## Shear-Induced Structure and Velocity Fluctuations in Particulate Suspensions Probed by Ultrasonic Correlation Spectroscopy and Rheology

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**Abstract.** We report measurements of the steady-state shear viscosity and shear-induced structure of neutrally buoyant non-Brownian particulate suspensions. 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 of the suspensions are discussed. A new ultrasonic technique was used to determine the relaxation time required for the particles to come to rest after the shear rate is set to zero.

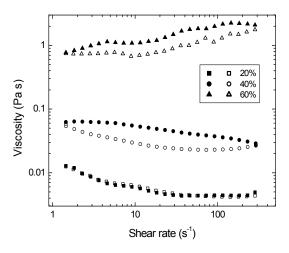
The flow properties and dynamics of particulate suspensions are of considerable current interest both scientifically and practically. Since the pioneering work of Gadala-Maria and Acrivos [1], much of this interest has focused on how the flow properties are influenced by the microstructure, the hydrodynamic inter-particle interactions and the redistribution of particles in the suspension under shear. 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 long-term viscosity decrease to shear-induced particle migration to low shear stress regions. The application of an NMR imaging technique [3] and a novel correlation method [4] have shed light on the evolution of particle concentration profiles and shear-induced self-diffusion in Couette flow. To further investigate this behaviour, we report measurements of the steady-state shear viscosity and particle velocities in neutrally buoyant suspensions of non-Brownian particles using rheology and Dynamic Sound Scattering [5].

The neutrally buoyant suspensions were assembled by first repeatedly sieving spherical borosilicate glass beads to obtain a nearly monodisperse size distribution of uniform-density particles (diameter  $d = 127 \pm 22 \,\mu\text{m}$ , density  $\rho = 2,220 \,\text{kg/m}^3$ ). The glass beads were then immersed in a density-matched liquid consisting of a mixture of LST heavy liquid (a

low viscosity aqueous solution of lithium heteropolytung states) and water. Suspensions with particle volume fractions  $\phi$  of 0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.55, and 0.6 were prepared.

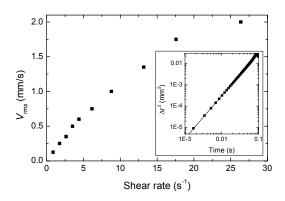
The viscosity measurements were carried out in a concentric cylindrical cell at a temperature of 27° C using a TA instruments AR2000 rheometer. The viscosity of the suspensions was investigated under steady shear flow at constant angular velocity from 0.1 to 20 rad/s for both increasing and decreasing angular velocity, in a series of steps or cycles that allowed long-term evolution of the suspension microstructure to be investigated.

Figure 1 illustrates general trends of the viscosity during Couette flow for the suspension at different volume fractions. Four distinct types of behaviour were found for the viscosity of the suspensions as the shear rate and concentration were varied. For dilute suspensions ( $\phi$  up to 10%), normal Newtonian viscosity was found, as expected. For  $\phi$  between 20 and 30%, reversible shear thinning was observed, suggesting that the particles rearrange in a layered structure to facilitate the flow. At higher  $\phi (\geq 40\%)$ , considerable hysteresis was found at increasing and decreasing shear rates, reflecting a slow dynamic evolution of the suspension microstructure. At even higher concentrations ( $\phi \geq 55\%$ ), the viscosity was found to increase with shear rate, as particle interactions and jamming tend to break up the layered structure that was favored at lower  $\phi$ .

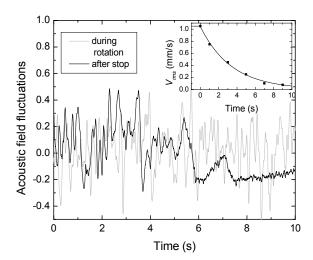


**FIGURE 1.** Measured viscosity as a function of shear rate for suspensions of neutrally buoyant glass beads at volume fractions of 0.2, 0.4, and 0.6. Solid and open symbols represent data at increasing and decreasing shear rates.

To measure the dynamics of the particles directly, we use Dynamic Sound Scattering (DSS), a new technique in ultrasonic correlation spectroscopy that allows the temporal evolution of the mean square displacement of the particles  $(\Delta r^2(\tau))$  to be determined from the fluctuations of singly scattered The DSS measurements were ultrasound [5]. performed in a cylindrical shear cell, with a stationary outer wall and a rotating inner wall, separated by a 4 mm gap. Figure 2 shows typical results taken in reflection using a focusing transducer with the beam axis perpendicular to the cell wall. In this orientation, the technique is still sensitive to the dominant motion of the particles in the direction of the shear flow because of the angular distribution of



**FIGURE 2.** Particle velocity as a function of shear rate. The inset shows the expected quadratic time dependence of  $\Delta r^2$  at one shear rate:  $\Delta r^2(\tau) = V_{rms}^2 \tau^2$ .



**FIGURE 3.** Field fluctuations during steady shear (grey line) and after the rotor was stopped (black line). The inset shows the decay of the particle motion after the shear rate was set to zero.

incident and scattered waves about the beam axis, allowing the average velocity of the particles  $V_{rms}$  parallel to the applied shear to be probed.

Figure 3 shows the field fluctuations both during shear and immediately after the rotor was stopped. It can be seen that the field fluctuations gradually slow down after the shear rate is set to zero, allowing the relaxation time of the particle dynamics, including the time required for shear-induced structures to break up, to be determined. From the exponential decay of  $V_{rms}$  shown in the inset, this relaxation time was found to be 3.4 s for this system at  $\phi = 0.30$ .

## **ACKNOWLEDGEMENTS**

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