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Hydrodynamic diffusion of suspended particles: a symposium

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Preamble

Hydrodynamic diffusion refers to the fluctuating motion of non-Brownian particles (or droplets or bubbles) in a dispersion, which occurs due to multiparticle interactions. For example, in a concentrated sheared suspension, particles do not move along streamlines but instead exhibit fluctuating motions as they tumble around each other (figure 1*a*). This leads to a net migration of particles down gradients in particle concentration and in shear rate, due to the higher frequency of encounters of a test particle with other particles on the side of the test particle which has higher concentration or shear rate. As another example, suspended particles subject to sedimentation or fluidization do not generally move relative to the fluid with a constant velocity, but instead experience diffusion-like fluctuations in velocity due to interactions with neighbouring particles and the resulting variation in the microstructure or configuration of the suspended particles (figure 1*b*). In flowing granular materials, the particles interact through direct collisions or contacts; these collisions also cause the particles to undergo fluctuating motions characteristic of diffusion processes. Although the existence and importance of hydrodynamic diffusion of particles have been embraced only in the past several years, the subject has already captured the attention of a growing number of researchers in several diverse fields (e.g. suspension mechanics, fluidization, materials processing, and granular flows).

An international symposium was held at the YMCA of the Rockies conference center in Estes Park, Colorado, during 22–25 July 1995 to discuss the current state of knowledge and the need for continued research on hydrodynamic diffusion. The symposium was sponsored by the International Union of Theoretical and Applied Mechanics, with co-funding from the National Science Foundation, the National Aeronautics and Space Administration, the Office of Basic Energy Sciences of the US Department of Energy, and the Centre National de la Recherche Scientifique. The symposium had 68 attendees from academic, government, and industrial research laboratories in 12 countries. The variety of backgrounds (applied mathematics, chemical engineering, fluid mechanics, materials, mechanical engineering, mechanics, and physics) led to lively discussion, as different points of view were expressed and debated.

The scientific committee and program committee for the symposium included Andreas Acrivos, John Brady, Robert Davis (Committee Chair), Francois Feuillebois, John Hinch, Jim Jenkins, Don Koch, David Leighton, Ron Phillips, Robert Powell, Werner Schneider Eric Shaqfeh, and Leen van Wijngaarden. The symposium began on Saturday evening with an opening talk followed by discussion and a reception. Each subsequent session included an invited lead-off presentation, followed by several related contributions. In addition to discussion following each talk, an open discussion

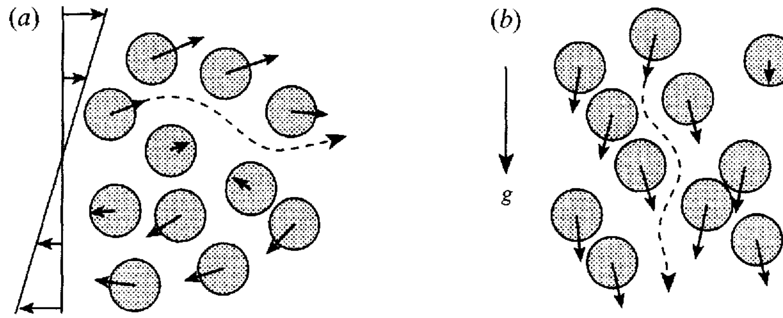

FIGURE 1. Schematic of particle velocities and the trajectory of a test particle in a suspension subject to (*a*) shear flow and (*b*) sedimentation.

period was included at the end of each session. The symposium also had a poster session held on the last evening; the format included an initial poster viewing period, followed by two-minute oral presentations for all posters, and then a second viewing period with refreshments. Extended abstracts were collected for all presentations, including the posters, and distributed in a booklet at the symposium.

This report summarizes the key findings and areas of discussion at the symposium. The report is organized around common themes which correspond to the session topics. I have attempted to extract the essence of the presentations and ensuing discussions, and I apologize to any participants whose points of view have been misinterpreted or overlooked.

1. Hydrodynamic diffusion during sedimentation and fluidization

Although simple in principle, sedimentation of particles presents perhaps the greatest challenge to our understanding of hydrodynamic diffusion. In his opening talk of the meeting "*What is so puzzling about hydrodynamic diffusion?*" Don Koch (Cornell University) noted that the slow spatial decay of the disturbance velocity fields surrounding small sedimenting particles in Stokes flow causes the particle velocity variance and hydrodynamic diffusivity in an infinite suspension of randomly distributed particles to diverge (Caflisch & Luke 1985; Hinch 1988); the inference drawn is that finite values must depend on the size of the container and/or on the establishment of non-random suspension microstructure (Koch & Shaqfeh 1991). This theme, and the discrepancy between experiments and simulations, was a subject of considerable discussion throughout the symposium.

