Effects of Particle Surface Roughness on Particle Interactions in Concentrated Suspensions

L. A. Mondy
Sandia National Laboratories
Division 1511
Albuquerque, NM 87185

A. L. Graham
Los Alamos National Laboratory WX-4
Los Alamos, NM 87545

P. Stroeve and A. Majumdar
Department of Chemical Engineering
University of California
Davis, CA 95616

Introduction

This study focuses on determining the relative importance of hydrodynamic and surface effects in the movements and interactions of particles in highly concentrated suspensions. We observe that even at high concentrations of solids ($\phi = 0.50$), both rough and smooth spheres falling through a suspension of other spheres experience the same mean stress field. That is, over the range of our data, all falling balls with different surface roughnesses experience the same average resistance to motion, as though the effects of the numerous surrounding spheres could be replaced by a single, hypothetical, one-phase fluid.

In the absence of wall effects, the terminal velocity of a sphere in creeping flow through a Newtonian liquid can be determined via Stokes' law:

$$v_t = \frac{d^2(\rho - \rho_l)g}{18\mu}. \tag{1}$$

Early experimental researchers of the drag on a sphere reported no effect of the surface roughness in creeping flow through Newtonian liquids (Landenberg, 1907; Arnold, 1911).

However, we do not know the effects of the surface characteristics of a sphere falling in a highly concentrated sea of other rough particles. At high concentrations the average distance between suspended particles is much less than the diameter of the particles. For example, if the suspended particles are spheres in a simple cubic arrangement, the relationship between the volume concentration of the solids in the suspension and the gap width between nearest neighbors can be expressed (Graham, 1981) as

$$\frac{d_s}{h} = \frac{(\phi/\phi_m)^{1/3}}{1 - (\phi/\phi_m)^{1/3}}. \tag{2}$$

The maximum packing concentration for simple cubic packing is $\pi/6 = 0.524$. Therefore, in a suspension of spheres, which on the average could be taken as being in such an arrangement, at ϕ of 0.5, the gap width is less than 2% of a suspended sphere diameter. Subsequent work on particle clustering in concentrated suspensions (Graham et al., 1984) showed that actual gap separation distances between particles in a sheared suspension can be much smaller than those predicted by simple cubic packing.

Our intent in this study was to test the hypothesis that at very small interparticle separation, surface characteristics would influence significantly the rheological behavior of the suspension. To address this question, we performed falling-ball experiments in a model suspension of neutrally buoyant, uniform spheres ($d_s = 0.32$ cm) at a volume fraction of solids of 0.50 in a Newtonian liquid. With these large suspended spheres, the hydrodynamic forces dominated the electrostatic and Brownian forces

Correspondence concerning this paper should be addressed to L. A. Mondy

(Jeffrey and Acrivos, 1976). The effect of surface roughness was examined by varying the surface characteristics of four, otherwise identical, falling balls.

Experimental Method

Background

An earlier study (Mondy et al., 1986a) showed that the average velocity of a ball falling through a suspension with ϕ up to 0.45 could be modeled with Eq. 1 as if it were falling through a Newtonian continuum characterized by the apparent suspension viscosity, provided that the container walls were far away. All falling balls in this previous study were commercially obtained ball bearings with the same surface roughness.

The discrete nature of the suspensions was very apparent in these experiments. Balls that were close in size to the suspended spheres fell erratically, slowing dramatically as they approached and circumvented suspended spheres and then speeding up in periods of free fall through the interstices between suspended spheres. Therefore, the terminal velocity of the falling ball was an indication of the effects of many interactions between the falling ball and the suspended spheres. Large balls falling through the suspension fell in a more constant and smooth fashion. Surprisingly, all size balls, both smaller and larger than the suspended balls, experienced the same average resistance to motion.

In these investigations, we used real-time radiography coupled with falling-ball rheometry to study the rheology of concentrated suspensions. The terminal velocities of balls falling through suspensions were used to calculate the average apparent viscosities of dilute and moderately concentrated suspensions ($0 \le \phi \le 0.45$). It was found that these average viscosities, extrapolated to correct for wall effects, agreed with apparent viscosities collected independently (Thomas, 1965) in shear and capillary rheometers, as shown in Figure 1. Although not shown in the figure, we note that they are also in excellent agreement with more recent data (Gadala-Maria and Acrivos, 1980; Chan and Powell, 1984).

