

PARTICLE MIGRATION IN A LINEAR SHEAR FLOW

by

EUGENE C. ECKSTEIN

B.S., Massachusetts Institute of Technology
(1970)

S.M., Massachusetts Institute of Technology
(1970)

SUBMITTED IN PARTIAL FULFILLMENT
OF THE REQUIREMENTS FOR THE
DEGREE OF

DOCTOR OF PHILOSOPHY

at the

MASSACHUSETTS INSTITUTE OF TECHNOLOGY

February 1975

Signature of Author.
Department of Mechanical Engineering, February 11, 1974

Certified by
.....

Accepted by
Chairman, Department Committee on Graduate Students



PARTICLE MIGRATION IN A LINEAR SHEAR FLOW

by

EUGENE C. ECKSTEIN

Submitted to the Department of Mechanical Engineering
on February 11, 1975 in partial fulfillment of the
requirements for the Degree of Doctor of Philosophy/
Master of Science.

ABSTRACT

The particles in a laminar slurry flow disperse through several types of migrational motions of the particles. The velocity shear field causes passing interactions among the particles and thereby drives one type of migrational motion. This type of migrational motion is stochastic in nature and can be described by a diffusion coefficient. Mass transfer characteristics of slurry flows are strongly affected by these diffusive motions.

Experimental measurements of the diffusion coefficient, D , for shear flows of spherical-particle slurries with volumetric concentrations of 20 to 50% are reported in this thesis. To minimize the other migrational motions, the experiments were conducted in a low-Reynolds-number (10^{-4} to 10^{-2}), linear shear flow established by a circular-cylinder Couette apparatus. To ascertain the correct form of the non-dimensional diffusion coefficient, $D/a^2\omega$, measurements were made using slurries with different average particle radii, a , and for three shear rates, ω . Using the ergodic hypothesis, diffusion coefficients were calculated from observations of the diffusive motion of a single labelled particle as it interacted with the other unlabelled particles of the slurry. Observations of the labelled particle were made without disturbing the diffusive motion by means of experimental apparatus which measured the rate of gamma-ray emission of a radioisotope affixed to the labelled particle. Construction of a suitable radioactively labelled particle posed significant difficulties. Two labelled particles were constructed; only one of these was wholly suitable.

Only a weak variation of the non-dimensional diffusion coefficient was observed over the range of volumetric concentration of 20 to 50%. Although the experiment was so difficult as to preclude high accuracy, the non-dimensional diffusion coefficient was established, to within a factor of two, at a value of $D/a^2\omega \approx 0.03$, over the concentration range mentioned.

Thesis Supervisor: Ascher H. Shapiro

Title: Ford Professor of Engineering

ACKNOWLEDGEMENTS

This investigation was supported from two sources: (1) a Public Health Service Grant 5P01H14322-03 from the National Heart and Lung Institute to the Harvard-MIT Program in Health Science and Technology; and (2) Grant GK-31183 from the National Science Foundation (Engineering Mechanics Program).

Many people have provided me with help and guidance during the course of this thesis. My advisor, Professor Ascher Shapiro gave his constant support and insight throughout the work. His encouragement, particularly during times of experimental difficulty, made the project much easier. Dr. William Hyman was my constant working companion during the first two and one-half years. Together, we performed the preliminary experiments described in Appendix II. Professor Mikic and Professor Colton, members of my thesis committee, served as a thoughtful and responsive sounding board.

It would have been impossible for me to assemble an experimental apparatus of this complexity without the expert help of the various MIT shops and laboratories. David Palmer, Richard Fenner and Dominic DiMartino were especially helpful during the construction of the apparatus.

Marjorie Joss patiently deciphered and typed both the drafts and final copies of the thesis.

My family, especially my wife Jane, has provided the very necessary encouragement to tackle and complete this work.

TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT	2
ACKNOWLEDGEMENTS	3
TABLE OF SYMBOLS	9
CHAPTER 1 - INTRODUCTION	11
1.1 - Definition and Characteristics of Slurry Flows	11
1.2 - Migrational Motion in the Lateral Direction	12
1.3 - Dimensional Analysis and Scaling	16
1.4 - Examples of Slurry Flows and the Effects of Migration of the Particulate Component	25
1.5 - Description of Mutually-Induced Migrations	30
1.6 - Scope of this Work	38
CHAPTER 2 - RELATED WORK BY OTHER INVESTIGATORS	40
2.1 - Particle Motion in Shear Flows	40
2.2 - Experiments on the Motions of Particles in a Slurry Flow	43
.1 - Couette Flows	43
.2 - Concentration and Velocity Profiles in Couette Flows	48
2.3 - Particle Motion in Poiseuille Flow	52
2.4 - Augmented Diffusion in Blood Flows	55
CHAPTER 3 - REQUIREMENTS FOR THE DESCRIPTION OF THE SLURRY AND THE SHEAR FLOW APPARATUS	61

TABLE OF CONTENTS (CON'T.)

	<u>Page</u>
3.1 - Use of the Dimensional Analysis to Establish the Requirements of the Slurry and the Experimental Apparatus	61
3.2 - Couette Flows	65
.1 - Linear-Belt Couette Apparatuses	66
.2 - Circular-Cylinder Couette Apparatuses	68
3.3 - Slurries	73
CHAPTER 4 - TECHNIQUES AND APPARATUS USED TO MEASURE THE DIFFUSIVE MOTION	76
4.1 - Techniques for the Measurement of Diffusion Coefficients	76
.1 - Multiple-Particle Techniques	76
.2 - Single-Particle Technique	78
4.2 - Methods of Application of the Single- Particle Technique in a Circular Cylinder Couette Apparatus	81
4.3 - Technical Requirements of the Single- Particle Technique	83
4.4 - Bench-Top Tests of Three Models of Labelled Particle and Corresponding Detector	85
.1 - Light-Emitting Particles	86
.2 - The Radioactively-Labelled Particle	90

TABLE OF CONTENTS (CON'T.)

	<u>Page</u>
4.5 - Description of the Apparatus to Measure Lateral Position	93
4.6 - Component Parts of the Apparatus to Measure Lateral Position	96
.1 - Radioactively-Labelled Particles	97
.2 - Collimating-Slit Viewing Windows	101
.3 - Radiation Detection Equipment	104
.4 - Coordinating Electronics	105
4.7 - Operating Considerations and Accuracy of the Detector	111
.1 - Determination of the Threshold Value	114
.2 - Alignment of the Axes of the Couette Apparatus and the Viewing Windows	117
.3 - Accuracy and Calibration of the Complete Detector System	119
CHAPTER 5 - PROCEDURES USED TO COLLECT AND REDUCE THE DATA	122
5.1 - Procedures Used to Collect the Data	122
.1 - Initial Preparation	122
.2 - Order of Events During an Experimental Run	126

TABLE OF CONTENTS (CON'T.)

	<u>Page</u>
5.2 - Calculation of the Diffusion Coefficient	128
.1 - Outline of the Computer Program	130
CHAPTER 6 - RESULTS AND DISCUSSION	138
6.1 - Experimental Results	138
6.2 - Summary of Error Analysis	139
.1 - Random Errors	140
.2 - Systematic Errors	144
.3 - Results of Computational Methods of Lesser Accuracy	148
6.3 - Summary	149
CHAPTER 7 - INVESTIGATIONS OF THE LATERAL CONCENTRATION DISTRIBUTION AND ITS RELATIONSHIP TO THE DETECTIONAL PROCEDURE	151
7.1 - Detection Procedures for Making Observations of Marked Particles	152
7.2 - Numerical Experiments	156
7.3 - Detection Processes in the Numerical Experiments	160
7.4 - Values of Parameters in the Numerical Experiments	160
7.5 - Results of Numerical Experiments	163
CHAPTER 8 - CONCLUSIONS	166

TABLE OF CONTENTS (CON'T.)

	<u>Page</u>
REFERENCES	167
CAPTIONS FOR FIGURES	171
FIGURES	172
APPENDIX I -- PROCEDURE FOR PLATING ^{58}Co ONTO COPPER- CLAD ABS PARTICLES	227
APPENDIX II -- TWO-DIMENSIONAL PARTICLE MIGRATION IN A LINEAR SHEAR FLOW	236
APPENDIX III -- CONSIDERATION OF ERROR	245
A3.1 - Systematic Errors	245
A3.2 - Random Errors	247
APPENDIX IV -- DIFFUSION COEFFICIENTS FOR SPHERICAL- PARTICLE SLURRIES OF LOW CONCENTRATION AND FOR DISCOID-PARTICLE SLURRIES	263
BIOGRAPHICAL SKETCH	264

TABLE OF SYMBOLS

a	particle radius
A	scaling coefficient
b	particle radius in Figures 2.3 and 2.5
B	scaling coefficient
c, c_0	area concentration of particles
D	diffusion coefficient
E	expectation
f	subscript denoting fluid
g	acceleration
i	subscript denoting order in a list
ℓ	diffusive length
m	mean of a sample
N	number of items
R	Couette-cylinder radius
s	subscript denoting solid
t	time
u	velocity parallel to the channel walls
V	variance, second moment of a distribution
w	channel width
x	direction parallel to the channel walls
y	direction perpendicular to the channel wall
δ	only as δt , time step in numerical experiments

Δ	difference between two quantities
χ	$\Delta Y/\sqrt{\Delta t}$, diffusive jump
ϕ	volumetric concentration
μ	fluid viscosity
μ_2	second moment of the infinite underlying distribution
ρ	mass density
ω	shear rate, $\partial u/\partial y$
$\wedge$	superscript denoting underlying infinite distribution

1. INTRODUCTION

1.1 Definition and Characteristics of Slurry Flows

Within this thesis the term "slurry" signifies a concentrated suspension of solid particles in a Newtonian liquid. Common usage implies that the average size of the particulate component of a slurry is small compared to a characteristic dimension of the flow. This size restriction allows the slurry to be treated for certain purposes as a quasi-homogeneous mixture with bulk properties such as local concentration. The bulk properties of the slurry are determined by the material properties of the components; these include the density and viscosity of the suspending fluid as well as the density, shape, and size distribution of the particles of the solid component. The volumetric concentration of the solid particles also influences importantly the other bulk properties of the slurry; for example, the apparent viscosity of a slurry increases by more than an order of magnitude as the concentration is increased from 30 to 40 percent.

Within a slurry flow there are interactions both among the solid particles and between the solid particles and the fluid which surrounds them. These interactions are the cause of flow phenomena which occur in slurries but do not occur in analogous laminar flows of Newtonian fluids. The dependence of the apparent viscosity upon concentration is an example of such phenomena on a scale of size of

the characteristic dimension of the flow. The term microrheology has been applied to detailed studies of fluid-solid interactions for the purpose of better understanding the macroscopic rheology of slurries [1].

Another example of such interactive phenomena is the irregular migrational motion of the individual particles of the solid component of the slurry which produces a lateral dispersion, or diffusion. Several different types of interactions produce such lateral migrational motions. In general, there is an interrelationship between the apparent viscosity and the migrational motion since migrations alter concentration profiles and thereby cause changes in apparent viscosity, while regions of higher or lower apparent viscosity influence the flow velocity profiles and thus the migrational motion. Simple examples of these concepts are discussed in sections 1.2 and 1.4.

1.2 Migrational Motion in the Lateral Direction

The several principal sources of those migrational motions that produce lateral dispersion are discussed below. For concreteness, they are discussed in the context of pipe flows, since this is a common method for transporting solid materials in suspension from a reservoir of uniform concentration to another location. In such pipe flows the particles tend to redistribute non-uniformly over the cross-section of the pipe. The region near the wall becomes depleted and the concentration in the axial core increases. At high concentrations the velocity profile tends to be flat in the core, with most of the

shear occurring in the depleted wall regions. There are three sources of the migrational motions responsible for the redistribution:

1) Mutually-Induced Velocity Fields. These produce a lateral irregular migration of the particles in a shear flow. The mutually-induced velocity fields are the result of two types of fluid and particle motions caused by the velocity gradient.

Because of the velocity gradient, each particle acquires a rotational motion, and this in turn tends to produce rotational flow in the nearby fluid.

Further, each particle also has a convective velocity, approximately the average of that of the fluid it replaces. Since the particles of a slurry are closely spaced laterally, particles must pass another if there is a velocity gradient. During a passing event the particles are transiently displaced laterally.

The actual motion of any one particle in a slurry is the sum of the undisturbed motion it would have if there were no other particles, plus the rotational and convective motions induced on the particle by its neighboring particles. The transient nature of the passing events and the random spacing between particles virtually preclude a deterministic description of the motion of an individual particle.

The phenomena described here are treated in more detail in Section 1.4. Note that, unlike lift forces and body forces (see below), the migrations induced by shear in the flow are stochastic in nature.

ii) Lifting Forces. Lateral motions are also caused by laterally-directed lift forces that may, in certain circumstances, act on a particle.

In inertia-free flows at zero Reynolds Number, rigid particles in the vicinity of a wall do not suffer lift forces, as may be seen by the principle of kinematic reversibility (see section 2.1). With deformable particles, however, lift forces exist in both the inertia-free and the inertial regime [2].

In inertial flows near walls, a lift force on a single rigid particle exists because of the asymmetry of the flow field [3,4]. Other lift forces originate by reason of the asymmetry that exists when the particle is in a flow where the velocity profile has both shear and curvature [3,4]. Thus, we see that laterally-directed lift forces occur for a single rigid particles in the inertial range of Reynolds Numbers and at any Reynolds Number for a single deformable particles. Table 1.1 summarizes the many situations in which these lift forces occur. It is reasonable to assume that similar lift forces will exist for slurry flows, i.e. for many particles.

iii) Body Forces. Lateral migrations may obviously be developed, in the case of a non-neutrally-buoyant slurry, by gravity or by centrifugal forces associated with curvature of the fluid streamlines.

Table 1.1. Occurrence of Lift Forces on Particles

The marked entries in the table denote conditions under which a laterally-directed lift force exists. The basic flow is an inertia-free, infinite linear shear flow with a single rigid particle; the basic flow generates no lift force on the particle. Entries in the table show the occurrence of a lift force for alterations of the basic flow. The alterations are denoted by the headings for the rows and columns of the entries. For example, using the entry in the second row and the third column, a lift force occurs for a deformable particle in an inertia-free, linear shear flow in the vicinity of a wall.

Basic Flow with $\rightarrow$ and with $\downarrow$	Curvature	Wall	Particle Deformability	Inertia
Curvature			x	x
Wall			x	x
Particle Deformability	x	x		x
Inertia	x	x	x	x

Relationship of Migrational Motions to Depleted Regions Near

Walls. A qualitative explanation of the origin of depleted regions in pipe flows of slurries, or the "plasmatic-zone" in blood flow, would be that the net lifting force and larger average lateral migration found near the wall in the zone of higher shear rate causes a redistribution of particles to the core. The lower shear rate in the region near the axis would, on the other hand, reduce the amount of lateral migration within and out of the core region. Such a mechanism is obviously concentration-limited by the maximum packing attainable in the core. The increased concentration in the core also produces

an increase in apparent viscosity there, which concomitantly reduces the shear rate necessary for a force balance. Thus lateral migration in the axial core is further reduced as compared to a Poiseuille velocity profile.

It should be stressed that the explanation given here is speculative. The lifting forces have been observed for single isolated particles, but have not been studied for many, closely-spaced particles. Experiments by other investigators demonstrate such lifting forces (see Chapter 2). More work needs to be done on the theoretical description of the forces and on the effect of multiple particles on the forces.

1.3 Dimensional Analysis and Scaling

In view of the great difficulty in theoretically describing the migrational events in a multi-particle slurry, dimensional analysis provides a rational means of structuring an experimental investigation: it reduces the number of parameters, suggests the scaling laws, helps toward an understanding of the relative roles of different parameters, and is all-important in organizing and interpreting the results.

Based on a consideration of the fluid dynamical events which occur when a cloud of rigid particles is convected through a channel, we may set down a list of the parameters that control the phenomena:

<u>Symbol</u>	<u>Parameter</u>	
a	particle radius	L
ω	first derivative of the velocity, shear rate	T^{-1}
$\partial^2 u / \partial y^2$	curvature of the velocity profile	$L^{-1} T^{-1}$
ϕ	average columetric concentration	L^3 / L^3 (no units)
μ	viscosity of the fluid	$M L^{-1} T^{-1}$
ρ_f	mass density of the fluid	$M L^{-3}$
w	characteristic width of the channel flow	L
y	distance from the channel wall	L
$g(\rho_s - \rho_f)a^3$	"reduced" weight (or centrifugal force) when g is the acceleration of gravity (or the centrifugal acceleration) and ρ_s is the mass density of the particle	$M L T^{-2}$

For a given flow geometry (e.g. the radius of a pipe flow or the channel width of a two-dimensional, cylindrical Couette flow) the foregoing independent parameters determine all events in the flow, including the magnitude of the diffusion coefficient for lateral migrational motions:

D	diffusion coefficient for lateral migrational motions	$L^2 T^{-1}$
-----	--	--------------

Accordingly, we may write the implicit physical relationship

$$D = f(a, \omega, \partial^2 u / \partial y^2, \phi, \mu, \rho_f, w, y, g(\rho_s - \rho_f)a^3) \quad (1.1)$$

By straightforward application of the Pi Theorem, the following dimensionless relationship can be obtained, involving three fewer parameters:

$$\frac{D}{a^2 \omega} = f\left(\phi, \frac{a}{w}, \frac{a}{y}, \frac{\rho_f a^2 \omega}{\mu}, \frac{a \partial^2 u / \partial y^2}{\omega}, \frac{g(\rho_s - \rho_f) a}{\mu \omega}\right) \quad (1.2)$$

Several of the groups on the right-hand side have simple physical significance, and consideration of these helps in our understanding of the problem.

Ratio of Particle Radius to Channel Width, a/w . There are two natural lengths which occur in the list of parameters, namely a and w . Obviously one non-dimensional group is the ratio of these two lengths, which is an index of the relative importance of wall effects.

When $a/w \ll 1$, the wall effects are confined to a small zone near the wall, to a boundary zone. Specifically, for very low concentration ϕ , small a/w produces the classical channel flow of a Newtonian fluid. At very high concentrations, a plug flow will exist in the core of the channel, with migrational phenomena restricted to thin regions near the walls.

When a/w approaches 1, the flow can no longer be considered a slurry since major regions of the flow are occupied by individual particles, thereby preventing establishment of the usual characteristics of a channel flow such as a velocity profile.

Examples of the effect of the ratio of a/w in a cylindrical Couette apparatus are shown in Figure 2.5 and discussed in Section 2.2.2.

Ratio of the Particle Radius to the Distance from the Channel Wall, a/y . The walls of a channel produce effects on the particle motions in a slurry flow by reason of the boundary conditions requiring that the normal and tangential velocities be zero there. Thus, from the viewpoint of defining quasi-homogeneous properties of a slurry, the wall regions may produce anomalous values of two types. One type of anomalous value is purely geometric; an example is the reduced concentration near the wall which occurs near the walls. The reduction occurs since some particle positions are excluded as they would require the particle to penetrate the wall. The second type of anomalous value is of a fluid mechanical origin. In the central region of a slurry flow each particle is surrounded by many other particles, while in the wall region many or all of the particles on one side are replaced by the wall. Thus, there is a fundamental asymmetry near the wall. This asymmetry and the geometric reduction of the concentration produce a higher wall shear rate in a channel flow of a slurry than in a corresponding flow of a Newton fluid. The ratio of a/y is a measure of the asymmetry; as the ratio becomes small, the wall effects will have a lesser part in the fluid mechanics of the slurry flow. Further discussion of this ratio and a review of pertinent experimental results by Karnis are contained in section 2.2.2.

Reynolds Number, $\rho_f a^2 \omega / \mu_f$. The Reynolds number appropriate to the properties of the slurry is based upon the particle size rather than the channel width. Physically, the appropriate Reynolds number is a measure of the ratio of the inertial stresses to the viscous stresses in a reference frame moving with the particle.

An appreciation of the quantities used to form the Reynolds number is best obtained by first considering a single spherical particle in a linear shear flow. As shown in Figure 1.1, the particle speed is nearly the same as the undisturbed fluid at the center of the particle. This must be so if there are no other forces, since in steady flow the drag caused by the "slip" velocity will tend to accelerate or decelerate the particle and, thus, eliminate the "slip" velocity. Thus, in this frame of reference the particle appears to be sitting in the center of the shear field. The characteristic velocity of the flow is the fluid velocity at the surface of the particle which is given by the product of the radius a , and the shear rate ω . Other characteristic parameters are the particle radius and the fluid viscosity μ_f and density ρ_f .

The effects of the other particles in a slurry or of distance to the channel wall are added to this otherwise over-simplified picture through the entry of the relevant parameters, ϕ , and y , on to the list of independent parameters of the dimensional analysis.

For the purposes of determining migrational motions, the particle Reynolds Number is an index of the time necessary for the particle to achieve the local fluid velocity after it has been

laterally displaced. Alternatively stated, the Reynolds number is an index of the time for the slip velocity to become negligible. For $\rho_f a^2 \omega / \mu \ll 1$ the particle very quickly achieves the local fluid velocity of its new region. For $\rho_f a^2 \omega / \mu \gg 1$ a relatively long time (or, equivalently, in a channel flow, a long distance) will be required before the particle reaches the local fluid velocity. This effect is augmented in shear flows with curvature since in these flows adaptation to local fluid conditions requires a change of rotational velocity as well as a change of translational velocity. In slurry flows the other independent parameters, especially ϕ and y , will play a part in describing the time for the slip velocity to become negligible.

Ratio of the Curvature of the Velocity Profile to the Particle Size, $a(\partial^2 u / \partial y^2) / \omega$. This non-dimensional group provides a measure of the normalized radius of curvature of the velocity profile, $(\partial^2 u / \partial y^2) / \omega$, to the particle radius. It may also be viewed as the fractional change in the shear rate over a distance of one particle radius

$$\frac{a \frac{\partial^2 u}{\partial y^2}}{\omega} = \frac{a \frac{\partial}{\partial y} \left(\frac{\partial u}{\partial y} \right)}{\frac{\partial u}{\partial y}} \approx \frac{a \frac{\Delta \left(\frac{\partial u}{\partial y} \right)}{\Delta y}}{\frac{\partial u}{\partial y}} \quad (1.3)$$

For values of $(a \partial^2 u / \partial y^2) / \omega \ll 1$ the velocity profile may be treated as virtually linear.

A lateral force which depends upon the curvature of the velocity profile is posited to explain the stable position of individual particles at a particular radius in a pipe flow of a dilute suspension (Segré-Silberberg effect). The ratio of this "curvature-dependent" force to other lateral forces would presumably increase monotonically with the magnitude of $(a \partial^2 u / \partial y^2) / \omega$.

Ratio of Reduced Particle Weight to a Viscous Drag Force,
 $g(\rho_s - \rho_f)a / \mu\omega$. Since slurry flows generally have a small particle Reynolds number, a quasi-static force balance prevails between the "reduced weight" of a particle and the viscous drag force associated with the "settling velocity", that is, the component of slip velocity in the direction of the "reduced weight". The "reduced" particle weight is the gravitational or centrifugal force acting on the particle less the buoyant force exerted by the pressure distribution in the fluid on the particle. It is the net force which is balanced by the viscous drag, and forms the numerator of the dimensionless parameter. The denominator, $\mu \cdot \omega \cdot a^2$ has the dimensional form of a viscous drag force, i.e., it is the product of an absolute viscosity, a velocity gradient, and an area. Thus the ratio reflects directly the relative magnitudes of the two forces.

A more physically meaningful interpretation derives from observing that the Stokes settling velocity of a single particle in an infinite sea of fluid is proportional to $g(\rho_s - \rho_f)a^2 / \mu$. The ratio of this settling velocity to ωa (which characterizes the

velocities induced on a particle in a shear flow) is precisely the dimensionless group above:

$$\frac{v_{\text{settling}}}{\omega a} \sim \frac{g(\rho_s - \rho_f)a}{\mu_f \omega} \quad (1.4)$$

This parameter shows that in order to reduce buoyancy or centrifugal-force effects to a negligible level, a larger mismatch between particle and fluid densities may be tolerated with small particles than with large particles.

From the viewpoint of the diffusional motion, this ratio is an index of the relative importance of gravitational or centrifugal settling compared with diffusional spreading. To illustrate, consider a cloud of non-neutrally buoyant particles in a cylindrical Couette apparatus with vertical axis. The particles settle in the direction of the vorticity associated with the shear flow. In a stationary fluid the particles settle as a well-defined group. But in the shear field the cloud spreads diffusively. Small values of the dimensionless parameter indicate that the diffusive spreading will be relatively more important than settling effects.

Care must be taken in the interpretation of this parameter for cases where the settling motion of particles is not parallel to the vorticity; the settling motions then generate a "slip" velocity over and above the shear field, and thereby cause lateral migrational motions. Brenner [5] provides an excellent review of this type of migrational motion.

Remarks on the Diffusion Coefficient. The presumed dimensional representation of the migrational motion in the form of a diffusion coefficient is somewhat arbitrary. It cannot be wholly rationalized for the case of pipe flows. An alternative representation would be to use a drift velocity on the basis that lifting forces, which are balanced by a Stokes drag, cause a part of the motion. Since, as will be seen later, we will limit the investigation to symmetrical linear shear flows far from the walls, in which case lifting forces are absent, we have formulated the problem in terms of a diffusion coefficient such as those of Fick's Law.

Lifting forces could be included in a diffusional representation by varying the diffusion coefficient with channel position. Such descriptions of a force as a gradient of the diffusion coefficient are used in the random-walk models of diffusion of atoms in crystal lattice [6]. However, the correct representation also includes the concentration gradients near the walls, since these gradients also provide a laterally directed "driving" force. All the necessary parameters for such a description are provided in the list of independent parameters; however, either a theoretical or experimental attack upon such a description would be very difficult.

Since the list of parameters does not include a distance from the origin of the channel the dimensionless groups can not describe any entry phenomena. To describe these entry phenomena the list of independent parameters would have to include the length from the beginning of the channel. Another length ratio, the particle

radius divided by the entry length, would appear among the non-dimensional groups which characterize the diffusion coefficient.

For the migrational motion in a cylindrical Couette flow, the classical diffusion coefficient seems to be, by symmetry considerations, the correct formulation for the case of small a/w , i.e., for small wall effects. Here the central region of the flow appears as a linear shear flow, which has no characteristic lateral direction. Thus the migrational motions are statistically haphazard, like those of a diffusing molecule in a gas.

In the case when a/w is not small, D will be a function of position within the Couette channel. These points are further treated in Section 1.5 and Section 7.2.

1.4 Examples of Slurry Flows and the Effects of Migration of the Particulate Component

Industrial Examples. Industrial applications for slurry flow are commonplace since slurries offer an economical way of transporting large volumes of solid particulate materials. Most processes use water or air as a suspending fluid because of the low cost and availability. The transportation of coal-water slurries through pipelines from the mining fields to gas or electricity generating plants may be significantly increased in the future as a result of the current high costs of oil. Other water-base slurries are found in the processes of ore mining and separation and in the pulp-

handling sections of processes used to manufacture paper products. A common method of industrial handling of plastics is to suspend the plastic beads or rods in air and move them through ducts as an air-borne suspension. It should be noted that although most industrial slurry flows have large pipe Reynolds numbers their particle Reynolds number are much smaller. For example under typical operating conditions the particle Reynolds number of a coal-water slurry pipeline, which have pipe Reynolds numbers of 10^6 or greater, is less than 10.

A continual problem in industrial use of slurry pipelines is the settling of the solid species such as occurs, for example, to coal particles between pumping stations. Also, should the pipeline be shut down before all the slurry is displaced from the pipeline by water or air, the solid component settles out and poses a major problem when the pipeline is restarted. A better understanding of the interparticle forces and of the migrational motions might provide solutions or allow alleviation of these problems. It is expected that such understanding will at least in part lead to replacement of the empirical design techniques which are, of necessity, now used.

A second application for knowledge of particle dynamics in a slurry flow would be in the design of separation techniques for different components of a slurry. Such separations might be based upon differences in particle behavior caused by differences in particle properties such as density, shape, or deformability.

Blood as a Slurry. Blood is a most interesting example of biological slurry. From the viewpoint of constituent components blood can be considered as a Newtonian fluid for flows with shear rates over 100 s^{-1} . In this simplified slurry the plasma supports a high concentration of red blood cells (RBC's) and a much smaller number of other cells and cell fragments, i.e. the various types of white blood cells and the platelets. In normal, healthy persons the concentration of RBCs, expressed as a volume fraction, is in the range of 0.35 to 0.45. The plasma is a water-base solution of salts, carbohydrates, lipids, and proteins.

Within the living organism blood serves a remarkable function in distributing metabolites and oxygen and in removing catabolites, carbon dioxide, and heat. In most of its functions blood is convected in bulk to a site where it delivers the relevant constituent component through diffusion as illustrated by the transport of oxygen from the lungs to the cells of the body. Through the chemical reactions of coagulation, which involve the proteins of the plasma and the platelets, blood is able to repair leaks in the circulatory system. The delivery of the clotting constituents such as fibrinogen or platelets to a thrombus are also examples of the use of bulk convection and final diffusive delivery.

Several complications are of course present in blood which are absent in most industrial slurries, as described in several excellent articles that deal with rheological effects in blood [7,8,9,10,11]. The cellular formed elements are flexible and easily

deformed, thereby causing effects not seen with solid particulates in slurries. The protein linkage between cells at low shear rates produces a non-Newtonian rheological behavior.

Despite the differences cited, it is hoped that there is enough similarity flows of solid particulates and flows of the cellular particles of the blood for the results to be of some significance in establishing the orders of magnitude of the self-induced diffusional fluxes of the formed elements of the blood.

Effects of Migration in Blood Flows. During extracorporeal blood flow, red blood cells (RBC's) are damaged at a greater rate than that which normally occurs in the circulation. This results in either direct or delayed hemolysis and, unless anticoagulents are used, initiates the physiologic clotting system. Clearly, the nature of the artificial surfaces in contact with the blood is essential. However, the rate of arrival of RBCs to the artificial surface is also important. To be significant in evaluating hemolysis caused by flow-induced contact between RBCs and an artificial surface, the rate of arrival of RBCs must be broken into categories such as the number of repeat arrivals at the surface by a RBC or according to the distance from which a RBC can travel to touch the surface. Observations of the lateral migrations of RBCs in tube flows of ghost cell suspensions (RBC's which have been drained of hemoglobin) have been made but a categorical classification of RBC behavior is not yet possible. A knowledge of the dynamics of RBC motion, it

is hoped, would make it possible to introduce the fluid mechanical aspects of hemolysis tests of artificial surfaces as a more explicit basis for the design of flow configurations for prosthetic devices.

The formation of platelet thrombi is partially dependent upon the delivery rate of platelets to the site of formation. (Platelets are present in blood at a one-tenth fraction of the number density of RBC's and are of smaller size, approximately 1-2 microns, as compared to the 8-micron major diameter of the RBC.) Lateral motion of the platelets can be assumed to be controlled by the rotation and migration of the more dominant RBC's. The occurrence of platelet thrombi presents a hazard because they may multiply greatly in size as a consequence of the strong triggering action of activated platelets upon the intrinsic clotting system. Fragments of platelet thrombi can have the same effect at distant locations. The basic problem is exceedingly complex because of the biochemical reactions involved. However the mechanical delivery-rate of the platelets to the thrombus for the clotting reaction may significantly affect reaction rates. Several papers dealing with this phenomenon are cited in Chapter 2.

Facilitated Diffusion. The simplest method of increasing the rate of mass transfer is to mix the unequilibrated species thereby increasing the concentration gradients. In a slurry which is used as a part of a transfer process the migration and rotation of the solid component achieves this mixing on a small scale.

Such increases produce what is usually termed augmented or facilitated diffusion since the mass transfer, from a bulk viewpoint, can be modelled with a diffusion coefficient larger than in a stationary system. Various methods of achieving increased transfer rates are used in fluidized beds, blood oxygenators, and kidney dialysis machines. The facilitated diffusion can be especially large when the solid component has a sink (or source) behavior. In the case of blood oxygenation the lateral motion of the RBC's acts, as it were, as a bucket brigade from the oxygen-rich to oxygen-poor areas. Chapter 2 contains a discussion of two blood oxygenators which achieve greater oxygen transfer per area of gas-exchange surface by using the RBC migrations.

1.5 Description of Mutually-Induced Migrations.

This thesis as discussed more fully later, is restricted to an experimental study of the diffusive lateral migrations which result from particle interactions in a linear shear flow. Furthermore, the experimental measurements have been restricted to regions of the shear field far from bounding walls where the concentration of the slurry is uniform. The lateral migration in this case is fundamentally different from that caused by lifting or body forces first, because it is stochastic in nature; and second, because it occurs by reason of interactions between the fluid and the particles. In contrast, migrations caused by lifting or body forces will have first, a

characteristic direction and magnitude; and second, occur by reason of interaction between particles, the fluid and either the boundaries, the curvature of the velocity profile, or the unbalanced buoyancy. The migration we are concerned with is a type of self diffusion in that it occurs within an infinite shear field of uniform concentration. Qualitative discussions of the fluid mechanical mechanisms which produce such a migration are given below.

Preliminary Considerations. Before considering how the lateral migration of an individual particle occurs it is helpful to examine the simple events experienced by spherical, rigid particles suspended in a linear shear flow containing a small number of particles. Furthermore, as there is a small number of particles they interact rarely and do not appreciably alter the flow. Since they come near each other only very rarely, the typical "collision" or interacting event occurs between two particles.

1) Each particle translates with a speed equal to the average speed of the fluid replaced by the particle. There is no slip velocity, at least when the two particles under consideration are not close together during a passing "collision". Parenthetically, we note that although it is convenient to describe an interaction event as a "collision" the particles do not really come into contact; there is always a layer of fluid separating them.

1i) Each particle rotates since the shear establishes a velocity gradient relative to the particle. Jeffries [12] has shown

that a single sphere in an infinite shear flow, and at zero particle Reynolds number, rotates at half the shear rate of the fluid.

iii) The rotational deviation from the infinite shear field produced by the presence of an individual particle decays as the inverse square of the distance from the center of the particle, i.e. $(a/R)^2$, so for low concentrations the interaction between particles is relatively weak. Table 1.2 gives the inter-particle spacing for various concentrations of particles arranged in a crystalline cubic array. At each concentration, the center-to-center distance is given as a multiple of the particle radius. The surface-to-surface distance, or the interparticle spacing, is given in the same fashion. At a concentration greater than a few percent, because of the close spacing of the particles, one would expect significant interaction of the rotational velocity fields of the particles. At such concentrations the interactions produce disturbances in the otherwise steady and uniform shear flow that resemble the effects of turbulence, hence the velocity gradient must be considered as an average quantity which characterizes the flow.

iv) The particles in a shear field pass each other, since they have different convective velocities. When the lateral spacing between two particles is relatively small the particles experience lateral motions, inasmuch as they tend to follow the streamlines of the disturbed flow relative to each particle. Put differently, the lateral motions allow the faster particle to pass the slower particle. Goldsmith, Mason, and co-workers [1] have performed experiments in

Table 1.2

Distances between centers or surfaces of spherical particles when the particles are arranged in a crystalline, cubic lattice at various concentrations.

<u>Volumetric concentration</u>	<u>Center-to-center distance of particles</u>	<u>Surface-to-surface distance of particles</u>
(a is the radius of the particles)		
3%	5a	3.0a
10%	3.5a	1.5a
20%	2.8a	0.8a
30%	2.4a	0.4a
40%	2.2a	0.2a
50%	2.03a	0.03a

shear flows showing the nature of these interactions. Figure 1.2, taken from their paper, illustrates an interaction between two particles in a shear flow of low particle Reynolds number. The coordinate frame used in the illustration has the average velocity of the particles. At the beginning of the interaction the faster particle comes to near contact with the slower particle and the two particles appear to form a doublet. This doublet rotates about the point of apparent particle contact to a location which is equal to and the opposite of the angle where the doublet first formed, and then uncouples. Thus, the faster particle moves ahead of the slower particle.

v) In the passing interaction in a linear shear flow with only two particles at zero particle Reynolds number there is only an exchange of position in the convective direction. No net lateral motion occurs, hence no net lateral dispersion. In shear flows of concentrated suspensions, on the other hand, passing events occur almost continuously because of the small distance between the particles. It is impossible to place a beginning or an end on the pass interaction, and, as shown in the following paragraph, net lateral migration may occur.

Qualitative Picture of Lateral Migration in an Infinite Linear Shear Flow. At this point, let us follow in a Lagrangian manner the path of one particle in an infinite linear shear flow of a high-concentration slurry. The particle has a rotation which is time-varying because of the changing influence of its several near neighbors. However, the average rotation is approximately half the average shear rate. The particle is continuously passing and interacting with its lower-speed neighbors, and is being passed by and interacting with its higher-speed neighbors. Since the average distance to a neighbor is less than a particle radius the beginnings and ends of the pass interactions are indistinct. Thus, part way through an interaction with one particle, an interaction with another particle may begin. The effects of the near neighbors may be greater on one side or the other during the course of an interaction, thereby preventing the particle from returning to its original lateral position.

It is exceedingly important to note that the near neighboring particles, which determine both the frequency of passing interactions and the number and size of disturbances during passing interactions, are continually changing. The particles with higher convective velocities move ahead while those of lower convective velocity drop behind. In brief, each passing interaction occurs with a new arrangement of near neighboring particles. Because average concentration is uniform, the number of times the near neighbors on the high velocity side dominate an interaction will equal the number of times the near neighbors on the low velocity side dominate an interaction. Thus there will be no directional preference in the migrations. On the other hand, a directional preference would occur if a concentration gradient were present, since there would be statistically a greater number of interactions dominated by the near neighbors on the side of higher concentration. In the flows we are considering, however, it would appear reasonable to assume that the arrangement of near neighbors is a continuously changing random event. Then, since the lateral migrational motions are created by many inputs from a random arrangement of near neighbors, the lateral dispersion, or self-diffusion of the particles, should exhibit the type of diffusional behavior produced, for instance, by random walk processes, and described quantitatively by Fick's Law.

Insights from Other Physical Phenomena. Two familiar phenomena provide some insight into the nature of diffusional lateral migration in a shear flow: molecular diffusion in a gas and, eddy diffusion in turbulence. Neither, however, is strictly analogous.

Molecular Diffusion. A billiard ball model of the self-diffusion which occurs in a dilute gas is sufficient for the purposes of this analogy. (See, for example, Vincenti and Kruger [13] or Jeans [14].) Since the diffusion in such a gas occurs because of the many collisions between the molecules of the gas it is a form of self-diffusion. As the collisions occur with no loss of mechanical energy, the phenomenon is reversible.

In name, the events which cause the diffusion in a shear flow are identical; that is, there are interactions or collisions between like particles and for inertia-free flow the diffusive motion is kinematically reversible. A closer view of the interactions, however, shows a large difference. In a dilute gas the molecules can recede from the collision in all directions without respect to their incoming direction. Furthermore it is the change of direction with each collision which results in the diffusive motion. For the particle in a linear shear flow the direction of motion is not changed as a result of an interaction. Instead the result of a interaction (or collision) is to change slightly the speed at which the particle leaves the interaction by its lateral motion. Thus, the significant

aspect of the interaction which causes net diffusion is an effect of second higher-order, rather than the actual first-order motion during the interaction.

Turbulence. The analogy to turbulence is almost transparent. The rotational motion of the particles are viewed as the eddies in a turbulent flow. Passing interactions between particles are viewed as eddies of larger size with a characteristic lifetime and rate of formation. The velocity of the suspending fluid has both average and time varying components. On these similarities, the diffusion of particles in a shear flow would parallel the diffusion of eddies in a turbulent flow.

The main flaw of this analogy is that it presumes a turbulent flow with an exceedingly limited spectrum of eddy sizes. Furthermore, the number of small-size eddies per unit volume is determined by the number concentration of particles. In contrast, true fluid mechanical turbulence, has a broad spectrum of eddy sizes and an ever-changing identity of the fluid particles which compose the eddies.

A comparison of the data from the experiments and a theoretical calculation by Buyevich [15] using an analogy to turbulent flow is presented in Section 6.3. The comparison is not favorable to the assumption of a turbulent-flow analogy.

1.6 Scope of This Work

This thesis is an experimental investigation of the diffusional type of migrational motion which is caused by the mutually-induced velocity fields that occur in a non-inertial, two-dimensional, infinite, linear shear flow of a slurry. The experiments were carefully designed to minimize the additional sources of migrational motions, described earlier, that might otherwise occur in slurry flows.

Specifically, the construction and operation of the experimental apparatus had as objectives and features the following simplifications of Equation 1.2 relating the dimensionless parameters of the problem:

- i) The particle Reynolds number, $\rho_f a^2 \omega / \mu$, was made sufficiently small as to be effectively zero, such that the flow was virtually inertia-free.
- ii) The velocity profile was sufficiently linear such that the parameter $a \partial^2 u / \partial y^2 / \omega$ was effectively zero, and curvature-driven migrational motions were negligible.
- iii) The density of the particles was sufficiently well-matched to that of the fluid as to make the parameter $g(\rho_s - \rho_f)a / \mu \omega$ effectively zero, thus eliminating migrational motion due to gravity or streamline curvature.
- iv) The parameter a/w was rendered unimportant by making it rather small (although it could not be made as small as had been hoped for).

v) The parameter a/y was eliminated as a variable by obtaining data only from the central region of the Couette gap in the zone of linear shear flow, and out of the range where the walls would produce lift forces.

vi) The particles were rigid spheres, thereby eliminating migrational effects relating to deformable particles.

These restrictions reduce the number of dimensionless groups in Eq. 1.2, such that it simplifies to

$$\frac{D}{a^2 \omega} = f(\phi) \quad (1.5)$$

This simple relationship expresses the physical concept that the diffusional motion of an infinite linear shear flow, where all lateral positions are equivalent, is a property only of the local flow conditions, i.e., the shear rate, the particle radius, and the concentration of particles.

Note that this is not a Brownian diffusion. Due to their size, the particles of a slurry have Brownian diffusion coefficients negligibly small compared with the diffusion coefficients produced by the mutually-induced velocity fields of the particles.

Details of the experimental technique and apparatus used to accomplish the simplifications herein outlined are contained in Chapters 3, 4 and 5 and Appendix 1.

2. RELATED WORK BY OTHER INVESTIGATORS

In the literature are several types of investigations related to this thesis. These types include (a) the basic fluid mechanics describing the motion of a single particle in shear flows, (b) experimental studies of particle motion in slurry flows, and (c) applications which hypothesize or demonstrate effects related to the migrational motion of particles in a slurry. The literature relating to each of these is reviewed in the following sections of this chapter. In addition, observations on the nature of particle motion in a linear shear flow of a slurry, which are relevant to the operating conditions used for this experiments, appear in Section 2.2.1.

2.1 Particle Motion in Shear Flows

The motion of individual particles in nearly inertia-free flows is well studied; see for instance, Brenner's thorough review article [5] or the book by Happel and Brenner [16].

Among the results of these studies are the following which are particularly relevant to the experimental investigations of this thesis:

- 1) In an infinite shear field containing an individual neutrally-buoyant particle, the convective velocity of the particle

is equal to the velocity of the fluid streamline which would pass through its center if the flow was undisturbed by the presence of the particle.

ii) An individual particle in a shear flow will have a rotational motion. In an infinite shear field containing an individual neutrally-buoyant, spherical particle, the rotational speed of the particle is exactly half the local shear rate of the fluid.

iii) The presence of a solid boundary parallel to the direction of the shear flow decreases both the rate of rotation and the rate of translation of the particle.

iv) In an inertial flow, the retardation of the particle due to a nearby wall produces a lifting force on the particle. In a totally inertia-free flow, on the other hand a rigid particle experiences no lift force even with the retarding effect of a boundary.

v) Flexible particles or liquid drops of an immiscible fluid experience lifting forces in laminar shear flows, whether in the inertia-free or inertial flow regimes.

On the Lack of a Lifting Force on a Rigid Particle in an Inertia-Free Flow. Considering the asymmetric geometry the lack of a lifting force for a translating, rotating rigid particle in an inertia-free flow seems amazing. That it is so, however, may be seen rather simply through a thought-experiment which uses the principle of kinematic reversibility for inertia-free flows.

Suppose that in a horizontal pipe flow there is a left-to-right flow which carries along, near the bottom, a neutrally-buoyant particle whose diameter is small compared to the pipe diameter. Assume, for the sake of the argument that the particle experiences an axis-directed (upward) motion due to a lift force. Now, by the principle of kinematic reversibility, when the flow is reversed, the particle motion and lift force must change from the axis-oriented direction to a wall-oriented direction. However, had one been viewing the flow as first proposed, but from the rear, the particle would still have had an axis-oriented direction, but with a right-to-left flow direction. This flow direction is identical to that in the kinematically reversed flow, but the particle motion is opposite to that occurring in the kinematically reversed flow. Thus, in order to satisfy the principle of kinematic reversibility that prevails in flows at zero Reynolds number, and avoid a paradoxical behavior in which the particle motion would depend upon the position of observation, the original presumption of a lift force must be abandoned. Only when the particle exhibits no vertical motion is the paradox removed. (The condition of neutral buoyancy is important in this argument because the gravitational force is not kinematically reversible. This lack of kinematical reversibility does not allow a comparison of the rear view of the particle motion with the motion when the flow is kinematically reversed.)

2.2 Experiments on the Motions of Particles in a Slurry Flow

Fundamental experiments on particle motions in a slurry flow are usually conducted in either a linear shear flow established by a Couette apparatus or in a Poiseuille flow established in a pipe. An excellent review article by Goldsmith and Mason [1] covers almost all the experiments on particle motion in both common types of shear flow. The two subsections that follow are categorized by the type of shear flow employed in the experiments.

2.2.1 Couette Flow Experiments. A number of experiments on the motion of particles in a circular-cylinder Couette apparatus are reported in the above-mentioned review article by Goldsmith and Mason. The experiments most relevant to this thesis are those performed by Karnis, Goldsmith, and Mason [17]. In addition to studying the nature of the interaction between two spherical particles in a linear shear flow, which was described in the previous chapter, Karnis, Goldsmith, and Mason have noted the occurrence of net lateral migration, or dispersion, in tube and linear shear flows of concentrated suspensions. They note that--unlike the case of two-body interactions--a lateral migration is possible when three or more particles participate in an interaction.

For an understanding of why the number of particles is of importance it is helpful to review the explanation of why there is no net migration for a two particle interaction. This explanation is very similar to that which demonstrates that there is no

lifting force on a body in an inertia-free flow near a boundary. Consider that a two-particle interaction occurs in an infinite linear shear flow, and postulate, for the sake of the argument that the particles experience some net lateral migration. Specifically, suppose the migration caused a relative separation of the particle, as shown in Figure 2.1. Then, because the flow is inertia-free and thus kinematically reversible, it is possible to reverse the flow and thereby undo the lateral migration. For the event in a shear flow in one direction the particles then end up further apart, but by reversing the flow they can be returned to their original locations. However, the rear view of the original shear flow has the same flow direction as the reversed shear flow. Upon re-starting the experiment but viewing from the rear side an increase in the separation of particles would still be hypothesized. This paradox, indeed physically impossible inconsistency, can only be avoided if the interaction produces no net migration. The argument is illustrated in Figure 2.1 with its conflicting possibilities.

When many particles are present and interacting, but with absolutely symmetric original locations (e.g. a crystalline array), interactions with no net migration can also occur, but these are highly improbable circumstances. For the usual multiple particle interaction, as illustrated in Figure 2.2, the relative geometric arrangement of the particles at the beginning of the kinematically-reversed flow of a three-particle interaction is not identical to the particle arrangement when the original flow is viewed from the

rear side. As shown in the figure, particles A and B are the first to interact during right-hand shear flow and the particles A and C are the last to interact. Thus, during the left-hand, kinematically-reversed shear flow, particles A and C interact first and particles A and B last. The kinematically-reversed interactions do restore the particles back to their original positions but are not equivalent to the interactions for flow in the reverse direction. Without this equivalence it is not possible to conclude that the interaction causes no net lateral motion of any of the particles. Indeed there is net lateral migration in a multi-particle interaction.

The effect of the interactions thus obviously depends upon the number density of particles. For very dilute slurries, exceedingly little net lateral migration occurs since almost all the interactions are between pairs of particles that are widely separated from other particles. As the concentration increases the probability of multiparticle interactions increases rapidly, and the amount of lateral migration will correspondingly increase. Figure 2.3 taken from Karnis, et al. [17] shows the change in the radial position of a labelled particle suspended in a slurry flow in a cylindrical Couette apparatus for different slurry concentrations. An experiment using only a single, neutrally-buoyant particle in the shear flow of a Newtonian liquid would trace out a horizontal line on the coordinates of Figure 2.3(a). The particle path in the upper half, (a), is under conditions of 3% concentration while the

lower tracing, (c), is for a concentration of 19%. At the 19% concentration, all particles are very closely spaced so the probability of an unencumbered two-, or even a three-particle, interaction is exceedingly small. The tracer particle is in continuous interaction with many near neighbors as previously discussed in Section 1.5. The particle motion in Figure 2.3(c) exhibits a chaotic, diffusional motion; to accurately measure a statistical value for the diffusion coefficient from such an experiment a very long period of observation of the motion of the tracer particle is necessary. Further, these measurements should be within a "cloud" of particles so the tracer particle can experience the full range of interparticle interactions.

