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MICROSTRUCTURE AND MOMENTUM TRANSPORT IN CONCENTRATED SUSPENSIONS

Lisa A. Mondy

Sandia National Laboratories, Dept. 9112, Albuquerque, New Mexico 87185-0834

Alan L. Graham

Los Alamos National Laboratory, ESA-EPE, Los Alamos, New Mexico 87545

Howard Brenner

Dept. of Chem. Eng., Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

ABSTRACT

This paper reviews several coupled theoretical and experimental investigations of the effect of microstructure on momentum transport in concentrated suspensions. An expression to predict the apparent suspension viscosity of mixtures of rods and spheres is developed and verified with falling-ball viscometry experiments. The effects of suspension-scale slip (relative to the bulk continuum) are studied with a sensitive spinning-ball rheometer, and the results are explained with a novel theoretical method. The first noninvasive, nuclear magnetic resonance imaging measurements of the evolution of velocity and concentration profiles in pressure-driven entrance flows of initially well mixed suspensions in a circular conduit are described, as well as more complex two-dimensional flows with recirculation, *e.g.* flow in a journal bearing. These data in nonhomogeneous flows and complementary three-dimensional video imaging of individual tracer particles in homogeneous flows are providing much needed information on the effects of flow on particle interactions and effective rheological properties at the macroscale.

INTRODUCTION

Many industrial processes include the transport of suspensions of solid particles in liquids, such as coal and other solid feedstock slurries. Oil, gas, and geothermal energy production rely on the transport of suspensions such as muds, cements, proppant, and gravel slurries in the drilling and completion of a well. Suspensions are also found in high-energy-consumption industrial processes such as found in pulp and paper manufacturing. The complex rheological response of suspensions often limit the efficiency of the design of such processes, causing loss of productivity, increased cost, and increased energy consumption. Because of

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the importance of particulate two-phase flows in the applications described above, the study of suspension rheology remains an important technical research topic for the Department of Energy.

This overview of our recent research supported by the Department of Energy, Office of Basic Energy Sciences, will focus on flow of suspensions of relatively large particles in which colloidal and inertial effects are negligibly small. There is growing evidence that even in this restricted range of flows, the rheology of a suspension with a nondilute particle concentration cannot be characterized by a material function. Instead, the microstructure of the suspension determines the overall macroscopic properties, and the flow history of the suspension determines aspects of the microstructure. Advances in the ability to predict the rheological response of concentrated suspensions depend on answering three broad questions: 1) How does the microstructure of a suspension affect the rheological properties? 2) How do boundaries, such as walls, affect the microstructure and properties? 3) How does the macroscopically imposed flow field affect the microstructure of a suspension? Aspects of these questions are being addressed in our work.

In the following section we will explore the first question by discussing the use of falling-ball rheometry as a means to circumvent the shear-induced changes in microstructure that can be encountered when using conventional rotational devices to measure suspension viscosity. We will discuss falling-ball rheometry used to determine the apparent viscosity of a suspension of particles of two shapes. In the third section we will discuss experimental and theoretical aspects of spinning-ball rheometry in otherwise quiescent suspensions and show that this can provide a sensitive measure of slip at the surface of a particle.

The fourth section of this paper focuses on efforts to develop capability to predict the evolution of concentration and velocity profiles of an initially well mixed suspension as it demixes when subjected to non-homogeneous shear flows. If the local concentration is known, one can then use the falling-ball information to determine the *local* viscosity in a flow field. Global behavior can then be determined by incorporating a spatially varying viscosity field into the usual balance equations. We will illustrate the existence of flow-induced microstructural changes with data on the time evolution of concentration and velocity profiles in suspensions undergoing flow in pipes and between counter-rotating eccentric cylinders (journal bearings). When the suspended particles are small in comparison to the characteristic dimensions of the flow apparatus, steady-state concentration and velocity profiles are in good agreement with predictions of the shear-induced migration model [1,2]. However, another avenue to modeling particle migration is to use a kinetic theory approach, which has been applied successfully in granular flows [3,4]. In this theory the intensity of the velocity fluctuations, caused by particle interactions, is characterized by a 'granular temperature' analogous to the temperature in classical kinetic theories and governed by a balance of fluctuation energy. Under some situations this approach leads to the same balance equations as with the first model, but with a hydrodynamic diffusion that can be determined in homogeneous flow fields. We will describe experiments where we use particle tracking techniques, originally developed in falling-ball studies, to determine the granular temperature of various suspensions undergoing homogeneous flow between parallel moving belts.

FALLING-BALL RHEOMETRY IN COMPLEX SUSPENSIONS

In previous work, we have shown that falling-ball rheometry is an excellent tool to probe the rheological properties of a suspension without significantly changing the properties through the very act of measuring them. Unlike conventional viscometers, which employ flow fields that tend to influence the microstructure of the suspension, falling-ball rheometry can be used to determine the macroscopic viscosity of a suspension with little effect on the microstructure [5]. This is especially useful for suspensions of particles with aspect ratio greater than one, whose alignment is especially sensitive to the flow field. We have recently begun to use falling-ball rheometry to study suspensions of particles with a mix of shapes.