The wall effects in these dilute to moderately concentrated suspensions were found to be indistinguishable from those predicted by Faxén (1921) for pure Newtonian fluids at concentrations below 0.30. At a $\phi \ge 0.45$, additional wall effects, relative to those in the pure Newtonian liquid, were observed when the falling ball diameter was larger than 0.1 of the cylinder diameter. Below this geometric constraint, wall corrections used for pure Newtonian liquids were also valid for the highly concentrated suspension. Furthermore, in the absence of wall effects, the average apparent viscosities of the dilute and moderately concentrated suspensions were not functions of the size of the falling ball relative to the suspended spheres ($d \ge 0.75 d_s$).

In the present study, in order to study surface effects in a suspension of yet higher concentration, we selected a falling ball size in the range where the apparent viscosity was not affected by additional wall effects. The additional wall effects were indicative of a change of mechanism that was not addressed in this study. First, we dropped falling balls in a range of sizes, but with similar surface characteristics, and determined a size for the falling balls that avoided additional wall effects identified earlier in highly concentrated suspensions. Next, we compared the apparent viscosities given by several balls with different surface characteristics but with similar diameters and densities. These falling balls with various roughnesses were also dropped

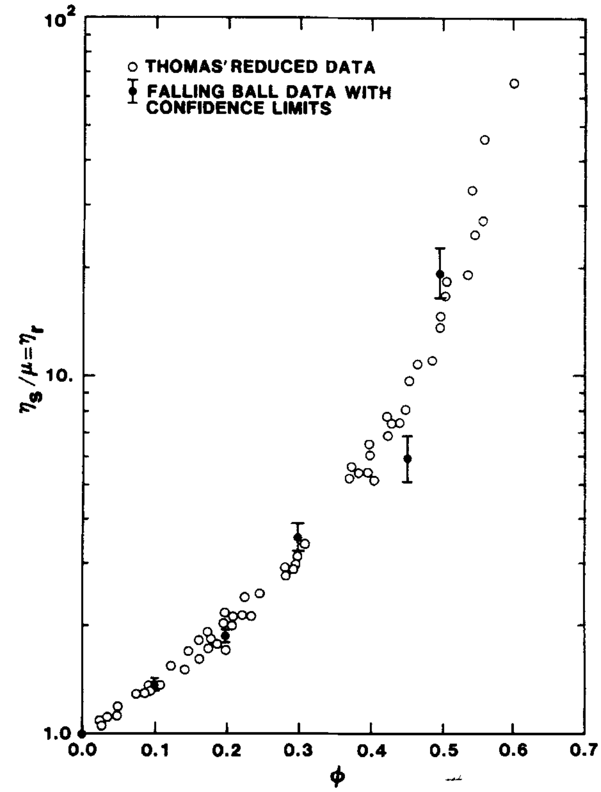

Figure 1. Relative viscosities from experiments compared with data from shear and capillary rheometers (Thomas, 1965).

Experimental data corrected for wall effects. Bars represent Student *t*-test estimates of reproducibility (95% confidence) of averages from independent sets of at least five ball drops for concentrations ϕ up to 0.45 (Mondy et al., 1986) and of 15 drops of $\phi = 0.50$ (present study).

in a pure Newtonian liquid to indicate if the differences in surface conditions, as well as small differences in size and weight, would result in measurable differences in the predicted viscosities. No statistically supported difference due to the surface roughness variations was observed in either the pure suspending liquid or in the suspension, under creeping flow conditions.

Apparatus

Because of the high concentration of particles in the suspensions studied, the falling ball cannot be observed optically. A technique for studying such opaque, highly concentrated suspensions was developed by Mondy et al. (1986b) using real-time radiography techniques. A fixed X-ray generator produces a beam that passes through the cylinder ($D = 15$ cm) containing the suspension in which a dense ball is falling. The falling ball attenuates the X-rays and forms a shadow on an image intensifier. The image intensifier converts the X-ray image to an optical image that can be recorded by a video system. This system is the Spin Physics SP-2000 Motion Analysis System by Eastman Kodak. With this system, any frame can be examined and digitized data can be taken for position as well as accurate elapsed time. The accuracy in position and time is estimated to be ± 0.04 cm and ± 0.001 s, respectively, with our geometry and frame rate.

