Int. J. Multiphase Flow Vol. 16, No. 4, pp. 651–662, 1990
Printed in Great Britain

0301-9322/90 $3.00 + 0.00
Pergamon Press/Elsevier

MEASUREMENTS OF THE VISCOSITIES OF SUSPENSIONS OF ORIENTED RODS USING FALLING BALL RHEOMETRY

L. A. MONDY,[1] T. G. MORRISON,[2] A. L. GRAHAM[3] and R. L. POWELL[2]
[1]Sandia National Laboratories, Division 1511, Albuquerque, NM 87185, U.S.A.
[2]Department of Chemical Engineering, University of California, Davis, CA 95616, U.S.A.
[3]Los Alamos National Laboratory, MEE-4, Los Alamos, NM 87545, U.S.A.

(*Received* 3 *July* 1989; *in revised form* 1 *November* 1989)

Abstract—Using falling ball rheometry, we measured the viscosities of suspensions of oriented rods (average aspect ratio = 19.83). The rods were neutrally buoyant in the suspending fluid and were sufficiently large so that Brownian and colloidal forces were negligible. The rods were oriented hydrodynamically by passing a fixture through the suspension which produced a local flow that tended to align the particles along the axis of the glass column containing the suspension. Balls were dropped along the axis of the cylinder and the average terminal velocity was measured using an eddy current detector. We find that the apparent viscosity for a suspension of rods that are approximately aligned is less than that for a suspension having the same volume fraction of rods that are randomly oriented. For example, at a volume fraction of 0.05, the relative viscosity of an oriented suspension is 1.52, whereas a randomly oriented suspension has a relative viscosity of 2.37. In addition, we find that the viscosity measured for the aligned rods closely correlates with the viscosities of suspensions of short fibers (having similar aspect ratios and concentrations) measured in shearing flows.

Key Words: short fiber suspensions, fiber orientation, falling ball viscometry

INTRODUCTION

In a recent series of papers (Graham *et al.* 1987; Milliken *et al.* 1989a, b; Powell *et al.* 1989), we have shown that falling ball rheometry can be used to measure the viscosity of suspensions of randomly oriented rods. In this paper we describe experiments in which this same technique is used to determine the viscosity of suspensions of rods that are approximately aligned. We find that falling ball rheometry is a tool with which the effects of particle orientation can be isolated. As used in these experiments, the size of a test sphere is of the order of the characteristic length of the suspended particles. As it falls, it disturbs the original microstructure of the suspension only slightly, and it experiences an effective resistance (viscosity) which is that of the undisturbed or slightly disturbed suspension. This is in contrast to conventional rheological measurements using shearing flow in which interacting rods that are not subject to randomizing rotary Brownian forces tend to align; hence, shearing measurements are performed on suspensions having flow-induced anisotropy.

In the falling ball measurements performed to date, the suspensions were stirred prior to each experiment to ensure that the suspensions were isotropic. It is possible, however, to use the falling ball technique on oriented suspensions. That is, rather than stirring the suspensions to achieve randomness, one can hydrodynamically induce alignment in the suspension. Once the particles are aligned, the terminal velocity of a ball falling through the suspension is measured. This work is motivated by the observation that such alignment may mimic the flow-induced orientation found in rotational rheometers. Indeed, as we shall demonstrate, the average axial viscosity determined by falling ball rheometry compares favorably with measurements made using conventional techniques and is lower than that measured for randomly oriented suspensions.

Theoretical Considerations

The theoretical basis for falling ball rheometry is Stokes' formula relating the terminal velocity of a ball falling under gravity through a Newtonian fluid and the viscosity, μ, of the fluid (Stokes 1851; Happel & Brenner 1983):

$$v_{\mathrm{cor}} = \frac{g d_{\mathrm{ball}}^2 (\rho_{\mathrm{ball}} - \rho)}{18\mu}, \qquad [1]$$

where g is the acceleration of gravity, d_{ball} is the ball diameter, ρ_{ball} is the density of the ball and ρ is the density of the fluid.

We use the symbol v_{cor} to accentuate the difference between the measured velocity, v, and the velocity which is corrected for the additional drag due to wall effects (Milliken *et al.* 1989a, b). For balls sufficiently large relative to the mean suspended particle size (ball diameter/rod diameter $\geqslant 8$), the balls experience a Newtonian continuum: the extra drag on the balls due to the presence of the cylinder walls is predicted by a Faxén correction for a Newtonian fluid, and the corrected velocity is proportional to the square of the diameter of the ball. Note that even these "sufficiently large" balls can be small compared to the suspended particle size (ball diameter/rod length < 1). This Faxén correction (Faxén 1923), due to Bohlin (1960), is to $O(d_{\text{ball}}/D_{\text{col}})^6$

$$v_{\text{cor}} = \frac{v}{1 - 2.104\left(\frac{d_{\text{ball}}}{D_{\text{col}}}\right) + 2.09\left(\frac{d_{\text{ball}}}{D_{\text{col}}}\right)^3 - 0.95\left(\frac{d_{\text{ball}}}{D_{\text{col}}}\right)^5}, \qquad [2]$$

where D_{col} is the diameter of the cylindrical column containing the suspension. We will show that this Faxén correction also appears to be appropriate for the suspension of approximately oriented rods.

