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Shear-Induced Structure of Concentrated Suspensions of Non-Brownian Particles in a Microgravity Environment

We report measurements of the steady-state shear viscosity of non-Brownian particulate suspensions in which the particles were meticulously density matched to the surrounding liquid to simulate a microgravity environment. Four distinct types of behavior were found for the viscosity of the suspensions as the shear rate and concentration were varied. Possible interpretations of the data in term of the shear-induced structure and dynamics of the suspensions are discussed. In addition, the shear rate dependence of the average particle velocity and its variance has been measured using a new technique in ultrasonic correlation spectroscopy called Dynamic Sound Scattering, demonstrating the potential of this technique for investigating the dynamics of sheared particulate suspensions.

INTRODUCTION

The properties of non-Brownian particles in a liquid are usually dominated by gravitational forces that cause the particles to sediment to the bottom of the container. In microgravity, however, sedimentation is greatly reduced, allowing stable suspensions to be formed, and providing a unique opportunity to investigate the many body hydrodynamic interactions that determine their basic properties. One of the most important properties of suspensions is their viscosity, and there has been a growing interest in the effects on viscosity of the microstructure, hydrodynamic interactions and concentration distribution of the particles since the pioneering work Gadala-Maria and Acrivos [1]. For concentrated suspensions of non-Brownian, neutrally buoyant solid particles they first reported that the apparent shear viscosity in a Couette viscometer decreased with time during prolonged shear. Leighton and Acrivos [2] attributed this slow viscosity decrease to shear-induced particle migration to low shear stress regions. The introduction of a NMR imaging technique [3] and a novel correlation method [4] has shed light on the evolution of suspension concentration profiles and shear-induced self-diffusion in Couette flow upon shearing. Here we present new data on the viscosity and dynamics of suspensions of non-colloidal spherical particles in a low viscosity density-matched liquid. Our goals are to gain new insights into the behaviour of the viscosity and the motion of the particles in suspensions under the influence of shear - phenomena that need to be examined in detail if the flow of suspensions in microgravity is to be understood, and possible applications in space technology developed and optimized.

MATERIALS AND EXPERIMENTAL METHODS

To create neutrally buoyant suspensions, uniform borosilicate glass beads (diameter a = 63 ± 11 µm, ρ = 2,220 kg/m^3) were immersed in a density-matched liquid consisting of a mixture of LST heavy liquid (a low viscosity aqueous solution of lithium heteropolytungstates) and water. The beads were sieved more than once to limit the particle size distribution and centrifuged to minimize the density variance. Before carrying out the viscosity measurements, the stability of the suspensions was monitored using a combination of video microscopy and Dynamic Sound Scattering (DSS) techniques [5]. From the particle velocities measured in these experiments, we were able to demonstrate that the density differences between the particles and the liquid were less than one part in 10^5, corresponding to buoyant forces on the particles that could only be achieved in 10 µg or less for suspensions of the same particles in water.

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The viscosity measurements were carried out for volume fractions ϕ of 0, 10, 20, 30, 40, 50, 55, and 60 % at a temperature of 27°C using a TA instruments AR2000 rotational viscometer equipped with a concentric cylindrical cell. The outer cylinder was fixed in position (stator), with the inner cylinder being the rotor, which had a conical bottom to facilitate loading. The inner and outer radii of the cylindrical cell were 14 and 15 mm respectively, corresponding to a gap width of 1 mm (approximately 10 particle diameters). While for such a relatively narrow gap, wall effects cannot be ruled out, previous experiments suggest that such effects are unlikely to be very significant [4]. The viscosity of the suspensions was investigated under steady shear flow at constant angular velocity from 0.1 to 20 rad/s (shear rate $\dot{\gamma}$ from 1.45 to 300 s^{-1}); the measurements were performed for both increasing and decreasing angular velocity, in a series of steps or cycles that allowed the effects of the evolution of the suspension microstructure on viscosity to be investigated. Initial measurements were performed in the standard mode of operation whereby the viscosity was recorded after monitoring it every 10 s until the variation in the last 3 measurements was less than 5%, or the maximum sampling time (set to 1 min) was reached. To further examine the time evolution of the viscosity, measurements were also performed at a constant shear rate for time intervals up to 30 minutes, allowing particle migration and microstructure evolution in the suspension to be investigated more fully. To reduce possible particle migration into the low shear region at the bottom of the cylindrical cell, the volume of this region was minimized by lowering the bottom of the rotor as close to the stator as possible. The volume of the gap was 3.8 ml and the volume in the bottom reservoir was 4.7 ml.

