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Interface pinning of immiscible gravity-exchange flows in porous media

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We study the gravity-exchange flow of two immiscible fluids in a porous medium and show that, in contrast with the miscible case, a portion of the initial interface remains pinned at all times. We elucidate, by means of micromodel experiments, the pore-level mechanism responsible for capillary pinning at the macroscale. We propose a sharp-interface gravity-current model that incorporates capillarity and quantitatively explains the experimental observations, including the $x \sim t^{1/2}$ spreading behavior at intermediate times and the fact that capillarity stops a finite-release current. Our theory and experiments suggest that capillary pinning is potentially an important, yet unexplored, trapping mechanism during CO$_2$ sequestration in deep saline aquifers.

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I. INTRODUCTION

Gravity-exchange flows, in which density differences induce fluid segregation and migration, occur in processes as diverse as debris flows [1], seawater intrusion into groundwater [2], and coating of substrates by thin films [3]. More recently gravity-current flows have emerged as fundamental descriptors of the fluid mechanics of carbon dioxide (CO$_2$) sequestration and coating of substrates by thin films [3]. More recently gravity-exchange flows in porous media have a long history [11, 12], and this prediction is in excellent agreement with experiments, including the $x \sim t^{1/2}$ spreading behavior at intermediate times and the fact that capillarity stops a finite-release current. Our theory and experiments suggest that capillary pinning is potentially an important, yet unexplored, trapping mechanism during CO$_2$ sequestration in deep saline aquifers.

II. LOCK-EXCHANGE EXPERIMENTS

We conducted lock-exchange experiments in rectangular, quasi-2D flow cells packed with glass beads. We constructed four flow cells with different heights (2.5 cm, 5.2 cm, 10.3 cm, 20 cm) and the same length (~55 cm) and thickness (0.9 cm). Each flow cell consists of three pieces of laser-cut acrylic: solid front and rear panels and a middle spacer that frames the working area. The spacer is clamped between the front and rear panels via bolts. Once assembled, we orient the cell “vertically” [Fig. 2(a)] and fill it with glass beads via a port on the spacer. We shake the cell during filling to generate a tight, consistent bead pack. The porosity of the packed flow cell ranges from 0.38 to 0.42. Once the cell is full, we plug this port. We use non-monodisperse glass beads with nominal bead sizes ranging from 0.36 mm to 2.1 mm (Fig. 3). The permeability of the packed flow cell for 1mm beads is 0.88 × 10$^{-5}$ cm$^2$.

In the miscible lock-exchange experiments, we used water with blue food dye as the buoyant fluid and a mixture of glycerol and water (77.5% glycerol by mass, $\rho = 1.2$ g/cm$^3$, $\mu = 0.47$ g/cm-s) as the ambient fluid. In the immiscible lock-exchange experiments, we used air as the buoyant fluid. We used one of silicone oil ($\rho = 0.96$ g/cm$^3$, $\mu = 0.48$ g/cm-s, $\gamma = 20$ dyn/cm), propylene glycol ($\rho = 1.04$ g/cm$^3$, $\mu = 0.46$ g/cm-s, $\gamma = 41$ dyn/cm), or the glycerol-water mixture (as above, $\gamma = 63$ dyn/cm) as the ambient fluid. We measured the advancing and receding contact angles of each of the fluid pairs used in our experiments (air and silicon oil; air and propylene glycol; and air and a glycerol-water mixture) on both glass and acrylic substrates. The measurements were made using the sessile drop method, on a ramé-hart Model 590 Advanced Automated Goniometer.

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lower hinge by "hinges" in the macroscopic interface. We denote the height of the air (dark) spreading over the same glycerol-water mixture. Part of predictions of sharp-interface models.

To add the fluids to the cell, we orient the cell vertically. We add the ambient fluid via a port near the bottom in the vertical orientation [Fig. 2(a)]. We inject the ambient fluid with a syringe pump in order to measure the volume injected. Once we close all ports. In the immiscible lock-exchange flow, the buoyant fluid does not need to be injected because it is air.

To initiate an experiment, we quickly rotate the cell by 90° so that it lies horizontally on the table, between an LED backlight and a digital camera [Fig. 2(b)]. We record the experiment as a sequence of still images. We disassemble the cell, discard the beads, and wash the acrylic plates after each experiment.

