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BRIEF COMMUNICATION

TECHNIQUES OF MEASURING PARTICLE MOTIONS IN CONCENTRATED SUSPENSIONS

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1. INTRODUCTION

The lack of knowledge concerning particle dynamics has made understanding disperse multiphase systems difficult. In concentrated suspensions, experimental observation of the interior of a flow field is obstructed by the particles themselves. In this paper, the authors describe two new techniques, one optical and one using X-rays, for observing particle dynamics in concentrated suspensions. First, we describe a method in which the refractive index of the suspended particles is matched to that of the suspending liquid. The motion and interactions of opaque particles subjected to various flow fields can then be seen. Discussed next is the use of real-time radiography to observe the motion of high density test particles in very concentrated systems. Third, we describe the high-speed video system used for recording data taken with either observation technique. Finally, we examine briefly an example study using falling-ball rheometry to demonstrate the techniques.

The analysis of falling-ball rheometry data is based on Stokes' solution of the terminal velocity (v_t) of a sphere falling in creeping flow through an unbounded, incompressible, Newtonian liquid (Lamb 1945):

$$\mu = 2R^2(\rho_s - \rho)g/9v_t. \qquad [1]$$

Here R is the radius of the sphere, ρ_s is the density of the sphere, ρ is the density of the fluid, and g is the acceleration due to gravity. In a non-Newtonian liquid, an apparent viscosity of the liquid can be measured as if the liquid were a hypothetical Newtonian fluid (Gottlieb 1979). Although Stokes derived [1] over 100 years ago, the principles of falling-ball rheometry have been used only rarely to study suspensions, because suspensions are opaque in general, and there is difficulty in determining the time-dependent position of the falling ball (Ward & Whitmore 1950). The two techniques described here permit an accurate observation of a ball falling through a suspension and thus provide a new tool for basic research into the nature of suspensions.

2. THE CREATION OF AN OPTICALLY-TRANSPARENT, NEUTRALLY-BUOYANT SUSPENSION

At high concentrations, if all the suspended spheres are opaque, only those at the apparatus boundary can be seen. Therefore, one would like to match the index of refraction of the suspended material and the suspending fluid, so that opaque spheres within the apparatus can be seen through a surrounding, transparent suspension. Ideally, in a falling-ball experiment, the suspension should be at rest so that the only forces on the suspension are created by the moving sphere. Therefore, the suspending fluid must also have the density of the suspended spheres.

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A mixture of UCON™ oil (a polyalkylene glycol manufactured by Union Carbide) and 1,1,2,2-tetrabromoethane (TBE) was used to match the index of refraction of PMMA spheres in experiments by Graham *et al.* (1984) to study particle clustering in concentrated suspensions subjected to a homogeneous shear field. However, they were not limited by the stringent requirement that the suspension be neutrally bouyant. A third component is needed to ensure that the density of the mixture matches that of the spheres.

To find a suitable third component, we assume that the refractive index is approximately linear with volume fraction, since the refractive index depends on number density. With three components we have three equations: the volume-averaged refractive index of the mixture equals the refractive index of PMMA; the volume-averaged density equals the density of PMMA; and the sum of the volume fractions equals one.

$$x_1\eta_1 + x_2\eta_2 + x_3\eta_3 = \eta_{\text{target}} \qquad [2]$$

$$x_1\rho_1 + x_2\rho_2 + x_3\rho_3 = \rho_{\text{target}} \qquad [3]$$

$$x_1 + x_2 + x_3 = 1 \qquad [4]$$

This system of equations assumes, of course, that there is no volume change with mixing.

Given three fluids, one can solve exactly for the composition. However, most combinations of three fluids will give impossible fractions, such as negative fractions. The third fluid must have a refractive index higher than that of the UCON oil but a density lower than that of the oil. Many aromatics, such as benzene and toluene, meet these constraints but are excellent solvents and will chemically attack PMMA. Terpineol also has suitable properties and will attack PMMA, but on a grossly slower time scale.

