective), as well as capillarity, is thus needed to resolve this issue. A numerical solution would enable prediction of the experimental results where inertia is thought to be important, such as in the case of laboratory experiments into 2.70 mm diameter capillary tubes. It would also provide a prediction for the centrifuge infiltration experiments into 1.33 mm and 2.70 mm diameter capillary tubes.
8.3.3 DNAPL Infiltration Centrifuge Experiments

8.3.3.1 Equipment Improvement

*Improved experimental setup to observe the early stages of the infiltration*

Similar to the laboratory experiments, a connection between reservoir tube and capillary tube was made using Teflon tape. This connection blocked the observation of the DNAPL infiltration at its early stages (see Section 8.3.2.1). In the case of centrifuge infiltration experiments, the holding rack (see Figure 6.10) also blocked part of the view, so that it was not possible to obtain infiltration data from the top of the capillary tube down to a depth of 20 mm-30 mm. Alternative modes of tube connection and support would be desirable in order to properly observe the early stages of the infiltration process.

Information of the early stages of the interface displacement (i.e. above a depth of 20 mm-30 mm) would be useful for the centrifuge experiments, as it is believed that the contact angle reduction and associated capillary force increase take place in this region. Experimental data in this region would thus enable one to verify whether capillary forces can be completely scaled in a reduced-scale centrifuge model. Images of the interface at the early stages of the infiltration would also show whether or not pre-infiltration initially takes place in centrifuge infiltration experiments. Finally, detecting infiltration at its early stages would improve the accuracy of the measurement of g-level at infiltration.

For the experimental program reported in this thesis, up to eight capillary tubes were set up in the centrifuge strong box, so that several infiltration experiments were run during a single centrifuge test. The centrifuge platform camera was mounted with a wide-angle camera, so that all of the experiments could be filmed simultaneously. This process provided large amounts of infiltration experiment data with relatively few centrifuge tests. On the other hand, the resolution of a single infiltration experiment was limited. An alternative to this experimental setup would be to film a single infiltration experiment per centrifuge test, thereby capturing closer-range images of the DNAPL/water interface, particularly at the early stages of infiltration. Obviously, the field of view of the camera must be larger than the total length of the capillary tube.

8.3.3.2 Suggestions for Further Research

*Capillary rise experiments to verify the scaling of capillary forces*

Although the influence of capillary forces was demonstrated for the centrifuge infiltration experiments reported in this thesis, the infiltration process of these experiments appeared mostly controlled by gravity and viscous forces, as well as
inertia forces when these were not negligible. Thus, this experimental program did not specifically prove that dynamic capillary forces were properly scaled in a reduced-scale model.

As discussed in Section 2.4.2, in experimental studies of capillary rise in sand columns, where the magnitude of the (driving) capillary forces is as important as that of the (resisting) viscous and gravity forces, scaling of the kinetics of capillary rise was quite inconclusive [Depountis et al., 2001]. This thesis hypothesized that scaling of dynamic capillary forces was complicated by the dependence of the dynamic contact angle upon velocity.

Thus, it is suggested that a series of capillary rise experiments be run in the centrifuge using vertical capillary tubes similar to those used for the experimental program of this research. A viscous liquid, such as a silicon oil of high viscosity, could be used to minimize the effects of inertia at the early stages of the rise [see, for example, capillary rise experiments of Joos et al., 1990]. Air could be used as the displaced, non-wetting fluid. A series of laboratory and reduced-scaled centrifuge experiments would be used to perform modeling-of-models and verify whether or not dynamic capillary forces can be successfully scaled using centrifuge modeling.

8.3.4 DNAPL Infiltration Using Rough-Walled Fracture Systems

8.3.4.1 Experimental Investigation

As previously discussed, the NICCA model was extended in Chapter 3 to rough-walled fractures where homogeneous hydraulic properties can be defined. Thus, experiments of infiltration in replicas of rough-walled fracture systems should be performed to verify if, and under which conditions, the model is valid. These experiments could also be used to investigate the relative importance of interfacial tension and pinning forces in rough-walled fracture systems, and measure capillary pressure-saturation relationships, as well as capillary pressure-relative permeability relationships. These experiments could be carried out both in the laboratory and the centrifuge environments.

In order to investigate the DNAPL infiltration kinetics, the fracture replicas should be made of a transparent material. The transparent material could be one of the following:

1. Transparent epoxy replicas [see, for example, Persoff and Pruess, 1995; Kneafsey and Pruess, 1998]. One of the drawbacks of this technique is the potential of DNAPLs for staining the epoxy and possibly attacking the epoxy surface. Also, unlike most real fracture surfaces, the epoxy surface may be somewhat hydrophobic [Wan et al., 2000]. To the author’s knowledge, there is no published study of DNAPL transport in fracture replicas made out of epoxy. Existing studies have
used air or an LNAPL as the non-wetting fluid [see, for example, Persoff and Pruess, 1995; Kneafsey and Pruess, 1998].

2. Roughen glass plates obtained by sand blasting [Schwille, 1988; Amundsen et al., 1999], etching with hydrofluoric acid [Theodoropoulou et al., 2001], or gluing a layer of glass beads [Fourar et al., 1993]. One drawback is that it is not clear whether roughen glass plates constitute realistic replicas of rough-walled fractures [Wan et al., 2000].

3. A glass cast of a rock fracture surface. A method was recently developed for fabricating transparent replicas of fracture surfaces using glass [Wan et al., 2000]. Although a very promising method, one drawback is that the fabrication process is rather long and difficult. Nonetheless, the cast surface provides a good reproduction of the fracture topography.

8.3.4.2 Numerical Investigation

Several studies have attempted to model porous media as a group of capillary tubes of various sizes or as a periodic tube, and made use of the Lucas-Washburn equation (see Section 2.3.2) to investigate the penetration of a liquid in the idealized porous medium. In one study, the porous medium was idealized as a circular section capillary tube with periodic step changes in its diameter [Dullien et al., 1977]. In another study, the idealized model consisted of a sequence of conical sections alternatively converging downwards and upwards [Levine et al., 1980]. Capillary rise was also considered between two sinusoidally corrugated plates, where the corrugations were in a plane normal to the flow direction [Borhan and Rungta, 1993].

Using an approach similar to that used above for porous media, one could model a rough fracture as a series of discrete rectangular capillary tube elements of an aperture that is the average aperture of the fracture at the location of the element. Using this technique, the larger the number of capillary tube elements, the smaller the fracture resolution. The NICCA, MFDV and NIVCA models could then be used to generate a prediction of the DNAPL infiltration kinetics in the idealized rough fracture. Capillary pressure-saturation relationships and relative permeability-saturation relationships could then be obtained and compared to the results of conventional percolation models (see Section 2.2.3.1). A similar approach could be developed for DNAPL remediation scenarios, such as hydraulic flushing or enhanced flushing.

Ultimately, the coupling of a fracture system model [e.g., Wu and Pollard, 1992; Ivanova, 1998] that describes the orientation and distribution of a fracture network at a site with a rough-walled fracture flow model could be applied at waste sites, where information from outcrops, boreholes and drilling cores would enable the estimation of average fracture properties. A combination of numerical developments, laboratory experiments and centrifuge experiments, as presented in this thesis, is one approach that can be used to advance the field in this direction.
8.4 References


APPENDIX A

CENTRIFUGE DATA ACQUISITION CODE

The following data acquisition program is written for the AD1170 data acquisition card in QuickBasic. The reader is referred to the discussion of Section 6.4.3.

DECLARE SUB Estimrpm ()
DECLARE SUB Estimglevel ()
'DECLARE SUB Switch ()
DECLARE SUB ReadFile (File$)
DECLARE SUB SetUpDataFile ()
DECLARE SUB WriteToFile ()
DECLARE SUB GetVoltage ()
DECLARE SUB PrintEngData ()
DECLARE SUB Pause ()
DECLARE SUB SetUpScreen ()
DECLARE SUB SetUpDAQ (Card)
DECLARE SUB IntroScreen ()
DECLARE SUB Printmask (Row!, Column!, Number!, Places!)
DECLARE SUB CenterText (Column!, Text$)
DECLARE SUB PrintLabels (Row!, Column!, Text$)
DECLARE FUNCTION Voltage (Channel!, Gain!, Card!)
DECLARE FUNCTION Time ()
DECLARE FUNCTION Max (Numbera, Numberb)
DECLARE SUB Introparam ()
DECLARE SUB GetTime ()
DECLARE SUB CreateFile ()
DECLARE SUB Choice ()

DIM SHARED lv!
DIM SHARED hv!
DIM SHARED cf
DIM SHARED rpm
'DIM SHARED glevel
DIM SHARED height
DIM SHARED AD1170(2)
DIM SHARED Multiplex(2)
DIM SHARED IntTime
DIM SHARED IntBit
DIM SHARED DAQFileName$
DIM SHARED InputFileName$
DIM SHARED TimeNext
DIM SHARED InitialTime
DIM SHARED DeltaTime
DIM SHARED TimeInterval
DIM SHARED StartDate$
DIM SHARED Clock

'Voltage reading from each channel
DIM SHARED LowVolt(6)
DIM SHARED Highvolt(6)

CLS
IntroScreen
Pause
Choice
Introparam
SetUpDataFile
Pause
SetUpScreen
InitialTime = TIMER
StartDate$ = DATE$
DO
    GetTime
    IF ((DeltaTime) / TimeInterval) > Clock THEN
        GetVoltage
        Estimrpm
        PrintEngData
        WriteToFile
    END IF
LOOP WHILE INKEY$ <> CHR$(27)

SUB CenterText (Column, Text$)
    length = LEN(LTRIM$(RTRIM$(Text$)))
    start = (80 - length) / 2
    PrintLabels Column, start, Text$
END SUB

SUB Choice
CLS
PRINT
PRINT
INPUT "Do you want to create a parameter file (y/n)? ", a$
IF a$ = "y" OR a$ = "Y" THEN
  CreateFile
ELSE
  CLS
  PRINT
  PRINT
  INPUT "Enter parameter file name: ", InputFileName$
END IF
END SUB

SUB CreateFile
  INPUT "Enter name of new parameter file: ", InputFileName$
  INPUT "Enter number of low voltage channels: ", a
  INPUT "Enter number of high voltage channels: ", b
  INPUT "Enter time interval between readings (sec): ", c
  PRINT "Integration time: ">
  PRINT "  1. 1 ms";
  PRINT "  2. 10 ms"
  PRINT "  3. 16.67 ms"
  PRINT "  4. 20 ms"
  PRINT "  5. 100 ms"
  PRINT "  6. 166.7 ms"
  PRINT "  7. 300 ms"
  INPUT "Select integration time: ", selection
  d = 15 + selection
  'calibration factor of the second tachometer
  cf = 100.0
  'below is the calibration factor for the first tachometer
  'code lines kept as comment'
  'INPUT "Enter maximum RPM of your test: ", rp
  'IF rp <= 75 THEN
  '  cf = 16.2444
  'ELSE
  '  IF rp > 75 AND rp <= 225 THEN
  '    cf = 46.0048
  '  ELSE
  '    IF rp > 225 AND rp <= 350 THEN
  '      cf = 72.7184
  '    ELSE
  '      cf = 119.682
'END IF
'END IF
'END IF

'INPUT "Do you wish to specify a particular radius to calculate the G-level (y/n)? ", a$
'IF a$ = "y" OR a$ = "Y" THEN
  'INPUT "Enter height (in cm) from base of platform at which you want the G-level to be calculated: ", height
'ELSE
  'PRINT "The G-level will be calculated at the center of gravity of the platform."
  'height = 8.255 'center of gravity
'END IF
'radin = 41.5 + 8.75 - height / 2.54
'radcm = 2.54 * radin
'PRINT "At high RPM, the radius at which the G-level will be calculated is: ", radin,
'PRINT " inches, or ",
'PRINT radcm,
'PRINT " cm"
OPEN InputFileName$ FOR OUTPUT AS #1
PRINT #1, a, b, c, d, cf
CLOSE #1
'Pause
END SUB

SUB Estimrpm
  rpm = cf * Highvolt(1)
  'glevel = (41.5 + 8.75 - height / 2.54) * rpm ^ 2
END SUB

SUB GetTime
  DeltaTime = Time - InitialTime
  TimeNext = -DeltaTime + Clock * TimeInterval
  IF TimeNext > 0 THEN
    Printmask 6, 63, TimeNext, 2
  ELSE
    Printmask 6, 63, 0, 2
  END IF
END SUB

SUB GetVoltage
FOR i = 1 TO lv!
    LowVolt(i) = Voltage(i - 1, 10, 1)
NEXT i
FOR i = 1 TO hv!
    Highvolt(i) = Voltage(i - 1, 1, 2)
NEXT i
END SUB

SUB Introparam
    CLS
    PRINT
    PRINT
    ReadFile InputFileName$
    IntBit = 15  'bit precision
    SetUpDAQ (1)
    SetUpDAQ (2)
    'Switch
    CLS
    INPUT "Enter output data file name: ", DAQFileName$
    Clock = 0  'Initializes counter for data sampling
END SUB

SUB IntroScreen
    CLS
    CenterText 10, "Centrifuge Program"
    CenterText 11, "Version 2.0 - 3/20/98"
    CenterText 12, "Written by Kurt Sjoblom,"
    CenterText 13, "But Tremendously Improved by"
    CenterText 14, "Catalina Marulanda and Laurent Levy"
END SUB

FUNCTION Max (Numbera, Numberb)
    IF Numbera > Numberb THEN
        Max = Numbera
    ELSE
        Max = Numberb
    END IF
END FUNCTION

SUB Pause
    CenterText 24, "Press any key to continue..."
DO
LOOP UNTIL INKEY$ <> ""
END SUB

SUB PrintEngData
FOR i = 1 TO lv!
    Printmask 2 + 2 * i, 12, LowVolt(i), 3
NEXT i
FOR i = 2 TO hv!
    Printmask 2 + 2 * i, 29, Highvolt(i), 3
NEXT i
Printmask 18, 37, rpm, 4
'Printmask 18, 40, glevel, 5
Printmask 4, 68, DeltaTime, 2 'prints elapsed time in seconds
'prints no. of readings written on data file
LOCATE 2, 63
PRINT Clock + 1
END SUB

SUB PrintLabels (Row, Column, Text$)
    LOCATE Row, Column
    PRINT Text$;
END SUB

SUB Printmask (Row, Column, Number, Places)
    Mask$ = "####.
    FOR i = 1 TO Places
        Mask$ = Mask$ + LTRIM$("#")
    NEXT i
    LOCATE Row, Column
    PRINT USING Mask$; Number;
END SUB

SUB ReadFile (File$)
    OPEN File$ FOR INPUT AS #1
    INPUT #1, lv!, hv!, TimeInterval, IntTime, cf
    'IntTime is the integration time of A/D converter
    'TimeInterval is the interval for data sampling in seconds
    CLOSE #1
END SUB
SUB SetUpDAQ (Card)
    'routine to setup data acquisition card
    'routine to be run once
    ' 768 = decimal I/O address of AD1170 A/D converter
    ' 776(old), 772(new) = decimal I/O address of multiplexer channel selector
    ' 15 = multiplexer connection to ground
    'StrawberryTree = 6928
    'Switch = StrawberryTree + 4
    Multiplex(Card) = 772 + (Card - 1) * 1024
    AD1170(Card) = Multiplex(Card) - 4
    'set default calibration time
    OUT AD1170(Card), 70: WAIT AD1170(Card), 1, 1
    'load data format into 2nd byte
    OUT AD1170(Card) + 1, IntBit
    'lock in the loaded data format
    OUT AD1170(Card), 48: WAIT AD1170(Card), 1, 1
    'begin background calibration
    OUT AD1170(Card), 176: WAIT AD1170(Card), 1, 1
END SUB

SUB SetUpDataFile
    OPEN DAQFileName$ FOR OUTPUT AS #1
    PRINT #1, "Centrifuge Program"
    PRINT #1, "Version 2.00, 3/20/98"
    PRINT #1, "Written by Kurt Sjoblom"
    PRINT #1, "But Tremendously Improved by"
    PRINT #1, "Catalina Marulanda and Laurent Levy"
    PRINT #1, DATE$, TIME$
    PRINT #1, "Time",
    FOR i = 1 TO lv!
        PRINT #1, "LowCh " + LTRIM$(STR$(i - 1)) + "",
    NEXT i
    FOR i = 1 TO hv!
        PRINT #1, "HighCh " + LTRIM$(STR$(i - 1)) + "",
    NEXT i
    PRINT #1,
    CLOSE #1
END SUB

SUB SetUpScreen
    SCREEN 10   '640x350 graphics
CLS
PrintLabels 2, 13, "Low Volt High Volt"
PrintLabels 3, 13, "-------- --------"
PrintLabels 4, 47, "Time at last reading:",
PrintLabels 4, 76, "secs"
PrintLabels 2, 47, "Last reading #:",
PrintLabels 6, 47, "Next reading in:",
PrintLabels 6, 71, "secs"
FOR i = 1 TO Max(lv!, hv!)
    PrintLabels 2 + 2 * i, 5, "Ch " + (LTRIM$(STR$(i - 1)))
NEXT i
PrintLabels 18, 13, "RPM at last reading:",
'PrintLabels 20, 13, "G-level at last reading:",
END SUB

'SUB Switch
'PRINT "Switch tachometer selector to position: ",
'SELECT CASE cf
    'CASE IS = 16.2444
        'PRINT "Up to 75 RPM"
    'CASE IS = 46.0048
        'PRINT "Up to 225 RPM"
    'CASE IS = 72.7184
        'PRINT "Up to 350 RPM"
    'CASE IS = 119.682
        'PRINT "Up to 400 RPM"
'END SELECT
'Pause
'END SUB

FUNCTION Time
    CurrentDate$ = DATE$
    IF StartDate$ = CurrentDate$ THEN
        Time = TIMER
    ELSE
        Time = 86400 + TIMER
    END IF
END FUNCTION

FUNCTION Voltage (Channel, Gain, Card)
    'Take regular readings
    OUT Multiplex(Card), Channel
OUT AD1170(Card), IntTime: WAIT AD1170(Card), 1, 1
'Convert to voltage
LowByte = INP(AD1170(Card) + 1)
MidByte = INP(AD1170(Card) + 2)
HiByte = INP(AD1170(Card) + 3)
Counts = LowByte + 256 * MidByte + 65536 * HiByte
'reenable background calibration
OUT AD1170(Card), 176: WAIT AD1170(Card), 1, 1
Voltage = (Counts * 10 / 2 ^ (IntBit + 7) - 5) / Gain
END FUNCTION

SUB WriteToFile
    OPEN DAQFileName$ FOR APPEND AS #1
    PRINT #1, DeltaTime,
    FOR i = 1 TO lv!
        PRINT #1, LowVolt(i),
    NEXT i
    PRINT #1, rpm,
    FOR i = 2 TO hv!
        PRINT #1, Highvolt(i),
    NEXT i
    PRINT #1,
    CLOSE #1
    Clock = Clock + 1
END SUB
APPENDIX B

CALCULATING G-LEVELS FROM RPM DATA

B.1 Background

This appendix describes in details how g-levels were calculated from centrifuge rotational velocity data acquired during the DNAPL infiltration centrifuge experiments (see experimental methodology in Section 6.3 and Section 6.4, and results presented in Section 7.3). Because the centrifuge rotational velocity at the onset of DNAPL infiltration was low, the effects of the Earth’s gravity were not always negligible and had to be accounted for in the derivation of the equivalent g-level. In Section B.2, the equations relating the g-level, the rotational velocity, the radius of gyration and the swinging platform inclination are derived. Section B.3 presents a series of applications of the equations derived in Section B.2.

