MACROSTATISTICAL HYDRODYNAMICS

Progress Report

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MACROSTATISTICAL HYDRODYNAMICS

Summary

Experimental, analytical, and numerical studies of falling-ball 'tracer' particle dynamics in concentrated suspensions composed of dispersed, neutrally-buoyant spheres and rods comparable in size to the tracer have been continued and extended.

Fluctuations in the mean sedimentation velocity of a falling ball through such suspensions were analyzed and successfully interpreted in terms of a "hydrodynamic diffusion" model. Diffusivity results agreed well with a theoretical model of the phenomenon, originally developed for dilute systems.

Spinning-ball rheometry in quiescent suspensions is being developed as a useful adjunct to prior falling-ball rheometric studies. This technique provides another experimental benchmark for numerical and analytical studies of microstructural effects in concentrated suspensions.

The pressure 'drop' incurred by a sphere falling through a quiescent neutrally-buoyant suspension, which constitutes another useful dynamical suspension parameter, was measured and found to agree well with theoretical results for the comparable pressure drop in a homogeneous Newtonian liquid.

In collaboration with Lovelace, NMR imaging was used to study the flow-induced migration of neutrally-buoyant suspensions of rods (of aspect ratio up to 20) dispersed in a Newtonian liquid undergoing inhomogeneous shear. Despite their very different geometry, the rod results were similar to those previously observed for spheres.

MACROSTATISTICAL HYDRODYNAMICS

Progress Report

Progress to Date

This work has been supported by the U.S. Department of Energy, Division of Engineering and Geosciences, Office of Basic Energy Sciences. During this time we studied suspensions of particles in Newtonian (and some non-Newtonian) liquids, embodying a combination of analysis, experiment, and numerical simulation. Portions of our accomplishments that directly relate to the proposed work are discussed below.

Falling Ball Rheometry

Experiments primarily involved tracking small balls falling slowly through otherwise quiescent suspensions of neutrally buoyant particles. A theory was developed relating the average terminal velocity of the ball to the macroscopic suspension viscosity (Brenner, Graham, Abbott, and Mondy, 1990). Detailed trajectories of the balls, obtained both with numerical (Mondy, Ingber, and Dingman, 1991) and new experimental techniques (Abbott, 1993), were statistically interpreted in terms of the mean settling velocity and the dispersion about that mean.

We showed that such falling-ball rheometry, using small balls relative to the suspended particles, can be a means of measuring the macroscopic zero-shear-rate viscosity without disturbing the

original microstructure significantly; therefore, falling ball rheometry can be a powerful tool to study the effects of microstructure on the macroscopic properties of suspensions. Conventional rotational viscometers, on the other hand, may induce changes in the microstructure over time; as such, the data from them may not provide accurate measures of the viscosity of the suspension originally introduced into the viscometer. For example, flow can effect both the distribution and orientation of rodlike particles in suspension. However, falling-ball measurements can be performed on suspensions of *randomly oriented* rods (Milliken, Gottlieb, Graham, Mondy, and Powell, 1989), with results agreeing with dilute theory (Brenner, 1974; Haber and Brenner, 1984) over a surprisingly large range of solids concentration. Furthermore, a specific orientation of rods can also be obtained by hydrodynamically aligning the particles (Graham, Mondy, and Guell, 1994) prior to the falling-ball experiment. Such a suspension was tested using falling ball rheometry (Mondy, Morrison, Graham, and Powell, 1990); results demonstrated the ability of the technique to determine a viscosity dependent on the measurement direction for anisotropic suspensions.

In addition to the mean settling velocity, we also measured the fluctuations in the sedimentation velocity as the falling ball interacts with individual suspended particles or clusters of suspended particles. Whereas the mean settling velocity predicts the continuum behavior of the suspension, the dispersion around that mean allows insight into the non-continuum behavior of the suspension caused by the presence of the suspended particles. We performed experiments which focused on the three-dimensional dispersion of a single ball settling through a suspension of neutrally buoyant particles. The detailed (three-dimensional) paths of falling balls were recorded from direct observations in transparent suspensions (in which the refractive index of the suspending fluid was matched to that of the suspended particles) and using real-time radiography in opaque suspensions. The primary experimental parameters were the relative size of the settling ball and suspended particles, and the concentration and shape of the suspended particles.