The particle dynamics of the suspensions under shear were investigated directly using Dynamic Sound Scattering [5], which is the acoustic analogue of Dynamic Light Scattering. The basic idea is to relate the fluctuations of the singly scattered ultrasonic waves to the motion of the particles via the temporal field autocorrelation function. In DSS, the field autocorrelation function can be measured directly due to the ability of ultrasonic detectors to measure the acoustic pressure rather than the intensity. As discussed by Cowan et al. [5], the field correlation function can be written

$$g_1(\tau)=\left\langle \exp\left[-i\Delta\phi_p(\tau)\right]\right\rangle=\left\langle \exp\left[-i\vec{q}\cdot\Delta\vec{r}_p(\tau)\right]\right\rangle$$

where $\Delta\phi_p(\tau)$ is the phase change of the scattered waves due to the p^{th} particle's motion, $\vec{q}=\vec{k}'-\vec{k}=2k\sin(\theta/2)$ is the scattering wave vector, θ is the scattering angle, $\vec{r}_p(\tau)$ is the change in position of the p^{th} particle during the time interval τ, and $\langle..\rangle$ denotes time average. Since $\Delta\phi_p(\tau)=\langle\Delta\phi_p(\tau)\rangle+\delta\phi_p(\tau)$, the correlation function for ballistic particle motion, where $\Delta\vec{r}_p(\tau)=\vec{V}_p\tau$, is

$$g_1(\tau)\simeq\cos\left(2k\sin(\theta/2)\langle V_{\vec{q}}\rangle\tau\right) \times\exp\left[-2k^2\sin^2(\theta/2)\langle\delta V_{\vec{q}}^2\rangle\tau^2\right] \quad (2)$$

Equation (2) shows that both the average particle velocity along $\vec{q}$, $\langle V_{\vec{q}}\rangle$ and the variance, $\langle\delta V^2\rangle=\langle V_{\vec{q}}^2\rangle-\langle V_{\vec{q}}\rangle^2$ can be measured by this technique. By orienting the generating and detecting transducers in different directions relative to the sample cell, all three components of the average particle velocity and the variance can be measured.

RESULTS AND DISCUSION

Figures 1 and 2 illustrate general trends of the viscosity during Couette flow for the suspensions at different concentrations. These measurements have enabled us to identify four different regimes for the behavior of the viscosity of non-Brownian suspensions as the shear rate and concentration are varied:

(1) For dilute suspensions of particles (up to 10%), the vis-

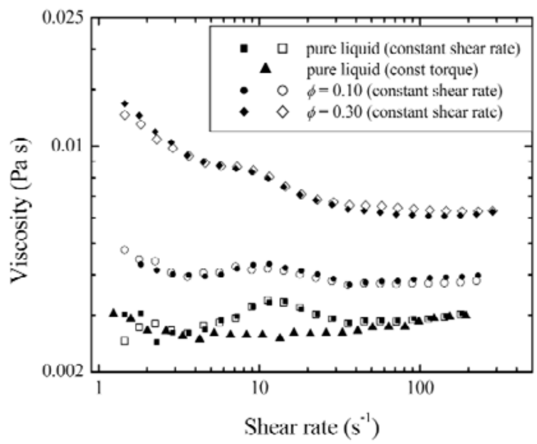

Fig. 1. Measured viscosity as a function of shear rate for the pure liquid and suspensions at volume fractions ϕ of 10% and 30%. Closed symbols: increasing $\dot{\gamma}$; open symbols: decreasing $\dot{\gamma}$.

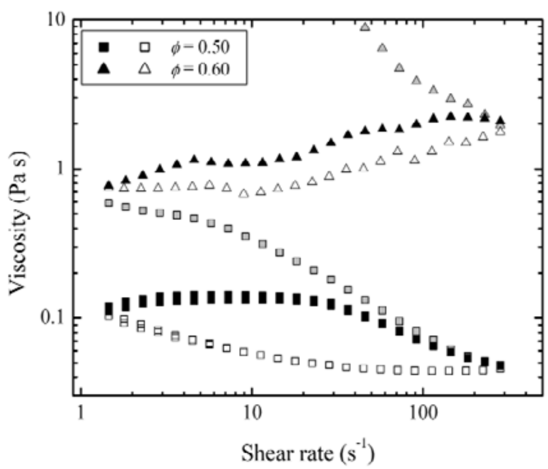

Fig. 2. Viscosity of the glass/LST suspensions at volume fractions of 50% and 60% for several cycles of increasing and decreasing shear rates. Closed symbols: increasing $\dot{\gamma}$; open symbols: decreasing $\dot{\gamma}$; filled grey symbols: first sequence of increasing $\dot{\gamma}$.

cosity is essentially independent of shear rate within experimental error, and the same behavior is seen for increasing and decreasing shear rates. The peaks seen near 10 s^{-1} for the pure liquid and dilute suspensions are instrumental artifacts for liquids with low viscosities that are close to the smallest values that can be detected by the rheometer in controlled shear rate mode. (Note that the viscosity of the pure liquid is only about 3 times the viscosity of water). For higher viscosities, these peaks vanish. To be certain that the pure liquid is Newtonian, additional experiments were performed in controlled torque mode, which has greater sensitivity. The results, shown by the solid triangles in Fig. 1, show that the spurious peaks seen in the controlled shear rate mode are no longer present and confirm that the pure liquid does exhibit normal Newtonian viscosity.