If the linear approximation does not result in a perfect match of the properties of the solid and liquid, then the composition must be adjusted. The technique of evolutionary operations (Box 1957) can be used to modify the UCON/TBE/terpineol mixture to reach the desired density and optical properties. In this case, the composition is near the linear estimate and was quickly perfected.

At 26°C, we match index of refraction and density of the PMMA with a mixture of 33.7 wt% UCON, 24.4 wt% TBE, 41.8 wt% terpineol and 0.1 wt% Tinuvin™, an inhibitor manufactured by Ciba Geigy. Dilution of the terpineol with the other components slows the reaction with the PMMA spheres. PMMA spheres in the same mixture ratio of components, but without the Tinuvin, suffered only a 0.01% weight loss in three days. Tinuvin, originally added to prevent discoloration due to the breakdown of TBE, further slows the solvent action so that negligible weight loss occurs within three days. However, if a neutrally-buoyant suspension must be saved for more than a few days, this three-component suspending fluid is not recommended.

The three-component solution is Newtonian. Samples of the solution were tested on a Rheometrics System Four Rheometer and a Deer Constant Stress Rheometer. No normal stresses or shear-rate dependence in the viscosity were detected.

The properties of the mixture are very dependent on temperature, and hence the experiments must be performed in a temperature-controlled environment. In order for one to see through a thickness of 15 cm of a concentrated (30% solids) suspension, the refractive index match must be perfect. The temperature in this case must be controlled to approximately ± 1°C. At lower concentrations the refractive index match and hence the temperature control is less important. Because of the critical temperature control and because of minor imperfections of the PMMA spheres, refractive index matching as an effective technique for most falling-ball experiments is limited to concentrations below about 30% solids.

3. REAL-TIME RADIOGRAPHY IN AN OPTICALLY OPAQUE SUSPENSION

For highly concentrated (over 30% solids) suspensions, another technique must be found to track the falling ball. Real-time radiography (Bryant 1983) offers an easy alternative to optical techniques (Karnis *et al.* 1966, Graham *et al.* 1984). Figure 1 is a diagram of a typical system. In this case, a fixed X-ray generator produces a beam with 150 kilovolts constant potential (kVCP), 20 milliamperes current and a 1.2 mm focal spot size. The beam

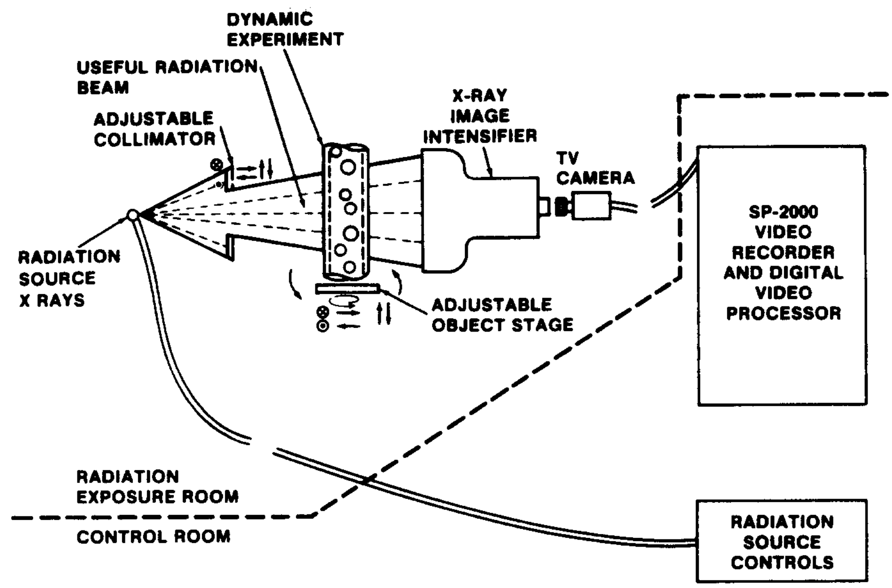

Figure 1. Diagram of real-time radiography equipment. X-rays, collimated by lead bricks, pass through a cylinder containing a suspension and are imaged by an image intensifier. This image is then recorded with a video system. For safety, the radiation source is controlled remotely. The video recorder and monitor are also in the control room, protected from radiation.