Falling Ball Rheometry for Suspensions

We have shown (Graham *et al.* 1987; Milliken *et al.* 1989a, b; Powell *et al.* 1989) that the average suspension viscosity measured by falling ball rheometry is reproducible even when relatively small balls are used. For example, for a suspension of rods having a nominal aspect ratio of 20 (31.65 mm long × 1.596 mm dia), measurements using balls of diameter > 0.3 times the rod length yielded an intrinsic viscosity that agrees well with predictions by Brenner (1974). These relatively small balls disturb the initial orientation only slightly. However, such a small ball must interact with a large number of suspended particles, sampling all orientations, in order for its average velocity to be accurately represented by [1] and [2], where μ is now the suspension viscosity. The ball also must travel over a sufficiently long length to average to a reproducible value over any inhomogeneities in the suspension. Because our sampling distance is fixed by the apparatus, we repeat the measurement of v_{cor} over this fixed distance several times with similar balls. Then we define the measured average viscosity of the suspension as

$$\mu_{\text{avg}} = \frac{1}{n}\sum \frac{g d_{\text{ball}}^2(\rho_{\text{ball}} - \rho)}{18 v_{\text{cor}}}, \qquad [3]$$

where n is the number of individual measurements for balls of similar diameter and mass.† Throughout this paper, we shall refer to μ_{avg} as simply the suspension viscosity, which will be symbolized by μ. In addition to μ, the viscosity of a suspension is often described by the relative viscosity, μ_r, which is the ratio of the viscosity of the suspension to the viscosity of the suspending fluid.

Suspensions of Rodlike Particles

Suspensions of rods and rodlike particles have recently received considerable attention. For suspensions in which Brownian forces are absent, Batchelor (1971) calculated the Trouton viscosity in uniaxial extensional flow in which the particles are completely aligned. He determined that for oriented suspensions, the dilute range could be identified by $\varepsilon n l^3 \ll 1$, where n is the number of particles per unit volume, l is one-half the length, L, of a rod, $\varepsilon = (\ln(2L/d))^{-1}$ and d is the rod diameter. The average spacing between particles, h, was found to be $h = (2nl)^{-1/2}$, and a theory for concentrated suspensions was proposed for $L \gg 2h \gg d$. The resulting formulas for the Trouton viscosity were shown by Batchelor (1971) to be consistent with the data of Weinberger & Goddard (1974) and were later found by Mewis & Metzner (1974) to describe their experimental results. For

†Note that the value of the viscosity using the average of $1/v_{\text{cor}}$ defined by [3] may be slightly different than the viscosity calculated using the inverse of the average of v_{cor}. This is only significant when the scatter in individual values of v_{cor} is large, although even then the difference is contained within the 95% confidence limits on the mean viscosity. The difference between the two averages is very small for the measurements presented in this paper.

rods subject to strong Brownian motion, Brenner (1974) obtained results for a wide variety of flows, which were specialized to the case of simple shearing. Results for slender bodies in strong flows (weak Brownian motion) were obtained by Leal & Hinch (1971). More recently, the interest has been in the behavior at higher concentrations. A recent theory of Berry & Russel (1987) examines the rheology of interacting slender rods. They include long-range interactions of rods subject to strong Brownian motion to obtain an expression for the viscosity which is valid to $O(n^2)$. Their theory holds for $\varepsilon nl^3 < 1$, and it may be seen as a bridge between theories for dilute suspensions where $\varepsilon nl^3 \ll 1$ and semi-concentrated theories where $\varepsilon nl^3 \gg 1$. Other works by Dinh & Armstrong (1984) and Bibbo *et al.* (1985) have attempted to use results from both the hydrodynamic theories (Batchelor 1971) and molecular theories (Doi & Edwards 1978a, b) to calculate the properties of suspensions of interacting rods on which only hydrodynamic forces act. Their model is limited in its range of concentrations to $L^{-3} < n < dL^{-2}$ for randomly oriented suspensions and $L^{-3} < n < Ld^{-2}$ for oriented suspensions.

Recent experimental results from conventional rheometrical tests are discussed in Milliken *et al.* (1989a), and for work prior to 1984 in Ganani & Powell (1984). One of the principal conclusions from these studies is that for suspensions of rigid rodlike particles, the effects of particle volume fraction, aspect ratio and orientation have yet to be clearly understood. For example, Milliken *et al.* (1989a) found that for suspensions of randomly oriented rods of a single aspect ratio, a_r, a dilute regime exists in which the relative viscosity is proportional to the volume fraction, ϕ, and a concentrated (or semiconcentrated) regime where $\mu_r - 1 \propto \phi^3$. For these $a_r = 19.83$ particles, the dilute regime extended to a volume fraction of approx. 0.12. In terms of the usual measure of concentration for rodlike particles, nL^3, the transition occurred at $nL^3 \approx 60$, which is surprisingly similar to the value predicted by recent theories of solutions of rodlike macromolecules (Keep & Pecora 1985; Magda *et al.* 1986; Bitsanis *et al.* 1988). The intrinsic viscosity calculated in the dilute regime compared to within 6% with the predictions of Brenner (1974) for dilute suspensions of randomly oriented rods. [The rigorous correspondence between measurements of the viscosity of suspensions of randomly oriented rods and Brenner's theory for suspensions subject to strong Brownian motion was made possible by the foundations laid by Haber & Brenner (1984).]

For all the data reviewed by Ganani & Powell (1984), it appeared that the aspect ratio rather than the volume fraction was the determining rheological parameter outside the dilute regime, although, as discussed subsequently by Ganani & Powell (1986), a more critical evaluation of the data suggests that experimental artifacts may be present in many of the reported data. However, the study of Powell *et al.* (1989) reinforces these observations on the importance of aspect ratio. Using particles with $a_r = 10$ and comparing our results with those of Milliken *et al.* (1989a) we showed that the aspect ratio has a dramatic effect on relative viscosity. For suspensions having $\phi = 0.05$, we found that the relative viscosity decreased from 2.37 to 1.45 when the aspect ratio was halved. In terms of the intrinsic viscosity, $[\mu]$, which can be defined through $\mu_r = 1 + [\mu]\phi$, this represents a decrease from $[\mu] = 27.6$, measured by Milliken *et al.* (1989a), to $[\mu] = 9$. These results were also predicted by Brenner (1974). In contrast, the recent work of Bibbo *et al.* (1985) appears to suggest that the particle aspect ratio may be of little importance. This would contradict our falling ball studies published to date (Milliken *et al.* 1989a; Powell *et al.* 1989) as well as recent work in our laboratory (Morrison 1989). Further Bibbo *et al.* (1985) claim that when using a parallel-plate rotational rheometer, the rheometer gap-to-particle length ratio needs only to be maintained at approximately one to ensure that instrumentation effects are eliminated. Our published falling ball studies strongly suggest that a characteristic container dimension (D_{col}) must be maintained at least 3 times larger than the particle length to allow free rotations of the rods as the ball sediments through the suspension and thereby eliminating geometrical effects. Further, our more recent studies (Morrison 1989) strongly suggest that a column 5 times larger must be used. These discrepancies may be a function of the alignment induced in shearing flow.