(2) For particulate suspensions with volume fractions in the range from 20 to 30%, marked shear thinning is observed (see Fig. 1). In colloidal suspensions, shear thinning is usually due to the effects of Brownian motion and interparticle interactions [6]. By contrast, for our suspensions, Brownian motion is negligible and the interactions between the spheres are expected to be "hard sphere" to an excellent approximation; hence shear thinning cannot be ascribed to either of these mechanisms, but is most likely due to the change in the configurations of the particles as the shear stress is applied to the suspension. For this range of particle concentrations, increasing the shear rate is believed to cause the initially random three-dimensional dispersion of the particles to be reorganized in a more two-dimensional layered arrangement. This layered arrangement facilitates the flow, and hence the viscosity decreases.

(3) For more concentrated particulate suspensions ($\phi \geq 40$ %), the shear rate dependence of the viscosity shows considerable hysteresis (Fig. 2), reflecting a slow evolution of the particle configurations in the suspension over time scales that are longer than the duration of each individual measurement (~ 1 min for the data in Fig. 2). In addition, for most of the data at high volume fractions, the viscosity decreases considerably as the shear rate is first increased after packing the suspension into the cell. This demonstrates that the initial 3D structure of the suspension, which is likely to be more or less random depending on the method of packing the suspension into the shear cell, rapidly becomes rearranged as a result of shearing the suspension in order to facilitate the flow, thereby reducing the viscosity.

(4) For the most concentrated suspensions ($\phi \geq 55$ %), the viscosity shows shear thickening behavior after the initial decrease in viscosity has occurred. This increase in the viscosity with shear rate is believed to be caused by interparticle interactions that tend to break up the layered structure formed at lower ϕ leading to more disordered configurations of the particles that impede the flow of the suspension and increase the viscosity [7].

To further investigate the viscosity evolution associated with the hysteresis, the time dependence of the viscosity was measured every second for up to 30 minutes at each shear rate. Typical results at $\phi = 0.50$ are shown in Fig. 3, which displays the time dependence of the viscosity at several shear rates (the complete sequence of shear rates was 0.5 s^{-1}, 1 s^{-1}, 2 s^{-1}, 5 s^{-1}, 15 s^{-1}, 100 s^{-1}, 15 s^{-1}, 5 s^{-1} and 2 s^{-1}). The solid curves are guides to the eye using the empirical relation $a + bt + c\exp[-t/\tau]$, where a, b and c are constants, and τ is a relaxation time. For high shear rates (> 5 s^{-1}), the viscosity decreases approximately exponentially with time, an effect that is likely due to the (reversible) migration of particles into the low shear region at the bottom of the cell. This effect has been modeled by Leighton and Acrivos as shear induced diffusion of the particles normal to the plane of shear, leading to a depletion of particles in the gap and a consequent reduction in the viscosity [2]. While their model gives a qualitative description of our data, there are substantial differences in the magnitude and shear rate dependence of the particle diffusion coefficients compared with those expected from Leighton and Acrivos's work, possibly reflecting additional effects due to the much smaller viscosity of the pure liquid in our experiments. At low shear rates, this migration effect is swamped by another mechanism, not reported in the previous experiments, that causes the viscosity to increase approximately linearly with time over the time window of these measurements. This slow increase in the viscosity may be related to the formation of higher density "blobs" in the suspension that remain deposited in a regular pattern around the rim of the stator after unloading the cell. Further work is required to model this effect quantitatively and determine the underlying mechanism, which may be related to particle rearrangements associated with precursors of the jamming transition that has been demonstrated at $\phi \approx 0.57$ under small oscillatory shear stresses [8].

We have also performed Dynamic Sound Scattering measurements to obtain additional information on how the particles actually move in response to shear. These experiments were carried out in a custom-made cylindrical shear cell on the 30% suspensions. The cell had a 4-mm-wide gap between the stationary outer wall and the rotating inner wall, which went straight to the