In her opening talk for the session on sedimentation and fluidization, Elisabeth Guazzelli (Ecole Supérieure de Physique et Chemie Industrielles de la Ville de Paris) described both self-diffusion and gradient diffusion. Self-diffusion refers to the fluctuating motion of a test particle in the interior of a suspension where the particle concentration is uniform. Using a system with matched refractive indices, but with a test particle rendered opaque by a thin silver coating, her group observed that particle velocity fluctuations were large, on the same order as the mean particle velocity. As shown in figure 2, particles sometimes moved upwards, against gravity (Nicolai *et al.* 1995). Hydrodynamic self-diffusivities over a broad range of concentrations were determined to be approximately $D^s_{\parallel} = 8aU$ in the vertical direction and $D^s_{\perp} = 2aU$ in the horizontal direction, where a is the particle radius and U is the mean hindered settling velocity, indicating a strong anisotropy in the hydrodynamic diffusivity.

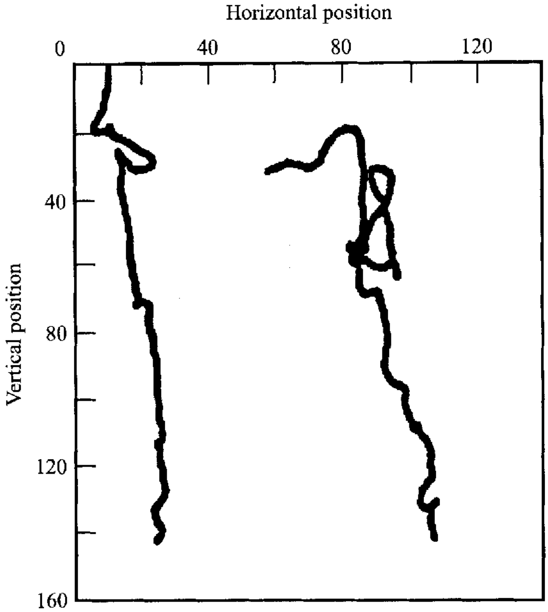

FIGURE 2. Trajectories of two test particles in the interior of a suspension subject to sedimentation; the positions are non-dimensionalized by the particle radius (adapted from Nicolai *et al.* 1995).

Above a particle concentration of 30% by volume, the values of the self-diffusivities decreased substantially. Guazzelli also reported that measured coefficients of self-diffusion did not vary significantly with container size (Nicolai & Guazzelli 1995). In contrast, John Hinch (Cambridge University) reported that computer simulations yield predicted diffusivities which do increase with increasing container size. The anisotropy in the self-diffusivity from the simulations and is similar to that observed in the experiments.

Anisotropy was also noted in experiments on a heavy sphere settling through lighter spheres, described by James Abbott (Los Alamos National Laboratory) and by Hélène Nicolai (Ecole Supérieure de Physique et Chemie Industrielles de la Ville de Paris). It was noted that the horizontal component of the hydrodynamic diffusivity for a heavy sphere falling through a dilute suspension of neutrally buoyant spheres is zero, since pairwise interactions give rise to no net horizontal displacements under creeping flow conditions (Davis & Hill 1992). A poster presentation by Shulin Zeng (University of Colorado) showed that microscopic surface roughness does give rise to net horizontal displacements, however, and consequentially a small ($D_\perp/D_\parallel = O(10^{-3}) - O(10^{-2})$) horizontal diffusivity is predicted for a heavy sphere falling through a dilute suspension of neutrally buoyant spheres (Davis 1992). In the experiments described by James Abbott, for which the lowest concentration of suspended particles was 15% by volume, the measured value of the anisotropy was $D_\perp/D_\parallel \approx 0.05$; the greater diffusivity in the horizontal direction than predicted by the dilute theory is likely to be due to multiparticle (as opposed to pairwise) hydrodynamic interactions.