B.2 Derivation of the G-Level-RPM Relationship

B.2.1 Statement of Equations

Consider the simplified schematic of the centrifuge rotary arm and main swinging platform shown in Figure B.1. The point of intersection of the arm and the centrifuge shaft is denoted A. The center of the platform trunnion axis is denoted C. The length of the arm from A to C is $r_c$. The height from the base of the platform to point C is $b_c$. Let $G$ be the center of gravity of the experimental package/platform system and $M$ a point of the axis of symmetry ($CG$) of the platform. The heights $y_G$ and $y$ are the distance from the base of the platform to point $G$ and point $M$, respectively. As shown in Figure B.1.a, when the centrifuge is not spinning, the platform is at rest and its axis of symmetry is vertical. When the centrifuge is spinning at a rotational velocity of $\omega$, the platform rotates about the trunnion axis, such that the resultant component of the Earth’s gravitational force and the rotational acceleration force is normal to the platform (see Figure B.1.b). Let $\psi$ be the angle made between the centrifuge arm and the axis of symmetry ($CG$) of the platform. Under these conditions, $\psi$ is equal to 90°
when the centrifuge is at rest, and decreases to $0^\circ$ when $\omega$ increases. The angle $\psi$ can be seen as the inclination of the centrifuge platform with the horizontal.

The purpose of this derivation is to estimate the magnitude of the gravitational force at point $M$, such that the equivalent $g$-level, $n$, at this point can be estimated. Let $r_G$ and $r_M$ be the radii of gyration at point $G$ and point $M$, respectively. The total gravitational acceleration, $a_t$, at point $M$ can be written as

$$a_t^2 = g^2 + (r_M \omega^2)^2,$$

Figure B.1. Schematic of the centrifuge arm and its main platform: a. At rest; b. During spinning at rotational velocity $\omega$. 

(b)
where the radius of gyration at point $M$ is given by

$$r_M = r_c + (b_c - y)\cos\psi.$$  \hfill (B.2)

The $g$-level at point $M$, $n$, is defined as

$$n = \frac{a_c}{g}.$$  \hfill (B.3)

Thus, making use of (B.1) and (B.2), $n$ can be written as

$$n = \sqrt{1 + \frac{\omega^4}{g^2} [r_c + (b_c - y)\cos\psi]^2}.$$  \hfill (B.4)

If the effects of gravity and platform inclination are neglected in (B.4), then $r_M\omega^2/g \gg 1$ and $\psi \approx 0$. Under these conditions, (B.4) is reduced to the classic $g$-level formula

$$n = \frac{(r_c + b_c - y)\omega^2}{g}.$$  \hfill (B.5)

Obviously, (B.4) tends to (B.5) if the centrifuge velocity, $\omega$, is large.

Equation (B.4) gives the value of the $g$-level at point $M$ as a function of its location $y$, the geometrical constants of the centrifuge $r_c$ and $b_c$, the centrifuge rotational velocity $\omega$, and the angle $\psi$. Since $\psi$ is an unknown, a second equation is needed for calculating $n$ as a function of the centrifuge velocity. Equilibrium of forces at the center of gravity of the experimental package/platform system leads to

$$\tan\psi = \frac{g}{r_c(\psi)\omega^2}.$$  \hfill (B.6)

Making use of (B.2) written at point $G$, and replacing in (B.6) gives

$$r_c \tan\psi + (b_c - y_G)\sin\psi = g/\omega^2.$$  \hfill (B.7)

Since $r_c$, $b_c$, and $y_G$ are all geometrical constants, (B.7) can be used to obtain the value of $\psi$ as a function of the centrifuge rotational velocity $\omega$. When $\omega$ is small, the solution of (B.7) is $\psi = 90^\circ$. On the other hand, if $\omega$ becomes very large, then the solution of (B.7) is $\psi = 0^\circ$. The solution $\psi$ of (B.7) can be used to compute the $g$-level, $n$, at point $M$ given by (B.4).
B.2.2 Calculation of Centrifuge Geometrical Constants

Before applying the equations derived in Section B.2.1, the values of the geometrical constants of the centrifuge, i.e. the values of $r_c$, $b_c$ and $y_G$, must be provided.

According to the centrifuge Genisco manual, the weight of each platform is approximately 68 kg (150 lb) and the center of gravity of each platform is located at the center of the platform under consideration, i.e. 235 mm (9-1/4 inches) below the axis of the trunnion. A thick aluminum plate is mounted on top of the main platform (see Section 6.3.2). The weight and dimensions of the plate are 10 kg and $533.4 \text{ mm} \times 520.7 \text{ mm} \times 12.7 \text{ mm}$ (21 inches $\times$ 20.5 inches $\times$ 0.5 inch), respectively. A quick calculation shows that the center of gravity corresponding to the plate/platform system is practically the same as that of the platform alone. Specifically, the center of gravity of the plate/platform system is located 234 mm below the axis of the trunnion, i.e. 1 mm above the plane of the platform.

The centrifuge strong box had external dimensions of approximately $370 \text{ mm} \times 130 \text{ mm} \times 260 \text{ mm}$, where 260 mm corresponds to the box height. The box was filled with water, such that its total weight was equal to 16 kg. Since the strong box was directly set up on the aluminum plate, the center of the box was located $235 \text{ mm} - 12.7 \text{ mm} - 260 \text{ mm}/2 \approx 92 \text{ mm}$ below the axis of the trunnion. It is assumed that no other experimental part but the box and its contents was significant to contribute to the overall weight of the experimental package. Based on this assumption, the center of gravity of the total system is found to be located $[92 \text{ mm} \times 16 \text{ kg} + 235 \text{ mm} \times (10 \text{ kg} + 68 \text{ kg})] / (16 \text{ kg} + 10 \text{ kg} + 68 \text{ kg}) \approx 210 \text{ mm}$ below the axis of the trunnion, or 12 mm above the top of the aluminum plate.

Taking the top of the aluminum plate as the reference plane, the values $y_G = 12 \text{ mm}$ and $b_c = 222 \text{ mm}$ are obtained. These values are the numerical parameters to be used in (B.7). As seen in Section 6.3.2, the value of $r_c$, also used in (B.7), is equal to 1067 mm (42 inches).

B.3 Application of Equations

B.3.1 G-Level at the Center of Gravity as a Function of RPM

Equation (B.7) is solved numerically to give the angle of inclination of the centrifuge platform, $\psi$, as a function of the centrifuge rotational velocity, $\omega$. The results are plotted in Figure B.2. As can be seen in the figure, the angle of inclination decreases sharply from $90^\circ$ to $11^\circ$ when the rotational velocity increases from 0 RPM to 60 RPM. The angle of inclination then proceeds to decrease more slowly, from $11^\circ$ to $0^\circ$, when the rotational velocity increases from 60 RPM to 400 RPM.
Figure B.2. Inclination of platform with horizontal (solid line) and radius of gyration of center of gravity of platform/experimental package system (dashed line) as a function of the centrifuge rotational velocity.

Figure B.3. $g$-Level at center of gravity as a function of centrifuge rotational velocity. The solid line includes the effects of gravity. The dashed line neglects gravity.
In the same figure, the radius of gyration of the center of gravity of the platform/experimental package system, \( r_G \), is plotted as a function of the centrifuge rotational velocity. The radius \( r_G \) is obtained by applying (B.2) at point \( G \). As can be seen in Figure B.2, \( r_G \) increases from \( r_C = 1067 \text{ mm} \) to a maximum of \( r_C + (b_c - y_G) = 1277 \text{ mm} \). However, it appears that large changes in radius of gyration are limited to the interval of rotational velocities 0 RPM-60 RPM. Indeed, at 60 RPM, \( r_G \approx 1273 \text{ mm} \). For any value of velocity larger than 60 RPM, the radius of gyration is close to its maximum of 1277 mm, with a difference of less than 0.4%. These results suggest that the Earth’s gravitational force has little influence over the overall gravitational force for a centrifuge rotational velocity larger than 60 RPM.

To further examine the effect of the Earth’s gravity, the \( g \)-level at point \( G \) is plotted in Figure B.3 versus the centrifuge velocity, \( \omega \). The solid line incorporates the effects of gravity and is obtained using (B.4) applied at point \( G \). The dashed line neglects the effects of gravity, assumes that the radius of gyration is always at its maximum of 1277 mm, and thus is obtained using (B.5) calculated at point \( G \). As can be seen in the figure, the effects of gravity are virtually negligible if the centrifuge rotational velocity exceeds 50 RPM. Specifically, the difference of \( g \)-level predicted by (B.4) and that predicted by (B.5) becomes less than 0.1 \( g \) if \( \omega \) exceeds a velocity of 55 RPM, for which the exact \( g \)-level at the center of gravity is equal to 4.3 \( g \).

### B.3.2 \( g \)-Level Variation Across a Capillary Tube

Because some of the DNAPL infiltration centrifuge experiments included tests run at \( g \)-levels below the velocity of 55 RPM, gravity was taken into account when computing the \( g \)-levels. Based on the dimensions of the strong box and tube holding rack (see Section 6.3.3), the location of the entrance to a capillary tube was estimated to be 140 mm above the aluminum plate, that is \( y = 140 \text{ mm} \). It was also assumed that the capillary tube was located at the axis of symmetry of the centrifuge platform. Although the capillary tube could be off the axis by a factor \( x \) and require a second correction on the radius of gyration equal to \( x \sin \psi \), this correction can be shown to be negligible at practical centrifuge rotational velocities.

For data reduction, it was assumed that the \( g \)-level was a constant along the capillary tube throughout the infiltration process. The \( g \)-level was taken equal to the \( g \)-level at the entrance to the capillary tube at the start of infiltration (see Section 6.3.4 and Section 7.3).

In practice, the radius of gyration varied along the capillary tube, and thus the \( g \)-level at a given point of the tube was also expected to be dependent upon the location of this point within the tube. To check whether the change in \( g \)-level along the tube could be significant, a 130 mm long capillary tube is considered (length of capillary tube of a typical test), such that \( y = 10 \text{ mm} \) at the lower end of the tube. In Figure B.4, the \( g \)-level calculated at both ends of the tube using (B.4) is plotted versus the centrifuge rotational velocity. As can be expected, the larger the centrifuge velocity,
the larger the $g$-level variation across the tube. At 400 RPM, the $g$-level contrast across the tube is 23 $g$ for a $g$-level of 206 $g$ at the entrance to the capillary tube. It should be noted, however, that the relative $g$-level contrast across the tube stays approximately constant at practical centrifuge velocity and increases from 10% to 11% when the rotational velocity increases from 50 RPM to 400 RPM. The relative contrast is expected to be smaller for shorter capillary tubes.

A $g$-level variation of 10% is still acceptable at this stage of the experimental program. Nonetheless, these results show that, when selecting the length of a capillary tube or of any fracture system for a centrifuge test, there is a trade-off between inertia effects (see Section 3.5.2.3 and Section 7.3) and $g$-level changes across the system.

![Figure B.4](image)

**Figure B.4.** $g$-Level across a 130 mm long capillary tube as a function of the centrifuge rotational velocity.
APPENDIX C

DATA OF DNAPL INFILTRATION LABORATORY AND CENTRIFUGE EXPERIMENTS

C.1 Explanatory Note on Test Reference Number

1. Test series reference number
   L: laboratory spontaneous infiltration test
   (example: L8h = test h of 8th series of laboratory infiltration tests)
   Lch: laboratory controlled-head infiltration test
   (example: Lch167.2 = controlled-head infiltration test with \( h = 167.2 \) mm)
   C: centrifuge infiltration test (example: C5 = 5th centrifuge test)

2. Capillary tube actual diameter (example 1.33 = 1.33 mm diameter capillary tube)

3. Type of reservoir tube
   st: reservoir tube without graduations (diameter varies, generally 8 mm ID)
   ct: reservoir tube with graduations (diameter varies)
   f: reservoir funnel
   lst: long reservoir tube without graduations
   lct: long reservoir tube with graduations

4. Total length, \( l_t \), of capillary tube (in millimeters) (capillary tube flow length, \( l \), depends on pre-infiltration)
C.2 4-Chlorotoluene Spontaneous Infiltration Laboratory Experiments

C.2.1 2.70 mm Diameter Capillary Tubes
Reference Number of Test: L9-2.70st-1222mm

Date of Test: June 30, 1999

Date of Data Reduction: July 8, 1999

Capillary tube diameter [mm]: 2.70
Total length of tube, \( l_t \) [mm]: 1222
Type of reservoir tube: 16.5 mm ID non-graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of pool at onset of infiltration, \( h_p \) [mm]: 36.1
Depth of pre-infiltration, \( h_{pi} \) [mm]: 20

Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]: 56.1
Reduced length of tube, \( l = l_t - h_{pi} \) [mm]: 1202
\( \alpha_t \)-number, \( \alpha_t = \frac{\Delta \rho \rho g r_0^4}{16 \mu w^2 l_t} \): 0.120

Notes regarding the test: Four 305 mm long capillary tubes were connected together to form the tube. The tank tube was constituted of a 25 mm/32 mm (ID/OD) PVC hose. Some clamps holding the hose as well as the connection Teflon tape partially obstructed the view of the infiltration process.

**Infiltration data, \( z_c = z_t - h_{pi} \) [mm]**

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**Reference Number of Test**

L9-2.70st-915mm

**Date of Test**

July 7, 1999

**Date of Data Reduction**

July 8, 1999

**Capillary tube diameter [mm]**

2.70

**Total length of tube, \(l_t\) [mm]**

915

**Type of reservoir tube**

16.5 mm ID non-graduated tube

**Name of DNAPL used**

4-chlorotoluene

**Pool height measurement method**

optical caliper

**Height of pool at onset of infiltration, \(h_p\) [mm]**

38.0

**Depth of pre-infiltration, \(h_{pi}\) [mm]**

6

**Total pool height at infiltration, \(h = h_p + h_{pi}\) [mm]**

44.0

**Reduced length of tube, \(l = l_t - h_{pi}\) [mm]**

909

**\(\alpha_t\)-number, \(\alpha_t = \Delta \rho \rho w g r_0^{4/16} \mu w^{2} l_t\)**

0.160

**Notes regarding the test**

Three 305 mm long capillary tubes were connected together to form the tube. The tank tube was constituted of a 25 mm/32 mm (ID/OD) PVC hose. Some clamps holding the hose as well as the connection Teflon tape partially obstructed the view of the infiltration process.

\[Infiltration\ data, \ z_c = z_t - h_{pi} [mm]\]

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**Reference Number of Test**
L9-2.70st-610mm

**Date of Test**
July 8, 1999

**Date of Data Reduction**
July 8, 1999

- **Capillary tube diameter [mm]**
  2.70

- **Total length of tube, \( l_t \) [mm]**
  610

- **Type of reservoir tube**
  16.5 mm ID non-graduated tube

- **Name of DNAPL used**
  4-chlorotoluene

- **Pool height measurement method**
  optical caliper

- **Height of pool at onset of infiltration, \( h_p \) [mm]**
  35.3

- **Depth of pre-infiltration, \( h_{pi} \) [mm]**
  4

- **Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]**
  39.3

- **Reduced length of tube, \( l = l_t - h_{pi} \) [mm]**
  606

- **\( \alpha_t \)-number, \( \alpha_t = \Delta \rho \rho g r_0^4/(16 \mu w^2 l_t) \)**
  0.240

**Notes regarding the test**
Two 305 mm long capillary tubes were connected together to form the tube. The tank tube was constituted of a 25 mm/32 mm (ID/OD) PVC hose. Some clamps holding the hose as well as the connection Teflon tape partially obstructed the view of the infiltration process.

**Infiltration data, \( z_c = z_t - h_{pi} \) [mm]**

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<td>17.52</td>
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Reference Number of Test: L9-2.70st-305mm
Date of Test: July 9, 1999
Date of Data Reduction: July 9, 1999

Capillary tube diameter [mm]: 2.70
Total length of tube, l_t [mm]: 305
Type of reservoir tube: 16.5 mm ID non-graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of pool at onset of infiltration, h_p [mm]: 31.5
Depth of pre-infiltration, h_{pi} [mm]: 4

Total pool height at infiltration, h = h_p + h_{pi} [mm]: 35.5
Reduced length of tube, I = l_t - h_{pi} [mm]: 301
α_t-number, \( \alpha_t = \Delta \rho \rho g r_0^4 / (16 \mu w^2 l_t) \): 0.480

Notes regarding the test: The tank tube was constituted of a 25 mm/32 mm (ID/OD) PVC hose. Some clamps holding the hose as well as the connection Teflon tape partially obstructed the view of the infiltration process.

Infiltration data, z_c = z_t - h_{pi} [mm]

<table>
<thead>
<tr>
<th>Data Point Number</th>
<th>Clock Time [s]</th>
<th>Absolute Depth z_t [mm]</th>
<th>Corrected Depth z_c [mm]</th>
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**Reference Number of Test**  
L1a-2.70ct-260mm  
**Date of Test**  
March 18, 1997  
**Date of Data Reduction**  
March 18, 1997  

<table>
<thead>
<tr>
<th>Parameter</th>
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<tbody>
<tr>
<td>Capillary tube diameter [mm]</td>
<td>2.70</td>
</tr>
<tr>
<td>Total length of tube, ( l_t ) [mm]</td>
<td>260</td>
</tr>
<tr>
<td>Type of reservoir tube</td>
<td>6.4 mm ID graduated tube</td>
</tr>
<tr>
<td>Name of DNAPL used</td>
<td>4-chlorotoluene</td>
</tr>
<tr>
<td>Pool height measurement method</td>
<td>volume graduations on res. tube</td>
</tr>
<tr>
<td>Height of pool at onset of infiltration, ( h_p ) [mm]</td>
<td>54.3</td>
</tr>
<tr>
<td>Depth of pre-infiltration, ( h_{pi} ) [mm]</td>
<td>not measured but small</td>
</tr>
<tr>
<td>Total pool height at infiltration, ( h = h_p + h_{pi} ) [mm]</td>
<td>54.3</td>
</tr>
<tr>
<td>Reduced length of tube, ( l = l_t - h_{pi} ) [mm]</td>
<td>260</td>
</tr>
<tr>
<td>( \alpha )-number, ( \alpha_t = \frac{\Delta \rho \rho g r_0^4}{(16 \mu_w^2 l)} )</td>
<td>0.563</td>
</tr>
</tbody>
</table>

**Notes regarding the test**  
The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.

---

**Reference Number of Test**  
L1b-2.70ct-260mm  
**Date of Test**  
March 18, 1997  
**Date of Data Reduction**  
March 18, 1997  

<table>
<thead>
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<th>Parameter</th>
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<tr>
<td>Capillary tube diameter [mm]</td>
<td>2.70</td>
</tr>
<tr>
<td>Total length of tube, ( l_t ) [mm]</td>
<td>260</td>
</tr>
<tr>
<td>Type of reservoir tube</td>
<td>6.4 mm ID graduated tube</td>
</tr>
<tr>
<td>Name of DNAPL used</td>
<td>4-chlorotoluene</td>
</tr>
<tr>
<td>Pool height measurement method</td>
<td>volume graduations on res. tube</td>
</tr>
<tr>
<td>Height of pool at onset of infiltration, ( h_p ) [mm]</td>
<td>55.9</td>
</tr>
<tr>
<td>Depth of pre-infiltration, ( h_{pi} ) [mm]</td>
<td>not measured but small</td>
</tr>
<tr>
<td>Total pool height at infiltration, ( h = h_p + h_{pi} ) [mm]</td>
<td>55.9</td>
</tr>
<tr>
<td>Reduced length of tube, ( l = l_t - h_{pi} ) [mm]</td>
<td>260</td>
</tr>
<tr>
<td>( \alpha )-number, ( \alpha_t = \frac{\Delta \rho \rho g r_0^4}{(16 \mu_w^2 l)} )</td>
<td>0.563</td>
</tr>
</tbody>
</table>

**Notes regarding the test**  
The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.
Reference Number of Test L1c-2.70ct-260mm
Date of Test March 18, 1997
Date of Data Reduction March 18, 1997

Capillary tube diameter [mm] 2.70
Total length of tube, $l_t$ [mm] 260
Type of reservoir tube 6.4 mm ID graduated tube
Name of DNAPL used 4-chlorotoluene

Pool height measurement method volume graduations on res. tube
Height of pool at onset of infiltration, $h_p$ [mm] 54.3
Depth of pre-infiltration, $h_{pi}$ [mm] not measured but small

Total pool height at infiltration, $h = h_p + h_{pi}$ [mm] 54.3
Reduced length of tube, $l = l_t - h_{pi}$ [mm] 260
$\alpha_t$-number, $\alpha_t = \Delta \rho \rho_g r_0^4 / (16 \mu_w^2 l_t)$ 0.563

Notes regarding the test The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.