Reynolds Number Criterion for Kinematic Reversibility.

Karnis, et al. report that the interaction between two particles is no longer reversible above a particle Reynolds number of 10^{-5} . This is a surprisingly low limit, and the inertial effects involved must be extremely small. The explanation of why the limit is so small lies in the method of decision for when the purely viscous regime has been left. Most such decisions are based upon a first-order criterion such as a velocity, flow, or pressure measurement, which represent instantaneous measures of the flow. However, the reversibility of a two-particle interaction is a decision based on a criterion of the return of the particles to their original location, and relates to an integral measure which can be

substantially affected by small deviations whose cumulative effect is of the order of the quantity being measured. More specifically, each of the deviations which occur during the interaction have the effect of making the interaction from that point on behave as if it had started from a different original arrangement of particles. Not only will this include all the inertial deviations but it will also include all experimental deviations from the perfect conditions required to achieve kinematic reversibility.

Instantaneous spatial positions and velocities are not measured; rather the measurement which is made to decide whether the property of kinematic reversibility has been attained, is the degree to which the particles return to their original locations. It is possible to perform this type of "almost-null" measurement very accurately by choosing an instrument with a small range of size of measurements since the accuracy of most instruments is restricted to a fraction of the total range. This measurement of the deviation from inertia-free flow, with its physically determined standard of zero net motion, is much more sensitive than that of other determinations such as of a velocity where a measured value is compared on an absolute scale to a theoretical value. Thus, both the integral criterion and the type of measurement allow a finer determination of the property of kinematic reversibility than is commonly performed.

For a particle in a concentrated slurry flow the Reynolds-number limit of kinematical reversibility would be much lower simply because of the much greater number of particles which would experience a motion influenced by a small amount of inertia. Each inertial deviation of any particle would influence all subsequent interactions. However, although the net result of inertial effects on an individual particle can be measured at very low particle Reynolds numbers, the viscous domination of the average properties of the slurry flow will be described by the limits of the Reynolds number.

A simple analogy of the effect of minor inertial forces is that they affect the diffusion of particles in a slurry as entropy affects the diffusion of gas molecules: to first-order both prevent a "time-reversal" of the phenomena. As in molecular diffusion where the original randomness of the velocity and positions of the molecules gives rise to the diffusive behavior of an individual molecule, in shear-driven diffusion the original randomness of location gives rise to the diffusive motion of a particle. In both, the multibody nature of the phenomena, which prevents a time-reversal of the motion, can be viewed as an extra degree of randomness.

2.2.2 Concentration and Velocity Profiles in Couette Flows. In the laminar flow of a Newtonian fluid, such as cane syrup, the linear velocity profile is used to define Couette flow.

The concentration profile of the solute, sugar, in the solvent, water, would be uniform across the channel. In a slurry flow the particles are many orders of magnitude larger than the molecules of the suspending fluid (or the solvent). Therefore such a simple concentration profile is not possible near channel walls because the walls exclude particles for a distance up to a particle radius. To obtain a physical view of this exclusion process, consider the sketch of Figure 2.4 where each particle has been represented as a circle with a center point. Observe the concentration in the various strip areas parallel to the channel wall; a concentration, for the purposes of this illustration will be defined as the number of center points of particles/strip area. Note that in the area near the wall, since particles cannot penetrate the wall, the number of center points decreases. (This region is sometimes termed the "Vand zone" after the investigator who first recorded the effect of it upon the rheological behavior of slurries [18].) Karnis, et al. [17] report that the concentration across a Couette flow of a slurry is uniform except for this Vand zone at each channel wall. Noting that a net redistributive force upon the particles at any lateral channel position would result in a non-uniform concentration profile, Karnis concluded no wall force of significance existed for low Reynolds number (effectively inertia-free) flows. As previously noted, this is consistent with the theory of zero Reynolds number slurry flow.

Karnis observed, however, that the velocity profile has a much more complex shape. As can be seen in Figure 2.5, it is not linear but rather has a linear center section with a zone of higher concentration at each wall. Parts (a), (b) and (c) of Figure 2.5 show the effect of the ratio of particle size to channel width a/w and concentration; at small ratios and low concentration the profile is approximately linear. At large ratios and high concentrations the effect is to alter dramatically the velocity profile. Most of the experimental measurements reported in this thesis are for ratios of particle size to channel width similar to those in part (b) of Figure 2.5; two measurements; which had relatively large associated errors, had a size ratio similar to that in part (a) of Figure 2.5, however, they had much higher particle concentrations.

The change in the velocity profile near the wall results in an increased shear rate in a region of decreased concentration. This increased shear rate occurs because the shear stress must of necessity be nearly constant across the gap. In a concentric cylinder Couette device mechanical equilibrium requires a constant moment of the shear force on each concentric cylindrical surface. Since the change in radius is small across the gap this is a condition of approximately equal shear stress on all cylindrical surfaces. The decreased concentration near the wall causes a decrease in the apparent viscosity which necessitates an increased shear rate to balance the shear stress on cylindrical surfaces near the wall.

Karnis, et al. [19] have also investigated the effect of inertia on the behavior of suspensions in Couette flow and Poiseuille flow. Their experiments, which cover a range of particle Reynolds number up to 1.0 for spherical particles with ratios of particle size to channel width, a/w of 0.007 to 0.03, show that in Couette flow there is no measurable effect on the rate of particle rotation. Although experiments are reported showing the redistribution of particles of various shapes in Poiseuille flow and showing preferred orientation of non-spherical particles in Couette flow, no parallel redistribution phenomenon is reported for Couette flow. To observe the establishment of an orientation direction for particles in Couette flow requires considerable time and effort; furthermore any orientation phenomenon would be affected by redistribution of the particles. Based upon the lack of reported experimental findings by Karnis et al. (who have performed some of the best and most thorough research in the area of particle motions) it seems reasonable to infer that no redistribution phenomena occur in Couette flows at these Reynolds numbers. Visual observations of the Couette flow of slurries during the course of this thesis have provided us no reason to believe in a net redistribution of particles in Couette flows with the particle Reynolds number less than 1.0.

Summary. To summarize, previous investigators have demonstrated that diffusional phenomena do indeed occur in Couette flow. Further, they show that the concentration is approximately uniform across a Couette gap with the exception of a region within a particle diameter of the wall, which is depleted slightly. The depletion produces a non-linear velocity profile in this region. No measurements of the coefficient of the diffusional motion, its scaling with flow parameters or particle size, or of its functional relationship to the particle concentration of the slurry are reported. A principle objective of the thesis is to provide an experimental measure of these properties of shear-driven diffusion of spherical particles.

2.3 Particle Motion in Poiseuille Flow.

Most previous work on the motion of particles has been done for tube flows because of their obvious application to industrial and biological situations. Much of this work has been directed toward a description of the net redistribution of particle concentration in the cross-section of the pipe. Net redistribution of the particles can be caused by either forces which act upon the particles or unequal lateral fluxes caused by gradients in the concentration and/or diffusion coefficient.

Segré-Silberberg Effect. In an exquisite series of experiments, Segré and Silberberg [4] showed that in a tube flow that originates from a reservoir containing a small concentration of neutrally-buoyant particles, the particles migrate to a stable position at a radius equal to approximately 60% of the tube radius, independently of their initial radial position as they entered the tube. These experiments were conducted with extremely low concentrations by volume ($< 0.5\%$), and a small ratio of particle radius to tube radius, 0.03 to 0.15. The tube Reynolds number was varied from 2 to 500.

In these experiments the particles assumed a location in the tube where there is significant curvature of the velocity profile. This location is significant to the lateral force balance necessary to achieve a preferred, stable, non-central radial location. Specifically, one can suppose a balance of forces which result from a slight inertial effect on a particle near a boundary in a shear flow with curvature of the velocity profile. The lift force generated by the linear shear flow around the particle in the presence of a wall is balanced by the lift force generated by the curvature of the velocity profile. In the region near the wall the "wall force" dominates, producing an inward redistributive motion, while in the central region the "curvature force" dominates, producing an outward redistribution of particles. During the writing of this thesis a theoretical explanation of these phenomena has been published by Leal and Ho [3]. Rubinow and Wohl [2]

have provided a theoretical description of the migration of deformable particle in a shear with curvature.

Sacks and Tickner [20] performed visual experiments to classify regimes of flows of neutrally-buoyant suspensions with higher concentrations. Their experiments show a "critical" tube Reynolds of 10, in the following sense. Above this Reynolds number a zone depleted of particles always develops near the walls of the tube. For flows of all Reynolds numbers a plug flow develops as the concentration is increased. Sacks and Tickner describe a "tumbling motion" which occurs among the particles at intermediate concentrations. They note that this motion is especially pronounced near the tube walls and that it allows faster particles to pass slower particles. This motion is essentially identical to the passing interaction which is illustrated in Figure 1. Karnis, et al. [17] also note the erratic motion of the particles near the wall in their work on tube flows of slurries at relatively low particle Reynolds number. With a slurry of 33% concentration, Karnis [17,19] observed a plug flow in the central region, and noted that there appeared to be little radial migration of an observed particle compared to the considerable lateral motion of a similar particle in the high-shear region near the tube walls. Although the particle motions in these studies are not solely the result of a shear-driven diffusion, the scale of the motions does correlate with the local shear rate. Similar observations of the migration of a red blood cell in the tube flow of a suspension of

ghosts have been made by Goldsmith [21].

Summary. In tube flows there is always a curvature of the velocity profile and therefore a non-uniform shear rate across the tube. The curvature is important since, by reason of the associated asymmetry in the resulting flow around the particle, it represents the origin of a lateral force which affects the migration of the particles. The importance of the curvature is demonstrated experimentally by the Segré-Silberberg experiments and dimensionally by the group $(a\partial u^2/\partial y^2)/\omega$ as shown in Section 1.3. For flows of concentrated slurries the interactions between particles will also come into play. It is the complex, position-variable combination of the several forces which makes experiments using tube flows impractical at this time for the isolation and description of any single one of them.

2.4 Augmented Diffusion in Blood Flows

The literature on biological fluid mechanics contains a number of papers which treat or postulate a flow-induced lateral motion of individual particles or the net effect of such motion on macroscopic transport phenomena.

In a theoretical treatment of experiments which measure thrombus formation in a stagnation-point flow, Petschek and Weiss [22] postulate a shear-driven diffusive motion of the platelets. The form and scaling of the diffusion coefficient are obtained

from a perfect-gas model of the RBC platelet motion. In the model the rotational motion of the RBC's is cited as being the primary cause of the increased platelet diffusivity. The increased diffusivity is scaled as $a^2\omega$, where a is the effective radius of the RBC and ω is the angular rate of shear. A comparison of their estimate of the platelet diffusion coefficient with measured values in a flowing system, however shows the theoretical estimate to be two orders of magnitude larger than the measured value.

In the interpretation of experiments which measured the uptake of radioactively labelled cholesterol from a tube wall by RBC's, Bernstein, Blackshear, and Keller [23] postulate the existence of a RBC diffusion coefficient which is proportional to shear rate. Steinbach [24] has measured the diffusion of RBCs using the method proposed by Taylor [25] for determining the diffusion coefficient through observation of labelled material during flow in a pipe. Steinbach's results show no shear-rate dependence of the diffusion coefficient. His experiments suggest a value of D of order 10^{-8} cm^2/s , and, since the Brownian diffusion coefficient for a large body like the RBC is $\sim 5 \times 10^{-10}$ cm^2/s , it would appear that the effective diffusion coefficient has been enhanced by the shear flow. The apparent conflict in these statements--that there is no shear-rate dependence of the diffusion and that the diffusion coefficient is enhanced by flow--may conceivably be resolved by considering the flexibility of the RBC. This flexibility may be a shear rate dependent property such that at low shear rates the RBCs exhibit

a passing interaction or a tumbling motion like rigid particles but at higher shear rates the RBCs may deform to avoid interactions and thereby limiting the amount of augmentation. Another possibility is that the redistribution phenomena and plug-like velocity profiles may prevent Taylor's method from useful application in these situations. The difficulty of experimental technique with resulting inaccuracies may also obscure any shear-rate dependence in the experimental results.

In a series of experiments which also used Taylor's method to measure the diffusion coefficient of platelets in flowing blood, Tumillo, Benis, and Leonard [26] found significant enhancement of the diffusion coefficient for a flowing system as compared to a static Brownian-motion diffusion coefficient. Because of a limited range of flow rates and statistically-limited sensitivity of the instrument used to count platelets they note that the experiments could not in a meaningful way detect a dependence of the diffusivity upon the shear rate or the hematocrit. Grabowski, Friedman, and Leonard [27] measured experimentally and modelled mathematically the effects of shear rate on platelet-thrombus formation. The results of this work indicate that platelet diffusivity is a shear-rate dependent quantity with a power-law relationship whose exponent is less than one. It should be remembered that the major cause of the increased diffusive motion of the platelets is rotational motion of the red blood cells and not the interaction between the platelets themselves. The rotation of the RBC tends to carry the

platelet from one side of the RBC to the other. At this station the platelet may be returned toward its original location or come under the rotational influence of another RBC. Since platelets exhibit chemototactic motion such experiments are hard to interpret in a purely fluid mechanical sense. However, they are extremely relevant from a medical viewpoint.

The migration of RBCs in shear flows offers a potential method for increased efficiency in blood oxygenators. This increased efficiency can be viewed as a form of augmented diffusion. In a blood oxygenator there are two possible sources of augmented diffusion. One source is the rotation of the RBCs which has the effect of locally stirring the surrounding plasma. Collingham [28], using solid particles has studied the rotational augmented diffusion for gas transfer across a tube wall. Most of the experiments were at volumetric concentrations of less than 15%; the experiments used neutrally-buoyant polystyrene-salt water slurries. An augmentation of 2 to 6 times the molecular diffusion coefficient was observed; the amount of augmentation was shown to scale as ω^2 . Parallel experiments in which heat transfer rates were measured were carried out by Singh [29].

In a series of experiments using blood flow through small-diameter tubing, Diller [30] has demonstrated augmentation of the diffusion coefficient of dissolved gases. A separation of the augmentation effects due to the rotational and migrational motions of the RBCs can be obtained by variation of the degree of oxygen

saturation of the hemoglobin in the RBCs as they enter the tube to undergo oxygenation. Due to the sigmoid shape of the saturation curve for hemoglobin as a function of the partial pressure of oxygen to which the hemoglobin is exposed, the RBC at low oxygen partial pressures, can act as a high-strength sink or a high-strength source. Thus, when they migrate from one area to another, these RBCs act as a bucket brigade carrying oxygen from areas of high concentration to areas of low concentration. For higher oxygen partial pressures this effect is minimal and the increase in oxygen transfer is due mainly to the rotational motion of the RBCs which mixes the plasma in its immediate vicinity. By pursuing this approach, Diller was able to show that the augmentation due to the migrational motion of the RBCs was nearly constant over the shear rates studied ($60\text{--}2500\text{ s}^{-1}$); the augmentation produced by this motion is proportional to the sink strength of the RBC and is, at maximum, about 2-1/2 fold. The relative independence of shear rate for the migrationaly-enhanced diffusion which was observed by Diller is consistent with the hypothesized flexibility limitation on RBC diffusion discussed previously. In this case it would represent a limit upon the amount of augmentation of the diffusion coefficient which would be achieved by introducing shear in oxygenators.

Two oxygenators using incorporating shear-augmented diffusion have been described in the literature. Keller and Overcash [31] have designed and tested a circular-cylinder Couette oxygenator. The blood path through this oxygenator is helical. Oxygen and

carbon dioxide transfer occur through the cylinder wall. Preliminary results indicate a 100% augmentation at a shear rate of 1000 sec^{-1} . The Tecna Corporation [32] has designed a disc-shaped blood oxygenator which has a geometry similar to that of a centrifugal pump; blood enters in the center of the casing and exits through a peripheral port. The casing and a rotating disc hold the membrane surfaces through which gas transfer occurs. Within the casing, the rotating disc sets up a radially-varying shear field between itself and the casing; the amount of shear, and with it, the augmentation, is proportional to the rate of rotation of the disc. Oxygen transfer has been demonstrated to be increased by rotation of the disc by up to a factor of three.

The above references which show a medical use for an increased understanding of shear-augmented diffusion provide a humane incentive for the pursuit of this investigation.

3. REQUIREMENTS FOR AND DESCRIPTION OF THE SLURRY AND THE SHEAR FLOW APPARATUS

The first section of this chapter describes the necessary requirements that the slurry and shear flow must satisfy to accomplish the aims set forth in Chapter 1. It also shows the ranges of values of the non-dimensional groups of Equation 1.2 used in the experiments. The second and third sections contain explicit descriptions of the Couette apparatus and the components of the slurry used in the experiments.

3.1 Use of the Dimensional Analysis to Establish the Requirements of the Slurry and the Experimental Apparatus

A guide to scaling of equipment and selection of materials can be obtained from the dimensional analysis of Chapter 1. For convenience, we repeat the equation resulting from the dimensional analysis:

$$\frac{D}{a^2 \omega} = f\left(\phi, \frac{a}{y}, \frac{a}{w}, \frac{\rho_f a^2 \omega}{\mu_f}, \frac{a \partial^2 u / \partial y^2}{\omega}, \frac{g a (\rho_s - \rho_f)}{\mu_f \omega}\right) \quad (1.2)$$

To obtain a guide to the ideal values of the dimensionless groups we compare the above equation with equation 1.5 of section 1.5, where the scope of this thesis was limited to a measurement of the diffusional motion caused by the mutually-induced velocity fields of

the particles in a linear shear flow of a slurry. In that section it was noted that this type of diffusional motion occurs in an inertia-free, zero-Reynolds-number, infinite linear shear flow of a neutrally-buoyant slurry of rigid particles. The diffusional motion is driven solely by the shear--it does not require bounding walls, curvature of the velocity profile or inertial effects to occur. The dimensionless equation for this ideal situation reduced to

$$\frac{D}{a^2 \omega} = f(\phi) \quad . \quad (1.5)$$

To achieve this simplicity, the terms which are not common to equations 1.2 and 1.5 must be zero or sufficiently small such that the phenomena they represent do not substantially influence the diffusive motion.

The selection of materials for the components of the slurry and of the experimental design and operating conditions to obtain a zero or near zero values of the dimensionless groups may be a difficult or even impossible task. For example, consider the dimensionless group

$$\frac{g\left(\frac{\rho_s - \rho_f}{\rho_f}\right)}{(\mu_f / \rho_f) \omega}$$

This is zero for particles that are exactly neutrally buoyant. Although the value of $\frac{(\rho_s - \rho_f)}{\rho_f}$ can in a practical sense be made exceedingly small (e.g. less than 0.1%), variations of density

among the particles occur as an intrinsic part of the manufacturing processes for plastics, and this sets a lower limit on $\frac{(\rho_s - \rho_f)}{\rho_f}$. To reduce the dimensionless ratio further, the suspending fluid should be of high viscosity. This also has the desirable effect of reducing the Reynolds number. On the other hand, reducing the buoyancy parameter by using large values of ω tends to produce large Reynolds numbers. Similarly, for large values of $a^2 \omega$ D will be large (see Eq. 1.2). Large values of D are desirable since the associated diffusive displacements tend to be large and, therefore, can be measured more accurately. However, increasing $a^2 \omega$ also has the undesirable, concomitant effect of increasing the Reynolds number.

Upon investigation of possible approaches to the experimental design, and the materials from which the slurries and experimental apparatus would be made, it became apparent that most of the ratios are limited by the availability of materials in certain sizes or with certain properties. To wit, the particle size must be relatively large in order to use the detection system described in chapter 4. Since this detection system follows the diffusive motion of one marked, identifiable particle, many other unmarked particles are necessary. These two restraints, in combination with the need for a viscous suspending fluid of equal density, determined the properties of the slurry.

Availability of materials and restrictions imposed by construction techniques limited the maximum width of the Couette flow apparatus used to establish the shear flow.

The scale of ω is set by the desire to have a relatively large diffusive motion which can be measured with a reasonable accuracy and a desire to collect data within a reasonable time frame.

Consideration of size of the diffusive motion also governed the choice of particle volumetric concentration. These restrictions and considerations produced the following ranges of the non-dimensional groups

ϕ	0.2, 0.3, 0.4, 0.5
$Re = \frac{\rho_f \omega a^2}{\mu_f}$	4×10^{-4} to 1×10^{-2}
$g(\rho_s - \rho_f)a/\mu\omega$	0.004 to 0.02
$\frac{a}{w}$	0.02 and 0.06

Values of the ratio of $a \frac{\partial^2 u}{\partial y^2} / \omega$ vary across the channel flow; actual velocity profiles for three values of a/w published by Karnis et al. [17] are discussed in Chapter 2. For the present purposes it is sufficient to note that the results of Karnis make it possible to relate the ratios, a/y and a/w , to the size of wall regions, thus indicating where the ratio $a \frac{\partial^2 u}{\partial y^2} / \omega$ is significantly different from zero. Based on those results, only diffusive motions beginning in the central one-fifth of the channel were used to calculate D , since in this region the velocity profile is nearly linear.

The ratios listed above represent an unavoidable compromise between the ideal experiments hypothesized in section 1.5 and the

realizable experiments. This compromise reflects itself in the fact that only the buoyancy parameter is wholly negligible, and the dimensionless expression which describes the experimental studies of the diffusive motion contains as weak parameters additional dimensionless groups i.e. beyond the concentration ϕ .

$$\frac{D}{a^2 \omega} = f\left(\phi, \frac{\rho_f a^2 \omega}{\mu_f}, \frac{a}{w}, \frac{a}{y}\right)$$

Care should be taken not to write off the ideal underlying the experiments as being unattainable; rather, at this point only the ideal experiments have not yet been attained. What remains is to test the effect of the weak non-dimensional ratios--if they do not significantly alter the experimental results then it is still possible to measure the shear-driven diffusive motion such as would occur in an infinite shear flow.

3.2 Couette Flows

One of the aims of Section 1.5 is to conduct the experimental measurements of the shear-driven diffusion in a linear shear field at a zero or near-zero particle Reynolds number to avoid inertial motions. There are two common types of experimental apparatus' used to establish a linear-shear or Couette flow; both establish and drive the flow through a difference in the velocity of two parallel walls. In the case of low-Reynolds-number flows of Newtonian fluids

both can produce, with appropriate precautions, exceedingly good approximations to a linear shear flow. Both the linear-belt Couette device and the circular-cylinder Couette device are described and discussed subsequently. The rationalization underlying the selection of a circular-cylinder apparatus for the purposes of these experiments will become apparent as a result of this discussion. The effects produced by a change from a Newtonian fluid to a concentrated slurry as measured by Karnis, et al. [17] were discussed in Chapter 2.

3.2.1 Linear-Belt Couette Apparatuses. At the beginning of this work we borrowed¹ a linear belt apparatus for the purpose of testing the quality of the linear shear flow of the slurry which is produced by this type of device. This device has a length of approximately four feet and a channel height of approximately 12 inches. The channel width is adjustable; a width of 1.00 inches was used for our preliminary tests. A sketch of the top view of the apparatus is shown in Figure 3.1a. The linear-belt Couette apparatus sat inside a large tank, T, which contained the fluid. Use of a tank is a very convenient feature since it eliminates the need for fluid seals at moving surfaces. The Couette belt, B, was

¹This device was borrowed from Prof. E. Mollo-Christiansen who, in conjunction with his graduate-student co-workers, used it to study the stability of Couette-flows of density-stratified salt water. The density stratification and much smaller channel width to height ratio used in their experiments resulted in an acceptable Couette flow for their purposes.

driven by a motor and v-belt drive connected to the axle of one of the belt-positioning pulleys, P. The two sides of the belt are maintained parallel to each other through tension in the belt which tends to pull them against metal support plates S,P. This arrangement was not satisfactory and restraining guides were installed on the metal support plates at the top and bottom edges of the belt. The shear rate produced in such a machine is twice the belt velocity divided by the distance between the inner surfaces of the belt, i.e. the channel width.

This device had two primary faults, i and ii, and a minor inconvenience, iii, namely:

i) the belts, despite much effort and many attempted improvements, were exceedingly difficult to maintain parallel to each other and against the supporting walls. Thus, the shear flow in the central region of the apparatus could not be made steady and uniform. This behavior of the belts is fundamental to the design; in many ways the belt is analogous to a string under tension: the force required to deflect the central region of the string is a very small fraction of the tension in the string. The maximum string or belt tension is limited by the materials from which the string or belt is made.

ii) the secondary flows between the stationary fluid under the Couette apparatus and the shearing fluid within the device were very severe. Use of a relatively inviscid bottom fluid damped the secondary circulation in the viscous fluid within the belt-region of

the Couette apparatus, however a satisfactorily low level of secondary flows could not be achieved in the preliminary experiments.

iii) since the fluid is recirculated at the ends of the apparatus there is an entry length at each end. This limits the period of observation over which the motion of an individual particle can be followed within a slurry.

Two-Belt Variation of the Linear Couette Device. Another type of the linear-belt Couette apparatus is common; in this type two belts are used and the linear shear flow occurs in the region between the outer surfaces of the two belts. This type of device is illustrated in Figure 3.1b. Although this type of linear-belt device allows a solution of iii by changing the belt speeds so the individual particle under observation is always at a position with zero fluid velocity, the other problems were expected to remain.

3.2.2 Circular-Cylinder Couette Apparatus. The second type of Couette flow apparatus was also borrowed and tested. This type, which, from a limited survey of the literature, appears to be the more popular type, consists of two concentric cylinders as shown in cross-section in Figure 3.2. The fluid is placed in the annular space between the cylinders; a shear flow is produced by rotating the cylinders at different speeds. When the gap distance between the cylinders is small compared to the radius of either cylinder, an approximately linear shear flow results. The exact form of this flow is easily calculated from theory when end effects are ignored; for a

derivation of the flow see Lamb [33]. In Appendix II, Figure A2.2 shows a comparison between the calculated profile and an average linear velocity profile for the borrowed circular-cylinder Couette device. A Couette device of similar dimensions, which will be described subsequently, was constructed for use in these experiments.

This type of Couette apparatus is slightly more difficult to construct than a linear belt apparatus. However, if the cylinder walls are rigid and concentric, they will generate no unsteady motion, such as caused by deflecting belts. In contrast to the belt-type apparatus, the concentric-cylinder Couette flow can be made highly accurate through careful design and painstaking construction. The range of available cylinder blanks from which the Couette cylinders can be machined is a significant limitation on the maximum economical size of this type of Couette apparatus. Thus, obtaining a small ratio of a/w via a large cylinder gap, w , is very costly.

Physical Description of the Concentric-Cylinder Couette

Apparatus. A cross-sectional view of the Couette apparatus ultimately used to create the linear shear flow is shown in Figure 3.2. It has four main parts: an outer cylinder (C), an inner cylinder (B) which is mounted on an axle, and upper and lower spacer plates (A and D respectively) which maintain the concentricity of the outer and inner cylinders through the bearings and the axle. The outer cylinder and the bottom spacer plate are epoxied together to form a cup which contains the fluid that is sheared. An oil seal is used at the axle

to allow rotation of the axle relative to the spacer plate without fluid leakage. The inner diameter of the outer cylinder is 13.05 inches and the outer diameter of the inner cylinder is 10.98 inches. The gap distance between cylinders, w , is 1.053 ± 0.002 inches. The total possible fluid height is 9.8 inches.

When the cylinders have different velocities a shear flow is created in the gap region between the cylinders. For the apparatus used in these experiments either or both cylinders could be driven. The outer cylinder was driven via a V-belt groove which was machined into the bottom spacer plate. The inner cylinder was driven via a sheave attached to the axle. Variable speed motors were used to drive both cylinders. All the experiments reported, except for the one with $\omega = 0.4 \text{ s}^{-1}$, were performed with the inner cylinder held stationary. For the experiment with $\omega = 0.4 \text{ s}^{-1}$ a co-rotational mode of operation was used, with the speed of the outer cylinder identical to that of the experiments having $\omega = 1.0 \text{ s}^{-1}$.

The Couette apparatus was mounted via the central axle in pillow-block bearings on a large, rigid steel frame. The steel frame provided an easy method of mounting the V-belt drive systems and variable speed motors which were used to drive the Couette cylinders. The rigidity of the frame ensured that the initial alignment of the Couette apparatus and the detection components were maintained throughout the experiments. As will be explained in Chapter 4 the alignment of the Couette apparatus and the detection components is important to the accuracy of the detection technique.

Secondary Flows. Secondary flows are a general problem of Couette devices. In the apparatus of Figure 3.2, by reason of the fact that the bottom spacer plate is epoxied to the outer cylinder to form a cup, the fluid near it cannot achieve the uniform shear rate of fluid in the central region of the Couette apparatus. The centrifugal force on the fluid in the viscous boundary layer produced by the bottom spacer plate exceeds that on the fluid outside this boundary layer, thus creating an outwardly-directed secondary flow near the bottom spacer plate. This flow is deflected upward by the outer cylinder. A secondary toroidal circulation is thus produced in a diametral plane as shown in Figure 3.3. The secondary-flow stream surfaces are doughnut-shaped, the secondary flow being outward near the lower plate, upward near the outer cylinder, inward near the upper free surface, and downward near the inner cylinder. If there is no air between the upper spacer plate and the liquid in the Couette apparatus, two secondary-flow cells develop, the bottom one having the direction described above and being half as tall; the upper secondary flow is a mirror image of the bottom secondary flow about the central horizontal plane perpendicular to the axle.

To reduce the secondary circulation layers of relatively inviscid fluids are used near the spacer plates. Besides being relatively inviscid the upper fluid layer must also be less dense than the slurry and correspondingly, the lower fluid layer must be more dense. Air, an obvious choice, was used near the upper spacer plate and a totally-fluorinated hydrocarbon (Fluorinert by 3M Co.) was used

near the lower spacer plate. Ethylene glycol was tried as a lower fluid but the relatively small density differences, compared to the slurry, i.e. approximately 15% was not sufficient to prevent globules of ethylene glycol from being entrained by the slurry. Fluorinert is approximately 65% more dense than the slurry.

These relatively inviscid fluid layers "buffer" the effects of the secondary flow upon the slurry. The secondary flow in the slurry is driven by the shear stress on the interface between the slurry and the end fluid. Thus, because of different viscosities the velocity gradient in the slurry is less than that in the end fluid by the ratio of the viscosity of the relatively inviscid end fluid to the viscosity of the slurry. In the experiments, the viscosity ratio of the suspending fluid of the slurry to the more viscous cap fluid was greater than 1000:1. The large effective viscosity of concentrated slurries as compared to the suspending fluid will further reduce the secondary flow velocity in the slurry.

Thermally driven convection is another source of unwanted motion. To avoid these motions the experimental equipment was operated in a room where the air temperature was thermostatically controlled to be at $70^{\circ}\text{F} \pm 2^{\circ}\text{F}$. The thermal mass of the Couette apparatus was sufficiently large such that no variation in slurry temperature could be measured using a thermometer with 0.1°F graduations.

With the above precautions no secondary or thermal flow effects could be detected through observation over a two-hour period of a

single, small, neutrally-buoyant particle in a Couette flow of the viscous suspending fluid.

3.3 Slurries

The slurries used in this thesis were assembled in the Couette apparatus as described in Chapter 5. Each slurry was made from a viscous suspending oil and either of two types of polystyrene particles. The several slurry components are treated below.

Suspending Fluid. A water-soluble artificial oil, Ucon 50-HB-5100 manufactured by Union Carbide, was used as the basis of the suspending fluid. This oil as supplied by the manufacturer is slightly more dense than most commercial polystyrene plastics, consequently water was added to the oil to match the density of the plastic particles. An adequate match of densities was arbitrarily set as a rise time of less than 1 cm/hr for a single large polystyrene particle in a large vessel; this corresponds to a value of $(\rho_s - \rho_f)$ of less than 0.002 gm/cm. As the amount of water added is almost the amount which this naturally hydroscopic oil would absorb from the air, the oil-water solution was quite stable. The viscosity of the oil-water solution used in the experiments was 21 poise at 70°F.

It is interesting to note that under the most severe operating conditions vertical gravitational settling would be observed before centrifugal separation. This agrees with the fact that the ratio of these accelerations (centrifugal to gravity) at a shear rate of 10

inverse seconds is 0.015. During all the experiments the interactions between particles were sufficiently strong to maintain a uniform vertical concentration, at least as judged by eye.

Large Polystyrene Particles. The larger size of polystyrene spheres, 0.125 inch diameter, were commercially manufactured using a centerless grinding process. Consequently, the sphericity and dimensional tolerances were very good, ∓ 0.002 inches. Thus, slurries made using the large spheres are almost truly monodisperse slurries.

Small Polystyrene Particles. The smaller-size spherical plastic particles used to assemble the slurries were sieved from an industrial product (Fosterene Type 50 polystyrene beads which are produced and were donated to us by the Foster-Grant Co.). These beads are polymerized from a monomer solution in a batch process using a suspension method. During polymerization, the surface tension of the plastic causes the beads to assume a spherical shape. The particles used in the experiments are the fraction of the suspension product which passed through a sieve with an inter-wire spacing of 0.045 inch and collected upon a sieve with an inter-wire spacing of 0.038 inch. Measurement of the diameters of two hundred particles showed approximately 6% of the particles with diameters outside of the range 0.038-0.045 inches. No particles with a diameter of less than 0.030 or greater than 0.050 inches were found. Visual inspection of the sieved beads showed occasional doublets, i.e. beads which were formed from two smaller beads, however most of the beads looked

and rolled like perfect spheres. The slurries constructed using these particles have a small range of sizes and may be viewed as having a narrow, top-hat distribution of particles radii.

4. TECHNIQUE AND APPARATUS USED TO MEASURE

THE DIFFUSIVE MOTION

4.1 Techniques for the Measurement of Diffusion Coefficients

4.1.1 Multiple-Particle Techniques. In the most common method of measuring diffusion coefficients an experimental set-up with an initially well-defined distribution of an identifiable diffusing material is used. Measurements are then made of the subsequent changes with time of the distribution, or of the flux of the diffusing material. The coefficient of diffusion is obtained by numerically fitting the measurements to a mathematical solution of the diffusion equation which uses the boundary conditions that describe the initial distribution of the diffusing material. The initial distribution must be truly well-defined to obtain accurate measurements using this method. Furthermore, to obtain easy numerical fits of the experimental measurements to a mathematical solution the initial distribution should lead to a readily-applied analytic solution of the diffusion coefficient.

Examples of Self-Diffusions Which Commonly Use a Multiple-Particle Technique. The shear-driven diffusion is a type of self-diffusion. Other examples of species with self-diffusion are the atoms in single element crystals or the individual molecules in gas of single composition. The common factor of all self diffusion is

that the interactions which give rise to the diffusion motions occur among equal species.

Radioisotopes are commonly used as identifiable, but equal species in order to experimentally measure the diffusive motion and thereby determine a diffusion coefficient. For diatomic gases self-diffusion coefficients are oft-times measured using another chemically-different gas of similar or identical molecular weight as the equal but identifiable diffusing material. Because of the equality of molecular weights the coefficient of binary diffusion for the two gases is a very good approximation of the coefficient of self diffusion.

For self-diffusion experiments using the above identifiable diffusing materials, the well-defined initial distribution is commonly a localized concentration of identifiable material.

Preliminary Experiments. The multiple-particle technique was used in preliminary experiments. The shear-driven diffusive motion which occurs in a single layer of particles positioned on the shearing upper liquid surface of a circular-cylinder Couette flow was measured in these experiments. The well-defined initial distribution of identity was a step change in the color of the particles at the mid-line of the Couette channel.

Rather than measure concentrations, the more accurate measurements of the flux of colored particles across the midline of the Couette channel were taken. The diffusive motion was calculated

from a numerical fit of the measurements to the solution of the diffusion equation using an infinite step as an initial condition. For short diffusion times, the solution for the annular geometry is approximately the solution for an infinite plane step. The limited channel width does not enter and the solution has the mathematical form of an error function. Analysis of the data showed that the effects of interfacial tension prevented a solely diffusive motion of the particles. Accordingly single-layer experiments were discontinued; a more thorough description of the experiments and the results is given in Appendix II.

The surface effects which prevented a close interpretation of particle diffusion in the preliminary experiments demonstrated the necessity of performing measurements of the shear-driven diffusive motion within a cloud of particles in a slurry. No method could be found to establish a well-defined distribution of particles, would would be identifiable by color or other label, within a cloud of particles in a Couette flow. Consequently, the experiments for this thesis were performed using a single identifiable particle as next described.

4.1.2 Single-Particle Technique. When the ergodic hypothesis is used, a second technique for the measurement of a self-diffusion coefficient exists. Instead of observing the statistical result of the diffusive motion of many particles over a short time, we may observe one particle over a very long time, and then reduce the data by appropriate statistical methods. Thus, we may make

many observations of the motion of a single, identifiable particle interacting with the other unidentifiable particles rather than performing one observation of many, identifiable particles interacting with other particles. The form of the data associated with such measurements of self-diffusion is a list of sequential positions and the times at which these positions are determined.

For the ergodic hypothesis to be valid the time between observations of the labelled particle should be long enough such that the change of position is not correlated with its motion at the last observation. From a simple viewpoint, the change of lateral position should not be due to the lateral velocity of the labelled particle at its last observation.

In a bounded system the simplest form of data reduction occurs when the changes of position do not involve interaction with or are not restricted significantly by the boundaries. Also, for systems with concentration gradients the changes of position should occur in regions of constant concentrations. (Measurements can be made in bounded or concentration-varying systems but in calculating the value of the diffusion coefficient these phenomena must be included.) Thus, in order to avoid effects of the boundaries or concentration gradients on the measurements an upper limit must be placed on the time interval between observations of the position of the identifiable particle.

Measurements of diffusion using the single-particle technique are commonly referred to as random-walk measurements because

of the irregular, seemingly haphazard particle movements and the probabilistic mathematical description. Chandrasekhar [35] provides a very good review of the probabilistic mathematics which underlies the physical diffusion process. His review article provides a derivation of the following relationship between the random-walk measurements and the diffusion coefficients:

$$D = \frac{1}{2N} \sum_{i=1}^N \chi_i^2 \quad (4.1)$$

where

$$\chi_i = Y_{i+1} - Y_i / (t_{i+1} - t_i)^{1/2} \quad (4.1a)$$

Here Y_i is position of the labelled particle at time t_i , and N is the total number of excursions observed. Note that D has the statistical form of a second moment of an appropriately scaled change of position. The distribution of the χ_i 's for a diffusive motion is shown in Chandrasekhar's article to be a normal distribution centered about χ equal to zero. Thus it is possible to obtain a check on the fundamental diffusive nature of the experimental phenomena simply by examining the distribution of the excursions formed from the data.

The single-particle technique is rarely used in the measurement of self diffusion because of the unavailability of a suitable tracer particle. In these experiments the relatively large size of the particles allowed construction of an identifiable tracer

particle with nearly the same diameter and density as the unidentifiable particles. Statistical measurements of this type have been used many times to measure the diffusion coefficient of Brownian motion.

Note that Brownian diffusion is not a self-diffusion since the motion of the Brownian particle is the resultant effect of many collisions between the much smaller surrounding molecules and the particle. Also, note that with the large size of the particles used in these experiments the Brownian motion of any of them is vanishingly small. The particles used in these experiments have a coefficient of Brownian diffusion of order $10^{-15} \text{ cm}^2/\text{s}$ as calculated from the Einstein relationship.

4.2 Methods of Application of the Single-Particle Technique in a Circular-Cylinder Couette Apparatus

For simplicity in data reduction one would prefer to design the experiments such that observations could be made at constant time intervals. Two possible methods, I and II, which would allow radial position measurements of a labelled particle to be taken at constant time intervals are described along with the reasons they were not selected for use. Method III which was used in the experiments, carries the associated liability of variable time intervals between observations.

Method I. An obvious method is to construct a detector which is capable of making a lateral (radial) position measurement at any circumferential location in the cylindrical Couette annulus. This method was rejected because the accuracy of measurement of the radial position is significantly reduced by reason of the fact that it is necessary to observe a very large area.

Method II. Without sacrificing the ability to make measurements at constant time intervals an increase in the accuracy of position measurements can be achieved by any method in which the area of observation is reduced because the labelled particle is kept within a limited zone of detection. This could be done by varying the velocity of the cylinder walls yet keeping their relative velocity, and therefore the shear rate, fixed. Another approach would be to "track" the labelled particle by moving the detector so as to keep the particle under the detector within its limited zone of detection. Such systems were not pursued because the changing convective velocity of the labelled particle would require complicated feedback controls to keep the particle within the zone of detection.

Method III. This method is the one which was chosen and is illustrated schematically in Figure 4.1. The relative positions of the several parts of the detection apparatus and the circular-cylinder Couette device are shown in Figures 4.2, 4.3 and 4.4. In this method the detector is limited to observing in a small

circumferential arc of the Couette annulus, and scans laterally the shear flow directly below it. The particle is observed only when it comes into the arc of detection by reason of its circumferential convective motion. Since the particle changes its circumferential velocity through diffusion, the time between observations is not constant. This technique, because of its dependence upon the convective velocity, does not produce an equal number of observations of the labelled particle at all lateral positions across the shear flow. The frequency of observation at a particular lateral location is partially determined by the convective velocity at that location and partially by the concentration, the wall force, and the nature of the diffusive motion near the wall. In Chapter 7, which deals with numerical experiments of the detection and migrational process, this point is discussed more thoroughly. Even though the experimental time intervals are not uniform, the observations provide both the actual time intervals and the actual position of the diffusing particle and thus lead to a measurement of D according to Eq. 4.1. In this equation the greater absolute displacements which tend to occur when observations are taken at longer time intervals are appropriately scaled by forming the quantity χ_1 .

4.3 Technical Requirements of the Single-particle Technique

To accurately measure the diffusion coefficient there are several technical requirements which must be met by the identifiable particle and the experimental apparatus. They are:

i) The identifiable particle must have the same diffusional motion as the unidentifiable particles. This requires that the identifiable particle have the same geometric shape and moments of inertia as the unidentifiable particles.

ii) The conditions governing the diffusive motion must be constant throughout the course of any experiment. Further, measurements should only be taken in regions of the Couette flow in which the diffusive motion is constant and well defined. This requires, for the Couette flows discussed in Chapter 3, that the measurements be taken in the central region of the Couette channel. In this way measurements are avoided in the wall regions of increased shear rate and a reduced concentration. Thus, the observations must be of the lateral position in the channel and not merely the length and direction of the excursion which has occurred since the last observation.

iii) The observations of lateral position should be reliable and accurate since many types of errors mimic diffusion in that they have an associated normal distribution about the true value of the event. To illustrate, consider the extreme case in which there is a vanishingly small diffusive motion which is to be measured by a very erratic apparatus. In this case the errors in measurements may well have a normal distribution; however, it is possible to determine that the apparent diffusion coefficient is erroneous. The apparent diffusion coefficient will decrease when calculated using longer times between the Y_i 's since their magnitudes, and

therefore those of the Δy_i 's, are determined by the observational technique and not by time. In a bounded system, restriction of the diffusive motion by walls will produce a similar decrease in a calculated diffusion coefficients. It is shown in Appendix III that any inaccuracy in measuring the lateral position statistically accumulates to cause an increase in the calculated diffusion. From a simple view this is a direct result of the squaring of the displacement.

4.4 Bench-Top Tests of Three Models of Labelled Particle and Corresponding Detector

Before completing designs or beginning the construction of an apparatus to measure the lateral position of a labelled particle, bench-top tests were conducted to examine the feasibility of concepts which, on paper, showed promise. In these tests only models of the labelled particle and the detection apparatus were used. No attempts were made during the bench-top tests to manufacture an "ideal" labelled particle or to assemble a detection apparatus suitable for the final experiments. Rather the emphasis was placed on testing the reliability and feasibility of the concept.

During preliminary calculations on many schemes, no feasible method could be found which used a passive identifiable particle, i.e. a particle which does not emit a signal. Detection of a passive particle typically involves measurements of the change in slurry properties caused by the presence of the identifiable particle which

is different in a physical property such as conductivity or color. Detection of passive particles pose problems which are analogous to deciding whether there is a "needle in the haystack."

All three bench-top tests used mock-ups of active particles. Two of the mock-up particles emitted light and the third emitted gamma rays.

4.4.1 Light-Emitting Particles.

Test One. The first test used a mock particle made from radio-luminescent paint such as that used on watch dials. For this test an area of radio luminescent paint on a microscope slide was masked to cross-sectional area equivalent to that of a proposed labelled particle. This mock-up was then presented to the detector. Masks with circular openings having diameters between 0.02 and 0.10 inches were used.

A refrigerated photo multiplier was used to detect the light emitted by the radio luminescent paint. The window allowing light to enter the photo multiplier was restricted to a slit of the same width as the proposed collimating slit which would be used to locate the particle. A cuvette containing a "transparent" slurry was placed between the mock-up of the labelled particle and the photomultiplier. The slurry was made transparent by matching the index of refraction of the suspending fluid to that of the particles via organic additives mixed into the suspending fluid. Thus, in an otherwise dark environment the light input to the photomultiplier simulated the fraction of light from a light-emitting particle in a "transparent"

slurry which would pass through a collimating slit to a photomultiplier. In the proposed detector the position of the light-emitting particle would be determined from the known position of the collimating slit.

The larger-size mock-ups of the light emitting particles produced a marginally acceptable signal but the smaller size particles could not be detected. It probably would have been possible to change to a photon-counting type of detection and use such a light-emitting particle. However, the complications and cost of such detection apparatus are enormous. Moreover, because radio-luminescent paint is much more dense than the suspending fluid, a hollow, spherical-shell construction of the labelled particle would be necessary to make it neutrally-buoyant. Several preliminary attempts to make a neutrally-buoyant particle using non-radioactive luminescent paint were unsuccessful. The manufacturing failures seemed to be due to the low mechanical strength of the paint.

This concept was dropped because of the two basic practical difficulties exposed during the bench-top tests.

Test Two. One of the reasons Test One was unsuccessful is that the amount of light available for detection was too small. In Test Two the amount of light available for detection was increased in two ways:

- 1) The light output of the labelled particle was made higher using a laser to excite a fluorescent compound distributed throughout a plastic sphere. By definition a fluorescent compound is one which

absorbs and emits light at different wavelengths. Thus, in this system there is a difference in wavelengths of the "signal" light emitted by the labelled particle and of the "energizing" light of the laser. The laser light when it strikes the photomultiplier is a form of noise since it carries no information as to the position of the labelled particle. In this system, because of the fluorescent shift of wavelength, it is possible to filter the input to the photomultiplier and thereby remove the noisy light originating from the laser.

ii) There is no need to restrict light input to the photomultiplier via collimating slits since the position of the labelled particle can be related to the position of the laser beam which is well-defined. Specifically, if the laser beam is perpendicular to the lateral and convective directions and if the slurry is truly transparent and non-refracting, then the labelled particle will only emit light when it is within the laser beam. Thus, there is a one-to-one relationship of the position of the laser beam and the position of the labelled particle.

This concept, as that in Test One, also demands a "transparent" slurry in order that the position of the laser beam can be used as an index of the position of the particle. Absorption of light without a loss of image coherence is not per se a bad feature; however, it does require a more powerful laser to achieve a given signal.

Bench top trials of this concept were done using the arrangement of laser, slurry, particle labelled with a fluorescent compound (rhodamine-6G), and photomultiplier with filter as shown in Figure 4.5. The initial trial worked exceedingly well. However, after a few minutes an apparent broadening of the laser beam could be observed. Concurrent with the obvious visual broadening of the output level of signal from the photomultiplier could be observed to decrease. The hypothesized cause for the broadening was an attack, enhanced by the energy available from the laser beam, upon the surfaces of the plastic particles by the organic additives used to match the indices of refraction of the suspending fluid and the plastic particles. Indeed, immersion of the particles into the organic additives did produce a tacky surface on the particles; however, volumetric dissolution did not occur.

The requirements on the "transparent" slurry are very strict-- it must be neutrally buoyant and have matched indices of refraction of the particles and the suspending fluid. Furthermore, it must be insoluble in some relatively inviscid fluid of greater density (at least 50% greater). In addition, it must be stable. Neither, the combination of ingredients in the slurry used by Karnis [17] or that used here, (CCl_4 , CH_2I_2 , Ucon 50HB5100 for the suspending fluid and polyvinyl acetate particles) met the criterion of stability when exposed to the intense irradiation of an argon laser. To the eye, however, these slurries are stable for periods of up to a month.

No other suitable slurry could be found. Consequently this concept was dropped from consideration even though it has great aesthetic appeal.

4.4.2 The Radioactively Labelled Particle.

Test Three. The gamma radiation of radioactive isotopes offers a much better signal than light since it is not significantly affected by the other, unlabelled particles or the suspending fluid. Gamma rays decay by interaction with atoms or molecules; the decay of a gamma ray is a probabilistic event whose chance of occurrence depends upon the distribution of atoms in the path of the gamma ray. Thus the characteristic penetration distance of a gamma ray depends upon the density of the material. For a material with a specific gravity of one g/cm³, fifty percent of the 810 Kev gamma rays (the principal emission of ⁵⁸Co) will pass undegraded through 8.5 centimeters of the material.

In the bench-top tests of a radioactively labelled particle a point source of radioactivity was separated from a gamma-ray detector (scintillation counter) by a collimating-slit viewing window. A top view of the arrangement of material and apparatus used in these bench-top tests is shown in Figure 4.6. The tests were designed to measure the resolution of the lateral position measurements and the necessary level of radioactivity for an actual experiment.