passes through a cylinder containing a suspension. The placement of the cylinder is adjustable, allowing the experimentalist to ensure that he is viewing a section of the cylinder where the falling-ball velocity is constant and without end effects. After passing through the test section, the X-rays strike an image intensifier. The image intensifier is made up of an input fluor that converts X-rays to light, a photoelectron layer that converts the light to electrons and an electron acceleration, or intensification, stage. Finally, the electrons are converted back to light at the output fluor of the image intensifier, producing a radiographic image. This image is recorded with a video system.

For the falling-ball experiments, with the radiographic technique, we need only a suspending fluid in which PMMA spheres are neutrally buoyant. Only a slight change in the composition of the UCON/TBE mixture used by Graham (1984) is needed to create a fluid with the density of PMMA. Because the solid and liquid have different coefficients of volume expansion, neutral buoyancy can only be maintained with careful temperature control. However, the density match is less sensitive to temperature changes than is the match of both density and index of refraction. Reproducible data can be taken with the temperature controlled to $\pm 4°C$.

With the relatively small X-ray focal spot, dense (such as steel, brass, tungsten carbide) falling balls as small as 0.3 cm in diameter, falling through a 15 cm diameter cylinder, can be viewed. At high concentrations the image is actually sharper than at lower concentrations, in contrast to the optical technique discussed in the previous section. This is because the liquid attenuates X-rays a small amount more than does PMMA. The attenuation property of a material depends on the density and atomic number of the material. The liquid, recall, contains bromine in the form of tetrabromoethane, and bromine has a high atomic number compared to the other components of the suspension. However, like the optical technique, real-time radiography has limitations. Due to the attenuation properties of the suspension, this technique is limited to cylinders of about 15 cm in diameter and to falling balls larger than about 0.3 cm in diameter, even at the maximum packing density.

4. HIGH-SPEED VIDEOGRAPHY FOR OPTICAL AND X-RAY IMAGING

With both imaging techniques, we record the path of the falling ball with a high-speed, digitized video system, the SP-2000™, manufactured by Spin Physics Division of Eastman Kodak. With the suspensions of interest, the extreme high-speed capability of this system is not needed, although faster falls are recorded at 200 frames per second and played back at 60 frames per second to minimize blurring. Primarily, this video system allows collection of accurate, digitized, position data for any desired frame. Figure 2 shows reticles positioned on a sphere, with the X- and Y-position values and the elapsed time indicated on the monitor. The accuracy in position and time are estimated to be $\pm$ 0.04 cm in this geometry and $\pm$ 0.0005 s, respectively. Since the data are in digital form, X- and Y-position data versus time can be input directly to a computer.

The SP-2000 can support two cameras simultaneously. In the optical techniques, the two images, which are views differing by a 90° angle, can be seen on the television monitor. This allows three-dimensional analysis of the path taken by the falling ball or of the motion of opaque PMMA suspended balls. The same video system can be used to monitor the output fluor of an X-ray-sensitive image intensifier; although, our real-time radiography system was limited to one view and hence two dimensions.

5. RESULTS

Suspensions with volume concentration (ϕ) ranging from 0 to 45% solids were studied in the initial series of falling-ball experiments performed with the radiographic technique. The optical technique was used to reproduce the data in a suspension with $\phi = 30\%$. The details of the data reduction, error analysis, and theoretical interpretation are beyond the scope of this brief paper and will be discussed in later publications (Mondy *et al.* 1985). However, a few examples of the collected data will be given here.

