J. Fluid Mech. (1987), *vol.* 181, *pp.* 415–439
Printed in Great Britain

The shear-induced migration of particles in concentrated suspensions

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(Received 21 June 1985 and in revised form 3 February 1987)

In the course of viscometric measurements of concentrated suspensions of spheres in Newtonian fluids using a Couette device, Gadala-Maria & Acrivos (1980) observed a decrease in the suspension viscosity after long periods of shearing even though the viscosity of the pure suspending fluid remained constant under identical conditions. In the present work we demonstrate that this phenomenon is due to the shear-induced migration of particles out of the sheared Couette gap and into the fluid reservoir, which reduces the particle concentration in the gap and thereby the observed viscosity. We show further that this rate of viscosity decrease is consistent with a gap-limited shear-induced diffusion process normal to the plane of shear, with the relevant diffusion coefficient being proportional to $a^2\dot{\gamma}$, where a is the particle radius and $\dot{\gamma}$ is the applied shear rate.

Additional experiments also uncovered a new phenomenon – a short-term increase in the viscosity upon initial shearing of a suspension in a Couette device – which was attributed to the diffusive migration of particles across the width of the Couette gap and thus was used to infer values of the corresponding diffusion coefficient within the plane of shear parallel to gradients in fluid velocity.

In the theoretical part we demonstrate that the particle migrations that led to these observed phenomena may be explained in terms of the irreversible interparticle interactions that occur in these suspensions. From simple arguments, these interactions are shown to lead to effective diffusivities both normal to the plane of shear and normal to the direction of fluid motion within the plane of shear whose estimated magnitudes are comparable with those that were inferred from the experimental measurements. Furthermore, these interactions should induce, within a shear flow, particle drifts from regions of high to low shear stress, which are estimated to be of sufficient intensity to account for the observed initial viscosity increase mentioned above.

1. Introduction

During the course of their study of the rheology of concentrated suspensions of neutrally buoyant spheres in Newtonian fluids, Gadala-Maria & Acrivos (1980) reported the puzzling observation that, at very high solids concentrations $\phi > 0.40$, the effective viscosity of the suspensions, as measured in the conventional Couette viscometer depicted in figure 1, decreased slowly with prolonged shearing and eventually reached an equilibrium value which then remained time independent (cf. figure 2). The relative magnitude of this decrease was both quite significant (about

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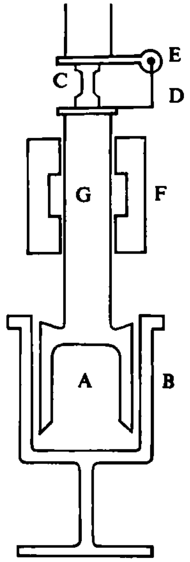

FIGURE 1. Schematic diagram of the R-17 Weissenberg Rheogoniometer torsion measurement apparatus. A bob, B cup, C torsion bar, D torsion arm, E transducer, F air bearing, G torsion shaft.

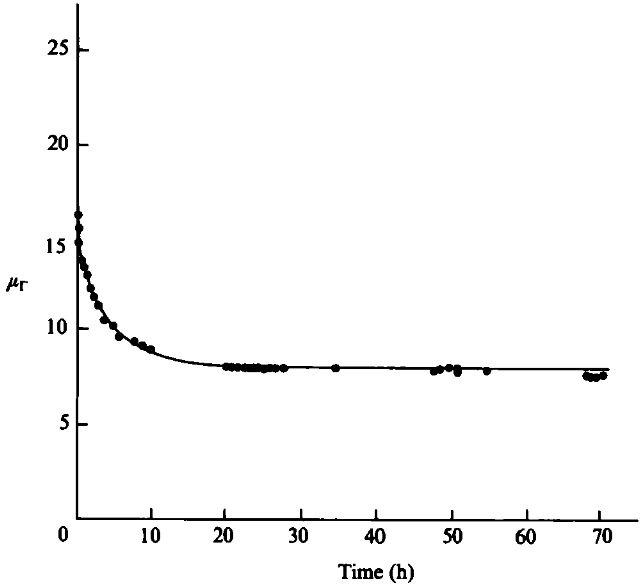

FIGURE 2. Relative viscosity of a $\phi = 0.45$ suspension as a function of the time that it had been sheared in the Couette device at $\dot{\gamma} = 24\ \mathrm{s}^{-1}$. Polystyrene spheres, 40–50 µm in diameter in a mixture of silicone oils (from Gadala-Maria 1979, figure 33).

a factor of 2 for 45 % suspensions) and a strong function of concentration. This drift in viscosity was quite unexpected since the suspending fluid was Newtonian and exhibited no degradation under experimental conditions, the particles were sufficiently large (40–50 µm) that Brownian or electroviscous forces would not be expected to play a significant role, and the total strain or fluid deformation associated with the viscosity decrease was very large – about 10^6. Those investigators were unable to account for this phenomenon.

We have repeated these experiments in our laboratory, and have shown that the observed decrease in viscosity resulted from a migration of particles out of the sample being sheared in the gap between the bob and the cup of the Couette device (cf. figure 1) and into the reservoir containing the stagnant part of the suspension. Clearly, such migrations would tend to decrease the particle concentration in the gap, and since in concentrated suspensions the viscosity is a very rapidly increasing function of ϕ, its measured value can be significantly affected by even small variations in the particle concentration. That this viscosity decrease was due to particle migrations was verified by sealing the bottom of the Couette gap with a layer of mercury, which thereby effectively prevented the escape of particles from the gap, and then repeating the experiments. It was found that, following a short equilibration period (shown to be associated with the sample loading procedure), the measured viscosity reached an equilibrium value, approximately equal to that measured initially in experiments with an unsealed gap but under otherwise identical conditions.

The mechanism responsible for this particle migration was, however, far from clear especially since the suspension both in the gap and in the reservoir could be safely assumed to have, initially, the same concentration. The most obvious possibility – settling due to gravity – was eliminated because the densities of the particles and of the suspending fluid were very closely matched, and in fact, if anything, the particles were shown to be very slightly lighter than the suspending liquid. Also, any migration due to inertia effects was ruled out by the low velocities and high viscosities in our experiments, which gave values of the particle Reynolds number $R = \dot{\gamma}a^2/\nu < 10^{-4}$ (with $\dot{\gamma}$ being the applied shear rate, a the particle radius, and ν the kinematic viscosity of the fluid), as well as by the observation that the equilibrium concentration in the gap (as inferred from the measured equilibrium viscosity) was independent of $\dot{\gamma}$. With these sources of drift having been eliminated, the observed particle migrations were therefore suggestive of the existence of some previously unsuspected mechanism.

In this paper we shall show that the effect discussed above can be explained as being due to a shear-induced particle drift arising from irreversible interparticle interactions. To begin with, we present experimental evidence to the effect that the migration of particles out of the Couette gap resulted from a diffusive process along the length of the gap. As a consequence, the observed rate of viscosity decrease was used to estimate the magnitude of the shear-induced diffusivity of a suspension normal to the plane of shear. A new phenomenon was also identified, in that when a suspension was sheared at a low shear rate just after loading, its effective viscosity was observed to increase over very short timescales (cf. figure 6), much shorter than those corresponding to the long-term viscosity decrease. Although it was not possible to directly measure the concentration profile in this case, the rate of viscosity increase was shown to be consistent with a mechanism of diffusive migration of particles across the gap, and thus was used to estimate the value of the shear-induced diffusivity within the plane of shear parallel to the gradient in fluid velocity. As will be described in detail, this initial viscosity-increase phenomenon is suggestive of the existence of a shear-induced particle migration from regions of high to low shear stress. This process of particle drift, whose strength was estimated from the magnitude of the initial viscosity increase, was also shown to be of sufficient intensity to explain the long-term viscosity-decrease phenomenon referred to earlier.

In §3 we propose a mechanism, based on the existence of irreversible interparticle interactions that occur as a suspension is sheared, which accounts for both the observed shear-induced diffusivity, and also for the inferred particle migrations from regions of high to low shear stress. First, we review the experimental evidence for the existence of irreversible interactions in concentrated suspensions of spheres and, from simple arguments, demonstrate that such interactions inevitably occur in sufficiently concentrated suspensions of real particles. We then demonstrate via a simple model how irreversible interactions must lead to migrations due to gradients in both concentration and applied shear stress. Although the complex nature of concentrated suspensions precludes any exact calculation of drift velocities, their estimated magnitudes are shown to agree closely with the values inferred from our experimental measurements.

The final section is devoted to a summary of our findings and contains suggestions for future work in this area.

2. Experiment

2.1. *Materials and experimental procedure*

All measurements were performed using the Couette geometry of a model R-17 Weissenberg Rheogoniometer, depicted in figure 1, with the samples to be analysed confined in the gap between the Couette bob A and cup B. Since our experiments involved only the determination of the relative viscosities of the suspensions, an accurate knowledge of the absolute magnitude of the suspension viscosity was not required and hence no attempt was made to precisely recalibrate the instrument. The rheometer was internally calibrated, however, by comparing the viscosity of glycerin as measured in the Couette device over shear rates from 0.24 s^{-1} to 240 s^{-1}.