Most investigations on the rheology of concentrated suspensions have focused on monodisperse suspensions of either spherical or rodlike particles. In practice, most suspensions contain particles that are polydisperse both in size and shape. Only a limited number of studies have been devoted to the problem of size polydispersity in suspensions of spherical particles, and even less is known about the behavior of suspensions composed of particles of different shapes.

Farris [6] develop a model for the viscosity of suspensions of spheres with multimodal diameter distributions. In his model, for each fraction of a given particle size, the smaller particles in suspension have the same effect as a homogeneous fluid with Newtonian viscosity similar to the viscosity of a suspension made up only of the fraction of smaller spheres. In other words, the smaller suspended particles do not interact with the larger particles and are 'sensed' by the larger particles as part of the continuous suspending fluid. We will apply these concepts to develop an equation for the relative viscosity of a suspension composed of a mixture of rodlike and spherical particles. If the rods are large enough relative to the spheres, we may consider the spherical particles as part of the homogeneous suspending continuum. Let us define an apparent sphere volume fraction $\phi_s^* = V_s / (V_0 + V_s) = \phi_s / (1 - \phi_r)$, where V is volume and the subscripts s , 0 , and r stand for the spheres, fluid, and rods, respectively. If we assume the viscosity of a suspension composed of spheres and rods is the same as the viscosity of a suspension of rods suspended in a Newtonian homogeneous fluid of viscosity identical to the viscosity of an equivalent suspension of spheres with a volume fraction ϕ_s^* , we may write (after Farris):

$$\mu_{\text{rel}}(\phi_T) = \mu_{\text{rel, spheres}}(\phi_s^*) \mu_{\text{rel, rods}}(\phi_r) \quad (1)$$

where the relative viscosity of a suspension is the viscosity of the suspension normalized by the viscosity of the suspending continuum. Several expressions are available for the relative viscosity of suspensions of spheres (e.g. those listed by Graham et al. [7]). Here we adopt the following empirical relation [8], which has agreed well with previous falling-ball measurements:

$$\mu_{\text{rel, spheres}}(\phi) = 1 + 2.5(\phi) + 10.05(\phi)^2 + 0.00273e^{(16.6\phi)} \quad (2)$$

For the viscosity of a suspension of randomly oriented rods, we have the following empirical relation for rods with aspect ratio of 20 [5]:

$$\mu_{\text{rel, rods}}(\phi) = 1 + 28.50\phi \quad \phi < 0.125 \quad (3a)$$

$$= 1 + 2.040\phi^3 \quad \phi > 0.125 \quad (3b)$$

The relative viscosity of a mixed suspension may now be calculated for any combination of rods (of aspect ratio 20) and spheres by the set of equations (1)-(3).

The falling-ball experimental apparatus, materials, and methodology have been described in detail previously [5,9]. The suspensions were composed of mixtures of poly(methyl methacrylate) particles in a Newtonian liquid. The particles were mixtures of spheres, with diameters of 3.175 mm, and aspect-ratio-20 rods, with length of 31.65 mm. The rod-sphere mixtures were suspended in a liquid solution with three primary ingredients (50wt% alkylaryl polyether alcohol, 35wt% polyalkylene glycol, and 15wt% tetrabromoethane). The weight fractions of the ingredients were adjusted so that the density and the refractive index of the fluid would match those of the particles. Three different suspensions were prepared with total solids volume fraction ϕ_T of 0.35 (volume fraction of rods $\phi_r=0.05$ and of spheres $\phi_s=0.30$), 0.40 ($\phi_r=0.10$ and $\phi_s=0.30$), and 0.45 ($\phi_r=0.05$ and $\phi_s=0.40$) respectively. The falling balls were either chrome-plated steel, monel, or tungsten-carbide ball bearings with diameters between 6.35 mm and 15.88 mm.

The trajectories of the falling balls were recorded on a high-speed digitizing video system. An average velocity for an individual experiment was determined by measuring the elapsed time for a ball to settle a known distance on the screen. The results of up to 40 individual experiments with a falling ball of one nominal size (not necessarily of one material) were averaged to determine a reproducible effective viscosity of the suspension. Two or three sizes of falling balls were used for each suspension and the results showed no significant effect of the relative sizes of the falling ball and the suspended particles over the size ranges listed above. The average apparent relative viscosity for each suspension was obtained by averaging the entire

set of experimental results (up to 120 individual experiments).

These results are shown in Figure 1 along with the values predicted by equations (1)-(3). The solid and dotted lines represent the relative viscosity for a suspension of randomly oriented rods of aspect ratio $a_r=20$ [eq. 3a and 3b] and for a suspension of spheres [eq. 2], respectively. The broken lines represent the calculated viscosity for mixed rod-sphere suspensions with the indicated fraction of rods (5%, 10%, and 15%) based on the predictions of eq. 1 combined with eqs. 2 and 3. The agreement between the falling-ball experimental points and the calculated lines is very good.

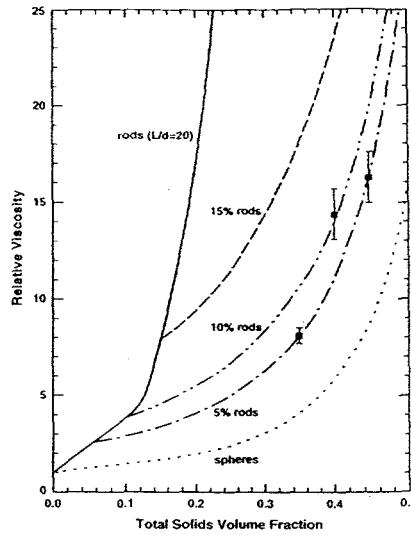


Figure 1. Falling-ball viscosities of mixtures of suspended rods and spheres.