For sufficiently large time intervals between successive observations, the settling paths of the balls were observed to disperse about their mean settling path in a random manner. Therefore, a hydrodynamic dispersivity exists which characterizes the linear growth with time of the variances in the position of a falling ball. However, for short time intervals, the vertical variances were dominated by deterministic behavior, which resulted in a quadratic growth of the variances. The short-time behavior of a ball falling in a suspension is not caused by the inertia of the ball, but rather by the time needed for the ball to change its local environment; in other words, it is caused by the local configuration of the suspended particles. To determine the dispersivity when the variances were deterministic, we estimated the time scale on which it takes the settling ball to change its local neighborhood and then used this time scale in a model, similar to that for Brownian tracers, relating the measured short-time variances to the dispersivity

In separate experiments we are currently measuring the (very small) pressure drop caused by a ball falling through suspensions of similar-sized particles (Graham, Tetlow, Abbott, Mondy, and Brenner, 1993). Like the falling-ball viscosity measurements, the pressure drop behavior in moderately concentrated suspensions is similar to that in a Newtonian single phase liquid, being independent of the relative sizes of the falling ball and suspended particles over a wide range of sizes. Using boundary element modeling, we had earlier predicted this to occur in dilute suspensions (Mondy, Ingber, and Dingman, 1991). At higher concentrations of solids, the pressure traces are more erratic with the maximum variation being observed for the very small falling balls. The average pressure drop is also slightly less than expected for a single phase liquid.

Although the falling ball is originally released along the axis of the cylinder, the ball spends a sig-

nificant amount of time away from the axis because of interactions with suspended particles (that lead to the dispersivities measured earlier). The wall effects, which influence the velocity of the ball, depend on the radial location of the ball. We have modeled the pressure drop in a suspension by combining measurements of the dispersivity of the falling ball with a randomized computational algorithm. The algorithm was based on the assumption that the ball settles with both horizontal and vertical displacements chosen from a Gaussian distribution weighted with experimentally measured dispersivities. Knowing the predicted instantaneous displacement, we calculated the pressure drop from the Newtonian fluid formula provided by Feldman and Brenner (1963), which includes the effects of the wall when the ball is displaced from the cylinder axis. The model predicts both the erratic nature of the pressure measurements and the lowering of the average pressure drop (due to wall effects).

Particle Migration in Flowing Suspensions

Recall that the dispersivity is due to interactions between the ball and a suspended particle or cluster of particles. The same physics is likely to govern the so-called "hydrodynamic diffusion" which occurs in concentrated suspensions undergoing flow, and which can result in overall particle migration (Leighton and Acrivos, 1987). This migration is thought to occur whenever particle interactions are stronger or 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. In collaboration with Lovelace Institutes (Albuquerque, NM), we used NMR imaging to study flow-induced migration of neutrally buoyant, monomodal, spherical particles suspended in Newtonian oils and undergoing flow in the annular space between rotating concentric cylinders (Abbott, Tetlow, Graham, Altobelli, Fukushima, Mondy, and Stephens, 1991). (The technique of NMR imaging of concentrated suspensions was developed in an independent grant to Lovelace by the Division of Engineering and Geosciences, Office of Basic Energy Sciences, Department of Energy.) The rotating concentric cylinders create perhaps the simplest flow field of any realizable nonhomogeneous shear flow. As such, this is a useful geometry in which to study the effects of nonhomogeneous shear on the microstructure of a concentrated suspension.

The primary data obtained from these experiments are NMR images of the concentration and velocity fields. From the comparison of initial concentration images to images taken after rotation of the inner cylinder, it is apparent that the particles migrate from the region of highest shear to the region of lowest. Furthermore, when suspensions of large (compared to the resolution of the images) spheres are studied, distinct shells of densely packed spheres can be seen after rotation occurs. This structure begins to appear very quickly: Significant migration can be detected after only 50 revolutions of the inner cylinder. It is important to note that this migration does not appear to be caused by inertial effects, which are negligible under conditions encountered in the experiments, namely the rates of rotation, the viscosity of the suspending liquid, and the particle sizes involved.

In addition to expanding our general understanding of the micromechanics of shear-induced migration, the primary purpose of the NMR imaging studies was to determine the dependence of the particle migration on a number of experimental parameters. These parameters included strain, shear rate, and viscosity of the suspending fluid, as well as concentration and diameter of the suspended particles. We found that the migration depends on the total strain and is independent of shear rate, as well as the viscosity of the (Newtonian) suspending liquid. The migration rate is sensitive to the particle size, increasing with approximately the square of the particle radius. The results of a constitutive model (Phillips, Armstrong, Brown, Graham, and Abbott, 1991) based on

Leighton and Acrivos' (1987) scaling arguments compared very favorably to these experimental results. In this model, a diffusion equation accounts for shear-induced particle migration through particle flux expressions derived by scaling arguments. A generalized Newtonian constitutive equations allows the viscosity of the suspension to increase with increasing volume fraction of particles. The good agreement with data verifies the scaling analysis and the basic mechanics incorporated into the constitutive description.