Three Couette cups with inside diameters 4.8776 cm, 5.0020 cm and 5.2524 cm were employed. The bob was 4.5080 cm in height and 4.7498 cm in diameter which, when combined with the Couette cups, yielded gap widths of 0.639 mm, 1.261 mm and 2.513 mm respectively. The earlier experiments by Gadala-Maria & Acrivos used only the Couette cup with gap width 1.261 mm. The volumes of the gaps were 4.36 ml, 8.71 ml and 17.80 ml respectively. The volume of the reservoir beneath the bob was variable, and depended on the relative depth to which the bob was immersed in the cup; however, for the majority of our experiments, the distance of the base of the bob to the bottom of the cup was held at 1.2 cm, thus giving values for the fluid reservoir volume equal to 21 ml, 22 ml and 24 ml. The ratios $\hat{V}$ of the gap volumes to those of the reservoir were therefore 0.21, 0.40 and 0.74 for the small, medium and large Couette cups respectively. The tilt and lateral positions of the cups were aligned with the axis of rotation of the rheometer so that the eccentricity was less than ± 10 μm. The rheometer was modified to allow accurate positioning of the upper axis so that the eccentricity of the bob with respect to the axis of rotation was also less than ± 10 μm.

Experiments were conducted at room temperature (approximately 22 °C) which was closely controlled, thereby obviating the need to directly control the temperature of the Couette device. The suspensions being analysed were protected from any rapid variations in temperature by a 1 in. layer of foam insulation surrounding the Couette cup and also by the large thermal mass of the rheometer. The temperature of the fluid was monitored by a digital thermometer probe mounted on the outside of the Couette cup beneath the insulation and typically fluctuated by less than 1 °C during each experiment.

The spheres used in our experiments were obtained from the Diamond Shamrock

Corporation of Redwood City, CA and consisted of two lots of cross-linked polystyrene–divinylbenzene copolymer with optically determined sizes of 46.0 ± 2.6 μm and 87.0 ± 5.8 μm. The density of the polystyrene was reported by the manufacturer to be 1.051 g/cm^3. The smaller polystyrene spheres were reclaimed from the suspensions used in the original experiments by Gadala-Maria & Acrivos, while the larger were wet sieved $-140+170$ in methanol from a $-100+200$ lot obtained directly from the manufacturer.

Since the total available quantity of polystyrene spheres was very small, it was necessary to reuse the particles in order to perform a complete set of experiments. Thus, after a suspension had been analysed in the viscometer, it was collected and rinsed with acetone to remove the suspending fluid. The particles were then placed in a medium fritted glass filter funnel (15 μm pore size) and rinsed at least three times with acetone to remove the last traces of the suspending fluid. The particles were then washed a minimum of three times with methanol to remove the acetone, and finally were allowed to dry overnight under aspiration. Before preparing a new suspension, the spheres were passed through a 180 μm sieve to break up any large clumps of particles.

The procedure of rinsing the particles with acetone and then drying them was not found to have any discernible effect on the 46 μm spheres; however, unless great care was taken to eliminate all of the acetone, a density loss of up to 0.05 g/cm^3 was noted with the 87 μm spheres. This decrease in density was attributed to an expansion of the spheres in the presence of acetone and could easily be estimated by measuring the rise velocity of the spheres in a suspending fluid that matched the density of the untreated spheres. Further experimentation showed that if the 87 μm spheres were dried for a longer period – about 3 days – then the density decrease could be confined to less than 0.01 g/cm^3.

The polystyrene spheres were suspended in a mixture of 11.83 wt % Dow Corning 556 fluid and 88.17 wt % Dow Corning 550 fluid (both fluids were polyphenylmethylsiloxanes). Two different lots of the latter fluid were used. The fluid that was prepared with lot BFK671 had a viscosity of 1.22 P and density of 1.051 g/cm^3 at 22 °C, whereas that prepared with lot AE043852 had a viscosity of 1.07 P and the same density.

Suspensions of spheres were loaded into the Couette device by first pouring them into the cup and then lowering the bob until the fluid was displaced, filling the gap. This flow of the suspension up the gap is akin to a channel flow behind an advancing meniscus and as such leads to the formation of a region of increased particle concentration immediately behind the meniscus (cf. Karnis & Mason 1967). The presence of this meniscus layer was found to have a significant effect on the observed viscosity, thus care was taken in our experimental procedure to flow sufficient extra fluid through the gap that the meniscus layer was eliminated. The implications of this meniscus effect, and other related phenomena arising from particle migrations, for the measurement of the shear viscosity of concentrated suspensions are discussed in more detail elsewhere (Leighton 1985). The viscosity of the suspension corresponding to the concentration initially loaded, which was required in interpreting the viscosity increase and decrease data, was taken to be that observed after the short-term viscosity increase had reached equilibrium, but before the long-term viscosity decrease became significant. This was easily accomplished, since the total fluid deformation over which each migration occurred was widely different.

Viscosity measurements made in this way were quite reproducible for each suspension in a particular gap width; however the values measured for the 87 μm spheres were somewhat different than those for the 46 μm spheres, and both were a function of the gap width and shear rate. The viscosity data were best represented by an Eiler's equation:

$$\frac{\mu}{\mu_0} = \left[1 + \frac{\frac{1}{2}[\mu]\phi}{1-\phi/\phi_{\max}}\right]^2, \tag{2.1}$$

where $[\mu]$ is the intrinsic viscosity and $\phi_{\max}$ the maximum particle concentration. The fitted values of these parameters were functions of particle size, gap width and shear rate; thus separate correlations were used in examining experiments where these varied. Using the values measured in all three gap widths and extrapolating to a zero ratio of particle diameter to gap width, the best-fit values for the suspensions of 46 μm polystyrene spheres sheared at 24 s^{-1} were found to be $[\mu] = 3.0$, $\phi_{\max} = 0.58$.

2.2. *Long-term viscosity-decrease experiments*

As described in §1, when concentrated suspensions of neutrally buoyant polystyrene spheres were sheared in a Couette device, the viscosity was found to decrease over long periods of time, this drift being eliminated when the base of the gap was sealed with a layer of mercury. It was concluded, therefore, that this effect was due to the migration of particles out of the sheared gap into the fluid reservoir.

In our experimental work, no attempt was made to directly measure the suspension concentration distribution in the Couette device owing to insurmountable experimental difficulties. Instead, this distribution was inferred from the measured apparent viscosity of the suspension when sheared which, as is well known, is a very sensitive function of concentration for concentrated suspensions and therefore can provide an accurate estimate of the changes in concentration.

2.2.1. *A diffusion model for the observed long-term viscosity decrease*

In this section we present a diffusion model that relates the observed viscosity of the suspension to the concentration distribution in the gap. We assume that, initially, the concentration throughout the gap has the uniform value ϕ_0 (that initially loaded into the gap) and that, upon shearing, the geometry of the flow at the base of the gap induces in some way a migration of particles out of that region and into the reservoir. Of course, this outward migration is, at steady-state, balanced by a diffusive drift back toward the gap, but it is likely that this equilibration between the concentration at the base of the gap and that in the reservoir occurs over a lengthscale comparable with the gap width. Thus, since the gap width was much smaller than the gap length, the characteristic time for equilibration at the base of the gap should have been much less than that associated with migration along the length of the gap, with the result that the primary limitation to migration into the reservoir is the migration of particles along the gap length.

We therefore model this process by assuming that the concentration at the base of the gap is in equilibrium with that in the reservoir at all times. We further suppose that the migration of particles through the length of the gap is controlled by diffusion, which implies that any secondary currents that may exist in the gap are assumed negligible.

The diffusion process as modelled above will be governed by the diffusion equation

$$\frac{\partial \phi}{\partial t} = \frac{\partial}{\partial z}\left(D_{\perp}\frac{\partial \phi}{\partial z}\right), \quad 0 < z < h \tag{2.2}$$

where the base and top of the gap are denoted respectively by $z = 0$ and $z = h$, and $D_\perp$ (the diffusivity normal to the plane of shear) is, of course, a function of concentration. Our assumption of a uniform initial concentration everywhere except at the base of the gap yields the initial condition

$$\phi|_{t=0} = \begin{cases} \phi_0, & 0 < z \leqslant h, \\ \phi^*, & z = 0, \end{cases} \tag{2.3}$$

where ϕ^*, the value of which will be inferred from the experimental data, is the concentration at the base of the gap in equilibrium with ϕ_0, the initial concentration in the reservoir. A reasonable boundary condition at $z = h$ is simply the zero-flux condition:

$$\left.\frac{\partial \phi}{\partial z}\right|_{z=h} = 0,$$

while our assumption that the reservoir is in equilibrium with the concentration at the base of the gap yields that, at $z = 0$,

$$\left.\frac{\partial \phi}{\partial t}\right|_{z=0} = \frac{\hat{V} D_\perp}{h} \left.\frac{\partial \phi}{\partial z}\right|_{z=0}, \tag{2.4}$$

where $\hat{V}$ is the ratio of gap to reservoir volume.