At the highest count of gamma rays registered by the scintillation counter the point source is in direct "line of sight" of

the detector through the collimating-slit viewing window. Since the full thickness of the lead which forms the viewing window will block almost all gamma rays, first-order estimates of the relationship between the count rate and the lateral position are given by the same formulas which describe collimation of a point source of light. However, the statistical nature of gamma-ray decay in matter causes the lead corners of the collimating slit to appear "fuzzy" i.e. the phenomena permit some of the incident gamma rays to pass through the lead. Therefore calculations are impractical as compared with tests on bench-top models. In addition, the efficiency of available radiation detection equipment could be evaluated directly.

A point source of radiation was made for the bench-top tests by sealing a small amount of solution containing ^{58}Co into a glass capillary of 0.010 inches internal diameter. The length of solution in the capillary was less than half an inch. The capillary was presented to the plane of the lead shielding in an end-on direction so as to appear as a point source when viewed by the detector, which was positioned approximately 6 inches away.

Standard three-inch lead cubes were used as shielding between the point source and the scintillation detector. A viewing window was established in the shielding by using plastic shims between the lead cubes. Clamps on the lead blocks maintained an accurate width of viewing window.

Results of the bench-top tests showed that a 0.020 inch diameter particle labelled with 25 mCi of activity should be detectable with an accuracy better than its radius with a distance of 10 inches between the detector and the labelled particle. Moreover, this accuracy was of an absolute nature, in the sense that a larger particle with a 0.020 inch core of radioactivity would be able to be detected to the same distance, i.e. ± 0.010 inches. The position of the radioactive particle would be given by the position of the viewing window, which since it would be out of the slurry could be easily measured.

Discussions with the Nuclear Products Division of 3M Corporation indicated that they would be able to manufacture such a labelled particle. They intended to use a technique which had been developed to make radioactive tracer material (3M Brand Tracer Microspheres) for use in blood circulation studies. Their technique involved an ion-exchange procedure to place the radio nuclide onto a polystyrene matrix. Subsequently the isotope is fixed to the polystyrene by a baking procedure. Thus, for a carrier-free isotope like ^{58}Co , a labelled particle with approximately the same density as normal polystyrene could be produced. It should be realized that there are large changes of scale between the normal particle size and amount of radioactivity customarily used by 3M and the much larger particle size and the much larger amount of radioactivity required to produce a suitable labelled particle for these experiments.

Based upon the success of the bench-top trials and the commercial source of expertise to make a labelled particle a decision was made to contract 3M to make a particle, on a "best-efforts" basis. Simultaneously design of an apparatus to measure the lateral position of a radioactively labelled particle was begun. Results and experimental components of Test Three, that is a scintillation detector, and collimating-slit viewing windows in the lead shielding, were used as fundamental elements in the design.

4.5 Description of the Apparatus to Measure Lateral Position

To use the radioactively labelled particle in the fashion described as Method III the viewing window in the lead shielding must scan a small circumferential arc of the Couette annulus. When the viewing window is over the radioactive particle, the signal rate from the detector is increased; the position of the labelled particle is then determined from the position of the viewing window. With the complication of measuring the signal rate and with a provision for automatically recording the observations the apparatus used to measure the lateral position is essentially that described in Section 4.4.2. Figures 4.2, 4.3, and 4.4 show various views of the component parts of the apparatus in relationship to the Couette flow device. For purposes of clarity of illustration almost all the lead shielding has been deleted from the figures. The only part of the lead shielding which remains is that which contains the viewing window(s), which, in the figures are denoted as part F. Part of

the lead shielding, which has been deleted for purposes of clarity, was placed around the sides of the scintillation detector, A, on the shelf, G. Another section of lead shielding was hung from the shelf and together with the lead wheel of viewing windows provided the "bottom" shielding for the scintillation detector.

The sectional top view, B-B, shows the upper lead wheel with many viewing windows. However, the collimating-slit viewing windows are sufficiently separated such that at any time only one can be over the circumferential arc of detection. Thus, when the lead wheel is rotated, the small arc of the Couette annulus is repeatedly scanned by the viewing windows, one at a time. Rather than make one thick wheel, two half-thickness lead wheels were made, thus allowing the height of collimation to be varied. The importance of alignment between the viewing windows and the Couette apparatus is treated later in this chapter.

The scintillation detector, part A, is placed directly over the Couette annulus. To prevent false observations when the labelled particle is not below the scintillation counter, it is surrounded by lead shielding (which is not shown in the figures). The radioactive particle flows in the slurry and so is restricted to the Couette annulus. Gamma rays emitted by the radioactive particle have a relatively decay-free path to the detector only when the radioactive particle is in direct "sight" of the detector through the viewing window. Thus, when there is a good alignment of the Couette apparatus, C, and the wheel of viewing windows, F, the position of

the radioactive particle is directly related to the position viewing window. For the highest rate of detection of gamma rays the radioactive particle is centered in the viewing window.

An electrical signal is generated in response to the high rate of gamma-ray counts registered by the radiation electronics when a viewing window is directly over the radioactive particle. It is produced by the rate-meter section of the coordinating electronics (described subsequently). Except for spurious electronic noise, when the threshold rate level of the coordinating electronics is properly set, this electrical signal can only occur when a viewing window is over the radioactive particle.

Each viewing window as it scans across the small arc of the Couette annulus allows passage of a light beam from a He-Ne laser, B and causes a second electrical signal related to the position of the viewing window. The light beam is reflected by a mirror, H, through each viewing window to the photodiode, D thereby producing an electrical signal of the Couette annulus just before that viewing window begins to scan the small arc of the Couette annulus. By design, the only path from the laser to the photodiode involves going through a viewing window. Since the position of the laser beam is fixed relative to the apparatus, this signal forms the zero, or reference point for the measurement of the position of the labelled particle.

The electrical signal from the photodiode zeros an electronic stopwatch, and the radiation-induced signal records the time on the

stopwatch. The time measured by the stopwatch, thus corresponds to the time required for the viewing window to move the lateral distance from the laser beam to the radioactive particle. Due to its mass, the lead wheel of viewing windows is very easily maintained at a constant speed of rotation. Thus, by measuring the time between two electrical signals produced in conjunction with the motion a viewing window, the lateral position of the radioactive particle is measured as a time. Physically the recorded time of the stopwatch is equal to lateral distance from the laser beam to the particle divided by the velocity of the viewing window.

Note that the measurements of the lateral particle position are relative to the position of the laser beam. Knowledge of locations of the walls of the Couette walls relative to the laser beam allows interpretation of the measurements as positions within the Couette channel.

4.6 Component Parts of the Apparatus to Measure Lateral Position

Each of the component parts of the apparatus used to measure the lateral position is now discussed in more detail. Where relevant, physical variables which aid in an understanding of the operation are provided.

4.6.1 Radioactively Labelled Particles. During the course of the thesis several radioactive particles were constructed. An underlying guideline during the construction of any radioactive particle was that the finished radioactive particle must have the same diffusional behavior as the unlabelled particles. Since the diffusional motion originates in the fluid-mechanically, mutually-induced motions of the slurry flow, this requires that the labelled particle must have the same specific gravity, shape, and moments of inertia as the unlabelled particles.

The specific gravity of available suspending fluids and unlabelled particles limits the possible values of specific gravity to a value near one g/cm^3 . Thus, since most radioisotopes have a density which is much greater than unity, the selection of a labelling isotope is limited to those which are carrier-free (i.e., consist only of the radioactive species) and are of high specific activity. In this way the labelling isotope contributes only a minute amount to increasing the apparent specific gravity of the labelled particle. The half-life of the isotope, the cost and available methods of attaching the radioisotope to the base particle limits the selection to ^{58}Co .

Two methods of incorporating the ^{58}Co into labelled particles were used. A characterization of each method is included with the description of a particle using that method. Particle One was used in the large-particle experiments; Particle Two was used in the small-particle experiments.

Particle One. The Nuclear Products Division of the 3M Co. was contracted on a "best efforts" basis to produce several particles of 0.020" diameter (approximate size) using an ion-exchange process to affix ⁵⁸Co to Dowex beads.* Due to unanticipated problems this process failed to incorporate a sufficient amount of radioactivity per bead. These problems were most likely a result of the greatly increased scales of activities and particle sizes, compared to 3M's normal production process. However, after several trial attempts 3M did produce seven beads of approximately two mCi activity each. These beads were returned to us.

Since these beads were of no use individually, we placed them inside a hollowed-out 0.125 inch diameter particle of the same type as the unlabelled particles used in the large-particle slurries. The fourteen mCi total activity proved sufficient to perform reliable detections. A small length of copper wire was also included to provide a rough match to the specific gravity of the slurry. The necessary amount of copper wire was estimated using a chemical balance. A low estimate of the amount of copper was made so that weight could be added to achieve a final match of specific gravity between the suspending fluid of the slurry and the radioactive particle. A thin layer of red paint was used to achieve a final matching of the specific gravity.

* This process is used by 3M to produce, on a regular commercial basis, tracer particles with diameters of 5 to 100μ for use in the study of blood circulation. In this product ⁶⁰Co, because of its low cost, it used.

This labelled particle was used in all of the experiments performed with large-particle slurries. During these experiments the labelled particle could occasionally be seen near the outer Couette cylinder due to the red paint. It would appear at all heights within the slurry region in the Couette apparatus and seemed to have no unusual characteristics as compared with the unlabelled particles. Furthermore, visual observations of the lateral position of the labelled particle, when it was near the top of the slurry in the Couette apparatus, agreed with the output of the detection apparatus.

Particle Two. In an effort to make a very small labelled particle, a second method in which ^{58}Co is electroplated onto a copper-clad ABS (acrylonitrile-butadiene-styrene) plastic sphere was developed by the author. This method produces an electroplated surface which is fragile. Therefore, the sphere is subsequently coated with epoxy to prevent radioactive contamination of the slurry. The matching of specific gravity is achieved by adding hollow glass microspheres to the epoxy coat. Details of the entire procedure, which is quite lengthy, tedious, and demanding, and which is fraught with a high risk of failure, appear in Appendix I.

Two attempts were made to produce a labelled particle with a diameter in the range of the small-particle slurry, i.e. 0.038 to 0.045 inch diameter. Neither attempt was totally successful. The first attempt was a diametric failure in which the resultant

particle was 0.090 inches diameter when neutrally buoyant. The second attempt produced a radioactive particle which, when neutrally buoyant was 0.052 inches average diameter. The radioactivity which was plated onto the copper-clad ABS sphere could not be sealed onto this particle with the first coat of epoxy. Accordingly, a second layer of epoxy was applied to seal the particle; this layer, though very thin, made the particle quite heavy relative to the suspending fluid. The specific gravity of the radioactive particle was reduced by attaching a layer of small hollow glass spheres (Ecospheres by Emerson-Cummins) with polystyrene cement. This second epoxy coating and the balancing glass spheres made the particle larger than desirable. However, it was used to obtain the experimental data presented here as small-particle slurries.

Furthermore, the coat of hollow glass beads on the epoxy-sealed radioactive particle came off several times during the small-particle experiments. At these times the labelled particle was removed from the Couette apparatus and its specific gravity readjusted to match the slurry. When the coat of hollow glass beads came off, it did so in a catastrophic fashion thereby causing the particle to sink quickly. Through close observations of the lowest region of the slurry, it was possible to determine when the buoyant coat was lost. Thus perserverance and close observation allowed collection of data during the times when the radioactive particle was neutrally buoyant.

4.6.2 Collimating-Slit Viewing Windows. A wheel of collimating-slit viewing windows was constructed from 30 cast lead blocks attached to a central core structure. Each collimating slit was formed by the adjoining faces of two lead blocks. Figure 4.7 shows details of this arrangement. One face of each block had a two-inch long protrusion with a 6 inch radius of curvature. The other, opposite face of each block had a two-inch long, 1/16 inch recessed intrusion with a six inch radius of curvature. Thus, when two blocks were put together a collimating-slit viewing window with a two inches length and 1/16 inch width and a six inch radius of curvature was formed. The six-inch radius of curvature matched the average radius of the Couette annular gap.

On each block there was a 12° angle between the two faces which formed the collimating slit. When the blocks were assembled they formed a segmented ring. The blocks were assembled and shimmed such that all viewing windows had parallel axes of sight. After assembly, the ring was cut perpendicular to its axis to form two identical halves. The halves were very carefully aligned and mounted onto a core structure. The core structure was designed to hold the rings relative to each other and to provide a central axle. Great care was taken during installation of the axle to insure that the axis of the axle and the axes of sight of the viewing windows were parallel. The amount of collimation could be varied by changing the spacing of the halves of the segmented ring on the axle.

The core structure also served as a portion of the radiation shielding for the scintillation detector. The four-inch total thickness of the lead blocks which form the wheel of viewing windows will degrade more than 99.9% of the incident 810 Kev photons, which is the major emission of ^{58}Co .

For experiments with the large-particle slurries a distance between the top of the upper wheel and bottom of the lower wheel was 7.0 inches. In the case of the large particles the width of the collimating-slit viewing window is equal to the radius of a particle. Thus, the corner-to-opposite-corner viewing angle produced a maximum deviation in the lowest portion of the slurry (14 inches from the detector) of approximately a particle radius.

For the experiments with the small particle slurries the height of collimation, i.e., the top-to-bottom distance was 4.5 inches. In addition, a sheet of 1/32" lead was inserted into the viewing windows of the upper wheel. Using the direction is given by the direction of rotation, this lead sheet was on the trailing side of the viewing window as is shown in Figure 4.10. This has appreciably the same effect as if both the upper and lower halves of the viewing windows were half blocked since the "sighting" of the radioactive particle is performed using the rise in rate of gamma-ray counts rather than the absolute maximum of the count rate or of the incident energy.

During the large particle experiments the two halves of the wheel of collimating-slit viewing windows were separated by 3 inches.

Thus, it was possible to have a portion of the lead shielding for the scintillation detector which protruded into the region between the halves of the wheel of viewing windows, as is shown in Figure 4.8. This method of shielding was very effective when the particle was in the region below the detector but was not directly in sight through a viewing window.

For the small-particle experiments with the half inch of space between the two halves a much thinner lead lip protruding between the halves of the lead wheel would have provided an insufficient shielding. The "noisy" radiation would reach the scintillation detector primarily because of the outer polygonal surface of the segmented ring. The flats on the ring would offer a path with a rather large cross-sectional area, compared to a viewing window. The outer cylindrical surfaces of the two halves of the polygonal wheel were machined between the end of the large particle experiments and the beginning of the small particle experiments. This resulted in good fit between the wheel and the shielding and therefore a reasonable level of background radiation. However, during this machining process a misalignment of the lead blocks occurred. Thus, the increased accuracy associated with restricted viewing windows was traded off against the relative loss of alignment of the viewing windows. In the following section which deals with operational considerations the effect of alignment of the viewing windows is discussed at length. Furthermore, section 4.7 contains a description of the calibration procedure during which the accuracy

of the detector, including a loss of accuracy due to alignment, is shown in quantitative terms.

A slurry depth of 4 inches was used in the small particle experiments. For the collimation height and the slurry depth the associated deviation caused by the corner-to-corner sighting through the viewing window was approximately 0.03 inches in the forward direction. However, because of the loss of alignment, this can not be directly equated to a level of accuracy.

4.6.3 Radiation Detection Equipment. The detection of the gamma rays produced by the decay of ^{58}Co was accomplished using a scintillation detector (Harshaw "Integral-Line" Type 7SF812) and a spectrometer (Baird-Atomic Model 530A). This type of radiation detection equipment was chosen because it produces a discrete output pulse for each gamma ray detected. The method of operation of the equipment is standard; Price [36] offers an excellent explanation of events during a detection and review of the electronics of the detector.

An important part of the spectrometer is the energy analyzer which can filter out pulses caused by gamma rays which are outside of a set range of energies. The energy analyzer was used during these experiments to eliminate the lower-energy gamma rays which are re-emitted whenever a gamma ray interacts with matter (such as the lead shielding). Specifically, the range of the energy analyzer set to encompass the major emission band of ^{58}Co , i.e. 810 Kev. A lower

"noise-level" is achieved through the use of an energy analyzer since it filters out the lower energy gamma rays which result from interaction with the lead shielding from the "signal" gamma rays from the radioactive particle. Although the ambient background radiation was small compared to the radiation emitted by the ^{58}Co in the labelled particle, the energy analyzer also removes most of this radiation since it has a very broad spectrum.

4.6.4 Coordinating Electronics. Several necessary components of standard electronic design were not conveniently available. Therefore they were constructed from TTL (transistor-transistor-logic) integrated circuits. These components were a variable time-base rate meter, two electronic stopwatches with digital output and a signal-activated interface circuit to record data on a teletype. The data consisted of two times. They are the time for the viewing window to move from the laser beam to the radioactive particle and the time interval since the last observation. These time intervals were measured by the electronic stopwatches.

Rate Meter. The input signal to the rate meter was the square wave pulse produced by the Baird Atomic spectrometer. Each time a gamma ray within the preset energy range was detected the spectrophotometer produced a pulse. These pulses were accumulated on a 3-digit counter, which was reset to zero at the end of a set time interval. The time interval used for these experiments was five milliseconds. If the counter registered a number greater than a

threshold level before the end of the five millisecond interval then the rate meter generated a square wave pulse. For correct threshold levels this pulse occurs only when the viewing window is over the labelled particle. This pulse was used to stop the electronic stopwatches and also to initiate the automatic recording of the times on the teletype. To provide flexibility the design of the rate meter included provisions for variation of the threshold level and the time-interval.

Stopwatches. Two electronic "stopwatches" were constructed as a portion of the TTL circuitry. Each stopwatch consisted of two main parts, a counter and a memory. To measure time intervals the stopwatches counted the wave forms of a pulse wave of known frequency which originated from a 6 MHz ($\pm .001\%$ accuracy) crystal oscillator. Frequency-division circuits were used to provide pulse waves of lower frequency.

To reset the stopwatch the number registered on the counter was set to zero. Under most circumstances the memory contained the number registered on the counter. The exception to this occurred in the recording of the time on a stopwatch. When the stopwatch was stopped to make an observation of the labelled particle the number registered in the counter was "latched" onto the memory. A time-dependent interlock was set to prevent the contents of the memory from being altered until the times in the memories of both stopwatches had been recorded by the teletype. The interlock, however, did not

prevent the counter section of the stopwatch from being reset to zero. Thus, the stopwatch could begin to time the next event while the memory held the record of the last. At the rates at which observations were made in these experiments, the number in the memory was always recorded by the teletype prior to the next observation.

As mentioned above one stopwatch was used to measure the time for the viewing window to move from the position of laser photodiode to a position above the labelled particle. This stopwatch is hereafter referred to as the position stopwatch. The other stopwatch was used to measure the time interval between observations of the position of the labelled particle and is referred to as the interval stopwatch.

The position stopwatch is reset to zero by the photodiode each time a viewing window allowed passage of the laser beam to the photodiode just before the viewing window begins to "scan" the arc of the Couette annulus. Since each reset involved using the viewing window about to pass over the Couette gap, the distance between individual viewing windows does not affect the accuracy of the measurements. The frequency counted by the position stopwatch is 10 KHz. However, the limit on accuracy of position measurements is the rate meter which produces the signal to stop the watch. The time over which the rate meter collects gamma ray counts in order to surpass a threshold is 5 ms. This interval or some portion of it is the fundamental limit on the accuracy of position measurements. The time

for the viewing window to move from wall to wall was approximately 200 ms. Thus the maximum position resolution of the system is approximately the ratio of 5 ms to 200 ms, 0.025.

The time-interval stopwatch was reset to zero and restarted immediately after the number registered on the counter was latched onto the memory. Thus, only the response time of the electronic circuitry was lost from the measurement of the time interval between observations. The response time was of order 100 nanoseconds; as the smallest time interval was three seconds this introduced almost no error. The frequency of the pulse wave counted by the interval stopwatch was 10^6 Hz. This forms the major limit on the accuracy of the measurements of the time interval between observations; it is ± 0.01 sec.

Interface Circuitry. The remaining portion of the coordinating electronics was interface circuitry which integrated the functions of the previously discussed electronic components with the teletype to provide automatic recording of observations. The rate-meter signal, which is generated when the level of radiation surpassed the threshold level, simultaneously sets a dead-time latch, an interlock, and records the times of the stopwatches on their memory sections. This signal also activates a code-conversion section which translates the internal electronic signals to a form which allows production of a paper-tape record by a teletype (Model ASR-33 by Teletype Corp.).

The interlock prevented the numbers recorded on the stop-watch memories from being changed while the teletype produced a paper-tape record of them. The interlock was activated for 2.4 seconds, i.e. the time required for the teletype to produce a record. Since the minimum time for the convective motion of the labelled particle to return to the zone of detection was greater than 4 seconds, the memory-teletype interlock did not prevent an observation of the labelled particle each time it passed through the zone of detection. It will, however, prevent any outside electrical noise from causing an erroneous record of the observations.

For experimental conditions in which not every consecutive, possible observation was desired, a dead-time latch was constructed as a portion of the interface circuitry. Each time an observation was made the latch was set and a timer was started. While set, the dead-time latch prevented recording of the stopwatches by (further) signals from the rate-meter. When the timer reached a pre-set value the dead-time latch was released thus allowing the next signal from the rate meter to record an observation.

The length of time before the timer would release the dead-time latch was selected according to experimental conditions. This value is set onto the coordinating electronics by the operation. For experiments using the large-particle slurries and having an ω of 10^{-5} , a 4 second dead-time was used. This was short enough such that the dead-time latch did not prevent any observations. At the

lower shear rate of 1 s^{-1} , the large-particle experiments were performed using a dead-time of 40 seconds. This also was shorter than the time required for the labelled particle to complete a "lap" around the Couette apparatus, and thus did not prevent any observations. In the experiments with small-particle slurries a dead time of 30 seconds was used to prevent some observations.

In experimental trials of measurements for the small-particle slurry, the use of short dead times showed that the observations which were suppressed with the 30 second dead-time were generally observations which produced extremely small changes of position of the labelled particle. Use of this data would produce diffusion coefficient with large errors because the changes of positions of the labelled particle would be small compared to the accuracy of the detector in measuring position. As is explained more fully in Chapter 6, when the data is reduced the observations actually used to calculate the diffusion coefficient would be taken at longer time intervals obtained by summing the time intervals of the actual data. The longer time intervals have larger average changes of position and, therefore, relatively smaller errors compared to the accuracy of particle-position measurements. Thus, the 30 second dead-time reduced the time involved in data reduction by eliminating the necessity to process many intermediate observations.

4.7 Operating Considerations and Accuracy of the Detector.

For any given radioactive particle there are two major factors which limit the accuracy of the apparatus to measure the lateral position. One is the threshold level at which the rate meter stops the stopwatches and records their times. The threshold level is the rate of radiation incident to the scintillation counter which causes the rate meter to "declare" the viewing window is positioned over the labelled particle. The second factor is the degree of alignment of the viewing axis of the wheel of collimating slits and the vertical axis of the Couette apparatus. These are treated in the first two parts of this section. The "whole-system" calibrations which measure the accuracy of the detection apparatus are reported in the last portion of this section. Prior to these sections a more thorough presentation of the shielding effects of a scanning collimating-slit viewing window is provided.

Discussion of Collimation by a Viewing Window. The relationship between the arrival rate of gamma rays at the detector and the relative positions of the radioactive particle and the collimating slit is complex. As the collimating slit scans over the radioactive particle the arrival rate of photons gradually increases, comes to a maximum and then gradually decreases. The gradual increase and decrease are due to areas of partial sight of the collimating slit as illustrated in Figure 4.9. Consider the effects of a scanning viewing window upon the arrival rate of photons at a light detector from a point source of light which represents the radioactive particle.

The source emits photons with equal probability in all directions. The first photon from the point source which the detector would see through the viewing window will be the one which barely misses both the closest bottom edge and the furthest top edge of the viewing window. As the relative lateral distance between the axis of the viewing window and the point source decreases more photons will reach the detector. At the point where the point source is directly under one face of the viewing window, the photon flux will be near a maximum. For all positions directly under the opening of the viewing window an approximately identical number of photons will reach the detector. The rate of arrival of photons will decrease by the reverse of the above process as the trailing face of the viewing window passes over the point source and the viewing window begins to scan away from the point source. The amplitude of the maximum will be inversely related to the square of the distance between the point source and the detector. It will be proportional to the area of the detector exposed by the collimating slit to the radiation. The length over which the point source will move during the gradual increase or decrease of photon arrival rate at the detector is proportional to the ratio of the distance of the point source below the collimating slit to the height of the collimating slit and directly proportional to the width of the collimating slit. For the case of these experiments, the radioactive particle is not a point compared to the width of the slit and the edges of the collimating slit do not block all

gamma rays striking them. Therefore, these relationships are good only for scaling experiments from models. Prior to construction of the experimental apparatus as described in section 4.4.2 the concept of collimating slits for accurate position measurement was tested in a static bench-top experiment using a small amount of radioactivity (300 μCi) and two lead blocks to collimate the radiation.

From these scaling considerations it is apparent that the threshold level at which the particle is declared to be under the slit must be set for conditions representative of the radioactive particle in the deepest region of the Couette annulus. It is also apparent that there will be some depth-dependent variation of the position of the collimating slit when the threshold is surpassed. The effect of these variations is accounted for as a system by the calibration procedure which is described lastly in this Chapter.

Effects of Partially-Blocking a Viewing Window. To perform the measurements of the diffusion of the small particles the upper half of the wheel of collimating-slit viewing windows was partially blocked by placing a 1/32 inch sheet of lead against the concave face of the collimating slit. This lowers the total energy which can reach the detector since it reduces the area of the collimating slit and creates unequal zones of partial sight as shown in Figure 4.10. The wheel of collimating-slit viewing windows was rotated so that the small zone of partial sight would scan across the Couette channel first. Since the detector triggers when the threshold is

crossed, more accuracy in the measurements would be expected. As previously mentioned, this was not realized because of the misalignment of the axes of sight of the viewing windows which occurred as a result of machining operation.

4.7.1 Determination of the Threshold Value. The rate of arrival of photons at the scintillation detector is a convenient measure of the amount of radioactive material in the viewing window because of the quantum nature of radioactive decay. The energy analyzer allows the detections of lower or higher energy photons to be eliminated from the signal which is used as the input to the rate meter.

Ideally, we wish to detect the labelled particle each time it passes through the limited region of detection. This involves inspecting many lateral locations within the small arc of the Couette apparatus which forms the limited region of detection. Thus, because of requirements imposed by the motion of the labelled particle and the need to measure accurately the location of the particle position at a known time, the time interval over which counts (gamma rays) are collected to form the rate measurement must be short. This requires that the detector work reliably with a small average number of counts per time interval. Since radioactive decay occurs in a random manner, the actual number of counts per time interval is a random event selected from a distribution. For count rates of moderate size the distribution is approximately Gaussian and hence the standard

deviation is proportional to the square root of the average count rate. Thus count rates from a distribution with a large average will be percentage-wise closer to the average than count rates from a distribution with a smaller average.

Note that for small average count rates the standard deviation is such a large portion of the average that measurements make little sense. Consider, for example, an average count rate of 3 counts per time interval.* Statistically any event within two standard deviations of the average will be relatively common occurrences. Accordingly count rates of 1 to 5 counts per time interval would have to be considered as characteristic count rates. Since the background radiation can cause a count rate of 1 count per time interval in any size time interval, an average count rate this low is unsuitable for a detection criteria.

Clearly an increase in the level of activity is one answer; however, if the background radiation count rate has a fractional value, then an increase in the size of the time interval is another answer. To illustrate, consider a situation in which the background count rate is 0.2 counts per time interval and the average count rate is, as above, three counts per time interval when the particle is under the viewing window. Then, by using a new time interval

* Actually, with count rates this low, Poisson statistics are the correct method. However, for purposes of simplicity of illustration, the Gaussian statistics adequately illustrate the principle.

which is three times longer, one would have average count rate levels of 0.6 and 9 counts per (longer) time interval. A detection criteria of 3 counts per time interval would now be reasonable since it would be two standard deviations below 9 counts per time interval and more than two standard deviations above the background count rate. Simply by changing the time interval it is possible to obviate the quantum nature of the background radiation.

Value of the Threshold Level. As previously mentioned incorporation of the radioactive cobalt into the beads in these experiments was a troublesome problem. Even without the limit imposed by the procedure a limit on the amount of radioactivity imposed by considerations of safety and cost would soon be reached. A typical activity of the marked particle used in the experiments reported here was 15 mCi. This activity produced an average rate of "signal" gamma rays of 20 counts/5 ms. A threshold for sighting the particle was set at 8 counts/5 ms which is approximately 3 standard deviations below the average. The background radiation level was on the order of 1 count/15 ms.

Use of a wheel of collimating-slit viewing windows was chosen since it provides a simple method to scan repeatedly the zone of detection by rotation of the wheel. The scanning speed of the viewing window due to rotation of the wheel is slow enough such that several separate measurements of the count rate can be performed in the time during which the slit moves a distance equal to the diameter

of a particle. The circumferential length of the collimating-slit viewing window is such that, for conditions at which the experiments were conducted, no convective velocity within the slurry is large enough to sweep the labelled particle through the zone of detection without the viewing window having been aligned over the labelled particle. Consequently the apparatus is capable of locating and measuring the lateral position of the labelled particle each time it passes through the zone of detection. A typical scanning speed of the collimating-slit viewing window was 12.7 cm/sec.

4.7.2 Alignment of the Axes of the Couette Apparatus and the Viewing Windows. To accurately measure the position of the particle, a high degree of alignment of the axes of the collimating-slit viewing windows relative to one another and to the axis of the Couette apparatus is necessary. As an illustration of this consider a measurement of the lateral position of the labelled particle when it is located in the zone of detection and not moving in any direction. A viewing window will pass over the laser beam and zero the position stopwatch. The rotation of the wheel of viewing windows will bring this viewing window to a position over the labelled particle. Here the rate of radiation received by the detector is sufficient for the rate-meter to issue a signal thereby recording the time on the position stopwatch. For purposes of this example, suppose this rate-meter signal always occurs when the labelled particle is directly in the line of sight of the collimating slit, i.e. on its axis.

(Variations from this condition will be treated as an error and are measured during the calibration procedure which is treated later in this section.) Now suppose that this viewing window, instead of having a line of sight parallel to the walls of the Couette apparatus, has a line of sight which is tilted into the direction of rotation of the wheel of collimating-slit viewing windows (see Figure 4.11). Then, this tilted viewing window will record the time on the stopwatch sooner than an aligned viewing window, i.e. one with a line of sight parallel to the walls of the Couette apparatus. This is shown in the illustrations of Figure 4.11. A variety of axes of sight of the collimating slits would produce a variety of position measurements for the location of a stationary particle. Clearly, the viewing windows must have a common axis of sight for a reliable detector.

The axes of the viewing windows must also be parallel to the walls of the Couette apparatus. The viewing window which had a line of sight tilted toward the direction of rotation will serve to illustrate this point. Suppose that viewing window recorded a position measurement for a particle near the top of the Couette fluid region. As mentioned above the time (position) measurement would be smaller than a time (position) measurement made with a viewing window whose line is parallel to the walls of the Couette apparatus. Now imagine, as shown in Figure 4.11 that the labelled particle is deeper in the slurry but at the identical spacing from the walls of the Couette apparatus. When the measurement is repeated with the same tilted viewing window an even smaller time (position) will be measured in

this case. The only way in which lateral-position measurements can be made independent of vertical position in the slurry is for the axes of the viewing windows to be parallel to the walls of the Couette apparatus and to each other.

Careful design and construction combined with optical checks on alignment of the components minimized the alignment contributions to the error. Any residual misalignment in the experimental apparatus was accounted for during the calibration measurements, which are discussed in detail next. The calibration achieved a unified measure of the error of the total scheme of detection by performing repeated measurements of the labelled particle while it was affixed to a wall.

4.7.3 Accuracy and Calibration of the Complete Detector

System. The detection apparatus was separately tested as a complete system to provide an estimate of the repeatability and accuracy of the measurements. During the tests the radioactively labelled particle was attached to a position on the wall of one of the Couette cylinders. Four calibration positions, at the top and bottom of the slurry region, and on both walls, were used. The outer cylinder wall was rotated during calibration at a speed which would approximate the most difficult experimental conditions. The inner cylinder wall was not rotated during the calibrations. Many observations were taken of the particle in each fixed position on the wall. The mean of the distribution was used as the value of the position of the particle at the wall and the width of the distribution was used as an indication of the accuracy of sighting the location of a particle.

In Figures 4.12 and 4.13 the distributions are shown for the large and small particle, respectively. For the large particle the position as detected was within plus or minus one particle radius 95% of the time. The accuracy of position determination for the small particle was much less; it was located by the detector within plus or minus two particle diameters of the true position 80% of the time. Although the observed position distribution of the small particle is much broader than that for the large particle; it nevertheless is still centrally distributed. This result is attributed to the misalignment of the individual collimating slits which was caused by the machining of the outer surface of the lead wheel as previously discussed.

A comparison of the sightings at the top and bottom of the Couette apparatus show the mean values to be nearly equal which indicates a good general alignment. In both figures the distribution of sightings taken further from the detector is broader than the distribution taken nearer the detector. This is partially caused by the zones of partial sight and the r^2 loss of photons. A compensating factor reducing the difference between the top and bottom positions may be that at the top where the zone of partial sight is small, the loss of definition caused by the statistical blocking of photons by lead is large both because of the geometry and greater number of photons per steradian. The opposite case is true for the situation where the radioactive particle is further from the detector.

In addition to providing a measure of the error which occurs in making a sighting of the radioactive particle, the calibration also provided values for the positions of the walls in relation to the laser and photodiode. When the data were reduced these absolute values of wall position made it possible to evaluate the diffusion coefficient using only changes of location which originated in the central portion of the Couette gap. This is discussed in detail in Chapter 6.

5. PROCEDURES USED TO COLLECT AND REDUCE THE DATA

In the first section of this chapter the procedures are described for collecting the many observations necessary to measure D . In the second section the computer program for evaluating D from the observations is described in outline form. An understanding of the logic built into the computer program provides a physical view of the "minimum time interval" and allows a better appreciation of the time-dependence of the errors, as discussed in Chapter 6.

5.1 Procedures Used to Collect the Data

Initial Preparation. The Couette chamber of the experimental apparatus was cleaned prior to the experiments. A small amount of mercury was placed in the bottom of the Couette cylinder. Because of its high surface tension, the mercury provided a leak-proof seal at the junction of the shaft of the inner cylinder and the oil seal in the bottom plate of the outer Couette cylinder.

Next a totally fluorinated hydrocarbon, Fluorinert (manufactured by 3M Co.), was placed in the Couette chamber, typically to a height of 4.5 inches. As discussed in Chapter 3, the relatively low viscosity of this dense liquid greatly reduces the secondary flows which would normally occur in a cylindrical Couette apparatus.

Since Fluorinert is rather volatile a thin layer of the suspending fluid of the slurry, Ucon 50-HB-5100 (manufactured by Union Carbide), was placed upon the Fluorinert.

The supporting feet on the steel frame were adjusted to bring the axis of the Couette apparatus into a vertical position. The approach of the Couette apparatus to a vertical position was judged by measuring the height of Fluorinert at various circumferential locations. A uniform final height of Fluorinert in the Couette chamber to a level ± 0.005 inch was achieved. The outer cylinder was rotated relative to the inner cylinder to ascertain that this rotation did not influence the uniformity of height. No changes in height due to the different relative positions of the cylinders could be measured using a vernier calipers.

Channel Width. A micrometer and a hole gauge were used to measure the width of the Couette channel at several circumferential locations and for several relative angular positions of the Couette cylinders. The average of this series of measurements was 1.053 inches. All measurements taken were within ± 0.002 of the average. The accuracy of the Couette channel in such a relatively large apparatus is a tribute to the craftsmanship of the machinists who built the experimental apparatus.

Apparent Concentration of the Slurry. Whenever the slurry in the Couette apparatus was changed the Couette chamber was cleaned and the Fluorinert level was restored. The slurry was added to the

Couette apparatus as two components. First, the suspending fluid was placed on the Fluorinert, then the particles were added. Rather than measure the individual volumes of the two materials, the heights of the suspending fluid alone and then of the slurry were measured while they were in the Couette apparatus. Because of the accurate construction of the Couette chamber, these heights are proportional to the volumes. Time was allowed for the viscous suspending fluid and the slurry to become uniformly distributed around the Couette annulus before height measurements were made. This height was very easy to measure since the menisci at the two interfaces of the suspending fluid were small. The teflon coatings on the Couette walls are probably the major reason for these small menisci.

With a fixed height of the suspending fluid, the total height of slurry necessary to produce a particular concentration was calculated from the formula

$$[\text{total height}] = \left[\frac{1}{1-\phi} \right] \cdot [\text{suspending-fluid height}] \quad (5.1)$$

where ϕ is the volumetric concentration. Particles were added until this desired height was reached.

When the Couette apparatus is filled using a pre-mixed slurry, unequal residual volumes of particles and suspending fluid are typically left in a volumetric container. This method of forming the slurry eliminates the inaccuracy caused by the unequal residual volumes.

The slurry was mixed by running the Couette apparatus for several hours. It is worth noting that the mixing, diffusive motions are sufficiently strong so that within minutes, the slurries were uniform to the eye.

Apparent Shear Rate. The speed controls on the motors which drive the Couette cylinders were set to values which would produce the desired shear rate for a particular run. Angular velocities of the Couette cylinders were typically determined from stopwatch measurements of the time necessary for ten revolutions of a cylinder. During the course of the early experiments, measurements of cylinder angular velocities were taken at intervals of approximately an hour. These measurements showed no changes in cylinder velocity, and hence for the later experiments, cylinder velocities were measured only at the beginning, middle and end of the experimental run.

Using the radii of the cylinders, the angular velocities were converted to cylinder velocities, V_{outer} and V_{inner} . The apparent shear rate, ω , is calculated from the cylinder velocities and the channel width, w , using the following equation:

$$\omega = \left| \frac{V_{\text{outer}} - V_{\text{inner}}}{w} \right| . \quad (5.2)$$

Speed of the Lead Wheel of Viewing Windows. The angular rate of rotation of the wheel of viewing windows was measured in a fashion similar to the angular velocity of a Couette cylinder. Using the

average radius of a viewing window, i.e. 5.50 inches, the velocity could be calculated for later use in determining the position of the labelled particle. A typical velocity of the viewing window was 4.90 in/s.

5.1.2 Order of Events During an Experimental Run. Before taking data for a particular experimental condition, the following preliminary steps were carried out:

i) A slurry of the desired concentration was mixed in the Couette chamber, as previously discussed. The radioactively labelled particle was placed in the slurry. This slurry was circulated in the Couette device for a period of at least an hour before beginning to collect observations of position of the labelled particle.

ii) A paper tape was loaded into the teletype. The date, apparent concentration of the slurry, the particle size, and the apparent shear rate were recorded onto the leading section of the tape. The teletype was left set to "local" i.e. made unresponsive to the coordinating electronics.

iii) A "dead time" was set on the coordinating electronics. The "dead times" used in the large-particle experiments were 4 s for $\omega = 10 \text{ s}^{-1}$; 40 s for $\omega = 1 \text{ s}^{-1}$ and $\omega = 0.4 \text{ s}^{-1}$. A "dead time" of 30 s was used in the small-particle experiments.

iv) The threshold radiation level was set previously; this threshold level determines the precision of the lateral position measurements. Before experiments with either labelled particle was

attempted a series of wall sightings (see Chapter 4) were performed using that labelled particle. At the time of the wall sightings, which are explained in Chapter 4, measurements of the sighting error were made for several values of the threshold level. For each labelled particle the threshold level of the experiments using that particle was the level which produced the least sighting error. Over the course of experiments reported here, it was unnecessary to adjust the threshold level.

v) The electrical power for all the electronics apparatus was turned on.

vi) The speeds of the Couette cylinders and lead wheel of viewing window were set and measured as previously discussed.

vii) A check was made to ascertain the proper operation of the laser-photo diode reset for the viewing windows. This is due primarily to see that no stray material is blocking a viewing-window thereby preventing the laser beam from resetting the electronic position stopwatch.

viii) The teletype was set to "remote" which allowed measurements to be recorded by the coordinating electronics.

From this stage on, the collection of data was automatic. However, using a mirror to avoid radiation exposure, the slurry flow behind the lead shield could be observed. This was done both to ascertain that the slurry was uniform and to verify the proper operation of the detection apparatus. Verifications of the proper operation were possible whenever the labelled particle could be observed

near the outer wall. Since its lateral motion was relatively slow compared to its circumferential motion this visual sighting near the outer wall provided a method of checking the position as reported by the teletype.

Data were collected for periods ranging from 10 to 60 hours. During the middle of most experimental runs, it was necessary to install additional paper tape. This caused the loss of some of the consecutive sightings. Such losses were noted on the tape and a non-sense number was manually entered as the first event of the new paper tape, signifying to the computer that the time interval between sightings was not known. This is covered more thoroughly in the subsequent section.

For the experiments with small particles, in which the labelled particle would on occasion lose its coating catastrophically, a continuous survey of the Fluorinert-slurry interface was necessary. In this way data collection could be stopped when the labelled particle reached the interface, thus avoiding the collection of false data. In such cases, the particle was retrieved, recoated and the experiment started again.

5.2 Calculation of the Diffusion Coefficient

The raw data at the end of an experimental run consisted of several papertapes each of which contained several hundred consecutive observations of the labelled particle separated by "control"

symbols. Each observation as recorded on the paper tape contained two time intervals.

As previously discussed, the first time interval is the time necessary for the viewing window to pass from the station of the laser and photodiode to its position over the labelled particle. It determines the lateral location of the particle in the Couette gap.

The second time interval is the time which has elapsed since the previous observation of the labelled particle.

In addition to their necessary functions for the proper operation of the teletype, the "control" symbols separated the series of observations recorded on paper tape by either the computer or the tape-to-card converter.

To be processed by the computer program which is outlined below, the experimental observations had to be read into file storage within the computer. In most of the experiments, the punched paper tape was first converted into punched cards by a tape-to-card interpreter. Each punched card produced by the interpreter contained one observation. For the last series of experiments the experimental data were read directly into computer storage using a paper-tape reader. In both cases, at the beginning of each paper tape a non-sensical observation was used as a code to instruct the computer that a new paper tape was about to be read.

5.2.1 Outline of the Computer Program. A computer program was written to ease the burden of reduction of the many observations into a diffusion coefficient. In evaluating a diffusion coefficient using Equation 4.1 one forms many changes of location ΔY_i from the experimental observations of the labelled particle. Since the time Δt_i between observations is not constant, each such change of location is scaled through division by the square root of the corresponding elapsed time between observations, thus producing an individual value of $\Delta Y_i / \Delta t_i^{1/2}$. For convenience, these scaled changes of location will be referred to as "jumps".

Because of the non-uniformity of the shear and concentration in the wall regions of the Couette channel, it is imperative that diffusion coefficients be evaluated using jumps occurring only in the central regions of the channel. To impose this condition and yet to avoid restrictions on possible jumps, the program logic used only those data for which the initial location of the jump was in the central one-fifth of the Couette channel.

In order to check that the walls and/or wall regions did not restrict the diffusive motion a series of diffusion coefficients were calculated from the experimental data. Each diffusion coefficient of the series had a specific minimum time interval between the observations used to calculate its jumps. For a diffusive motion the root-mean-square change of location of a particle between observations increases as the square root of the time interval between the observations. Therefore, for large minimum time intervals, the diffusion

coefficients are calculated from larger average changes of location. Note that since the process of observation does not disturb the particle, changes of location with long elapsed time intervals can be formed from the experimental data by summing the intermediate, reported time intervals between only two reported observations. The larger average changes of location are correctly scaled when the jumps are formed. However, for some large minimum time the displacement of the labelled particle will be limited by the walls. Diffusion coefficients calculated using such a minimum time will be spuriously low. Note however, that until the diffusive motions are restricted by the walls or altered by the non-uniformities of the wall regions, the jumps will be properly scaled as the square root of the time interval. A minimum time interval which produces relatively large, yet unrestricted changes of location produces more accurate values of D than a smaller minimum time interval. The diffusion coefficient is more accurate since the sighting error is a proportionately lesser part of the larger average change of location. The choice of a minimum time interval is discussed further in Chapter 6.

Thus, inputs to the computer program consist of: i) the experimental observations, ii) a range of lateral positions in which the initial observation used to form a jump must fall, i.e. the values of position-time which denote the central the channel, and iii) a minimum time interval between the observations used to form the jump. Due to the nature of the detection process, the observations which form the experimental data occur with various time

interval; it is for this reason that only a minimum time interval is specified.

Most of the computer program is an exercise in sorting the observations with the object of finding pairs of observations which suit the restrictions cited above, and from which values of $(\Delta Y_{OBS})^2 / \Delta t_{OBS}$ can be formed. When the computer has discovered a jump with a satisfactory initial position and a correct minimum time interval, it computes the value of $(\Delta Y_{OBS})^2 / \Delta t_{OBS}$ for that jump and adds it to a cumulative variable. It also increments by unity a counter, and thereby counts the number of jumps which have been summed by the cumulative variable.

The diffusion coefficient, given by Equation 4.1 is one-half the cumulative variable divided by the number of jumps which were used to form the cumulative variable.

Outline of the Computer Program. The following outline is a "bare-bones" description of the computer program used to reduce the data:

- I. All cumulative variables and counters within the computer program are set to zero.
- II. The computer program is given the following input information:
 - A. The locations, as times, of the Couette walls.
 - B. The range of locations within the Couette channel, as a range of times, in which the initial observation of a jump must occur. For the values of D shown in Figures 1, 6.2,

6.3, 6.6 and 6.7, all initial observations come from the central one-fifth of the Couette channel.

- C. The desired minimum time interval between observations which are used to form jumps.
- D. The file name and computer location of experimental data, which are lists of observations of the labelled particle and the elapsed time since the prior observation. These data were stored in the computer memory using a separate program at an earlier time.

III. The computer now begins to evaluate the diffusion coefficient; it does so by first forming a working table of 100 observations of the labelled particle and the associated elapsed time interval, taken in order from the experimental data. As each observation is placed onto the working table, it is checked to see that the format is correct and that the value of the time corresponding to the lateral position is reasonable. When an unreasonable format or value is encountered, that datum is not used on the working table. Furthermore, the process of entering observations onto the working table is ended. The observation previous to the unreasonable value is the last observation entered on the working table. Use of a shortened working table prevents the computer program from mechanically "bridging" gaps in the experimental data by forming diffusive jumps using erroneous values of either position or elapsed time interval. Specifically, the method was built into the program logic to

eliminate errors caused by misreading of the paper tape when it was converted to cards. It also detected the nonsensical values punched onto the paper tape when tapes were changed. In general, several observations are lost while the tape is loaded; this method prevented the program from using data from paper tapes since jumps formed from observations on separate tapes would, in general, have an incorrect time interval. Use of a shortened working table is described in Section IV C.

IV. The computer inspects the first entry on the working table to see if the position of the labelled particle is within the central region of the Couette channel. If this entry is within the correct range, then it will be used as the initial observation of a jump.

A. If the first entry is not within the central region of the Couette channel, the computer eliminates the first entry on the working table, shifts up all other entries one position, and reads (and checks for format) a new entry to fill the last position on the working table. The program now returns to step IV above.

B. If the first entry is within the central region of the Couette channel, the computer compares the elapsed time interval between the first and second observations on the table with the minimum time interval. This time interval is the elapsed time interval of the second entry on the working table.

1. If the minimum time interval is smaller than or equal to the time interval between the first and second observations, the computer forms the quantity $[(Y_2 - Y_1)^2 / \Delta t]_{\text{obs}}$ and adds this value to the cumulative variable. It also increments by unity, a counter and thereby sums the number of jumps which have been used to form the cumulative variable. The computer also forms $[(Y_2 - Y_1) / \sqrt{\Delta t}]_{\text{obs}}$ and classifies it into one of 18 size ranges. A counter representing jumps of this size range is incremented by unity.
2. If the minimum time interval is greater than the time interval between the first and second observations, the computer forms the net elapsed time interval between the first and third observations by adding the elapsed time intervals of the second and third entries. The net elapsed time interval is compared to the minimum time interval. If the net elapsed time interval equals or exceeds the minimum time interval then a diffusive jump $[(Y_3 - Y_1)^2 / t]_{\text{obs}}$ is formed and variables are incremented as in IV.B.1. If the net elapsed time interval is less than the minimum time interval, then a net time interval between entry 4 and entry 1 is calculated. This process is continued until a diffusive jump which has a net time interval which exceeds or equals the minimum time interval is located. The 100 observations

entered on the working table have a sufficiently great net time interval between the first and the last observations so that a suitable jump can always be formed from the table. When a suitable jump is found the computer, as in step IV.B.1, increments the cumulative variable, the number counter and the appropriate size-range counter. At this point the working table is reformed by deleting all observations up to the observation which was used as the second part of the jump. This observation is now used as the first entry on the "new" working table. All the observations after this entry on the table are shifted up and new observations are read sequentially from the experimental data to fill the table. The new observations are checked for format and numerical value as in section III. When the working table is refilled, the computer begins anew at IV.