We also studied flow-induced migration of nonspherical particles (Mondy, Brenner, Altobelli, Graham, and Abbott, 1994). Somewhat surprisingly, there was no effect of particle shape on the steady-state concentration profiles produced in the flow between concentric rotating cylinders. Fibers of aspect ratio up to 20 migrated to produce the same final concentration as did spheres.

Spinning ball rheometry

We have recently developed a spinning ball rheometer with which to study slip/stick phenomena in suspensions. In the falling ball experiments described earlier, 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 can instead measure the mean torque on a spinning ball. This scheme is more sensitive to slip at the ball boundary: the torque on a ball with perfect slip at the boundaries is zero! Again, we use balls comparable in size with the suspended particles.

We have developed equipment that allows the measurement of small angles of deflection of a thin, carefully calibrated torque wire suspending a ball (see Figure 1). The ball is placed in a rotating cylinder holding otherwise quiescent liquid. A mirror that is mounted between the torque wire and the suspended ball reflects a laser beam to a detector, which measures the angle of deflection between the initial beam position and the position when the cylinder rotates at a constant rate. This equipment, a spinning ball rheometer, was tested in a single-phase Newtonian liquid of known viscosity. The viscosity measured with the spinning ball rheometer was within one to three percent (depending on the ball size and rotation rate) of that measured with a conventional capillary viscometer.

Preliminary data on a moderately concentrated suspension (20% by volume of solids) has been taken. This suspension was created from uniform 1/4-inch-diameter spheres neutrally buoyant in a viscous Newtonian oil. The measured angle, like the measured terminal velocity of a falling ball, depends on the configuration of the suspended particles; therefore, several measurements must be repeated to obtain a mean apparent viscosity. We also note, that like the falling ball rheometry measurements, the spinning ball measurements include a meaningful fluctuation about the mean. Further experiments, in which the spinning ball size and the suspended particle concentration are varied, are expected to be completed this year.

One must note that this flow geometry has the potential to induce changes in the suspension microstructure over time. This results in a measurable decrease of the torque as the particles migrate away from the spinning ball. The instrumentation allows us to record a time history of the torque (the angle of deflection) as a function of the number of rotations of the platter holding the suspension-filled cylinder. The long-time measurements will complement the dispersion measurements taken with NMR imaging.

Boundary Element Modeling

We have used boundary element modeling to simulate the hydrodynamic interaction among suspended particles in falling ball experiments (Mondy, Ingber, and Dingman, 1991; Tullock, Phan-

Figure 1. Equipment to measure the angle of deflection of a torque wire connected to a ball suspended in a spinning, but otherwise quiescent, suspension of like spheres.

Thien, and Graham, 1992) as well as in more concentrated suspensions of particles arranged in periodic arrays (Phan-Thien, Tran-Cong, and Graham, 1991). These boundary element methods couple the Stokes equations for the fluid with the equilibrium equations for individual particles. These numerical studies have provided further insight into the connection between the micromechanics of suspensions and their bulk properties. For example, results of studies in which simple cubic arrays of spheres, spheroids, cubes, and clusters of spheres were subjected to a bulk simple shearing flow showed that the volume-averaged properties of these suspensions displayed only a weak dependence on the particle shape, despite large differences in the local micromechanics of stress and velocity fields. These studies (Phan-Thien, Tran-Cong, and Graham, 1991) show that valuable information about the relationship between the microstructure and the macroscopic properties can be acquired through numerical (or analytical) modeling of periodic systems.

In another example, by calculating the resistance to the fall of a heavy ball, as neutrally buoyant particles were added one at a time, we demonstrated that only the suspended particles in the "near field" significantly affected the heavy ball's velocity. Many studies are expected to be much easier in this "numerical rheometer" than in the laboratory. In addition, dynamic simulations may lead to more understanding of the relationship among fluctuations in the falling ball's velocity and the microstructure of the suspension. Such dynamic simulations have been shown to be possible (Mondy, Ingber, and Dingman, 1991; Dingman, Ingber, Mondy, Abbott, and Brenner, 1992), and the results to be accurate even when the particles become very close (Ingber and Mondy, 1993);

however, only preliminary testing has been done to date because of the large computational resources (time and memory) required for dynamic simulations. One promising avenue to enhancing the numerical modeling capabilities and greatly increasing the size of tractable problems is to implement the method on a computer with massively parallel architecture. Preliminary progress has been made in this endeavor (Ingber, Womble, and Mondy, 1994).