In writing boundary condition (2.4) we have taken into account the increase of the reservoir concentration as the particles migrate out of the gap by assuming that the concentration in the reservoir is homogeneous. This, of course, requires that the fluid reservoir be well mixed on a timescale that is short compared to the characteristic time for diffusive migration along the length of the gap. This appears to be a reasonable assumption because the geometry of the Couette reservoir is such that the rotation of the cup will induce an inertially driven secondary current analogous to that found in a parallel-plate geometry. Although it is not possible to ascertain the strength of this flow without a detailed numerical analysis of this complex geometry, we estimate from dimensional scaling arguments a mixing timescale of about 10 min, which is much shorter than the observed diffusion time of about 100 min for typical experimental parameters. In any event, the assumption of a well-mixed reservoir appears to agree well with our experimental results.

The boundary condition (2.4) also assumes that the rate of increase in the equilibrium concentration at the base of the gap is equal to that in the reservoir. This should be a good approximation since the total change in concentration in the reservoir, being proportional to the volume ratio $\hat{V}$, was of the order of 3% of the initial concentration for our experiments. Of course, if the volume of the reservoir is infinite ($\hat{V} = 0$), then, according to our model, the concentration at $z = 0$ would remain constant throughout the migration process, and hence the effect of a finite value of $\hat{V}$ is to increase the rate at which the concentration distribution in the Couette device will approach equilibrium over that obtained for an infinite reservoir.

We shall further simplify our model by treating the diffusion coefficient as constant throughout the migration process. Although, as we shall show, $D_\perp$ is a strong function of concentration, this approximation is reasonable provided the total variation in concentration in the gap is small. In any case the solution of (2.2), when fitted to our experimental results, will yield some average diffusion coefficient which should correspond to that of the average concentration in the gap during the migration. Further discussion of the effect on our results of assuming a constant diffusivity will be deferred until §2.2.2(*e*).

Subject to the assumed boundary conditions, the solution to (2.2) is then given by

$$\phi(z,t)=\phi^*+(\phi_0-\phi^*)\left[\frac{\hat{V}}{1+\hat{V}}+\sum_{n=1}^{\infty}\frac{2\exp(-\lambda_n^2 D_\perp t/h^2)}{(\lambda_n^2+\hat{V}(1+\hat{V}))}\left(\lambda_n \sin\frac{\lambda_n z}{h}-\hat{V}\cos\frac{\lambda_n z}{h}\right)\right], \tag{2.5}$$

where the eigenvalues λ_n satisfy

$$\tan\lambda_n+\frac{\lambda_n}{\hat{V}}=0. \tag{2.6}$$

The decrease in concentration in the Couette gap given by (2.5) induces a decrease in the observed viscosity of the suspension, which is simply the average viscosity in the gap:

$$\mu_{\text{obs}}=\frac{1}{h}\int_0^h \mu\,\mathrm{d}z \tag{2.7}$$

where, when fitting the model to our experimental data, the dependence of the viscosity on concentration was determined from the initial viscosity measurements as mentioned earlier.

Since the initial and equilibrium values of the concentration in the gap may be determined from the initial and equilibrium values of the observed viscosity, the rate at which the viscosity approaches equilibrium is governed solely by the diffusion coefficient $D_\perp$. We may thus match the viscosity decrease predicted by (2.5) and (2.7) to our experimental data to obtain an estimate of the diffusion coefficient $D_\perp$ and of the equilibrium depletion $\phi_0-\phi^*$ for our suspensions.

2.2.2. *Experimental results*

We have conducted long-term viscosity-decrease experiments with suspensions of 46 and 87 μm polystyrene spheres using the small, medium and large Couette gaps. Experiments were performed at initial concentrations from 30 % to 50 % and at shear rates from 7.6 to 240 s^{-1}.

(*a*) *General features*

In the experiments with the small and medium Couette gaps, it was found that the model gave an accurate description of the decrease of the viscosity with shearing and yielded values of the diffusion coefficient that were quite reproducible. Figure 3 presents the observed viscosity as a function of time together with the fitted curve from the model for a typical experiment, that of a 45% suspension of 46 μm polystyrene spheres in silicone oil using the medium Couette gap at a shear rate of 24 s^{-1}. The diffusion coefficient was found to be proportional to $\dot{\gamma}a^2$, as was expected for shear-induced diffusion, and to be a strong function of concentration, increasing from a dimensionless value of about 0.2 for a 30% suspension to 5 for a 50% concentration. Figure 4 presents the observed dimensionless diffusion coefficient $\hat{D}_\perp \equiv D_\perp/\dot{\gamma}a^2$ for the polystyrene-sphere suspensions as a function of the average concentration during the migration using the data taken with the small and medium Couette gaps.

The equilibrium concentration depletion, calculated from the observed initial and equilibrium values of the suspension viscosity, was also found to be quite reproducible. The depletion was an increasing function of $\hat{a}$, the ratio of particle diameter to gap width, and, more surprisingly, was approximately independent of concen-

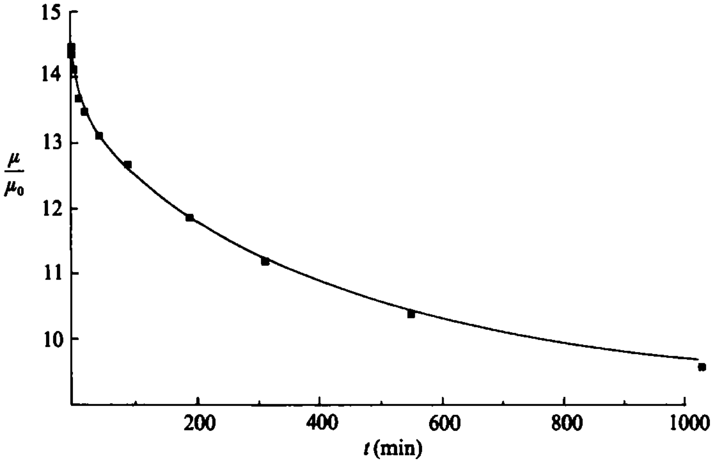

FIGURE 3. Long-term viscosity decrease. Data are for 46 μm polystyrene spheres in silicone oil, 45 % initial concentration. The suspension was sheared at 24 s^{-1} in the 1.261 mm Couette gap. Model parameters were $\phi_0 - \phi^* = 0.0490$, $D_\perp = 1.50$.

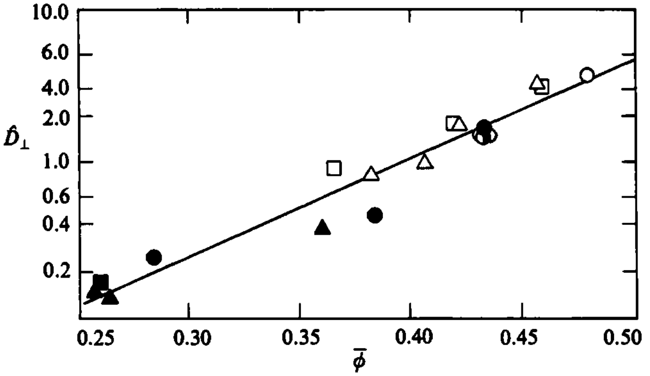

FIGURE 4. Diffusion coefficients calculated from the long-term viscosity decrease experiments: □, 46 μm polystyrene in 0.639 mm gap; ○, 46 μm polystyrene in 1.261 mm gap; △, 87 μm polystyrene in 1.261 mm gap. Shear rates were: filled symbols, 76 s^{-1}; open symbols, 24 s^{-1}; half-filled symbols, 7.6 s^{-1}.

tration. Figure 5 presents the observed values of the equilibrium depletion for 30 % and 45 % suspensions of both large and small polystyrene spheres as a function of the size ratio $\hat{a}$. We shall now discuss in more detail the dependence of this viscosity decrease on the experimental parameters.