It is well-known that in pure straining motion, long rigid particles in suspension will align along the direction of the highest principal rate of extension (Batchelor 1971). In shearing flow, rods rotate at a rate proportional to the vorticity (Salem & Fuller 1985) producing periodic stresses in dilute suspensions (Ivanov *et al.* 1982a, b) which decay at long times due to slight deviations from an exactly monodisperse population of particles. Both flows produce particular realizations of the suspension microstructure which, on the average, determine the macroscopic stresses. For a given suspension, however, there are infinitely many realizations depending upon the details of the particle orientation distribution. Previously, we have used falling ball rheometry to probe a particular realization of the microstructure of suspensions of rods: the randomly oriented suspension. In this paper, we use falling ball rheometry to isolate the effects of particle orientation on the viscosity of suspensions of nonspherical particles and thereby develop a method for measuring the rheological properties of a particular realization of the microstructure. We use the same fluids and particles as those used previously. Now, however, rather than stirring to produce randomly oriented suspensions (Graham *et al.* 1987; Milliken *et al.* 1989a, b; Powell *et al.* 1989), we align the rods using hydrodynamic forces (Shaqfeh & Koch 1988). This is described in the Experimental section. In the following section, results for two suspensions, $\phi = 0.02$ and 0.5, are presented.

EXPERIMENTAL

General descriptions of the experimental apparatus have appeared elsewhere (Graham *et al.* 1987; Milliken *et al.* 1989a, b; Powell *et al.* 1989), and here we include details only as they apply to these measurements.

Suspensions were made using the same fluid/particle systems described by Milliken *et al.* (1989a). The particles (31.65 mm long × 1.596 mm dia polymethyl methacrylate rods) have an aspect ratio of 19.83 with a standard deviation of 0.73. These rods are rigid and they are sufficiently large that Brownian effects can be neglected (Milliken *et al.* 1989a). They are neutrally buoyant in the suspending liquid at a temperature of 20.6°C. The viscosity of the suspending fluids, as measured by falling ball rheometry, was found to be 11.15 Pa s at 20.4°C. All measurements affirm that the suspending fluid is Newtonian (Milliken *et al.* 1989a) in the shear rate range of interest in these experiments.

The experimental apparatus consisted of a glass column, 510 mm high × 46 mm dia. The column was placed in an insulated tank connected to a water bath and controller. The temperature was maintained at 20.6 ± 0.2°C, which was determined by trial-and-error to be the temperature at which no appreciable settling of rods occurred over a 24 h period. We recorded the temperature in the bath frequently during the experiments. Independently, we measured the viscosity of the suspending fluid as a function of temperature using falling ball rheometry. For each experiment, it was then possible to calculate the viscosity of the suspending fluid, and thereby, the relative viscosity.

The falling balls were brass ball bearings having nominal diameters of 12.70, 15.88 and 19.05 mm. These diameters were used because previous studies had shown that the viscosity of suspensions of randomly oriented rods could be measured if the relations $d_{\text{ball}}/d > 6$ and $d_{\text{ball}}/L > 1/3$ (Milliken *et al.* 1989b) were satisfied. The mass and diameter of each ball were determined to within ± 0.2 mg and ± 0.0025 mm, respectively. Prior to the experiments, the balls were placed in the water bath to thermally equilibrate to the temperature of the suspensions. The balls were dropped along the centerline of the column and measurements of the terminal velocity were made using an eddy current detector system (Powell *et al.* 1989).

The crucial difference between the current study and our earlier ones is the treatment of the suspensions between individual experiments. In our previous work, our objective was to determine the viscosity of suspensions of randomly oriented rods. Between each experiment, the suspension was stirred to ensure that the rods were randomly oriented and that the suspension was homogeneous. In the present study, we aligned the rods in the suspension prior to each experiment, while maintaining a uniform distribution of particles. This alignment was achieved hydrodynamically. After stirring the suspension, we passed a fixture, designed to align rods, along the axis of the cylinder through the suspension prior to each experiment. This fixture was an irregular grid, aligned parallel to the bottom of the cylinder, suspended on fine supports, and passed slowly up and down the cylinder through the suspension. A sketch of this fixture is shown in figure 1. The exact geometry was developed by trial-and-error, and it will likely change for rods of other diameters and lengths. The central idea behind this technique is that since the rods are neutrally buoyant and since, in the quiescent suspension, they are undisturbed by interparticle or Brownian forces, any alignment induced by flow will remain once the flow is stopped.

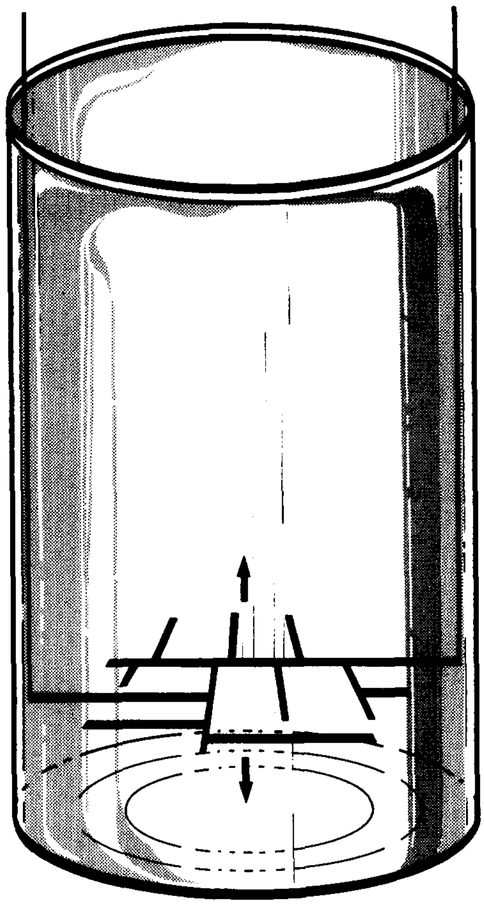

Figure 1. Sketch of the fixture used to align the rods.