SLIP STUDIES

Spinning-Ball Experiments

In the falling-ball experiments described in the section above, the drag on the ball appeared to be that found in a Newtonian liquid with *no slip* at the ball boundaries. Instead of measuring the mean velocity of a falling ball, we could instead measure the mean torque on a spinning ball. This geometry is more sensitive to slip at the ball boundary. Whereas the force F on a ball moving slowly through an unbounded Newtonian liquid without slip can be described as $F=6\pi a\mu v$ (where μ is the viscosity of the liquid and a and v are the radius and velocity of the ball, respectively), the force with perfect slip is $4\pi a\mu v$. In contrast, the torque T on a ball spinning slowly in a Newtonian liquid is described by Kirchoff's law, $8\pi a^3\mu\Omega$. (where Ω is the angular velocity of the ball); however, the torque on a ball with perfect slip at the boundaries is zero [10].

Kunesh and coworkers studied the torque on balls spinning in single-phase Newtonian liquids, verified the formula above, and quantified the effects of the free surface [11]. We have completed similar experiments to measure the torque on balls spinning in otherwise quiescent suspensions. We measured the torque on three sizes of balls (0.32, 1.27, and 2.54 cm in diameter) spinning in various suspensions. Suspension with solids volume fractions of 0.25, 0.40, and 0.50 were studied. Three sizes of suspended spheres (0.07, 0.32, and 0.64 cm in diameter) were used in the suspending oil described earlier. The suspensions were well mixed prior to the start of an experiment.

Typical traces of the torque on a 1.27 cm-diameter ball in terms of the relative viscosity (the measured spinning-ball viscosity normalized by capillary viscosity measurements of the suspending liquid) in the suspending liquid and in a suspension with $\phi=0.5$ is shown in Figure 2. The suspending liquid measurements agree well with capillary measurements and show no variation with time (number of revolutions). On the

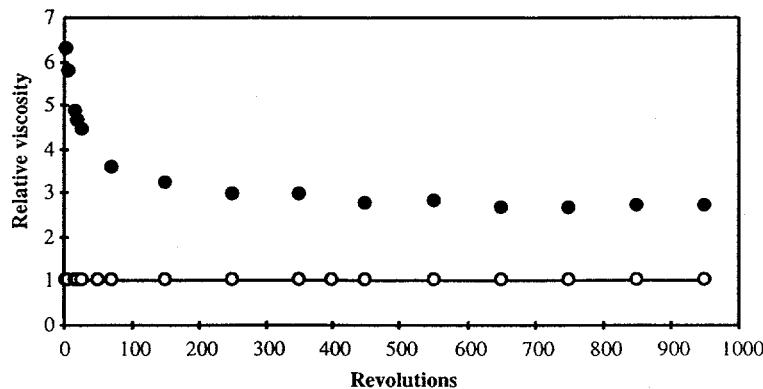


Figure 2. Spinning-ball viscosities (relative to capillary measurement of the suspending liquid viscosity) for the suspending liquid (○) and for a suspension with $\phi=0.50$ (●).

other hand, the suspension measurements show a distinct fall off in the measured viscosity with increasing numbers of revolution. This is expected, as this nonhomogeneous flow induces particle migration (which will be discussed in the following section). The short-time behavior (an average torque for the first four revolutions) is taken as an indication of any apparent slip at the ball's surface in the still homogeneous suspension. The effects of the relative sizes of the spinning ball and the suspended spheres are shown in Fig. 3. Here, all data are taken in suspensions with $\phi=0.5$; however, the suspended-sphere size varies as well as the spinning-ball size. As the suspended spheres become small compared to the spinning ball, the spinning-ball viscosity increases. The torque experienced (initially) on a 2.54-cm-diameter ball spinning in a suspension of 0.07-cm-diameter particles is correspondent to the viscosity measured with falling-ball rheometry. Conversely, when the spinning ball and the suspended spheres are comparable in diameter, the presence of significant 'Kirchoff-law slip' is observed.

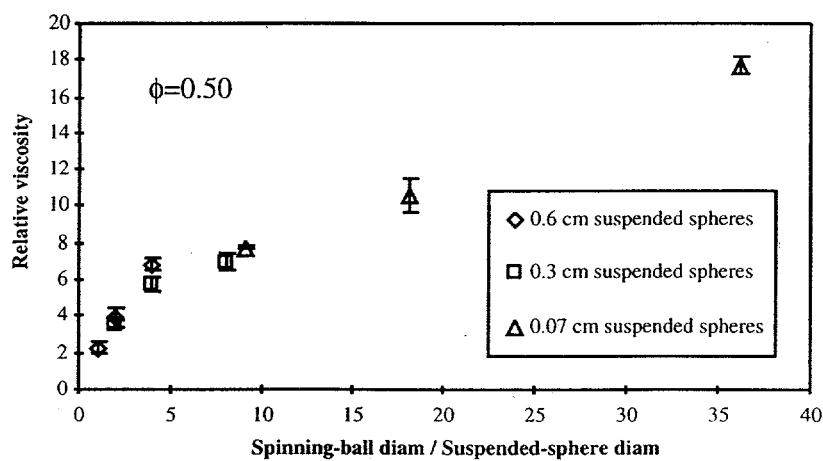


Figure 3. The effect of the relative sizes of the spinning ball and the suspended spheres on the initial apparent viscosity measured assuming Kirchoff's law. The data point to the far right is close to the value predicted by falling-ball studies [23] and conventional rheometry [8].

