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Title: A Fundamental Study of Dispersed Multiphase Flows at Small Scales
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Final Report

During the two-year period covered by this report, our research on Dispersed Multiphase Flows at Small Scales focused on two broad areas: I) The Rheology of Suspensions of non-Colloidal Particles; and, II) The Transport of a Nano-Sized Particle through a Fluid-Filled Nano-Channel. These two parts of our program will be considered separately.

I. Here we extended our earlier studies in the following two directions: First, by means of Stokesian Dynamics simulations, we investigated in considerable detail the dynamics of concentrated suspensions and discovered, much to our surprise, that their statistical properties could be described, to a surprising degree of accuracy even for particle concentrations up to 20%, by means of an analytical theory of pair –particle interactions, provided that pair-doublets were assumed to be totally absent. This result has important implications concerning the modeling of such suspensions. In parallel, we also investigated further the fascinating phenomenon of particle axial segregation which was observed when a suspension (and a very dilute one at that) containing neutrally buoyant particles was sheared in a partially filled horizontal Couette device or a rotating horizontal circular cylinder. This mystery was eventually solved my means of a combined experimental, analytical & numerical study, where it was shown that this banding phenomenon results from a two-step instability in which the particles first segregate radially and then form bands in the axial direction due to an axial particle concentration perturbation that induces an axial variation in the effective viscosity of the suspension..

II. Here we used molecular dynamics simulations to study the behavior of closely fitting spherical and ellipsoidal particles moving through a fluid-filled cylinder at nanometer scales. The particle, the cylinder wall, and the fluid solvent were all treated as atomic systems, and special attention was given to the effects of varying the wetting properties of the fluid. Although the modification of the solid-fluid interaction led to significant changes in the microstructure of the fluid, its transport properties were found to be the same as in bulk. Independently of the shape and the relative size of the particle, we found two distinct regimes as a function of the degree of wetting, with a sharp transition between them. In the case of a highly wetting suspending fluid, the particle moved through the cylinder with an average axial velocity in agreement with that obtained from the solution of the continuum Stokes equations. In contrast, in the case of less-wetting fluids, only the early time motion of the particle was found to be consistent with continuum dynamics. At later times, the particle was eventually adsorbed onto the wall and subsequently executed an intermittent stick-slip motion. The force on the particle and the system's Helmholtz free energy were found to depend on the particle's history as well as on its radial position and the wetting properties of the fluid, even when the particle's motion occurs on time scales much longer than the spontaneous adsorption time. The hysteresis is associated with changes in the fluid density in the gap between the particle and the wall, and these structural rearrangements persist over surprisingly long times. The force and free energy exhibit large oscillations with distance when the lattice of the structured nanoparticle is held in register with that of the tube wall, but not if the particle is allowed to rotate freely. Adsorbed particles are trapped in free energy minima in equilibrium, but if the particle is forced along the channel the resulting stick-slip motion alters the fluid structure and allows the particle to desorb. Our studies, which are of particular relevance to the field of micro- and nano-fluidics, underline some of the issues arising when particle dynamics at the nanoscale are examined in quantitative detail.

These and many other results are further described in the attached list of publications which acknowledge support of the DOE Grant DE-FG02-03ER46068.

List of publications acknowledging the support of DE-FG02-03ER46068.

1. Jin, B. and Acrivos, A. "Rimming flows with an axially varying viscosity", *Phys. Fluids*, **16**, 633, (2004).
2. Jin. B. and Acrivos, A. "Theory of particle segregation in rimming flows of suspensions containing neutrally buoyant particles", *Phys. Fluids* **16**, 641, (2004).
3. Drazer, G., Koplik, J., Khusid, B., and Acrivos, A., Microstructure and velocity fluctuations in sheared suspensions ", J. Fluid. Mech. **511**, 237 (2004).
4. Acrivos, A. and Bo, J., "Rimming flows within a rotating horizontal cylinder: asymptotic analysis of the thin-film lubrication equations and stability of their solutions", *J. Engineering Math*. **50**, 99 (2004).
5. Drazer, G., Khusid, B., Koplik, J., Acrivos, A.," Wetting and particle adsorption in nanoflows", *Phys. Fluids,* **17**, 017102-1-18 (2005).
6. Drazer, G., Khusid, B., Koplik, J., Acrivos, A., "Hysteresis, force oscillations and non- equilibrium effects in the adhesion of spherical nanoparticles to atomically smooth surfaces", submitted to *Physical Review Letters*, Submitted, January, 2005.
7. Jin, B., and Acrivos, A., "The drag-out problem in rimming flows", *Phys. Fluids,* Under review.