Figure 4 shows snapshots from the contact angle goniometer for measurements on a glass substrate. The advancing and receding contact angles of silicon oil on acrylic are both zero.

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the viscosity ratio ranges from $\mathcal{M} \approx 250$ to 2500. The liquids are all wetting to glass, and the choice of liquid exhibits only a small difference on the receding and advancing contact angles: $\theta_r \in [0^\circ, 16^\circ], \theta_a \in [0^\circ, 23^\circ]$ (Fig. 4). Our experiments and analysis below show that, in this range, the contact angles are not leading-order parameters in the problem.

We understand the immiscible lock-exchange problem as a finite perturbation with respect to the miscible problem at the same value of $\mathcal{M}$. Assuming that the pinning length increases linearly with the strength of capillary forces relative to gravity, we suggest that $(h_s - h')/H \sim Bo^{-1}$, where $h_s/H$ is exclusively a function of $\mathcal{M}$. This scaling relation is confirmed experimentally (Fig. 5). The value of $h_s/H$ depends only weakly on the viscosity ratio $\mathcal{M}$ [12], taking values between 0.585 and 0.587 for the viscosity ratios corresponding to our experiments (Fig. 5, inset). The lower hinge approaches the tilting point of the miscible problem when capillarity is negligible relative to gravity ($h' \to h_s$ as $Bo^{-1} \to 0$) and is equal to zero when the balance between capillary and gravity forces exceeds a certain threshold $(h' = 0$ for $Bo^{-1} \gtrsim 0.14$). In this latter scenario, the entire interface is pinned by capillarity and does not tilt, regardless of the precise value of contact angle.

**B. Physics of capillary pinning**

The scaling of the hinge height, however, says little about the underlying reason why the hinge and the pinned interface exist. We next show that the mechanistic cause of the pinning behavior is capillary pressure hysteresis. In our immiscible gravity-exchange flows, air spreads along the top boundary of the cell (a drainage front) while the viscous liquid spreads along the bottom (an imbibition front). For the air to invade each pore throat, its pressure $P_{\text{nw}}$ (assumed atmospheric, $P_{\text{atm}}$) must, locally, exceed the pressure $P_w$ in the liquid by an amount larger than or equal to the drainage capillary entry pressure $P_{\text{cap}}$ [16]. Similarly, along the imbibition front, the liquid invades if $P_{\text{nw}} - P_w$ is less than the imbibition capillary pressure $P_{\text{imb}}$, which is always less than the drainage capillary pressure $2\rho g h_c$ (Fig. 6).

Although hysteresis in capillary pressure is sometimes caused by hysteresis in contact angle, such as for a raindrop pinned on a window, this is not the case here. In fluid flows through porous media, capillary pressure hysteresis is caused by the fundamental difference in the details of fluid invasion between drainage and imbibition: Invasion of nonwetting fluid produces strongly curved interfaces, whereas invasion of wetting fluid produces much flatter interfaces [16–20]. As a result, capillary pressure hysteresis is present even in the absence of contact angle hysteresis.

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1. Micromodel experiments

The presence of capillary pressure hysteresis in the absence of contact angle hysteresis is well illustrated by the gravity-exchange flow of air and silicone oil in a thin acrylic cell, etched to form a regular pattern of cylindrical posts (Fig. 6). This system serves as a porous medium analog in the sense of introducing microstructure, but takes away the complexity of a random medium and permits visualizing the flow at the pore level.

At the top of the pinned portion of the interface, the air-silicone oil interface is just at the threshold of air invading and displacing the silicone oil. For slow flows, in which pore-scale dynamic effects due to the intermittent pressurization of the viscous fluid are small [21,22], the pressure difference between the fluids is equal to the drainage capillary entry pressure, \( P_c^{dr} \), which, for zero contact angle (\( \theta = 0^\circ \)), is equal to \( 2\gamma/d \), where \( d \) is the distance between two neighboring posts (and, in our micromodel system, also equal to the diameter of the posts).