Suspension

The suspension was made from polymethyl methacrylate (PMMA) spheres suspended in a Newtonian liquid. The commercial availability of large quantities of suspended particles was a limiting factor in the experimental design of this study. The surface condition of the suspended spheres was fixed by the grinders that produced the spheres from PMMA rod. The PMMA spheres were manufactured by Clifton Plastics and were monodisperse (0.317 cm) in diameter. The manufacturer's published tolerances of these balls were ±0.005 cm dia. and 0.63 μm in maximum peak-to-valley surface roughness. The density of the PMMA spheres depended on the particular batch manufactured and usually varied between 1.17 and 1.19 g/cm^3.

The suspending liquid was a solution of approximately 84 wt. % polyalkylene glycol (UCON-75H-9500 oil) manufactured by Union Carbide and 16 wt. % 1,1,2,2-tetrabromoethane (TBE). (Readers are cautioned not to use this mixture in the presence of aluminum, as there is speculation that a potentially dangerous, highly exothermic reaction of the aluminum with the halogenated hydrocarbon may be possible.) The UCON oil provided a highly viscous suspending liquid, and the TBE allowed the density of the mixture to match that of the suspended spheres. The antioxidant Tinuvin 328, made by Ciba-Geigy, was added (≈0.1 wt. %) to inhibit the dissociation of TBE when exposed to ultraviolet radiation. The density of this liquid mixture was also a function of temperature. By precisely setting the temperature, we could control the density of the liquid to match exactly the density of a particular batch of PMMA spheres and could make a neutrally buoyant suspension. For these experiments, neutral buoyancy was obtained at 30.0°C. At this temperature, the viscosity of the suspending liquid was 34.2 poise (342 Pa · s). With careful control of the temperature to ±0.1°C, the time scale of the experiment was always well over four orders of magnitude faster than the time scale of any settling or rising of the suspended particles.

Falling balls

The first set of balls dropped in the suspension were brass ball bearings ranging in size from 0.24 to 0.95 cm. The balls were grade 200 (standard of the Anti-Friction Bearing Manufacturers Association), which corresponds to surface roughness of 0.2 μm. These balls were used only to determine whether or not Newtonian wall corrections could be used at a ϕ of 0.5. A set of four balls with various surface roughnesses was then manufactured at the Los Alamos Target Fabrication Facility of Los Alamos National Laboratory. These balls were made of copper with a relatively thick (>50 μm) gold plating. They were roughened and at the same time made highly spherical by a three-point polishing procedure. The surface roughness was dependent on the size of grit used in this process.

Great care was taken to characterize these spheres accurately. The peak-to-valley height of the surface, e, was measured at three widely spaced points on each ball with a sensitive (±0.001 μm) mechanical stylus. The average values of e of the four balls were 0.027, 0.325, 2.41, and 6.30 μm. The diameters of these balls were measured with an electronically controlled micrometer and were, respectively, 0.50924, 0.50944, 0.51059, and 0.51015 cm.

Once these balls had been manufactured and characterized, care was taken to prevent damage to the surface finishes. Special techniques were developed to handle these balls. The balls were routinely picked up with a Teflon cup attached to a vacuum hose. Soft, surgical gloves were worn when it was necessary to touch the balls directly.

Experimental procedure

We measured the terminal velocity of the characterized brass balls and the four gold balls in the suspension with ϕ of 0.50. There were at least 15 trials using each size of brass ball and at least 10 trials using each gold-plated ball. Each falling ball was allowed to equilibrate thermally with the liquid before being dropped. Each ball was also wetted with the liquid to prevent trapping an air bubble on the ball's surface. It was then dropped down the centerline of a 50 cm high cylinder via a guide tube and allowed to settle to the bottom. The balls were dropped remotely from a vacuum cup; therefore, there was no variation in the initial conditions of each drop. Each ball was retrieved through a valve at the bottom of the cylinder. The suspension was stirred between each drop. A vertical distance of approximately 6 cm around the midplane of the cylinder of each fall was recorded. The terminal velocity of each ball was averaged over this distance. The viewed section was always at least one cylinder diameter from the start and from the end of the fall. The cylinder was maintained at a constant temperature ±0.1°C at all times.