(b) *Effect of gap height and gap width*

Two critical assumptions in deriving (2.5) were that the primary factor controlling the rate of particle migration out of the gap was their rate of migration throughout the length of the gap and, further, that this migration could be modelled as a diffusion process. We have tested the hypothesis that the migration is gap limited by observing the effect of varying the gap height h on the rate of the viscosity decrease. Two experiments were conducted: one with the bob fully immersed in the Couette cup and the other with the bob immersed to only half its length. The rate of decrease

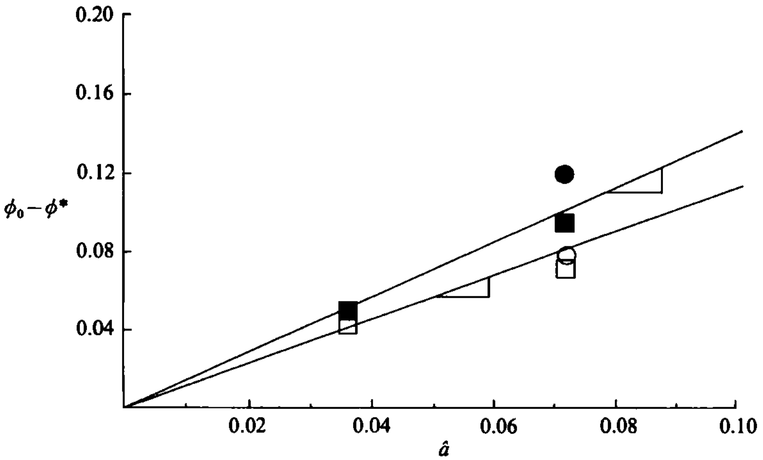

FIGURE 5. Equilibrium concentration depletion at the base of the Couette gap: □, 46 μm polystyrene; ○, 87 μm polystyrene. Solid symbols, $\phi_0 = 0.30$; open symbols, $\phi_0 = 0.45$.

in the observed viscosity in the half-height experiment was roughly four times that when the bob was fully immersed, indicating that the timescale for the migration was proportional to h^2 as had been expected from (2.5). If, on the other hand, the rate of migration out of the gap had been limited by a process of equilibration with the fluid reservoir at the base of the gap rather than by diffusion along the length of the gap, then the rate of viscosity decrease would have been found to be proportional to the total quantity of particles in the gap and thus linear in the gap height. We may conclude, therefore, that the migration was gap limited.

The second hypothesis, the assumption that the migration through the length of the Couette gap could be modelled as a diffusion process, was tested by varying the gap width and observing the rate of decrease in viscosity. Thus, if, for example, the migration through the gap had been controlled by secondary currents, rather than by diffusion, we should have expected a large effect from varying the gap width, since the intensity of these currents should be a strong function of this parameter. But, as may be seen from the data in figure 4, the experiments that we performed with the small and medium gaps yielded essentially identical diffusion coefficients; thus we conclude that for these gap widths secondary currents did not play a significant role. In the experiments with the largest gap, however, the calculated diffusion coefficients were a factor of as much as three greater than those for the same suspensions when measured in the smaller gaps, thus it is possible that secondary currents were becoming important in this case. This should not have been very surprising since, for the large cup, the ratio of the gap width to the Couette cup radius was only 1 in 10 and the ratio of the gap width to the distance between the base of the bob and the bottom of the cup was only 1 in 4.6. In contrast, for the smallest cup, corresponding ratios were 1 in 40 and 1 in 18.4 respectively.

No effect on the equilibrium depletion in concentration was observed for variations in the gap height, except that which could be accounted for by variations in $\hat{V}$, the ratio of gap volume to reservoir volume. Of course, as $\hat{V}$ becomes larger, the total change in concentration in the gap decreases since, as particles migrate out of the gap, the reservoir concentration increases, thereby also increasing the equilibrium concentration at the base of the gap. This effect has been taken into account in deriving (2.5), however, and we find from our experiments that, as would have been anticipated, $\phi_0 - \phi^*$ remained independent of the gap height.

(*c*) *Effect of shear rate*

In all cases, the time constant for the observed viscosity decrease was found to be inversely proportional to the applied shear rate in that experiments at shear rates of 7.6 and 76 s^{-1} yielded essentially the same dimensionless value for the diffusion coefficient $\hat{D}_\perp$. On the other hand, when suspensions of 46 μm polystyrene spheres were sheared in the medium Couette gap at 240 s^{-1}, a very strange phenomenon was observed. Instead of being time dependent as had been expected, the viscosity was found to remain approximately constant in time. Moreover, if the shear rate was then reduced to 76 s^{-1}, the viscosity was observed to decrease rapidly (the transient conforming to the diffusion model with the same value of $\hat{D}_\perp$ found at the other shear rates), but if the shear rate was raised again to 240 s^{-1}, the viscosity increased to approximately its initial value.

The most likely explanation for the lack of viscosity decrease at high shear rates is that the equilibrium normally attained between the fluid at the base of the gap and the reservoir was disrupted in some way, perhaps due to the onset of an instability or to an inertially driven secondary current. Since the equilibrium is most likely established over a region that is of the order of the gap width (and thus is much smaller than the gap height) the flow in this narrow region need not have significantly affected the measured suspension viscosity. Except for this effect, the equilibrium-concentration depletion in the device was independent of the magnitude of the applied shear rate.

(*d*) *Effect of particle size*

As was mentioned earlier, the diffusion coefficient associated with the viscosity decrease was found to be proportional to the square of the particle radius, and thus the dimensionless diffusion coefficient was independent of a, at least in experiments with the medium Couette gap. During experiments with suspensions of 87 μm spheres in the small Couette device, however, a dimensionless value of the diffusion coefficient a factor of as much as two greater than that obtained for the 46 μm spheres was found. Although the cause of this discrepancy was not explicitly determined, it was attributed to the large size of the spheres relative to the gap width (about 1 in 7). For such large values of the ratio $\hat{a}$ it was very difficult to remove the meniscus layer entirely from the gap, thereby increasing the scatter in both the observed initial viscosity and the diffusion coefficient.

(*e*) *Effect of concentration*

The diffusion coefficient was found to be a very strong function of concentration, approximately paralleling the increase in suspension viscosity with concentration. At the higher concentrations, the variation of viscosity with concentration was such that even a very small change in the concentration in the gap could easily be detected. At lower concentrations, however, the dependence of viscosity on concentration was not as great, thus the influence of experimental errors in the viscosity measurement on the determination of $D_\perp$ were much more significant, especially when combined with the long times necessary for these suspensions to reach equilibrium. Consequently, the values of $D_\perp$ at the lower concentrations are somewhat uncertain.

Due to the strong dependence of the diffusivity on concentration, it is appropriate to re-examine the error that is introduced in deriving (2.5) by assuming that the

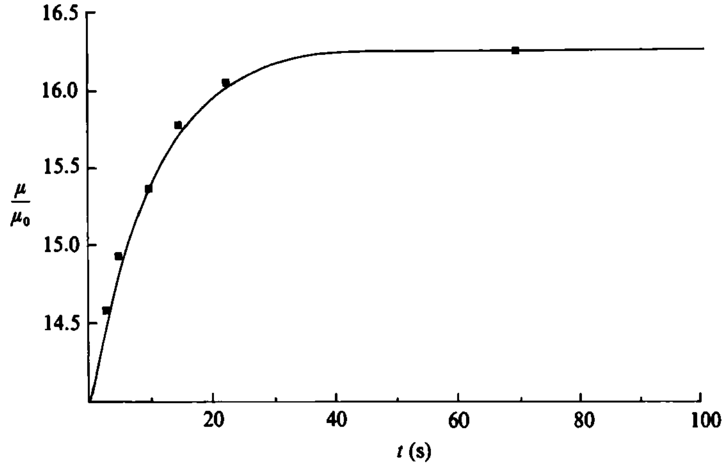

FIGURE 6. Short-term viscosity increase with shearing, 46 μm polystyrene suspension. Model parameters were: $\bar{\phi} = 0.45$, $\dot{\gamma} = 2.4\ \mathrm{s}^{-1}$, $\hat{D}_{\parallel} = 1.8$, $B_1 = 0.062$.

diffusion coefficient is constant throughout each experiment. As a typical case, consider the data depicted in figure 3 for the 46 μm spheres in the 1.261 mm Couette gap at an initial concentration $\phi_0 = 0.45$. For this experiment, the diffusion coefficient $\hat{D}_{\perp}$ was estimated to equal 1.5 and the concentration at steady state (e.g. the concentration in the gap at the end of the experiment) was $\phi = 0.415$. Note that this differs from ϕ^*, the concentration at the base of the gap in equilibrium with the initial concentration in the reservoir, owing to the increase in the reservoir concentration arising from the migration of particles out of the gap. On the other hand, from the observed dependence of $\hat{D}_{\perp}$ on concentration shown in figure 4, the estimated diffusivities at the initial and steady-state concentrations equal 1.8 and 1.2 respectively, a variation of ±20 % from the estimated constant value, which is not sufficient to significantly affect the profile of the viscosity decrease with time. In fact, errors due to assigning the estimated diffusion coefficient to the average concentration during the migration are likely to be less than 10 % in this instance, and represent an insignificant source for the scatter in the experimental results shown in figure 4.

2.3. *Short-term viscosity increase*

When measuring the viscosity of a concentrated suspension of polystyrene spheres in silicone oil, it was found that upon shearing the sample from rest the viscosity would increase rapidly over a short time interval and achieve some apparent steady-state value (cf. figure 6). Of course, if the suspension was sheared long enough, the long-term migration of particles out of the gap led to a decrease in the observed viscosity; however, since in our experiments the viscosity increase reached equilibrium after a total strain of about 10^2 while the viscosity decrease did not become apparent until after a strain of about 10^3, these two phenomena were well separated in the strain appropriate to each migration. We shall now attempt to model this phenomenon.