- C. For situations with a shortened working table steps IV A or IV B are carried out until no more suitable jumps can be formed. In cases with a shortened working table from which a suitable jump cannot be formed, the remaining observations on the table are discarded. A new table is formed beginning with the first reasonable observation after the nonsensical observation. The computer then begins again at step IV above.
- V. When the computer has sorted through the experimental data, it reports a value of D , which is one-half the cumulative variable

divided by the number of jumps. The value of D reported by the computer program has units of time since the jumps were formed using changes of position-time. A value of D in standard units is obtained by multiplying D as reported by the computer program by the square of the velocity of the viewing window. It also reports the numbers of jumps which fell into each of the 18 size ranges. These numbers provide a frequency distribution of the various jump sizes which were used to calculate D . They were used in the chi-squared tests of normality reported in Chapter 6.

6. RESULTS AND DISCUSSION

In the first section of this chapter the diffusion coefficients, determined as described in Chapter 5, are presented without consideration of error.

A summary of the various error sources and resultant effects upon the data are presented in the second section. The detailed analysis is presented in Appendix III.

In the third and final section are presented the best estimate of corrected data together with an estimated band of confidence. A comparison of the data to a theory by Buyevich [15] is also shown.

6.1 Experimental Results

Sets of experimental observations of the position of a radioactively labelled particle were collected for various apparent shear rates and volumetric concentrations of the slurry. From each set of observations a value of the diffusion coefficient was calculated using the methods of Chapter 5. The principal result of the experimental investigation, which is shown in Figure 6.1, is a plot of the non-dimensional diffusion coefficient, $D/a^2\omega$, versus the concentration ϕ .

To better illustrate the linear relationship of D to ω a plot of D/a^2 versus ω is included as Figure 6.2. (The bars shown on the graph represent error limits which are qualitatively described in Section 6.2 and quantitatively described in Appendix III.) A line with a slope of one has been included as a reference.

Among the data in Figure 6.1 there is no major ordering according to the Reynolds number of the Couette flow in which the sets of observations were collected. To wit, the uppermost and lowermost points at a concentration of forty percent are the two lowest Reynolds numbers.

Before proceeding to further discussion of the data presented in Figure 6.1, it is necessary to consider the errors which occur in collecting the observations since the scatter in the results of Figure 6.1 turn out to be of the same order as the error.

6.2 Summary of Error Analysis (see Appendix III for details)

Definition of Terms. This section is split into two parts which respectively discuss the systematic and random errors. The term systematic errors refers to those errors whose size and sign are explicitly fixed by either the apparatus, instrumentation, or technique. An example of a systematic error would be the error in a measurement of the distance which would occur if a bent or distorted scale were used.

On the other hand the term random errors is used to describe those errors that have a variable sign and size and which, consequently, are most commonly described using statistics. A familiar example of a random error is the variation of lengths which occurs with repeated micrometer measurements of the length of an object.

6.2.1 Random Errors. Ideally, the observations of the labelled particle would be only measurements of its "momentary average" position. If, for example, at the time of observation the labelled particle is laterally displaced because it is engaged in a passing interaction with its neighboring particles it has a "false" temporary lateral position from which it will recover. There is also a difference between the actual observed position and the position reported by the detector, which will be termed "the sighting error". In Figure 6.3 these three component parts of the reported lateral position reported at each observation of the labelled particle are shown. They are:

- i) the "momentary average" position, i.e. the lateral position which the labelled particle would take when it "finishes" its current interaction with its neighbors,

- ii) the passing-motion component, which is the lateral distance the labelled particle is displaced from its average position because it is currently interacting with its neighbors, and

iii) the sighting error component, which is the error in the detection of the true, instantaneous position of the labelled particle. Thus, every observation of the position of the labelled particle can be represented as

$$Y_{\text{obs}} = Y_{\text{average}} + Y_{\text{passing}} + Y_{\text{sighting}} \quad (6.1)$$

and the difference in observed position of the labelled particle at any two times can be expressed as:

$$\Delta Y_{\text{obs}} = \Delta Y_{\text{average}} + \Delta Y_{\text{passing}} + \Delta Y_{\text{sighting}} \quad (6.2)$$

where the Δ denotes the difference between the values of a quantity at the two times.

Values of the diffusion coefficient are calculated as described in Chapter 5 using $(\Delta Y_{\text{obs}})^2$. In Appendix III, it is shown that the form of the diffusion coefficient when ΔY_{obs} is represented using Eq. 6.2 is

$$D_{\text{obs}} = \frac{1}{2N} \sum_{i=1}^{i=N} \frac{(\Delta Y_{\text{ave}_i})^2}{\Delta t_i} + \frac{1}{2N} \sum_{i=1}^{i=N} \frac{(\Delta Y_{\text{passing}_i})^2}{\Delta t_i} + \frac{1}{2N} \sum_{i=1}^{i=N} \frac{(\Delta Y_{\text{sighting}_i})^2}{\Delta t_i} \quad (6.3)$$

In the derivation of Eq. 6.3, two assumptions based upon the physical origin of the components were made. The first is that the differences in the three components are independent variables. The second is that the mean value of each component ΔY is zero. Physically

this implies equal distributions of the lateral motions to the left and right. These assumptions appear to be physically meaningful.

Only the first term on the right hand side is a true diffusion. The other terms represent "error" diffusions. It is interesting to note that because of the "squared" form of the ΔY_{obs} which enter into calculations of the diffusion coefficient, the random errors are always additive, positive numbers.

The error diffusions have an interesting property since the magnitudes of their ΔY^2 terms are not functions of time. Rather they are determined either by the detection apparatus or by the nature of a "single" passing event in a shear flow. Thus, when values of D_{obs} are calculated using longer average times between observations of the position of the labelled particle these error diffusions will have smaller values. Physically, this is a reflection of the fact that for longer times the errors are a smaller fraction of the distance moved as a result of diffusion. The errors are caused by inaccuracies of sighting the particle or are spurious contributions caused by the passing motion, and since these events are independent of time so are the errors which result from them. This time-dependent behavior of the value of D_{obs} is shown in Figure 6.4 for a forty percent concentration of large-particle slurry and a shear rate of ten inverse seconds. The time interval shown on the abscissa is the

minimum time interval between the observations used to form jumps during the calculation of D_{obs} . Also shown are values of $D^*/a^2\omega$, i.e. values of $D_{\text{obs}}/a^2\omega$ which have been corrected for the sighting error and estimated passing motion error. The details of calculations for these corrections appear in Appendix III. Ideally, these corrections should produce a horizontal straight line. However, the form assumed for the passing-motion correction is very simple; a triangular distribution of the possible values of $\Delta Y_{\text{passing}}$ with a maximum value of $\Delta Y_{\text{passing}}$ equal to a particle radius was used. Thus, this correction may not be entirely adequate.

An obvious solution to this problem of time-dependent error is to wait long enough such that the curve becomes approximately flat. However, for a long minimum time interval between observations, such as 100 seconds, the bounds imposed by the channel walls begin to restrict the range of the possible diffusive motion. One test for wall restriction is to statistically compare the distribution of measured diffusive jumps with the ideal normal distribution. Of the data shown in Figure 6.4, only the points which have a minimum time interval of 50 s or less have a five percent level of confidence when chi-squared tested for fit to a normal distribution. Another test for wall restriction uses a calculation of the average length a particle can be expected to move during a time interval. In Appendix III, the use of this test to rationalize the selection of a "20 second" minimum time

interval is presented. Basically, with this time interval the diffusion length from the central fifth of the channel to the wall is sufficiently large that 95% of the jumps are unrestricted by the walls. Other than Figure 6.4 and 6.8, the data presented in this thesis for large particle slurries at $\omega = 10 \text{ s}^{-1}$ have been calculated using a minimum time interval between observations of 20 s. Using the scaling given by $D/a^2\omega$ the minimum time intervals for other shear rates and particle sizes are increased so these data are collected with the same "average" jump length; a more detailed discussion of the minimum time interval is contained in Appendix III. However, depending upon the error criteria used to select a minimum time interval, there is an uncertainty of value of the diffusion coefficient which is a factor of approximately 2 about the corrected value of the diffusion coefficient.

In Figure 6.5 the shaded symbols show the uncorrected values of the non-dimensional diffusion coefficient, while the open symbols show the values when corrected for passing and sightings using 20 s for the minimum time/ interval. Error bars in Figure 6.2, 6.4 and 6.5, which are 95% confidence limits, are shown with each point; the derivation of these error bars is covered in Appendix III.

6.2.2 Systematic Errors. Of the three sources of systematic error which occur in the measurements, there are two sources that occur for slurries of both particle sizes but are largest in

the case of the large-particle slurries. These two systematic errors are the differences between the true and apparent concentrations.

Apparent Shear Rate. All the data of this thesis have been given using the apparent shear rate, i.e. the shear rate calculated as the difference in Couette cylinder velocities divided by the width of the Couette channel. The previously discussed data of Karnis, et al. [17] show that the velocity profile in the middle of the Couette channel is approximately linear for small a/w but Karnis, in his work, did not seek or obtain an exact relationship describing the shear rate in the middle of the Couette channel. However, as described in Appendix III, it is possible to estimate the actual shear rate in the central region of Couette channel by interpolating between the data of two of the experiments whose results are presented by Karnis. If the data were plotted using this value of shear rate, the diffusion coefficients would be increased. The amount by which they would be increased is represented in Figure 6.6 by the vertical arrows which are attached to each data point. The position of the diffusion coefficient when normalized using the estimated true shear rate would be at the tip of the vertical arrow.

Apparent Volumetric Concentration. Because of an uncertainty in the actual concentration of the slurry in the central section of the channel the data as presented are plotted using the apparent volumetric concentration i.e., the volume of particles in the slurry

divided by the total volume of the slurry. It is obvious from the description of Chapter 5, that this apparent concentration can be measured very accurately.

The actual concentration in the central portion of the channel is larger than the apparent or loading concentration because this region contains the additional particles which are excluded by the wall, i.e. the Vand effect as discussed in Chapter 2. An upper limit on the actual concentration can be calculated assuming that all the particles within one radius of the channel wall have been redistributed to the center of the channel. In Figure 6.6 the horizontal arrows indicate the new positions which the data would take if they were plotted versus the estimated upper limit of actual concentration instead of versus the apparent concentration.

Systematic Error Specific to the Experiments Using Small-Particle Slurries. The third source of systematic error, pertains only to the data for the small-particle slurries. Because there is a range of particle sizes both among the unlabelled particles and also between the labelled particle and the unlabelled particles, there is a question concerning the appropriate value of a to be used when the data is non-dimensionalized. The range of radii of the unlabelled particles in these experiments was 0.019 inch to 0.023 (Chapter 3 contains a more thorough description of the particles). The radius of the radioactively labelled particle was larger than the radii of the unlabelled particles; it had an average radius of

0.026 inch. The small-particle data of the thesis have been plotted using a value of $a = 0.023$ inches; which is the largest radius of the unlabelled particles. This was selected as a compromise between radii of the labelled and unlabelled particle sizes.

In Figure 6.6 smaller, open circles are shown both above and below each data point obtained using the small-particle slurries. These smaller, open circles show the values of the non-dimensional diffusion coefficient when evaluated using $a = 0.019$ in. (upper circle) and 0.026 in. (lower circle).

Due to the mechanics of interactions between small and large particles it is possible that the upper smaller circles more truly represent the non-dimensional diffusion coefficient. To understand the reason for this, consider two particles about to undergo a passing interaction in an otherwise particle-free Couette flow. Furthermore, one of the particles is to be much smaller than the other. When the two particles undergo a passing interaction the small particle will approximate a fluid element, and will just go around the large particle. Thus, the small particle will undergo almost all the lateral displacement while the large particle will undergo almost none. As the sizes of the particles become more equal the relative amounts of motion will also become more equal. However, the larger particle will continue to have the smaller lateral motion. In a concentrated slurry the diffusive motion occurs by a mechanism of interrupted passing events. Then, the diffusive motion of larger particles is smaller since they have smaller passing motions which

can be interrupted. A reasonable approximation for the ratio of the sizes of the passing motions would be that they scale inversely as the respective radii of the particles. Thus, in the small-particle slurries with a large labelled particle, one might conjecture that the appropriate radius a to use in forming the dimensionless $D/a^2\omega$ would be that of the small unlabelled particles. This scaling is shown in Figure 6.3 by the upper small open circles.

6.2.3 Results of Calculational Methods of Lesser Accuracy.

For completeness in the presentation of the experimental data, graphs of the diffusion coefficients obtained from two other, less accurate methods of calculation of the diffusion coefficient have been included.

One method of calculation involved forming jumps using initial observations from the central three-fifths of the Couette channel. The second observation of each jump was then the observation which occurred after the minimum time interval of twenty seconds had elapsed. The diffusion coefficients which result from calculations performed using these selection criteria are shown in Figure 6.7.

Using these selection criteria produces jump distributions for the fifths adjoining the central one-fifth region which are so obviously skewed that no statistical test for wall restriction is necessary. Yet the values of the diffusion coefficient produced by these calculations are not grossly different from the results of

Figure 6.1 (Figure 6.1 used the same minimum time interval but had a restriction of the initial observation of a jump must occur in the central one-fifth of the Couette channel).

The second method of calculation involved using only observations from the central fifth of the Couette channel. However the second observation was chosen after a minimum time interval of four seconds had elapsed. Figure 6.8 shows the results of the calculations using these criteria. Although the values of D_{obs} are higher than those of Figure 6.1, they have the same general distribution. As discussed, both previously and in Appendix III, this larger value is caused primarily by the associated larger "apparent diffusions" caused by the sighting and passing motion errors.

It deserves emphasis that the most correct calculational procedure is probably the one presented in section 6.2.2, i.e., the one which uses the central one-fifth of the Couette channel as the location of the initial observation and selects a second observation after a minimum time of twenty seconds when jumps are formed.

6.3 Summary.

The effects of the systematic and random errors when estimated and for a time between observations of twenty seconds are approximately cancelled since they are of opposite sign and size. However, the choice of a 20-second minimum time between the observations used to calculate the diffusion coefficient is somewhat arbitrary; and thus,

an overall accuracy of measurement of a factor of two is appropriate. Further, there is a spread within the non-dimensional data of approximately a factor of two. Using this guideline, a band of confidence is shown in Figure 6.9 with the uncorrected data.

Within this level of accuracy it appears that there is only a weak variation of the non-dimensional diffusion coefficient with either concentration (in the range 0.20 to 0.50) or Reynolds number.

Also shown in Figure 6.9 are three open diamonds which are values of the shear-driven diffusion predicted by Buyevich [15] using a theoretical model which he derived using an analogy to turbulent flow. It is interesting that for the range of concentration from 30 to 50% the diffusion coefficient in this model approximately doubles. However, it is also approximately an order of magnitude smaller than the measured diffusion motion.

7. INVESTIGATION OF THE LATERAL CONCENTRATION

DISTRIBUTION AND ITS RELATIONSHIP

TO THE DETECTIONAL PROCEDURE

Introductory Comments. On first thought it seems reasonable that if the observations of the labelled particle are sorted according to lateral location, the relative frequency of observations at a location should be related to the local concentration. Then on this hypothesis, it would be possible to obtain a measurement of the concentration distribution across the Couette channel from the experimental data. Attempts to do this produced spurious results which were later understood to be caused by the interplay of the diffusive motions of the labelled particle and of the role of convection in the detectional process. This chapter contains a discussion of numerical experiments which examined the interplay and which demonstrate why the frequency distribution of observations across the channel width (produced using the detector described in Chapter 4) can not be simply related to the distribution number density across the channel width.

Several subjects relevant to the numerical experiments are treated in the chapter in the following order: i) detectional methods of making observations of a labelled particle; ii) convective and diffusive motions in the numerical experiments, iii) models of the detectors used in the numerical experiments.

- iv) values of coefficients in the numerical experiments, and
- v) results and conclusions of the numerical experiments.

7.1 Detectional Procedures for Making

Observations of Marked Particles

There are several procedures, at least in principle, by means of which a single labelled particle is used to measure the coefficient of lateral diffusion, D , and, which in some cases, can also be used to measure the local number density or concentration. Two of these procedures are discussed below. The first technique discussed was selected because it directly produces a measure of number density and is also easily interpreted in a physical sense. The second technique discussed is the one which was used in these experiments; as is made evident in Chapter 4, it was selected for use over other procedures because the others seemed impracticable in the laboratory.

Constant-Time-Interval Technique. By the ergodic hypothesis, the simplest single particle method is equivalent to the standard method of measuring number density. In the standard method a region of convenient volume is defined and the particles which are in the region are counted. The number density is then the result of dividing the number of particles by the volume of the region.

In the "constant-time-interval technique" observations of a single labelled particle lead to the concentration distribution

because, according to the ergodic hypothesis, the many individual observations of the single labelled particle can be combined mathematically to form the equivalent of a single observation of many labelled particles. Physically, an observation of many particles is "assembled" by combining the observations of the single labelled particle, which, during each one of the many observations, simulates one of the many labelled particles.

To use the ergodic hypothesis the many observations of the single labelled particle must be independent of one another, and must be taken without bias. Typically, this is achieved by performing the many consecutive observations with a constant time interval between observations, hence the name given to this technique. From the viewpoint of the diffusive motion, the condition of independence requires that the time between the observations must be sufficiently long for the particle to have diffusively interacted with the wall several times and thus to have lost the positional bias of its past history within the Couette channel. Thus, in order to measure concentration distribution, the time interval between successive observations of the lateral position of the labelled particle must be much larger than that for the measurement of the diffusion coefficient.

A key feature of the "constant-time-interval technique" is that the labelled particle(s) are observed and the position(s) are measured at constant time intervals. This feature requires the ability to make position measurement(s) at any place within the Couette apparatus.

"Viewing Window Technique". In this technique observations of the labelled particle are made only when it comes into a restricted region of detection through the convective motion of the Couette flow. In the restricted region of the Couette flow the lateral position of the labelled particle can be measured through a viewing window. Since the diffusive motion of the labelled particle changes its lateral position, it has a time-varying convective motion. Thus, the time to return to the region of detection is not constant and the time interval between observations is not constant.

Before a further consideration of the "viewing-window technique", it is important to understand that measurements made using this technique must be transformed to produce a measurement of the concentration distribution. Consider determining the lateral distribution of particles using a single labelled particle and the viewing-window technique. Furthermore, suppose that rather than a Couette flow, we have instead a flow with discrete velocities in a small number of discrete, concentric annuli as shown in Figure 7.1 that is, a sort of multi-ringed roulette wheel. Suppose further that the velocities in all the annuli, except in one, are very small compared with that in the exceptional annulus. In this annulus, it is so large that when the labelled particle lands in this annulus, it is carried below the viewing window before it can jump off. Now, let the many particles, of which only one is labelled, jostle each other and change annuli in such fashion that there is always an equal concentration in all annuli. Accordingly, each particle,

whether or not it is labelled, spends an equal amount of time in each annulus. Thus, observations of the lateral position using the "constant-time-interval technique" will show equal frequency of observations in all annuli. With the viewing window technique, on the contrary, the distribution of observations of the labelled particle will not coincide with the distribution of concentration. Indeed, most of the observations will be in the high-speed annulus. To transform the distribution of observations produced by a viewing window detector to a distribution which is proportional to concentration, it is necessary to divide the number of observations at a particular station by the mass flow under that station of the viewing window.

The mass-flow transformation can be explicitly demonstrated through a conversion via the ergodic hypothesis of the single-particle measurements to a single many-particle measurement. Again we consider a situation of uniform concentration, however, all the particles are labelled. Furthermore each of the particles will be detected only once by the detector each time it goes through the region of detection. Since there is constant concentration at each lateral location it is readily apparent that if the particles are frozen into their lateral locations that the multiple-particle measurement at a location will be proportional to the mass flow under the detector at that location. Now, if the particles are unfrozen, the same result must occur since for constant concentration there is no net change in the number of particles in any lateral location.

Each particle which diffuses out of a zone is replaced by a particle which diffuses into that zone. Thus, the number of observations at any location is affected only by the number of particles which flows under that location.

When the mass-flow transformation above is made, the concentration distribution determined with a single labelled particle and the viewing window technique must be the same as with the constant-time-interval technique.

Figure 7.2 is a bar-histogram depicting the frequency distribution of experimental observations of the labelled particle according to the lateral position at which they were taken. The observations were collected during large-particle experiments at a 40% concentration and a 10 s^{-1} shear rate. Also shown are a horizontal dashed line which is the expected result of a photographic technique of collecting the frequency distribution and an upward, sloping line which illustrates an untransformed frequency distribution as collected by a viewing-window technique. It is apparent that neither fits the experimental data.

7.2 Numerical Experiments

Numerical experiments were selected to investigate the difference between the experimental data and the expected results because they offered the chance to add or delete details of the physical events found in the actual experiments such as wall effects.

Also numerical experiments provide verification of the features which characterize observations taken with a viewing-window technique or with a constant-time interval technique. Since this portion of the work was undertaken as a verification of ideas, it was not made rigorously exact in all physical details. Rather only the primary, essential physical details were included so as to demonstrate the characteristics of the detection process with relationship to the measurements of the concentration

In the numerical experiments the slurry flow was modelled as mutually perpendicular convective and diffusive steps. As is illustrated in Figure 7.3 one step of each type occurred during each time step of the simulation. All the determinate features of a Couette flow were included in the convective steps, C, and all the interactions between the particles were included in the diffusive step, D.

Convective Steps. At the beginning of each time step the current lateral position of the labelled particle was used to obtain a convective velocity from a linear velocity profile. Each time step in the numerical experiments resulted in convective displacement which was the product of the time-step interval and the convective velocity associated with the lateral position at that time. This is illustrated in Figure 7.3 for several convective steps where the horizontal arrows with a number and the letter C are the convective motions.

Note that since the material of a particle would prevent its center from being at the Couette wall there are no positions of zero particle velocity in either the numerical or actual experiments even though one of the Couette walls is stationary.

Diffusive Steps. Physically, the diffusive steps are best viewed as the outcome of a random walk executed by the labelled particle during the course of a time step. Since the outcome of a diffusive step is not dependent upon the initial location, it is best described as a probabilistic distribution of the possible changes of location from the original location. The diffusive step is selected from a triangular distribution (approximating a normal distribution) by a computer subroutine which use a random number generator. Figure 7.4 shows the fit of the triangular distribution used in the numerical experiments to data from large-particle experiments for $\omega = 10 \text{ s}^{-1}$ and $\phi = 40\%$.

Wall Effects. The initial position of a particle undergoing a random walk must have a bearing on its outcome in two special cases which are of concern here. One case is when the labelled particle is near a wall; obviously the labelled particle cannot have a change of location which places it into or on the other side of the wall. In fact, what occurs is, that like the labelled particle, the distribution of changes of location is reflected, as it were, off the wall. The reflected distribution is obtained by first placing a "mirror-image" distribution of changes of location on the

opposite side of the wall. The centers of both distributions and the center of the particle are the same distance from the wall. Then, as shown in Figure 7.5 the reflected distribution of changes of location is the sum of the original and mirror distribution, with the restriction that the changes of location of the labelled particle must not result in a change of position to the other side. Chandrasekhar [35] shows that this treatment of a random walk is equivalent to the boundary condition of no mass flux through the wall.

The second case in which the outcome of a random walk is position-dependent is when the labelled particle is in a region with either a concentration gradient, a diffusion-coefficient gradient, or in a uniform-force field. All three of these situations result in a preferred direction for the changes of location and, in fact, as Manning [6] demonstrates, are of similar mathematical form. A simple way of picturing the preferred direction is to first consider the changes of direction which would result for an isolated (no other particles) labelled particle in a region with a uniform force field. Here the result during a time step is a systematic, directed change of location of the particle from its original location. For the situation of the labelled particle interacting with many other particles in either a gradient or in a uniform force field, there will be distribution of changes of location which is still symmetric. However, as shown in Figure 7.6, the distribution is now centered about the average change of location which would be caused solely by gradient or the uniform force field.

7.3 Detection Processes in the Numerical Experiments

Two types of detection systems were modelled in the numerical experiments. Each of the detection processes was modelled as occurring in the middle of a time step, between the convective motion and the diffusion motion. The order of events in the main loop of the numerical experiments is shown in Figure 7.6.

One detection system was based on "constant-time-interval" technique; this detector measured both the lateral and longitudinal positions of the center of the labelled particle. These observations were categorized by location according to whether the center of the labelled particle fell within equal-sized lateral spaces on $0 \leq Y \leq W$ and into equal-sized longitudinal spaces on $0 \leq X \leq L$.

The second detection system was based on the "viewing-window" technique. In reference to Figure 7.3, the viewing-window detectors in the numerical experiments were lines of zero width spaced at intervals, L . They could locate the labelled particle each and every time it crossed a detector line. These observations were also categorized into groups according to the lateral section of the detector line on which the observations were made.

7.4 Values of Parameters in the Numerical Experiments

In the numerical experiments, scaling values of parameters were selected to correspond to the experimental conditions. The channel width and lap length of the Couette device of the numerical

experiments correspond to the experimental sizes while the magnitude of the diffusion coefficient and the convective velocities were scaled to match experimental results obtained in large-particle experiments at 40% concentration and a ten-inverse-second shear rate. Values of D and $V_{\max}$ used are $0.8 \times 10^{-2} \text{ cm}^2/\text{s}$ and 29 cm/s .

A time step, δt , with a value of 0.04 seconds was used; with this time step a labelled particle executing only convective steps would require 100 consecutive steps to traverse the lap length of the Couette device at the maximum convective velocity. During the course of the numerical experiments separate runs were conducted in which this time element was increased or decreased by a factor of three. No change of outcome of the experiments was noted, thereby signifying that the size of the time element did not bias the outcome of the numerical experiments.

Time intervals of 40 and 400 seconds were used between the observations collected using the photographic technique. Both time intervals produced statistically-equivalent frequency plots thereby demonstrating that the time interval between photographic observations was sufficiently long to satisfy the criterion of independence which is necessary for the validity of the ergodic hypothesis.

In the numerical experiments which included the effects of gradients of concentration or diffusion coefficient or of force fields, rather than using an explicit description of the physical

events, the effects were approximated by a description of the shift of probability distribution which describes the diffusive motions under these circumstances. The shift of the probability distribution from its zero mean value (shown in Figure 7.6) was selected such that it decreased with the distance from the wall. Specifically, the shift was made to decay exponentially with the distance from the nearest Couette wall. It was of the form:

$$\delta Y_{\text{shift}} = B(D\delta t)^{1/2} e^{-AY/(D\delta t)^{1/2}} \quad (7.1)$$

where Y is the distance from the nearest Couette wall,

δY_{shift} is the average value of the change of location which the labelled particle would experience purely under the influence of a gradient or a force. The variables A and B of Equation 7.1 are scaling coefficients; for the data of Figure 7.8 values of A and B are 0.67 and 0.5 respectively. As is later shown, these values provide similar distributions for the numerical and actual experiments. It should be realized that with the numerical values of the experiments, the exponential decay produces no appreciable shift of position due to the gradient or force in the region greater than one large particle diameter from the wall. Furthermore, due to loss of significant digits by the computer, there is no shift in the central half of the Couette channel.

7.5 Results of Numerical Experiments

The bar histogram of Figure 7.7 shows the results of numerical experiments under conditions of uniform concentration and diffusion coefficient, and with no force fields. The open, wide bars are the values for the "viewing-window" detector and the closed, narrow bars are the results for the "constant-time-interval detector". With the viewing-window detector, which corresponds to the experimental apparatus, the number of observations taken by the detector in a small lateral zone was found to be directly proportional to the average convective velocity of that lateral zone. The constant-time-interval detector, however, gave a uniform distribution of sightings throughout the channel; that is when the region within a radius of the wall was excluded, no statistically significant deviation from the average could be found. The distribution of sightings in the longitudinal direction taken by the constant-time-interval detector was also independent of location, as would be expected. The results provide confirmation that the numerical experiments are in agreement with the physical facts described in the introduction of this chapter.

Figure 7.8 shows the results of the numerical experiments using Equation 7.1 to express the effect of concentration or diffusion-coefficient gradients or force fields near the walls. The differences between the results shown in the figure and the previous results are quite obvious. Now, the detector using the

constant-time-interval technique produces a histogram which is symmetric, with decreased frequency of observation closer to the walls. The histogram produced using the viewing-window technique has changed from the previous triangular shape and now decays at both walls. As necessary via conservation the viewing-window technique now produces a relative surplus of sightings in the center region.

Discussion. The results of the numerical experiments shown in Figure 7.8 are much closer to the actual experimental results than those of Figure 7.7. Furthermore, the nature of the slurry flow near the wall points to the necessity of including "wall events." According to the previously discussed results of Karnis [17] the slurry flow in the experiments has in the wall region a decreased concentration and an increased shear rate. Since D is proportional to ω the variable shear rate will produce an inwardly directed gradient of diffusion coefficient; the decreased concentration at the wall will have an associated concentration gradient. It is unlikely that motions induced by these two gradients will exactly cancel one another. If they do not, then some average directed gradient-induced motion will exist in the wall region. Accordingly, the small inwardly directed average shift of position, which produced the results of Figure 7.8, may be a reasonable representation of the behavior of particles in slurry flows near wall.

It, thus, seems reasonable to assign the seemingly spurious experimental results of Figure 7.2 to an interplay of the biased diffusive motion characteristic of the wall region with the convection dependence of the viewing-window detector. Physically, one might say that the wall regions have a propensity to "kick" the labelled particle to the central regions of the channel where it can easily be convected to the viewing-window.

The numerical experiments do provide a satisfying degree of verification of the measurements of the thesis in the following sense: the values used in the numerical experiments were taken from an actual experiment and converted, via an exceedingly simple, but reasonable computational scheme, into a gross measurement which compares favorably with a similar measurement produced by the actual experiments. Although this internal consistency is not in itself a proof, it is a necessary condition for the validity of the experimental work.

8. CONCLUSIONS

As a result of the experiments of this thesis, the following conclusions about shear-driven diffusion in low Reynolds Number shear flow may be drawn:

- i) The diffusion coefficient scales as $a^2 \omega$.
- ii) In the range of volume concentration from 20% to 50%, the diffusion coefficient appears to be only weakly dependent upon the volumetric concentration of the slurry.
- iii) The nondimensional diffusion coefficient, has a value approximately $D/a^2 \omega$, 3×10^{-2} .

REFERENCES

1. Goldsmith, H.L. and Mason, S.G., "The Microrheology of Dispersions" in Rheology - Theory and Applications, Vol. 4, Eirich, Frederick R. editor, Academic Press, N.Y. (1967)
2. Wohl, P.R. and Rubinow, S.I., "The Transverse Force on a Drop in an Unbounded Parabolic Flow," J. Fluid Mechanics, Vol. 62, Pt. 1, pp. 185-207 (1974)
3. Ho, B.P. and Leal, L.G., "Inertial Migration of Rigid Spheres in Two-Dimensional Unidirectional Flows," J. Fluid Mechanics, Vol. 65, Pt. 2, pp. 365-400 (1974)
4. Segré, G. and Silberberg, A., "Behavior of Macroscopic Rigid Spheres in Poiseuille Flow," J. Fluid Mechanics, Vol. 14, p. 115 and p. 136 (1962)
5. Brenner, H., Chem. Eng. Sci., Vol. 18, p. 2 (1963)
6. Manning, J.R., Diffusion Kinetics for Atoms in Crystals, D. Van Nostrand, Princeton, N.J. (1968)
7. Fung, Y.C., "Blood Flow in the Capillary Bed," J. Biomechanics, Vol. 2, p. 353 (1969)
8. Whitmore, R.L., "Hemorheology and Hemodynamics," Biorheology, Vol. 1, p. 20 (1963)
9. Merrill, E.W., Gilliland, E.R., Cokelet, G., Shen, H., Britten, A., Wells, R.E., "Rheology of Human Blood Near and at Zero Flow..." Biophysical Journal, Vol. 3, No. 3, p. 199 (1963)

22. Petschek, H.E. and Weiss, R.F., "Hydrodynamic Problems in Blood Coagulation," AIAA Paper No. 70-143 presented at AIAA 8th Aerospace Sciences Meeting, New York, January 1970
23. Bernstein, E.F., Blackshear, P.L., Keuer, K.H., American Journal of Surgery, Vol. 114, p. 126 (1967)
24. Steinbach, J.H., "Red Cell Diffusion by the Taylor Dispersion Method," Ph.D. thesis, University of Minnesota, March, 1974
25. Taylor, G.I., Proc. Roy. Soc. A 219, p. 186 (1953)
26. Turitto, V.T., Benis, A.M., and Leonard, E.F., "Platelet Diffusion in Flowing Blood," Ind. Eng. Chem. Fundam., Vol. 11, No. 2, p. 216 (1972)
27. Grabowski, E.F., Friedman, L.I. and Leonard, E.F., "Effects of Shear Rate on the Diffusion and Adhesion of Blood Platelets to a Foreign Surface," Ind. Eng. Chem. Fundam., Vol. 11, No. 2, p. 224 (1972)
28. Collingham, R.E., "Mass Transfer in Flowing Suspensions," Ph.D. thesis, University of Minnesota, March, 1968
29. Singh, A., "The Thermal Conductivity of Stationary and Flowing Suspensions," Ph.D. thesis, University of Minnesota, March, 1968
30. Diller, T.E., "Augmentation of Oxygen Diffusion in Blood Due to Shear-Induced Red Blood Cell Motion," S.M. thesis, MIT, May, 1974
31. Keller, K.H., "Effect of Fluid Shear on Mass Transport in Flowing Blood," Federation Proceedings, Vol. 30, No. 5, p. 1591 (1971)

10. Copley, A.L., Hemorheology, Oxford, England, Pergamon Press (1966)
11. Charm, S.E., Blood Flow and Microcirculation, Wiley, New York (1974)
12. Jeffrey, G.B., Proc. Roy. Soc. A 102, p. 161 (1922)
13. Vincenti, W.G. and Kruger, C.H., Jr., Introduction to Physical Gas Dynamics, Wiley, New York (1965)
14. Jeans, J.H., The Dynamical Theory of Gases, Dover, New York (1954)
15. Buyevich, Yu. A., "Statistical Hydromechanics of Disperse Systems; Part 3, Pseudo-turbulent Structure of Homogeneous Suspensions," J. Fluid Mechanics, Vol. 56, Pt. 2, pp. 313-336 (1972)
16. Happel, J., and Brenner, H., Low Reynolds Number Hydrodynamics Noordhoff (1973)
17. Karnis, A., Goldsmith, H.L. and Mason, S.G., "The Kinetics of Flowing Dispersions; I. Concentrated Suspensions of Rigid Particles," Journal of Colloid Interfacial Science, Vol. 22, p. 531 (1966)
18. Vand, V., J. Phys. and Colloid, Chem., Vol. 52, pp. 277-300 (1948)
19. Karnis, A., Goldsmith, H.L. and Mason, S.G., "The Flow of Suspensions Through Tubes; V. Inertial Effects," Canadian Journal of Chem. Eng., Vol. 44, p. 181 (1966)
20. Sacks, A.H. and Tickner, E.G., "Laminar Flow Regimes for Rigid-sphere Suspensions," Hemorheology, Proceedings of the First International Conference, p. 277, Pergamon Press, New York (1967)
21. Goldsmith, H.L., "Motion of Particles in a Flowing System," Vascular Factors and Thrombosis, p. 70, Schatlaue-Verlag, Stuttgart

32. Tecna - Hill, J.D., Iatridis, A., O'Keefe, R., Kitrilakis, S.,
"Technique for Achieving High Gas Exchange Rates in Membrane
Oxygenation", Trans. ASAI0, Vol. 20 (1974)
33. Lamb, H., Hydrodynamics, Dover, New York (1945)
34. Cramer, H., Mathematical Methods of Statistics, Princeton Univer-
sity Press, Princeton, N.J. (1946)
35. Chandrasekhar, S., "Stochastic Problems in Physics and Astronomy,"
Rev. Mod. Phys., Vol. 15, No. 1, p. 3 in Selected Papers on Noise
and Stochastic Processes, Nelson Wax, editor, Dover, N.Y. (1954)
36. Price, W.J., Nuclear Radiation Detection, McGraw Hill, New York
(1964)
37. Bailey, D.G., "Migration of Disks in Couette Flow and Application
to Blood Oxygenator Design," M.E. thesis, MIT, February, 1975

CAPTIONS FOR FIGURES

Figure 1.1 - Sketch showing a single particle in a shear flow. In frame (a) the particle is viewed from a stationary reference frame; frame (b) is shown in a reference frame moving at the velocity of the particle.

Figure 1.2 - "Two-body standard collisions between equal-sized rigid spheres as seen in the X_2X_3 plane (a) and the X_1X_3 plane (b). The stages shown are (i) approach, (ii) apparent contact at $\phi_1^0 = -60^\circ$; $\phi_2^0 = -59^\circ$, (iii) mid-point of doublet rotation as a rigid spheroid, (iv) point of separation at $\phi_1^0 = 60^\circ$, $\phi_2^0 = 59^\circ$ and (v) recession." from Goldsmith and Mason [1].

Figure 2.1 - Sketches of hypothesized interactions of two particles. In (a) the particle positions are shown prior to the interaction. During the interaction the particles will follow the arrows resulting in the hypothesized separation. In Frame (b) the positions and paths are shown when they are viewed from the rear of (a). In a zero-Reynolds number, inertia-free flow, the principle of kinematic reversibility requires that the paths of (a) and (b) be identical. Therefore, the hypothesized separation is incorrect.

Figure 2.2 - Sketches of a three-particle interaction in the reference frame of particle A. The initial positions of the particles are shown in (a), the final positions in (b), and a rear view of (a) is shown in (c). Since (b) and (c) are not geometrically equivalent, the

principle of kinematic reversibility cannot be used to show that there is no net lateral migration. Frame (d) shows symmetric arrangement of particles in which there will be no net lateral migration as a result of the interaction.

Figure 2.3 - Variation of the position of a tracer sphere in a slurry flow in a circular cylinder Couette apparatus. The outer wall of the Couette apparatus is denoted by the symbol R_2 . The ordinate is measured as the position r within the Couette channel relative to the outer wall. In frames (b) and (c) the ordinate maximum distance on the ordinate is the channel width, and the particle, with radius b , is illustrated in proportion to the channel width. Experimental conditions are:

"(a) volumetric concentration, $c = 0.035$, $b = 0.0172$ cm; for the lower particle $\omega = 0.403 \text{ sec}^{-1}$ and for the upper $\omega = 0.277 \text{ sec}^{-1}$. The arrows indicate the occurrence of 3-body or higher order collisions causing a change in $\bar{r}$.

(b) $c = 0.07$, $b = 0.172$ cm, $\omega = 0.05$ to 0.42 sec^{-1} ; readings at 5 minute intervals.

(c) $c = .19$, $b = 0.172$ cm, $\omega = 0.05$ to 0.42 sec^{-1} ; readings at 30 sec intervals", from Karnis, et al. [17].

Figure 2.4 - Sketch illustrating the nature of the Vand zone. If the number density is measured by counting the number of particle centers which fall into a given length of strip A, then the number density of zero will result. However, strips away from the wall, as E and F, would give normal results.

Figure 2.5 - Dimensionless plots of the velocity profiles observed in Couette flows of slurries. The relative channel position is shown on the ordinate and the relative velocity is shown in the abscissa. As formed, dimensionless groups rectify minor non-linearities caused by the circular geometry of a circular-cylinder Couette apparatus with radii R_1 and R_2 . Experimental conditions are:

"(a) volumetric concentration $c = 0.01$, $R_1 = 4.644$ cm, $R_2 = 5.795$ cm, rotational speed of cylinder 1, $\Omega_1 = 0.00543$ rad. sec⁻¹ rotational speed of cylinder 2, $\Omega_2 = 0.00841$ rad. sec⁻¹ and $2b/\Delta R = 0.026$.

(b) $c = 0.38$, $R_1 = 13.942$ cm, $R_2 = 14.625$ cm, and $2b/\Delta R = 0.083$; experimental points are open circles: $\Omega_1 = 0.00259$ rad. sec⁻¹, $\Omega_2 = 0.0205$ rad. sec⁻¹; open triangles: $\Omega_1 = 0.0149$ rad. sec⁻¹, $\Omega_2 = 0.00614$ rad. sec⁻¹

(c) $c = 0.38$, $R_1 = 14.427$ cm, $R_2 = 14.624$ cm, and $2b/\Delta R = 0.287$; open circles: $\Omega_1 = 0.00422$ rad. sec⁻¹, $\Omega_2 = 0.00851$ rad. sec⁻¹ and closed circles: $\Omega_1 = 0.00422$ rad. sec⁻¹, $\Omega_2 = 0.0129$ rad. sec⁻¹" for Karnis, et al, [17].

Figure 3.1 - (a) Sketch of the borrowed linear-belt Couette apparatus, (b) a two-belt version of (a). Parts denoted by letters are: T, tank which holds the fluid and Couette apparatus; B, moving belt which establishes the Couette flow; P, pulleys which position and tension the belt; and SP, support plates which fix the maximum distance between surfaces of the belt.

Figure 3.2 - Cross-section view of Couette apparatus used in the experiments. Parts A and D are upper and lower spacer plates which

maintain the concentricity of the outer cylinder, C, relative to the inner cylinder and axle, B,

Figure 3.3 - Sketch of the secondary-flow direction in the Couette flow of a liquid with an air interface when the Couette flow is driven by rotation of the outer cylinder and bottom spacer plate.

Figure 4.1 - Method III in which detection occurs when the convective motion brings the labelled particle D into a small arc of the Couette apparatus. The cylinders of the Couette apparatus are shown by letters A and B. For clarity the undetectable, unlabelled particles are now shown.

Figure 4.2 - Front view of the experimental apparatus. The following identifying letters for parts of the experimental apparatus are used in 4.2, 4.3, 4.4, and 4.8, 4.9, 4.10, 4.11. A. Scintillation counter, B. He-Ne laser, C. Couette apparatus, D. Photodiode, E. Steel frame, F. Lead wheel of collimating-slit viewing windows, G. Shelf for mounting of lead shielding. (For clarity the lead shielding which sits on the shelf has been omitted from the illustrations.) H. Mirror, I. V-belt drive for the outer Couette cylinder, K. Lead shielding, and L. Lead sheet.

Figure 4.3 - Side sectional view of the experimental apparatus.

Figure 4.4 - Top view of the lead wheel of viewing windows.

Details of the Couette apparatus have been suppressed.

Figure 4.5 - Illustration of Bench-top Test II. A. Laser with beam represented as a dashed line. B. Photomultiplier in refrigerated housing. C. Cuvette with transparent slurry and with E. Particle which

is labelled with a fluorescent compound, and F. Filter which passes the fluorescent light.

Figure 4.6 - Arrangement of Components for the Bench-Top Test III. A. Radioactive solution used as the mock-up of the labelled particle. B. Lead shielding with viewing window. C. and D. Scintillation detector.

Figure 4.7 - Top view of lead block used to form the wheel of collimating-slit viewing windows. Phantom lines show relationship to another lead block.

Figure 4.8 - Position of the lead shielding, K. used for the large-particle experiments. Dashed lines represent the path of least decay for gamma rays to reach the detector from the Couette chamber.

Figure 4.9 - Sketch showing the areas of partial sight, P, and complete sight, C, of a collimating-slit viewing window.

Figure 4.10 - Sketch showing the effect of a blockage of part of one slit. Note that the reduced zone of partial sight is in the forward direction of the scanning motion which is shown by the arrow.

Figure 4.11 - Effect of misalignment of the axis of the collimating-slit viewing window and the axis of the Couette apparatus. Numbers 1 and 2, denote the cases of upper and lower positions of the labelled particle which are discussed in the text. Direction of the scanning motion is shown by the arrow.

Figure 4.12 - Distribution of sightings of the large particle when it is fixed to a Couette wall. All sightings are references to the mean value of the sightings taken at the upper position on the

outer wall which is the zero for that distribution and for the lower position on the outer wall. The zero position on the inner wall is calculated from the scanning speed of the viewing window and particle position given as the mean value of the distribution on the outer wall.

Figure 4.13 - Distribution of sightings of the small particle when it is fixed to a Couette wall. Zero values of the distribution are determined as in Figure 4.12.

Figure 6.1 - Graph showing $D_{\text{obs}}/a^2\omega$ versus concentration. Symbols for small-particle experiments, $a = 0.019$ to 0.026 inches, are circles, $\omega = 10 \text{ s}^{-1}$, for large particle experiments, $a = 0.0625$ inches, at $\omega = 10 \text{ s}^{-1}$ squares; at $\omega = 1 \text{ s}^{-1}$ triangle; and at $\omega = 0.4 \text{ s}^{-1}$ hexagon.

Figure 6.2 - Graph showing $D_{\text{obs}}/a^2\omega$ or D^*/a^2 versus the shear rate, ω . Same symbol notation as Fig. 6.1; open symbols are values of D^* derived from the corresponding value of D_{obs} shown as a solid symbol.

Figure 6.3 - Sketch illustrating the definitions of the values of Y_{obs} , Y_{pass} , Y_{sight} , and Y_{ave} . The dashed line shows the path of the labelled particle. At the time the apparatus makes an observation the position of the particle is denoted by the open circle. The position, Y_{obs} , reported by the detector is shown by the "x". Note that due to its passing interactions with the neighboring particles the labelled particle is displaced laterally from its "momentary" average location.

Figure 6.4 - Graph showing the effect of using different minimum time intervals, Δt , on the calculated value of the diffusion coefficient and the corrected diffusion coefficient.

Figure 6.5 - Graph of the non-dimensional diffusion coefficient versus the concentration. The open symbols are values of the corrected diffusion coefficient, $D^*/a^2\omega$ described in Appendix III. The solid symbols are values of $D_{obs}/a^2\omega$. Definition of symbol shapes is the same as Figure 6.1.

Figure 6.6 - Graph showing the estimated corrections for two types of systematic error. The horizontal arrows are a correction for the apparent concentration and the vertical arrows are a correction for the apparent shear rate. The small closed circles are values of $D^*/a^2\omega$ obtained using the extreme radii of the distribution of particle radii of the small-particle slurry experiments.

Figure 6.7 - Graph showing corrected and observed non-dimensional diffusion coefficients calculated using a 20 second minimum time and the central one-fifth of the Couette channel as the location of the initial observation of the labelled particle. Definition of symbols is the same as Figures 6.1 and 6.2.

Figure 6.8 - Graph showing values of $D_{obs}/a^2\omega$ calculated using a 4 second minimum time interval. Definition of symbols is the same as Figures 6.1 and 6.2.

Figure 6.9 - Graph of $D_{obs}/a^2\omega$ versus concentration with a band of confidence and three points (open diamonds) from the theory of Buyevich [15].

Figure 7.1 - Sketch illustrating the hypothesized flow system. Annular spaces each have a velocity denoted by the arrows, V or v . One velocity V , is sufficiently great so that when the labelled particle, L , lands in that annulus, it will always be carried under the viewing-window detector, VW .

Figure 7.2 - Bar histogram of the frequency distribution of experimental observations of the labelled particle according to the lateral position in which the observations occurred.

Figure 7.3 - Motion of the labelled particle in the numerical experiments. The convective steps, C , are the horizontal arrows and the diffusive steps, D , are the vertical arrows and the viewing-window detectors are denoted by the letter V . (The proportions have been altered for increased clarity.)

Figure 7.4 - Bar histogram showing distribution of jumps for a 15 sec. minimum time interval and experimental conditions of $\phi = 40\%$ and $\omega = 10 \text{ s}^{-1}$. Vertical bars represent the frequency of jumps to a lateral position plus or minus $0.005 \text{ in. s}^{-1/2}$.

Figure 7.5 - Illustrations of alteration of the distribution of jumps when an undisturbed distribution could result in interference with the wall. a) a distribution for which there is no wall effect. b) and c) distributions after "reflection" from the wall. The mirror image distribution is shown in long dashed lines; the additional component of the distribution from the mirror-image distribution is cross-hatched. The area of the additional component is equal to the portion

of the original distribution which would pass through the wall, shown by the short dashed lines.

Figure 7.6 - Effect of a gradient or force field upon the size distribution of jump.

a) unshifted distribution of jumps.

b) variation of the gradient or force field with channel positions. For a uniform gradient or force field this would be a horizontal line and the magnitude of the shift illustrated in (c) would not depend upon channel location).

c) During the convective step the position has a lateral position, y_0 . In the presence of the gradient illustrated in (a) the distribution of diffusive steps will be selected from the frequency distribution of jumps shown by the solid line. The dashed line is the unshifted frequency distribution which would be used if there were no gradient or force field.

Figure 7.7 - Bar histogram of results of the numerical experiments with no gradients or force fields. Thin, closed bars show the frequency of observation with the constant-time-interval detector. Wide, open bars show frequency of observation for the viewing-window detector.

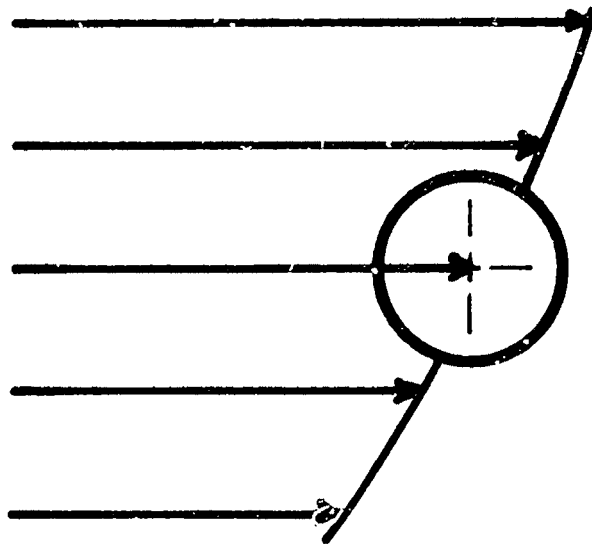
Figure 7.8 - Bar histogram of results of numerical experiments with a gradient or force field. Notation of bars is identical to that in Fig. 7.8.