The alignment induced by the fixture was checked by making the suspensions using both transparent and dyed rods. Except for their opacity, the dyed rods were identical to the transparent ones. Approximately 1 out of every 10 rods was opaque. After passing the aligning fixture through the suspension, we photographed the suspension for subsequent anaysis of the orientation distributions. A typical photograph is shown in figure 2. Usually each picture showed 25–30 dyed rods which could be used for orientation distribution analysis and for assurance that no significant concentration gradients resulted from the alignment procedure. We analyzed these photographs by making a composite of several photographs and formulating a probability density distribution (i.e. determining the probability of finding a rod in a particular orientation), using Givler's (1986) analysis [see also McGee (1982) and Lovrich & Tucker (1985)]. This analysis is based on the fact that the Fraunhofer diffraction pattern is identical to the Fourier transform of a pattern of oriented rods. If the original image serves as a two-dimensional diffraction grating, azimuthal profiles of the intensity of the diffraction pattern are proportional to the rods' orientation distribution function. Likewise, these profiles can be calculated from a Fourier transform of a high-contrast digital image, produced from a photograph of tracer rods. Digitally processed images composed of composites of several photographs were used in order to have over 100 rods in view, thereby giving a more accurate representation of the average orientation distribution. This analysis is, of course, only of the two-dimensional projection of our three-dimensional suspended particles. Nevertheless, we feel that it gives a strong indication of the orientation state of the particles.

Typical results, obtained in this way, for the orientation distribution of rods are given in figures 3 ($\phi = 0.02$) and 4 ($\phi = 0.05$). These distributions are normalized so that the total area under each curve is 1. The "r"-values represent the number of pixels, measured from the center of the Fourier transformed image. Uncertainties in the image analysis process lead to slight differences in the probability distributions for the various r-values. Generally, however, the peaks in the probability distributions are at an orientation angle of approximately $\pi/2$, which represents the angle for which

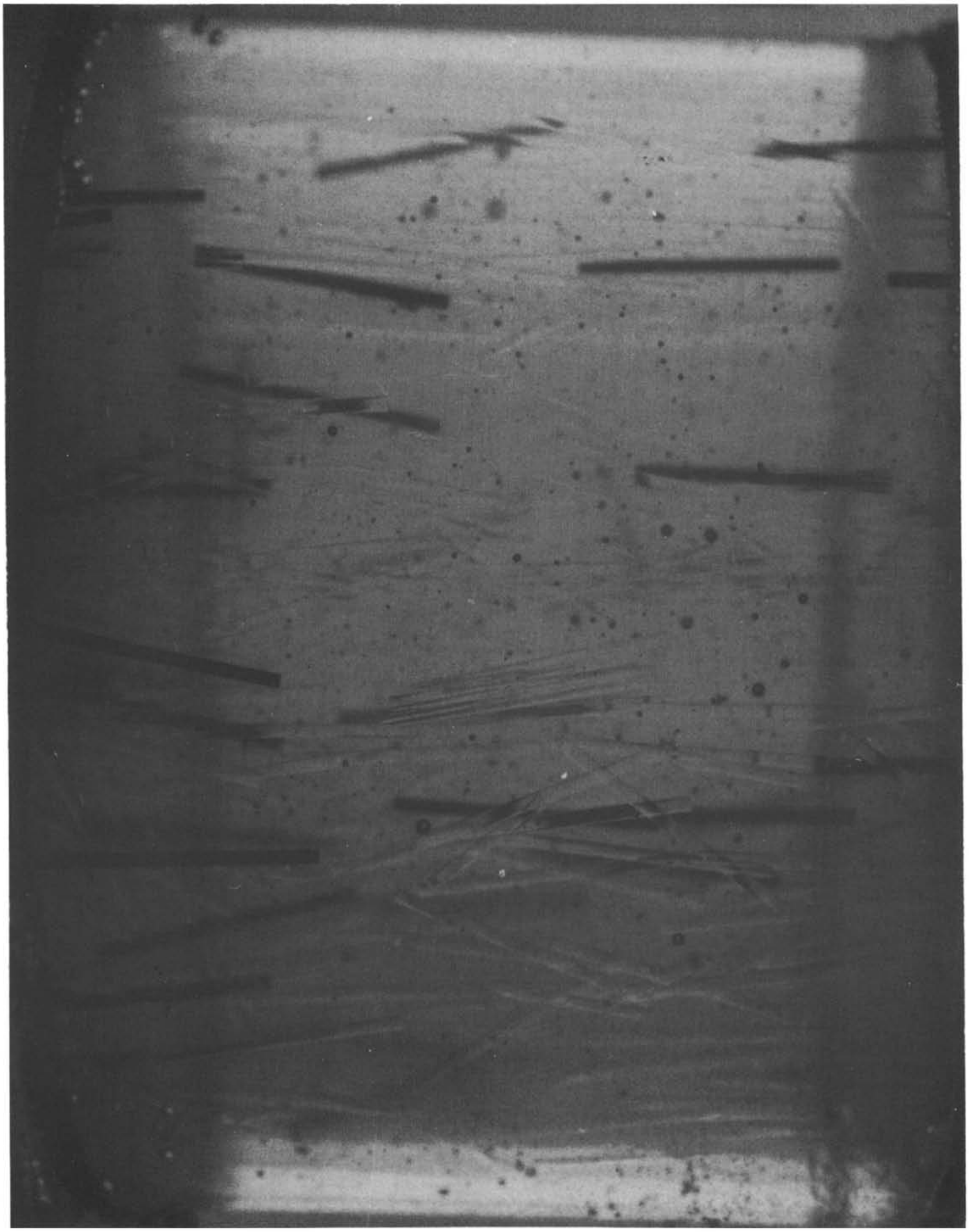

Figure 2. Typical photograph taken after aligning the suspended rods and prior to dropping a ball.