2.3.1. *Diffusion model for the observed short-term viscosity increase*

The observed timescale appropriate to the initial viscosity-increase phenomenon was found to be inversely proportional both to the shear rate $\dot{\gamma}$ and to the square of ratio of the particle diameter to gap width $\hat{a}$; thus this effect could be most easily explained in terms of a shear-induced migration of particles across the width of the Couette gap analogous to the long-term viscosity-decrease phenomenon just examined. This could only be the case, however, if the suspension flowing into the gap during the loading procedure acquired a concentration distribution across the gap that was different from the equilibrium profile corresponding to Couette flow. Then, upon shearing, this particle distribution would diffuse into that appropriate for the Couette flow, and thereby induce a change in the observed viscosity.

To examine this effect in more detail, we first note that if the concentration profile is only slightly non-uniform and is assumed to be symmetric about the centreline of the gap, then the observed viscosity will be given by

$$\frac{\mu_{\mathrm{obs}}}{\bar{\mu}}-1=\frac{1}{\bar{\mu}}\left[\frac{1}{b}\int_0^b\frac{1}{\mu}\,\mathrm{d}y\right]^{-1}-1=-\left[\left(\frac{1}{\mu}\frac{\mathrm{d}\mu}{\mathrm{d}\phi}\right)^2-\frac{1}{2}\frac{1}{\mu}\frac{\mathrm{d}^2\mu}{\mathrm{d}\phi^2}\right]\Bigg|_{\bar{\phi}}\frac{1}{b}\int_0^b(\Delta\phi)^2\,\mathrm{d}y+O(\langle(\Delta\phi)^3\rangle), \tag{2.8}$$

where we have simply expanded the viscosity $\mu(\phi)$ in a Taylor series about $\phi=\bar{\phi}$. In the above, $\bar{\mu}$ is the viscosity that corresponds to a uniform concentration $\bar{\phi}$, $\Delta\phi=\phi(y)-\bar{\phi}$ is the deviation from the average concentration across the gap, $y=0$ denotes the centreline of the gap, and $y=\pm b$ the walls. Thus, as expected, the variation in viscosity is proportional to the average value of $(\Delta\phi)^2$ across the gap for small fluctuations in concentration.

The viscosity function on the right-hand side of (2.8) may be calculated from the observed dependence of viscosity on concentration (cf. (2.1)). When this is done, it is seen that any non-uniformity in concentration across the gap leads to a decrease in the observed viscosity, the magnitude of this decrease being a strongly increasing function of $\bar{\phi}$. Thus, from the observation that the viscosity increases after shearing, in the manner of a diffusive process, we may conclude that the initial concentration distribution after the sample loading is more non-uniform than that corresponding to Couette flow.

It is instructive to consider at this point possible mechanisms by which this hypothesized initial non-uniform concentration distribution was established. We first recall that the suspension to be analysed was loaded by lowering the bob into the Couette cup, thereby inducing a flow that consisted of a converging flow entering the narrow gap followed by channel flow up the gap. The non-uniformity of concentration inferred from our measurements could of course be created in either of these two regions; however since the gap length was much greater than its width (a ratio of about 70:1 for the narrow gap) it seems likely that it was created by the channel flow, with inhomogeneities attributable to the entrance flow being confined to a narrow region at the base of the gap. Thus, if this picture is correct, the initial viscosity increase should result from a difference in the equilibrium concentration distribution that is achieved in a channel and in a Couette flow in the same geometry. But because the particle-to-gap-width ratios were the same in both cases, wall effects should not have been responsible for this difference. Similarly, the channel Reynolds number of the flow was sufficiently low ($2\bar{u}b/\nu=O(10^{-1})$, where $\bar{u}$ is the average velocity and b the gap width) that inertia-induced drifts such as the Segré–Silberberg effect should have been unimportant. As a consequence, the phenomenon is suggestive of the existence of particle migrations induced by gradients in shear stress. To be sure, it is impossible to tell from these experiments whether the particles accumulated at the centre or on the sides of the channel and thus whether the drift was towards or away from regions of high shear stress. In §3, however, we shall propose a mechanism according to which particles would be expected to drift from regions of high to low shear stress and which is sufficient to account for the observed phenomenon.

Therefore, if a suspension with a non-uniform concentration profile is sheared, the particles will diffuse and attain a uniform distribution and hence the viscosity will increase to some equilibrium value. Since this diffusion process occurs in the radial direction (the direction of increasing fluid velocity) we may use the observed viscosity data to estimate both the magnitude of the concentration fluctuation across the channel as well as the diffusion coefficient in this direction.

We model the process by assuming that the diffusion coefficient across the gap is constant throughout the migration. Again, this approximation is acceptable if the variation in concentration is sufficiently small, and in any case the experiment will yield some average value of the diffusion coefficient for the migration. We denote the initial concentration profile by $\phi(0,y)$, and that after a long period of shear by $\bar{\phi}$ = constant (assumed), and obtain for the concentration at all times

$$\phi(y,t) = \bar{\phi} + \sum_{n=1}^{\infty} B_n \cos\frac{n\pi y}{b} \exp\left(\frac{-n^2\pi^2 D_{\parallel} t}{b^2}\right), \tag{2.9}$$

where

$$B_n = \frac{1}{b}\int_0^b \phi(0,y)\cos\frac{n\pi y}{b}\,\mathrm{d}y$$

and $D_{\parallel}$ is the diffusivity within the plane of shear parallel to gradients in fluid velocity.

To further simplify the model, we approximate (2.9) by its limiting form at large strain, i.e. we let

$$B_1 \neq 0; \quad B_n = 0, \quad n \neq 1, \tag{2.10}$$

in which case, to obtain accurate values of the diffusion coefficient under this assumption, we fit (2.9) only to data taken after sufficient time has elapsed for the terms that we have neglected to become unimportant. But, since these higher-order terms decrease exponentially with a rate constant at least four times that of the leading-order term, this requirement is easily met. In any case, the first viscosity measurements used in calculating $D_{\parallel}$ were taken after a strain of at least 3. It should be noted in this context that Gadala-Maria & Acrivos (1980) demonstrated that, upon shearing, suspensions acquire a short-range order over a characteristic strain of approximately 3, and that this ordering affects the observed viscosity. By taking measurements after this short-range ordering had reached steady state, however, we ensure that any further increase in the viscosity would have to be due to the migration of the particles across the gap.

To fit our data to the model, we need to calculate the effective viscosity

$$\mu_{\text{obs}} = b\left[\int_0^b \mu^{-1}\,\mathrm{d}y\right]^{-1}. \tag{2.11}$$

Thus, the model describing the short-term viscosity increase contains three adjustable parameters: the equilibrium uniform concentration; the amplitude of the initial variation in concentration B_1; and the diffusion coefficient. The first of these, however, is fixed by the equilibrium viscosity corresponding to the concentration of the suspension initially loaded; thus the rate at which the viscosity approaches its equilibrium value and the magnitude of the deviation between this and its value at the start of the experiment should yield the diffusion coefficient and the approximate initial concentration variation across the gap.

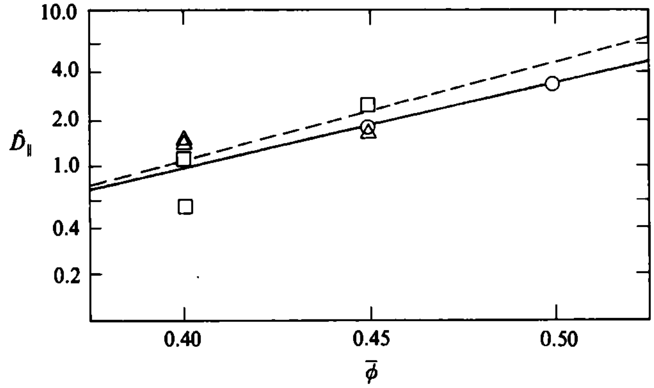

FIGURE 7. Diffusion coefficient observed in the short-term viscosity-increase experiments: ○, 46 μm polystyrene in medium gap; □, 46 μm polystyrene in small gap; △, 87 μm polystyrene in medium gap. Dashed line is the diffusion coefficient observed in the long-term viscosity-decrease experiments.