The solids were relatively large (diameter $d_s = 0.32$ cm) PMMA spheres. All of the suspensions were tested in cylindrical vessels 15 cm in diameter and 56 cm high. To test for wall effects, the suspensions with $\phi = 20\%$ and $\phi = 45\%$ were also tested in cylinders with diameters equal to 6, 9, and 11 cm. We dropped steel balls, with seven different diameters (d_f) spanning the range $1 \leq d_f/d_s \leq 12$. This range of ball diameters corresponded to Reynolds numbers ($d_f v_f \rho/\mu$) of 1.5×10^{-4} to 0.14 in the pure liquid. The Reynolds numbers decreased as the concentrations of solids, hence the effective viscosities, increased.

Each type of ball, as classified by its diameter, was dropped along the centerline axis of the cylinder and recorded at least five times. On route through a suspension, the smaller balls often drifted up to two ball diameters off the centerline due to interactions with the suspended spheres; however, this displacement on average was zero. Several falls under identical conditions then gave a mean terminal velocity for one type ball in a cylinder of a given diameter. The mean velocities were then extrapolated in order to estimate the velocity of a ball falling through an infinite medium (no wall effects present). Figure 3 shows the mean velocities, of many sizes of balls falling in four different cylinders, collapsed onto one graph and extrapolated to a zero value for the ratio of the falling ball diameter to the cylinder diameter, representing an infinitesimally small ball falling in an infinitely large cylinder. This extrapolated value was then used in [1] to predict the Stokes viscosity of the suspension.

The Stokes viscosity of the suspension can be normalized by the viscosity of the pure suspending liquid to give a reduced viscosity. The reduced viscosity can then be compared to reduced viscosities measured on other suspensions consisting of other particles in different suspending liquids. Measurements taken in the suspension made with the three-component solution ($\phi = 30\%$) give the same reduced viscosity as those taken with real-time radiography in the two-component solution. Because the PMMA may be degraded slowly by the suspending liquid, it is important to compare the measurements taken optically to those in which no chemical reactions are taking place. The reduced viscosities in all suspensions tested compare closely (Mondy *et al.* 1985) to existing literature values.

The data taken in different cylinders provide information about the effects of the wall on the velocity of a ball falling through a suspension. At lower concentrations ($\phi < 20\%$), the data agree closely with calculations by Faxén of the wall effect for a sphere in a Newtonian liquid (Faxén 1921, Gottlieb 1979). However, preliminary analyses show that wall effects become stronger at higher concentrations ($\phi > 20\%$) than those experienced by a sphere falling through a Newtonian liquid (Mondy *et al.* 1985).