Reference Number of Test L1d-2.70ct-260mm
Date of Test March 18, 1997
Date of Data Reduction March 18, 1997

Capillary tube diameter [mm] 2.70
Total length of tube, $l_t$ [mm] 260
Type of reservoir tube 6.4 mm ID graduated tube
Name of DNAPL used 4-chlorotoluene

Pool height measurement method volume graduations on res. tube
Height of pool at onset of infiltration, $h_p$ [mm] 52.8
Depth of pre-infiltration, $h_{pi}$ [mm] not measured but small

Total pool height at infiltration, $h = h_p + h_{pi}$ [mm] 52.8
Reduced length of tube, $l = l_t - h_{pi}$ [mm] 260
$\alpha_t$-number, $\alpha_t = \Delta \rho \rho_g r_0^4 / (16 \mu_w^2 l_t)$ 0.563

Notes regarding the test The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.
**Reference Number of Test**  
**Date of Test**  
**Date of Data Reduction**

| Description                              | Value                  
|------------------------------------------|------------------------
| Capillary tube diameter [mm]             | 2.70                   
| Total length of tube, \( l_t \) [mm]     | 260                    
| Type of reservoir tube                   | 6.4 mm ID graduated tube  
| Name of DNAPL used                       | 4-chlorotoluene         
| Pool height measurement method           | volume graduations on res. tube  
| Height of pool at onset of infiltration, \( h_p \) [mm] | 52.8                  
| Depth of pre-infiltration, \( h_{pi} \) [mm] | not measured but small  
| Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm] | 52.8                  
| Reduced length of tube, \( l = l_t - h_{pi} \) [mm] | 260                    
| \( \alpha \)-number, \( \alpha_t = \Delta \rho g r_0^4 / (16 \mu_w^2 l_t) \) | 0.563                  

**Notes regarding the test**  
The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.

**Reference Number of Test**  
**Date of Test**  
**Date of Data Reduction**

| Description                              | Value                  
|------------------------------------------|------------------------
| Capillary tube diameter [mm]             | 2.70                   
| Total length of tube, \( l_t \) [mm]     | 136                    
| Type of reservoir tube                   | small funnel           
| Name of DNAPL used                       | 4-chlorotoluene         
| Pool height measurement method           | optical caliper        
| Height of pool at onset of infiltration, \( h_p \) [mm] | 67.3                  
| Depth of pre-infiltration, \( h_{pi} \) [mm] | not measured but small  
| Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm] | 67.3                  
| Reduced length of tube, \( l = l_t - h_{pi} \) [mm] | 136                    
| \( \alpha \)-number, \( \alpha_t = \Delta \rho g r_0^4 / (16 \mu_w^2 l_t) \) | 1.076                  

**Notes regarding the test**  
Tube set up in the centrifuge strong box. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection. Test was not filmed.
<table>
<thead>
<tr>
<th>Reference Number of Test</th>
<th>L4-2.70ct-136mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Date of Test</td>
<td>May 27, 1997</td>
</tr>
<tr>
<td>Date of Data Reduction</td>
<td>May 27, 1997</td>
</tr>
</tbody>
</table>

**Capillary tube diameter [mm]** 2.70

**Total length of tube, \( l_t \) [mm]** 136

**Type of reservoir tube** 7.8 mm ID graduated tube

**Name of DNAPL used** 4-chlorotoluene

**Pool height measurement method** optical caliper

**Height of pool at onset of infiltration, \( h_p \) [mm]** 63.5

**Depth of pre-infiltration, \( h_{pi} \) [mm]** not measured but small

**Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]** 63.5

**Reduced length of tube, \( l = l_t - h_{pi} \) [mm]** 136

\[
\alpha_t \text{-number, } \alpha_t = \frac{\Delta \rho g r_0^4}{(16 \mu_r^2 l_t)} \]

**Notes regarding the test** Tube set up in the centrifuge strong box. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection. Test was not filmed.

<table>
<thead>
<tr>
<th>Reference Number of Test</th>
<th>L5-2.70ct-136mm</th>
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</thead>
<tbody>
<tr>
<td>Date of Test</td>
<td>May 28, 1997</td>
</tr>
<tr>
<td>Date of Data Reduction</td>
<td>May 28, 1997</td>
</tr>
</tbody>
</table>

**Capillary tube diameter [mm]** 2.70

**Total length of tube, \( l_t \) [mm]** 136

**Type of reservoir tube** 7.8 mm ID graduated tube

**Name of DNAPL used** 4-chlorotoluene

**Pool height measurement method** optical caliper

**Height of pool at onset of infiltration, \( h_p \) [mm]** 66.5

**Depth of pre-infiltration, \( h_{pi} \) [mm]** not measured but small

**Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]** 66.5

**Reduced length of tube, \( l = l_t - h_{pi} \) [mm]** 136

\[
\alpha_t \text{-number, } \alpha_t = \frac{\Delta \rho g r_0^4}{(16 \mu_r^2 l_t)} \]

**Notes regarding the test** Tube set up in the centrifuge strong box. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection. Test was not filmed.
<table>
<thead>
<tr>
<th>Reference Number of Test</th>
<th>L4-2.70ct-13xmm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Date of Test</td>
<td>May 27, 1997</td>
</tr>
<tr>
<td>Date of Data Reduction</td>
<td>May 27, 1997</td>
</tr>
</tbody>
</table>

| Capillary tube diameter [mm] | 2.70            |
| Total length of tube, $l_t$ [mm] |                |
| Type of reservoir tube       | non-graduated tube |
| Name of DNAPL used           | 4-chlorotoluene   |

| Pool height measurement method | optical caliper |
| Height of pool at onset of infiltration, $h_p$ [mm] | 57.9            |
| Depth of pre-infiltration, $h_{pi}$ [mm]             | not measured but small |

| Total pool height at infiltration, $h = h_p + h_{pi}$ [mm] | 57.9            |
| Reduced length of tube, $l = l_t - h_{pi}$ [mm]        |                |
| $\alpha_t$-number, $\alpha_t = \Delta \rho g r_0^4/(16 \mu_w^2 l_t)$ |                |

Notes regarding the test
Tube set up in the centrifuge strong box. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection. Test was not filmed.

<table>
<thead>
<tr>
<th>Reference Number of Test</th>
<th>L5-2.70ct-13xmm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Date of Test</td>
<td>May 28, 1997</td>
</tr>
<tr>
<td>Date of Data Reduction</td>
<td>May 28, 1997</td>
</tr>
</tbody>
</table>

| Capillary tube diameter [mm] | 2.70            |
| Total length of tube, $l_t$ [mm] |                |
| Type of reservoir tube       | non-graduated tube |
| Name of DNAPL used           | 4-chlorotoluene   |

| Pool height measurement method | optical caliper |
| Height of pool at onset of infiltration, $h_p$ [mm] | 63.8            |
| Depth of pre-infiltration, $h_{pi}$ [mm]             | not measured but small |

| Total pool height at infiltration, $h = h_p + h_{pi}$ [mm] | 63.8            |
| Reduced length of tube, $l = l_t - h_{pi}$ [mm]        |                |
| $\alpha_t$-number, $\alpha_t = \Delta \rho g r_0^4/(16 \mu_w^2 l_t)$ |                |

Notes regarding the test
Tube set up in the centrifuge strong box. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection. Test was not filmed.
Reference Number of Test: L3-2.70f-130mm
Date of Test: May 8, 1997
Date of Data Reduction: May 8, 1997

Capillary tube diameter [mm]: 2.70
Total length of tube, \( t \) [mm]: 130
Type of reservoir tube: small funnel
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of pool at onset of infiltration, \( h_p \) [mm]: 66.0
Depth of pre-infiltration, \( h_{pi} \) [mm]: not measured but small

Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]: 66.0
Reduced length of tube, \( l = t - h_{pi} \) [mm]: 130
\( \alpha_t \)-number, \( \alpha_t = \Delta \rho \frac{g}{16 \mu_w^2} \frac{h_p}{l} \): 1.125

Notes regarding the test: Tube set up in the centrifuge strong box. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection. Test was not filmed.
**Reference Number of Test**  
L8a-2.70st-130mm

**Date of Test**  
June 23, 1999

**Date of Data Reduction**  
June 23, 1999

- **Capillary tube diameter [mm]**: 2.70
- **Total length of tube, \( l_t \) [mm]**: 130
- **Type of reservoir tube**: 8 mm ID non-graduated tube
- **Name of DNAPL used**: 4-chlorotoluene

**Pool height measurement method**: optical caliper

- **Height of pool at onset of infiltration, \( h_p \) [mm]**: 40.9
- **Depth of pre-infiltration, \( h_{pi} \) [mm]**: 27

- **Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]**: 67.9
- **Reduced length of tube, \( l = l_t - h_{pi} \) [mm]**: 103

- **\( \alpha_t \)-number, \( \alpha_t = \frac{\Delta \rho \rho g \rho_0}{4 (16 \mu_w^2 l_t)} \)**: 1.125

**Notes regarding the test**  
Setup similar to L8a-1.33st-122mm. The Teflon tape was located between graduations 5 and 15 mm. One mark was missing at the bottom of the tube.

**Infiltration data, \( z_c = z_t - h_{pi} [mm] \)**

<table>
<thead>
<tr>
<th>Data Point</th>
<th>Clock Time [s]</th>
<th>Absolute Depth [mm]</th>
<th>Corrected Depth [mm]</th>
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<td>9</td>
<td>11.38</td>
<td>112.5</td>
<td>85.5</td>
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<tr>
<td>10</td>
<td>11.55</td>
<td>130</td>
<td>103</td>
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</tbody>
</table>
Reference Number of Test
L8b-2.70st-130mm

Date of Test
June 23, 1999

Date of Data Reduction
June 23, 1999

Capillary tube diameter [mm]
2.70

Total length of tube, \( l_t \) [mm]
130

Type of reservoir tube
8 mm ID non-graduated tube

Name of DNAPL used
4-chlorotoluene

Pool height measurement method
optical caliper

Height of pool at onset of infiltration, \( h_p \) [mm]
39.1

Depth of pre-infiltration, \( h_{pi} \) [mm]
42

Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]
81.1

Reduced length of tube, \( l = l_t - h_{pi} \) [mm]
88

\( \alpha_t \)-number, \( \alpha_t = \frac{\Delta \rho g r_0^4}{(16 \mu w^2 l_t)} \)
1.125

Notes regarding the test
This test immediately followed L8a-2.70st-130mm after rinsing tube with acetone and water. Note the large pre-infiltration.

Infiltration data, \( z_c = z_t - h_{pi} \) [mm]

<table>
<thead>
<tr>
<th>Data Point Number</th>
<th>Clock Time [s]</th>
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<th>Corrected Depth ( z_c ) [mm]</th>
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</thead>
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</tr>
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<td>6.99</td>
<td>65</td>
<td>23</td>
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<td>4</td>
<td>7.95</td>
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<td>9.47</td>
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<tr>
<td>10</td>
<td>9.79</td>
<td>130</td>
<td>88</td>
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</tbody>
</table>
C.2.2 2.20 mm Diameter Capillary Tubes
**Reference Number of Test** | L1a-2.20ct-260mm
---|---
**Date of Test** | March 18, 1997
**Date of Data Reduction** | March 18, 1997

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
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<td>Capillary tube diameter [mm]</td>
<td>2.20</td>
</tr>
<tr>
<td>Total length of tube, $l_t$ [mm]</td>
<td>260</td>
</tr>
<tr>
<td>Type of reservoir tube</td>
<td>8 mm ID graduated tube</td>
</tr>
<tr>
<td>Name of DNAPL used</td>
<td>4-chlorotoluene</td>
</tr>
<tr>
<td>Pool height measurement method</td>
<td>volume graduations on res. tube</td>
</tr>
<tr>
<td>Height of pool at onset of infiltration, $h_p$ [mm]</td>
<td>66.8</td>
</tr>
<tr>
<td>Depth of pre-infiltration, $h_{pi}$ [mm]</td>
<td>not measured but small</td>
</tr>
<tr>
<td>Total pool height at infiltration, $h = h_p + h_{pi}$ [mm]</td>
<td>66.8</td>
</tr>
<tr>
<td>Reduced length of tube, $l = l_t - h_{pi}$ [mm]</td>
<td>260</td>
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<tr>
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**Notes regarding the test**
The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.

---

**Reference Number of Test** | L1b-2.20ct-260mm
---|---
**Date of Test** | March 18, 1997
**Date of Data Reduction** | March 18, 1997

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**Notes regarding the test**
The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.
### Reference Number of Test
L1c-2.20ct-260mm

### Date of Test
March 18, 1997

### Date of Data Reduction
March 18, 1997

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### Notes regarding the test
The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.

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### Reference Number of Test
L1d-2.20ct-260mm

### Date of Test
March 18, 1997

### Date of Data Reduction
March 18, 1997

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The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.
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C.2.3 1.33 mm Diameter Capillary Tubes
Reference Number of Test: L10-1.33lct-918mm
Date of Test: July 30, 1999
Date of Data Reduction: August 2, 1999

Capillary tube diameter [mm]: 1.33
Total length of tube, \( l_t \) [mm]: 918
Type of reservoir tube: 7.5 mm ID 232 mm l. grad. tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of pool at onset of infiltration, \( h_p \) [mm]: 131.6
Depth of pre-infiltration, \( h_{pi} \) [mm]: 6

Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]: 137.6
Reduced length of tube, \( l = l_t - h_{pi} \) [mm]: 912
\( \alpha_t \)-number, \( \alpha_t = \Delta \rho g r_0^4/(16 \mu w^2 l_t) \): 0.00938

Notes regarding the test: Three 305 mm long capillary tubes were connected together to form the tube. The tank tube constituted a long glass tube. The Teflon tape at the tube connections partially obstructed the view of the infiltration process. Photographs of the reservoir tube were taken during the infiltration process, and showed that the DNAPL volume change was equal to the volume of capillary tube infiltrated by DNAPL. This suggests that water was almost entirely drained by DNAPL.

Infiltration data, \( z_c = z_t - h_{pi} \) [mm]:

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L6-1.33st-610mm

**Date of Test**  
August 12, 1998

**Date of Data Reduction**  
August 12, 1998

**Capillary tube diameter [mm]**  
1.33

**Total length of tube, l_t [mm]**  
610

**Type of reservoir tube**  
8 mm ID non-graduated tube

**Name of DNAPL used**  
4-chlorotoluene

**Pool height measurement method**  
optical caliper

**Height of pool at onset of infiltration, h_p [mm]**  
130.0

**Depth of pre-infiltration, h_{pi} [mm]**  
not measured but small

**Total pool height at infiltration, h = h_p + h_{pi} [mm]**  
130.0

**Reduced length of tube, l = l_t - h_{pi} [mm]**  
610

**α_t-number, \( \alpha_t = \Delta \rho g r_0^2 / (16 \mu w^2 l_t) \)**  
0.0141

**Notes regarding the test**  
Two 305 mm long capillary tubes were connected together to form the tube. The tank tube was constituted of a long glass tube. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection. It was probably in the order of 10 mm.

**Infiltration data, z_{c} = z_t - h_{pi} [mm]**

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Reference Number of Test
L12-1.33st-305mm

Date of Test
December 21, 2001

Date of Data Reduction
December 21, 2001

Capillary tube diameter [mm]
1.33

Total length of tube, l_t [mm]
305

Type of reservoir tube
8 mm ID non-graduated tube

Name of DNAPL used
4-chlorotoluene

Pool height measurement method
optical caliper

Height of pool at onset of infiltration, h_p [mm]
80.0

Depth of pre-infiltration, h_{pi} [mm]
40

Total pool height at infiltration, h = h_p + h_{pi} [mm]
120.0

Reduced length of tube, l = l_t - h_{pi} [mm]
265

α_t-number, α_t = \Delta \rho \rho g r_0^4/(16 \mu w^2 l_t)
0.0282

Notes regarding the test
The tank was constituted of a long glass tube.

Infiltration data, z_c = z_t - h_{pi} [mm]

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**Reference Number of Test**  
L14-1.33slt-305mm

**Date of Test**  
May 15, 2002

**Date of Data Reduction**  
May 15, 2002

**Capillary tube diameter [mm]**  
1.33

**Total length of tube, \( l_t \) [mm]**  
305

**Type of reservoir tube**  
32.5 mm ID non-graduated tube

**Name of DNAPL used**  
4-chlorotoluene

**Pool height measurement method**  
optical caliper

**Height of pool at onset of infiltration, \( h_p \) [mm]**  
> 120.4

**Depth of pre-infiltration, \( h_{pi} \) [mm]**  
25

**Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]**

**Reduced length of tube, \( l = l_t - h_{pi} \) [mm]**

\[ \alpha_t = \frac{\Delta \rho g \rho_0}{4/(16 \mu_w^2 l)} \]

\[ \alpha_t = 0.0282 \]

**Notes regarding the test**
The tank was constituted of a long glass tube. The connection between the reservoir tube and the capillary tube was made using Teflon tape reinforced by a series of short concentric glass tubes sandwiched between Teflon layers. Despite the large reservoir diameter, pre-infiltration took place. Final infiltration height reading could not be made prior to the onset of infiltration.