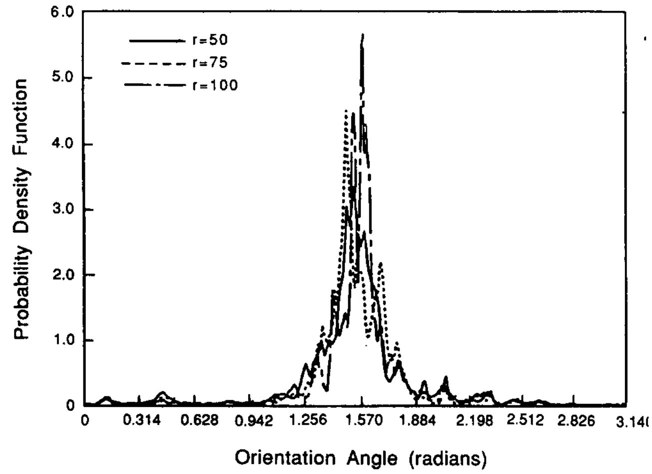

Figure 3. Probability density function of the orientation angle of the rods in a $\phi = 0.02$ suspension.

the rods are aligned along the gravity vector, and hence, along the path of the ball. Since other composite photographs gave analogous results, we concluded that the rods were predominantly aligned.

As with our previous studies, the velocity or viscosity we report is the average value obtained from several experiments. In between each measurement, the suspension is aligned. The photographs confirmed that each ball saw a different configuration of rods. Therefore, each measurement yielded a different value for the viscosity. These values did not correlate well with the degree of alignment determined with Givler's (1986) analysis on the individual photographs. This is not surprising, however, because the difference in the degree of alignment was small from photograph to photograph and Givler's analysis requires more rods to be in view than appeared in an individual photograph. The reported values were obtained by dropping, usually, 10 balls of the same size in each suspension. In our earlier studies, we generally found that data from 10 experiments would give a mean value of velocity or viscosity which would not change significantly if more experiments were performed (cf. Milliken *et al.* 1989a). This was confirmed in these studies, where we compared the mean values obtained using groups of 5 experiments and the overall average obtained using data from all 10 experiments and found no significant difference. Along with each average measurement, we report the 95% confidence interval (Johnson 1984) based on a standard Student t-distribution.

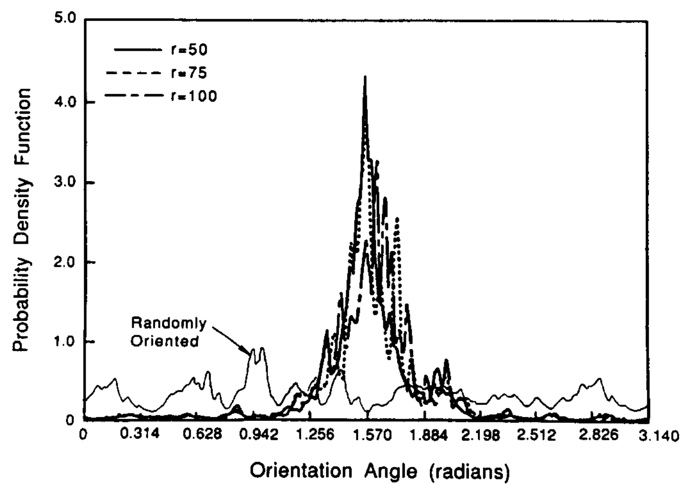

Figure 4. Probability density function of the orientation angle of the rods in a $\phi = 0.05$ suspension. Also shown is a probability density function for a suspension of randomly oriented rods at $\phi = 0.05$.

Table 1. Summary of data for suspensions of oriented rods

Volume fraction	Ball dia (mm)	No. of observations	Corrected velocity (mm/s)	Viscosity (Pa s)	Relative viscosity
0.02	12.7	10	49.42 ± 2.5	12.67 ± 0.50[a]	1.156 ± 0.044
0.02	15.88	9	73.90 ± 5.96	13.17 ± 0.81	1.192 ± 0.061
0.02	19.05	10	107.2 ± 4.6	13.12 ± 0.40	1.188 ± 0.039
0.02	**Composite**	**29**		$\mathbf{12.98 \pm 0.30}$	$\mathbf{1.178 \pm 0.025}$
0.05	12.7	10	37.01 ± 3.8	17.03 ± 1.21	1.542 ± 0.107
0.05	15.88	10	58.73 ± 7.03	16.69 ± 1.45	1.518 ± 0.127
0.05	19.05	10	86.9 ± 11.7	16.43 ± 1.61	1.489 ± 0.139
0.05	**Composite**	**30**		$\mathbf{16.71 \pm 0.70}$	$\mathbf{1.516 \pm 0.061}$

[a]Errors refer to 95% confidence limits.