At the bottom of the pinned portion of the interface, the air-silicone oil interface adopts a “diagonal” configuration between posts 2 and 4 in Fig. 3(d)], which is stable as long as the air-silicone oil interface adopts a “diagonal” configuration [be-

### FIG. 7. (Color online) Interface configuration (solid blue) on the imbibition side of the exchange flow. We solve for the progression of the interface arcs as they advance through the pore by enforcing pressure continuity between posts 1, 2 and 3, 4 (i.e., \( r_1 = r_2 \)). The interfaces merge and advance to the next set of posts when the two interface arcs touch (i.e., \( \phi_1 + \phi_2 = 3/4\pi \)). The diameter of the posts is \( d \), and each post is one diameter away from its closest neighbors, the same as the design of the micromodel [Fig. 6(b)].

function of the contact angle \( \theta \) and the subtended angles (\( \phi_1 \) and \( \phi_2 \)):

\[
\begin{align*}
 r_1 &= \frac{[2 - \cos(\phi_1)] d}{\cos(\phi_1 + \theta)}/2, \\
 r_2 &= \frac{[2\sqrt{2} - \cos(\phi_2)] d}{\cos(\phi_2 + \theta)}/2.
\end{align*}
\]

In drainage, \( \theta = \theta_r \) and the drainage capillary entry pressure corresponds to the minimum radius of curvature \( r_1 \): \( r_{dr} = \min_{\phi_1, \phi_2} r_1(\phi_1; \theta_r) \).

In imbibition, \( \theta = \theta_a \), and a sequence of stable configurations can be obtained by equating \( r_1 = r_2 \). This leads to a one-parameter family of solutions shown in Fig. 6(d). The imbibition capillary pressure corresponds to the configuration for which the two interfaces touch, that is, \( r_{imb} = r_1(\phi_1; \theta_a) \) such that \( r_1(\phi_1; \theta_a) = r_2(\phi_2; \theta_a) \) and \( \phi_1 + \phi_2 = 3\pi/4 \).

In Fig. 8 we show the dependence of the dimensionless quantity \( d/r_{dr} - d/r_{imb} \) as a function of advancing contact

### FIG. 8. (Color online) Dependence of the dimensionless quantity \( d/r_{dr} - d/r_{imb} \) controlling the magnitude of the pinned interface in our micromodel system, as a function of advancing contact angle \( \theta_a \), assuming \( \theta_r = 0^\circ \).
angle $\theta_r$, assuming $\theta_r = 0^\circ$. This quantity exhibits only a small dependence on the contact angles.

IV. INTERFACE PINNING: DYNAMICS

A. Macroscopic sharp-interface model

To capture the dynamic macroscopic behavior of immiscible gravity-exchange flows, we propose a simple theoretical model based on the classical sharp-interface model [11,12]. The mathematical model is a partial differential equation for the height of the interface, which assumes a hydrostatic pressure distribution everywhere (known as vertical flow equilibrium, or VFE [23]) and Darcy flow to link flow velocity and pressure gradients. In a related context, the classical model has been modified to replace the sharp interface by a capillary transition zone [24–26]. Here we extend the model to include the height of the interface: $h_1 + h_2 = H$, which also represents the thickness of the porous layer everywhere, where $h_1 + h_2 = H$. By assuming hydrostatic pressure in both fluids, we can express the pressure distribution in the layer as

$$P = \begin{cases} P_1 - \rho g (z - h_2) & \text{for } z > h_2 \\ P_1 - P_e + (\rho + \Delta \rho) g (h_2 - z) & \text{for } z \leq h_2 \end{cases},$$

where $P_1$ is the unknown pressure at the interface and $g$ is the gravitational acceleration. By definition, the pressure difference across the interface between the nonwetting fluid and the wetting fluid is the capillary pressure $P_e$.