Theoretical Developments

Einstein's [12] classical analysis of the rheology of a dilute suspension related the increased viscosity of the suspension to the additional dissipation occurring within a 'suspension cell' owing to the perturbing

presence of a freely suspended sphere in an otherwise uniform shear field. However, scalar dissipation arguments are viable only in cases where the suspension behaves macroscopically as a homogeneous isotropic fluid. In particular, these methods are inapplicable in circumstances where the suspension-scale stress/strain-rate relationship is anisotropic. Batchelor [13] and Brenner [14] developed a general theory from which the stress/strain-rate relation may be obtained. Their methods are based on averaging over a 'suspension cell' the interstitial-scale stress and velocity gradient tensors. Higher-order terms in the relative viscosity/suspended-particle concentration expansion have been obtained by Batchelor & Green [15,16], based on an 'ensemble average' approach. Each of the above methods is essentially local in nature; that is, effects of bounding walls as well as spatial nonhomogeneities in the ambient velocity gradient are neglected. When the ensemble-average approach is applied, and the existence of walls ignored, nonconvergent integrals arise, which require *ad hoc* renormalization methods to overcome.

Recently, a new method has been developed for rheologically homogenizing a dilute suspension composed of freely suspended spherical particle dispersed in a Newtonian fluid [17]. The method is global in nature; that is, wall effects and spatial dependence of both the ambient flow and the particle number density are encountered, enabling known classical results for the suspension viscosity to be obtained without the need for renormalization.

When the ambient flow is singular (as for example in the case of a small sedimenting or rotating ball comparable in diameter to the suspended spheres) it is possible to use this technique to estimate the velocity at points far from the singularity. In a recent paper, it is shown that even far from the singularity (relative to the freely suspended sphere radius), the suspension does *not* behave like a homogeneous medium [17]. Specifically, due to interparticle hydrodynamic interactions, the average extra-torque exerted on a ball rotating at a given angular velocity (and, conversely, the average reduction in angular velocity experienced by a sphere on which a given torque is exerted), are not related by the Kirchoff's law linear factor $8\pi d^3\mu$, instead a suspension-scale 'slip' occurs at the surface of the spinning ball in agreement with the experimental work discussed above. Furthermore, the extra-torque felt by a ball held at constant angular velocity in a suspension and the reduction in angular velocity felt by the same ball held at constant torque do not correspond directly. In fact, when the ratio of the spinning-ball diameter to the suspended-sphere diameter is roughly one, the dimensionless extra-torque is almost 25 per cent larger than the comparable reduction in angular velocity. This phenomenon cannot occur in a homogeneous medium for which the constitutive stress/rate-of-strain relationship is an intrinsic material property of the system.

In the case of a sedimenting ball, the 'apparent viscosity' obtained experimentally by the supposed applicability of Stokes law agrees with the viscosity of the suspension measured by standard viscometric methods if the ball is the same size or larger than the suspended spheres [9]. However, if the ball is somewhat smaller, the reduction in sedimentation velocity is *less*, apparently because of a 'slip' at the surface of the sedimenting ball [18]. Recent theoretical results show this as well for dilute suspensions [19]. It is interesting to note that the appearance of 'slip' occurs over a smaller range of the ratio of the tracer (in this case, falling) ball to the suspended particles than in the rotating ball case. Furthermore, the theory predicts that if the falling ball is yet smaller relative to the suspended spheres than those studied experimentally, the reduction in sedimentation velocity then becomes significantly *higher*. This contrasting behavior arises from the difference in the respective probability density functions for the cases of sedimenting and rotating spheres. For the case of a rotating sphere, the probability density function $P(x_1/s_0)$ is constant (where x_1 is the location of the center of a suspended sphere and s_0 corresponds to the domain inside a sedimenting or rotating sphere) and independent of the relative sizes of the rotating and suspended spheres. In contrast, in the case of a sedimenting sphere, it exhibits large gradients near s_0 for very small sedimenting spheres, rendering the near-field contribution dominant. Since the settling velocity decreases significantly when the settling and freely suspended spheres nearly touch, the reduction in sedimentation velocity increases proportionally. In other words, whereas the rotating sphere has only one 'interaction' mechanism (namely that the overall effect of hydrodynamic interactions decays monotonically with decreasing ratio of the rotating-to-suspended sphere diameter since the domain in which the effect of the suspended sphere is sensible shrinks), the settling sphere has two competing mechanisms. The first is similar to that of the rotating

sphere. The second is the increase of probability density function in the vicinity of the singularity with decreasing ratio of the rotating-to-suspended sphere diameter, which makes the near-field contribution dominant in spite of the fact that the domain in which this effect is significant shrinks.