Results and Discussion

Brass falling balls of five sizes between 0.24 and 0.95 cm all gave statistically the same viscosity of the suspension, when the apparent viscosity was corrected for wall effects. The apparent viscosity given by each ball was calculated via the relationship given in Eq. 1, using the measured characteristics and terminal velocity of the ball. The apparent viscosity of the suspension was then determined from the average of at least 15 drops for each ball size. The apparent viscosity for each falling-ball size was then corrected for wall effects by extrapolating the data to estimate the apparent viscosity that would be given in a cylinder of infinite diameter. We note that results obtained using Faxén's correction matched the extrapolated data closely, implying that no additional wall effects beyond those that would be felt in a Newtonian continuum were present. The apparent viscosity for a suspension with ϕ of 0.50 is presented in Figure 1. Also in Figure 1 are those apparent viscosities previously measured with falling-ball rheometry (Mondy et al., 1986a), as well as independent shear and capillary data (Thomas, 1965).

The apparent viscosity values, calculated from individual measurements of the terminal velocity of a ball, exhibited a large standard deviation; velocity measurements from any two successive trials could vary by as much as 100%. This variation was due primarily to the statistical nature of the interactions of the falling ball and the suspended spheres. Only by averaging the measurements from a large number of trials does the continuum nature of the suspension become apparent. In this system, for example, averages of velocity measurements from seven or more trials were reproducible. The vertical bars on the data shown in Figures 1 and 2 represent the confidence intervals, or reproducibility at 95% confidence as determined by a Student t-test, of the mean values of apparent viscosity. We defined these confidence intervals to be $ts/\sqrt{n}$ (Walpole and Myers, 1972). Note that these confidence limits are inversely proportional to the square root of the number of ball drops and would vanish only in the limit of an infinite number of trials. Thus, for example, if 60 trials were performed instead of 15 and the value of s did not change appreciably, the value of the confidence limits could be reduced by about 50%. Time and equipment limitations precluded such an extension of this study.

The gold-plated balls with characterized surface roughnesses were made to have a diameter of 0.51 cm, within the range of sizes of the brass balls used above. The relative roughnesses, e/d, for the falling balls varied over three orders of magnitude. For the four balls, these roughness values were about 5×10^{-6}, 6×10^{-5}, 5×10^{-4}, and 1×10^{-3}. These relative roughnesses were comparable to those used to determine friction factors for flow in pipes, where the relative roughness ratio is usually defined as the ratio of the surface roughness to the diameter of the pipe. However, if one normalized by the approximate distance between nearest neighbors, h as given by Eq. 2, in a suspension with ϕ of 0.5, one would get relative roughnesses e/h of up to 0.1, much larger than any roughness studied in pipe flow. We represent our results in this study in terms of e/d, not e/h, to avoid the ambiguity of an assumed packing for the randomly distributed particles.

Each of the four balls was dropped in the pure Newtonian suspending liquid five times. The viscosity of the liquid, calculated using the average terminal velocity and the measured diameter and mass of a particular ball, varied at most 1% from the mean given by all four balls. This gave us high confidence in our characterization procedures and in our velocity measurements. It also showed that no significant difference caused by variations in surface roughness could be seen in the pure liquid. This observation that the surface condition of the ball has little effect on its creeping flow terminal velocity in Newtonian liquids is in agreement with the observations of early researchers (Landenburg, 1907; Arnold, 1911), who measured the terminal velocities of balls with pitted or corroded surfaces as well as balls with polished surfaces. They found no appreciable change in the terminal velocity that could be attributed to surface irregularities. Numerical simulations underway in our group also support this observation (Wagner et al., 1986). The effects of surface conditions at higher Reynolds numbers are outside the scope of the present work.