The volumetric flux per unit width of fluid phase $i$ is given by Darcy’s law $q_i = -k_i \partial P / \partial x$, where $k_i = k_{r i} / \mu_i$ is the mobility of the fluid phase and $k_{r i}$ is the relative permeability to that phase. Since we assume the two fluid phases to be completely segregated, the relative permeabilities are equal to one. The flow rate is given by the product of the thickness of the fluid phase and its volumetric flux, $Q_i = h_i q_i$. Hence, we obtain

$$Q_1 = -h_1 k_{r 1} \left( \frac{\partial P_1}{\partial x} - \rho \frac{\partial h_1}{\partial x} \right),$$

$$Q_2 = -h_2 k_{r 2} \left( \frac{\partial P_2}{\partial x} - (\rho + \Delta \rho) \frac{\partial h_2}{\partial x} - \frac{\partial P_2}{\partial x} \right).$$

Imposing global volume conservation ($Q_1 + Q_2 = 0$), we solve for $\partial P_1 / \partial x$ and substitute into Eq. (6) to obtain the flow rates of both fluids. Since $h_1 + h_2 = H$, we can express the flow rates in terms of only $h_2$, which also represents the height of the interface:

$$Q_2 = \frac{\Delta \rho g k_2}{\mu_2} (1 - f) h_2 \left( \frac{\partial h_2}{\partial x} - \frac{\partial P_2 / \Delta \rho g}{\partial x} \right),$$

$$f = \frac{h_2}{h_2 + \mathcal{M} (H - h_2)}$$

where $f$ is the fractional flow function and $\mathcal{M} = \mu_2 / \mu_1$ is the viscosity ratio. To obtain an equation for the evolution of the interface, we consider the conservation of volume of the dense fluid over region $\Delta x$ and time $\Delta t$ (Fig. 9). The change in volume of the dense fluid is given by

$$\Delta V_2 = \Delta h_2 \Delta x \phi = (Q_{2x+\Delta x} - Q_{2x}) \Delta t.$$  

Inserting Eq. (7) into Eq. (8) and taking limits for small $\Delta x$ and $\Delta t$, we get the partial differential equation for the evolution of the interface height $h_2$:

$$\frac{\partial h_2}{\partial t} - \kappa \frac{\partial}{\partial x} \left[ (1 - f) h_2 \left( \frac{\partial h_2}{\partial x} - \frac{\partial h_2}{\partial x} \right) \right] = 0,$$

where $\kappa = \Delta \rho g k / (\mu_2 \phi)$ is the characteristic buoyancy velocity and $h_2 = P_e / \Delta \rho g$ is the capillary height.

The function $g(h; \mathcal{M}) \equiv (1 - f (h; \mathcal{M})) h$ in Eq. (9) can be interpreted as a nonlinear diffusion coefficient. It is well behaved for all finite (even large) values of $\mathcal{M}$ (Fig. 10). In the

FIG. 9. (Color online) We develop a sharp interface model for the exchange flow between a buoyant, nonwetting fluid and a dense, wetting fluid, separated initially by a vertical interface. We assume Darcy flow within each fluid and hydrostatic pressure distribution everywhere. We then solve for the model by enforcing conservation of volume in a control volume (dashed blue). The resulting model is a partial differential equation for the height of the interface as a function of time.

FIG. 10. (Color online) The dimensionless nonlinear diffusion coefficient $g(h; \mathcal{M}) / H \equiv (1 - f (h; \mathcal{M})) h / H$ in Eq. (9) as a function of the dimensionless height of the interface $h / H$ for several values of viscosity ratio $\mathcal{M}$. 

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limit $\mathcal{M} \to \infty$, application of the L'Hôpital rule indicates that $g_\infty(h) \equiv \lim_{\mathcal{M} \to \infty} g(h; \mathcal{M}) = h$, which is the well-known limit for a viscous fluid slumping against a horizontal surface in an inviscid ambient fluid [13,27]. Our experiments, however, always correspond to a finite value of $\mathcal{M}$.

We take $P_c$ to be some characteristic drainage capillary pressure $P_c^{dr}$, where the nonwetting fluid displaces the wetting fluid, and some characteristic imbibition capillary pressure $P_c^{imb}$, where the wetting fluid displaces the nonwetting fluid. Across the pinned interface, $P_c$ jumps from $P_c^{dr}$ to $P_c^{imb}$. Since we have constant capillary pressures along the active drainage and imbibition fronts, $\partial h_c/\partial x = 0$. Therefore, $\partial h_c/\partial x \neq 0$ only along the pinned interface. In other words, it introduces a jump $[h_c]$ in the interface height of magnitude $(P_c^{dr} - P_c^{imb})/\Delta \rho$.