Figure A2.1 - Cross-sectional view of circular-cylinder Couette apparatus used in the preliminary experiments. Dimensions are $h = 8.0$ in., $w = 1.25$ in., and $R_1 = 5.50$ in.

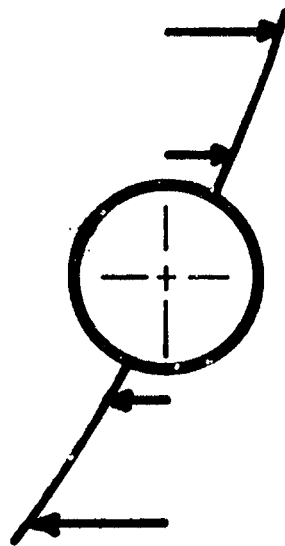
Figure A2.2 - Theoretical velocity profile and velocity measurements in the channel of the circular-cylinder Couette apparatus shown in Figure A2.1.

Figures A2.3 and A2.4 - Graphs of the number of particles per unit length of barrier which have crossed the barrier t seconds after the barrier is lifted. The concentration, c_o , is the area concentration of colored particles on either side of the barrier at the beginning of the experiment. In Figure A2.3 $c_o = 20\%$; in Figure A2.4 $c_o = 40\%$.

Figure A4.1 - Graph of $D/a^2\omega$ from Bailey [37] showing the data gathered by him and the data of this thesis. In the notation of Bailey, $D_{obs} = D$.



(a)



(b)

FIGURE 1.1

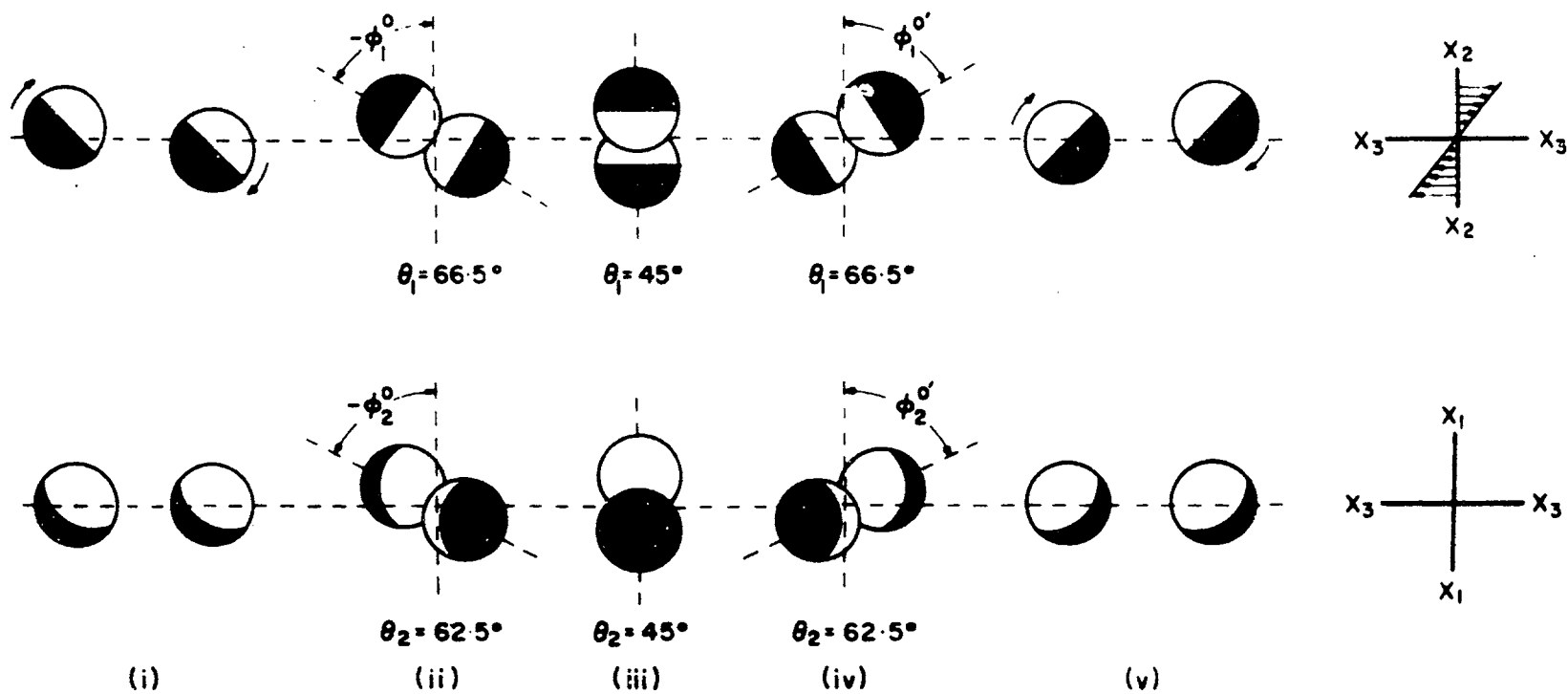
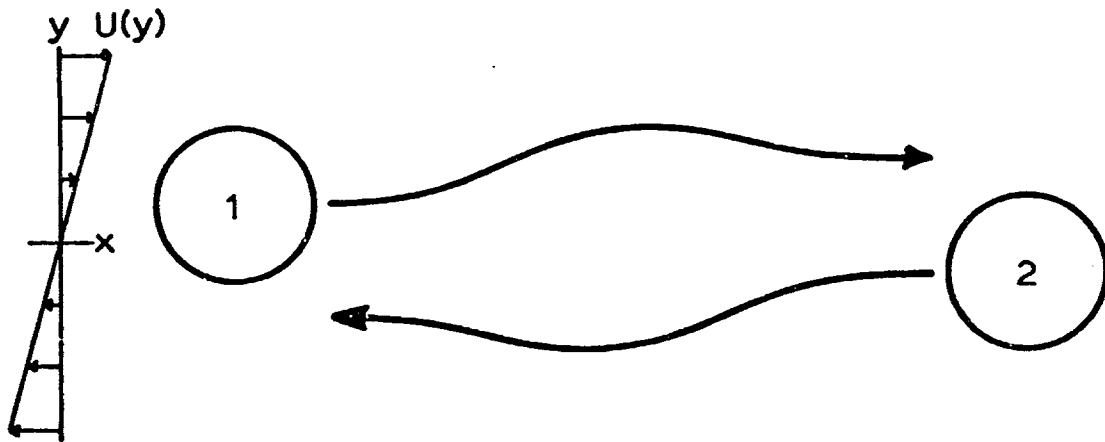
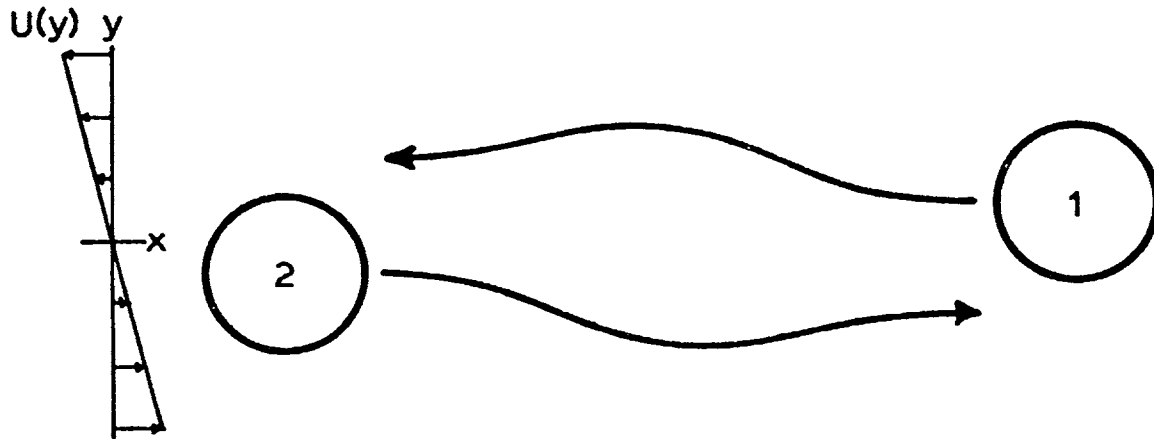


FIGURE 1.2

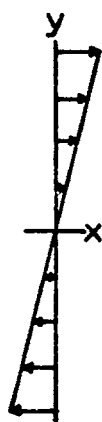


A. Front view showing an increase in the lateral separation.

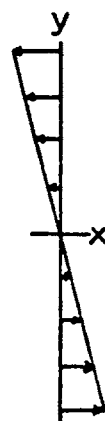


B. Rear view of (A), which should be identical to the reversed flow of (A).

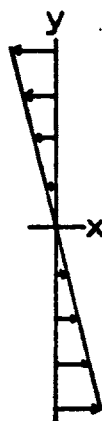
FIGURE 2.1



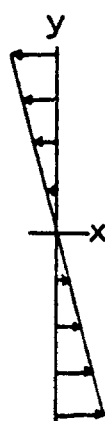
A. Front view of three particles about to undergo an interaction.



B. Relative positions after the interaction. Reversed flow will restore the positions in (A).



C. Rear view of (A).



D. Symmetric arrangement of particles which will undergo interaction without net lateral displacement.

FIGURE 2.2

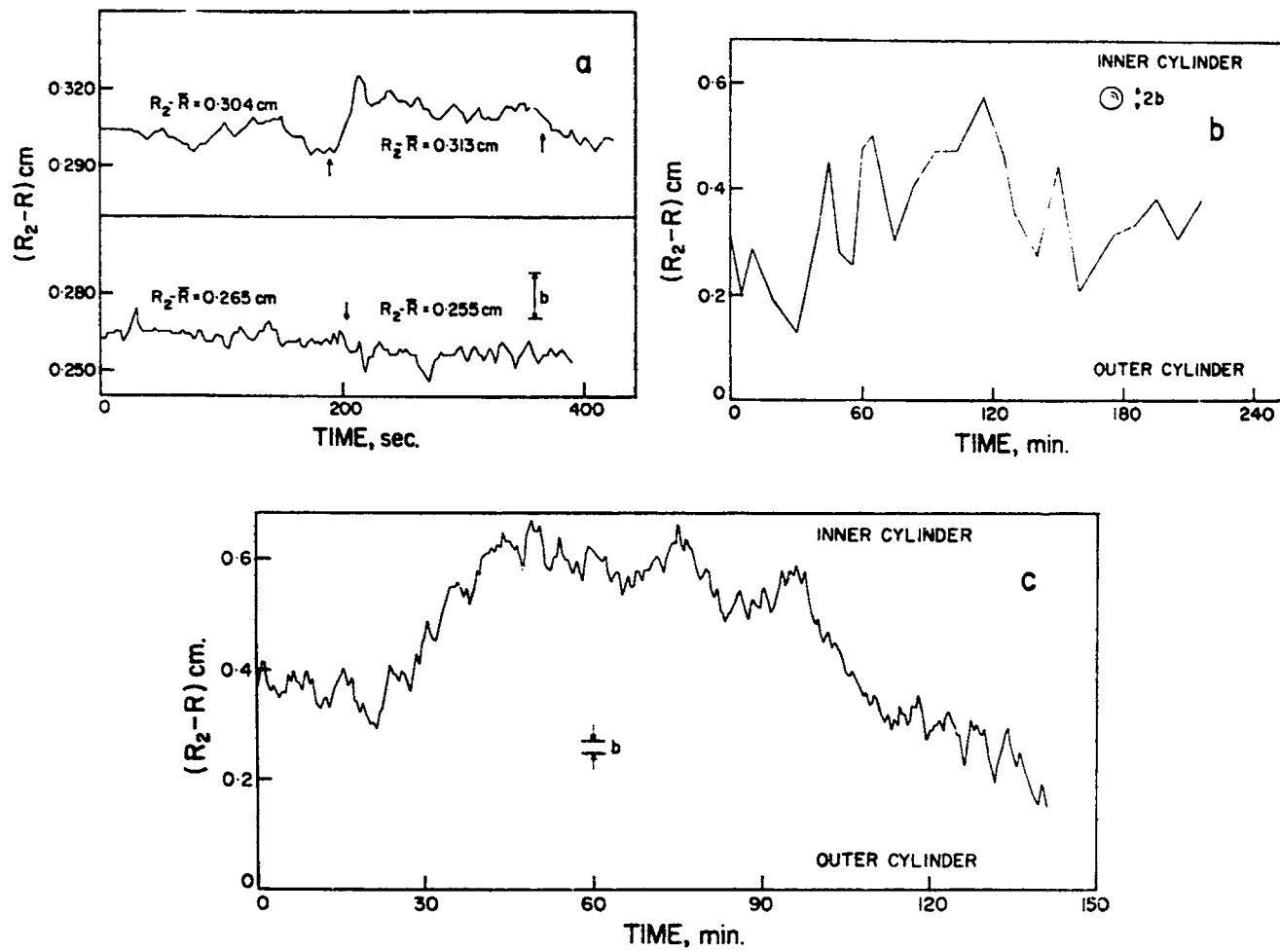


FIGURE 2.3

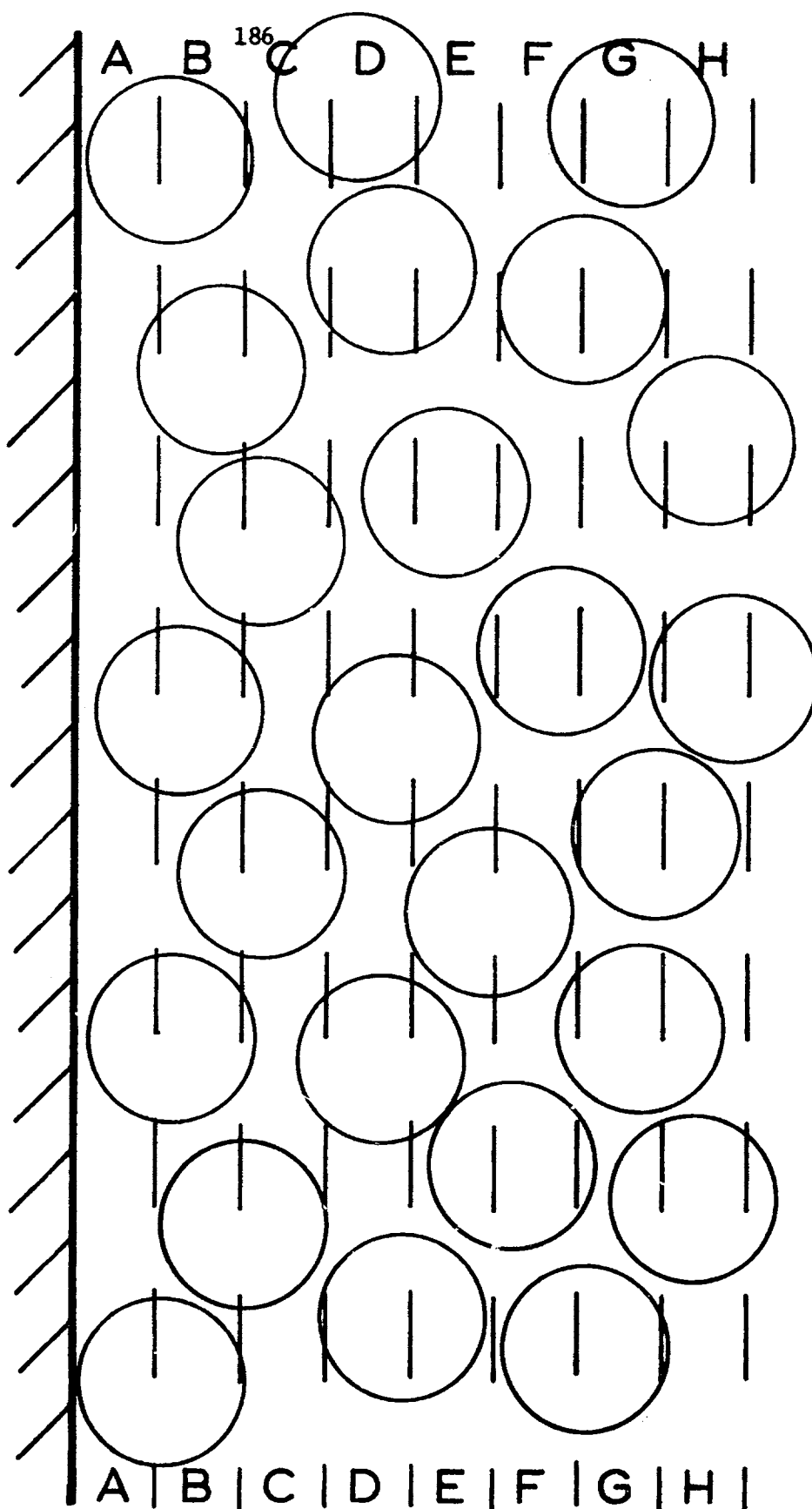


FIGURE 2.4

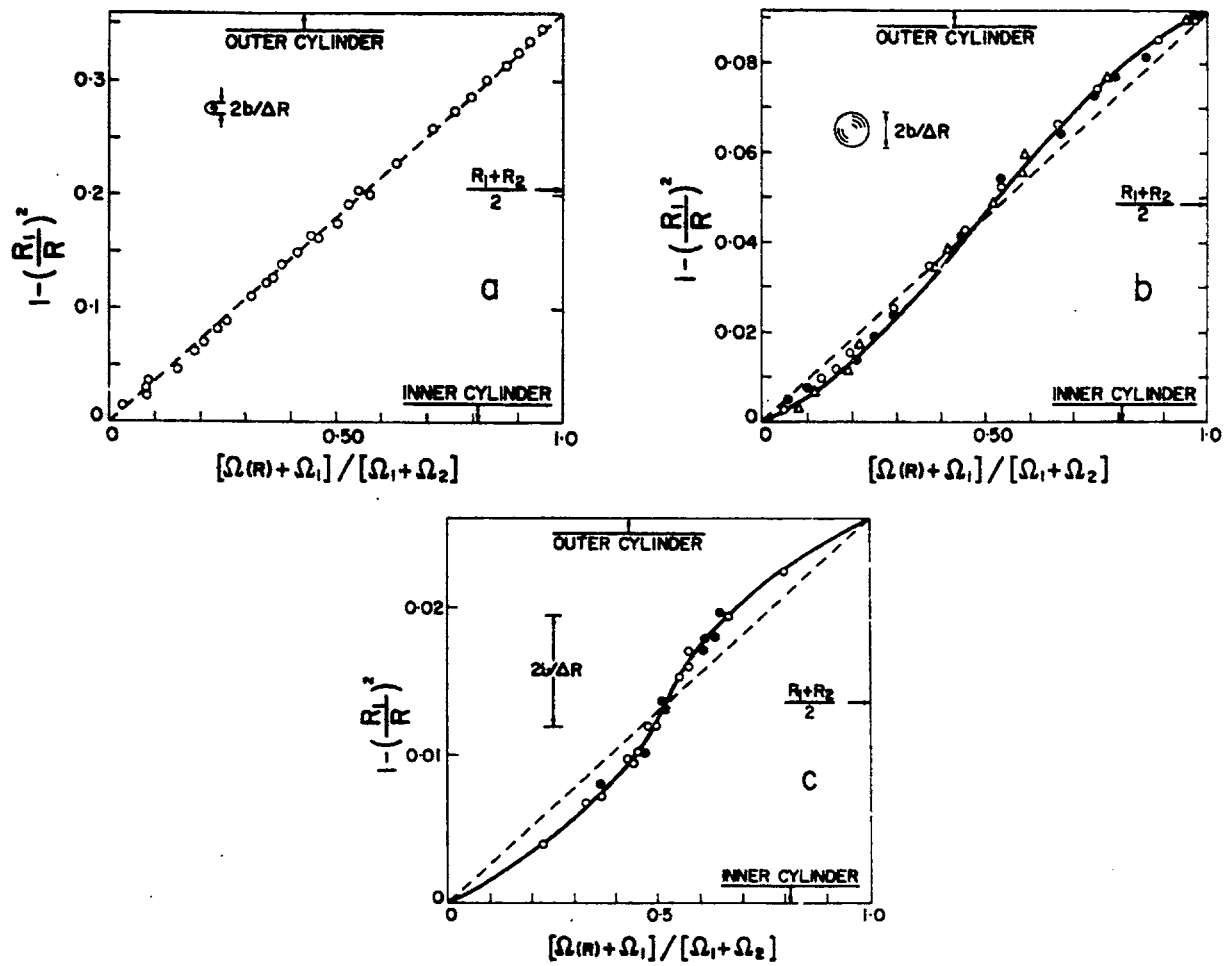
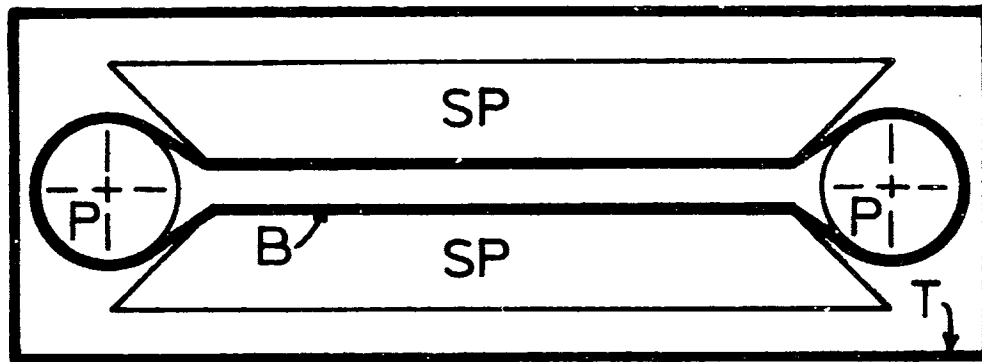
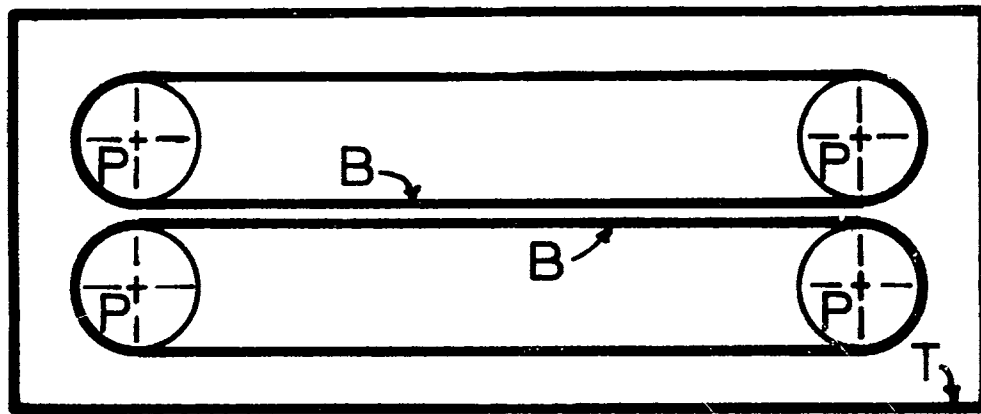


FIGURE 2.5



(a)



(b)

FIGURE 3.1

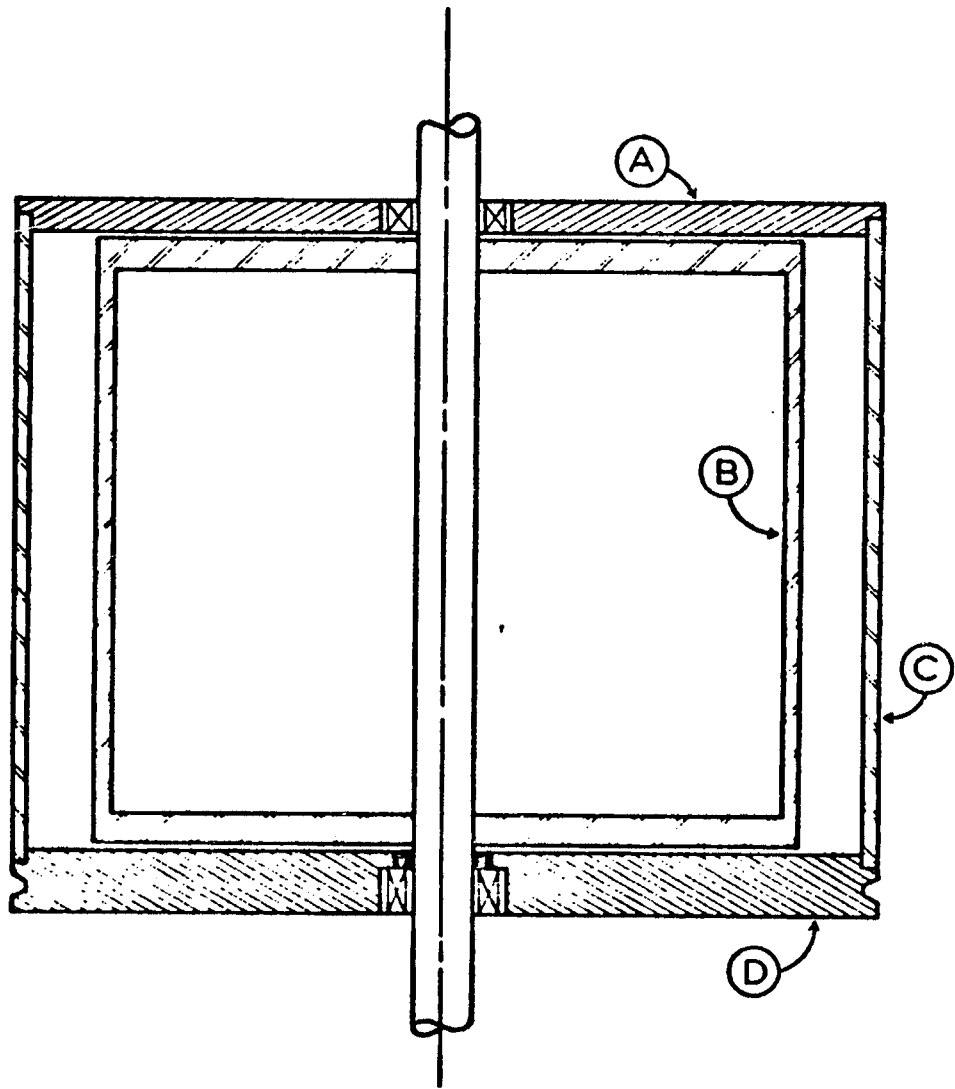


FIGURE 3.2

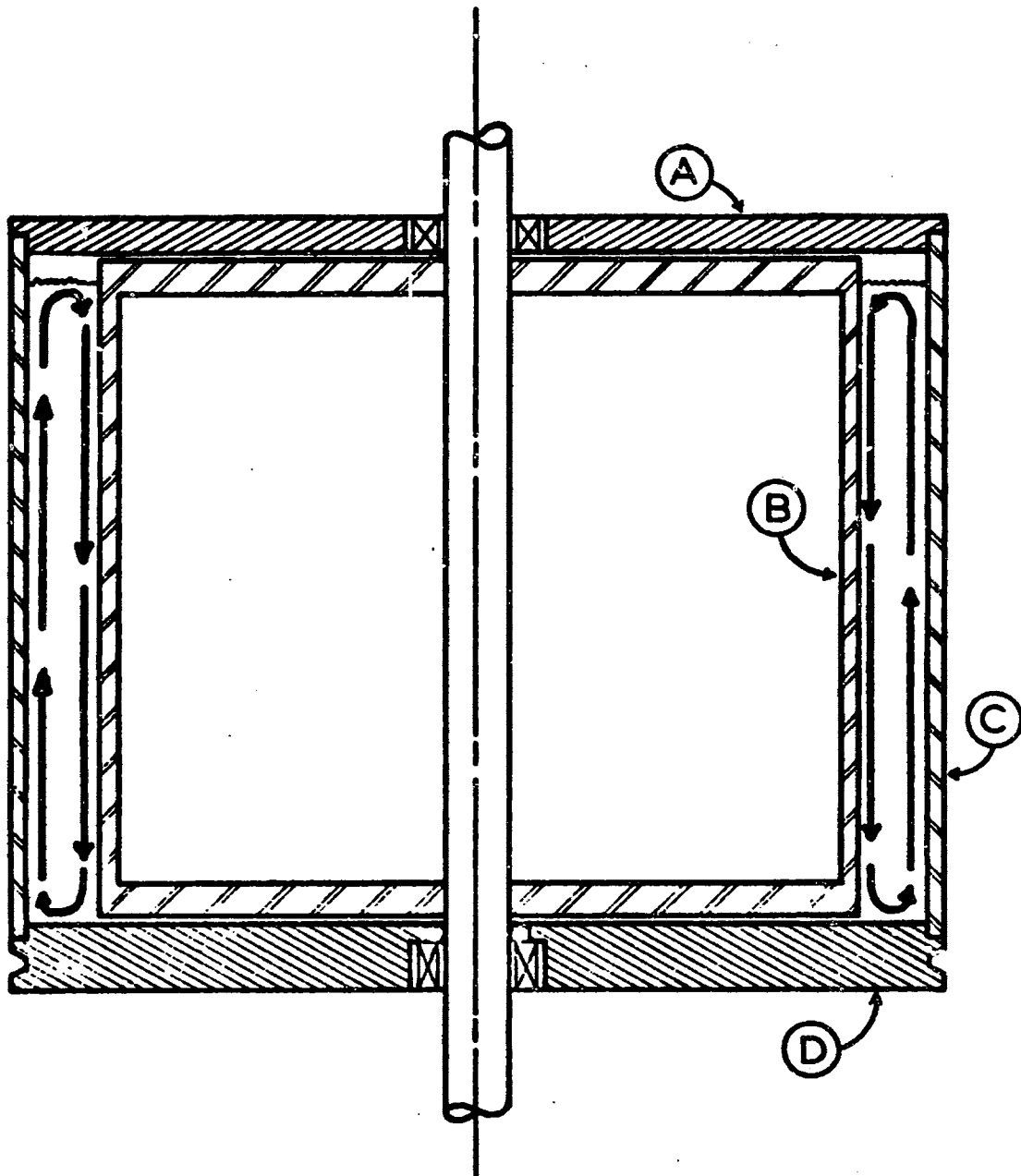


FIGURE 3.3

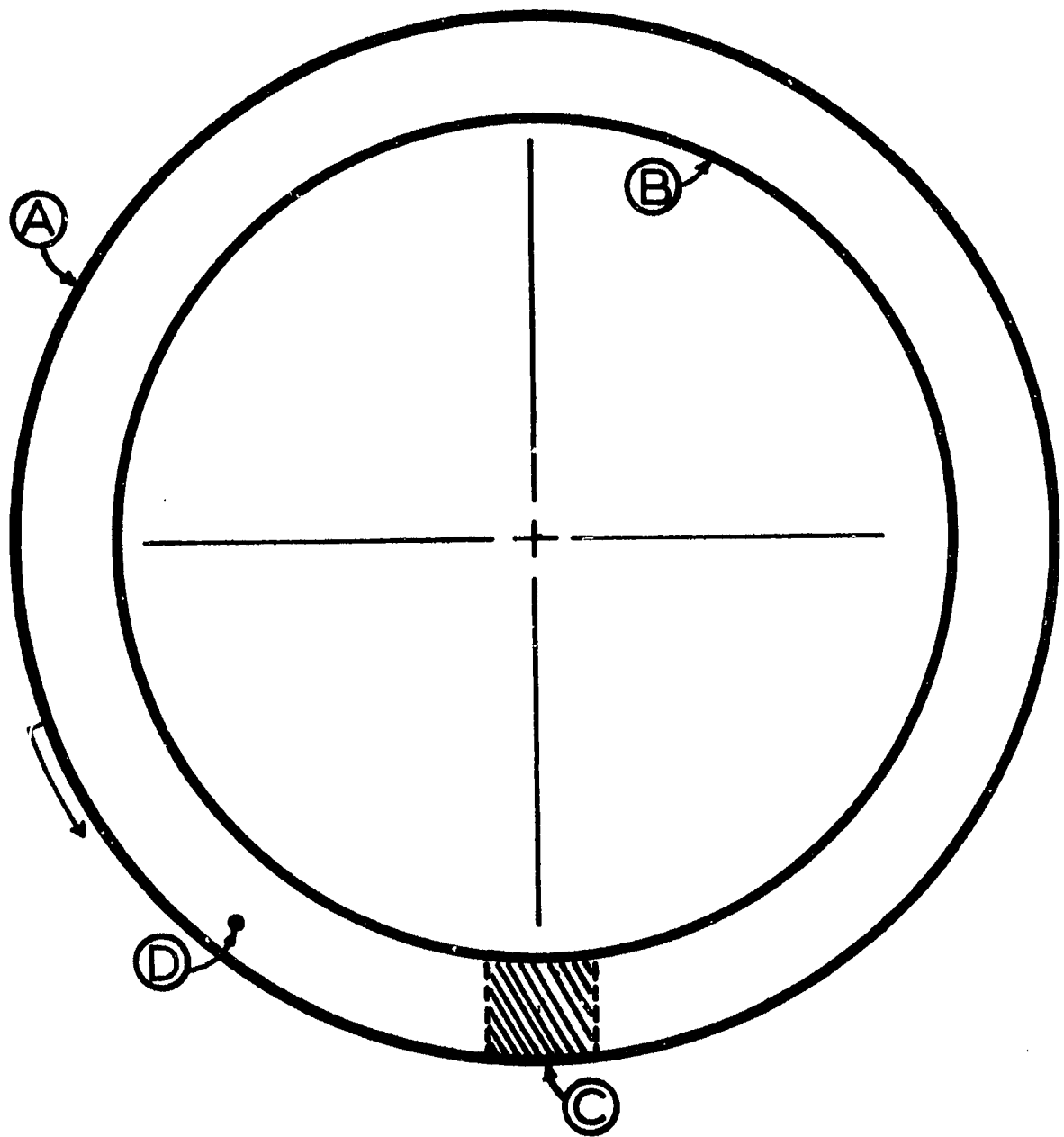


FIGURE 4.1

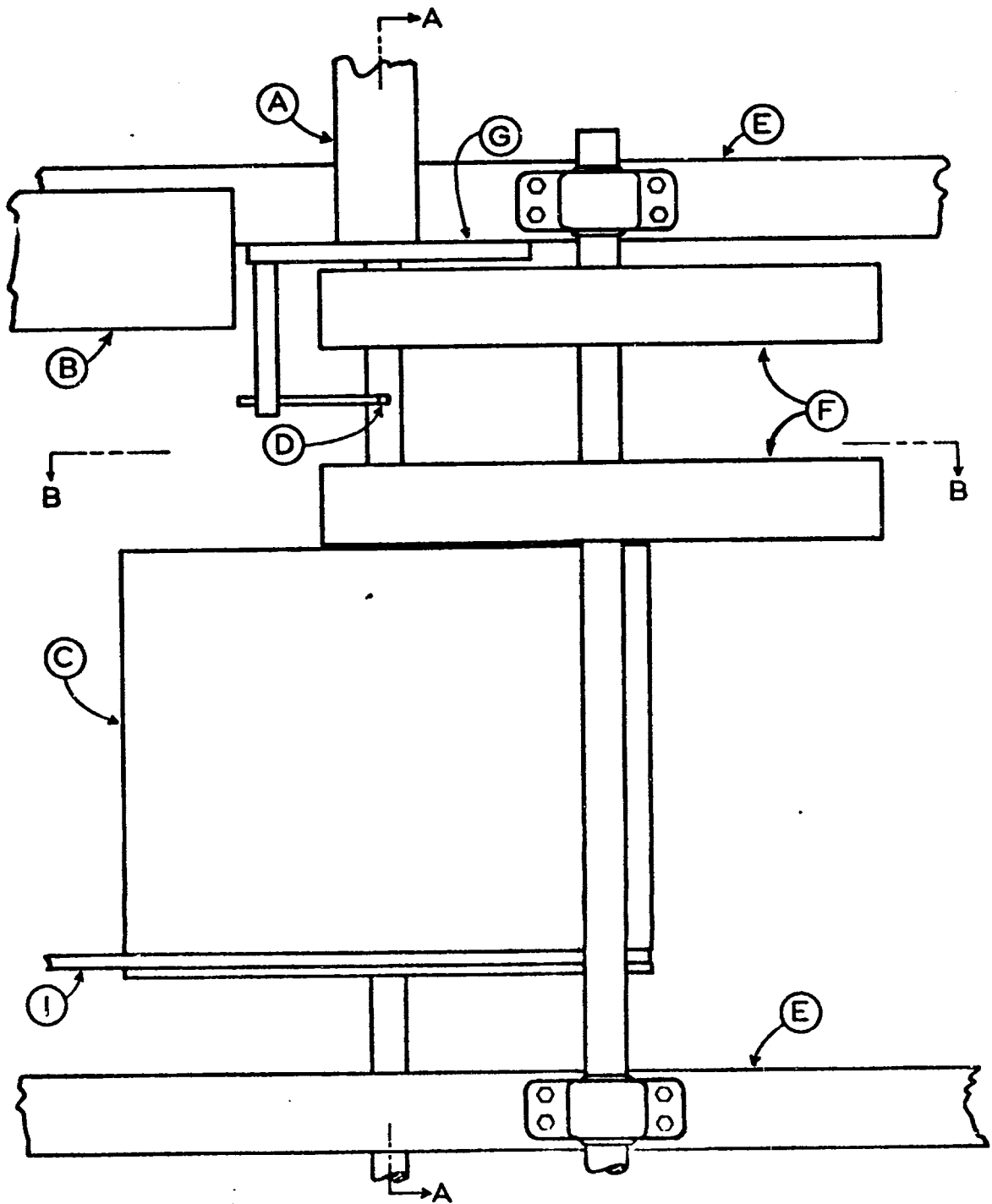
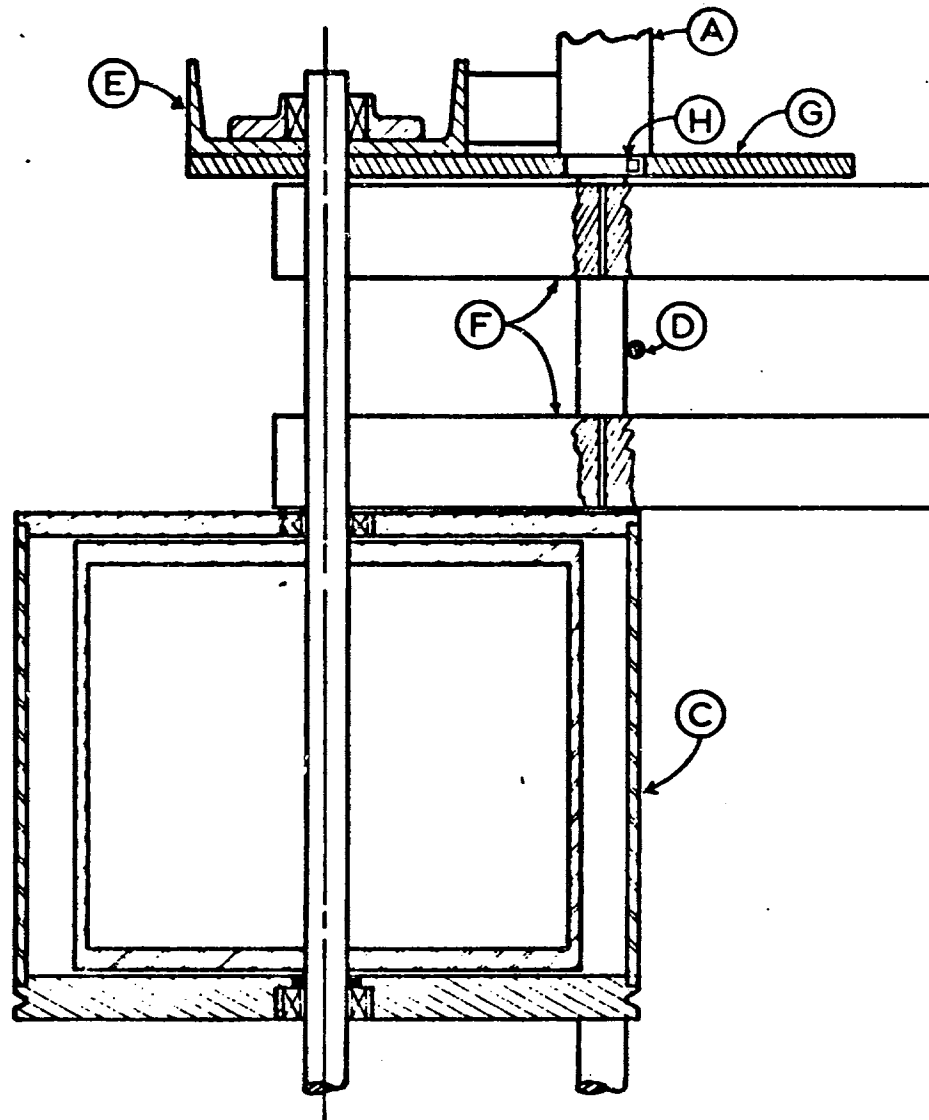
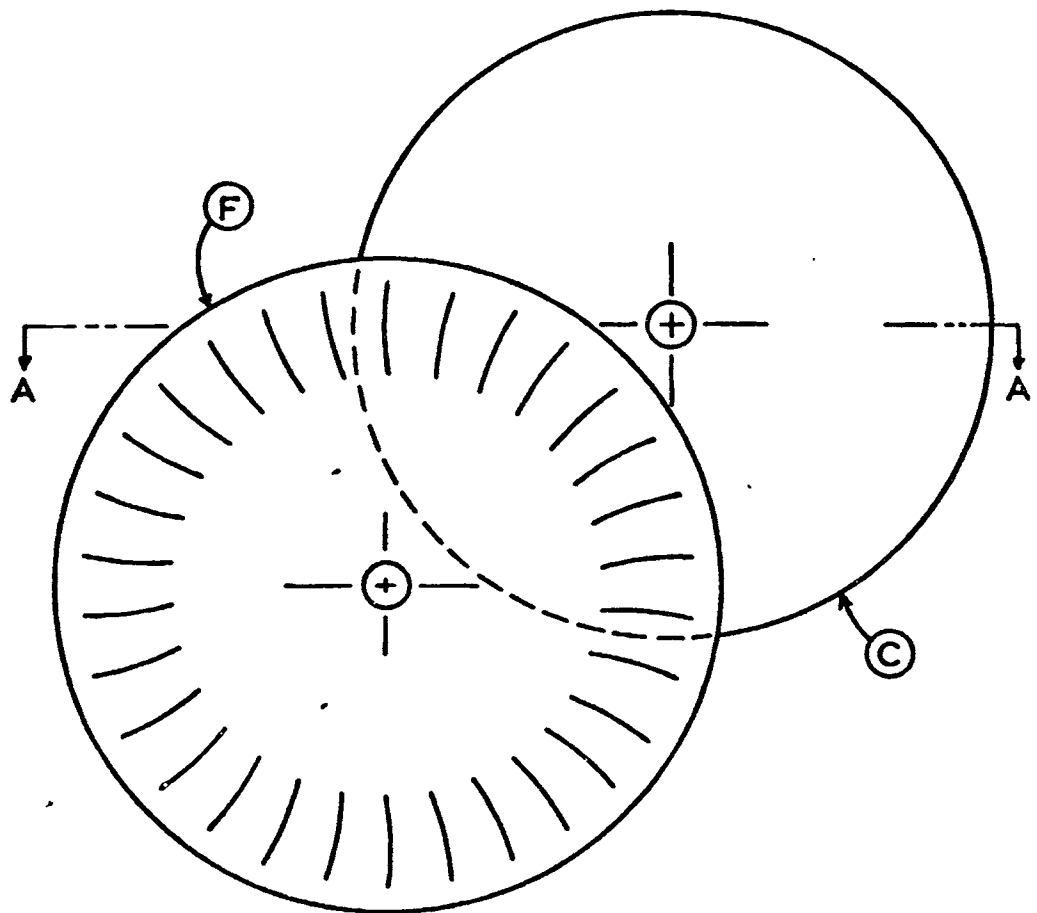


FIGURE 4.2



SECTIONAL VIEW A-A

FIGURE 4.3



SECTIONAL VIEW B-B

FIGURE 4.4

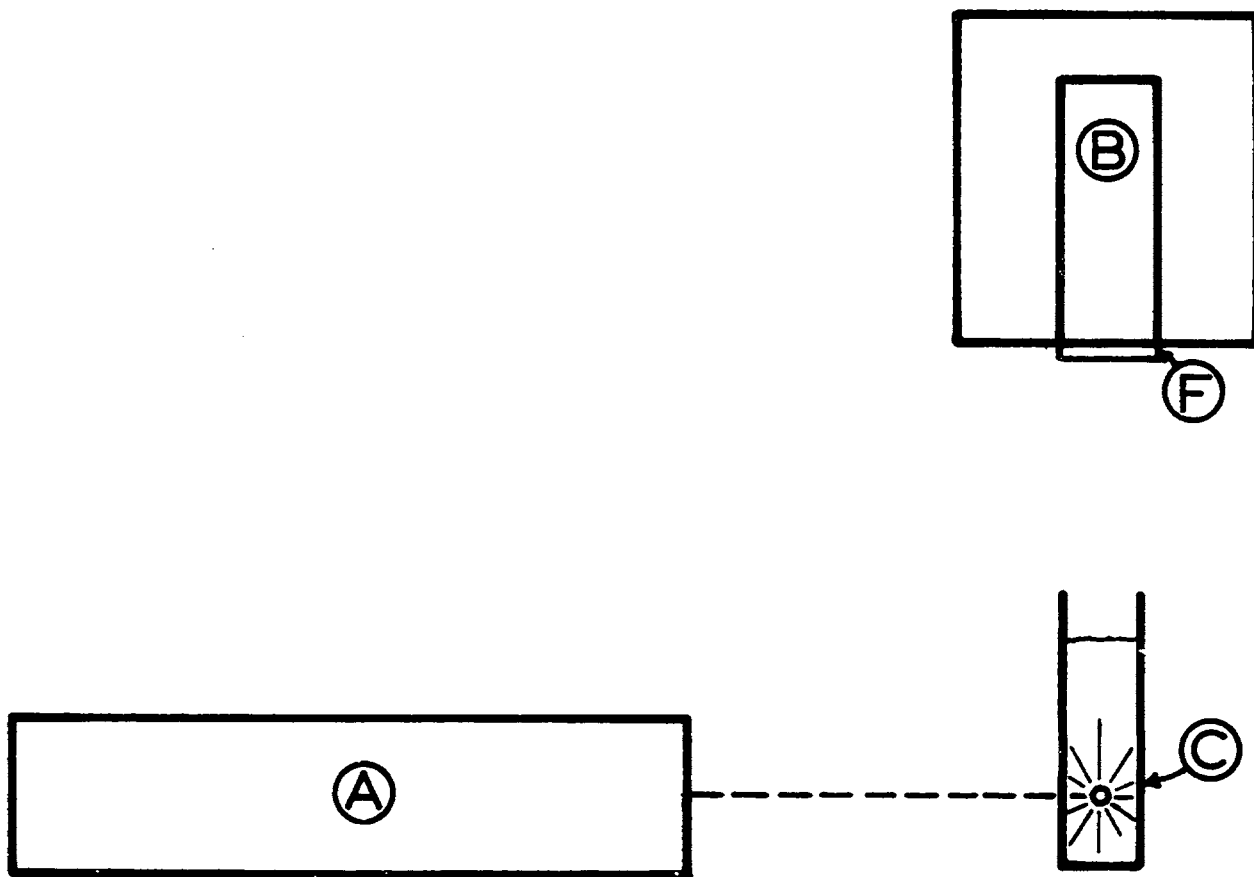


FIGURE 4.5

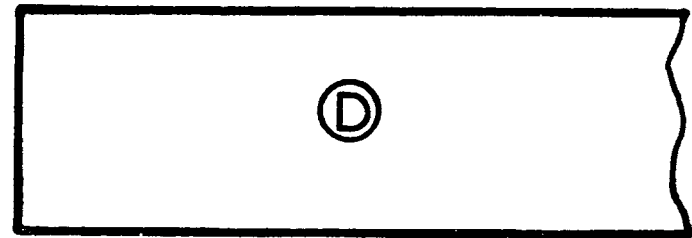
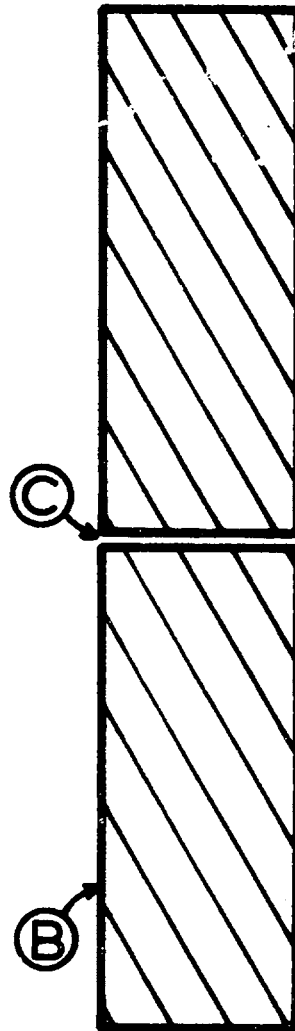
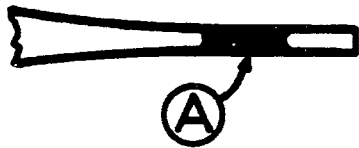