EFFECTS OF FLOW ON THE MICROSTRUCTURE OF SUSPENSIONS

Pressure-Driven Pipe Flow

Flow-induced migration of suspended particles is thought to occur whenever particle interactions are more frequent in one part of a flow field than in another, as could occur in the presence of spatially varying shear rate, concentration, or viscosity fields. The spatial distribution of suspended particles present in concentrated suspensions is difficult to measure because most suspensions are opaque even at relatively low particle concentrations. However, under the auspices of the Department of Energy, Office of Basic Energy Sciences, noninvasive techniques based on nuclear magnetic resonance (NMR) imaging have been developed by Fukushima and coworkers to study both concentration and velocity profiles in multiphase flows [20,21]. We have employed these NMR imaging techniques to study the flow-induced migration of particles in suspension when subjected to a variety of flow fields.

One of our more recent studies involved low-particle-Reynolds-number pressure-driven flow in a circular conduit of suspensions ranging in solids volume fraction ϕ of 0.1 to 0.45. Measurements were made using 3.175-mm-diameter particles in a 50.4-mm-diameter tube ($a/R = 0.0625$) and 675-mm-diameter particles in a 25.4-mm-diameter tube ($a/R = 0.0266$). The primary data obtained from these experiments were NMR images of the concentration (ϕ) and velocity (v) fields at various locations downstream of an in-line mixer.

During flow development, significant migration to the axis of the tube and 'plug-like' velocity profiles were observed at all solids volume fractions. Full flow development occurred sooner than predicted by existing scaling arguments. Evidence suggests that, at higher concentrations ($>30\%$), evolution of the ϕ and v profiles occur on different length scales. Two flow rates were tested (9.89 mm/s and 197.7 mm/s). The development of the ϕ and v profiles were independent of flow rate.

Example steady-state profiles are shown in Figure 4. At the lower ratio of a/R the ϕ profile achieves a cusp at the center of the flow. The higher a/R (0.0625) is significantly above the ratio suggested by Seshardi & Sutera [22] and Mondy et al. [23] to be the upper limit of continuum behavior. Particle size effects manifest themselves as somewhat more blunted concentration profiles at larger a/R . The depletion in ϕ apparent near the wall would result in a 'layer' of lower viscosity and a reduction in the pressure drop required to flow the suspension, which is also consistent with the findings of Mondy et al. [23].

Piston-Driven Pipe Flow

In contrast to pressure-driven pipe flow, piston-driven pipe flow is not unidirectional. At the surface of the moving piston the velocity profile is necessarily uniform, yet downstream the velocity profile becomes parabolic for a Newtonian liquid. In order for this to occur, continuity requires that liquid near the pipe walls be swept into the center of the pipe. We find that this complex flow leads to particle migration in both the radial and axial directions.

Recently, we have studied the flow of a concentrated suspension in a 38 cm long by 5 cm diameter pipe equipped with a driving piston at one end and a freely moving piston at the other. This geometry results in a closed system ideal for an NMR imaging study of a two-dimensional flow. Two suspensions were imaged, one of 0.07-cm-diameter spheres and one of 0.32-cm-diameter spheres, both at an overall solids volume fraction of 0.50. Figure 5A shows an image of the suspension of smaller spheres in the region near the driving piston after the piston has traveled approximately 5 pipe diameters. A region of high liquid content has formed near the pipe walls and has been swept to the pipe axis along the piston face. Figure 5B shows the radially averaged solids volume fraction, in the suspension of larger spheres, along the axis of the pipe from

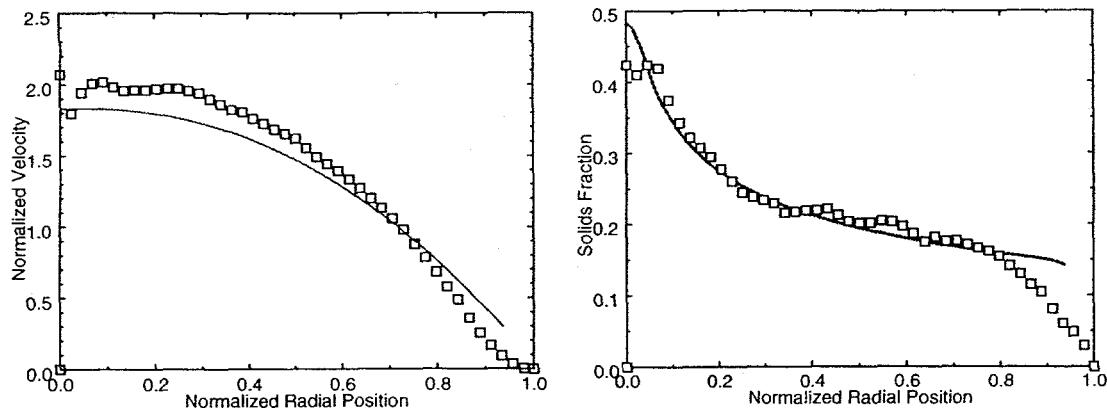


Figure 4. Steady-state v and ϕ profiles for $a/R = 0.0625$ at $\phi = 0.20$ as measured with NMR (symbols) and predicted values (lines) from the improved shear-induced migration model.

the driving piston to the freely moving one. The particles 'lead' the fluid and concentrate at the far end of the flow (farthest from the driving piston).