RESULTS AND DISCUSSION

Table 1 gives the average velocity corrected for wall effects using [2], v_{cor}, for each ball diameter along with the 95% confidence limits on these data. Data for two volume fractions are reported, $\phi = 0.02$ and 0.05, and they are shown in figure 5 as v_{cor} vs d^2. The best-fit straight lines passing through the data and having the form $v_{cor} = \text{const} \times d^2$ are also shown in figure 5. These lines are consistent with [1] in that $v_{cor} \propto d^2$. Hence, these suspensions are behaving as Newtonian continua in the sense that the additional drag due to the container walls is given by theoretical results for Newtonian fluids; and [2] relates the average terminal velocity to the ball diameter. This behavior is the same as that observed for suspensions of randomly oriented rods (Graham *et al.* 1987; Milliken *et al.* 1989a, b; Powell *et al.* 1989). Therefore, it is possible to use [3] to calculate the viscosity of the suspension in such a way that the results are independent of the diameter of the test ball.

The viscosities of the suspensions, as calculated with [3], are given in table 1 and plotted in figure 6 as a function of the nominal ball diameter. As expected by the preceding discussion, to within the error of these experiments, the viscosity is independent of the ball diameter. No trend with ball diameter is observed, which indicates that ball size effects such as those found by Milliken *et al.* (1989b) are not present here. Also given in table 1 and shown in figure 6 as the solid horizontal lines are the average viscosities for each volume fraction, which were calculated using the data from all the ball sizes.

Figure 7 presents the principal result of this study, namely, the relative viscosity of the two suspensions of aligned rods as a function of the dimensionless ball diameter, d_{ball}/L. The solid horizontal lines in figure 7 are data obtained by Ganani & Powell (1986) for the relative viscosity of suspensions of rodlike particles, having $a_r = 25$, in shearing flow. The similarity is striking. The falling ball and shearing measurements yield very similar results, indicating that the mean orientation imposed in our experiments approximately corresponds to the mean orientation induced by the macroscopic flow in the shearing experiments. The difference in the rheological properties implied by this orientation is demonstrated by the data shown in figure 7 for the relative viscosities of suspensions of randomly oriented rods, as determined by falling ball rheometry

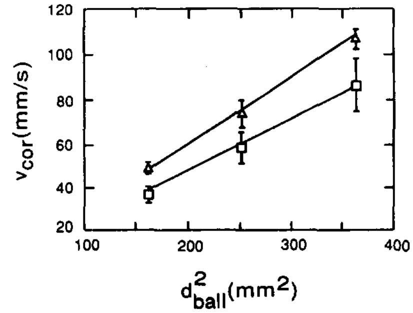

Figure 5. The average velocity corrected using [2], v_{cor}, as a function of the square of the ball diameter, d_{ball} for suspensions of oriented rods at $\phi = 0.02$ (△) and $\phi = 0.05$ (□). The lines represent the best-fits to [1].

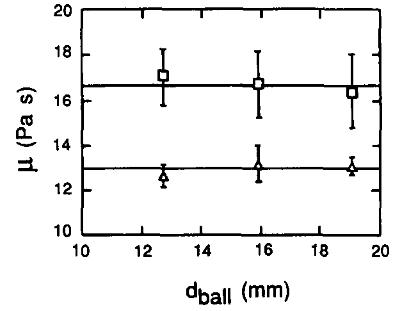

Figure 6. The average viscosity calculated using [3], μ, as a function of the ball diameter, d_{ball}, for suspensions of oriented rods at $\phi = 0.02$ (△) and $\phi = 0.05$ (□).

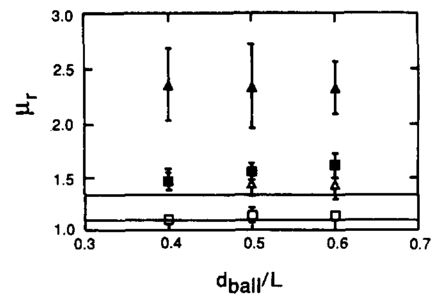

Figure 7. The relative viscosity, μ_r, vs dimensionless ball diameter, d_{ball}/L, for suspensions of oriented rods as measured using falling ball rheometry; $\phi = 0.02$ (□), $\phi = 0.05$ (△). Also shown are the relative viscosities of randomly oriented suspensions, $\phi = 0.02$ (■) and $\phi = 0.05$ (▲), and lines representing data obtained in shearing flows.

(Graham *et al.* 1987; Milliken *et al.* 1989a). At $\phi = 0.02$ the viscosity of the randomly oriented suspension is 30% larger than that of the aligned suspension, whereas for the $\phi = 0.05$ suspension it is nearly 60% larger. This difference is well above the uncertainties in the measurements. In terms of the specific viscosity, $\mu_{sp} = (\mu_r - 1)/\phi$, the difference is even more dramatic. The intrinsic viscosity of the randomly oriented suspension, which is the zero concentration limit of μ_{sp}, is 27.2. Table 2 shows that this value is over 250% higher than the specific viscosity of the oriented suspensions (≈ 10).

In our earlier studies of suspensions of randomly oriented rods (Milliken *et al.* 1989a; Powell *et al.* 1989), we found that, in the dilute range, the relative viscosity measured by falling ball rheometry compared quite well with Brenner's (1974) theory for the viscosity of suspensions of rodlike particles subject to strong Brownian motion. The justification for this comparison was based upon the work of Haber & Brenner (1984) who showed that the shear viscosity of a suspension of randomly oriented particles was the same as the shear viscosity of a suspension of particles subject to strong Brownian motion if the particle is a body of revolution. For suspensions of oriented rods, there appears to be no immediate connection between the results from falling ball rheometry experiments and existing theories. The most appealing choices for comparing our results are those theories which predict the viscosity for a suspension of rods which, in the mean, are aligned. In all such theories, however, the alignment is flow induced, with the viscosity increase due to the presence of the particles being caused by their disturbing the imposed macroscopic flow. No such flow is present in our experiments, and, hence, our comparisons must be viewed with some caution.