**Reference Number of Test**  
L1a-1.33ct-230mm

**Date of Test**  
March 18, 1997

**Date of Data Reduction**  
March 18, 1997

**Capillary tube diameter [mm]**  
1.33

**Total length of tube, \( l_t \) [mm]**  
230

**Type of reservoir tube**  
7.8 mm ID non-graduated tube

**Name of DNAPL used**  
4-chlorotoluene

**Pool height measurement method**  
volume graduations on res. tube

**Height of pool at onset of infiltration, \( h_p \) [mm]**  
129.7

**Depth of pre-infiltration, \( h_{pi} \) [mm]**  
not measured but small

**Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]**  
129.7

**Reduced length of tube, \( l = l_t - h_{pi} \) [mm]**  
230

\[ \alpha_t = \frac{\Delta \rho g \rho_0}{4/(16 \mu_w^2 l)} \]

\[ \alpha_t = 0.0374 \]

**Notes regarding the test**
The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.
**Reference Number of Test**  
L1b-1.33ct-230mm

**Date of Test**  
March 18, 1997

**Date of Data Reduction**  
March 18, 1997

Capillary tube diameter [mm]  
1.33

Total length of tube, $l_t$ [mm]  
230

Type of reservoir tube  
7.8 mm ID non-graduated tube

Name of DNAPL used  
4-chlorotoluene

Pool height measurement method  
volume graduations on res. tube

Height of pool at onset of infiltration, $h_p$ [mm]  
131.8

Depth of pre-infiltration, $h_{pi}$ [mm]  
not measured but small

Total pool height at infiltration, $h = h_p + h_{pi}$ [mm]  
131.8

Reduced length of tube, $l = l_t - h_{pi}$ [mm]  
230

$\alpha_t$-number, $\alpha_t = \Delta \rho g r_0^{4/3} (16 \mu w^2 l)$  
0.0374

Notes regarding the test  
The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.

---

**Reference Number of Test**  
L1c-1.33ct-230mm

**Date of Test**  
March 18, 1997

**Date of Data Reduction**  
March 18, 1997

Capillary tube diameter [mm]  
1.33

Total length of tube, $l_t$ [mm]  
230

Type of reservoir tube  
7.8 mm ID non-graduated tube

Name of DNAPL used  
4-chlorotoluene

Pool height measurement method  
volume graduations on res. tube

Height of pool at onset of infiltration, $h_p$ [mm]  
127.6

Depth of pre-infiltration, $h_{pi}$ [mm]  
not measured but small

Total pool height at infiltration, $h = h_p + h_{pi}$ [mm]  
127.6

Reduced length of tube, $l = l_t - h_{pi}$ [mm]  
230

$\alpha_t$-number, $\alpha_t = \Delta \rho g r_0^{4/3} (16 \mu w^2 l)$  
0.0374

Notes regarding the test  
The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.
Reference Number of Test  L1d-1.33ct-230mm
Date of Test  March 18, 1997
Date of Data Reduction  March 18, 1997

Capillary tube diameter [mm]  1.33
Total length of tube, \( l_t \) [mm]  230
Type of reservoir tube  7.8 mm ID non-graduated tube
Name of DNAPL used  4-chlorotoluene

Pool height measurement method  volume graduations on res. tube
Height of pool at onset of infiltration, \( h_p \) [mm]  127.6
Depth of pre-infiltration, \( h_{pi} \) [mm]  not measured but small

Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]  127.6
Reduced length of tube, \( l = l_t - h_{pi} \) [mm]  230
\( \alpha_t \)-number, \( \alpha_t = \Delta \rho \rho g r_0^4 / (16 \mu_w^2 l_t) \)  0.0374

Notes regarding the test  The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.

Reference Number of Test  L1e-1.33ct-230mm
Date of Test  March 18, 1997
Date of Data Reduction  March 18, 1997

Capillary tube diameter [mm]  1.33
Total length of tube, \( l_t \) [mm]  230
Type of reservoir tube  7.8 mm ID non-graduated tube
Name of DNAPL used  4-chlorotoluene

Pool height measurement method  volume graduations on res. tube
Height of pool at onset of infiltration, \( h_p \) [mm]  129.7
Depth of pre-infiltration, \( h_{pi} \) [mm]  not measured but small

Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]  129.7
Reduced length of tube, \( l = l_t - h_{pi} \) [mm]  230
\( \alpha_t \)-number, \( \alpha_t = \Delta \rho \rho g r_0^4 / (16 \mu_w^2 l_t) \)  0.0374

Notes regarding the test  The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.
Reference Number of Test  L7a-1.33st-184mm
Date of Test  May 25, 1999
Date of Data Reduction  May 25, 1999

Capillary tube diameter [mm]  1.33
Total length of tube, \( l_t \) [mm]  184
Type of reservoir tube  8 mm ID non-graduated tube
Name of DNAPL used  4-chlorotoluene

Pool height measurement method  optical caliper
Height of pool at onset of infiltration, \( h_p \) [mm]  112.0
Depth of pre-infiltration, \( h_{pi} \) [mm]  37

Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]  149.0
Reduced length of tube, \( l = l_t - h_{pi} \) [mm]  147
\( \alpha_t \)-number, \( \alpha_t = \frac{\Delta \rho \kappa g r_0^{4/3}}{(16 \mu \omega^2 l_t)} \)  0.0468

Notes regarding the test  The tank tube was constituted of a glass cylinder. Note the extremely large pre-infiltration.

Infiltration data, \( z_c = z_t - h_{pi} \) [mm]

<table>
<thead>
<tr>
<th>Data Point Number</th>
<th>Clock Time [s]</th>
<th>Absolute Depth ( z_t ) [mm]</th>
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Ref: L7b-1.33st-184mm

Date of Test: May 25, 1999
Date of Data Reduction: May 25, 1999

Capillary tube diameter [mm]: 1.33
Total length of tube, \( l_t \) [mm]: 184
Type of reservoir tube: 8 mm ID non-graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of pool at onset of infiltration, \( h_p \) [mm]: 112.8
Depth of pre-infiltration, \( h_{pi} \) [mm]: 33

Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]: 145.8
Reduced length of tube, \( l = l_t - h_{pi} \) [mm]: 151

\( \alpha_t \)-number, \( \alpha_t = \Delta \rho \rho g \frac{r_0}{(16 \mu_w l_t)} \): 0.0468

Notes regarding the test: This test immediately followed L7a-1.33st-184mm after rinsing tube with acetone and water. The tank tube was constituted of a glass cylinder. Note again the extremely large pre-infiltration.

Infiltration data, \( z_c = z_t - h_{pi} \) [mm]

<table>
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<tr>
<th>Data Point Number</th>
<th>Clock Time [s]</th>
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</table>
**Reference Number of Test**  
L8a-1.33st-122mm

**Date of Test**  
June 8, 1999

**Date of Data Reduction**  
June 8, 1999

**Capillary tube diameter [mm]**  
1.33

**Total length of tube, \( l \) [mm]**  
122

**Type of reservoir tube**  
8 mm ID non-graduated tube

**Name of DNAPL used**  
4-chlorotoluene

**Pool height measurement method**  
oneoptical caliper

**Height of pool at onset of infiltration, \( h_p \) [mm]**  
103.9

**Depth of pre-infiltration, \( h_{pi} \) [mm]**  
52

**Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]**  
155.9

**Reduced length of tube, \( l = l - h_{pi} \) [mm]**  
70

\[ \alpha_t\text{-number, } \alpha_t = \Delta\rho \rho g r_0^4/(16 \mu r^2 l) \]  
0.0706

**Notes regarding the test**  
The tank tube was constituted of a glass cylinder. Note the extremely large pre-infiltration. Infiltration took place after slightly raising the DNAPL pool level with the needle.

**Infiltration data, \( z_c = z_t - h_{pi} \) [mm]**

<table>
<thead>
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<th>Data Point Number</th>
<th>Clock Time [s]</th>
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<th>Corrected Depth ( z_c ) [mm]</th>
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**Reference Number of Test**  
L8b-1.33st-122mm

**Date of Test**  
June 8, 1999

**Date of Data Reduction**  
June 8, 1999

**Capillary tube diameter [mm]**  
1.33

**Total length of tube, \( l_t \) [mm]**  
122

**Type of reservoir tube**  
8 mm ID non-graduated tube

**Name of DNAPL used**  
4-chlorotoluene

**Pool height measurement method**  
optical caliper

**Height of pool at onset of infiltration, \( h_p \) [mm]**  
105.2

**Depth of pre-infiltration, \( h_{pi} \) [mm]**  
50

**Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]**  
155.2

**Reduced length of tube, \( l = l_t - h_{pi} \) [mm]**  
72

\[ \alpha_t - \text{number}, \alpha_t = \frac{\Delta \rho_p \rho g \rho_0^4}{(16 \mu_w^2 l_t)} \]

0.0706

**Notes regarding the test**  
This test immediately followed L8a-1.33st-122mm after rinsing tube with acetone and water. Note again the extremely large pre-infiltration.

**Infiltration data, \( z_c = z_t - h_{pi} \) [mm]**

<table>
<thead>
<tr>
<th>Data Point Number</th>
<th>Clock Time [s]</th>
<th>Absolute Depth ( z_t ) [mm]</th>
<th>Corrected Depth ( z_c ) [mm]</th>
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<tr>
<td>6</td>
<td>30.33</td>
<td>115</td>
<td>65</td>
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</tbody>
</table>
### Reference Number of Test

| L8c-1.33st-122mm |

### Date of Test

| June 8, 1999 |

### Date of Data Reduction

| June 8, 1999 |

### Capillary tube diameter [mm]

| 1.33 |

### Total length of tube, \( l_t \) [mm]

| 122 |

### Type of reservoir tube

| 8 mm ID non-graduated tube |

### Name of DNAPL used

| 4-chlorotoluene |

### Pool height measurement method

| optical caliper |

### Height of pool at onset of infiltration, \( h_p \) [mm]

| 112.3 |

### Depth of pre-infiltration, \( h_{pi} \) [mm]

| 18 |

### Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]

| 130.3 |

### Reduced length of tube, \( l = l_t - h_{pi} \) [mm]

| 104 |

### \( \alpha_t \)-number,

\[
\alpha_t = \frac{\Delta \rho g \rho_0}{\left(16\mu w^2 l_t \right)^{1/4}}
\]

| 0.0706 |

### Notes regarding the test

This test immediately followed L8b-1.33st-122mm after rinsing tube with acetone and water. Pre-infiltration hidden by Teflon tape and estimated at 18 mm (lower limit of Teflon tape). Note pre-infiltration lower than that of L8b-1.33st-122mm. The film is of poor quality.

---

### Reference Number of Test

| L8d-1.33st-122mm |

### Date of Test

| June 10, 1999 |

### Date of Data Reduction

| June 10, 1999 |

### Capillary tube diameter [mm]

| 1.33 |

### Total length of tube, \( l_t \) [mm]

| 122 |

### Type of reservoir tube

| 8 mm ID non-graduated tube |

### Name of DNAPL used

| 4-chlorotoluene |

### Pool height measurement method

| optical caliper |

### Height of pool at onset of infiltration, \( h_p \) [mm]

| 118.9 |

### Depth of pre-infiltration, \( h_{pi} \) [mm]

| 10 |

### Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]

| 128.9 |

### Reduced length of tube, \( l = l_t - h_{pi} \) [mm]

| 112 |

### \( \alpha_t \)-number,

\[
\alpha_t = \frac{\Delta \rho g \rho_0}{\left(16\mu w^2 l_t \right)^{1/4}}
\]

| 0.0706 |

### Notes regarding the test

Setup similar to L8a-1.33st-122mm, but the Teflon tape was located between graduations 35 and 45 mm, such that pre-infiltration was perfectly visible. The infiltration was not filmed.
Reference Number of Test  L8e-1.33st-122mm
Date of Test  June 10, 1999
Date of Data Reduction  June 10, 1999

Capillary tube diameter [mm]  1.33
Total length of tube, l_t [mm]  122
Type of reservoir tube  8 mm ID non-graduated tube
Name of DNAPL used  4-chlorotoluene

Pool height measurement method  optical caliper
Height of pool at onset of infiltration, h_p [mm]  106.9
Depth of pre-infiltration, h_{pi} [mm]  48

Total pool height at infiltration, h = h_p + h_{pi} [mm]  154.9
Reduced length of tube, l = l_t - h_{pi} [mm]  74

\[ \alpha_t = \frac{\Delta \rho g r_0^4}{(16 \mu_w^2 l_t)} \]

Notes regarding the test  This test immediately followed L8d-1.33st-122mm after rinsing tube with acetone and water. Prior to complete infiltration, DNAPL crept over the course of two hours from a total pool height of 123.7 mm with pre-infiltration of 15 mm to a total pool height of 154.9 mm with pre-infiltration of 48 mm. The infiltration was not filmed.
**Reference Number of Test**

L8f-1.33st-122mm

**Date of Test**

June 15, 1999

**Date of Data Reduction**

June 15, 1999

**Capillary tube diameter [mm]**

1.33

**Total length of tube, \(l_t\) [mm]**

122

**Type of reservoir tube**

8 mm ID non-graduated tube

**Name of DNAPL used**

4-chlorotoluene

**Pool height measurement method**

optical caliper

**Height of pool at onset of infiltration, \(h_p\) [mm]**

131.6

**Depth of pre-infiltration, \(h_{pi}\) [mm]**

18

**Total pool height at infiltration, \(h = h_p + h_{pi}\) [mm]**

149.6

**Reduced length of tube, \(l = l_t - h_{pi}\) [mm]**

104

\(\alpha_t\)-number,

\[
\alpha_t = \frac{\Delta \rho \rho g r_0^4}{16 \mu \omega^2 l_t}
\]

0.0706

**Notes regarding the test**

Setup similar to L8a-1.33st-122mm but the Teflon tape was located between graduations 5 and 15 mm.

**Infiltration data, \(z_c = z_t - h_{pi}\) [mm]**

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<th>Data Point Number</th>
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</table>
**Reference Number of Test**  L8g-1.33st-122mm  
**Date of Test**  June 15, 1999  
**Date of Data Reduction**  June 15, 1999  

**Capillary tube diameter [mm]**  1.33  
**Total length of tube, \( l_t \) [mm]**  122  
**Type of reservoir tube**  8 mm ID non-graduated tube  
**Name of DNAPL used**  4-chlorotoluene  

**Pool height measurement method**  optical caliper  
**Height of pool at onset of infiltration, \( h_p \) [mm]**  105.4  
**Depth of pre-infiltration, \( h_{pi} \) [mm]**  30  

**Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]**  135.4  
**Reduced length of tube, \( l = l_t - h_{pi} \) [mm]**  92  
**\( \alpha_t \)-number, \( \alpha_t = \frac{\Delta \rho \rho g \rho_0^2}{16 \nu w^2 l_t} \)**  0.0706  

**Notes regarding the test**  This test immediately followed L8f-1.33st-122mm after rinsing tube with acetone and water. The Teflon tape was located between graduations 35 and 45 mm. The infiltration was not filmed.

---

**Reference Number of Test**  L8h-1.33st-122mm  
**Date of Test**  June 15, 1999  
**Date of Data Reduction**  June 15, 1999  

**Capillary tube diameter [mm]**  1.33  
**Total length of tube, \( l_t \) [mm]**  122  
**Type of reservoir tube**  8 mm ID non-graduated tube  
**Name of DNAPL used**  4-chlorotoluene  

**Pool height measurement method**  optical caliper  
**Height of pool at onset of infiltration, \( h_p \) [mm]**  128.0  
**Depth of pre-infiltration, \( h_{pi} \) [mm]**  9  

**Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm]**  137.0  
**Reduced length of tube, \( l = l_t - h_{pi} \) [mm]**  113  
**\( \alpha_t \)-number, \( \alpha_t = \frac{\Delta \rho \rho g \rho_0^2}{16 \nu w^2 l_t} \)**  0.0706  

**Notes regarding the test**  This test immediately followed L8g-1.33st-122mm after rinsing tube with acetone, Alconox and water. The Teflon tape was located between graduations 35 and 45 mm. The infiltration was not filmed.
**Reference Number of Test** | L8i-1.33st-122mm
---|---
**Date of Test** | June 15, 1999
**Date of Data Reduction** | June 15, 1999

| Capillary tube diameter [mm] | 1.33 |
| Total length of tube, $l_t$ [mm] | 122 |
| Type of reservoir tube | 8 mm ID non-graduated tube |
| Name of DNAPL used | 4-chlorotoluene |

**Pool height measurement method** | optical caliper
**Height of pool at onset of infiltration, $h_p$ [mm]** | 127.8 |
**Depth of pre-infiltration, $h_{pi}$ [mm]** | 9 |

| Total pool height at infiltration, $h = h_p + h_{pi}$ [mm] | 136.8 |
| Reduced length of tube, $l = l_t - h_{pi}$ [mm] | 113 |
| $\alpha_t$-number, $\alpha_t = \Delta \rho \rho g r_0^4 / (16 \mu_w^2 l_t)$ | 0.0706 |

**Notes regarding the test** | This test immediately followed L8h-1.33st-122mm after rinsing tube with acetone and water. The Teflon tape was located between graduations 35 and 45 mm. The infiltration was not filmed.

---

**Reference Number of Test** | L8j-1.33st-122mm
---|---
**Date of Test** | June 15, 1999
**Date of Data Reduction** | June 15, 1999

| Capillary tube diameter [mm] | 1.33 |
| Total length of tube, $l_t$ [mm] | 122 |
| Type of reservoir tube | 8 mm ID non-graduated tube |
| Name of DNAPL used | 4-chlorotoluene |

**Pool height measurement method** | optical caliper
**Height of pool at onset of infiltration, $h_p$ [mm]** | 121.9 |
**Depth of pre-infiltration, $h_{pi}$ [mm]** | 11 |

| Total pool height at infiltration, $h = h_p + h_{pi}$ [mm] | 132.9 |
| Reduced length of tube, $l = l_t - h_{pi}$ [mm] | 111 |
| $\alpha_t$-number, $\alpha_t = \Delta \rho \rho g r_0^4 / (16 \mu_w^2 l_t)$ | 0.0706 |

**Notes regarding the test** | This test immediately followed L8i-1.33st-122mm after rinsing tube with acetone and water. The Teflon tape was located between graduations 35 and 45 mm. The infiltration was not filmed.
**Reference Number of Test**  
L8k-1.33st-122mm

**Date of Test**  
June 15, 1999

**Date of Data Reduction**  
June 15, 1999

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**Notes regarding the test**  
This test immediately followed L8j-1.33st-122mm after rinsing tube with acetone and water. The Teflon tape was located between graduations 35 and 45 mm. After 1 hour, the pre-infiltration was down to 12 mm with a slight lowering of the pool top. After 12 hours, the pre-infiltration was down to 15 mm with a lowering of the pool top of less than 1 mm. No change observed after 24 hours.
**Reference Number of Test**  
L8a-1.33st-60mm  
**Date of Test**  
June 9, 1999  
**Date of Data Reduction**  
June 9, 1999

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</table>

**Notes regarding the test**  
The tank tube was constituted of a glass cylinder. Pre-infiltration hidden by Teflon tape and estimated at 18 mm (lower limit of Teflon tape). The infiltration was not filmed but was observed to be extremely slow.