FIGURE 4.6

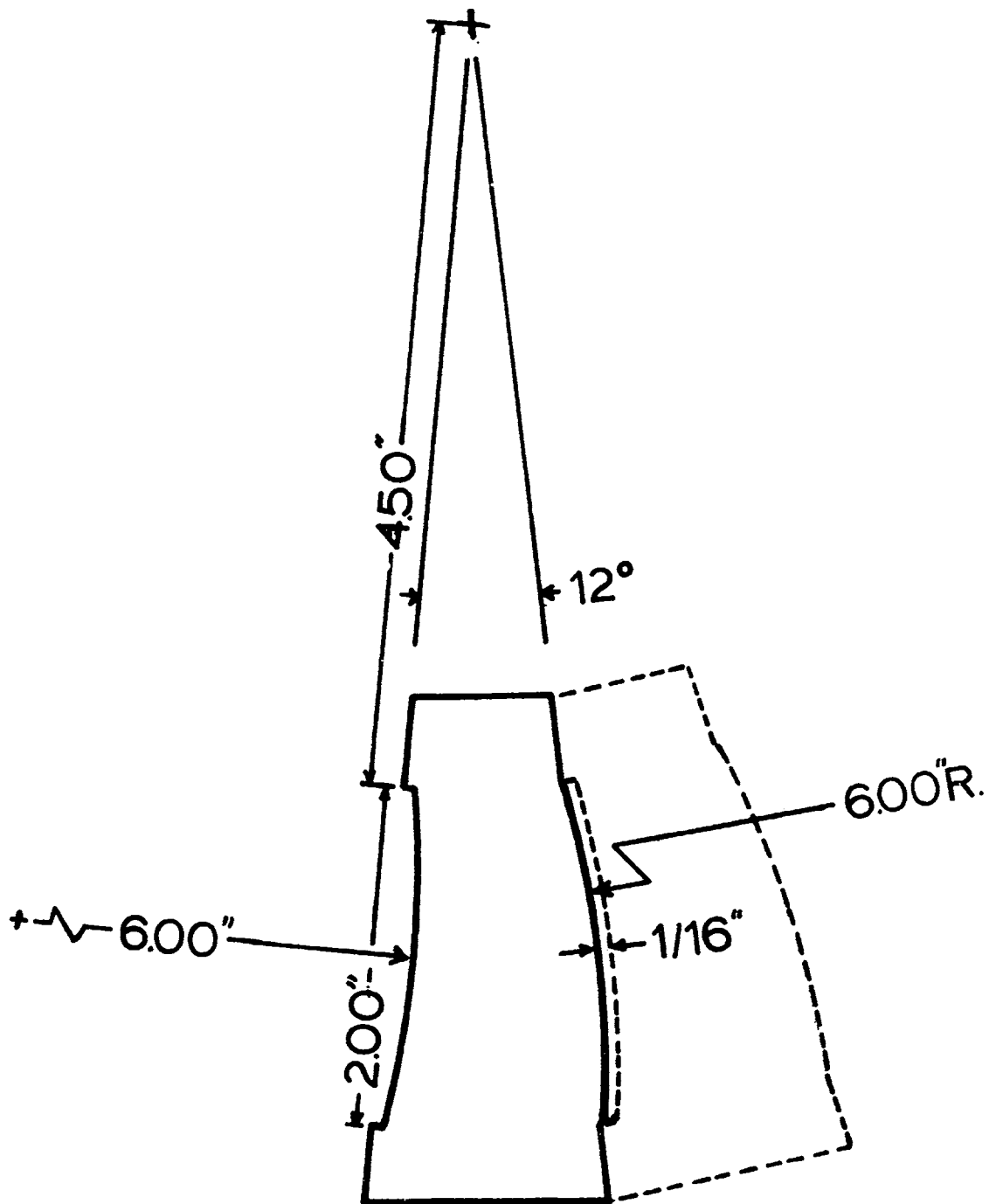


FIGURE 4.7

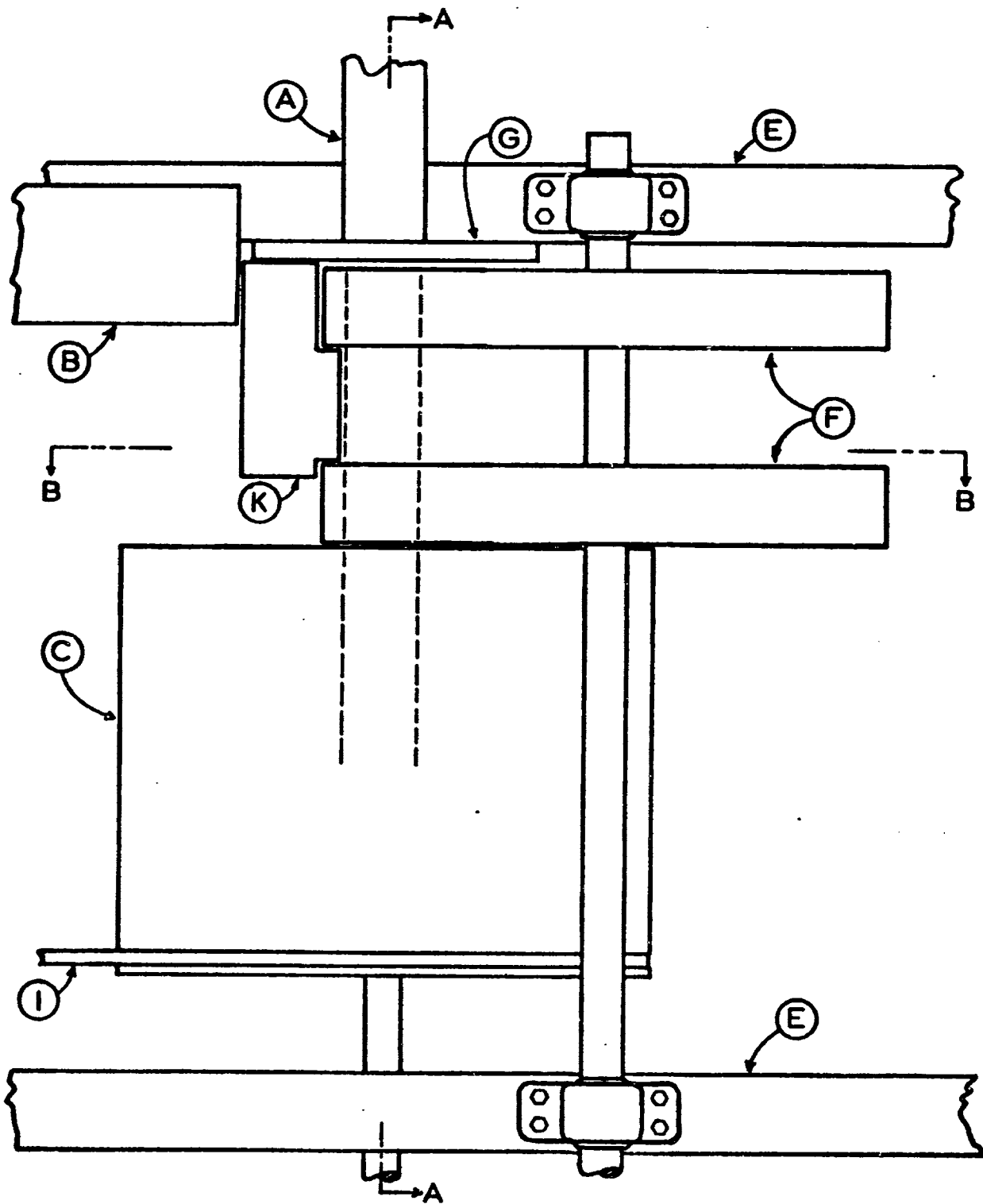


FIGURE 48

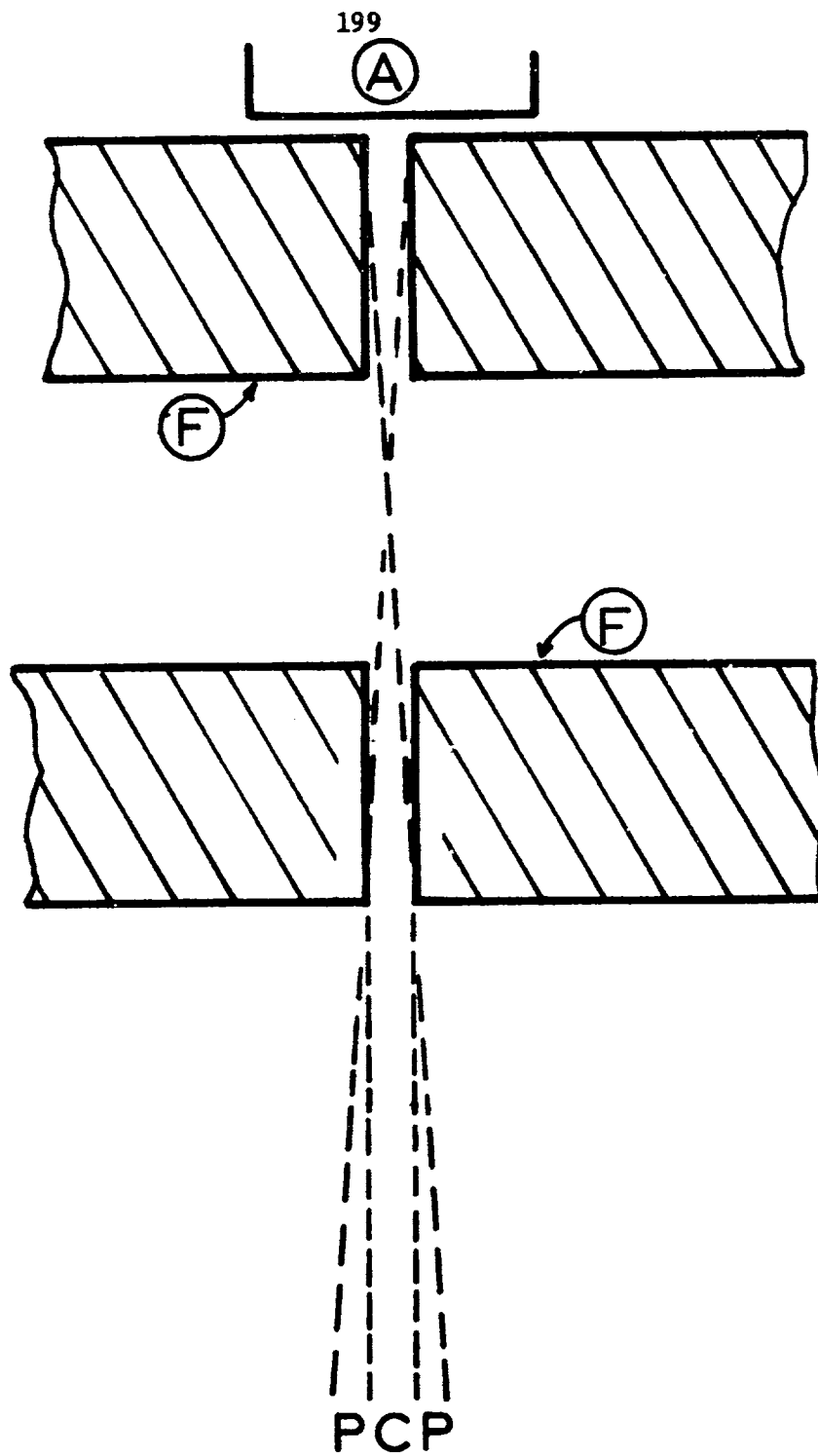


FIGURE 4.9

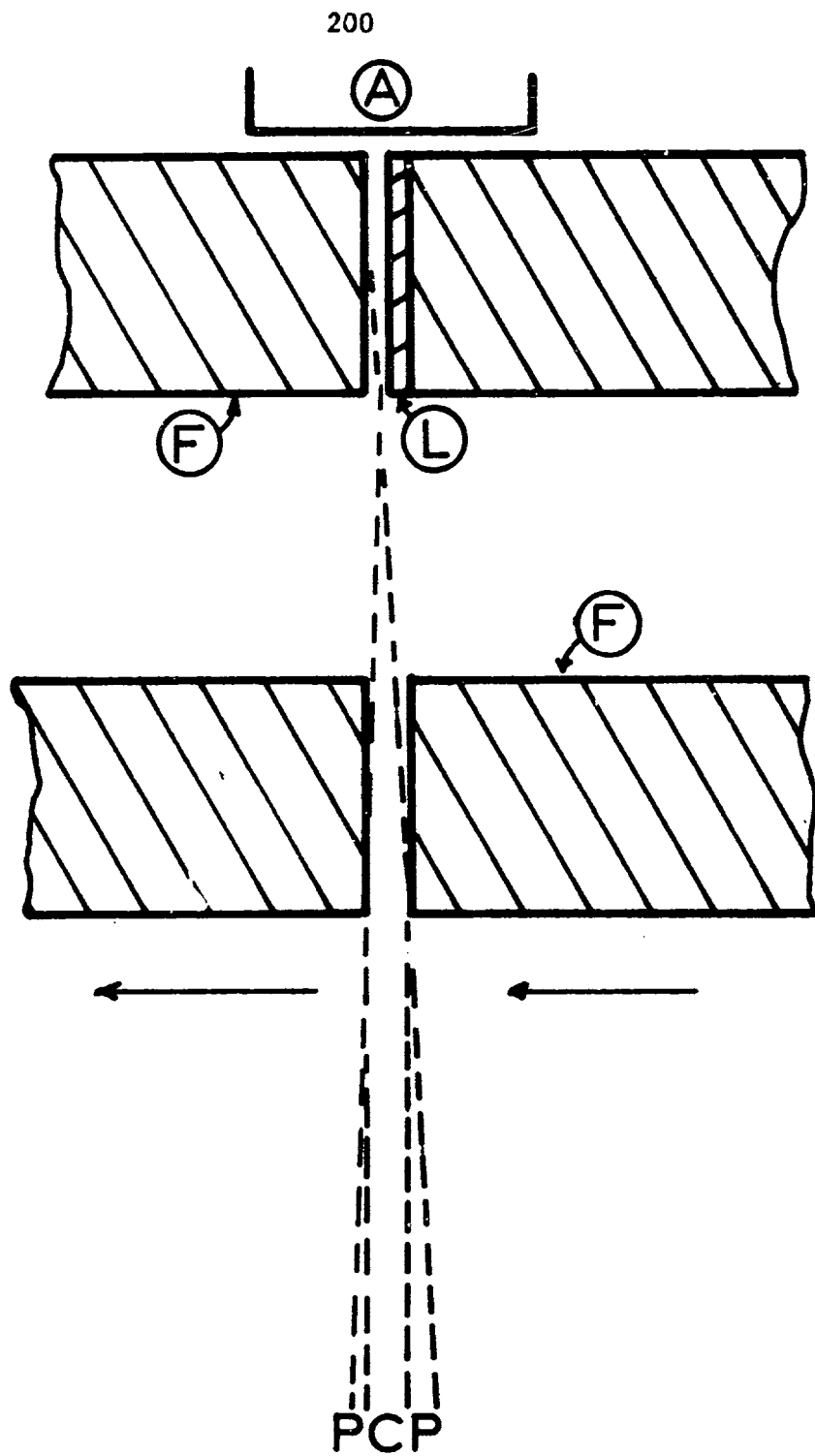


FIGURE 4.10

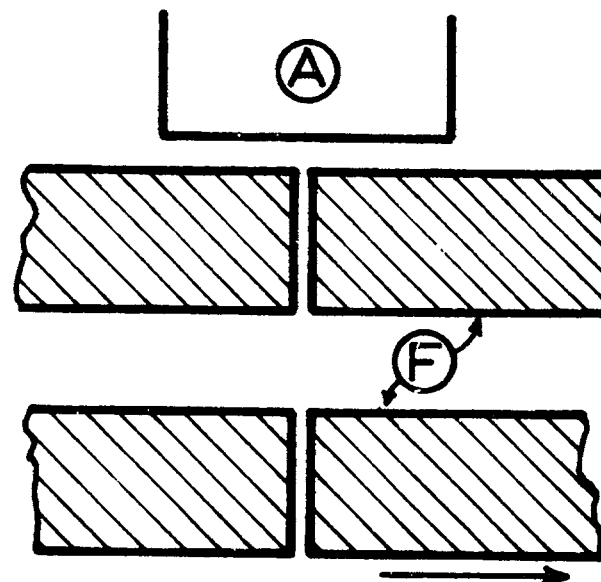
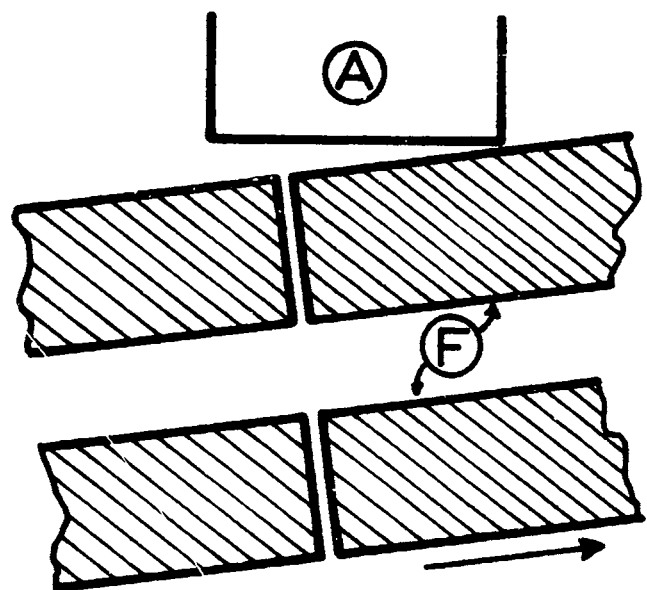
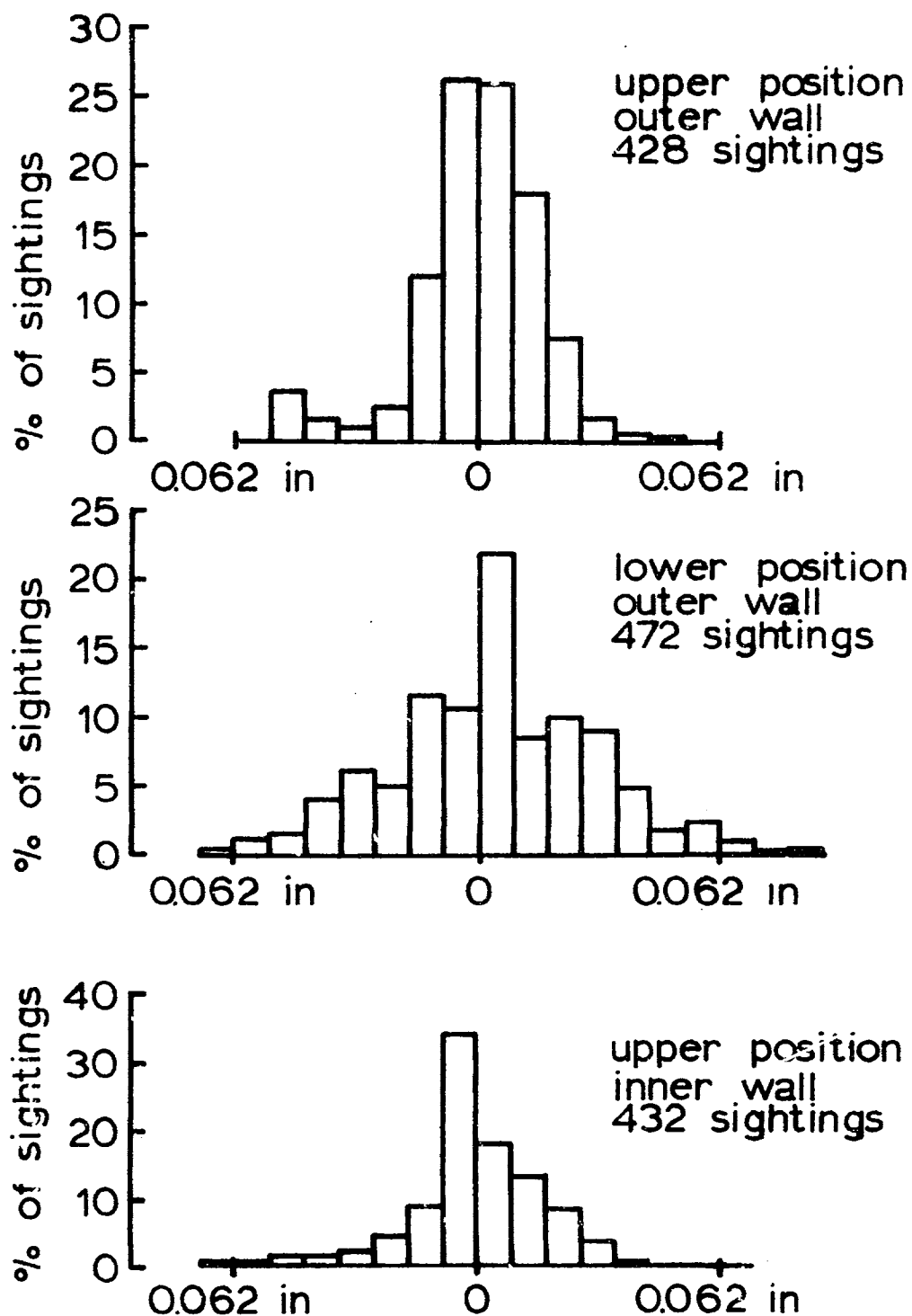
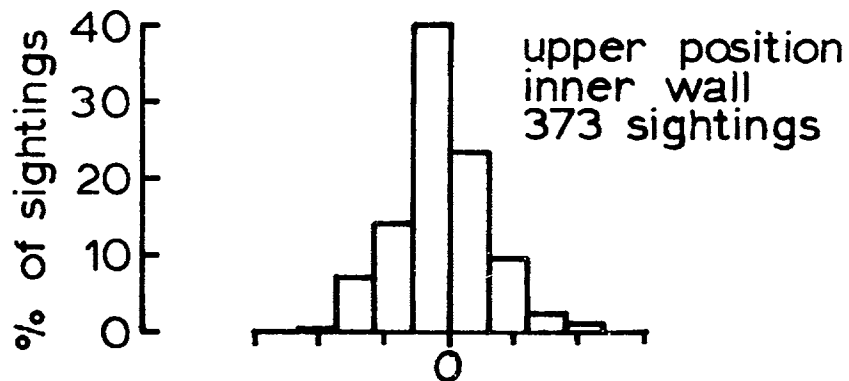
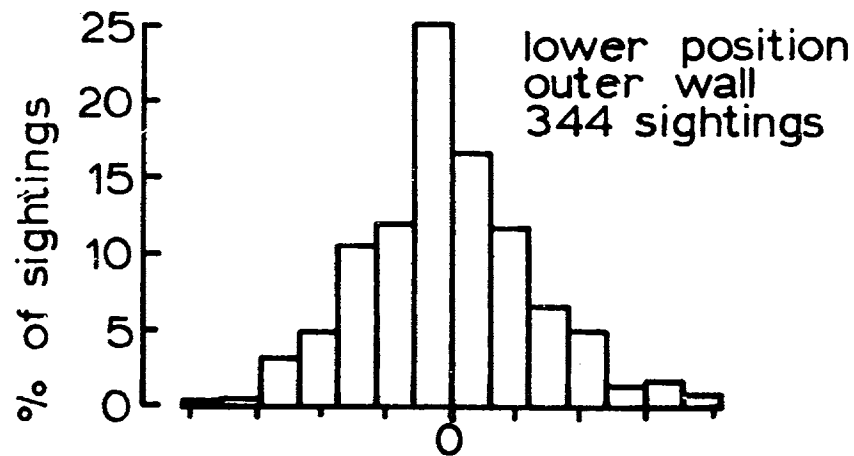
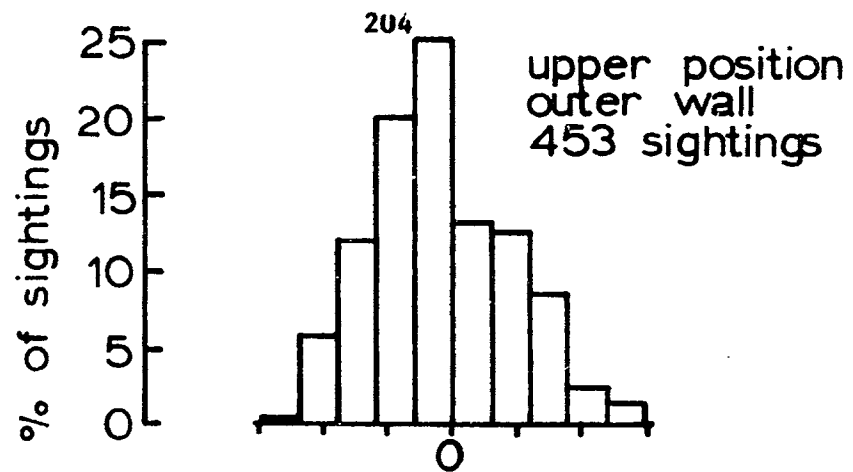