Batchelor's (1971) theory for the rheology of suspensions of rods provides estimates for the Trouton viscosity both for dilute and concentrated suspensions. Table 2 compares our results with the predictions of Batchelor (1971). We have obtained the values in table 2 using Batchelor's results for concentrated suspensions, which hold when $15.825 \gg h \gg 0.798$ mm, and we have divided the Trouton viscosity by 3. Values for h are given in table 2, where it is seen that the $\phi = 0.05$ suspension most closely fits within the "concentrated" category. The discrepancy between the measurements and the theory implies that our experiments are determining a property of aligned suspensions different from their resistance to straining motions. Our data can also be compared with the model of Bibbo *et al.* (1985). For aligned rods, their predictions are valid in the range $L^{-3} < n < Ld^{-2}$. In our case, this criterion is $3.1 \times 10^{-5} < n < 1.25 \times 10^{-2}\,\mathrm{mm}^{-3}$. Table 2 shows that both of our suspensions fit within these limits, and, it also gives the theoretical predictions which agree reasonably well with our results.

Table 2. Comparison between data for oriented suspensions and theoretical results

ϕ	n (mm^{-3})	$\epsilon n l^3$	h (mm)	μ_{sp}	Relative viscosity (1)[a]	(2)[a]	(3)[a]	(4)[a]	(5)[a]
0.02	3.2×10^{-4}	10.2	9.9	8.9	1.178	1.53	1.70	1.088	1.19
0.05	8×10^{-4}	25.5	6.3	10.3	1.516	2.46	2.15	1.22	1.34

[a](1) Suspensions of aligned particles (this study). (2) Randomly oriented suspensions (Milliken *et al.* 1989a). (3) Batchelor (1971). (4) Leal and Hinch (1971). (5) Bibbo *et al.* (1985).

Another class of theories with which it might be reasonable to compare our results are those which consider particles subject to weak Brownian motion. In shearing flow the particles tend to align along the streamlines, somewhat analogous to our oriented suspension. For spheroidal particles, Leal & Hinch (1971) give results for the intrinsic viscosity for $a_r = 20$, $[\mu] = 4.403$. This value is lower than ours by a factor of 2. Further, as discussed by Cox (1979), a comparison between results for spheroidal and cylindrical bodies should include a correction for the geometrical difference between the two particles. For rods having $a_r = 20$, the equivalent aspect ratio for a spheroid is 14.2 (Milliken *et al.* 1989a). The associated intrinsic viscosity would therefore be even lower than 4.403. This large discrepancy between our measurements and theoretical results for weak Brownian motion may result from the range of concentrations used in our studies. Leal & Hinch's (1971) theory is for dilute suspensions. By the classic measures, for example $\epsilon nl^3 \ll 1$, our suspensions are not dilute. However, as with the randomly oriented suspensions (Milliken *et al.* 1989a; Powell *et al.* 1989), we expect that the dilute range, as defined by a linear viscosity–volume fraction relationship determined using falling ball rheometry, may extend beyond the classically defined limits for oriented suspensions. Since we have data at only two concentrations, it is not possible to ascertain at this time whether such an assumption is valid.

CONCLUSIONS

Falling ball experiments in suspensions of rods which were hydrodynamically aligned prior to each experiment result in measured specific viscosities which are less than the specific viscosities of suspensions of randomly oriented rods by two-thirds. This result has been obtained for a single aspect ratio particle at two volume fractions. In both cases, our measurements agree well with measurements of the shear viscosity, made in shearing flow, of suspensions of fibers having a similar aspect ratio and the same volume fractions. As opposed to the case of randomly oriented rods, where a well-founded basis exists for comparing falling ball results with existing theories (cf. Milliken *et al.* 1989a), no such basis exists here. Available theories, do, however, lead to the conclusion that suspensions which experience flow-induced alignment during shearing have a lower viscosity than suspensions which stay random during flow (strong Brownian motion). These results further demonstrate the usefulness of falling ball rheometry as a tool for probing the macroscopic rheology of suspensions. They also raise several questions, including the effect of orientation on the concentration at which the dilute to semiconcentrated transition occurs, the effect of aspect ratio on the viscosity of oriented suspensions, and the directional dependence of the viscosity, all of which will be the subject of future studies.

Acknowledgements—This work was supported by grants from the National Science Foundation (CBT8718261) and the Division of Engineering and Geosciences of the Office of Basic Energy Science, U.S. Department of Energy (KC04106). The work at Sandia National Laboratories was sponsored by the U.S. Department of Energy under Contract DE-AC04-46DP00789. Those portions of this work accomplished at Los Alamos National Laboratory were sponsored by the U.S. Department of Energy under Contract W-7405-ENG-36. We also acknowledge the support of the Center for Nonlinear Studies at Los Alamos and the INCOR Program sponsored by the University of California. We are especially appreciative of the many helpful comments of Drs William J. Milliken and Rick Givler during the course of these studies.

REFERENCES

Batchelor, G. K. 1971 The stress generated in a non-dilute suspension of elongated particles by pure straining motion. *J. Fluid Mech.* **46,** 813–829.

Berry, D. H. & Russel, W. B. 1987 The rheology of dilute suspensions of slender rods in weak flows. *J. Fluid Mech.* **180,** 475–494.

Bibbo, M. A., Dinh, S. M. & Armstrong, R. C. 1985 Shear flow properties of semiconcentrated fiber suspensions. *J. Rheol.* **29,** 905–929.

Bitsanis, L., Davis, D. T. & Tirrell, M. 1988 Brownian dynamics of nondilute solutions of rodlike polymers. 1. Low concentrations. *Macromolecules* **21,** 2824–2835.

BOHLIN, T. 1960 On the drag on a rigid sphere moving in a viscous liquid inside a cylindrical tube. *Trans. R. Inst. Technol (Stockh.)* **155,** 1–63.

BRENNER, H. 1974 Rheology of a dilute suspension of axisymmetric Brownian particles. *Int. J. Multiphase Flow* **1,** 195–341.

COX, R. G. 1970 The motion of long slender bodies in a viscous fluid. Part I. General theory. *J. Fluid Mech.* **44,** 791–810.