---

**Reference Number of Test**  
L8b-1.33st-60mm  
**Date of Test**  
June 9, 1999  
**Date of Data Reduction**  
June 9, 1999

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**Notes regarding the test**  
This test immediately followed L8a-1.33st-60mm after rinsing tube with acetone and water. Pre-infiltration right at bottom of Teflon tape. Infiltration observed to move very abruptly.
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</table>

| **Capillary tube diameter [mm]** | 1.33 |
| **Total length of tube, \(l_t\) [mm]** | 60 |
| **Type of reservoir tube** | 8 mm ID non-graduated tube |
| **Name of DNAPL used** | 4-chlorotoluene |

**Pool height measurement method**  optical caliper

| **Height of pool at onset of infiltration, \(h_p\) [mm]** | 126.5* |
| **Depth of pre-infiltration, \(h_{pi}\) [mm]** | 30 |

| **Total pool height at infiltration, \(h = h_p + h_{pi}\) [mm]** | 156.5* |
| **Reduced length of tube, \(l = l_t - h_{pi}\) [mm]** | |
| **\(\alpha_t\)-number, \(\alpha_t = \Delta \rho g r_0^4 / (16 \mu w^2 l)\)** | 0.144 |

**Notes regarding the test**

This test immediately followed L8b-1.33st-60mm after rinsing tube with acetone and water. Although the total pool height was 156.5 mm, no infiltration was observed. After waiting 6 hours, the pre-infiltration progressed down to 39 mm with a slight lowering of the pool top. After 20 hours, the pre-infiltration was down to 38 mm with a 4 mm lowering of the pool top. No infiltration took place.
C.2.4 0.66 mm Diameter Capillary Tubes
Reference Number of Test: L11-0.66lct-1221mm
Date of Test: September 7, 1999
Date of Data Reduction: September 8, 1999

Capillary tube diameter [mm]: 0.66
Total length of tube, \( l_t [\text{mm}] \): 1221
Type of reservoir tube: 5.4 mm ID graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: reservoir pool graduations
Height of pool at onset of infiltration, \( h_p [\text{mm}] \): 288
Depth of pre-infiltration, \( h_{pi} [\text{mm}] \): 20

Total pool height at infiltration, \( h = h_p + h_{pi} [\text{mm}] \): 308
Reduced length of tube, \( l = l_t - h_{pi} [\text{mm}] \): 1201
\( \alpha_t \)-number, \( \alpha_t = \Delta \rho \rho g r_0^{4/16 \mu^2 l} \): 0.000428

Notes regarding the test: Four 305 mm long capillary tubes were connected together to form the tube. The tank tube was constituted a long glass tube. The optical caliper was not used because the pool height was larger than the range of the caliper. The reservoir had volume graduations from which the corresponding height could be obtained. The Teflon tape at the tube connections partially obstructed the view of the infiltration process.

Infiltration data, \( z_c = z_t - h_{pi} [\text{mm}] \)

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Reference Number of Test: L1a-0.66ct-xmm
Date of Test: March 18, 1997
Date of Data Reduction: March 18, 1997

Capillary tube diameter [mm]: 0.66
Total length of tube, \(l_t\) [mm]: 6.4 mm ID non-graduated tube
Type of reservoir tube: 6.4 mm ID non-graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: volume graduations on res. tube
Height of pool at onset of infiltration, \(h_p\) [mm]: 248.8
Depth of pre-infiltration, \(h_{pi}\) [mm]: not measured but small

Total pool height at infiltration, \(h = h_p + h_{pi}\) [mm]: 248.8
Reduced length of tube, \(l = l_t - h_{pi}\) [mm]:
\[\alpha_t - \text{number}, \quad \alpha_t = \frac{\Delta \rho \rho g r_0^4}{16 \mu_w^2 l_t}\]

Notes regarding the test: The tank tube was constituted of a glass cylinder. The capillary tube were extremely short (around 30 to 40 mm). Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.

Reference Number of Test: L1b-0.66ct-xmm
Date of Test: March 18, 1997
Date of Data Reduction: March 18, 1997

Capillary tube diameter [mm]: 0.66
Total length of tube, \(l_t\) [mm]: 6.4 mm ID non-graduated tube
Type of reservoir tube: 6.4 mm ID non-graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: volume graduations on res. tube
Height of pool at onset of infiltration, \(h_p\) [mm]: 234.8
Depth of pre-infiltration, \(h_{pi}\) [mm]: not measured but small

Total pool height at infiltration, \(h = h_p + h_{pi}\) [mm]: 234.8
Reduced length of tube, \(l = l_t - h_{pi}\) [mm]:
\[\alpha_t - \text{number}, \quad \alpha_t = \frac{\Delta \rho \rho g r_0^4}{16 \mu_w^2 l_t}\]

Notes regarding the test: The tank tube was constituted of a glass cylinder. The capillary tube were extremely short (around 30 to 40 mm). Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.
C.3 1,1,1-Trichloroethane Spontaneous Infiltration Laboratory Experiments

C.3.1 2.20 mm Diameter Capillary Tubes
Reference Number of Test | L2a-2.20ct
---|---
Date of Test | March 25, 1997
Date of Data Reduction | March 25, 1997

Capillary tube diameter [mm] | 2.20
Total length of tube, $l_t$ [mm] | 8 mm ID graduated tube
Type of reservoir tube | 1,1,1-trichloroethane
Name of DNAPL used | volume graduations on res. tube

Pool height measurement method | 10.1
Height of pool at onset of infiltration, $h_p$ [mm] | not measured but small
Depth of pre-infiltration, $h_{pi}$ [mm] | 10.1

Total pool height at infiltration, $h = h_p + h_{pi}$ [mm] | 10.1
Reduced length of tube, $l = l_t - h_{pi}$ [mm] | $\alpha_t = \Delta \rho \rho g \rho_0^4/(16\mu_w^2 l)$

Notes regarding the test | The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.

Reference Number of Test | L2b-2.20ct
---|---
Date of Test | March 25, 1997
Date of Data Reduction | March 25, 1997

Capillary tube diameter [mm] | 2.20
Total length of tube, $l_t$ [mm] | 8 mm ID graduated tube
Type of reservoir tube | 1,1,1-trichloroethane
Name of DNAPL used | volume graduations on res. tube

Pool height measurement method | 10.1
Height of pool at onset of infiltration, $h_p$ [mm] | not measured but small
Depth of pre-infiltration, $h_{pi}$ [mm] | 10.1

Total pool height at infiltration, $h = h_p + h_{pi}$ [mm] | 10.1
Reduced length of tube, $l = l_t - h_{pi}$ [mm] | $\alpha_t = \Delta \rho \rho g \rho_0^4/(16\mu_w^2 l)$

Notes regarding the test | The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.
C.3.2 1.33 mm Diameter Capillary Tubes
Reference Number of Test | L2a-1.33ct  
--- | ---  
Date of Test | March 25, 1997  
Date of Data Reduction | March 25, 1997  

Capillary tube diameter [mm] | 1.33  
Total length of tube, \( l_t \) [mm] |  \( 6.4 \text{ mm ID graduated tube} \)  
Type of reservoir tube |  \( 6.4 \text{ mm ID graduated tube} \)  
Name of DNAPL used |  \( 1,1,1\text{-trichloroethane} \)  

Pool height measurement method | volume graduations on res. tube  
Height of pool at onset of infiltration, \( h_p \) [mm] | 23.3  
Depth of pre-infiltration, \( h_{pi} \) [mm] |  \( \text{not measured but small} \)  

Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm] | 23.3  
Reduced length of tube, \( l = l_t - h_{pi} \) [mm] |  
\( \alpha_t \)-number, \( \alpha_t = \Delta \rho_w g r_0^4/(16 \mu w^2 l_t) \) |  

Notes regarding the test | The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.

Reference Number of Test | L2b-1.33ct  
--- | ---  
Date of Test | March 25, 1997  
Date of Data Reduction | March 25, 1997  

Capillary tube diameter [mm] | 1.33  
Total length of tube, \( l_t \) [mm] |  \( 6.4 \text{ mm ID graduated tube} \)  
Type of reservoir tube |  \( 6.4 \text{ mm ID graduated tube} \)  
Name of DNAPL used |  \( 1,1,1\text{-trichloroethane} \)  

Pool height measurement method | volume graduations on res. tube  
Height of pool at onset of infiltration, \( h_p \) [mm] | 24.8  
Depth of pre-infiltration, \( h_{pi} \) [mm] |  \( \text{not measured but small} \)  

Total pool height at infiltration, \( h = h_p + h_{pi} \) [mm] | 24.8  
Reduced length of tube, \( l = l_t - h_{pi} \) [mm] |  
\( \alpha_t \)-number, \( \alpha_t = \Delta \rho_w g r_0^4/(16 \mu w^2 l_t) \) |  

Notes regarding the test | The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.
C.3.3 0.66 mm Diameter Capillary Tubes
Reference Number of Test: L2a-0.66ct
Date of Test: March 25, 1997
Date of Data Reduction: March 25, 1997

Capillary tube diameter [mm]: 0.66
Total length of tube, $l_t$ [mm]:
Type of reservoir tube:
Name of DNAPL used: 1,1,1-trichloroethane

Pool height measurement method: volume graduations on res. tube
Height of pool at onset of infiltration, $h_p$ [mm]: 53.0
Depth of pre-infiltration, $h_{pi}$ [mm]: not measured but small

Total pool height at infiltration, $h = h_p + h_{pi}$ [mm]: 53.0
Reduced length of tube, $l = l_t - h_{pi}$ [mm]:

Notes regarding the test: The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.

Reference Number of Test: L2b-0.66ct
Date of Test: March 25, 1997
Date of Data Reduction: March 25, 1997

Capillary tube diameter [mm]: 0.66
Total length of tube, $l_t$ [mm]:
Type of reservoir tube:
Name of DNAPL used: 1,1,1-trichloroethane

Pool height measurement method: volume graduations on res. tube
Height of pool at onset of infiltration, $h_p$ [mm]: 53.0
Depth of pre-infiltration, $h_{pi}$ [mm]: not measured but small

Total pool height at infiltration, $h = h_p + h_{pi}$ [mm]: 53.0
Reduced length of tube, $l = l_t - h_{pi}$ [mm]:

Notes regarding the test: The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.
### Reference Number of Test

**L2c-0.66ct**

### Date of Test

March 25, 1997

### Date of Data Reduction

March 25, 1997

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<td>volume graduations on res. tube</td>
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<tr>
<td>Depth of pre-infiltration, ( h_{pi} ) [mm]</td>
<td>not measured but small</td>
</tr>
<tr>
<td>Total pool height at infiltration, ( h = h_p + h_{pi} ) [mm]</td>
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<td>Reduced length of tube, ( l = l_t - h_{pi} ) [mm]</td>
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**Notes regarding the test**
The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.

---

### Reference Number of Test

**L2d-0.66ct**

### Date of Test

March 25, 1997

### Date of Data Reduction

March 25, 1997

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<td>Depth of pre-infiltration, ( h_{pi} ) [mm]</td>
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**Notes regarding the test**
The tank tube was constituted of a glass cylinder. Pre-infiltration took place but could not be seen because of the upper Teflon tape connection.

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533
C.4 4-Chlorotoluene Controlled-Head Infiltration Laboratory Experiments (1.33 mm Diameter Capillary Tubes)
Reference Number of Test: Lch178.1-1.33st-305mm
Date of Test: April 28, 1998
Date of Data Reduction: April 30, 1998

Capillary tube diameter [mm]: 1.33
Total length of tube, \( l_t \) [mm]: 305
Type of reservoir tube: 8 mm ID non-graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Controlled Height of pool, \( h \) [mm]: 178.1

\( \alpha \)-number, \( \alpha = \frac{\Delta \rho \omega g r_0^2}{(16 \mu_w^2 l)} \): 0.0282

Notes regarding the test: View of early stage and end stage of infiltration obstructed by Teflon tape collar. Some pre-infiltration observed despite shut valve.

Infiltration data

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**Reference Number of Test**
Lch174.1-1.33st-305mm

**Date of Test**
April 28, 1998

**Date of Data Reduction**
April 30, 1998

**Capillary tube diameter [mm]**
1.33

**Total length of tube, l [mm]**
305

**Type of reservoir tube**
8 mm ID non-graduated tube

**Name of DNAPL used**
4-chlorotoluene

**Pool height measurement method**
optical caliper

**Controlled Height of pool, h [mm]**
174.1

\[ \alpha = \Delta \rho \frac{g r_c^4}{(16 \mu_w l)} \]

**α-number**, \( \alpha = 0.0282 \)

**Notes regarding the test**
View of early stage and end stage of infiltration obstructed by Teflon tape collar.

**Infiltration data**

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Reference Number of Test: Lch171.8-1.33st-305mm
Date of Test: March 10, 1998
Date of Data Reduction: March 10, 1998

Capillary tube diameter [mm]: 1.33
Total length of tube, l, [mm]: 305
Type of reservoir tube: 8 mm ID non-graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Controlled Height of pool, h [mm]: 171.8

\[ \alpha \text{-number, } \alpha = \frac{\Delta \rho \rho g \rho_0^2}{16 \mu_w^2 l} \]

\[ 0.0282 \]

Notes regarding the test: View of early stage and end stage of infiltration obstructed by Teflon tape collar.

Infiltration data:

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Reference Number of Test: Lch167.2-1.33st-305mm
Date of Test: March 17, 1998
Date of Data Reduction: March 17, 1998

Capillary tube diameter [mm]: 1.33
Total length of tube, l, [mm]: 305
Type of reservoir tube: 8 mm ID non-graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Controlled Height of pool, h [mm]: 167.2

$\alpha$-number, $\alpha = \frac{\Delta \rho g r_0^4}{(16 \mu_r^2 l)}$: 0.0282

Notes regarding the test: View of early stage and end stage of infiltration obstructed by Teflon tape collar.

Infiltration data

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Date of Test: March 17, 1998
Date of Data Reduction: March 17, 1998

Capillary tube diameter [mm]: 1.33
Total length of tube, l [mm]: 305
Type of reservoir tube: 8 mm ID non-graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Controlled Height of pool, h [mm]: 164.2

$\alpha$-number, $\alpha = \frac{\Delta \rho_{\text{w}} g r_0^2}{16 \mu_{\text{w}}^2 l}$: 0.0282

Notes regarding the test: View of early stage and end stage of infiltration obstructed by Teflon tape collar.

Infiltration data

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Reference Number of Test: Lch150.9-1.33st-305mm
Date of Test: March 10, 1998
Date of Data Reduction: March 10, 1998

Capillary tube diameter [mm]: 1.33
Total length of tube, l, [mm]: 305
Type of reservoir tube: 8 mm ID non-graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Controlled Height of pool, h [mm]: 150.9

α-number, \( \alpha = \frac{\Delta \rho g r_c^4}{16 \mu_c^2 l} \): 0.0282

Notes regarding the test: View of early stage and end stage of infiltration obstructed by Teflon tape collar.

Infiltration data

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C.5 4-Chlorotoluene Infiltration Centrifuge Experiments

C.5.1 2.70 mm Diameter Capillary Tubes
Reference Number of Test c2-2.70ct-136mm
Date of Test April 18, 1997
Date of Data Reduction April 18, 1997

Capillary tube diameter [mm] 2.70
Model length of tube, l [mm] 136
Type of reservoir tube 6.4 mm ID graduated tube
Name of DNAPL used 4-chlorotoluene

Pool height measurement method volume graduations on res. tube
Height of model pool, h [mm] 27.9

Centrifuge rotational velocity at infiltration [RPM] 41.8
Angle of inclination of centr. platform, ψ [degrees] 22.08
Computed g-level at infiltration, n [gravities] 2.45
Height of prototype pool, H = nh [mm] 68.3
Prototype length of tube, L [mm] 332.8
αₙ-number, αₙ = Δρρₑ₀(ng)r₀⁴/(16µₑ₀²l) 2.632

Notes regarding the test Interface displacement not recorded.

Reference Number of Test c3-2.70ct-136mm
Date of Test April 25, 1997
Date of Data Reduction April 25, 1997

Capillary tube diameter [mm] 2.70
Model length of tube, l [mm] 136
Type of reservoir tube 6.4 mm ID graduated tube
Name of DNAPL used 4-chlorotoluene

Pool height measurement method optical caliper
Height of model pool, h [mm] 18.7

Centrifuge rotational velocity at infiltration [RPM] 53.2
Angle of inclination of centr. platform, ψ [degrees] 13.96
Computed g-level at infiltration, n [gravities] 3.76
Height of prototype pool, H = nh [mm] 70.4
Prototype length of tube, L [mm] 511.9
αₙ-number, αₙ = Δρρₑ₀(ng)r₀⁴/(16µₑ₀²l) 4.049

Notes regarding the test Interface displacement not recorded.
Reference Number of Test: c4-2.70ct-136mm
Date of Test: May 14, 1997
Date of Data Reduction: May 14, 1997

Capillary tube diameter [mm]: 2.70
Model length of tube, l [mm]: 136
Type of reservoir tube: 7.8 mm ID graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 15.0

Centrifuge rotational velocity at infiltration [RPM]: 50.7
Angle of inclination of centr. platform, \( \psi \) [degrees]: 15.33
Computed g-level at infiltration, n [gravities]: 3.44
Height of prototype pool, \( H = nh \) [mm]: 51.6
Prototype length of tube, L [mm]: 468.2
\( \alpha_n \)-number, \( \alpha_n = \frac{\Delta \rho p w (n g r_0^4)}{16 \mu_w^2 l} \): 3.703

Notes regarding the test: Interface displacement not recorded. Very small pool height at infiltration possibly due to pre-infiltration.
Reference Number of Test: c7-2.70ct-136mm
Date of Test: October 15, 1997
Date of Data Reduction: November 13, 1997

Capillary tube diameter [mm]: 2.70
Model length of tube, l [mm]: 136
Type of reservoir tube: 7.8 mm ID graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 10.9

Centrifuge rotational velocity at infiltration [RPM]: 62.9
Angle of inclination of centr. platform, $\psi$ [degrees]: 10.06
Computed g-level at infiltration, n [gravities]: 5.18
Height of prototype pool, $H = nh$ [mm]: 56.4
Prototype length of tube, $L$ [mm]: 703.8
$\alpha_n$-number, $\alpha_n = \Delta \rho \rho_r (n g) r_0^3/(16 \mu_w^2 l)$: 5.567

Notes regarding the test

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**Reference Number of Test**
c8-2.70ct-136mm

**Date of Test**
November 25, 1997

**Date of Data Reduction**
November 25, 1997

- **Capillary tube diameter [mm]**: 2.70
- **Model length of tube, l [mm]**: 136
- **Type of reservoir tube**: 7.8 mm ID graduated tube
- **Name of DNAPL used**: 4-chlorotoluene

**Pool height measurement method**: optical caliper

**Height of model pool, h [mm]**: 15.5

- **Centrifuge rotational velocity at infiltration [RPM]**: 53.1
- **Angle of inclination of centr. platform, ψ [degrees]**: 14.01
- **Computed g-level at infiltration, n [gravities]**: 3.75
- **Height of prototype pool, H = nh [mm]**: 58.1
- **Prototype length of tube, L [mm]**: 510.1

\[ \alpha_n = \frac{\Delta \rho \rho_w (n g/r_0^4)}{16 \mu_w l} \]

4.035

**Notes regarding the test**

**Infiltration data**

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Reference Number of Test  c9-2.70ct-136mm
Date of Test  February 12, 1998
Date of Data Reduction  February 22, 1998

Capillary tube diameter [mm]  2.70
Model length of tube, l [mm]  136
Type of reservoir tube  7.8 mm ID graduated tube
Name of DNAPL used  4-chlorotoluene

Pool height measurement method  optical caliper
Height of model pool, h [mm]  21.3

Centrifuge rotational velocity at infiltration [RPM]  36.5
Angle of inclination of centr. platform, ψ [degrees]  28.20
Computed g-level at infiltration, n [gravities]  1.97
Height of prototype pool, H = nh [mm]  42.0
Prototype length of tube, L [mm]  267.9
αn-number, \( \alpha_n = \frac{\Delta \rho \rho (ng) r_0^2}{16 \mu_a^2 l} \)  2.119

Notes regarding the test  Very small pool height at infiltration possibly due to pre-infiltration.