FIGURE 4.11



Distributions of sightings of the large
particle fixed to a wall

FIGURE 4.12



unit distance on the abscissas
represents 0.050 inch

Distributions of sightings of the small
particle fixed to a wall

FIGURE 4.13

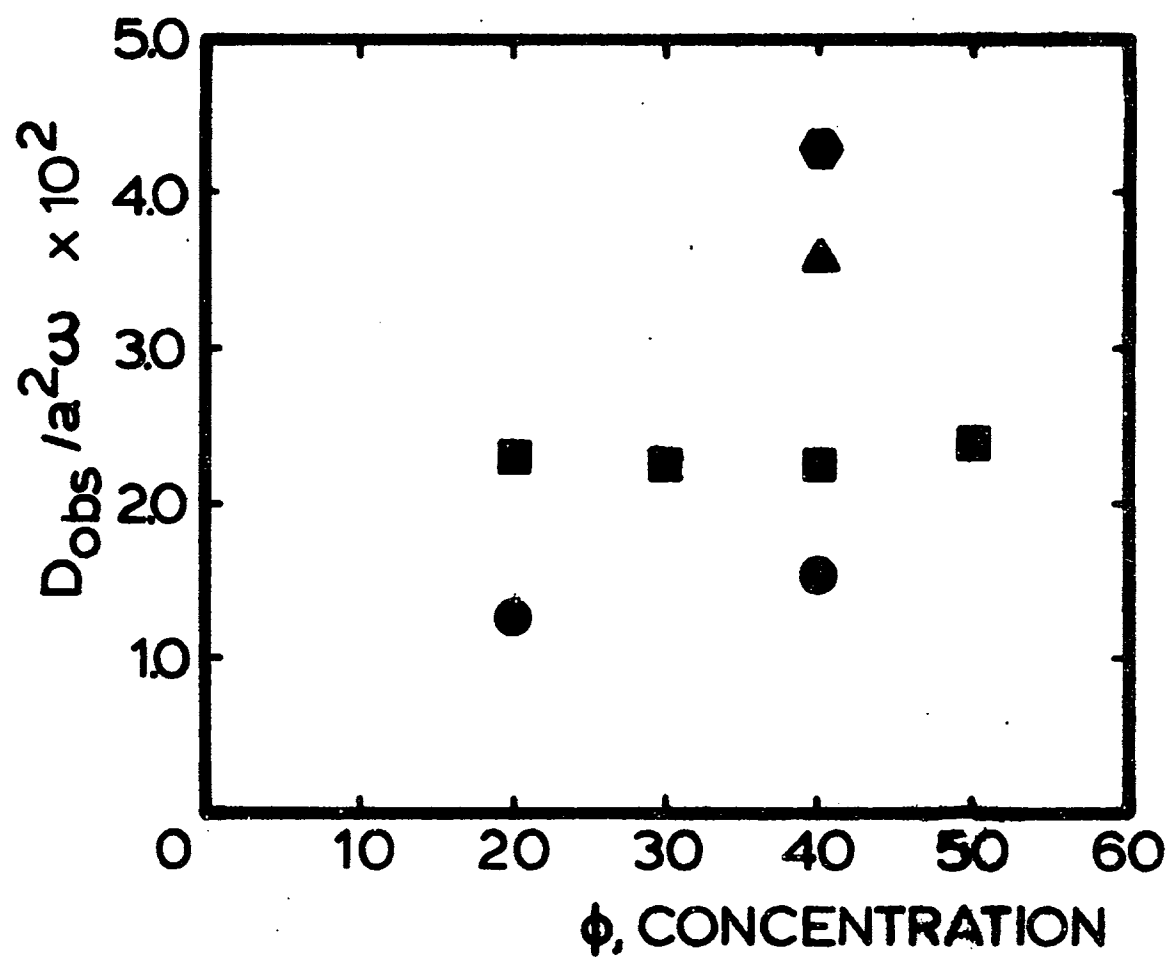


FIGURE 6.1

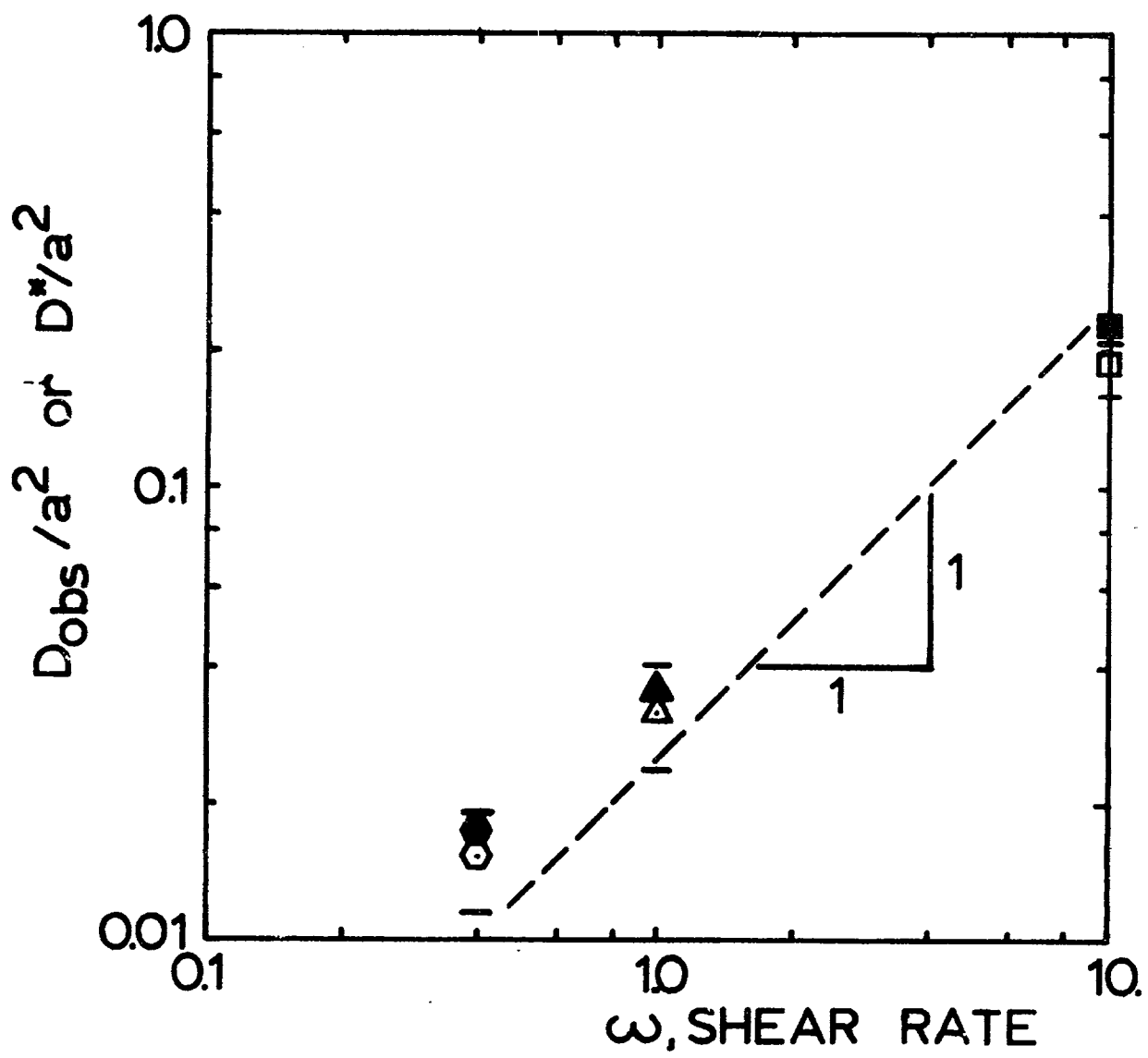


FIGURE 6.2

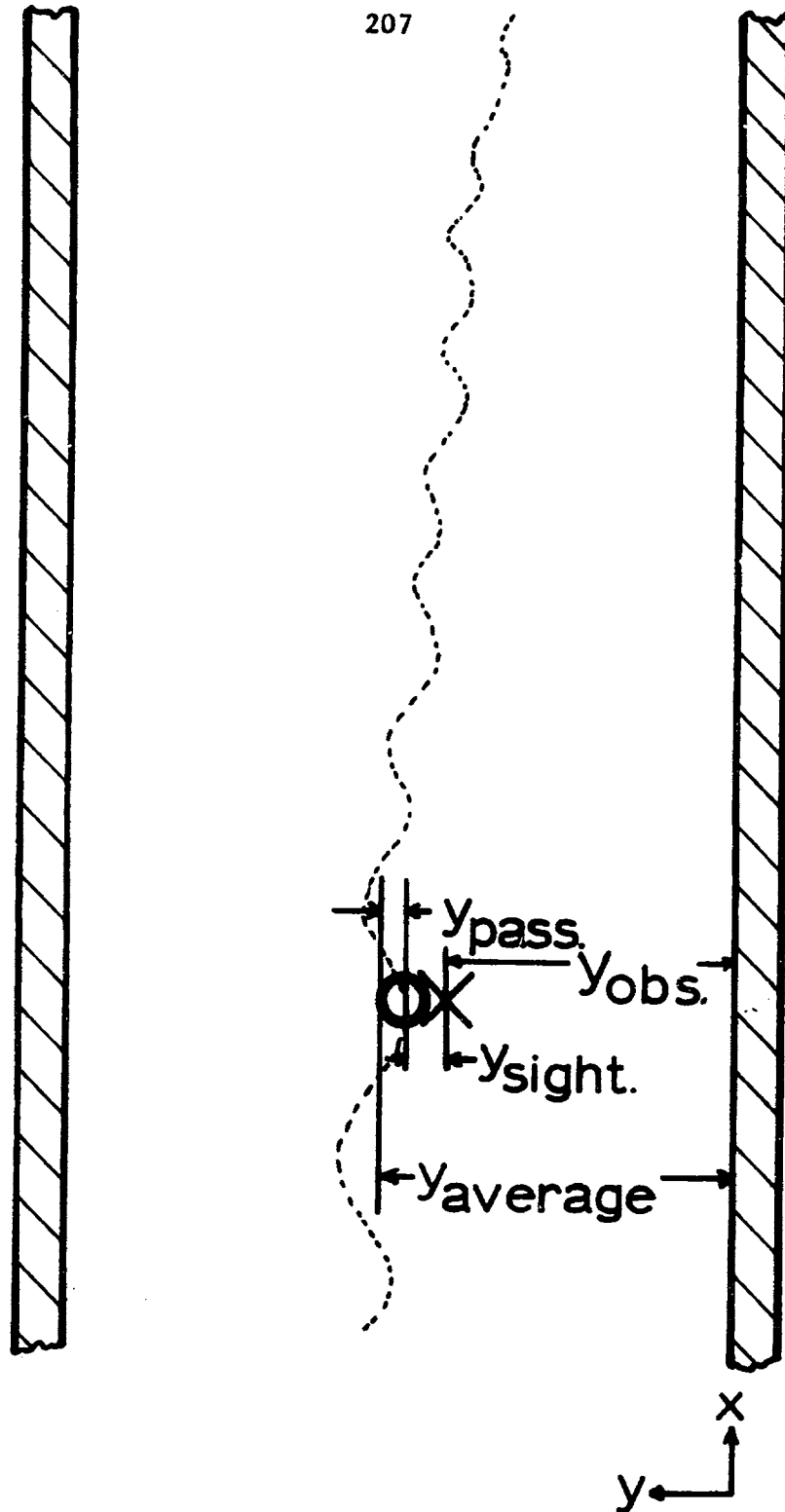


FIGURE 6.3

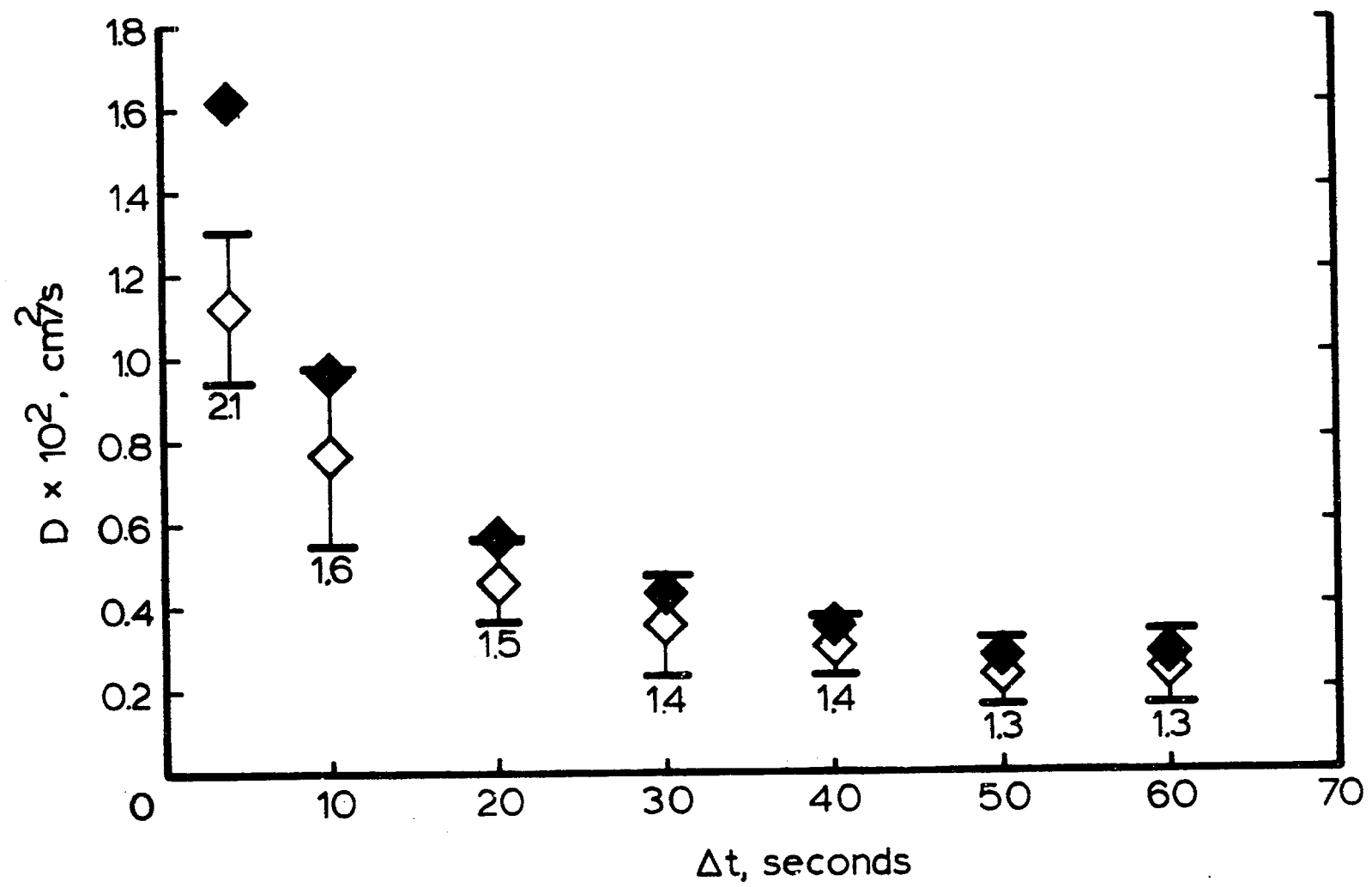


FIGURE 6.4

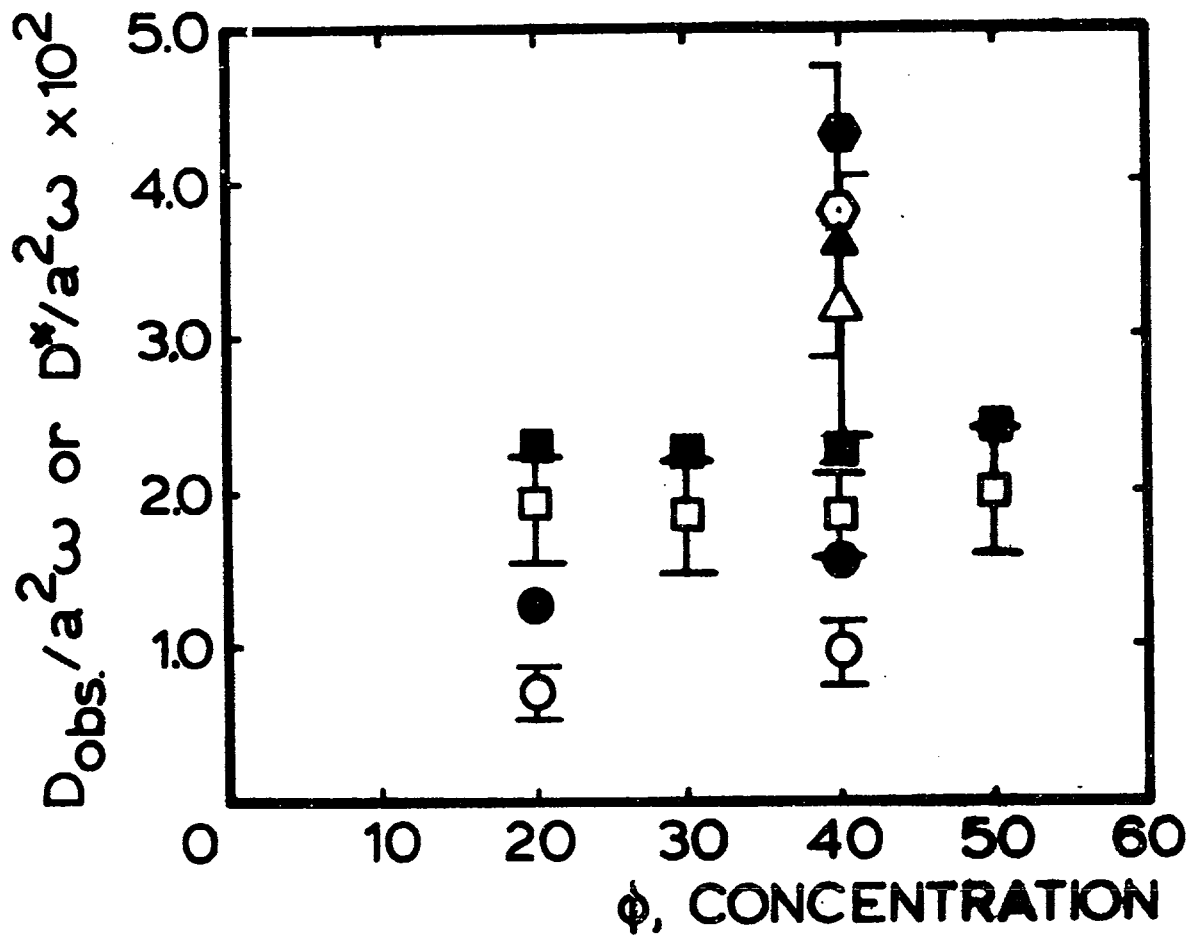


FIGURE 6.5

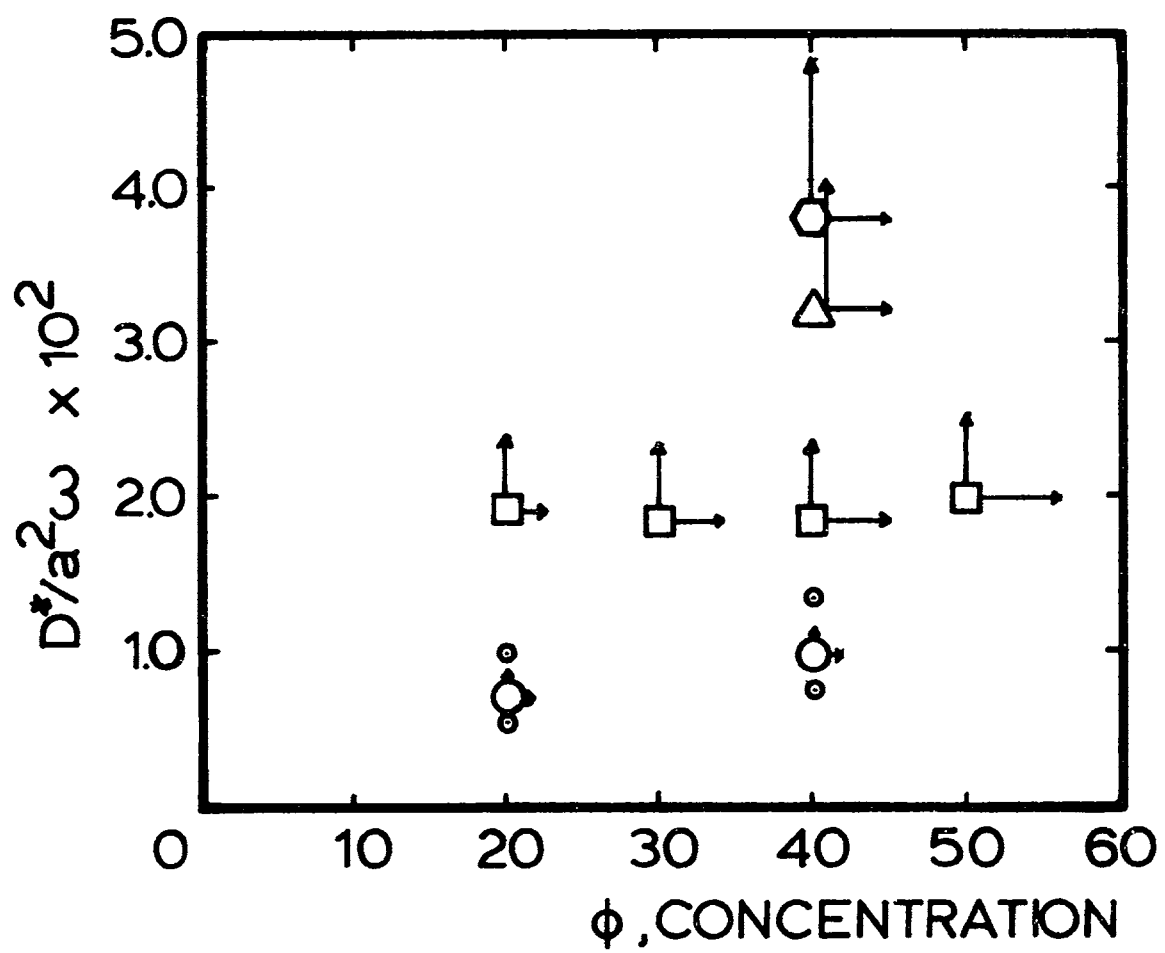


FIGURE 6.6

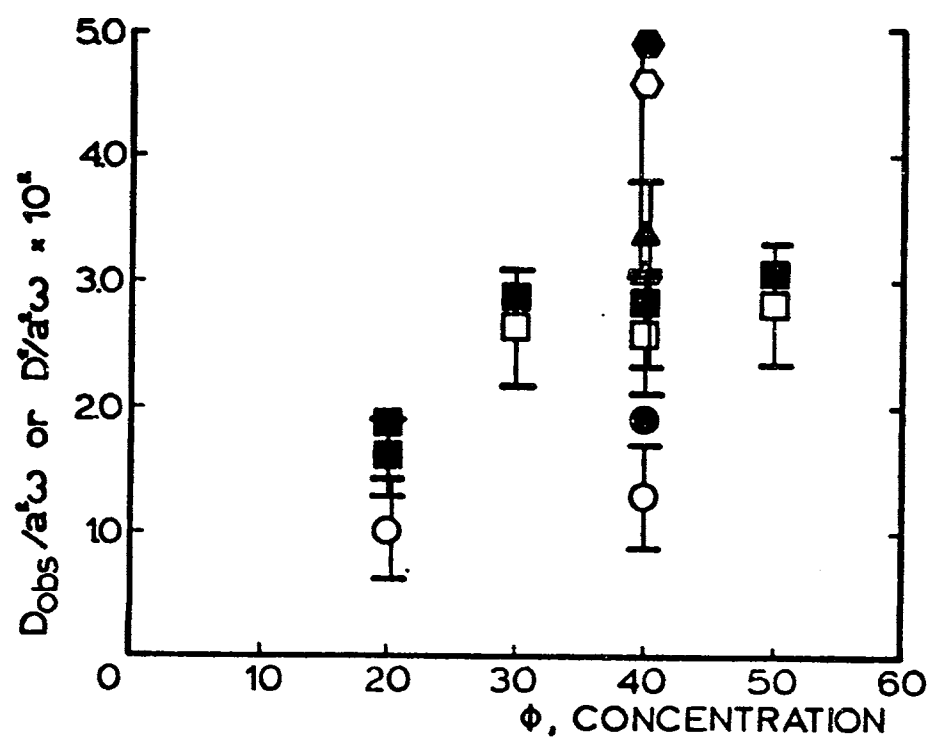


FIGURE 6.7

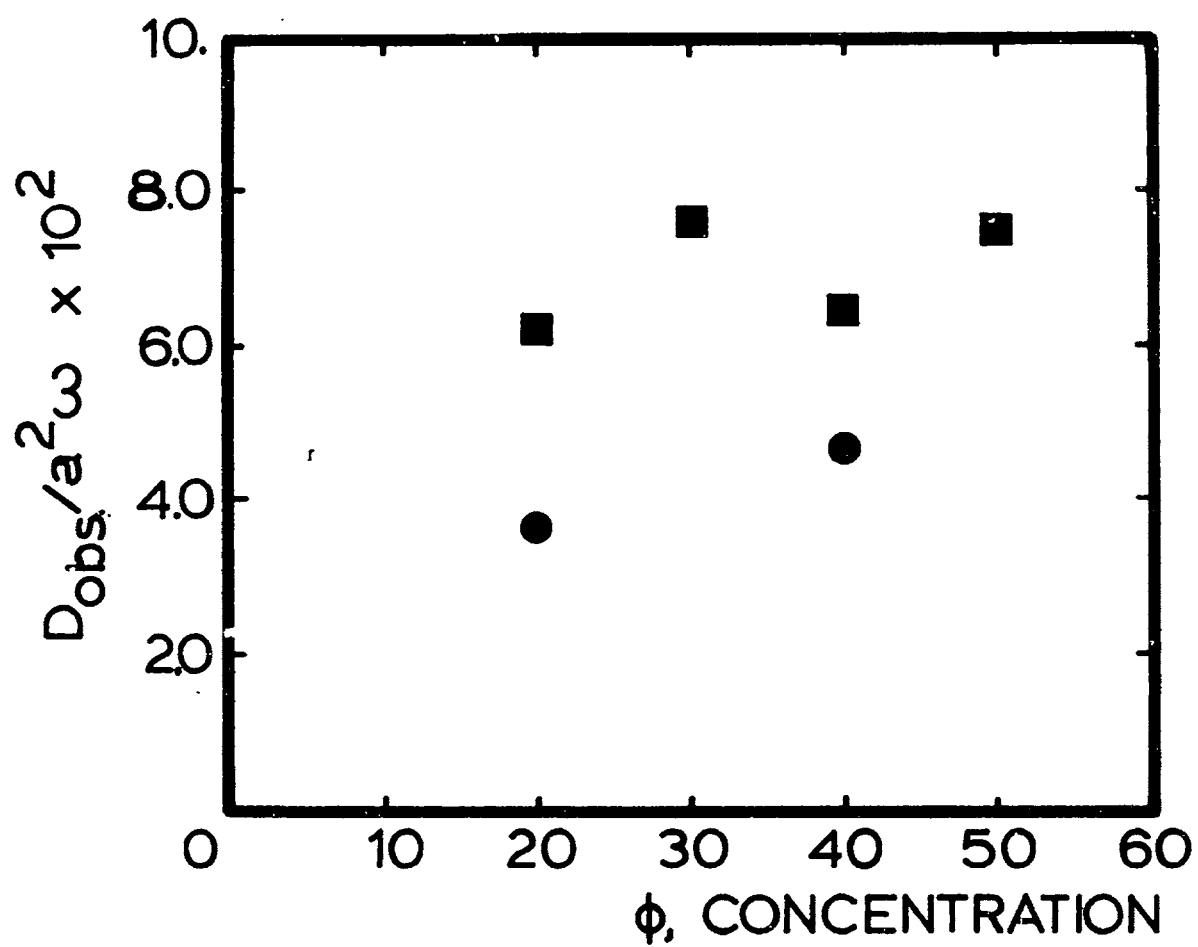


FIGURE 6.8

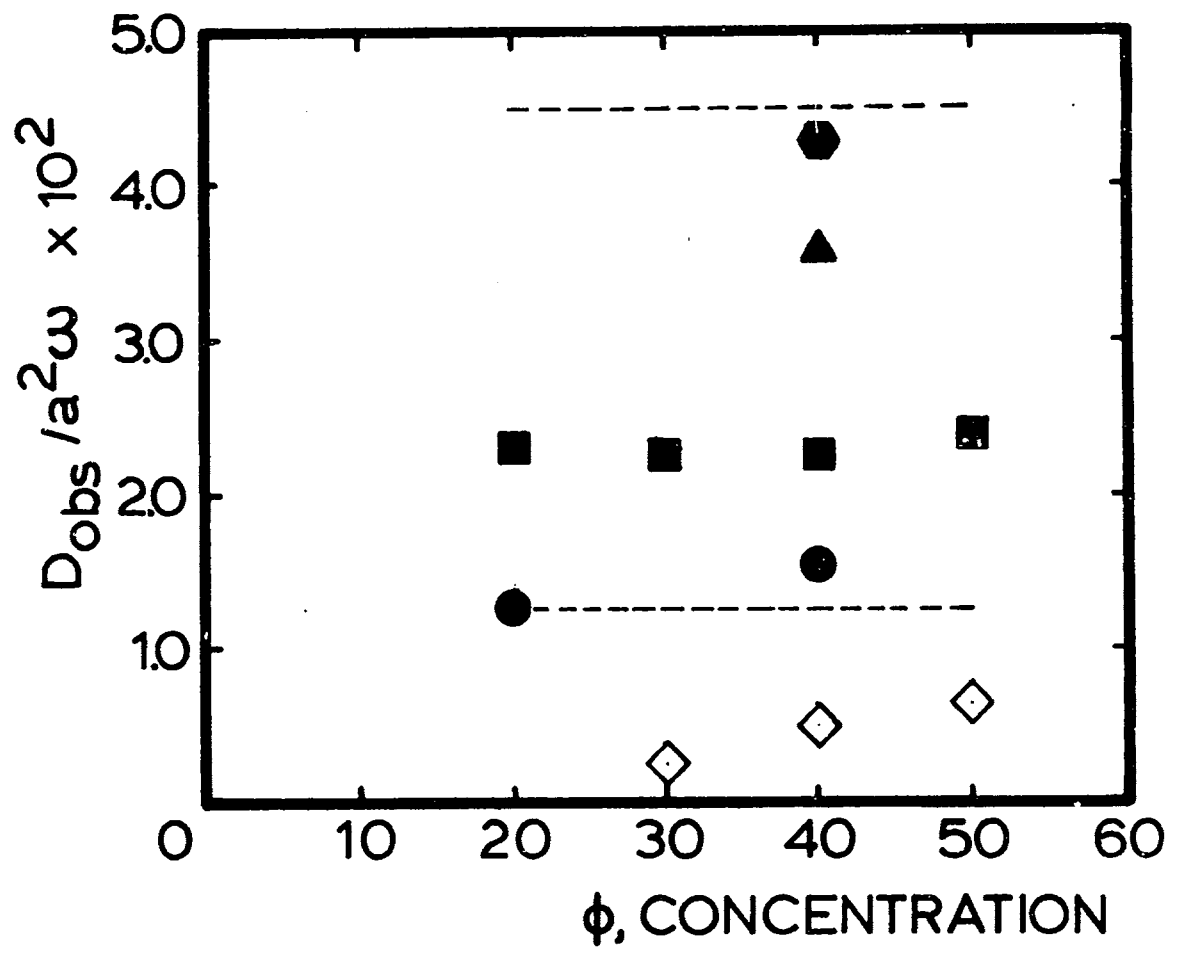


FIGURE 6.9

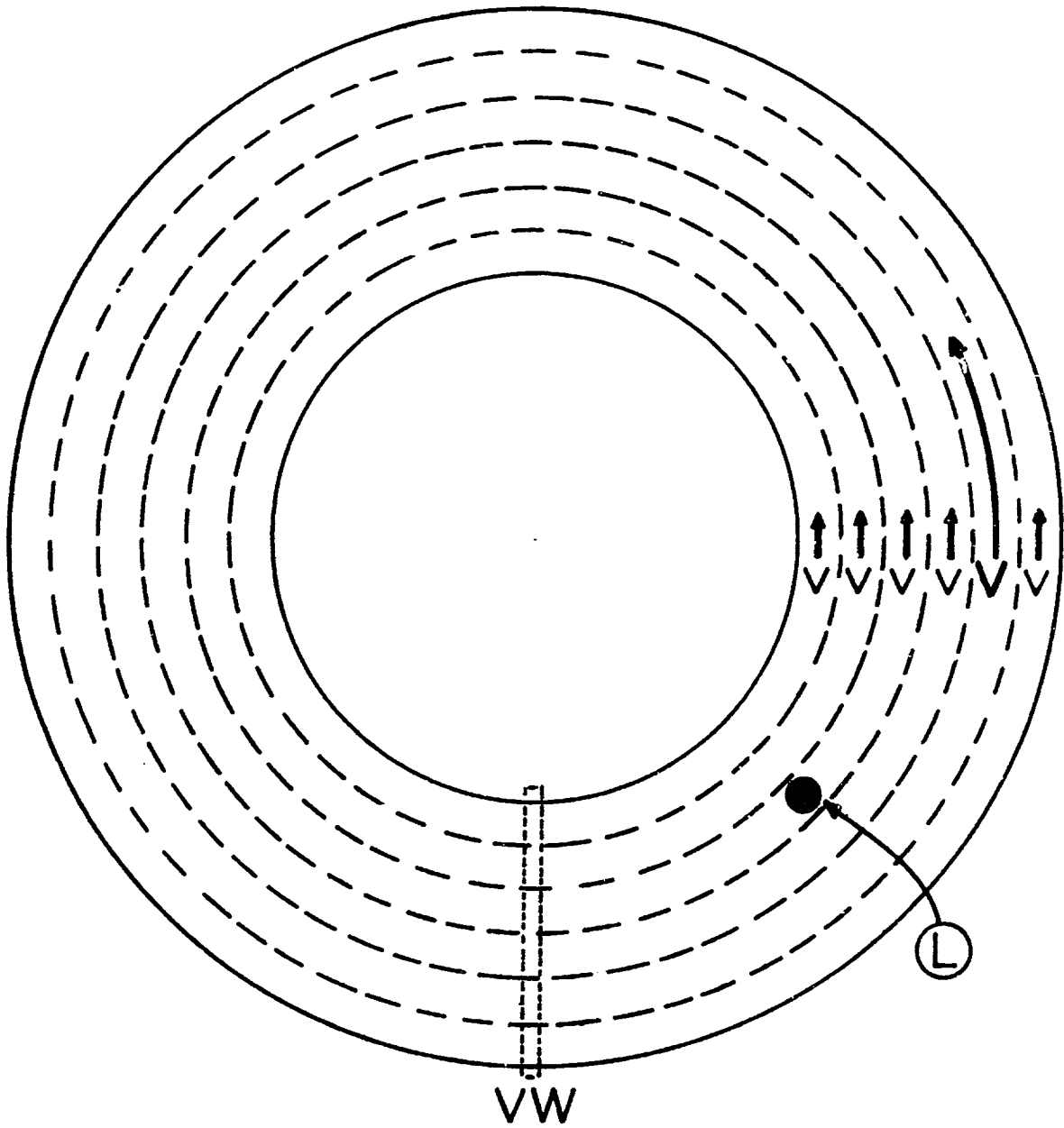


FIGURE 7.1

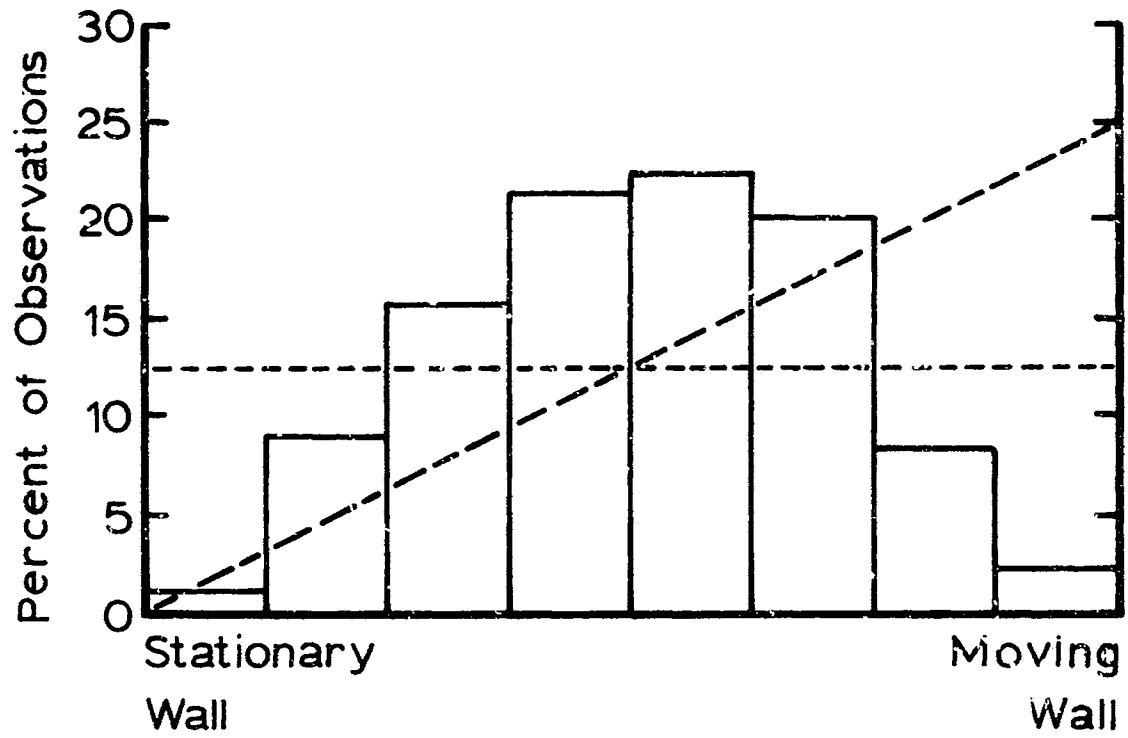


FIGURE 7.2

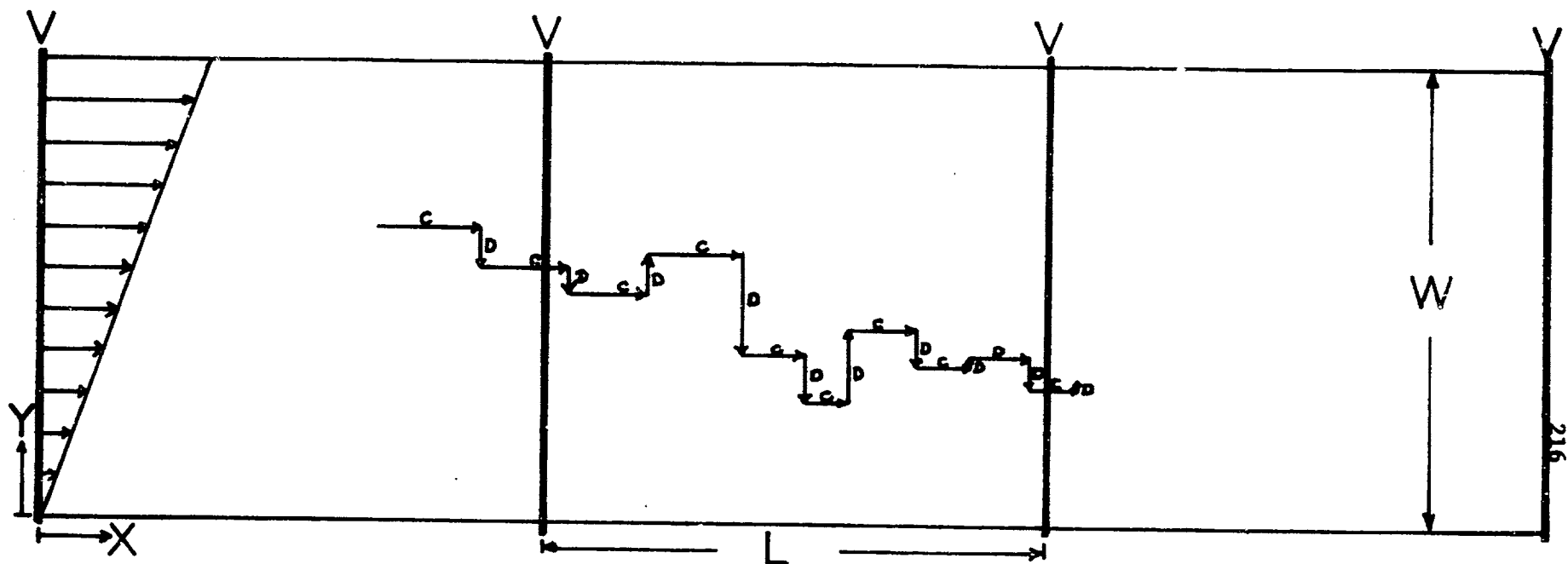
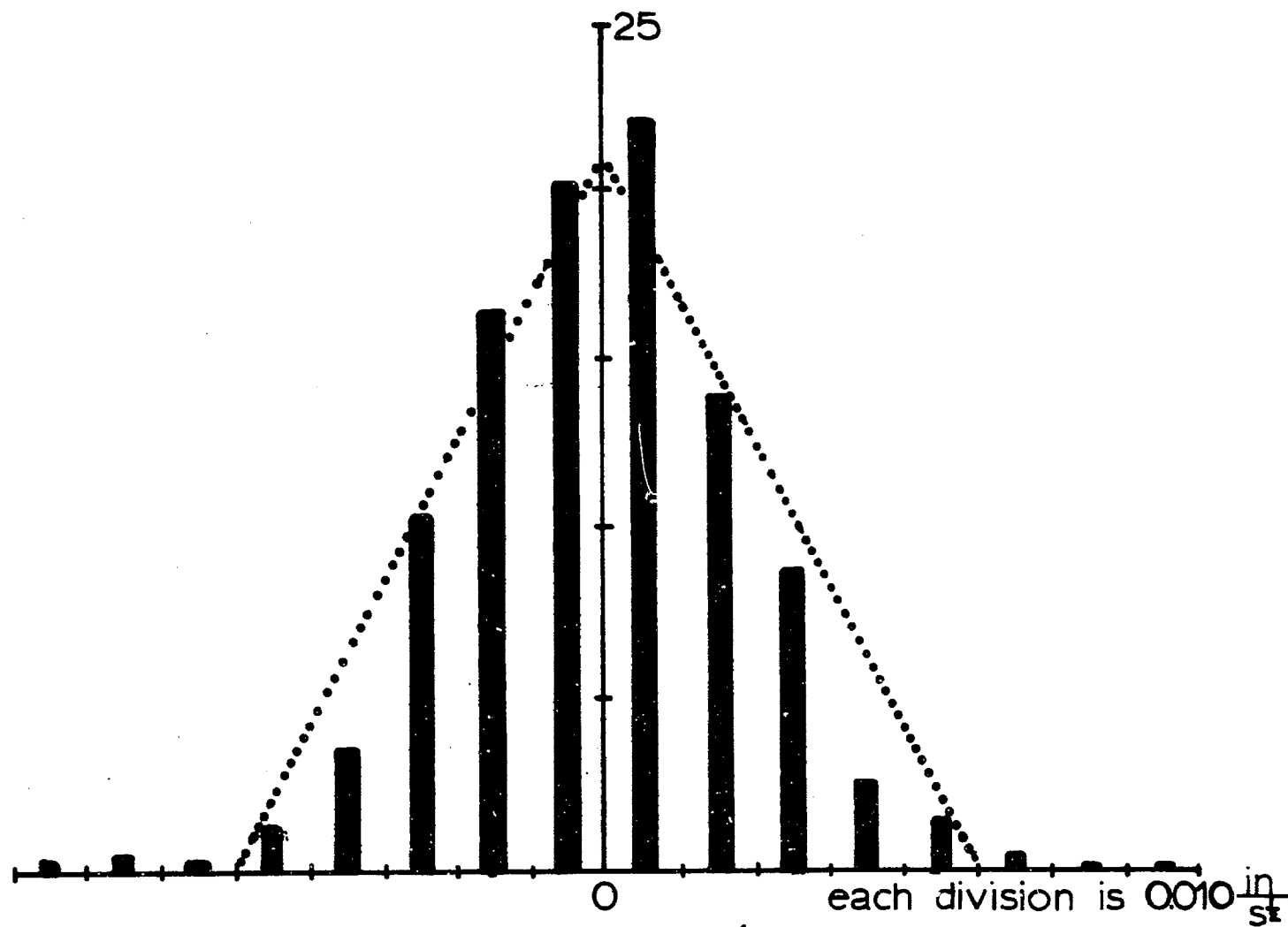
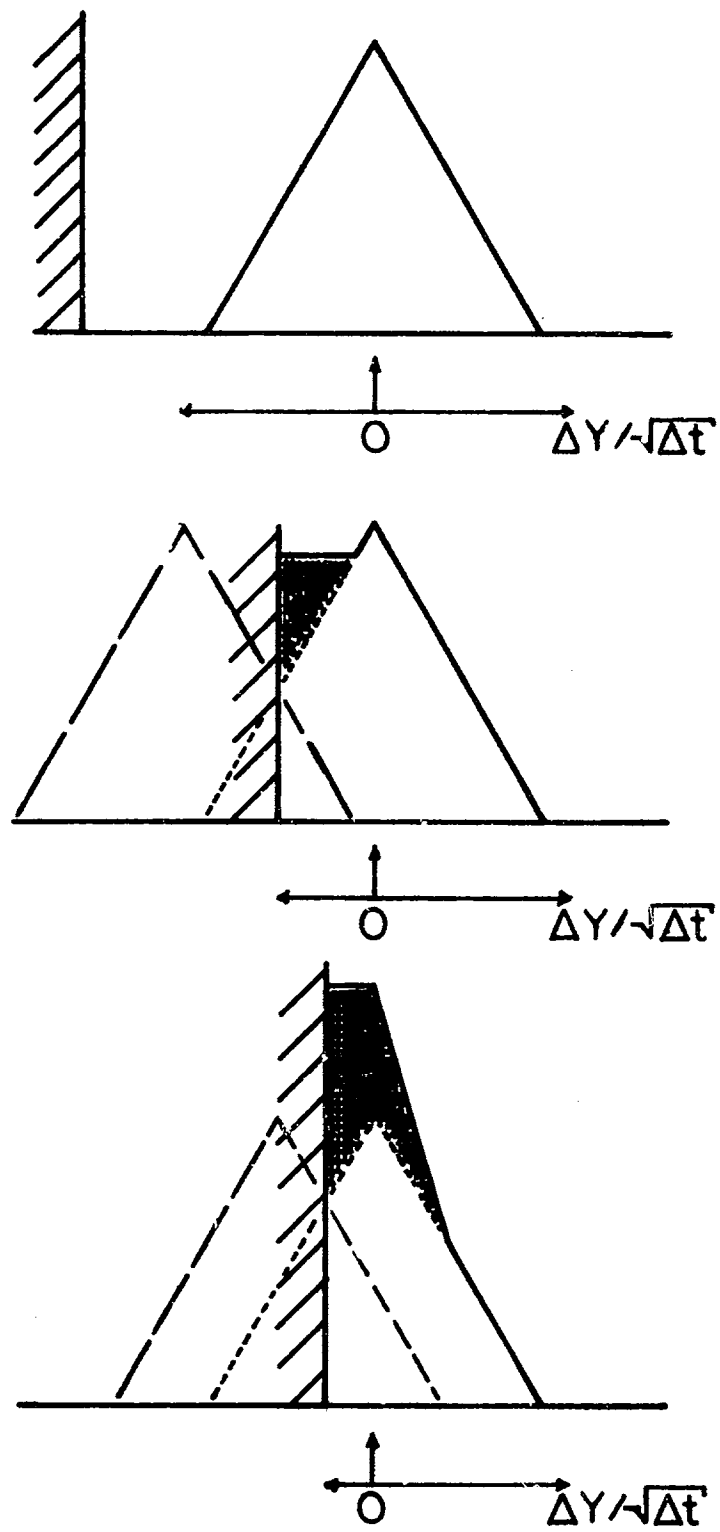


FIGURE 7.3



Distribution of Δx for $\phi = 40\%$, $\omega = 10 \text{ s}^{-1}$, and $a = 0.062 \text{ in.}$

FIGURE 7.4



Distributions of diffusion steps.

FIGURE 7.5

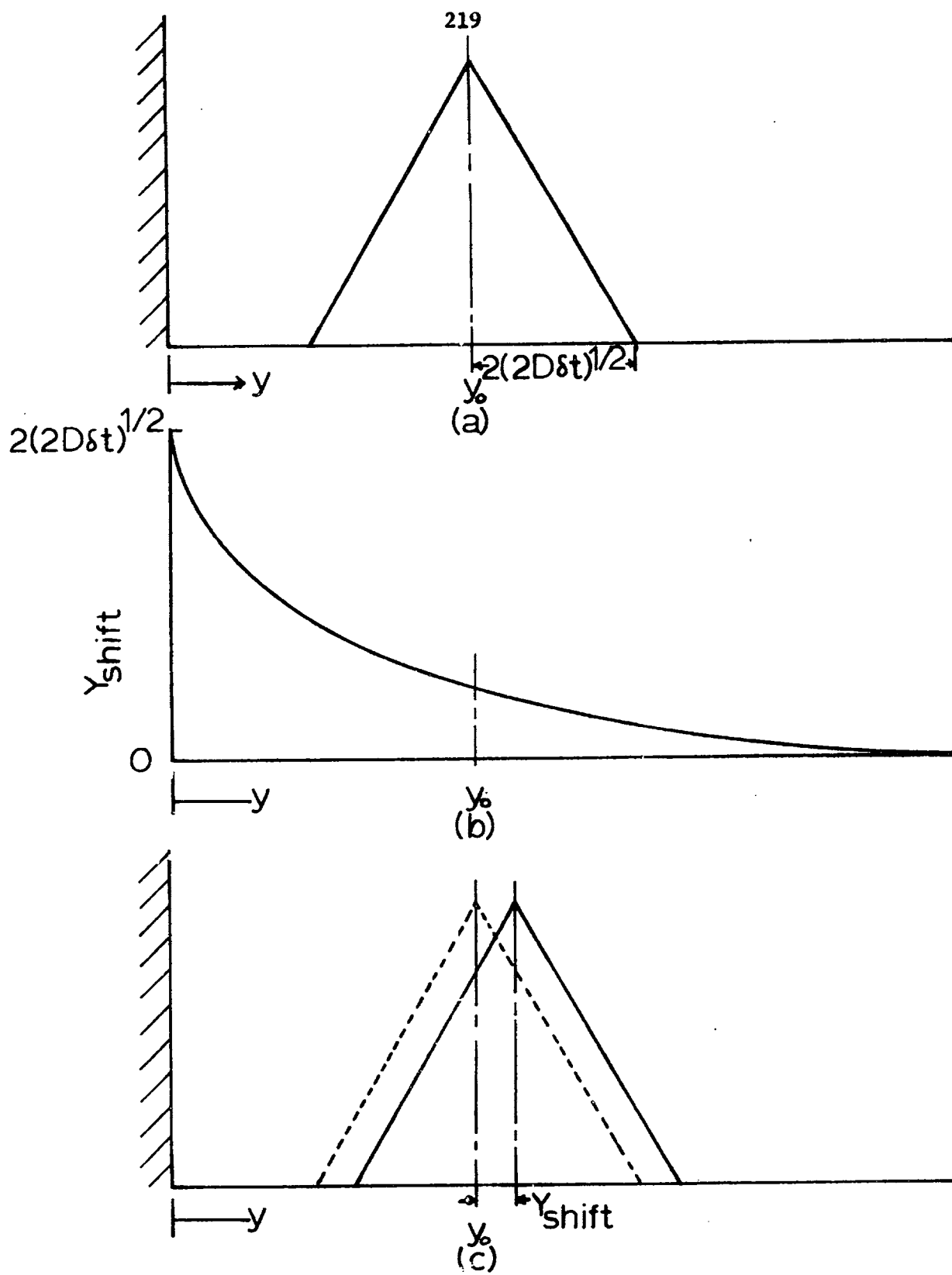


FIGURE 7.6

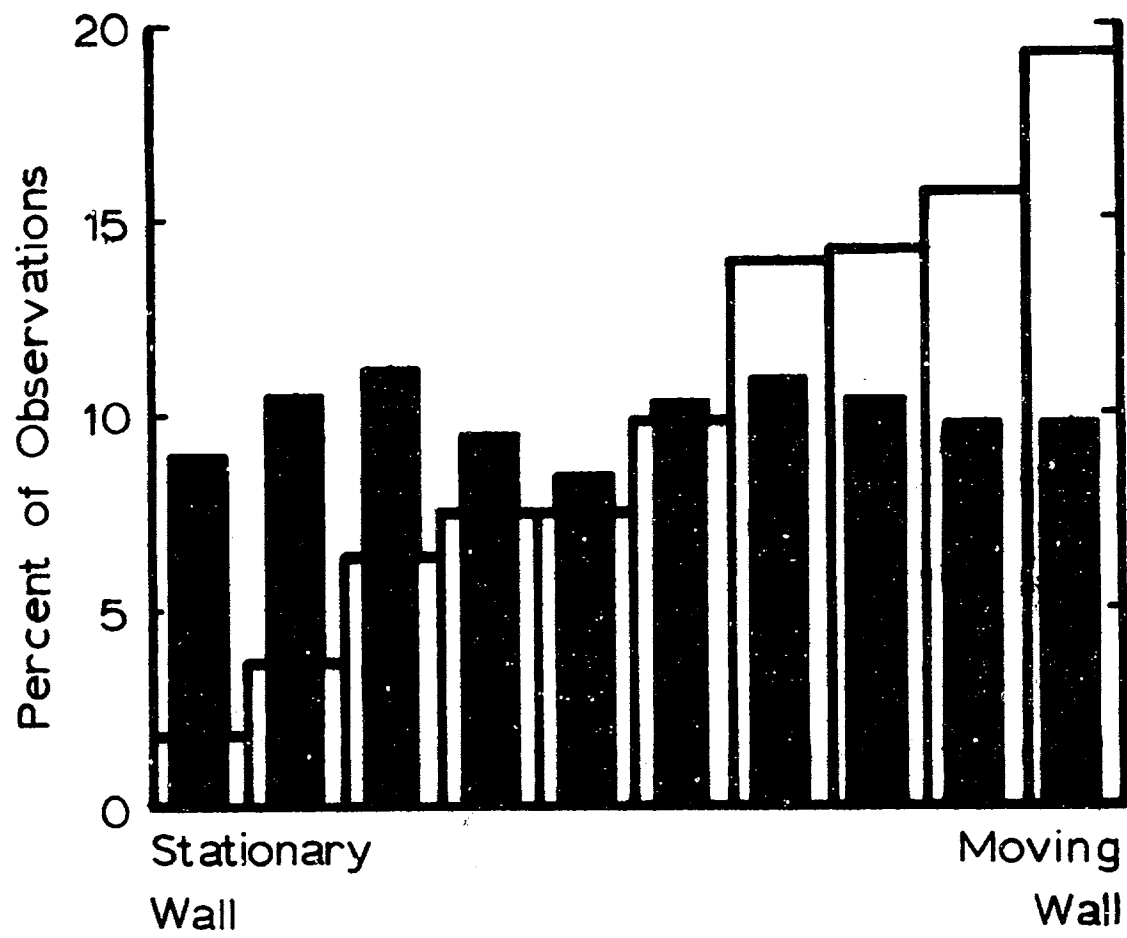


FIGURE 7.7

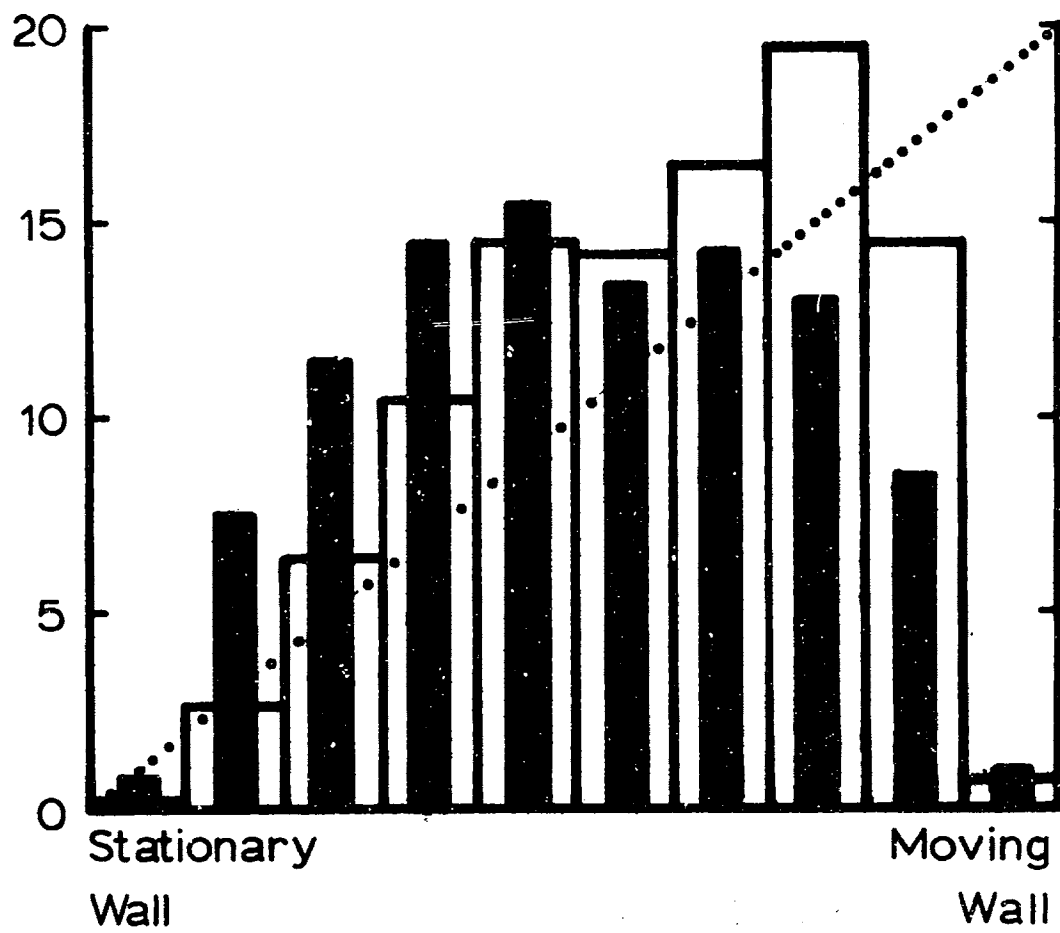


FIGURE 7.8

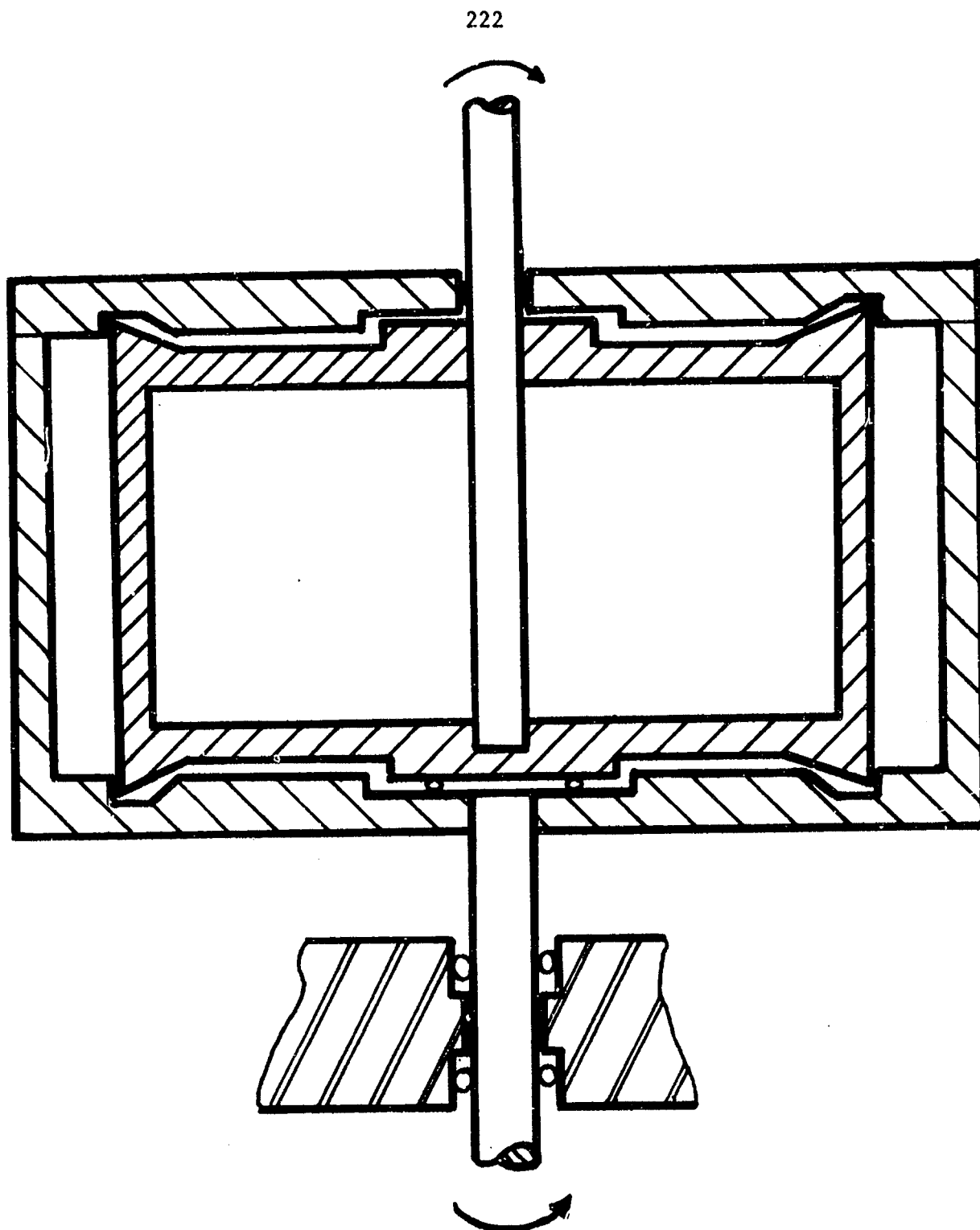


FIGURE A2.1

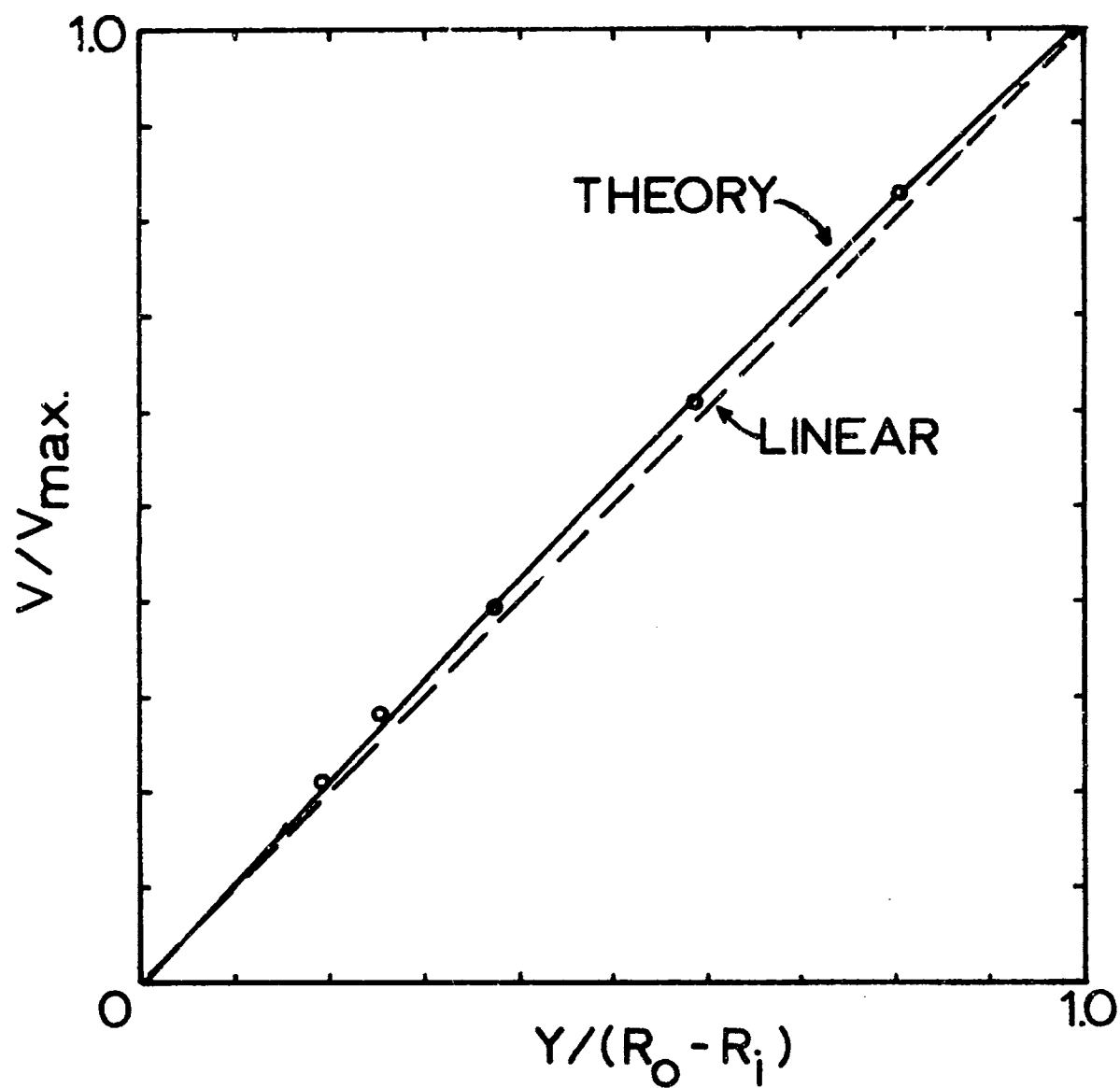


FIGURE A2.2

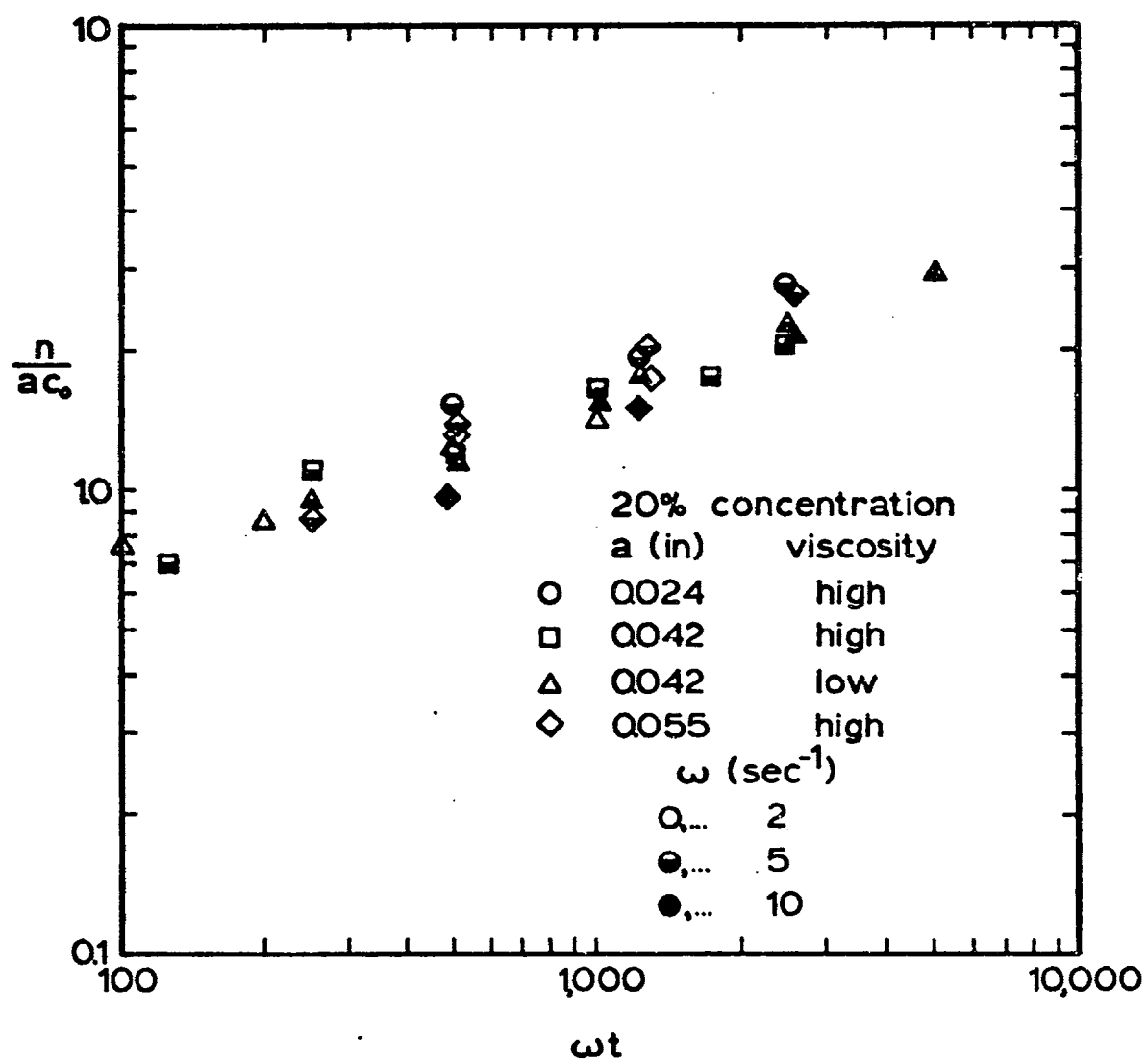


FIGURE A2.3

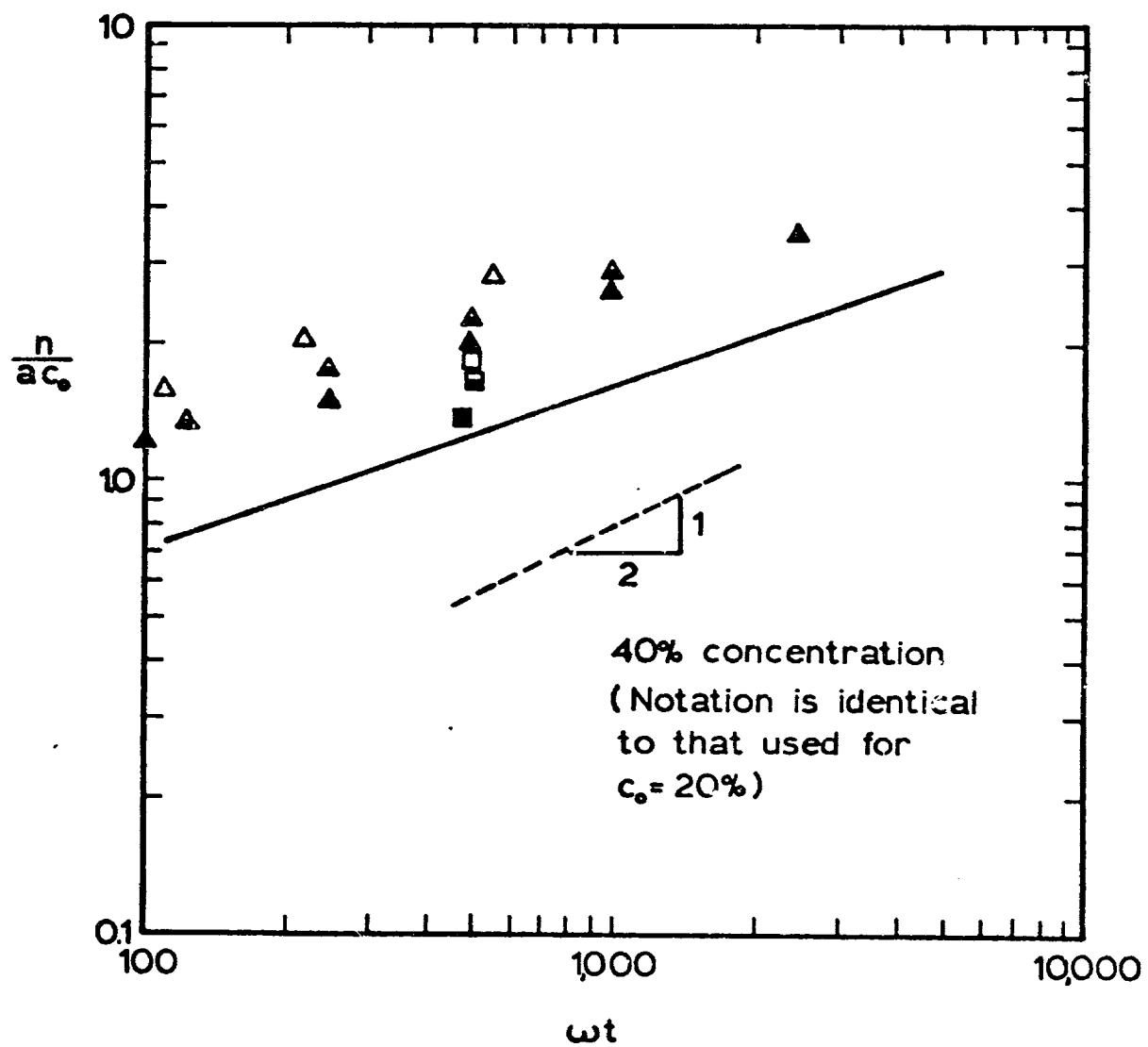


FIGURE A2.4

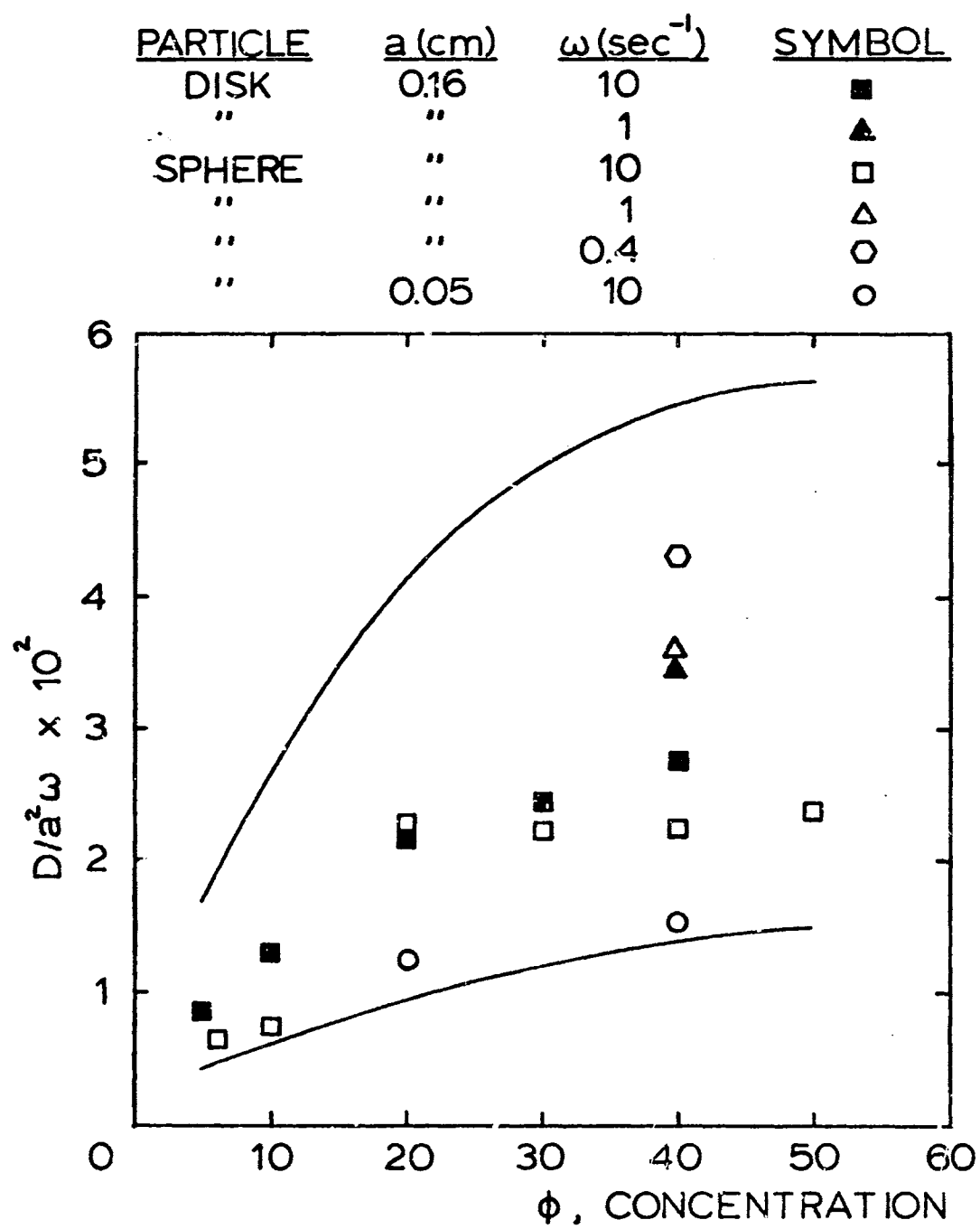


FIGURE A4.1

APPENDIX I. PROCEDURES FOR PLATING ^{58}Co
ONTO COPPER-CLAD ABS PARTICLES

This is a general outline of the procedure used to electroplate carrier-free ^{58}Co onto a copper-clad ABS (acrylonitrile-butadiene-styrene) sphere. Results of this procedure are extremely dependent upon cleanliness and generally good lab technique. The basic procedure separates into two parts -- i) an ion exchange process to remove the iron decay-product from the ^{58}Co and ii) the plating process.