We solve Eq. (9) for the interface height $h$, subject to an initial condition that approximates a step function from $h = 0$ for $x < 0$ to $h = 1$ for $x > 0$, and no-flow boundary conditions ($\partial h/\partial x = 0$) at the ends of the cell. We solve the equation numerically using a centered finite volume method in space with forward Euler time integration.

The capillary pressure term vanishes everywhere except at the nodes that separate the portion of the interface that is in drainage from the portion that is in imbibition. Here the capillary pressure term contributes a finite jump in the flux across these two nodes, and this jump causes a sharp step in the height of the interface.

### B. Simulation of lock-exchange flows

The shape of the interface predicted by our model exhibits good agreement with the experiment (Fig. 1). We measure a priori all the parameters in the model (porosity, permeability, fluid densities, and viscosities) except for $[h_c]$, which we obtain by measuring the length of the pinned interface in the experiment.

Additionally, the model predicts quantitatively the time evolution of the nose position of both currents: the buoyant nonwetting current [Fig. 11(a)] and the dense wetting current [Fig. 11(b)]. It is encouraging that our mathematical description, which assumes horizontal flow, predicts accurately a lock exchange where clearly there is vertical flow since a portion of the interface remains pinned (and therefore vertical) at all times. Capillarity reduces the gravity-exchange flow rate (and as a result slows down the process), but the nonwetting and wetting currents still advance in a way that is well described by the $x \sim t^{1/2}$ scaling of the miscible lock exchange before they hit the cell boundaries (Fig. 11).

### C. Simulation of finite-release gravity currents

The impact of interface pinning effects due to capillary hysteresis is most pronounced when the initial interface is close to one of the lateral boundaries. In this case, one of the currents hits the boundary early and the process models a finite-volume release. The finite release of a miscible buoyant fluid spreads indefinitely [12]. In contrast, a finite volume of immiscible fluid spreads up to a finite distance at which the hydrostatic pressure difference that drives the flow is exactly balanced by the difference in capillary pressures at the drainage and imbibition fronts. In other words, capillary hysteresis stops the gravity current (Fig. 12).

### V. DISCUSSION: RELEVANCE TO CO2 SEQUESTRATION

The potential for capillary pinning to stop a spreading gravity current is particularly relevant in the context of CO2 sequestration, where CO2 is injected into aquifers saturated with denser and more viscous groundwater. During and after injection, the difference in density between the two fluids will drive a gravity-exchange flow [5–7], increasing the lateral extent of the CO2 plume and the risk that the CO2 will encounter a fault or fracture through which it could leak. Deep saline aquifers are naturally water-wet and, therefore, the hysteresis behavior will be similar [28–30]. The vertical-flow
equilibrium assumption is also appropriate in the field setting due to the very large aspect ratio $L/H$ of aquifers. Therefore, transfer of the results depends on the values of $Bo^{-1} = \gamma/(\Delta \rho g H d)$ in the field. The value of the interfacial tension $\gamma$ is relatively insensitive to depth and is ~ 0.025 N/m [31], and the density difference $\Delta \rho$ varies between 250 and 500 kg/m$^3$ for deep and shallow aquifers, respectively [32]. Large capillary effects occur in aquifers that are deep ($\Delta \rho \sim 250\text{kg/m}^3$), either thin or vertically disconnected by low-permeability shale layers ($H \sim 10\text{m}$), and have small grain sizes ($d \sim 10\mu\text{m}$). In contrast, small capillary effects correspond to aquifers that are shallow ($\Delta \rho \sim 500\text{kg/m}^3$) and thick ($H \sim 100\text{m}$) and have large grain sizes ($d \sim 50\mu\text{m}$). Thus, $Bo^{-1} \in [0.001, 0.1]$, a range that exhibits a large overlap with our experimental conditions (Fig. 5). Even though the shape of the CO$_2$ plume after the injection period does matter quantitatively [33], the fundamental qualitative impact of the capillary pinning phenomenon is already present in the lock-exchange flow configuration. This suggests that capillary pinning is possibly an important, yet unexplored, trapping mechanism during geologic CO$_2$ sequestration.

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