Infiltration data

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**Reference Number of Test**  c10-2.70ct-136mm  
**Date of Test**  March 25, 1998  
**Date of Data Reduction**  June 2, 1998  

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**Pool height measurement method**  optical caliper  
**Height of model pool, h [mm]**  9.9

- Centrifuge rotational velocity at infiltration [RPM]  64.5  
- Angle of inclination of centr. platform, \( \psi \) [degrees]  9.58  
- Computed g-level at infiltration, \( n \) [gravities]  5.43  
- Height of prototype pool, \( H = nh \) [mm]  53.8  
- Prototype length of tube, \( L \) [mm]  738.8  
- \( \alpha_n \)-number, \( \alpha_n = \frac{\Delta \rho \rho_w (n g f_0^4)}{16 \mu_w l} \)  5.844

**Notes regarding the test**

**Infiltration data**

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Reference Number of Test: c11-2.70ct-136mm
Date of Test: May 5, 1998
Date of Data Reduction: June 2, 1998

Capillary tube diameter [mm]: 2.70
Model length of tube, l [mm]: 136
Type of reservoir tube: 7.8 mm ID graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 19.8

Centrifuge rotational velocity at infiltration [RPM]: 45.0
Angle of inclination of centr. platform, ψ [degrees]: 19.24
Computed g-level at infiltration, n [gravities]: 2.78
Height of prototype pool, H = nh [mm]: 55.0
Prototype length of tube, L [mm]: 377.8

αₙ-number, αₙ = Δρρₜₚₚ(wg)r₀^3/(16μₜₚₚ²l)

2.988

Notes regarding the test

Infiltration data

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Date of Test May 12, 1998
Date of Data Reduction May 12, 1998

Capillary tube diameter [mm] 2.70
Model length of tube, l [mm] 136
Type of reservoir tube 7.8 mm ID graduated tube
Name of DNAPL used 4-chlorotoluene

Pool height measurement method optical caliper
Height of model pool, h [mm] 21.0

Centrifuge rotational velocity at infiltration [RPM] 46.7
Angle of inclination of centr. platform, ψ [degrees] 17.94
Computed g-level at infiltration, n [gravities] 2.97
Height of prototype pool, H = nh [mm] 62.3
Prototype length of tube, L [mm] 403.4
αn-number, αn = Δρρw(ng)r0^4/(16µw^2l) 3.191

Notes regarding the test

Infiltration data

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**Reference Number of Test**
c4-2.70ct-13xmm

**Date of Test**
May 14, 1997

**Date of Data Reduction**
May 14, 1997

Capillary tube diameter [mm] 2.70
Model length of tube, l [mm] 130
Type of reservoir tube non-graduated tube
Name of DNAPL used 4-chlorotoluene

Pool height measurement method optical caliper
Height of model pool, h [mm] 15.0

Centrifuge rotational velocity at infiltration [RPM] 53.1
Angle of inclination of centr. platform, ψ [degrees] 14.01
Computed g-level at infiltration, n [gravities] 3.75
Height of prototype pool, H = nh [mm] 56.3

Prototype length of tube, L [mm]
αₙ-number, $\alpha_n = \frac{\Delta \rho \rho_w (ng) r_0^3}{16 \mu_w^2 l}$

Notes regarding the test Interface displacement not recorded. Very small pool height at infiltration possibly due to pre-infiltration.

**Reference Number of Test**
c4-2.70f-130mm

**Date of Test**
May 14, 1997

**Date of Data Reduction**
May 14, 1997

Capillary tube diameter [mm] 2.70
Model length of tube, l [mm] 130
Type of reservoir tube small funnel
Name of DNAPL used 4-chlorotoluene

Pool height measurement method optical caliper
Height of model pool, h [mm] 15.0

Centrifuge rotational velocity at infiltration [RPM] 63.4
Angle of inclination of centr. platform, ψ [degrees] 9.91
Computed g-level at infiltration, n [gravities] 5.25
Height of prototype pool, H = nh [mm] 78.8

Prototype length of tube, L [mm]
αₙ-number, $\alpha_n = \frac{\Delta \rho \rho_w (ng) r_0^3}{16 \mu_w^2 l}$

Notes regarding the test Interface displacement not recorded.
**Reference Number of Test**
c7-2.70f-130mm

**Date of Test**
October 15, 1997

**Date of Data Reduction**
November 13, 1997

- **Capillary tube diameter [mm]**
  2.70

- **Model length of tube, l [mm]**
  130

- **Type of reservoir tube**
  small funnel

- **Name of DNAPL used**
  4-chlorotoluene

- **Pool height measurement method**
  optical caliper

- **Height of model pool, h [mm]**
  9.9

- **Centrifuge rotational velocity at infiltration [RPM]**
  71.9

- **Angle of inclination of centr. platform, ψ [degrees]**
  7.73

- **Computed g-level at infiltration, n [gravities]**
  6.71

- **Height of prototype pool, H = nh [mm]**
  66.5

- **Prototype length of tube, L [mm]**
  872.6

- **αₙ-number, αₙ = Δρρw(ng/r₀)/(16μw²l)**
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**Notes regarding the test**

**Infiltration data**

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**Reference Number of Test**
c8-2.70f-130mm

**Date of Test**
November 25, 1997

**Date of Data Reduction**
November 25, 1997

- **Capillary tube diameter [mm]**
  2.70
- **Model length of tube, l [mm]**
  130
- **Type of reservoir tube**
  small funnel
- **Name of DNAPL used**
  4-chlorotoluene

**Pool height measurement method**
optical caliper

**Height of model pool, h [mm]**
15.2

**Centrifuge rotational velocity at infiltration [RPM]**
59.1

**Angle of inclination of centr. platform, \( \psi \) [degrees]**
11.37

**Computed g-level at infiltration, \( n \) [gravities]**
4.59

**Height of prototype pool, \( H = nh \) [mm]**
69.8

**Prototype length of tube, \( L \) [mm]**
596.9

\[ \alpha_n \text{-number, } \alpha_n = \frac{\Delta \rho \rho_w (ng) r_0^2}{16 \mu_w l \psi} \]
5.167

**Notes regarding the test**

**Infiltration data**

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**Reference Number of Test**
c9-2.70f-130mm

**Date of Test**
February 12, 1998

**Date of Data Reduction**
February 22, 1998

**Capillary tube diameter [mm]**
2.70

**Model length of tube, l [mm]**
130

**Type of reservoir tube**
small funnel

**Name of DNAPL used**
4-chlorotoluene

**Pool height measurement method**
optical caliper

**Height of model pool, h [mm]**
15.2

**Centrifuge rotational velocity at infiltration [RPM]**
47.0

**Angle of inclination of centr. platform, ψ [degrees]**
17.72

**Computed g-level at infiltration, n [gravities]**
3.00

**Height of prototype pool, H = nh [mm]**
45.6

**Prototype length of tube, L [mm]**
390.0

\[ \alpha_n = \frac{\Delta \rho \rho_w (ng)^4}{16 \mu_w^2 l} \]

\[ \alpha_n = 3.376 \]

**Notes regarding the test**
Very small pool height at infiltration possibly due to pre-infiltration.

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**Infiltration data**

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c10-2.70st-130mm  
**Date of Test**  
March 25, 1998  
**Date of Data Reduction**  
June 2, 1998  

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**Pool height measurement method**  
optical caliper  
**Height of model pool, h [mm]**  
19.8  

**Centrifuge rotational velocity at infiltration [RPM]**  
49.6  
**Angle of inclination of centr. platform, \( \psi \) [degrees]**  
15.99  
**Computed g-level at infiltration, n [gravities]**  
3.31  
**Height of prototype pool, \( H = nh \) [mm]**  
65.5  
**Prototype length of tube, L [mm]**  
429.9  
**\( \alpha_n \)-number, \( \alpha_n = \Delta \rho \rho_w (ng) r_0^3 / (16 \mu_w t) \)**  
3.721

**Notes regarding the test**

**Infiltration data**

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Date of Test: May 5, 1998
Date of Data Reduction: June 2, 1998

Capillary tube diameter [mm]: 2.70
Model length of tube, l [mm]: 130
Type of reservoir tube: 8 mm ID non-graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 10.2

Centrifuge rotational velocity at infiltration [RPM]: 64.9
Angle of inclination of centr. platform, \( \psi \) [degrees]: 9.46
Computed g-level at infiltration, n [gravities]: 5.50
Height of prototype pool, \( H = nh \) [mm]: 56.1
Prototype length of tube, L [mm]: 714.8
\( \alpha_n \)-number, \( \alpha_n = \Delta \rho \frac{ng}{r_0^3} \frac{1}{\mu_w l} \): 6.187

Notes regarding the test

Infiltration data

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**Reference Number of Test**: c12-2.70st-130mm  
**Date of Test**: May 12, 1998  
**Date of Data Reduction**: May 12, 1998

- **Capillary tube diameter [mm]**: 2.70  
- **Model length of tube, l [mm]**: 130  
- **Type of reservoir tube**: 8 mm ID non-graduated tube  
- **Name of DNAPL used**: 4-chlorotoluene  
- **Pool height measurement method**: optical caliper  
- **Height of model pool, h [mm]**: 10.4

- **Centrifuge rotational velocity at infiltration [RPM]**: 62.9  
- **Angle of inclination of centr. platform, Ψ [degrees]**: 10.06  
- **Computed g-level at infiltration, n [gravities]**: 5.18  
- **Height of prototype pool, H = nh [mm]**: 53.8  
- **Prototype length of tube, L [mm]**: 672.8  
- **αn-number, αn = Δρρw(ng)r0^(3/2)/(16µw^2l)**: 5.824

**Notes regarding the test**

**Infiltration data**

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C.5.2 2.20 mm Diameter Capillary Tubes
Reference Number of Test: c2-2.20ct-136mm
Date of Test: April 18, 1997
Date of Data Reduction: April 18, 1997

Capillary tube diameter [mm]: 2.20
Model length of tube, l [mm]: 136
Type of reservoir tube: 8 mm ID graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: volume graduations on res. tube
Height of model pool, h [mm]: 29.4

Centrifuge rotational velocity at infiltration [RPM]: 44.2
Angle of inclination of centr. platform, \( \psi \) [degrees]: 19.90
Computed g-level at infiltration, \( n \) [gravities]: 2.69
Height of prototype pool, \( H = nh \) [mm]: 79.1
Prototype length of tube, \( L \) [mm]: 366.1
\( \alpha_n \)-number, \( \alpha_n = \frac{\Delta \rho \rho_w (ng) r_0^2}{16 \mu_w l} \): 1.276

Notes regarding the test: Interface displacement not recorded.

Reference Number of Test: c3-2.20ct-136mm
Date of Test: April 25, 1997
Date of Data Reduction: April 25, 1997

Capillary tube diameter [mm]: 2.20
Model length of tube, l [mm]: 136
Type of reservoir tube: 8 mm ID graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 20.8

Centrifuge rotational velocity at infiltration [RPM]: 49.6
Angle of inclination of centr. platform, \( \psi \) [degrees]: 15.99
Computed g-level at infiltration, \( n \) [gravities]: 3.31
Height of prototype pool, \( H = nh \) [mm]: 68.8
Prototype length of tube, \( L \) [mm]: 449.7
\( \alpha_n \)-number, \( \alpha_n = \frac{\Delta \rho \rho_w (ng) r_0^2}{16 \mu_w l} \): 1.568

Notes regarding the test: Interface displacement not recorded.
**Reference Number of Test** | c4-2.20ct-136mm  
**Date of Test** | May 14, 1997  
**Date of Data Reduction** | May 14, 1997

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**Notes regarding the test**  
Interface displacement not recorded.
**Reference Number of Test**
c5-2.20ct-136mm

**Date of Test**
June 5, 1997

**Date of Data Reduction**
November 19, 1997

**Capillary tube diameter [mm]**
2.20

**Model length of tube, l [mm]**
136

**Type of reservoir tube**
8 mm ID graduated tube

**Name of DNAPL used**
4-chlorotoluene

**Pool height measurement method**
optical caliper

**Height of model pool, h [mm]**
10.2

**Centrifuge rotational velocity at infiltration [RPM]**
78.3

**Angle of inclination of centr. platform, ψ [degrees]**
6.52

**Computed g-level at infiltration, n [gravities]**
7.94

**Height of prototype pool, H = nh [mm]**
81.0

**Prototype length of tube, L [mm]**
1079.4

\[ \alpha_n, \text{number}, \alpha_n = \Delta \rho \rho_w (ng) r_0^5 / (16 \mu_w^2 l) \]
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**Notes regarding the test**

**Infiltration data**

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| **Capillary tube diameter [mm]**  | 2.20                    |
| **Model length of tube, l [mm]**  | 133                     |
| **Type of reservoir tube**        | small funnel            |
| **Name of DNAPL used**            | 4-chlorotoluene         |

| **Pool height measurement method**| optical caliper         |
| **Height of model pool, h [mm]** | 15.0                    |

| **Centrifuge rotational velocity at infiltration [RPM]** | 62.8                   |
| **Angle of inclination of centr. platform, ψ [degrees]** | 10.09                  |
| **Computed g-level at infiltration, n [gravities]**     | 5.16                   |
| **Height of prototype pool, H = nh [mm]**               | 77.4                   |
| **Prototype length of tube, L [mm]**                   | 686.2                  |
| **αₙ-number, αₙ = ∆ρpω(ng)ro⁴/(16μw²l)**                | 2.501                  |

**Notes regarding the test**
Interface displacement not recorded.
Reference Number of Test: c5-2.20f-133mm
Date of Test: June 5, 1997
Date of Data Reduction: November 19, 1997

Capillary tube diameter [mm]: 2.20
Model length of tube, l [mm]: 133
Type of reservoir tube: small funnel
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 10.2

Centrifuge rotational velocity at infiltration [RPM]: 75.4
Angle of inclination of centr. platform, ψ [degrees]: 7.03
Computed g-level at infiltration, n [gravities]: 7.37
Height of prototype pool, H = nh [mm]: 75.2
Prototype length of tube, L [mm]: 980.0

αₙ-number, αₙ = Δρρₙ(ng)r₀²/(16μₙ²l)

3.573

Notes regarding the test

Infiltration data

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C.5.3 1.33 mm Diameter Capillary Tubes
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**c7-1.33ct-135mm**

**Date of Test**

**October 15, 1997**

**Date of Data Reduction**

**November 13, 1997**

**Capillary tube diameter [mm]**

**1.33**

**Model length of tube, l [mm]**

**135**

**Type of reservoir tube**

**6.4 mm ID graduated tube**

**Name of DNAPL used**

**4-chlorotoluene**

**Pool height measurement method**

**optical caliper**

**Height of model pool, h [mm]**

**15.5**

**Centrifuge rotational velocity at infiltration [RPM]**

**86.7**

**Angle of inclination of centr. platform, ψ [degrees]**

**5.33**

**Computed g-level at infiltration, n [gravities]**

**9.71**

**Height of prototype pool, H = nh [mm]**

**150.5**

**Prototype length of tube, L [mm]**

**1310.4**

**α_n-number, α_n = Δρρ_w(ng)r_0^4/(16μ_w l)**

**0.619**

**Notes regarding the test**

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**Model length of tube, l [mm]**
135  
**Type of reservoir tube**
6.4 mm ID graduated tube  
**Name of DNAPL used**
4-chlorotoluene  
**Pool height measurement method**
optical caliper  
**Height of model pool, h [mm]**
23.9  
**Centrifuge rotational velocity at infiltration [RPM]**
75.4  
**Angle of inclination of centr. platform, \(\psi\) [degrees]**
7.03  
**Computed g-level at infiltration, n [gravities]**
7.37  
**Height of prototype pool, \(H = nh\) [mm]**
176.1  
**Prototype length of tube, L [mm]**
994.8  
**\(\alpha_n\)-number, \(\alpha_n = \Delta \rho p_w (ng)^{1/2}/(16 \mu_w l)\)**
0.470  

**Notes regarding the test**

**Infiltration data**

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Date of Test  February 12, 1998
Date of Data Reduction  February 22, 1998

Capillary tube diameter [mm]  1.33
Model length of tube, \( l \) [mm]  135
Type of reservoir tube  6.4 mm ID graduated tube
Name of DNAPL used  4-chlorotoluene

Pool height measurement method  optical caliper
Height of model pool, \( h \) [mm]  41.4

Centrifuge rotational velocity at infiltration [RPM]  54.0
Angle of inclination of centr. platform, \( \psi \) [degrees]  13.56
Computed g-level at infiltration, \( n \) [gravities]  3.87
Height of prototype pool, \( H = nh \) [mm]  160.2
Prototype length of tube, \( L \) [mm]  522.5
\( \alpha_n \)-number, \( \alpha_n = \Delta \rho g \mu_0 (ng)^{4/16} (\mu_0 \mu_w^2 l) \)  0.247

Notes regarding the test

Infiltration data

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Reference Number of Test: c11-1.33ct-135mm
Date of Test: May 5, 1998
Date of Data Reduction: May 7, 1998

Capillary tube diameter [mm]: 1.33
Model length of tube, l [mm]: 135
Type of reservoir tube: 6.4 mm ID graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 40.1

Centrifuge rotational velocity at infiltration [RPM]: 55.1
Angle of inclination of centr. platform, \( \psi \) [degrees]: 13.04
Computed g-level at infiltration, \( n \) [gravities]: 4.02
Height of prototype pool, \( H = nh \) [mm]: 161.2
Prototype length of tube, \( L \) [mm]: 542.7
\( \alpha_n \)-number, \( \alpha_n = \Delta \rho \rho_w (ng)r_0^{2/3}/(16\mu_w^2l) \): 0.256

Notes regarding the test:

Infiltration data

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Reference Number of Test: c12-1.33ct-135mm

Date of Test: May 12, 1998
Date of Data Reduction: May 12, 1998

Capillary tube diameter [mm]: 1.33
Model length of tube, l [mm]: 135
Type of reservoir tube: 6.4 mm ID graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 39.8

Centrifuge rotational velocity at infiltration [RPM]: 53.4
Angle of inclination of centr. platform, ψ [degrees]: 13.86
Computed g-level at infiltration, n [gravities]: 3.79
Height of prototype pool, H = nh [mm]: 150.9
Prototype length of tube, L [mm]: 511.7

αn-number, \( \alpha_n = \Delta \rho \rho_w T_0^4/(16 \mu_w^2 l) \): 0.242

Notes regarding the test

Infiltration data

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Reference Number of Test: c14-1.33ct-135mm
Date of Test: August 18, 1998
Date of Data Reduction: March 10, 1999

Capillary tube diameter [mm]: 1.33
Model length of tube, l [mm]: 135
Type of reservoir tube: 6.4 mm ID graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 30.7

Centrifuge rotational velocity at infiltration [RPM]: 56.1
Angle of inclination of centr. platform, \( \psi \) [degrees]: 12.59
Computed g-level at infiltration, \( n \) [gravities]: 4.16
Height of prototype pool, \( H = nh \) [mm]: 127.7
Prototype length of tube, \( L \) [mm]: 540.6
\( \alpha_n \)-number, \( \alpha_n = \Delta \rho \rho_w (ng)_0 r_0^2/(16 \mu_w l) \): 0.276

Notes regarding the test: Noticed pre-infiltration down to about 3 mm prior to spinning of centrifuge.