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Appendix A

PUBLICATIONS ACKNOWLEDGING DOE SUPPORT

(April 15, 1993 - April 14, 1994)

Howard Brenner

Journal Articles Published

Lue, A., and Brenner, H., "Phase flow and statistical structure of Galton board systems," *Phys. Rev. E.*, **47**, 3128-3144 (1993).

Batycky, R.P., and Brenner, H., "Fictitious initial conditions for effective medium descriptions of transient, nonconservative systems," *AMD (Applied Mechanics Division of the ASME)* **160**, 15-21 (1993).

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Thesis

Bryden, Michelle D., "Macrotransport Processes in Branching Networks: Modeling Convective-Diffusive Phenomena in the Lung." M.S. Thesis, Massachusetts Institute Of Technology (1994).

Appendix B

SEMINARS AND TECHNICAL PRESENTATIONS

(April 15, 1993 - April 14, 1994)

Presentations by H. Brenner *et al.*1993

with A.L. Graham and L.A. Mondy), Eleventh Symposium on Energy Engineering Sciences, DOE, Argonne National Laboratory, Argonne, Illinois (May).

Los Alamos National Laboratory, Macrostatistical Hydrodynamics Group, Los Alamos, New Mexico (May).

(with R.P. Batycky), Joint ASCE-ASME-ASCE Summer Applied Mechanics Meeting, Symposium on Recent Advances in Structured Continua, University of Virginia, Charlottesville, Virginia (June).

National Institute of Science and Technology (NIST), Polymer and Biotechnology Divisions, Gaithersburg, Maryland (August).

(with A.L. Graham, N. Tetlow, J.R. Abbott and L.A. Mondy), International Symposium on Advances in Structured and Heterogeneous Continua, Institute of Applied Mechanics, Russian Academy of Sciences, Moscow, Russia (August).

(with L.A. Mondy and A.L. Graham), 65th Annual Meeting, Society of Rheology, Boston, Massachusetts (October).

(with J. Camacho), 1993 AIChE Annual Meeting, St. Louis, Missouri (November).

(with A. Lue), 1993 AIChE Annual Meeting, St. Louis, Missouri (November).

¹American Physical Society, Fluid Dynamics Division, Annual Meeting, Albuquerque, New Mexico (November).

1994

University of Alabama, Department of Mathematics, Tuscaloosa, Alabama (January).

Brown University, Division of Biology and Medicine, Graduate Program in Artificial Organs, Biomaterials and Cellular Technology, Providence, Rhode Island (March).

McGill University, Department of Civil Engineering and Applied Mechanics, Series of "Seminars in Mechanics," Montreal, Canada (March).

¹Invited Lecturer

Appendix C

SIGNIFICANT PROFESSIONAL ACTIVITIES

by H. Brenner

(April 15, 1993 - April 14, 1994)

Consulting Editor, Chairman of the Advisory Board, The Butterworth-Heinemann Series on Chemical Engineering.

Member, Scientific Committee, IUTAM Symposium on "Liquid-Particle Interaction in Suspension Flow," Grenoble, France (April, 1994).

Member, Scientific Committee, International Symposium on *Advances in Structured and Heterogeneous Continua*, Moscow, Russia. (August, 1993).

Chairman of the session on "*Suspensions II*," 1993 Annual Meeting, Fluid Dynamics Division, American Physical Society, Albuquerque, New Mexico (November, 1993).

Member, International Advisory Committee, The Caribbean Congress of Fluid Dynamics.

Member, Selection Committee, Society for Natural Philosophy.

Member, Interfacial Phenomena Committee (Area 1C), National Program Committee, American Institute of Chemical Engineers.

Editorial Advisory Board, *International Journal of Multiphase Flow* (Pergamon).

Member, Awards Committee, American Chemical Society.

Member, Kluwer book series advisory board on *Theory and Applications of Transport in Porous Media*.

Member, Scientific Committee, *International Symposium on Advances in Structured Continua II*, Moscow, Russia (August, 1995).