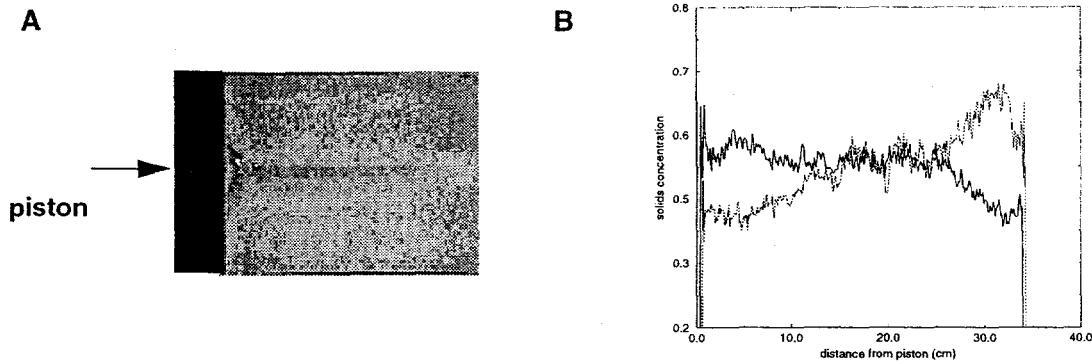


Figure 5. A. NMR image near the driving piston. Dark areas represent regions of higher liquid fraction. B. Radially averaged solids volume fraction along the axis of the pipe from the driving piston to the freely moving one.

Numerical Modeling and Constitutive Equation Development

The NMR data discussed above has been compared with the results of a constitutive model developed by Phillips et al. [2] after Leighton & Acrivos [1]. This constitutive model consists of both a Newtonian constitutive equation, in which the viscosity depends on the local particle volume fraction, and a diffusive equation that accounts for shear-induced particle migration. Two adjustable parameters arise in the diffusive equation, which describe the relative strength of the mechanisms for particle migration. These two rate parameters can be evaluated empirically with experimental measurements of velocity and concentration profiles in a wide-gap Couette apparatus. We have recently determined these parameters as functions of ϕ .

One criticism of this model was its prediction of a cusp-shaped concentration profile in pressure-driven pipe flow. However, the recent NMR experiments have shown that such concentration profiles can occur, as shown in Figure 4, which is of one of the more blunted obtained. The original Phillips formulation [2] has been modified so that the shear rate is averaged over the size of a particle. This results in different fully

developed profiles for different particle sizes, in agreement with the data. Examples of the predictions and the close agreement with data can be seen in Figure 4. However, the original formulation and the new model both overpredict the entrance length in comparison to the data.

The second criticism was the inherently one-dimensional treatment of the particle migration (it is dependent on gradients in a scalar shear-rate). Although this could still be an impediment to generalizing the model to complex flows, we have found that this simple model can often predict the very complex behavior of suspensions. The constitutive expression previously described by Phillips et al. [2] has been expanded to two-dimensional flows by describing the flow in terms of the strain rate tensor \mathbf{D} and the migration in terms of gradients in the generalized shear rate $\dot{\gamma} = (2 \operatorname{tr} \mathbf{D}^2)^{1/2}$. The equation set was then solved numerically and the predictions compared to NMR imaging data. NMR imaging has also been used to study the flow of concentrated suspensions in the gap between a rotating inner cylinder placed eccentrically within an outer fixed cylinder (a journal bearing). We reported earlier [24] that this model, when coupled with a finite volume solver, failed to capture the qualitative nature of this two-dimensional flow. Specifically, at certain values of the eccentricity a very slow recirculation occurs and concentrated suspensions evolve a concentration profile with the maximum concentration of solids occurring, not at the outer wall, but inside the gap. The earlier numerical results always predicted a monotonic increase in solids volume fraction from the rotating inner cylinder to the outer wall, and no recirculation zone was predicted. However, recently we have used a finite element technique with more success. Figure 6 shows the development of a spatially varying solids volume fraction as the number of turns of the inner cylinder increases. The predicted profiles are remarkably similar to the NMR images. In addition, the calculations do indeed predict a recirculation zone.

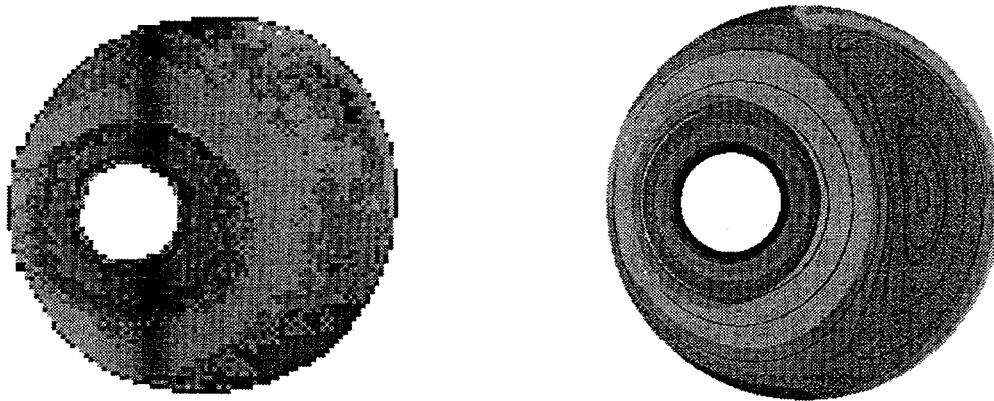


Figure 6. NMR image of liquid volume fraction contours (left) and predicted contours of liquid volume fraction and streamlines (right) for a suspension undergoing flow in a cylindrical journal bearing after 1000 turns.