Infiltration data

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**Reference Number of Test**  c15-1.33ct-135mm

**Date of Test**  August 18, 1998

**Date of Data Reduction**  March 10, 1999

- **Capillary tube diameter [mm]**  1.33
- **Model length of tube, \( l \) [mm]**  135
- **Type of reservoir tube**  6.4 mm ID graduated tube
- **Name of DNAPL used**  4-chlorotoluene

**Pool height measurement method**  optical caliper

**Height of model pool, \( h \) [mm]**  17.0

- **Centrifuge rotational velocity at infiltration [RPM]**  81.5
- **Angle of inclination of centr. platform, \( \psi \) [degrees]**  6.02
- **Computed g-level at infiltration, \( n \) [gravities]**  8.59
- **Height of prototype pool, \( H = nh \) [mm]**  146.0
- **Prototype length of tube, \( L \) [mm]**  1116.6
- **\( \alpha_n \)-number, \( \alpha_n = \Delta \rho \rho \omega (n g)\rho_0^3 / (16 \mu_\omega^2 l) \)**  0.569

**Notes regarding the test**

**Infiltration data**

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**Reference Number of Test**
c16-1.33ct-135mm

**Date of Test**
January 26, 1999

**Date of Data Reduction**
March 9, 1999

Capillary tube diameter [mm] 1.33
Model length of tube, l [mm] 135
Type of reservoir tube 8 mm ID graduated tube
Name of DNAPL used 4-chlorotoluene

Pool height measurement method optical caliper
Height of model pool, h [mm] 13.0

Centrifuge rotational velocity at infiltration [RPM] 82.8
Angle of inclination of centr. platform, \( \psi \) [degrees] 5.84
Computed g-level at infiltration, \( n \) [gravities] 8.86
Height of prototype pool, \( H = nh \) [mm] 115.2
Prototype length of tube, \( L \) [mm] 1152.0

\[ \alpha_n\text{-number, } \alpha_n = \Delta\rho_p \rho_p (ng) r_0^2/(16 \mu_w^2 l) \]

0.587

**Notes regarding the test**

**Infiltration data**

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**Reference Number of Test**
c17-1.33ct-135mm

**Date of Test**
February 4, 1999

**Date of Data Reduction**
March 9, 1999

**Capillary tube diameter [mm]**
1.33

**Model length of tube, l [mm]**
135

**Type of reservoir tube**
8 mm ID graduated tube

**Name of DNAPL used**
4-chlorotoluene

**Pool height measurement method**
optical caliper

**Height of model pool, h [mm]**
14.7

**Centrifuge rotational velocity at infiltration [RPM]**
84.6

**Angle of inclination of centr. platform, \( \psi \) [degrees]**
5.59

**Computed g-level at infiltration, n [gravities]**
9.25

**Height of prototype pool, \( H = nh \) [mm]**
135.9

**Prototype length of tube, L [mm]**
1202.1

\[ \alpha_n = \Delta \rho_p \rho_w (ng)^{\frac{\alpha_n}{16 \mu_w}} \]
\[ \alpha_n = 0.613 \]

**Notes regarding the test**
Lower part of the capillary tube difficult to see.

**Infiltration data**

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Reference Number of Test c7-1.33f-129mm

Date of Test October 15, 1997

Date of Data Reduction November 13, 1997

Capillary tube diameter [mm] 1.33
Model length of tube, l [mm] 129
Type of reservoir tube small funnel
Name of DNAPL used 4-chlorotoluene

Pool height measurement method optical caliper
Height of model pool, h [mm] 15.0

Centrifuge rotational velocity at infiltration [RPM] 94.4
Angle of inclination of centr. platform, ψ [degrees] 4.50
Computed g-level at infiltration, n [gravities] 11.49
Height of prototype pool, H = nh [mm] 172.4
Prototype length of tube, L [mm] 1482.3

Notes regarding the test

Infiltration data

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Reference Number of Test  c8-1.33f-129mm
Date of Test  November 25, 1997
Date of Data Reduction  November 25, 1997

Capillary tube diameter [mm]  1.33
Model length of tube, l [mm]  129
Type of reservoir tube  small funnel
Name of DNAPL used  4-chlorotoluene

Pool height measurement method  optical caliper
Height of model pool, h [mm]  24.6

Centrifuge rotational velocity at infiltration [RPM]  70.4
Angle of inclination of centr. platform, ψ [degrees]  8.06
Computed g-level at infiltration, n [gravities]  6.44
Height of prototype pool, H = nh [mm]  158.5
Prototype length of tube, L [mm]  831.0
αn-number, αn = Δρρw(ng)^(1/4)/(16μw^2l)  0.430

Notes regarding the test

Infiltration data

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Reference Number of Test: c9-1.33f-129mm
Date of Test: February 12, 1998
Date of Data Reduction: February 22, 1998

Capillary tube diameter [mm]: 1.33
Model length of tube, l [mm]: 129
Type of reservoir tube: small funnel
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 36.6

Centrifuge rotational velocity at infiltration [RPM]: 51.9
Angle of inclination of centr. platform, ψ [degrees]: 14.65
Computed g-level at infiltration, n [gravities]: 3.59
Height of prototype pool, H = nh [mm]: 131.6
Prototype length of tube, L [mm]: 463.7

\( \alpha_n \)-number, \( \alpha_n = \frac{\Delta \rho \rho_w (ng) r_0^3}{(16 \mu_w)^2 l} \): 0.240

Notes regarding the test

Infiltration data

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Reference Number of Test c10-1.33st-129mm
Date of Test March 25, 1998
Date of Data Reduction June 2, 1998

Capillary tube diameter [mm] 1.33
Model length of tube, I [mm] 129
Type of reservoir tube 8 mm ID non-graduated tube
Name of DNAPL used 4-chlorotoluene

Pool height measurement method optical caliper
Height of model pool, h [mm] 40.1

Centrifuge rotational velocity at infiltration [RPM] 54.2
Angle of inclination of centr. platform, ψ [degrees] 13.47
Computed g-level at infiltration, n [gravities] 3.90
Height of prototype pool, H = nh [mm] 156.3
Prototype length of tube, L [mm] 502.8
αn-number, αn = Δρρw(ng)r0^4/(16μw^2l) 0.260

Notes regarding the test

Infiltration data

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Reference Number of Test  
c11-1.33st-129mm

Date of Test  
May 5, 1998

Date of Data Reduction  
May 7, 1998

Capillary tube diameter [mm]  
1.33

Model length of tube, I [mm]  
129

Type of reservoir tube  
8 mm ID non-graduated tube

Name of DNAPL used  
4-chlorotoluene

Pool height measurement method  
optical caliper

Height of model pool, h [mm]  
32.8

Centrifuge rotational velocity at infiltration [RPM]  
58.7

Angle of inclination of centr. platform, $\psi$ [degrees]  
11.53

Computed $g$-level at infiltration, n [gravities]  
4.53

Height of prototype pool, $H = nh$ [mm]  
148.7

Prototype length of tube, $L$ [mm]  
584.7

$\alpha_n$-number, $\alpha_n = \Delta \rho \rho w (ng) r_0^3/(16 \mu w^2 l)$  
0.303

Notes regarding the test

Infiltration data

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**Reference Number of Test**
c12-1.33st-129mm

**Date of Test**
May 12, 1998

**Date of Data Reduction**
June 2, 1998

- **Capillary tube diameter** [mm]: 1.33
- **Model length of tube, l [mm]**: 129
- **Type of reservoir tube**: 8 mm ID non-graduated tube
- **Name of DNAPL used**: 4-chlorotoluene

- **Pool height measurement method**: optical caliper
- **Height of model pool, h [mm]**: 33.2

- **Centrifuge rotational velocity at infiltration [RPM]**: 59.2
- **Angle of inclination of centr. platform, ψ [degrees]**: 11.34
- **Computed g-level at infiltration, n [gravities]**: 4.61
- **Height of prototype pool, H = nh [mm]**: 152.9
- **Prototype length of tube, L [mm]**: 594.2
- \[ \alpha_n \text{-number, } \alpha_n = \Delta \rho \rho_w (ng) r_0^4 / (16 \mu_w l) \]
- **Value**: 0.308

**Notes regarding the test**

**Infiltration data**

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**Reference Number of Test**  c13-1.33st-120mm  
**Date of Test**  July 2, 1998  
**Date of Data Reduction**  July 14, 1998  

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**Pool height measurement method**  optical caliper  
**Height of model pool, h [mm]**  30.0

**Centrifuge rotational velocity at infiltration [RPM]**  60.3  
**Angle of inclination of centr. platform, ψ [degrees]**  10.93  
**Computed g-level at infiltration, n [gravities]**  4.77  

**Height of prototype pool, H = nh [mm]**  143.1  
**Prototype length of tube, L [mm]**  572.6  
**αn-number, αn = Δρρw(ng)r0^3/16μw^2l**  0.342  

**Notes regarding the test**  Lower part of the capillary tube difficult to see.  

**Infiltration data**

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**Reference Number of Test**  
c14-1.33st-120mm  
**Date of Test**  
July 8, 1998  
**Date of Data Reduction**  
July 14, 1998  

Capillary tube diameter [mm]  
1.33  
Model length of tube, \( l \) [mm]  
120  
Type of reservoir tube  
8 mm ID non-graduated tube  
Name of DNAPL used  
4-chlorotoluene  

Pool height measurement method  
optical caliper  
Height of model pool, \( h \) [mm]  
30.2  

Centrifuge rotational velocity at infiltration [RPM]  
58.8  
Angle of inclination of centr. platform, \( \psi \) [degrees]  
11.49  
Computed \( g \)-level at infiltration, \( n \) [gravities]  
4.55  
Height of prototype pool, \( H = nh \) [mm]  
137.3  
Prototype length of tube, \( L \) [mm]  
545.7  
\( \alpha_n \)-number, \( \alpha_n = \Delta \rho \rho_w (ngr_0^4/(16\mu_w^2l)) \)  
0.326  

Notes regarding the test  
Lower part of the capillary tube difficult to see.  

**Infiltration data**  

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**Reference Number of Test**
c16-1.33st-120mm

**Date of Test**
January 26, 1999

**Date of Data Reduction**
February 1, 1999

**Capillary tube diameter [mm]**
1.33

**Model length of tube, l [mm]**
120

**Type of reservoir tube**
8 mm ID non-graduated tube

**Name of DNAPL used**
4-chlorotoluene

**Pool height measurement method**
optical caliper

**Height of model pool, h [mm]**
13.0

**Centrifuge rotational velocity at infiltration [RPM]**
81.1

**Angle of inclination of centr. platform, \( \psi \) [degrees]**
6.08

**Computed g-level at infiltration, \( n \) [gravities]**
8.51

**Height of prototype pool, \( H = nh \) [mm]**
110.6

**Prototype length of tube, \( L \) [mm]**
1020.7

**\( \alpha_n \)-number, \( \alpha_n = \Delta \rho \rho_w (n g) r_0 \frac{v}{(16 \mu_w l)} \)**
0.611

**Notes regarding the test**
Lower part of the capillary tube difficult to see.

**Infiltration data**

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Date of Test  February 4, 1999
Date of Data Reduction  March 9, 1999

Capillary tube diameter [mm]  1.33
Model length of tube, l [mm]  120
Type of reservoir tube  8 mm ID non-graduated tube
Name of DNAPL used  4-chlorotoluene

Pool height measurement method  optical caliper
Height of model pool, h [mm]  30.0

Centrifuge rotational velocity at infiltration [RPM]  57.6
Angle of inclination of centr. platform, ψ [degrees]  11.96
Computed g-level at infiltration, n [gravities]  4.37
Height of prototype pool, H = nh [mm]  131.2
Prototype length of tube, L [mm]  524.6
αₙ-number, \( \alpha_n = \Delta \rho \rho_w (n g r_0^4 / (16 \mu_w l)) \)  0.314

Notes regarding the test  Lower part of the capillary tube difficult to see.

Infiltration data

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Reference Number of Test: c13-1.33st-60mm
Date of Test: July 2, 1998
Date of Data Reduction: July 14, 1998

Capillary tube diameter [mm]: 1.33
Model length of tube, l [mm]: 60
Type of reservoir tube: 8 mm ID non-graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 15.0

Centrifuge rotational velocity at infiltration [RPM]: 77.3
Angle of inclination of centr. platform, ψ [degrees]: 6.69
Computed g-level at infiltration, n [gravities]: 7.74
Height of prototype pool, H = nh [mm]: 116.1
Prototype length of tube, L [mm]: 464.3

αn-number, αn = Δρρw(ng)r0⁵/(16µw²I)

1.111

Notes regarding the test

Infiltration data

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Reference Number of Test: c14-1.33st-60mm
Date of Test: July 8, 1998
Date of Data Reduction: July 14, 1998

Capillary tube diameter [mm]: 1.33
Model length of tube, l [mm]: 60
Type of reservoir tube: 8 mm ID non-graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 15.2

Centrifuge rotational velocity at infiltration [RPM]: 80.2
Angle of inclination of centr. platform, ψ [degrees]: 6.22
Computed g-level at infiltration, n [gravities]: 8.32
Height of prototype pool, H = nh [mm]: 126.5
Prototype length of tube, L [mm]: 499.3
αₙ-number, αₙ = Δρρw(ng)²/(16μw²l): 1.194

Notes regarding the test

Infiltration data

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Reference Number of Test

Date of Test

Date of Data Reduction

Capillary tube diameter [mm]
Model length of tube, l [mm]
Type of reservoir tube
Name of DNAPL used

Pool height measurement method
Height of model pool, h [mm]

Centrifuge rotational velocity at infiltration [RPM]
Angle of inclination of centr. platform, ψ [degrees]
Computed g-level at infiltration, n [gravities]
Height of prototype pool, H = nh [mm]
Prototype length of tube, L [mm]

αn-number, αn = Δρρw(ng)r0^3/(16νw^2l)

Notes regarding the test

Infiltration data

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**Reference Number of Test**  
c16-1.33st-60mm

**Date of Test**  
January 26, 1999

**Date of Data Reduction**  
March 9, 1999

**Capillary tube diameter [mm]**  
1.33

**Model length of tube, l [mm]**  
60

**Type of reservoir tube**  
8 mm ID non-graduated tube

**Name of DNAPL used**  
4-chlorotoluene

**Pool height measurement method**  
optical caliper

**Height of model pool, h [mm]**  
12.4

**Centrifuge rotational velocity at infiltration [RPM]**  
87.0

**Angle of inclination of centr. platform, ψ [degrees]**  
5.29

**Computed g-level at infiltration, n [gravities]**  
9.77

**Height of prototype pool, H = nh [mm]**  
121.2

**Prototype length of tube, L [mm]**  
586.4

\[
a_n = \frac{\Delta \rho \rho_w (ng)^{\frac{3}{2}}}{(16 \mu_w^2 l)}
\]

\[
1.403
\]

**Notes regarding the test**

**Infiltration data**

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Reference Number of Test: c17-1.33st-60mm
Date of Test: February 4, 1999
Date of Data Reduction: March 9, 1999

Capillary tube diameter [mm]: 1.33
Model length of tube, l [mm]: 60
Type of reservoir tube: 8 mm ID non-graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 15.0

Centrifuge rotational velocity at infiltration [RPM]: 87.3
Angle of inclination of centr. platform, \( \psi \) [degrees]: 5.25
Computed g-level at infiltration, \( n \) [gravities]: 9.84
Height of prototype pool, \( H = nh \) [mm]: 147.6
Prototype length of tube, \( L \) [mm]: 590.4
\( \alpha_n \)-number, \( \alpha_n = \frac{\Delta \rho \rho_w (ng) r_0^6}{16 \mu_w^2 l} \): 1.413

Notes regarding the test

Infiltration data

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Reference Number of Test: c13-1.33st-40mm
Date of Test: July 2, 1998
Date of Data Reduction: July 14, 1998

Capillary tube diameter [mm]: 1.33
Model length of tube, l [mm]: 40
Type of reservoir tube: 8 mm ID non-graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 9.9

Centrifuge rotational velocity at infiltration [RPM]: 97.3
Angle of inclination of centr. platform, ψ [degrees]: 4.23
Computed g-level at infiltration, n [gravities]: 12.20
Height of prototype pool, H = nh [mm]: 120.8
Prototype length of tube, L [mm]: 488.1

αₙ-number, \( \alpha_n = \Delta \rho \rho w (ng)^3 / (16 \mu_w^2 l) \): 2.628

Notes regarding the test

Infiltration data

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Reference Number of Test

Date of Test

Date of Data Reduction

Capillary tube diameter [mm] 1.33
Model length of tube, l [mm] 40
Type of reservoir tube 8 mm ID non-graduated tube
Name of DNAPL used 4-chlorotoluene

Pool height measurement method optical caliper
Height of model pool, h [mm] 9.9

Centrifuge rotational velocity at infiltration [RPM] 91.5
Angle of inclination of centr. platform, ψ [degrees] 4.78
Computed g-level at infiltration, n [gravities] 10.80
Height of prototype pool, H = nh [mm] 106.9
Prototype length of tube, L [mm] 432.0
α_n-number, \( α_n = \Delta \rho \rho_w (ng)^{0.5}(16\mu_w^2 l) \) 2.326

Notes regarding the test
Test interrupted during spinning after DNAPL level in reservoir tube dropped following displacement of an air bubble initially trapped in Teflon tape collar. Reservoir was refilled to target height and test allowed to resume.