DINH, S. & ARMSTRONG, R. C. 1984 A rheological equation of state for semi-concentrated fiber suspensions. *J. Rheol.* **28,** 207–227.

DOI, M. & EDWARDS, S. F. 1978a Dynamics of rod-like macromolecules in concentrated solution. Part 1. *Trans. Faraday Soc.* **2,** 560–570.

DOI, M. & EDWARDS, S. F. 1978b Dynamics of rod-like macromolecules in concentrated solution. Part 2. *Trans. Faraday Soc.* **2,** 918–932.

FAXÉN, H. 1923 Die Bewegung einer starren Kugel längs der Achse eines mit zäher Flüssigkeit gefüllten Röhres. *Ark. Mat. Astr. Fys.* **17,** 1–28.

GANANI, E. & POWELL, R. L. 1984 Suspensions of rodlike particles: literature review and data correlations. *J. composite Mater.* **19,** 194–215.

GANANI, E. & POWELL, R. L. 1986 Rheological behavior of rodlike particles in Newtonian and non-Newtonian fluids. *J. Rheol.* **30,** 995–1013.

GIVLER, R. C. 1986 Quantitative description of *in situ* fiber orientation state with image processing techniques. Report SAND86-0075, Sandia National Labs, Albuquerque, N.M.

GRAHAM, A. L., MONDY, L. A., GOTTLIEB, M. & POWELL, R. L. 1987 Rheological behavior of a randomly oriented suspension of rods. *Appl. Phys. Letts.* **50,** 127–129.

HABER, S. & BRENNER, H. 1984 Rheological properties of dilute centrally symmetric Brownian particles at small shear-rates. *J. Colloid Interface Sci.* **97,** 496–514.

HAPPEL, J. & BRENNER, H. 1983 *Low Reynolds Number Hydrodynamics.* Nijhoff, Boston, Mass.

IVANOV, Y., VAN DE VEN, T. G. M. & MASON, S. G. 1982a Damped oscillations in the viscosity of suspensions of rigid rods. I. Monomodal suspensions. *J. Rheol.* **26,** 213–230.

IVANOV, Y., VAN DE VEN, T. G. M. & MASON, S. G. 1982b Damped oscillations in the viscosity of suspensions of rigid rods. II. Bimodal and polydisperse suspensions. *J. Rheol.* **26,** 231–244.

JACKSON, W. C. & TUCKER, C. L. 1983 In *Conf. Proc. SPE ANTEC 83,* 2–5 May, pp. 751–759.

JOHNSON, R. 1984 *Elementary Statistics,* 4th edn. Duxbury Press, Boston, Mass.

KEEP, G. T. & PECORA, R. 1985 Reevaluation of the dynamic model for rotational diffusion of thin, rigid rods in semidilute solution. *Macromolecules* **18,** 1167–1173.

LEAL, L. G. & HINCH, E. J. 1971 The effect of weak Brownian rotations on particles in shear flow. *J. Fluid Mech.* **46,** 685–703.

LOVRICH, M. L. & TUCKER, C. L. 1985 Automated measurement of fiber orientation in short fiber composites. In *Conf. Proc. SPE ANTEC 85,* 29 Apr.–2 May, pp. 1119–1122.

MAGDA, J. J., DAVIS, H. T. & TIRRELL, M. 1986 The transport properties of rod-like particles via molecular dynamics. I. Bulk fluid. *J. chem. Phys.* **85,** 6674–6685.

MCGEE, S. H. 1982 The influence of microstructure on the elastic properties of composite materials. Ph.D. Dissertation, Dept of Chemical Engineering, Univ. of Delaware, Newark, Del.

MEWIS, J. & METZNER, A. B. 1974 The rheological properties of suspensions of fibres in Newtonian fluids subjected to extensional deformations. *J. Fluid Mech.* **62,** 593–600.

MILLIKEN, W. J., GOTTLIEB, M., GRAHAM, A. L., MONDY, L. A. & POWELL, R. L. 1989a The viscosity volume fraction relation for suspensions of randomly oriented rods by falling ball rheometry. *J. Fluid Mech.* **202,** 217–232.

MILLIKEN, W. J., MONDY, L. A., GOTTLIEB, M., GRAHAM, A. L. & POWELL, R. L. 1989b The effect of the diameter of falling balls on the apparent viscosity of suspensions of spheres and rods. *PhysicoChem. Hydrodynam.* **11,** 341–355.

MONDY, L. A., GRAHAM, A. L. & JENSEN, J. 1986 Continuum approximations and particle interactions in concentrated suspensions. *J. Rheol.* **30,** 1031–1051.

MORRISON, T. G. 1989 Experimental investigation of the mechanics of suspensions of elongated particles using falling-ball rheometry. M.S. Thesis, Dept of Chemical Engineering, Univ. of California, Davis, Calif.

POWELL, R. L., MONDY, L. A., STOKER, G. G., MILLIKEN, W. J. & GRAHAM, A. L. 1989 Development of a falling ball rheometer with applications to opaque systems: measurements of the rheology of suspensions of rods. *J. Rheol.* **33,** 1173–1188.

SALEM, A. J. & FULLER, G. G. 1985 Small angle light scattering as a probe of flow-induced particle orientation. *J. Colloid Interface Sci.* **108,** 149–157.

SHAQFEH, E. S. G. & KOCH, D. L. 1988 The effect of hydrodynamic interactions on the orientation of axisymmetric particles flowing through a fixed bed of spheres of fibers. *Phys. Fluids* **31,** 728–743.

STOKES, G. G. 1851 On the effect of the internal friction of fluids on the motion of pendulums. *Trans. Camb. phil. Soc.* **9,** 8–106.

WEINBERGER, C. B. & GODDARD, J. D. 1974 Extensional flow behavior of polymer solutions and particle suspensions in a spinning motion. *Int. J. Multiphase Flow* **1,** 465–486.