The finite element model was also used to predict the behavior of concentrated suspensions undergoing piston-driven flow. Figure 7 shows the predicted spatially varying concentration obtained under the same conditions as the experiments described in the previous subsection. The contour plot is remarkably similar to the NMR results shown in Figure 5.

Microrheological Observations

Despite some successes with the above model, we feel that other avenues should continue to be explored to ensure that the particle migration phenomena is adequately understood and appropriately generalized to multiple dimensions. One such avenue recently suggested is to use a kinetic theory approach, which has

been applied successfully in granular flows [3,4]. In this theory the intensity of the velocity fluctuations, caused by particle interactions, is characterized by a "granular temperature" analogous to the temperature in classical kinetic theories and governed by a balance of fluctuation energy. This approach emphasizes the importance of measuring not only average behavior of suspensions but the details of the fluctuations about those averages. Under some situations this approach leads to the same balance equations as with the first model, but with a hydrodynamic diffusion that can be determined in homogeneous flow fields.

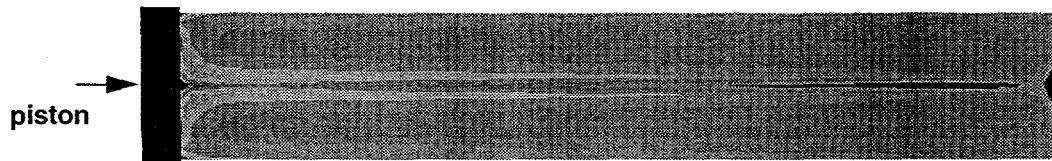


Figure 7. Predicted concentration contours in piston-driven flow using a finite element code.

Currently, we are using particle tracking techniques, originally developed in falling-ball studies, to determine the granular temperature of various suspensions undergoing homogeneous flow between parallel moving belts. The suspensions consist of poly(methyl methacrylate) 0.635-cm-diameter spheres neutrally buoyant in the oil mixture described earlier. The suspended spheres are primarily transparent, with the same index of refraction as the suspending liquid. A few opaque tracer spheres, otherwise identical to the others, are added to the suspension.

To date, 100 detailed three-dimensional trajectories of the tracer spheres in a suspension with $\phi=0.20$ undergoing flow at two different shear rates have been recorded. Figure 8A shows one such trajectory (in only two of the three directions), where the y-direction is parallel to the belt and in the direction of motion and the x-direction is in the direction of the overall velocity gradient. The origin is at the center of the device. Figure 8B shows the velocity fluctuations in the x-direction for an ensemble of 50 particles at a shear rate of 3.34 sec^{-1} . From our preliminary measurements, it appears that the velocity fluctuations are more or less isotropic. The granular temperature in each direction is defined as $T = \langle u' \cdot u' \rangle_p$, where u' is the velocity fluctuation of a particle about its local mean velocity and the angled brackets denote ensemble averaging over all the particles. Preliminary results for T_x (in the x-direction) at two shear rates are shown in Figure 8C.

Currently we are instrumenting the homogeneous flow apparatus with piezoelectric pressure sensors that will allow the measurement of the frequency of particle-wall interactions, as well as the additional pressure due to the presence of the particles. These are pieces of information critical to the evaluation of granular-flow based suspension rheology models.

CONCLUSIONS

We have performed a variety of experimental, theoretical, and numerical studies to elucidate the linkage between the microstructure and the macroscopically observed responses of suspensions of particles in liquids. NMR imaging studies and visual observations have confirmed that a suspension's microstructure can change dramatically during flow. Falling-ball viscometers, on the other hand, can be used (under certain circumstances) to determine an apparent viscosity of a *homogeneous* suspension, without significantly affecting the microstructure during the measurement. Quiescent suspensions can also be used to examine effects of boundaries. We have described one such measurement: the torque on a rotating ball in otherwise quiescent suspensions. Recent theoretical results have also shed light on experimental results indicating Stokes law and Kirchoff's law could be presumed to hold only under limited circumstances.

Information obtained with quiescent suspensions can be combined with information about the evolving microstructure in a flow to predict the spatial variations in viscosity and the global behavior. We have had successes in modeling multidimensional flows with an approach that describes shear-induced particle migration with a diffusive equation. However, further studies of the details of particle interactions are needed