Gradient diffusion refers to the drift of particles down a concentration gradient as a result of the fluctuating motion of the particles. A manifestation of gradient diffusion is the spreading of the interface at the top of a sedimenting monodisperse suspension. Guazzelli noted that Davis & Hassen (1988) measured this spreading and inferred maximum values of $D_\parallel^g \approx 8aU$ for the gradient diffusivity, but that corrected values of $D_{\parallel}^{g} \approx 20aU$ result when hindered settling (which reduces the spreading by a self-sharpening process) are accounted for (Lee *et al.* 1992; Martin, Rakotomalala & Salin 1994). Jerome Martin (Université de Paris Sud) reported that the opposing effects of self-sharpening and hydrodynamic diffusion lead to propagating concentration profiles of steady shape within a fluidized bed after a step increase or decrease in the fluidization velocity. From the shape of these profiles, Martin, Rakotomalala & Salin (1995) inferred that $D_{\parallel}^{g}/aU$ increases with concentration for dilute beds, reaches a value of approximately 60 over the range of 15–30% particles by volume, and then decreases at higher particle concentrations. The three-fold-higher gradient diffusivity in the fluidization case over the sedimentation case evoked some discussion, without a conclusive reason provided.

A lively discussion at the end of the session focused on the discrepancy between experiments and simulation on the dependence of the velocity variance and the hydrodynamic diffusivity on container size. It was concluded that, if experiments show no dependence on container size, then there must be some screening or cutoff length scale in the suspension microstructure which does not depend on the container size but does depend on the particle concentration. The origin and nature of such a microstructure is not certain, however.

2. Shear-induced diffusion and particle migration

John Brady (California Institute of Technology) gave an overview of hydrodynamic diffusion of non-Brownian particles in sheared suspensions. He also emphasized the difference between self-diffusion in a uniform suspension and gradient diffusion in the presence of a concentration gradient. The latter is responsible, for example, for the viscous resuspension of a settled layer of particles when subject to a shear flow (Leighton & Acrivos 1986). Ironically, the supposedly stochastic fluctuating motion of particles arises from deterministic hydrodynamic interactions between the particles. The reversibility of the creeping flow equations implies that the particles would retrace their paths upon reversal of the flow (i.e. a resuspended bed would resettle if the shear was reversed). However, even a slight amount of irreversibility (e.g. Brownian motion, particle surface roughness, particle deformability, or non-continuum effects) is enough to cause a memory loss after a particle undergoes many encounters.

The shear-induced diffusivity scales as $\dot{\gamma}a^2$, where $\dot{\gamma}$ is the local shear rate. Analytical expressions for both the self-diffusivity and gradient diffusivity were presented by David Leighton (University of Notre Dame) for dilute colloidal hard-sphere suspensions which do not interact hydrodynamically and for which Brownian motion is negligible. Francisco Da Cunha (University of Brasilia) presented results for the transverse shear-induced self-diffusivity and gradient diffusivity in the direction of the velocity gradient (i.e. across bulk streamlines) for simple shearing of a dilute suspension with pairwise hydrodynamic interactions. In order for pairwise interactions to give net displacements of particles across streamlines, a small amount of surface roughness allowing for contact was assumed, which results in a self-diffusivity of approximately $0.005\dot{\gamma}a^2\phi$ for roughness heights of 0.01a (Da Cunha & Hinch 1996); the gradient diffusivity is about five times larger. In contrast, net displacements do occur in the longitudinal direction (i.e. along bulk streamlines) due to pairwise interactions of smooth spheres, and a much larger longitudinal shear-induced self-diffusivity results because of the very large displacement of a test sphere caused by a second sphere which is far away but moving along a nearby streamline (Acrivos *et al.*

1992). A related poster on the longitudinal gradient diffusivity in a dilute suspension undergoing shear was presented by Yongguang Wang, Andreas Acrivos, and Roberto Mauri (City College of the City University of New York).

In a tag-team presentation, Ludwig Nitsche (University of Illinois) and Johannes Nitsche (State University of New York at Buffalo) noted that net lateral displacements could also occur for porous or deformable particles. A poster by Michael Loewenberg (Yale University) presented theoretical results for the transverse shear-induced self-diffusivities of dilute dispersions of deformable drops in the direction of the velocity gradient and in the direction of the vorticity; anisotropy was predicted, with the self-diffusivity in the direction of the velocity gradient being about five times larger than that in the vorticity direction.