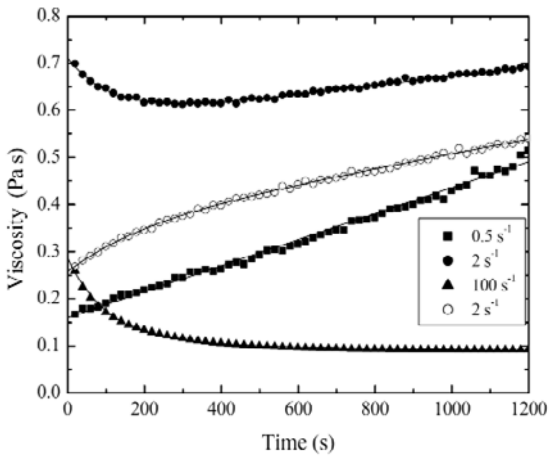

Fig. 3. Evolution of the viscosity under steady shear at ϕ = 0.50. Closed symbols: $\dot{\gamma} > \dot{\gamma}_{previous}$, open symbols $\dot{\gamma} < \dot{\gamma}_{previous}$.

bottom of the cell, thereby eliminating any low shear rate fluid reservoir at the bottom. The ultrasonic scattering measurements were performed in reflection mode using a focusing transducer whose orientation relative to the cell could be varied. Figure 4 shows the measured field autocorrelation functions for three orientations of the transducer: beam axis perpendicular to the cell wall (radial), beam axis inside the cell inclined at 15.5° with respect to the normal in the plane perpendicular to the cylindrical axis (z) (contains tangential and radial velocity components), beam axis at 60° to the *z*-axis (contains radial and vorticity velocity components). Comparing Eq. (2) to these data reveals that only in the tangential direction is the average velocity non-zero and that the variance in the particle velocities is appreciable in all three directions. Since we use a pulsed technique, the particle velocities can be measured as a function of depth inside the cell, which in this case corresponds to the middle of the gap. Figure 5 shows the shear rate dependence of the average velocity parallel to the shear, as well as the standard deviation of the velocity fluctuations σ_V along the three orthogonal directions. The average velocity in the middle of the gap is

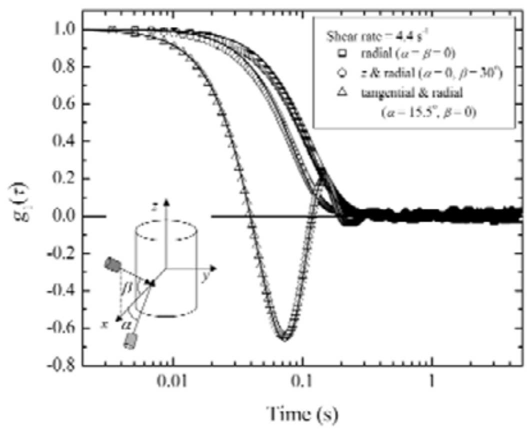

Fig. 4. Autocorrelation functions of the acoustic fieldfluctuations for three beam directions in the glass/LSTsuspensions at f = 0.30. Solid lines are fits of Eq. (2)to the data (symbols). The insert shows the transducerorientations relative to the cylindrical cell.

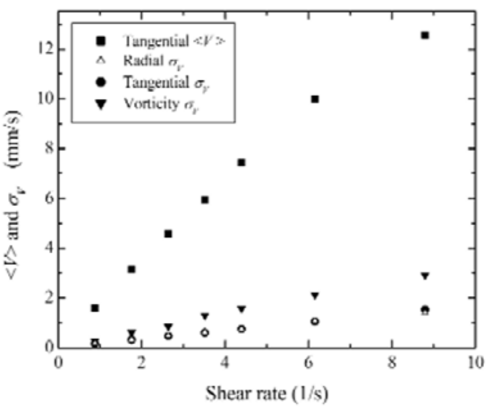

Fig. 5. DSS measurements of the shear rate dependence of the average particle velocity in the tangential direction compared with all three components of the standard deviation of the velocity fluctuations.

roughly half the rotor velocity as expected, but increases less rapidly than the shear rate, suggesting that the velocity profile across the cell may be nonlinear at high shear rates for this low viscosity system. By contrast, σ_V is proportional to the shear rate within experimental error and is isotropic in the plane perpendicular to the cylindrical axis, its magnitude being approximately 10% of the average velocity. Interestingly, the standard deviation of the velocity fluctuations in the vorticity direction is twice as large. These results illustrate the potential of DSS for probing the dynamics of non-Brownian particles under shear; this technique may be especially useful for studying suspensions in which the particles and fluid cannot be index matched optically, or are even opaque.

CONCLUSIONS

The viscosity of neutrally buoyant suspensions of non-Brownian particles in a low viscosity liquid has been investigated as the shear rate and concentration were varied. These data show that the flow behavior of particulate suspensions is intimately related to the particle configurations, and that the motion of the particles as they rearrange their relative positions in response to applied shear is critical in determining the viscosity. The potential of a new ultrasonic scattering technique, Dynamic Sound Scattering, for obtaining quantitative information on the particle dynamics has been illustrated. Future work will use this technique to determine the particle velocity profiles and fluctuations over a wide range of concentrations and applied shear rates for both steady and oscillatory flows.

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