2.3.2. *Experimental results*

Initial-viscosity-increase experiments were performed with suspensions of both 46 and 87 μm polystyrene spheres at concentrations from 30 % to 50 % using all three Couette gap widths. Figure 6 presents the observed viscosity increase for a 45 % suspension of the smaller polystyrene spheres in the medium Couette gap. The fit of the data to the model was excellent; however, owing to the many assumptions that were necessary in deriving (2.9), the calculated values of the diffusion coefficient and concentration fluctuation across the gap must be considered only approximate. The diffusion coefficient was found to be proportional to $\dot{\gamma}a^2$ and, as in the case of diffusion normal to the plane of shear measured in experiments described earlier in this section, was also a strong function of concentration. A plot of the diffusion coefficient as a function of concentration is given in figure 7, where the dashed line is the diffusion coefficient normal to the plane of shear estimated from experiments described earlier in this section and depicted in figure 4. The fact that, as may be seen, the measured values of the two diffusion coefficients were almost identical provides us with added confidence that the viscosity increase was interpreted correctly as resulting from particle migration across the gap width.

The short-term viscosity-increase phenomenon was observable only for a narrow range of particle diameters, suspension concentrations and Couette gap widths. This was due in part to the fact that, since the diffusion coefficient was found to be proportional to the square of the particle radius, the total length of shearing necessary to reach equilibrium was inversely proportional to $\hat{a}^2$, the square of ratio of the particle diameter to gap width. Thus, for very large values of $\hat{a}$ (such as were obtained for experiments with the 87 μm polystyrene spheres in the narrow Couette gap) the strain associated with the migration was sufficiently short that it was not possible to reliably separate fluctuations in the viscosity due to migrations across the gap from those due to the initial equilibration of any short range order in the suspension. Similarly, the timescale for the viscosity increase was also much too short for experiments with the 46 μm spheres in the narrow gap at a 50 % concentration, owing to the high value of the dimensionless diffusion coefficient found at this concentration.

At 30 % solids concentration, a different experimental difficulty was encountered in that, although the total fluctuation in concentration across the gap may have been the same as that observed for more concentrated suspensions, the resultant variation in the observed viscosity was too small to be accurately measured. This problem was much more severe in the short-term viscosity-increase experiments than in those involving the long-term viscosity decrease at the same concentration. Indeed, from the observed dependence of viscosity on concentration, it can be shown that the same concentration fluctuation across the width of the gap affects the observed viscosity at an average concentration of 30% by an amount that is less by an order of magnitude than that at 50%.

Finally, for some combinations of concentration, particle diameter and gap width, it was likely that the fully developed concentration profile for channel flow up the gap had not been reached. But, if the initial non-uniform concentration was in fact due to channel flow rather than to entrance effects, then we would have expected that the initial-viscosity-increase phenomenon would have been absent or at least diminished in strength if the concentration distribution was not fully developed. We shall now examine this point further. First, the concentration distribution of the suspension when loaded into the gap should be that resulting from flow in the entrance region and hence should attain its asymptotic shape only beyond some distance up the gap that is a function of the diffusion coefficient, the gap width and the particle radius. The average distribution through the length of the gap will therefore approach that at equilibrium provided that the equilibration distance is much less than the total length of the gap. This places a restriction on the diffusion coefficient, particle radius, gap width and gap length that must be satisfied before the particle-concentration profile can be assumed to be everywhere fully developed.

We shall derive an approximate expression for this restriction by assuming that the flow up the Couette gap when loading the sample is everywhere parabolic, i.e.

$$u = \tfrac{3}{2}\bar{u}\left(1-\frac{y^2}{b^2}\right), \quad -b < y < b. \tag{2.12}$$

Moreover, we shall require that the diffusion length for a particle at the mid-points of the gap half-widths ($y = \pm\frac{1}{2}b$), equal to $(2\hat{D}_{\parallel}\,\dot{\gamma}a^2h/u)^{\frac{1}{2}}$, where h is the height of the gap, and u and $\dot{\gamma}$ are the local fluid velocity and shear rate, be much greater than $\frac{1}{2}b$, the distance to the wall and to the centreline. For parabolic flow, this condition reduces to

$$\tfrac{32}{3}\hat{D}_{\parallel}\frac{a^2h}{b^3} \gg 1, \tag{2.13}$$

which is a strong function of the gap width and is independent of the flow rate of the suspension into the gap. For a 45% suspension of 46 μm polystyrene spheres in the narrow gap, the observed diffusion coefficient is such that the parameter in (2.12) has a value of about 14. Thus, although for this combination of gap width, particle radius and diffusion coefficient, it is likely that the concentration profile has attained its fully developed form in the gap, this may not have been the case at lower concentrations, owing to the lower value of the diffusion coefficient, or for larger gap widths. In fact, since the initial-viscosity-increase phenomenon was observable only for values of this parameter greater than about unity, this suggested that the initial inhomogeneous concentration distribution was due to the channel flow rather than entrance effects. Further experimentation is necessary to demonstrate this conclusively, however. Table 1 presents the calculated amplitude of the concentration fluctuation across the channel for those experiments where it was measureable.

Particle diameter (μm)	Gap width (mm)	ϕ	$\hat{D}_{\parallel}$	B_1
46	0.639	0.40	1.1	0.131
		0.40	0.54	0.094
		0.45	2.45	0.075
	1.261	0.45	1.7	0.062
		0.50	3.2	0.081
87	1.261	0.40	1.4	0.050
		0.40	1.4	0.075
		0.45	1.6	0.063

TABLE 1. Estimated diffusion coefficient and amplitude of concentration fluctuation for the short-term viscosity-increase experiments

3. Mechanisms leading to shear-induced migrations

Thus far, we have presented experimental evidence for the existence of shear-induced particle migrations arising from gradients in concentration and shear stress. What has yet to be shown, however, is a mechanism by which these migrations take place. Let us first examine the case of migration due to gradients in particle concentration.

Consider a single marked sphere of radius a in a suspension of otherwise identical spheres undergoing the viscous linear shear flow $u = \dot{\gamma} y$, where u is the velocity in the x-direction. As the sphere interacts with its neighbours in the shear flow, it will experience a series of displacements in both the y- and z-directions with a characteristic length proportional to a and a frequency proportional to the shear rate $\dot{\gamma}$. In the absence of any gradient in concentration, these displacements will be random with zero mean (i.e. on average the particle will remain on its initial streamline), and thus will constitute a random walk. But, as is well known, in the presence of a concentration gradient such a random walk will lead to a diffusive flux and thus may be characterized by a diffusion coefficient, in this case with the dimensional scaling $\dot{\gamma}a^2$, the same as was observed in the experiments described here. It is important to note that in a dilute suspension the spheres will return to their initial streamlines at the end of all two-particle interactions owing to the linearity of the viscous-flow equations when only viscous forces are present. As a consequence, at least three particles must interact to yield the permanent displacements that lead to a random walk, and therefore, since the rate at which two particles interact with the marked sphere is proportional to $\dot{\gamma}\phi^2$, the diffusion coefficient must be proportional to $\phi^2\dot{\gamma}a^2$ in the dilute limit.

This source of diffusive flux, termed shear-induced self-diffusion, may be measured by examining the random walk of a single marked particle in a homogeneous suspension. Experiments carried out along these lines have been conducted by Eckstein, Bailey & Shapiro (1977), and more recently by Leighton & Acrivos (1987), and have demonstrated that the diffusion coefficient is indeed proportional to $\dot{\gamma}a^2$. Leighton & Acrivos (1987) also obtained a dimensionless value of the diffusivity of about $\phi^2/2$ in the dilute limit, in agreement with the scaling predicted by the theory. The measured coefficient of self-diffusion, however, was found to be a much weaker function of concentration and, at a concentration of 40 %, had a value nearly an order of magnitude lower than the diffusivity that was calculated from the experiments described earlier in this work. The discrepancy between the observations of self-diffusion and effective diffusivity in the presence of a gradient in concentration suggests, therefore, that the presence of a concentration gradient in some way induces a drift of particles from regions of high to low concentration in addition to that provided by random self-diffusion.

The most likely source of this additional drift is that interparticle interactions in the presence of a gradient in concentration lead to an average displacement of the marked sphere from regions of high to low concentration. It is not clear at this stage whether interactions in the presence solely of viscous forces can lead to such a drift. Direct calculations of the drift for such a suspension would require consideration of interactions involving at least three spheres in a dilute suspension and, in concentrated suspensions where the average interparticle separation distance is very small, the interaction of many spheres would have to be taken into account. Such calculations are far beyond the capabilities of current analytical techniques, and are also quite difficult to deal with numerically owing to the very large number of particles that must be included in any computation.

As we shall presently demonstrate, however, for sufficiently concentrated suspensions of real non-colloidal particles, it is not necessary or even appropriate to consider only the influence of purely viscous hydrodynamic forces because, in such systems, the particles are driven sufficiently close together by the flow that irreversible surface contact will occur as a consequence of surface-roughness effects, thereby destroying the macroscopic reversibility of the purely viscous interactions. In this section we shall therefore discuss both theoretical and experimental evidence for the existence of such irreversible interactions and demonstrate that they may lead to the diffusivities observed here.