3. Applications of hydrodynamic diffusion in suspension flows

The third session covered several applications of suspension flows in which hydrodynamic diffusion plays important roles. In the lead-off lecture, Andreas Acrivos (City College of the City University of New York) described how hydrodynamic diffusion or migration of particles from regions of high shear to regions of low shear is responsible for the previously unexplained observation by Karnis, Goldsmith & Mason (1966) of the blunting of the velocity profile for laminar flows of concentrated suspensions in channels or tubes. Recent experiments by Koh, Hookham & Leal (1994) have verified that the particle concentration becomes higher in the centre of a channel or tube, and reaches a steady profile in which hydrodynamic diffusion away from the walls due to the gradient in shear rate is balanced by hydrodynamic diffusion away from the centre due to the gradient in particle concentration.

Acrivos also noted that shear-induced hydrodynamic diffusion and particle migration are responsible for a variety of results in viscometric measurements (Leighton & Acrivos 1987*a*), viscous resuspension of settled particles (Leighton & Acrivos 1986; Schaflinger, Acrivos & Zhang 1990; Acrivos, Mauri & Fan, 1993), and sediment flow down an inclined plate (Nir & Acrivos 1990; Kapoor & Acrivos 1995). Applications of these concepts to oil recovery were described by Uwe Schaflinger (Technical University of Vienna) and by Iftikar Miskin (University of Leeds), and in a poster by Susan McCaffery (University of Leeds). A poster on the effects of aggregation, sedimentation, and shear-induced hydrodynamic diffusion on viscometry was presented by Wolter Wolthers and J. Mellema (University of Twente).

Andrea Chow (Lockheed Missiles and Space Company) described the results of tube-drawing experiments which show that particles in a suspension drawn into an empty tube undergo shear-induced migration toward the tube centre, and then move toward the advancing meniscus due to the higher velocities near the tube centre. For suspensions containing different particle sizes, the larger ones migrate faster than the smaller ones, yielding an enrichment of the larger particles in the meniscus region. Size segregation due to shear-induced hydrodynamic diffusion was also reported in a poster by Gokul Krishnan (Massachusetts Institute of Technology) and David Leighton (Notre Dame University).

Discussion at the end of this session included the need for constitutive models which include shear-induced hydrodynamic diffusion and the resulting inhomogeneous concentration profiles. Constitutive relations describing the shear-induced particle flux in unidirectional flows have been proposed by Leighton & Acrivos (1987*b*) and by Phillips *et al.* (1992), but it is unclear how well they may be adapted to describe the behaviour of more complex suspension flows.

4. Particle migration and segregation in granular flow

The symposium also included one session on granular flows, for which hydrodynamic forces are not important but particles undergo collision-induced fluctuating motions and net migrations which have similarities to hydrodynamic diffusion. Hans Buggisch (University of Karlsruhe) gave the opening talk and described how random particle motions superimposed on the bulk motion lead to net particle migrations and size segregation in both rapid granular flows (in which particles collide and bounce off each other with some similarities to gases) and slow granular flows (in which adjacent particles remain in prolonged contact). Descriptions of rapid granular flow by Campbell (1990) and by Jenkins & Savage (1983) were reviewed. Buggisch also reported on his own work on particle mixing (related to self-diffusion in monodisperse suspensions) and demixing (related to gradient diffusion and size segregation in polydisperse suspensions) for slow granular flows (Buggisch & Löffelmann 1989; Peciar, Buggisch & Renner 1993).

Melany Hunt (California Institute of Technology) then spoke on heat transfer applications in granular flows, such as heating and drying of particulates, photocopying, and gas-fluidized beds. Her focus was on the contribution of particle convection to heat transfer through particle collisions and mixing or diffusion. Expressions for the self-diffusion coefficient in rapid granular flows have been developed by Hsiau & Hunt (1993) and by Savage & Dai (1993) using dense-gas kinetic theories. Hunt also described how the effective thermal conductivity is related to the 'granular temperature' or kinetic energy of the particles.

Jim Jenkins (Cornell University) described a kinetic theory (Jenkins & Mancini 1989) for the separation of binary mixtures of smooth, inelastic spherical particles in rapid granular flows. Nuclear magnetic resonance imaging (NMRI) experiments to quantify size segregation in a rotating cylinder partially filled with a binary granular medium were described by Masami Nakagawa (Sandia National Laboratories). Since NMRI generally requires liquid present for sufficient signals, pharmaceutical pills containing vitamin oil were used in these studies (Nakagawa 1994).