Infiltration data

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<th>Model</th>
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Reference Number of Test  c15-1.33st-40mm
Date of Test  August 18, 1998
Date of Data Reduction  March 10, 1999

Capillary tube diameter [mm]  1.33
Model length of tube, l [mm]  40
Type of reservoir tube  8 mm ID non-graduated tube
Name of DNAPL used  4-chlorotoluene

Pool height measurement method  optical caliper
Height of model pool, h [mm]  9.9

Centrifuge rotational velocity at infiltration [RPM]  92.8
Angle of inclination of centr. platform, \( \psi \) [degrees]  4.65
Computed g-level at infiltration, \( n \) [gravities]  11.11
Height of prototype pool, \( H = nh \) [mm]  110.0
Prototype length of tube, \( L \) [mm]  444.3
\( \alpha_r \)-number, \( \alpha_r = \Delta \rho p_w (ng)^{1/2}/(16 \mu_w l) \)  2.392

Notes regarding the test

Infiltration data

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Reference Number of Test  
c16-1.33st-40mm

Date of Test  
January 26, 1999

Date of Data Reduction  
March 9, 1999

Capillary tube diameter [mm]  
1.33

Model length of tube, l [mm]  
40

Type of reservoir tube  
8 mm ID non-graduated tube

Name of DNAPL used  
4-chlorotoluene

Pool height measurement method  
optical caliper

Height of model pool, h [mm]  
12.7

Centrifuge rotational velocity at infiltration [RPM]  
82.7

Angle of inclination of centr. platform, \(\psi\) [degrees]  
5.85

Computed g-level at infiltration, n [gravities]  
8.84

Height of prototype pool, \(H = nh\) [mm]  
112.3

Prototype length of tube, \(L\) [mm]  
353.6

\(\alpha_n\)-number,  
\[\alpha_n = \frac{\Delta \rho \rho_w (ng)^r_0}{16\mu_w^2 l}\]  
1.904

Notes regarding the test

Infiltration data

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Reference Number of Test: c17-1.33st-40mm
Date of Test: February 4, 1999
Date of Data Reduction: March 9, 1999

Capillary tube diameter [mm]: 1.33
Model length of tube, l [mm]: 40
Type of reservoir tube: 8 mm ID non-graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 10.2

Centrifuge rotational velocity at infiltration [RPM]: 88.4
Angle of inclination of centr. platform, ψ [degrees]: 5.12
Computed g-level at infiltration, n [gravities]: 10.09
Height of prototype pool, H = nh [mm]: 102.9
Prototype length of tube, L [mm]: 403.5
αn-number, αn = Δρρw(g)r0^4/(16µw^2l): 2.172

Notes regarding the test

Infiltration data

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C.5.4 0.66 mm Diameter Capillary Tubes
Reference Number of Test: c3-0.66ct-137mm
Date of Test: April 25, 1997
Date of Data Reduction: April 25, 1997

Capillary tube diameter [mm]: 0.66
Model length of tube, l [mm]: 137
Type of reservoir tube: 5.4 mm ID graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 44.7

Centrifuge rotational velocity at infiltration [RPM]: 71.5
Angle of inclination of centr. platform, ψ [degrees]: 7.81
Computed g-level at infiltration, n [gravities]: 6.64
Height of prototype pool, H = nh [mm]: 296.8
Prototype length of tube, L [mm]: 909.6

αₙ-number, αₙ = Δρₚw(ng)½r₀⁴/(16μₚ²l) = 0.0253

Notes regarding the test: Interface displacement not recorded.
Reference Number of Test: c7-0.66ct-137mm
Date of Test: October 15, 1997
Date of Data Reduction: November 13, 1997

Capillary tube diameter [mm]: 0.66
Model length of tube, l [mm]: 137
Type of reservoir tube: 5.4 mm ID graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 31.2

Centrifuge rotational velocity at infiltration [RPM]: 89.8
Angle of inclination of centr. platform, ψ [degrees]: 4.97
Computed g-level at infiltration, n [gravities]: 10.41
Height of prototype pool, H = nh [mm]: 324.7
Prototype length of tube, L [mm]: 1425.7
αn-number, αn = ∆ρρw(ng)r0^2/(16μw^2l): 0.0397

Notes regarding the test

Infiltration data

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**Reference Number of Test**
c8-0.66ct-137mm

**Date of Test**
November 25, 1997

**Date of Data Reduction**
November 25, 1997

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<td>Pool height measurement method</td>
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<td>Height of model pool, h [mm]</td>
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**Notes regarding the test**
Infiltration displacement very difficult to observe on film.
**Reference Number of Test**  
c9-0.66ct-137mm

**Date of Test**  
February 12, 1998

**Date of Data Reduction**  
June 2, 1998

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**Pool height measurement method**  
optical caliper

**Height of model pool, h [mm]**  
50.0

**Centrifuge rotational velocity at infiltration [RPM]**  
72.3

**Angle of inclination of centr. platform, ψ [degrees]**  
7.64

**Computed g-level at infiltration, n [gravities]**  
6.79

**Height of prototype pool, H = nh [mm]**  
339.3

**Prototype length of tube, L [mm]**  
929.7

**αn-number, αn = ∆ρρw(ng)r0^3/(16µw^2l)**  
0.0259

**Notes regarding the test**

**Infiltration data**

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Reference Number of Test: c11-0.66ct-137mm
Date of Test: May 5, 1998
Date of Data Reduction: June 2, 1998

Capillary tube diameter [mm]: 0.66
Model length of tube, l [mm]: 137
Type of reservoir tube: 5.4 mm ID graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 42.7

Centrifuge rotational velocity at infiltration [RPM]: 75.9
Angle of inclination of centr. platform, ψ [degrees]: 6.94
Computed g-level at infiltration, n [gravities]: 7.46
Height of prototype pool, H = nh [mm]: 318.8
Prototype length of tube, L [mm]: 1022.7
α_n-number, α_n = Δρρ_r(ng)r_0^4/(16μ_r^2l): 0.0285

Notes regarding the test

Infiltration data

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Reference Number of Test: c7-0.66f-126mm

Date of Test: October 15, 1997

Date of Data Reduction: November 13, 1997

- **Capillary tube diameter [mm]**: 0.66
- **Model length of tube, l [mm]**: 126
- **Type of reservoir tube**: small funnel
- **Name of DNAPL used**: 4-chlorotoluene
- **Pool height measurement method**: optical caliper
- **Height of model pool, h [mm]**: 30.5
- **Centrifuge rotational velocity at infiltration [RPM]**: 88.6
- **Angle of inclination of centr. platform, \( \psi \) [degrees]**: 5.10
- **Computed g-level at infiltration, n [gravities]**: 10.13
- **Height of prototype pool, \( H = nh \) [mm]**: 309.0
- **Prototype length of tube, L [mm]**: 1276.7
- **\( \alpha_n \)-number, \( \alpha_n = \Delta \rho \rho_w (n g) r_0^3 / (16 \mu_w^2 l) \)**: 0.0420

**Notes regarding the test**

**Infiltration data**

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**Reference Number of Test**
c8-0.66f-126mm

**Date of Test**
November 25, 1997

**Date of Data Reduction**
November 25, 1997

**Capillary tube diameter [mm]**
0.66

**Model length of tube, l [mm]**
126

**Type of reservoir tube**
small funnel

**Name of DNAPL used**
4-chlorotoluene

**Pool height measurement method**
optical caliper

**Height of model pool, h [mm]**
46.5

**Centrifuge rotational velocity at infiltration [RPM]**
70.8

**Angle of inclination of centr. platform, ψ [degrees]**
7.97

**Computed g-level at infiltration, n [gravities]**
6.51

**Height of prototype pool, H = nh [mm]**
302.9

**Prototype length of tube, L [mm]**
820.7

\[ α_n = \frac{\Delta \rho \rho (ng)r_0^4}{16 \mu_w l} \]

\[ 0.0270 \]

**Notes regarding the test**
Infiltration displacement very difficult to observe on film.

---

**Reference Number of Test**
c9-0.66f-126mm

**Date of Test**
February 12, 1998

**Date of Data Reduction**
February 22, 1998

**Capillary tube diameter [mm]**
0.66

**Model length of tube, l [mm]**
126

**Type of reservoir tube**
small funnel

**Name of DNAPL used**
4-chlorotoluene

**Pool height measurement method**
optical caliper

**Height of model pool, h [mm]**
42.9

**Centrifuge rotational velocity at infiltration [RPM]**
71.4

**Angle of inclination of centr. platform, ψ [degrees]**
7.83

**Computed g-level at infiltration, n [gravities]**
6.62

**Height of prototype pool, H = nh [mm]**
284.1

**Prototype length of tube, L [mm]**
834.3

\[ α_n = \frac{\Delta \rho \rho (ng)r_0^4}{16 \mu_w l} \]

\[ 0.0274 \]

**Notes regarding the test**
Infiltration displacement very difficult to observe on film.
Reference Number of Test
Date of Test
Date of Data Reduction
Capillary tube diameter [mm]
Model length of tube, l [mm]
Type of reservoir tube
Name of DNAPL used
Pool height measurement method
Height of model pool, h [mm]
Centrifuge rotational velocity at infiltration [RPM]
Angle of inclination of centr. platform, ψ [degrees]
Computed g-level at infiltration, n [gravities]
Height of prototype pool, H = nh [mm]
Prototype length of tube, L [mm]
\(\alpha_n\)-number, \(\alpha_n = \Delta\rho\rho_w(ng)r_0^3/(16\mu_w l)\)
Notes regarding the test

Infiltration data

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Reference Number of Test c11-0.66st-126mm

Date of Test May 5, 1998

Date of Data Reduction June 2, 1998

Capillary tube diameter [mm] 0.66
Model length of tube, l [mm] 126
Type of reservoir tube 8 mm ID non-graduated tube
Name of DNAPL used 4-chlorotoluene

Pool height measurement method optical caliper
Height of model pool, h [mm] 56.1

Centrifuge rotational velocity at infiltration [RPM] 65.9
Angle of inclination of centr. platform, ψ [degrees] 9.18
Computed g-level at infiltration, n [gravities] 5.66
Height of prototype pool, H = nh [mm] 317.7
Prototype length of tube, L [mm] 713.6

$\alpha_n$-number, $\alpha_n = \Delta \rho \rho_w (n g) r_0^4 / 16 \mu_w l$
0.0235

Notes regarding the test

Infiltration data

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Reference Number of Test

Date of Test

Date of Data Reduction

Capillary tube diameter [mm]

Model length of tube, \( I \) [mm]

Type of reservoir tube

Name of DNAPL used

Pool height measurement method

Height of model pool, \( h \) [mm]

Centrifuge rotational velocity at infiltration [RPM]

Angle of inclination of centr. platform, \( \psi \) [degrees]

Computed g-level at infiltration, \( n \) [gravities]

Height of prototype pool, \( H = nh \) [mm]

Prototype length of tube, \( L \) [mm]

\( \alpha_n \)-number,

\[ \alpha_n = \frac{\Delta \rho \rho_w (ng)r_0^2}{16 \mu_w^2 l} \]

Notes regarding the test

Test interrupted at its early stage because water was flowing out of the box.

Infiltration data

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**Reference Number of Test**  
c18b-0.66ct-119mm

**Date of Test**  
August 11, 1999

**Date of Data Reduction**  
August 12, 1999

**Capillary tube diameter [mm]**  
0.66

**Model length of tube, l [mm]**  
119

**Type of reservoir tube**  
5.4 mm ID graduated tube

**Name of DNAPL used**  
4-chlorotoluene

**Pool height measurement method**  
optical caliper

**Height of model pool, h [mm]**  
25.4

**Centrifuge rotational velocity at infiltration [RPM]**  
79.6

**Angle of inclination of centr. platform, ψ [degrees]**  
6.31

**Computed g-level at infiltration, n [gravities]**  
8.20

**Height of prototype pool, H = nh [mm]**  
208.2

**Prototype length of tube, L [mm]**  
975.6

\[ \alpha_n = \Delta \rho \rho (n g_r)^{\frac{4}{16 \mu_w^2 l}} \]

\[ \alpha_n = 0.0360 \]

**Notes regarding the test**  
Test interrupted at its early stage because water was flowing out of the box. Prototype pool is lower than expected possibly due to pre-infiltration into the tube.

**Infiltration data**

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Reference Number of Test c18c-0.66ct-119mm
Date of Test August 11, 1999
Date of Data Reduction August 12, 1999

Capillary tube diameter [mm] 0.66
Model length of tube, l [mm] 119
Type of reservoir tube 5.4 mm ID graduated tube
Name of DNAPL used 4-chlorotoluene

Pool height measurement method optical caliper
Height of model pool, h [mm] 25.7*

Centrifuge rotational velocity at infiltration [RPM] 67.8
Angle of inclination of centr. platform, ψ [degrees] 8.68
Computed g-level at infiltration, n [gravities] 5.99*
Height of prototype pool, H = nh [mm] 153.8*
Prototype length of tube, L [mm] 712.3*
αₙ-number, αₙ = Δρρw(ng)r₀²/(16µw²l) 0.0263

Notes regarding the test Test interrupted at its early stage because water was flowing out of the box. Significant pre-infiltration observed on this tube after test was interrupted, probably because water in box dropped below level of water inside the reservoir tube creating a water head difference across the tube. Point at depth 115 mm difficult to see on film.

Infiltration data

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Reference Number of Test: c19a-0.66ct-119mm
Date of Test: August 18, 1999
Date of Data Reduction: August 19, 1999

Capillary tube diameter [mm]: 0.66
Model length of tube, I [mm]: 119
Type of reservoir tube: 5.4 mm ID graduated tube
Name of DNAPL used: 4-chlorotoluene

Pool height measurement method: optical caliper
Height of model pool, h [mm]: 27.2

Centrifuge rotational velocity at infiltration [RPM]: 91.5
Angle of inclination of centr. platform, ψ [degrees]: 4.78
Computed g-level at infiltration, n [gravities]: 10.80
Height of prototype pool, H = nh [mm]: 293.8
Prototype length of tube, L [mm]: 1285.3
αn-number, \( \alpha_n = \Delta \rho p \omega^4 r_0^4 / (16 \mu_w^2 l) \): 0.0474

Notes regarding the test:
Infiltration data

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Reference Number of Test c19b-0.66ct-119mm
Date of Test August 18, 1999
Date of Data Reduction August 19, 1999

Capillary tube diameter [mm] 0.66
Model length of tube, l [mm] 119
Type of reservoir tube 5.4 mm ID graduated tube
Name of DNAPL used 4-chlorotoluene

Pool height measurement method optical caliper
Height of model pool, h [mm] 25.9

Centrifuge rotational velocity at infiltration [RPM] 88.3
Angle of inclination of centr. platform, \( \psi \) [degrees] 5.14
Computed g-level at infiltration, \( n \) [gravities] 10.06
Height of prototype pool, \( H = nh \) [mm] 260.7
Prototype length of tube, \( L \) [mm] 1197.7
\[ \alpha_n \text{-number, } \alpha_n = \Delta \rho p_w (n g r_0^2)/(16 \mu_w^2 l) \] 0.0442

Notes regarding the test Point at depth 65 mm very hard to see because of brightness of back light.

Infiltration data

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**Reference Number of Test**
c20a-0.66ct-119mm

**Date of Test**
December 21, 1999

**Date of Data Reduction**
December 21, 1999

- **Capillary tube diameter [mm]**: 0.66
- **Model length of tube, l [mm]**: 119
- **Type of reservoir tube**: 5.4 mm ID graduated tube
- **Name of DNAPL used**: 4-chlorotoluene

- **Pool height measurement method**: optical caliper
- **Height of model pool, h [mm]**: 20.1

- **Centrifuge rotational velocity at infiltration [RPM]**: 103.8
- **Angle of inclination of centr. platform, \( \psi \) [degrees]**: 3.72
- **Computed g-level at infiltration, \( n \) [gravities]**: 13.88
- **Height of prototype pool, \( H = nh \) [mm]**: 278.9
- **Prototype length of tube, \( L \) [mm]**: 1651.4
- **\( \alpha_n \)-number, \( \alpha_n = \Delta \rho \rho_w (n g) r_0^4 / (16 \mu_w^2 l) \)**: 0.0609

**Notes regarding the test**

**Infiltration data**

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| **Capillary tube diameter [mm]** | 0.66 |
| **Model length of tube, l [mm]** | 119 |
| **Type of reservoir tube**       | 5.4 mm ID graduated tube |
| **Name of DNAPL used**           | 4-chlorotoluene |

| **Pool height measurement method** | optical caliper |
| **Height of model pool, h [mm]**  | 20.8 |

| **Centrifuge rotational velocity at infiltration [RPM]** | 90.7 |
| **Angle of inclination of centr. platform, Ψ [degrees]** | 4.87 |
| **Computed g-level at infiltration, n [gravities]**      | 10.61 |
| **Height of prototype pool, H = nh [mm]**                | 220.8 |
| **Prototype length of tube, L [mm]**                     | 1263.1 |
| **\( \alpha_n \)-number, \( \alpha_n = \Delta \rho \rho_w (ng) r_0^2/(16 \mu_w^2 l) \)** | 0.0466 |

**Notes regarding the test**

Height of prototype pool at infiltration was somewhat low. Pre-infiltration could explain this behavior but was not observed prior to spinning.
Reference Number of Test  c21-0.66ct-100mm
Date of Test  December 22, 1999
Date of Data Reduction  December 22, 1999

Capillary tube diameter [mm]  0.66
Model length of tube, l [mm]  100
Type of reservoir tube  7.8 mm ID graduated tube
Name of DNAPL used  4-chlorotoluene

Pool height measurement method  optical caliper
Height of model pool, h [mm]  48.8

Centrifuge rotational velocity at infiltration [RPM]  64.0
Angle of inclination of centr. platform, ψ [degrees]  9.72
Computed g-level at infiltration, n [gravities]  5.35
Height of prototype pool, H = nh [mm]  261.2
Prototype length of tube, L [mm]  535.2
αn-number, αn = Δρρw(ng)4/(16μw2l)  0.0280

Notes regarding the test  Pre-infiltration down to 3 mm observed prior to spinning.
This was taken into account in the values of h, l and zc.

Infiltration data

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BIOGRAPHICAL NOTE

Laurent Claude Levy, the son of Drs. Jean Bernard and Françoise Levy, was born in Paris, France, on March 6, 1973. He graduated from Lycée Chaptal, Paris, in June 1990 with a Baccalauréat C (mathematics and physics). He attended engineering preparatory school at the Lycées Condorcet and Henri IV. He was admitted to Ecole Centrale Paris (ECP), located in Châtenay-Malabry, in the fall of 1992, where he majored in civil engineering. He graduated from ECP in June 1995 with a Diplôme d’Ingénieur as well as a Diplôme d’Études Avancées (DEA) in Modélisation des Structures et Ouvrages dans leur Environnement (modeling of civil engineering structures in their environment). As part of his undergraduate curriculum, he worked in Birmingham, England, in the summer of 1994 as a field engineer for Christiani & Nielsen, a civil engineering contractor. In the spring of 1995, he worked in Paris as a research engineer for the SNCF Eole project (an express subway line). He was also a teaching assistant (“colleur”) in physics at the Lycée Lavoisier, Paris, from fall 1993 to spring 1995.

Levy started his graduate studies at MIT in the fall of 1995, and passed his doctoral exam in January 1997. In 1998, he received the Robert Guenassia award for outstanding graduate work. While a research assistant for Prof. P. J. Culligan and Dr. J. T. Germaine throughout his studies at MIT, he was also a teaching assistant in a geotechnical engineering undergraduate course in the fall of 1997, and in a probability and statistics undergraduate course in the fall of 1998, for which he received the departmental effective teaching assistant award. In the spring of 2001, he was the lecturer for a graduate course on waste containment and remediation technologies. Levy is a member of the American Society of Civil Engineers and a member of the scientific research society Sigma Xi.

Additionally, during December 1997, Levy and his friend Kurt J. Sjoblom (PhD, 2000) drove non stop from Boston to San Francisco in a 1978 Olds mobile Delta 88, which had to be junked upon arrival. The trip took 62 hours.

Levy took a year away from MIT in 2000 for his military service, and served as a commissioned officer and platoon leader in the French army at the 5ème Régiment du Génie (5th Engineering Regiment) in Versailles.


Publications


Presentations