A list of references from which this procedure was drawn is included. A cursory reading of the starred references is recommended before attempting to plate ^{58}Co using this procedure.

Piatinitskii, I.V., Analytical Chemistry of Cobalt, Jerusalem Israel Program for Translations, 1966

Cobalt Monograph, Center D'Information du Cobalt, Brussels (published jointly with Batelle Memorial Institute), 1960

Young, E. S., The Analytical Chemistry of Cobalt, New York, Pergamon Press, 1966

- * Lingane, J.J., Electroanalytic Chemistry, New York, Interscience, 1958
 - * Lingane, J.J. and Page, J.A., "Coulometric Determination of Nickel and Cobalt", *Analytica Chimica Acta*, 13, (1955) pp. 281-287
 - * Hague, J.L., Maczkowska, E.E., Bright, H.A., "Determination of Nickel, Manganese, Cobalt, ...," *Journal of Research of the National Bureau of Standards*, 53, 6 (December 1953) pp. 353-359
- Bate, L.C. and Leddicotte, G.W., The Radiochemistry of Cobalt, U.S. Government Publications NAS-NS3041 Office of Technical Services, Dept. of Commerce, Washington, D.C. 1961

I. PREPARATION AND CONDITIONING OF THE ION-EXCHANGE COLUMN

A. Column should be set up for conditioning several hours prior to its required use. Prepare the column by:

1. Place a glass wool plug at bottom of ion-exchange column.
2. Using demineralized, distilled water, suspend a quantity of Dowex-1 ion exchange beads in a clean beaker.
3. Pour the suspension of beads into the ion-exchange column until a 5-10 cm height of settled beads results.
4. Place a glass wool plug on top of beads.
5. Attach an outflow tube (e.g. a long Pasteur pipette) in a manner which allows the hydrostatic head across the column to be varied.

B. Conditioning of the Ion-Exchange Column.

1. Wash column with demineralized distilled H_2O until the output is clear. Use a very slow flow rate and a setup in which the Dowex beads cannot dry out. Changing the position of the outflow tube will vary the hydrostatic head and therefore, will adjust the flow rate through the column. OVERWASH IF ANYTHING!!!
2. Wash the column with concentrated HCl until output is clear. Follow a procedure similar to 1.

3. Adjust the outflow tube to leave 1 cm of HCl on top of the column. Into this acid, the 5 ml of $^{58}\text{Co-HCl}$ solution, which is prepared as outlined in section II, will be added.

CAUTION: Never allow the column to dry out; always maintain acid or water over all the beads! If the column does dry out, recondition it by starting at B.1.

NOTE: The amount of washing of the column prior to addition of the Co organic contaminants which can wash off with the Co. Certain of these contaminants can inhibit uniform deposition of cobalt onto the cathode. Overwash the column if anything.

II. PREPARATION OF $^{58}\text{Co-HCl}$ SOLUTION FOR ION-EXCHANGE PROCEDURE

- A. Prepare fresh solutions of HCl (conc. 6M, 4M, 0.5M), conc. HNO_3 and 30% H_2O . Clean the necessary glassware well with soap scrubbing and chromic acid bath. Rinse thoroughly (!!) using demineralized, distilled H_2O . Turn on the hot plate.
- B. Place cobalt solution in 25 ml Erlenmeyer flask and reduce the volume of solution on the hot plate to 3-5 ml. Do not boil, only simmer slowly. For all heating steps use an aspirator to collect acid and water vapors and to collect any unintentional spatter. Use of an aspirator speeds the volumetric reductions.

- C. If the ^{58}Co solution contains volatile contaminants, take the solution to a salt and then take up the salt up in 3-5 ml 6M HCl. Always allow the flask to cool before adding solutions to it. If this step is deemed unnecessary because the cobalt solution is relatively pure, proceed directly to step D from B.
- D. Reduce the hot plate temperature. Add 2-3 drops of conc. NH_4OH to the Co solution. Evaporate to a wet salt (near dryness). This will digest any organic contaminants. A greenish-blue wet spot may appear-- a good sign of a fairly pure solution.
- E. Take up in 4 ml 6M HCl. Evaporate to near dryness.
- F. Repeat step E twice. This is to remove any nitrates from the cobalt solution.

NOTE: Do not bake the salts! Cobalt nitrate salts sublime at comparatively low temperatures. $\text{Co}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$ sublimates at 55°C . $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ sublimates at 110°C .

- G. Take up the residue in 2 ml of 6M HCl. Increase volume to 10 ml with demineralized, distilled H_2O .
- H. Add 4-6 drops of 30% H_2O_2 . Digest at low heat (steam bath temperature).
- I. When reaction stops add 4-6 more drops of H_2O_2 . Evaporate to dryness.
- J. Take up the residue in 4 ml of 6M HCl. Evaporate to dryness.

- K. Repeat step J.
- L. Take up the residue in 5 ml of conc. HCl. The solution is now column-ready. All the Fe will be in the Fe^{+++} state because of the H_2O_2 treatment. The ion-exchange column will hold the cobalt and iron at a high molarity of HCl. The nickel will wash through at high molarity. Fe^{+++} will stay on the column until approximately 1 M. However, Fe^{++} will come off with the Co^{++} . Thus, the H_2O_2 treatment is very important for a successful ion-exchange separation.

III. ION-EXCHANGE PROCEDURE

- A. Approximately 1 cm of conc. HCl should have been left above the beads in the ion-exchange column. Into this acid pipette the ^{58}Co . Carefully wash the sides of the 25 ml Erlenmeyer flask using 10 ml of conc. HCl. Pipette this acid into the column using it to police the sides of the column and to wash all ^{58}Co into the beads. (This could be broken into a couple of repeat steps to insure that all ^{58}Co is extracted from the flask and washed into the column.)
- B. Using 15-30 ml of conc. HCl wash the Co onto the column and wash any Ni through the column. Collect the elutate in a 250 ml Erlenmeyer flask. Adjust the output pipette to achieve a slow drip as the output of the column.

- C. Elute the Co fraction using 4M HCl. Up to 60 ml of 4M HCl may be necessary. Use a survey meter to determine when the ^{58}Co appears at the output. When it does, switch to a clean 250 ml Erlenmeyer flask. Use of this procedure should avoid any Ni which might first come out with the conc. HCl. Continue surveying both the output and the ion exchange column. Cease eluting with 4M HCl, when all the Co has been collected in the Erlenmeyer flask.
- D. Add 1-2 ml of conc. HNO_3 to the cobalt solution eluted from the column. Slowly reduce the volume of the ^{58}Co solution to ~ 5 ml on a low heat. This step will aid in digesting any organic contaminants eluted from the ion-exchange beads of the column.
- E. Transfer the Co solution to a 25 ml Erlenmeyer flask. Wash the 250 ml flask with less than 15 ml of 6M HCl and add this to the 25 ml flask. Slowly evaporate to a dry salt.
- F. Take up the salt with 1-2 ml of 6M HCl and dilute to 4-5 ml with demineralized, distilled H_2O . Add 4-6 drops of H_2O_2 and place on low heat. Evaporate to dryness. Repeat the uptake in 6M HCl only and reduce the solution to dryness 3 times to remove any nitrates. Perform the last reduction to dryness in a 5 ml beaker. This beaker will be used as a plating vessel.

IV. PREPARATION OF COPPER-CLAD ABS PARTICLE FOR PLATING

- A. Impale the ABS particle* on a glass-coated tungsten needle (obtained from a glass blower). The tip of the needle should be sharp and no unnecessary tungsten should be exposed. Square shoulders on the glass coat seem to prevent cobalt plate from growing out from a copper surface of the particle over the glass. The electrical connections to the particle are made through the tungsten needle. A short length of nickel wire can be spot welded to the tungsten needle; the nickel wire will provide a material to which solder will adhere.
- B. Clean the particle in 15% H_2SO_4 . Rinse the bead in demineralized, distilled H_2O and do not allow it to dry since no trace of an oxide layer may be left on the bead. The copper should have a reddish color.

NOTE!!!: The cleanliness of the copper surface is crucial. The amount of Co to be laid down is only several atomic mono-layers if ideally distributed, therefore the condition of the surface is probably the single most important factor in achieving a quality plate. Do not let the fresh copper surface dry--that will re-establish the oxide layer.

* Mr. David Maluchnik of the Shipley Co., Newton, Ma. graciously plated many ABS particles for us.

- C. A strike coat of copper was applied to the electrodes deposit of copper on the ABS particle. Although this step increases the weight of the particle it provides a good check on the electrical connection between the electroless copper shell and the tungsten needle. Furthermore, much of this copper can be carefully etched away in the acid bath.

V. PLATING PROCEDURE

- A. At the time of the last volumetric reduction, make plating solution of 1M pyridine, 0.2M hydrazine. Adjust the pH to 7 to 8 using HCl. Remove scavenger ions using two platinum electrodes with a voltage between them of approximately 1.6 V. Run to a stable background current.
- B. Take up cobalt salt in 5 ml beaker in a minimal amount of 0.5M HCl (2-3 drops). Add 2-3 ml of plating solution. Neutralize solution using hydrazine or HCl solutions. A primary aim is to keep the volume of plating solution as small as possible in order to maintain the maximum concentration of Co.
- C. Add the platinum anode. With a 1-2 V potential between the platinum anode and the copper particle (cathode), place the copper particle into the plating solution. Place a chloride electrode in close proximity to the copper head. The potential of the copper

particle relative to the solution is measured on an electrometer using a chloride electrode.

- D. Plate out the cobalt at between 4-6 volts between the chloride electrode and the bead. For small particles a plating current of several milliamperes will be observed. Most of this current is background current from ancillary reactions in the plating solution. This voltage should be monitored with an electrometer. Use a flea and magnetic stirrer to keep the plating solution well mixed. Maintain the pH as close to 7 as possible. At lower pH's, the cobalt is complexed to pyridine rather than hydrazine. Cobalt-pyridine complexes will plate out more evenly and at lower voltages than cobalt-hydrazine complexes. A decrease in rate of plating is observed for non-neutral pH's. The hydrazine prevents competing reactions at the anode by depolarizing it.
- E. Maintain the potential between the particle and the anode when the particle is removed from the plating solution. Immediately rinse the particle with demineralized, distilled water. Plating of 15 mCi of ^{58}Co from a 30 mCi bath may take from 12 to 24 hours. Larger cathode areas would presumably require less time to plate out half of the ^{58}Co .

APPENDIX II. TWO-DIMENSIONAL PARTICLEMIGRATION IN A LINEAR SHEAR FLOW

Prior to the experiments described in the main body of the thesis a series of experiments were performed to study the migration within a single layer of particles in a linear shear flow. The major purpose of these experiments was to gain an understanding and a sense of scale for the diffusional phenomena. A circular cylinder Couette device was used to establish the linear shear flow. The dimensions of the device are illustrated in Figure A2.1. As in the single-labelled-particle experiments a two-fluid system was used to reduce the secondary flows. Water was used as the base fluid. During the course of the experiments two silicon oils with viscosities of 35 and 300 poise were used for the upper, viscous fluid. Figure A2.2 shows a velocity profile obtained from measurements of the speed of a single small particle. Great care was exercised to select a neutrally-buoyant particle.

The experiments on particle migration used particles which floated on the upper surface of the silicon oil thereby producing a single layer of particles in which the diffusive motion could be studied. Initially, spherical particles of low-density polyethylene were chosen for the experiments. These particles were approximately 8% lighter than the silicon oil. When the shear flow was started these particles would sink below the surface of the silicon oil.

Observations of a single particle showed that it would sink vertically to a stable position about two diameters below the surface. Observations of a large number of particles, which in static circumstances would form a single layer, showed that they would sink and form a cloud of particles several diameters thick. Polyethylene particles were abandoned because of these phenomena.

To prevent the sinking effects, very low-density styrofoam beads were used. (These particles are expanded polystyrene of the type which is molded into disposable hot-drink cups or packaging.) These particles did form a single layer in the shear flow; however, during some particle interactions a particle would be seen to reduce its interaction by sinking into the silicon oil.

The particles were sieved into three size groups with mean sizes of 0.024 inch, 0.042 inch, and 0.055 inch; the maximum width of the particle-size distributions was 18%. The particles were lightly painted in various colors to allow identification of the particles of the same size range.

In these experiments an initial step distribution in the color of the particles of one size was established and its change with time was measured. By comparison of experimentally observed quantities with quantities from the mathematical solution of an analogous diffusion problem, the diffusion coefficient for the process can be established.

Rather than measure at some time the concentration of various distances from the initial step, the number of particles which

crossed from one side of the step to the other was measured. Mathematically, the number is an integral over time of the flux at the step. Physically, it is a very easy quantity to measure accurately. However, several types of flux could produce the same numerical value of the integral. Therefore, to check that the phenomena is truly diffusive, the number which cross the step must be measured for different time intervals after the diffusive process is started. This is mathematically equivalent to integrating the flux for different lengths of time. As is shown in the section dealing with analysis of the experiments, the number of particles that cross the step varies as the square root of the time for a diffusive process.

This type of experiment is easier than the major experiments of the thesis because it requires a smaller amount of data collection and reduction. However, the experimental technique is possible only for a single layer of particles on a surface since well defined initial distributions must be established.

The initial distribution of particles of two different colors was established using a steel band which separated the Couette channel into two equal widths. The band rotated at the speed of the fluid underneath it and could be raised out of or lowered into the silicon oil while rotating. At the beginning of an experiment the lower edge of the band was submerged approximately one particle diameter into the silicon oil. Equal number densities of particles were placed on the upper surface of both halves of the Couette channel; different colors of particle were used on the two halves.

The Couette apparatus was started and the shear flow established; time was allowed for the particles on each side to disperse in the shear flow. The steel band was lifted quickly and smoothly without introducing a disturbance. At the time the steel band left the oil, a stopwatch was started. At a later time the steel band was lowered into the silicon oil and the stopwatch stopped as the band entered the oil. The recorded time on the stopwatch was used as the time over which diffusion occurred. The numbers of particles that had crossed from each half to the other were counted and recorded. The particles were separated by color before beginning another run.

Analysis of Results

Dimensional analysis shows that the concentration distribution of color would involve a solution of the form

$$\frac{c}{c_0} = f\left(\frac{y}{a}, \omega t, \phi, \frac{y}{R}, \frac{\omega^2 a}{\nu}\right) \quad (\text{A2.1})$$

where c is the concentration and c_0 the initial concentration. The coordinate y is measured from the steel band, a is the particle radius, ω the shear rate, ϕ the area fraction of spheres, R the radius of the steel band and ν the fluid viscosity. The last group is the particle Reynolds number and can be neglected for the conditions of slow viscous flow employed in these experiments. The term y/R is a measure of the effect of curvature. For short

times curvature can be neglected since the problem can be approximated by diffusion from an infinite plane step.

Rather than measure the concentration distribution itself it is easier to measure the total number of particles having crossed per unit length of circumference. This is given by the following integral

$$n = \int_0^{y_L} c(y,t) dy \quad (A2.2)$$

where y_L is taken large enough for c to approach zero. In terms of the non-dimensionalization given above this becomes

$$\frac{n}{ac_0} = F(\omega t, \phi) . \quad (A2.3)$$

A solution to the diffusion equation for the cylindrical step function valid for small Dt/R^2 is given by Lykov*

$$\frac{c(r,t)}{c_0} = \left(\frac{R}{r}\right)^{1/2} \operatorname{erfc} \frac{1 - \frac{r}{R}}{2\sqrt{Dt}/R} + \dots, \quad \frac{r}{R} > 1 . \quad (A2.4)$$

Letting $y = r - R$ and retaining terms to zero order in y/R this reduces to

* Lykov, A.V., Analytical Heat Diffusion Theory, Ed. by J.P. Hartnett, New York, Academic Press, 1968.

$$\frac{c(x,t)}{c_0} = \operatorname{erfc} \frac{y}{2\sqrt{Dt}} . \quad (\text{A2.5})$$

This is identical to the solution for the infinite plane step.

Performing the integration of Equation A2.2 to determine the total quantity for $x > 0$ the result is the following

$$n = \frac{1}{\sqrt{\pi}} c_0 (Dt)^{1/2} . \quad (\text{A2.6})$$

Comparing (A2.3) and (A2.6) it is seen that if the process under study is truly diffusional $F(\omega t, \phi)$ must take the form

$$F(\omega t, \phi) = \left(\frac{D}{\pi a^2 \omega} \right)^{1/2} (\omega t)^{1/2} . \quad (\text{A2.7})$$

Experimental Results

The quantity n/aC_0 as a function of dimensionless time ωt is shown in Figure A2.3 for an area fraction of particles of 20%. Data is given for three particle sizes, three shear rates and two values of viscosity.

From the figure it can be seen that the correlation given in Eq. A2.3 is confirmed. Furthermore, it seems that the dependence on ωt follows a power law with constant exponent over the range investigated (i.e. a straight line on a log-log plot).

Figure A2.4 shows a similar plot for an area fraction of 40%. The straight line is a best fit by eye to the 20% data of Figure A2.3. It can be seen that a straight line parallel to the 20% line is a reasonable fit to the low viscosity data at $\phi = .40$. However, within the low viscosity data there is a systematic variation with the shear rate, the flux decreasing as the shear rate increases. Also a set of high viscosity data at $\omega t \sim 500$ falls well below the low viscosity results and shows the same systematic variation with shear rate. The lack of correlation here may be due to deviations in the vertical distribution of the particles. This deviation is a result of particles being pushed below the surface by the unknown forces discussed earlier as well as by the collision mechanism under consideration here. Shear rate and viscosity would control the relative recovery time for the disturbed layer. In order to examine this question the following dimensionless group which compares viscous forces to gravitational forces was formulated

$$N_1 = \frac{\mu \omega}{a \Delta \rho g} \quad . \quad (A2.8)$$

Here $\Delta \rho$ is the difference between the density of the particles and that of the fluid. The density of the particles was approximately 0.02 gm/cc so that $\Delta \rho$ is very nearly the density of the fluid alone. In this case N_1 can be rewritten

$$N_1 = \frac{\nu \omega}{ag} \quad (A2.9)$$

where ν is the kinematic viscosity. For increasing values of N_1 the vertical dispersion would be expected to increase. In the experiments at $\phi = .40$ (Fig. 4) ν and ω are the only quantities varied. For the set of points near $\omega t = 500$ the respective values of $\nu\omega$, scaled to the lowest value, are given in the following table:

<u>Symbol</u>	<u>$\nu\omega/(\nu\omega)_0$</u>
Δ	1.0
$\blacktriangle$	2.4
$\blacktriangle$	4.5
$\square$	8.0
$\blacksquare$	21
$\blacksquare$	39

It can be seen that the ordering of the points is consistent with this parameter. It is apparent that minimizing the shear rate and the viscosity would lower this parameter and presumably a true single layer configuration could be approached. Unfortunately, both of these possibilities were limited. The shear rate was limited by the mechanical system employed. Reducing viscosity introduced the additional undesirable parameter of surface tension which causes the particles to form clumps and to approach the walls. The dimensionless group by which surface tension effects could be considered is the following

$$N_2 = \frac{\mu\omega a}{\sigma} \quad (A2.10)$$

where σ is the surface tension. In order to reduce the effect of surface tension $\mu\omega$ should be increased which is the opposite requirement from that given above. Reduction of the surface tension with surfactants would achieve little since the air-silicone oil surface tension is already quite low (≈ 20 dyne/cm). The opposing requirements indicated above limited the possibility of approaching the single layer condition hypothesized.

Discussion

The dotted line in Figure A.4 has a slope of one half. The slope of the experimental data is clearly seen to be less than this. For a purely diffusive motion to be governing the particle motion the slope of the experimental results must be one half as shown in Equation A2.7. From the correlations shown with non-dimensional quantities N_1 , and N_2 , it is clear that not only shear-driven forces act on the particles.

A value of the non-dimensional diffusion coefficient calculated for the 40% concentration experiments at a time interval, $\omega t = 1000$ is 2×10^{-2} . This is nearly equal to of the value obtained in the experiments using a single labelled particle. Considering the unknown relationship between two and three dimensional experiments and the extraneous effects, this agreement is quite amazing.

APPENDIX III. CONSIDERATION OF ERROR

A.3.1 Systematic Errors

The simplest errors to assign size and direction to are the systematic errors. In these experiments there are three origins of systematic error which are apparent concentration, apparent shear rate and particle size. The text below contains estimates of the first two types of systematic error. The systematic error relating to particle size is treated in Chapter 6.

Apparent Concentration. The concentration used in this thesis is the apparent concentration, i.e., the volume of particles contained in the Couette apparatus divided by the volume of slurry in the Couette apparatus. The error in measuring the apparent concentration is negligibly small. (Measurements of the apparent concentration were made using the heights of particle-free suspending fluid in the Couette apparatus and the slurry in the Couette apparatus as explained in Chapter 5. Since the Couette channel is a well-controlled dimension, these heights are directly proportional to the respective volumes.) The systematic error in concentration occurs because of the Vand effect as reported by Karnis, et al. whose work was discussed in Chapter 2. Their work on Couette flows showed that the concentration is uniform across the Couette channel except for a depleted region near the walls from which particles

are physically excluded by the wall.

An estimate of the maximum increase in the actual concentration over the apparent concentration can be obtained by modelling all the particles in the Couette apparatus to be within a volume reduced from the original slurry volume by a space near each Couette wall one particle radius wide. For the large particles this would cause an increase in the central concentration of $\sim 14\%$; in the case of the small particles the increase would be less than 5%.

Apparent Shear Rate. As the experiments of Karnis, et al. [17] show, the Vand effect leads to a non-linear velocity profile across the Couette gap. From their data, shown in Figure 2.5 (b,c) it appears that this non-linearity is restricted to a zone near each wall with a width of approximately one particular diameter. The data of Karnis have values of the ratio of particle radius to channel width, a/w , which bracket the value of a/w for the large-particle experiments. Furthermore, the data of Karnis is for slurry flows with a high volumetric concentration of 38%. Interpolation between the data of Karnis provides an estimate of 21% for the decrease in the shear rate of the central channel region from the apparent shear rate for the large-particle experiments. Extrapolation from the data of Karnis provides an estimate of an 8% decrease of the shear rate of the central channel region for the small-particle experiments. These estimates of the shear rate are reasonable for the experimental conditions of forty percent

concentration. No comparable data could be found for other concentrations. Accordingly, these estimates have been applied to all concentrations as the systematic error caused by use of the apparent shear rate.

A.3.2 Random Errors

Relationship of the Sample to the Infinite Distributions.

From a mathematical viewpoint, the experiments are merely an empirical method for defining the probability function which describes the diffusive motion of the labelled particle. Since all the particles are equivalent, except for the label, the diffusive motion of the labelled particle therefore is common to all the particles. In the probabilistic sense, observations of the position of the labelled particle Y_i , taken at time t_i , are formed into scaled changes of position

$$\chi_i = (Y_i - Y_{i-n}) / \sqrt{t_i - t_{i-n}} \quad .$$

If the diffusive motion is not restricted or interfered with there is no need to use subsequent observations; hence n can be any positive integer 1 or greater. The relative frequency of occurrence of a class of χ_i forms the basis for defining the probability distribution function. The diffusion coefficient, D , is equal to one-half of the second moment of this probability distribution, that is

$$D = \frac{1}{2} E(\chi^2) \quad (\text{A3.1})$$

Here E denotes a general expectation, i.e.

$$E(a^b) = \frac{1}{N} \sum_{i=1}^{i=N} a_i^b p(a_i)$$

and a_i is a member of a set of N items which has the observed probability $p(a_i)$.

There are two basic questions concerning the amount of error in D which have statistical answers and therefore use the above statistical picture of diffusion. The first question is how well does the sample, that is the group of values of χ_i measured during an experiment, reflect the underlying infinite distribution of all possible diffusive motions from which the sample was presumably drawn. This is treated directly in most texts on statistics. One characteristic of the sample is its variance, $m_2 = E(a^2)$. Cramer [34] provides the following formulae which relate the expected value of the variance of a sample of size N , $E(m_2)$, to the well-defined variance of the underlying infinite distribution, $\mu_2 = \int_{-\infty}^{+\infty} \alpha^2 p(\alpha) d\alpha$ where $p(\alpha)$ is the probability-distribution function which describes the infinite distribution of α ; they are

$$E(m_2) = \frac{N-1}{N} \mu_2 \quad (\text{A3.2})$$

or for the case of the diffusion coefficient, which equals half the second moment

$$E(D) = \frac{N-1}{N} \hat{D} \quad (\text{A3.2a})$$

(The circumflex, $\hat{}$, will be used to denote moments of the underlying infinite distribution from which the observations used to define the uncircumflexed moment are drawn.) The relationship between the variance of m_2 and the second and fourth central moments, μ_2 and μ_4 , of the underlying distribution is:

$$V(m_2) = \frac{\mu_4 - \mu_2^2}{N} - \frac{2(\mu_4 - 2\mu_2^2)}{N^2} + \frac{\mu_4 - 3\mu_2^2}{N^3} \quad (\text{A3.3})$$

where $V(m_2) = E(m_2^2)$. Or, using Equation A3.1,

$$4 V(D) = \frac{\hat{E}(X^4) - 4\hat{D}^2}{N} - \frac{2(\hat{E}(X^4) - 8\hat{D}^2)}{N^2} + \frac{\hat{E}(X^4) - 12\hat{D}^2}{N^3} \quad (\text{A3.3a})$$

If χ has a normal distribution, then $\hat{E}(X^4) = 3\hat{E}^2(X^2) = 12\hat{D}^2$; in this case the third term on the right hand side of Equation A3.3 is zero and the first two terms have a much simpler form,

$$V(D) = \frac{2(N-1)}{N^2} \hat{D}^2 \quad (\text{A3.4})$$

Equations A3.2 and A3.4 illustrate two common features of statistics. The mean value of a sample moment, for large samples, is, according to Equation A3.2, just slightly less than the corresponding

moment of the infinite population. The second feature is that the standard deviation of the sample moment, which is the square root of $V(D)$ as given by Eq. A3,4 decreases as the inverse square root of N . The number of scaled changes of position χ_1 in the experimental samples used to calculate D ranged from 150 to 640. A range of possible observed values of D which would be produced by 99% of all possible samples is given by the mean value, $\hat{D}$, plus or minus two standard deviations, which for these sample sizes would represent 14 to 27% of D . Note, however, that this only deals with the fit of the sample to the underlying distribution. It does not verify that the underlying distribution is composed wholly of diffusive motions.

A property of the probability distribution functions which describe diffusive motions is that they are normal (or Gaussian). The chi-squared-test for goodness of fit was positive at a 0.10 level of significance for the probability distributions for which D was calculated.

One must bear in mind that so far the discussion of random errors has focused upon the relationship of the sample to the underlying infinite distribution. This is not equivalent to the accuracy of the experiments since the underlying distribution includes the effects of error and the lateral passing motion of the labelled particle.

Effect of Sighting-Error and Passing-Motion Components. To

further examine the nature of the distribution of χ let us return to its definition and examine the manner in which observations are taken. As before,

$$\chi_i = (Y_{\text{obs}_i} - Y_{\text{obs}_{i-1}}) / \sqrt{t_{\text{obs}_i} - t_{\text{obs}_{i-1}}} \quad (\text{A3.5})$$

in which the Y_{obs_i} 's are observations of the lateral position of the labelled particle at time t_i as reported by the detector. The detector measurements do not necessarily record the true instantaneous position of the labelled particle, therefore, there is some distribution of errors among the observed Y 's. The time will be modelled as being an exact measurement since the electronic interval stopwatch is recorded by the same signal which records the position. Thus each Y_{obs} can be expressed as a sum of the true position, Y_{true} , and an error Y_{sighting} . Equation A3.5 then becomes

$$\begin{aligned} \chi_i &= \frac{(Y_{\text{true}} + Y_{\text{sighting}})_i - (Y_{\text{true}} + Y_{\text{sighting}})_{i-1}}{\sqrt{t_i - t_{i+1}}} \\ &= \frac{\Delta Y_{\text{true}_i} + \Delta Y_{\text{sighting}_i}}{\sqrt{\Delta t_i}} \end{aligned} \quad (\text{A3.6})$$

(The delta subscripted with an i , Δ_i , will be used subsequently to indicate a difference between the values of a quantity at observations $i+1$ and i .)

However, the true instantaneous lateral position of the labelled particle is not the correct variable for the diffusive motion as can be seen if one considers the motion of the labelled particle in the slurry. The labelled particle diffuses because it has many interactions with its neighbors. These interactions allow the faster particles to pass ahead of close, slower particles. During a passing interaction, both principal particles are laterally displaced; diffusion of a particle results from many incomplete recoveries of the particle from the lateral displacements which occur during the passing interactions. The random positions of the neighbors cause random amounts of incomplete recovery. This, in turn, causes the net changes in lateral position to have a diffusive nature. Thus, in the motion of the labelled particle there is a large amplitude lateral passing-motion about a momentary average lateral position, which diffusively changes over time due to the random incompleteness of the passing interactions. This has an effect upon the distribution of χ since the Y_{obs} 's which form χ are measurements of the instantaneous lateral position rather than measurements of the momentary average lateral position. To estimate the effects of this on D , the momentary average lateral position and the instantaneous lateral position can be related by a variable correction, Y_{passing} , described by an assigned probability distribution, thus,

$$Y_{\text{true}} = Y_{\text{ave.}} + Y_{\text{passing}}$$

When introduced into Equation A3.6 this yields

$$\chi_1 = \frac{\Delta Y_{ave_1} + \Delta Y_{passing_1} + \Delta Y_{sighting_1}}{\sqrt{\Delta t_1}}$$

To examine the results which the sighting error and passing-motion component have upon the computed diffusion coefficient, D , it is necessary to understand the form and nature of their distributions. Due to the separate origins of the three components, it is appropriate to assume them to be independent. The error distribution originates in a detector with limited accuracy. The passing-motion correction for the true position originates with the interactions between the particles at any one time while the diffusion, i.e. the change in Y_{ave} is caused by the summation of many interactions of particles. Note that the distribution of $\Delta Y_{ave_1}/\sqrt{\Delta t_1}$ will be a normal distribution with mean value of zero and standard deviation of $\sqrt{2\hat{D}^*}$, the true diffusion coefficient for the motion.

Distribution of the Sighting Error. The distribution of $Y_{sighting}$ is obtained from the "wall-sightings" error calibration which is described in Chapter 4. In the calibration procedure the labelled particle is fixed to the wall and thus, there is no passing-motion correction for each observation. The mean value of all the observations taken as wall sightings has been used as the true lateral position of the wall. Subtracting this value of $Y_{average}$ from the other observations produces a distribution of the error, $Y_{sighting}$. The distribution has zero mean value due to the manner in which it was obtained. A chi-

square test of goodness of fit of a normal distribution to Y_{sighting} has a level of significance of .10 . Accordingly, a normal distribution has been assumed for the distribution of sighting error. The standard deviation of Y_{sighting} for the lower sightings of the larger labelled particle is 0.030 inch. For similar conditions with the smaller labelled particle the standard deviation is 0.064 inch.

Assuming that the values of Y_{sighting} used to form $\Delta Y_{\text{sighting}}$ are independent of each other, the distribution of $\Delta Y_{\text{sighting}}$ is directly defined by the distribution of Y_{sighting} . Both will be normal distributions with zero mean values. The standard deviation of $\Delta Y_{\text{sighting}}$ will be twice that of Y_{sighting} .

Distribution of Passing-Motion Error. The distribution of $\Delta Y_{\text{passing}}$ is estimated from a statistical model of the necessary correction of the true instantaneous position to achieve the average position. For simplicity of calculation the passing-motion correction was assumed to have a probability distribution which is uniform over a range of two particle radii and centered about zero, i.e. $P(Y_{\text{passing}}) = \frac{1}{2a}$ for $-a \leq Y_{\text{passing}} \leq +a$ with zero probability for all values of Y_{passing} . Physically this says that any instantaneous measurement is within plus or minus one radius of the average lateral position.

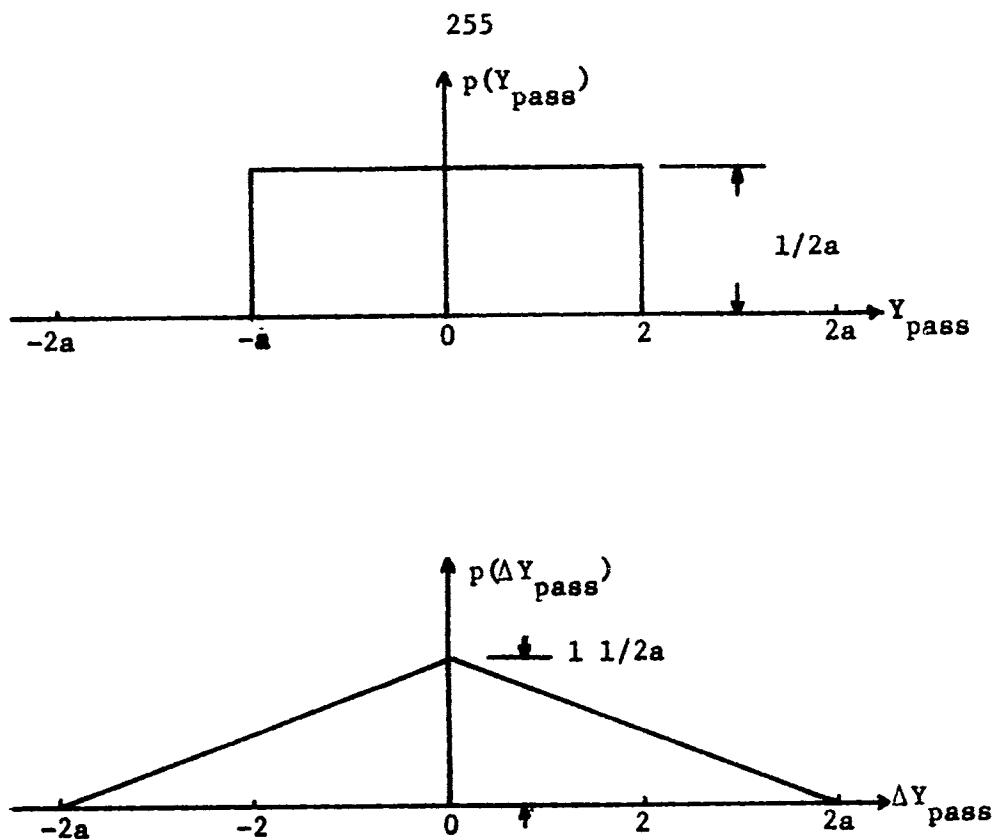


Figure A3.1 Distributions of Y_{pass} and ΔY_{pass} .

The above distribution of passing-motion error does not incorporate all the physical parameters upon which the passing motion is dependent. For example, concentration must play a significant role. If a passing-motion distribution were established for very low concentration, most of the distribution's mass would fall on the point of zero passing motion, simply because of very low concentrations the collisions which produce the passing motion are infrequent events.

At higher concentrations the passing motion distribution would have to include contributions from multiple-particle events since they provide situations where the labelled particle is displaced more than one radius as it passes a cluster of particles. All of the experiments reported here are for high concentrations and have a mean inter-particle distance of less than a radius. Thus, it is expected that the distribution described above will underestimate the phenomena since it does not include a multi-particle effects. However, it should provide a first view of the passing motion error and its effect upon the diffusion coefficient.

With this distribution of Y_{passing} , the distribution of $\Delta Y_{\text{passing}}$ will be triangular and centered about $\Delta Y_{\text{passing}} = 0$. The maximum values of $\Delta Y_{\text{passing}}$ are $\pm 2a$; there is zero probability of $\Delta Y_{\text{passing}}$ being greater than or less than $2a$. Since the distribution of $\Delta Y_{\text{passing}}$ has a mean value of zero and is symmetric, both positive and negative passing corrections are equally probable. Its second moment is $2/3 a^2$ and its fourth moment is $8/15 a^4$.

Effect of Passing-Motion and Sighting Errors upon D. At this point, using the descriptions of the individual distributions which contribute to the total distribution of χ_1 , it is possible to determine a relationship between D as given by one-half of the second moment of the observations and D^* , the true diffusion coefficient which is solely due to changes of Y_{ave} . By definition

$$\begin{aligned}
D &= \frac{1}{2} E(\chi^2) \\
&= \frac{1}{2} E \left[\frac{(\Delta Y_{\text{average}} + \Delta Y_{\text{passing}} + \Delta Y_{\text{sighting}})^2}{\Delta t} \right] \quad (\text{A3.9})
\end{aligned}$$

By considering all the χ_i to have a common average time, Δt , the statistical treatment is made much simpler. If the Δt used is the minimum time interval used by the computer program in compiling the χ_i as described in Chapter 5 the error predicted by this treatment will be slightly larger than the true error.

It is shown in all statistics texts that the expectation of a sum is the sum of the expectations; using this theorem Equation A3.9 can be expressed as

$$\begin{aligned}
D &= \frac{1}{2\Delta t} [E(\Delta Y_{\text{average}}^2) + E(\Delta Y_{\text{passing}}^2) + E(\Delta Y_{\text{sighting}}^2) + \\
&\quad E(\Delta Y_{\text{average}} \Delta Y_{\text{passing}}) + 2E(\Delta Y_{\text{passing}} \Delta Y_{\text{sighting}}) + \\
&\quad 2E(\Delta Y_{\text{sighting}} \Delta Y_{\text{average}})] \quad (\text{A3.10a})
\end{aligned}$$

By assuming that $\Delta Y_{\text{average}}$, $\Delta Y_{\text{sighting}}$, and $\Delta Y_{\text{passing}}$ are independent, the expectations of the cross terms may be split into products of expectations, i.e. $E(\Delta Y_{\text{sighting}} \Delta Y_{\text{passing}}) = E(\Delta Y_{\text{sighting}}) \cdot E(\Delta Y_{\text{passing}})$. The zero mean value of the first moments of all quantities will produce zero values for the expectations of cross terms. Thus, Equation A3.10a will have only even order terms.

$$D \approx D^* + \frac{1}{2\Delta t} E(\Delta Y_{\text{passing}}^2) + \frac{1}{2\Delta t} E(\Delta Y_{\text{sighting}}^2) \quad (\text{A3.10})$$

This result is quite interesting since it shows that D will always be larger than D^* , the true diffusion coefficient.

The last two terms on the right hand side of Equation A3.10 might be considered as apparent diffusions due to passing and sighting error. It also shows that the measurements when compiled as D will depend upon the minimum time allowed between observations, since the distributions of sighting error and passing error are independent of time.

The confidence limits on the value of D can be obtained from the standard deviations which are obtained by substitution of a constant time interval approximation for t into the variance as given by the first term of Equation A3.3a. One obtains after simplification:

$$\begin{aligned} \frac{V(D)}{D^{*2}} &\approx \frac{2}{N} \left[1 + \frac{E(\Delta Y_{\text{sighting}}^2)}{D^* \Delta t} + \frac{E^2(\Delta Y_{\text{sighting}}^2)}{4 D^{*2} \Delta t^2} \right. \\ &\quad + \frac{E(\Delta Y_{\text{passing}}^2)}{D^* \Delta t} + \frac{E(\Delta Y_{\text{passing}}^2) E(\Delta Y_{\text{sighting}}^2)}{D^{*2} \Delta t^2} \\ &\quad \left. + \frac{E(\Delta Y_{\text{passing}}^4) - E^2(\Delta Y_{\text{passing}}^2)}{4 D^{*2} \Delta t^2} \right] \\ &\approx \frac{2}{N} [1 + R] \end{aligned} \quad (\text{A3.11})$$

The error bars plotted on Figures 6.2, 6.4, and 6.5 are two-standard deviation limits as given by the square root of $V(D)$, Equation A3.11. The limits of the error bars provide a range of values which with 95% probability includes the true value of $\hat{D}^*$.

As formed in Equation A3.11, the non-dimensional variance would go to a value representing just the degree of fit between the sample and the finite distribution if the sum of all passing correction and sighting error terms, R , were zero. Beneath each lower error bar in Figure 6.4 is a number which represents the value of the term, $1 + R$ of Equation A3.11. The values of this term may be used as a criterion of error since its value reflects the increased size of the error brackets of D^* by the sighting error and passing motion. However, in the application of this criterion the limitations of the simple distribution of passing motion should be remembered.

Selection of a Minimum Time Interval. One possible method of achieving a small value of the term $1 + R$ is to select a very large value of the minimum time interval, Δt . However, since the diffusive motion increases as the square root of time, such a method is likely to result in a spuriously low value of the diffusion coefficient. The low value is caused by the restriction of the diffusive motion by the walls of the Couette channel.

Due to the fact that each jump may assume a random size only the distribution of jumps can be checked for restriction by the walls. There are two ways of checking the distribution of jumps. One way is

the chi-square statistical test for goodness of fit. In this test the distribution is tested against the ideal, normal distribution. Chi-square tests of the underlying distributions for the data shown in Figure 6.5 have a level of confidence of 0.05 for maximum times of less than 50 seconds. The level of confidence is the fraction of improbable samples which could be drawn from a normal distribution. One says that the data test to a level of significance when they do not belong to the improbable group. Stated alternatively, the data belong to the 95% of the possible samples which could be drawn from a normal distribution. The chi-square test does not specifically test the shape of the distribution; it only tests whether the allotment of elements of the sample from the underlying infinite distribution is probable. Accordingly, it is not a very sensitive test of the amount of wall restriction since wall restriction gradually changes the jump distribution.

The second type of test for wall restriction has a more physical interpretation. A typical diffusion length, ℓ , can be defined as

$$\ell = \sqrt{2D\Delta t}$$

This length is equivalent to the standard deviation of the jump distribution which would be compiled using a time interval, Δt , between the observations which form the jump. Therefore, when the Couette channel is measured in terms of this natural length the amount of wall restriction can be gauged by the amount of the jump distribution which would interfere with the wall. Specifically, the initial position of each of

the diffusive jumps is restricted to the central fifth of the Couette channel, as was done in reducing the data of Figures 6.1, 6.4, 6.5, and 6.9. Then for some minimum time interval Δt , the distance from the edge of the initial region to the channel walls is calculated in diffusion lengths, and the amount of restriction is estimated. Performing this calculation using the diffusion coefficient, D^* , for the minimum time of 4 seconds in Figure 6.4, the distance from the edge of the central fifth of the channel to the wall is found to be three diffusion lengths. Thus, ninety-nine percent of the possible diffusive motions could occur without wall interference. However, in evaluating this diffusion length it should be remembered that the value of D^* used is probably unduly large because of the imperfect correction for the passing motion error.

Accordingly, the same measure of the width of the Couette channel is performed for the minimum time interval of twenty seconds. For this case there are slightly more than two diffusive lengths from the edge of the central one-fifth to the wall of the Couette channel. Thus, over ninety-five percent of the jump distribution for the twenty-second minimum time interval are unrestricted jumps. It also has a smaller amount of apparent diffusion due to sighting errors and the passing motion. For these two reasons all the large-particle data for a shear rate of ten inverse seconds has been reduced using a minimum time interval of twenty seconds.

Data taken for other shear rates and with the small-particle were reduced using "equivalent" minimum times. When the minimum time interval is chosen what is actually selected is a value of the diffusion

length ℓ . Thus, to achieve a constant value of ℓ , the minimum time must scale inversely as the diffusion coefficient, i.e. it must scale as $1/a^2\omega$.

APPENDIX IV. DIFFUSION COEFFICIENTS FOR SPHERICAL-
PARTICLE SLURRIES OF LOW CONCENTRATION
AND FOR DISCOID-PARTICLE SLURRIES

During the time this thesis was written, Bailey [37] has made measurements of the diffusion coefficient for linear shear flows of slurries of spherical particles with volumetric concentrations of 6 and 10 percent. Also he has measured the diffusion coefficient for shear flows of discoid-particle slurries over range of volumetric concentrations from 5 to 40 percent. With the exception of a new, and better, wheel of viewing windows these measurements were performed using the equipment described in Chapter Four. For the convenience of readers interested in Bailey's data, a summary graph of his work has been included as Figure A4.1.

BIOGRAPHICAL SKETCH

The author is the eldest of four children of Robert and Catherine Eckstein. He was reared and attended school in northern Ohio, a region unique only because it is typically "American." Since September, 1964 he has attended MIT. In June of 1970 he received the S.B. and S.M. degrees in Mechanical Engineering.

At the present time the author is healthy, married and has a son. He currently holds the position of Associate in Medicine (Bioengineering) at the Peter Bent Brigham Hospital of Boston, Mass.