5. Computer simulation techniques

A talk and companion poster by Yidan Lan and Tony Rosato (New Jersey Institute of Technology) touched on the widespread use of computer simulation techniques for describing granular flows. Although developed separately, computer simulations for suspension flows have similar features to those for granular flows in terms of tracking the positions and velocities of large numbers of particles with fluctuating motions due to particle–particle interactions.

Tony Ladd (Lawrence Livermore National Laboratory) gave an invited overview of simulation techniques applied to suspensions. He noted that the Stokesian dynamics techniques based upon multipole moments and pioneered by Brady & Bossis (1988) are computationally intensive and limited to about 10^2 particles. Grouping together particles which are far away from a given sphere provides a significant reduction in computer requirements (Sangani & Mo 1995). Ladd then described a lattice-Boltzmann simulation technique which uses time-dependent hydrodynamics (Ladd 1993, 1994*a,b*). Citing results of this technique for up to 8000 spheres in a periodic box, Ladd returned to an earlier theme of the symposium and discussed the hydrodynamic self-diffusivity during sedimentation. He reported good agreement with the experiments of Nicolai *et al.* (1995) for the particle velocity fluctuations, but the predicted anisotropy ratio $D^s_{\parallel}/D^s_{\perp} \approx 20$ is several-fold higher than that observed experimentally. In addition, the simulations did not provide evidence of a screening microstructure, such as described by Koch & Shaqfeh (1991), and the simulation results for the velocity variance and hydrodynamic diffusivity appear to diverge as the size of the periodic box is increased.

Ashok Sangani (Syracuse University) described further simulations of sedimentation for which particle inertia is important (finite Stokes number) but fluid inertia is still negligible (low Reynolds number). Such a formulation is appropriate for particles with diameters of approximately 0.1 mm sedimenting in a gas. Hydrodynamic interactions were included, but lubrication forces were assumed to break down when particle surfaces approached within a distance comparable to the mean free path of the gas molecules, thus allowing collisions to occur between particles. A suspension temperature (kinetic energy) formulation reminiscent of granular flow simulations was used. The simulations again predict that the hydrodynamic diffusivity increases with the size of the periodic box employed, but the particle velocity variance is independent of the box size and decreases with increasing particle concentration and Stokes number. The results are in good agreement with the asymptotic theory of Koch (1990, 1992) for large Stokes numbers, corrected to take into account the effects of finite particle volume fractions.

The session concluded with a talk by Gerald Ristow (Philipps-Universität) which led to some discussion of the elastohydrodynamic theory of collisions of particles in a fluid (Davis, Serayssol & Hinch 1986). Viscous dissipation during such collisions might reduce the coefficient of restitution and hence the behaviour of, for example, wet granular materials. A related poster on computer simulations was presented by Wolfgang Kalthoff (Ecole Supérieure de Physique et Chemie Industrielles de la Ville de Paris).

6. Orientational and configurational dispersion

Although it is now well-known that hydrodynamic and contact interactions between particles cause diffusion and migration of the centres of mass of particles, there is much less appreciation of the effects of such interactions on the configuration or orientation distribution of complex bodies such as non-spherical particles, deformable drops, and macromolecules. In an invited lecture, Eric Shaqfeh (Stanford University) reviewed the work that he, Don Koch, and their collaborators have undertaken in this area. Shaqfeh started by describing the flow of complex fluids (i.e. ones containing fibres, polymer molecules or deformable drops) in dilute fixed beds, which have the simplifying features of the configuration of bed particles being fixed and of the Brinkman screening in the bed acting to cut off the long-range hydrodynamic interactions which would otherwise give indeterminate results for the long-term orientation distributions. In all cases, highly anisotropic (i.e. aligned particles) steady states were predicted and observed (Shaqfeh & Koch 1988, 1992; Frattini *et al.* 1991; Evans, Shaqfeh & Frattini 1994). He then discussed the effects of particle interactions on the orientational distribution of fibres in extensional and shear flows. For extensional flows, interactions lead to a dispersion of the fibre orientations about the mean alignment with the principal axis of extension (Shaqfeh & Koch 1990). For shear flows, non-interacting elongated particles remain in their original distribution of Jeffery orbits, whereas particle–particle interactions lead to a steady orientation distribution that is independent of the initial distribution (Rahnama, Koch & Shaqfeh 1995). Shaqfeh concluded with a discussion of the sedimentation of fibres. He showed by simulations that the fibres tend to align with gravity due to particle–particle interactions, and that such systems are unstable and exhibit mean sedimentation velocities which increase with time due to clumping of the particles.