Although there are variations of about ±6% in the mean apparent viscosity of the highly concentrated suspension as given by the four different balls, the differences are well within the confidence limits of the measurements. Figure 2 shows the apparent viscosity averaged over at least 10 drops of each ball vs. the relative surface roughness (e/d) of each ball. A Student t-test shows the four viscosity values to be statistically indistinguishable to 95% confidence. We note also that an F-test shows that the standard deviations of each group of trials are statistically indistinguishable, again to 95% confidence (Walpole and Myers, 1972).

There may exist effects of surface roughness within the confidence limits reported here (approximately ±15%). However, the existence of such effects is not supported by any trend in the average viscosities. We cannot support statistically the existence of any effects of surface roughness with our present data. As discussed earlier, the resolution of any possible effects within the present confidence limits can only be made with a vastly expanded data base.

In suspensions in which viscous hydrodynamic forces dominate, these results show that there are no significant effects of surface roughness in falling-ball rheometry over the range of our data ($\phi = 0.50$, $10^{-6} < e/d < 10^{-3}$, and $e/d_s = 10^{-4}$). As the suspension approaches maximum packing density, is constrained in a small cylinder, or is in the inertially dominated regime, surface effects may become important. These are potential areas for future study.

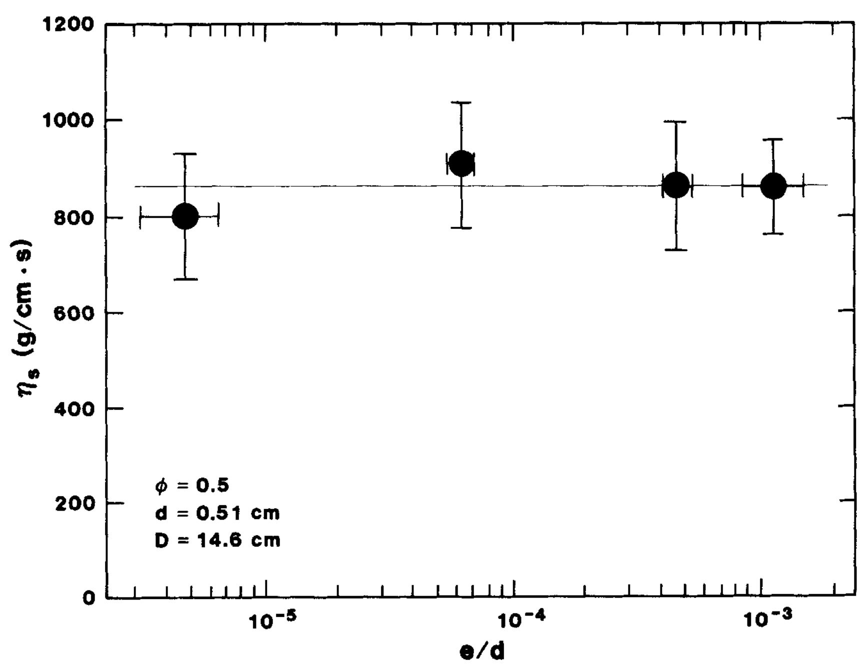

Figure 2. Apparent viscosity vs. relative roughness.

No correction for wall effects; $\phi = 0.50$ suspension, creeping flow conditions ($Re \approx 10^{-4}$). Horizontal bars represent range of e/d measurements; vertical bars represent 95% confidence limits on viscosity measurements. Within accuracy of data, results are indistinguishable.

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Notation

d = diameter of falling sphere
d_s = diameter of suspended spheres
D = diameter of cylindrical container
e = peak-to-valley height of surface
g = acceleration due to gravity
h = gap width between nearest-neighbor suspended spheres
n = number of tests (drops)
Re = Reynolds number; $\rho_l dv/\eta_s$, where v is the terminal velocity of the ball
s = standard deviation of sample
t = 95% confidence level from standard statistical t tables

Greek letters

η_s = apparent viscosity of suspension
μ = viscosity of Newtonian liquid
ρ = density of falling sphere
ρ_l = density of liquid
ϕ = solids volume fraction of a suspension
ϕ_m = solids volume fraction at maximum packing

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