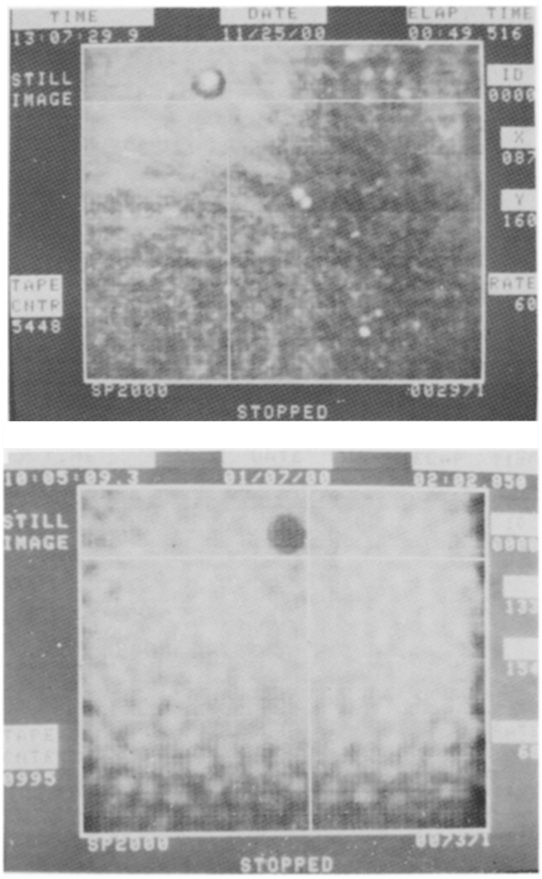

Figure 2. Video monitor screen in each technique (optical above and radiographic below), showing a ball falling through a suspension of 30% solid spheres. Horizontal and vertical reticles are shown crossing the screen, marking the position of the ball. Framing each screen, in clockwise direction from upper left, are the time, date, elapsed time from the beginning of recording to the particular frame being viewed, the X-coordinate, the Y-coordinate, recording in frames per second and the frame count. With the radiographic methods, the background attenuates the X-rays slightly more than the spheres do.

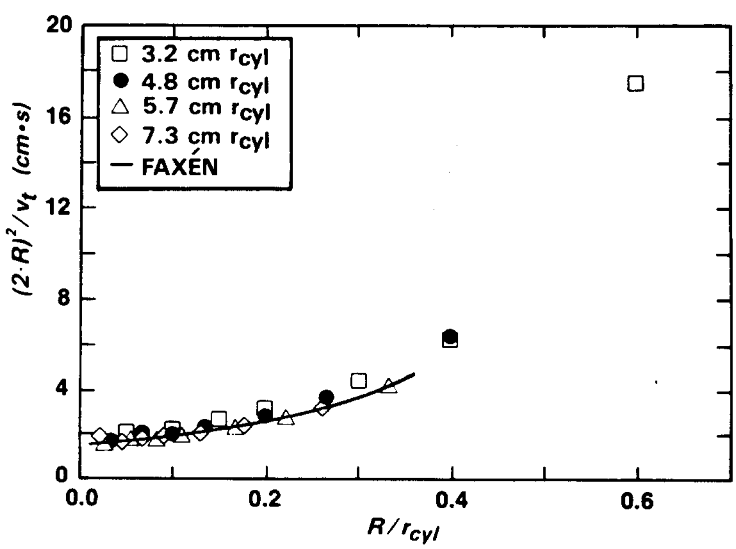

Figure 3. The falling ball diameter squared and divided by the terminal velocity (proportional to the Stokes viscosity) versus the falling-ball radius divided by the radius of the cylinder. The data represented here were taken in a suspension of 20% solids. The data from seven ball sizes in four cylinders collapse onto one graph. The solid line indicates the values predicted by Faxén's wall correction for Newtonian liquids. It appears that the suspension can be approximated by a Newtonian continuum characterized by the Stokes viscosity.

6. CONCLUSIONS

Two methods have been developed to view the interior of a concentrated suspension and have been used in a falling-ball experiment. The first involves creating a transparent suspension. The suspending fluid is a three-component liquid mixture that exactly matches the index of refraction and density of suspended PMMA spheres. With a high-speed video, precise observations can be made of the falling sphere. Real-time radiography is the key to the second method. X-rays are used to form a radiographic image of a dense ball falling through a neutrally-buoyant suspension. This image is visualized in real time by a video system. In both methods, the velocity of the ball is then calculated from the position versus time data taken from the video monitor. The Stokes viscosity of the suspension can be determined from the velocity data.

The two techniques have been used successfully in measuring the viscosity of suspensions of 10 to 45% solids. It was noted that the data from balls and cylinders of many sizes can be collapsed as in a Newtonian fluid. This implies that a continuum approximation and Stokes' law apply. At these small shear rates, the apparent viscosity is independent of the shear rate. Also, measurements have been made that will allow the determination of wall effects and, hence, the assessment of the distance that a velocity disturbance propagates in concentrated suspensions. These basic observations will give valuable clues to understanding the physics behind the rheological behavior of suspensions.

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