The session also included talks by Oliver Harlen (University of Leeds) on the effects of polymer solutions on the alignment and orientational diffusion of suspended fibres (Harlen & Koch 1992, 1993), and by R. Sundararajakumar (Cornell University) on the effects of mechanical contacts between particles on the orientational dispersion of fibres and on the diffusion of a ball settling through a suspension of neutrally buoyant fibres. Since the rheological properties of complex suspensions are strongly affected by even small changes in the orientational distribution of the inclusions, the conclusion of the session was that this is an important area for further research.

7. Experimental techniques

The symposium concluded with a session on experimental techniques for observing and quantifying particle migration and diffusion in suspensions and granular media. Dominique Salin (Université de Paris Sud) gave an overview of this subject. He noted that tracking single particles may be accomplished with video or X-ray cameras. The fluid and particles have to be transparent to the corresponding rays, and so such techniques are often restricted to very dilute systems or to materials for which the refractive indices of the fluids and particles are matched (Nicolai *et al.* 1995; Mondy, Graham & Jensen 1986). Fluorescent and radioactive tracer methods may also be used. Salin also discussed another class of techniques, which uses light, X-ray or acoustic attenuation through suspensions to measure particle concentrations (Davis & Hassen 1988; Martin *et al.* 1995).

Lisa Mondy (Sandia National Laboratories) then described the use of nuclear magnetic resonance (NMR) imaging, which has the advantage of being able to measure both velocity and concentration profiles. NMR requires expensive and sophisticated equipment, and so its use in studying suspension and granular flows has been limited to relatively few laboratories. Several talks and posters, based on work using the NMR equipment at the Lovelace Institutes in New Mexico, were presented during the symposium by a group of collaborators which includes Masami Nakagawa and Lisa Mondy of Sandia National Laboratories, Steve Altobelli and Eiichi Fukushima of the Lovelace Institutes, and Alan Graham and James Abbott of Los Alamos National Laboratory. Presentations based on NMR experiments were also made by Andrea Chow (Lockheed Missiles and Space Company) and Robert Powell (University of California at Davis). The experiments give clear evidence of particle migration and size segregation, non-uniform concentration profiles, and deviations from Newtonian velocity profiles, which result from hydrodynamic diffusion in suspension flows (Graham *et al.* 1991; Abbott *et al.* 1991; Phillips *et al.* 1992). As noted previously, liquid-state NMR techniques have also been adapted to measure velocity profiles, concentration profiles, and particle diffusion in granular media, by using pharmaceutical pills or plant seeds which contain encapsulated liquid (Nakagawa *et al.* 1993; Nakagawa 1994).

Mike Lyon (University of California at Santa Barbara) and Avi Nir and Anat Shauly (Technion) each reported on the recent development of laser-Doppler anemometry (LDA) or velocimetry (LDV) techniques for measuring particle velocity and concentration profiles in concentrated suspensions for which the refractive indices of the particles and fluids are matched. They were able to measure the migration of particles toward the centre line in pressure-driven flow in a rectangular duct, and to quantify the blunting of the velocity profiles due to this shear-induced hydrodynamic diffusion. The measurements were compared with qualified success to predictions from the diffusion-based and statistical mechanical constitutive models of Phillips *et al.* (1992) and Nott & Brady (1994), respectively.

8. Concluding remarks

The symposium on hydrodynamic diffusion was timely. Various phenomena related to hydrodynamic diffusion have been studied for a time period (just under 10 years) which is long enough for many interesting results to have been presented but short enough that there are many features which are not yet understood and evoked much discussion and speculation. Certainly, a predictive understanding of hydrodynamic diffusion during sedimentation is still needed, as are constitutive models which account for hydrodynamic diffusion and can be applied to complicated flows. Orientational and configurational diffusion in complex fluids is a fruitful area for additional research, and improvements in simulation and experimental techniques will provide important tools. Finally, continued exchange between the suspension and granular flow communities will help advance the subject.

I give special thanks to Professors Andreas Acrivos and George Batchelor for initiating interest in the symposium. Appreciation is also extended to the sponsors of the symposium.

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