3.1. *Irreversible interactions in concentrated suspensions*

Consider a sphere interacting with a second sphere in a simple shear flow. As the two spheres approach one another, the viscous stresses in the fluid act to drive the spheres together. Under purely viscous conditions, when the interparticle separation distance is very small this approach is resisted by the lubrication layer between the particles, in which the resistance is inversely proportional to the separation distance. Thus, although two mathematically smooth interacting spheres may never touch, hydrodynamic theory predicts that, over a certain range of initial configurations, they will approach one another very closely even in a dilute suspension. On the other hand, in the presence of a finite amount of surface roughness on the spheres (as is the case for any real particles), the interaction may be significantly modified. Indeed, Arp & Mason (1977) found that even for two isolated interacting spheres, the existence of a small degree of surface roughness was sufficient to eliminate the closed orbits predicted for purely viscous interactions.

This effect of surface roughness is accentuated for concentrated suspensions. Specifically, since the forces driving spheres together in the flow depend on the bulk fluid stresses, they are proportional to the bulk suspension viscosity, which in turn is a strong function of concentration. In contrast, the lubrication forces resisting this approach remain proportional to the pure-fluid viscosity since the presence of other particles in the suspension does not affect the flow in the narrow gap between particles. This imbalance, combined with some finite surface roughness, implies that in sufficiently concentrated suspensions particles will simply approach one another without any significant displacement from their original streamlines until they come into physical contact, following which they will rotate owing to the vorticity of the shear flow, and finally separate. Moreover, since the interaction is no longer reversible owing to the surface contact it will also no longer be symmetric and thus will lead to permanent displacements of the particles from their original streamlines at the end of each interaction. A more complete discussion of the evidence for irreversible interactions and their effect on the rheology of concentrated suspensions is given by Leighton (1985).

3.2. *Particle drift arising from irreversible interactions*

There are several ways in which the irreversible interactions described above can lead to drift in the presence of gradients in concentration or shear stress. To see this, consider a test sphere at the origin which is immersed in a suspension undergoing shear. We shall assume that the bulk flow is in the x-direction with a constant shear stress σ_{yx} and a linear concentration gradient in the z-direction, normal to the plane of shear. Under these conditions, the viscosity, and hence the shear rate, will be constant within the plane of shear. Consequently, when the particles are not mathematically smooth, the test sphere will be irreversibly displaced upwards following an interaction with another sphere approaching it from below, and conversely if approached from above. Thus, in the presence of a higher particle concentration on one side of the test sphere than on the other, this sphere will experience a drift towards the region of lower concentration since it interacts with more particles on one side than on the other. Moreover, the displacement after each interaction will be proportional to the particle radius; thus since the excess rate of interactions from regions of higher concentration is proportional to $\dot{\gamma} a \,\mathrm{d}\phi/\mathrm{d}z$, the particle flux resulting from this source of drift should be proportional to $\dot{\gamma} a^2 \phi \,\mathrm{d}\phi/\mathrm{d}z$, which is the same scaling expected for shear-induced diffusion.

A second source of drift normal to the plane of shear arises from the gradients in suspension viscosity brought about by gradients in concentration. First we note that in the absence of a gradient in viscosity, two touching spheres in a shear field will rotate about their centre of mass (the point of contact). But, in the presence of a viscosity gradient, the centre of mass will no longer be the centre of rotation and the particles will, on average, be displaced during an interaction from regions of high to low viscosity. The magnitude of this displacement during each irreversible interaction will scale as the relative variation in viscosity across the particle, i.e. it will be proportional to $(a/\mu)\,\mathrm{d}\mu/\mathrm{d}z$, multiplied by the particle radius. Since, for small gradients in concentration, the variation in viscosity is linear in the concentration gradient, the above result, when multiplied by the rate of interactions $\dot{\gamma}\phi$, gives the corresponding drift velocity. In turn, when multiplied by ϕ and divided by $\mathrm{d}\phi/\mathrm{d}z$, the drift velocity leads to an expression for the effective diffusivity arising from this source of drift $D_\perp \sim \dot{\gamma} a^2 (\phi^2/\mu)\,\mathrm{d}\mu/\mathrm{d}\phi$. The total effective diffusivity normal to the plane of shear is then the sum of the two mechanisms outlined above, plus that due to shear induced self-diffusion.

For concentrated suspensions, the function $(1/\mu)\,\mathrm{d}\mu/\mathrm{d}\phi$ is very large, with a value of about 15 for a 45 % suspension; thus drift due to gradients in viscosity is likely to dominate the diffusivity. Recognizing this, we therefore let

$$D_\perp = K_\perp \frac{\phi^2}{\mu}\frac{\mathrm{d}\mu}{\mathrm{d}\phi}\dot{\gamma} a^2 \tag{3.1}$$

be the expected form for the effective diffusivity normal to the plane of shear at high concentrations, where $K_\perp$ is a dimensionless parameter whose value will depend on

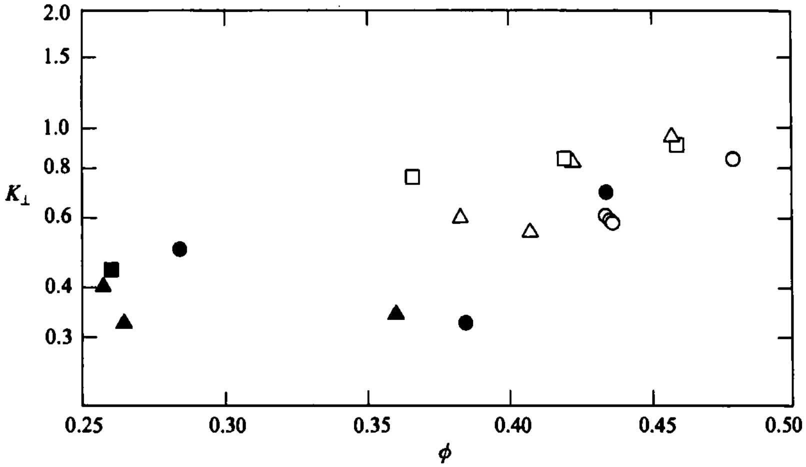

FIGURE 8. Value of $K_\perp$ *vs.* concentration: □, 46 μm polystyrene in 0.639 mm gap; ○, 46 μm polystyrene in 1.261 mm gap; △, 87 μm polystyrene in 1.261 mm gap. Open symbols, $\dot{\gamma} = 24\ \mathrm{s}^{-1}$; filled symbols, $\dot{\gamma} = 76\ \mathrm{s}^{-1}$; half-filled symbols, $\dot{\gamma} = 7.6\ \mathrm{s}^{-1}$.

the exact geometry of the interactions. As shown in figure 8, $K_\perp$, as determined from the results of our long-term viscosity experiment, was found to be a relatively weak function of concentration with a value of about 0.7 at 45 %. The scatter in the data is, of course, indicative of the simplicity of our model.

The contribution of irreversible interactions to drift within the plane of shear is quite similar to that found above for the case of drift normal to the plane of shear. Again, displacements will lead to random self-diffusion, to drift arising from a higher rate of interactions on one side than on another due to the concentration gradient (assuming a uniform shear rate), and drift from regions of high to low viscosity. In addition to these sources of drift, however, variations in viscosity within the plane of shear will, in the case of a uniform applied shear stress, lead to variations in the local shear rate. Thus, in regions of low concentration and low viscosity the shear rate will be greater, with the consequence that a test sphere will, on average, experience a greater number of interactions from the region of lower concentration than would otherwise be the case. Since the shear rate is inversely proportional to the local viscosity for a constant shear stress, the excess rate of such interactions will be proportional to $(\phi\dot{\gamma}a/\mu)\,(\mathrm{d}\mu/\mathrm{d}y)$, which in turn will reduce the drift velocity from regions of high to low concentration by an amount proportional to $(\dot{\gamma}a^2\,\phi/\mu)\,(\mathrm{d}\mu/\mathrm{d}\phi)\,(\mathrm{d}\phi/\mathrm{d}y)$, an expression for the drift velocity identical with that due to gradients in viscosity with uniform shear.

Although this drift due to the shear rate gradient effect will certainly reduce the magnitude of the effective diffusivity (the sum of all contributions due to drift and random walk) it appears unlikely that the two terms due to gradients in fluid viscosity will exactly cancel out. Thus, as in the case of diffusion normal to the plane of shear, we obtain for the limiting expression of the effective diffusivity at high concentrations

$$D_\parallel \approx K_\parallel \frac{\phi^2}{\mu}\frac{\mathrm{d}\mu}{\mathrm{d}\phi}\dot{\gamma}a^2, \tag{3.2}$$

where the value of $K_\parallel$ may be determined from the initial-viscosity-increase experiments. The results are shown in figure 9 from where it is seen that, in spite of the

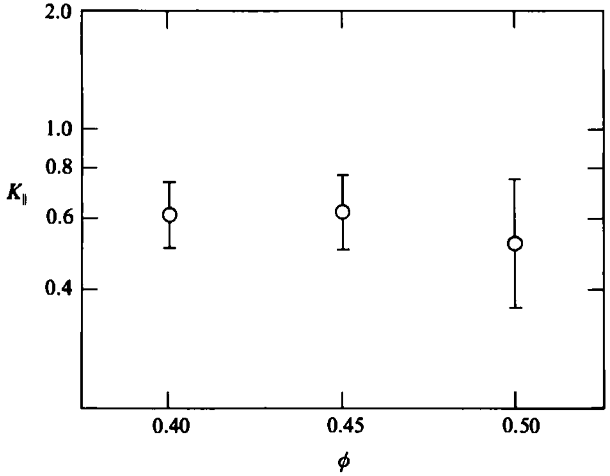

FIGURE 9. Average value of $K_\parallel$ *vs.* concentration calculated from the data in table 1. Error bars denote one standard deviation as estimated from the scatter in the experimental results.

scatter, which again is indicative of the simplicity of the model, $K_\parallel$ appears to be relatively independent of concentration and approximately equal to 0.6.