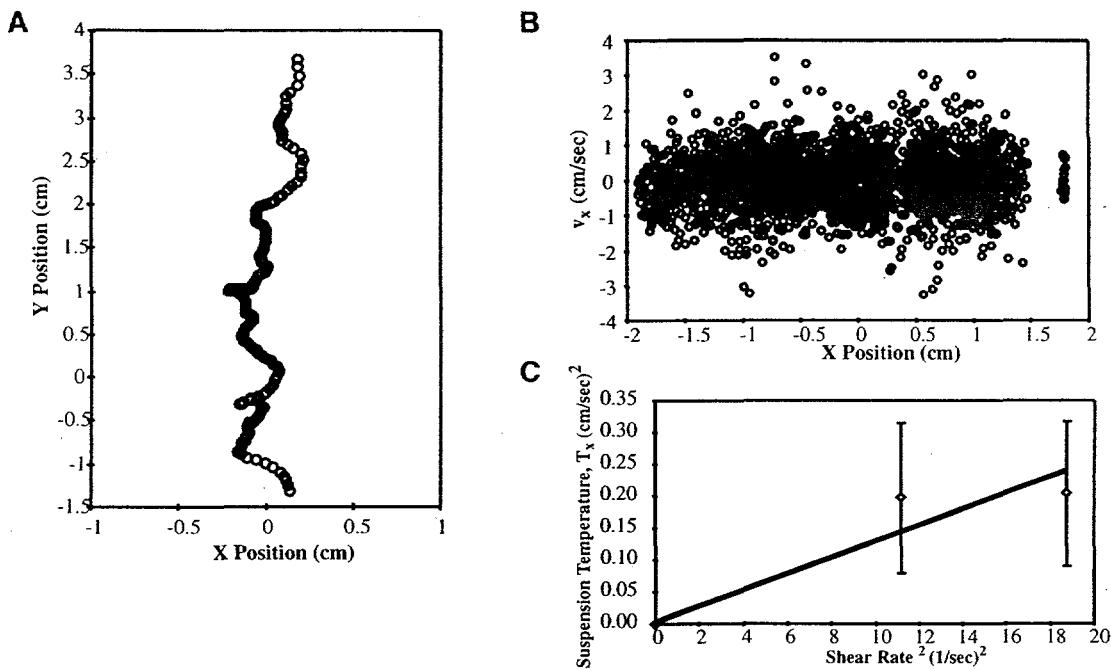


Figure 8. A. The trajectory of a tracer sphere in a suspension with $\phi=0.20$ undergoing uniform shear. B. Velocity fluctuations of 50 particles in this flow. C. Resultant granular temperatures at two shear rates.

before definitive predictive capabilities can be developed. Measurement of the detailed fluctuations of the velocity of particles in suspension undergoing flow is an example of one such study.

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REFERENCES

1. Leighton, D. and Acrivos, A., *J. Fluid Mech.* 275, 155-199 (1987).

2. R. J. Phillips, R. C. Armstrong, R. A. Brown, A. L. Graham, J. R. Abbott, *Phys Fluids A* 4, 30-40 (1992).
3. D.F. McTigue and J. T. Jenkins, "Channel flow of a concentrated suspension," In *Advances in Micromechanics of Granular Materials*, H. H. Shen et al., Editors, Elsevier Science Publishers, New York, 1992.
4. P. R. Nott and J. F. Brady, *J. Fluid Mech.*, 275, 157-199 (1994).
5. W. J. Milliken, L. A. Mondy, M. Gottlieb, A. L. Graham, and R. L. Powell, *J. Fluid Mech.* 202, 217 (1989).
6. R. J. Farris, *Trans. Soc. Rheol.* 12, 281 (1968).
7. A. L. Graham, R. D. Steele, and R. B. Bird, *Ind. Eng. Chem. Fundam.* 23, 420 (1984).
8. D. G. Thomas, *J. Colloid Sci.* 20, 267 (1965).
9. L. A. Mondy, A. L. Graham, and J. Jensen, *J. Rheol.* 30, 1031 (1986).
10. H. Lamb, *Hydrodynamics*, Dover Publications, New York, 6th edition, 1945.
11. J. G. Kunesh, H. Brenner, M. E. O'Neill, and A. Falade, *J. Fluid Mech.* 154, 29-42 (1985).
12. A. Einstein, *Ann. Physik* 19, 289-306 (1906). Errata, *Ann. Physik* 34, 591-592 (1911).
13. G. K. Batchelor, *J. Fluid Mech.* 41, 545-570 (1970).
14. H. Brenner, *Chem. Engng. Sci.* 27, 1069-1107 (1972).
15. G. K. Batchelor, and J. T. Green, *J. Fluid Mech.* 56, 375-400 (1972).
16. G. K. Batchelor, and J. T. Green, *J. Fluid Mech.* 56, 401-427 (1972).
17. Y. Almog and H. Brenner, "Renormalization-free homogenization of a dilute suspension of spheres," preprint (1996).
18. W. J. Milliken, L. A. Mondy, M. Gottlieb, A. L. Graham, and R. L. Powell, *Phys. Chem. Hydro.* 11, 341 (1989).
19. Y. Almog and H. Brenner, "Non-continuum anomalies in the apparent viscosity experienced by a test sphere moving through an otherwise quiescent suspension," preprint (1996).
20. A. Caprihan and E. Fukushima, *Physics Reports* 198, 195-235 (1990).
21. P. D. Majors, R. C. Givler, and E. Fukushima, *J. Magn. Reson.* 85, 235-243 (1989).
22. V. Seshadri and S. P. Sutera, *Trans. Soc. Rheol.* 14, 351 (1970).
23. L. A. Mondy, A. L. Graham, and M. Gottlieb, *Proceedings of the Xth International Congress on Rheology*, Sydney, Australia 2, 137 (1988).
24. N. Phan-Thien, A. Graham, S. A. Altobelli, J. R. Abbott, and L. A. Mondy, *Ind. Eng. Chem Res.* 34, 3187-3194 (1995).

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