The same mechanism due to gradients in shear rate that reduced the effective diffusivity within the plane of shear should also lead to drift from regions of high to low shear stress in a homogeneous suspension. This is because, for the case of uniform particle concentrations and uniform suspension viscosities, the local shear rate is proportional to the local shear stress σ; hence the excess rate of particles interacting with the test sphere from regions of high shear stress is proportional to $(\phi\dot{\gamma}a/\sigma)/(\mathrm{d}\sigma/\mathrm{d}y)$, yielding a particle flux

$$N_y = K_\sigma \frac{\phi^2}{\sigma}\frac{\mathrm{d}\sigma}{\mathrm{d}y}\dot{\gamma}a^2, \tag{3.3}$$

where again K_σ is some order-one function of concentration. At steady state this flux will be balanced by diffusive migrations, thereby creating a steady non-uniform concentration profile. Thus, we may use the measured amplitude of the initial-viscosity-increase experiments and the values estimated for $K_\parallel$ to infer K_σ.

3.3. *Particle concentration profiles in a channel flow*

To see how K_σ may be estimated, consider a concentrated suspension flowing through a channel of width $2b$ with mean velocity $\bar{u}$. For channel flow, the shear stress is simply proportional to y, hence

$$\frac{1}{\sigma}\frac{\mathrm{d}\sigma}{\mathrm{d}y} = \frac{1}{y} \tag{3.4}$$

and, therefore, the particle flux due to gradients in shear stress will equal

$$N_y = K_\sigma \frac{\phi^2}{y}\dot{\gamma}a^2, \tag{3.5}$$

where $\dot{\gamma}$ is of course a function of y. Since the flow is bounded, the particle flux given by (3.5) will lead to the formation of a region near the walls having a reduced particle

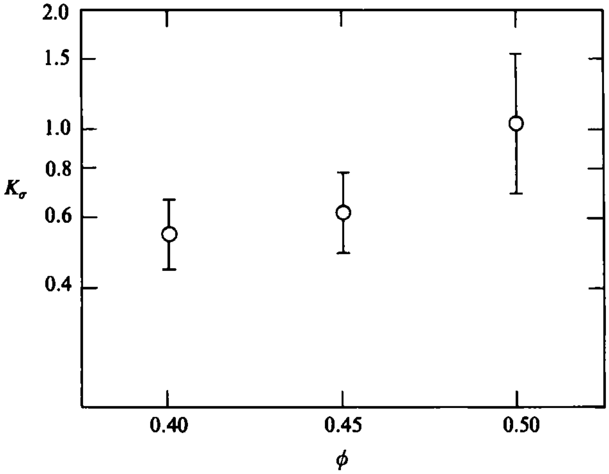

FIGURE 10. Average value of K_σ *vs.* concentration calculated from the data in table 1. Error bars are one-standard-deviation error estimated from the scatter in the experimental results.

concentration and a diffusive flux towards the walls will be induced. At steady state the net flux is zero, thus

$$\frac{\mathrm{d}\phi}{\mathrm{d}y} = \frac{K_\sigma \phi^2}{y\hat{D}_\parallel} = \frac{K_\sigma}{yK_\parallel}\left[\frac{1}{\mu}\frac{\mathrm{d}u}{\mathrm{d}\phi}\right]^{-1}. \tag{3.6}$$

The above should not be expected to hold at the centre of the channel since in this region the shear rate, drift velocity and the diffusive flux all vanish.

We may use the steady-state gradient in concentration predicted by (3.6) to estimate the deviation of the concentration profile from the average value $\bar{\phi}$. To accomplish this, we assume that the predicted concentration gradient at the midpoints of the channel half-widths approximates the average gradient across each half-width. Hence, given a channel width $2b$, the estimated amplitude of the concentration fluctuation across the channel is

$$\frac{\phi_{\max}-\bar{\phi}}{\bar{\phi}} \approx \left.\left\{\frac{1}{\phi}\frac{K_\sigma}{K_\parallel}\left[\frac{1}{\mu}\frac{\mathrm{d}\mu}{\mathrm{d}\phi}\right]^{-1}\right\}\right|_{\phi=\bar{\phi}}. \tag{3.7}$$

Note that, according to (3.7), the concentration distribution is independent of the particle radius and gap width, as was found in the experiments described in §2.

Next, if we recall that the initial concentration distribution inferred from the initial-viscosity-increase experiments was that due to channel flow, we may write, with B_1 having been defined following (2.9),

$$K_\sigma = B_1 K_\parallel \frac{1}{\mu}\frac{\mathrm{d}\mu}{\mathrm{d}\phi}, \tag{3.8}$$

which provides a means for estimating K_σ. The results of our calculations are plotted in figure 10, which, allowing for the considerable scatter in the experimental data, indicates that K_σ is a relatively weak function of concentration with a value of about 0.6 at $\phi = 0.45$.

In a bounded Poiseuille flow (channel or tube), any migration of particles toward the centre leads to an increase in the viscosity in this region and hence to a blunting of the parabolic velocity profile that applies for Newtonian fluids. Indeed, the occurrence of such blunting in the flow of concentrated suspensions through tubes was reported by Karnis, Goldsmith & Mason (1966); however, it is not clear that, in their case, the effect was due to the migration of particles. Those authors did not detect a non-uniform concentration distribution across the tube, although owing to the statistical nature of their measurement technique, a fluctuation on the order of 10% could not have been observed. Specifically, their measurements involved counting the number of marked spheres that were present in each of four divisions of the half-width of a tube cross-section, and since only a small fraction of the spheres were marked, their observations were subject to Poisson statistics, which dictate that the one-standard-deviation error in the number of spheres that were counted is equal to the square root of that number. But since the total number of spheres that were counted in each region was rather small (less than 80), the concentrations reported by Karnis *et al.* (1966) had a one-standard-deviation error greater than 10%. Further measurements of the concentration distribution in Poiseuille flow are therefore needed in order to determine the source of the observed blunting of the velocity profiles.

3.4. *Particle concentration profiles in a Couette device*

The migration from regions of high to low shear stress described in this work may also account for the observed decrease in concentration in a Couette device after long periods of shear. Consider again the Couette device depicted in figure 1. Note that at the base of the sheared gap there exists a transition region in which the applied shear stress decreases from its maximum value (that within the sheared gap) to nearly zero in the fluid reservoir. As a consequence, we should expect that particles would migrate out of the Couette gap and thereby create a region of depleted concentration at the base of the gap. At steady state, this drift would be balanced by a diffusive drift back towards the gap.

From this model we would predict that the equilibrium depletion at the base of the gap would be insensitive to ratio of the particle diameter to gap width, whereas experimentally this depletion was found to be approximately proportional to this ratio. The measured magnitude of the depletion was, however, of the same order as that inferred from the initial-viscosity-increase experiments; thus the rate of particle migration due to gradients in shear stress appears to be of sufficient strength to account for this depletion. Clearly though, further experiments that directly probe the behaviour of particles at the base of the gap are needed in order to determine the factors that govern this migration.

4. Summary

Our understanding of the behaviour of concentrated suspensions has been significantly broadened by our study of the rheology of these systems. In this section, we shall summarize the most significant results of our work and provide suggestions for future research in this area.

4.1. *Experimental findings*

In the course of our experimental study of the behaviour of concentrated suspensions of neutrally buoyant particles in Newtonian fluids using the Couette device, we demonstrated that, even under creeping flow conditions, particles may migrate across streamlines and against concentration gradients when undergoing bounded shear flow. Specifically, we have shown that the long-term decrease in suspension viscosity with shearing in a Couette device observed by Gadala-Maria & Acrivos (1980) was in fact due to the particles in the suspension migrating out of the sheared gap and into the reservoir even though both were initially at the same concentration. Further, our examination of the short-term viscosity-increase phenomenon suggests that, in a concentrated suspension flowing in a channel, particles will migrate owing to gradients in shear stress, and we have therefore provided the first evidence for the existence of a non-uniform equilibrium concentration distribution in a suspension undergoing Poiseuille flow at zero Reynolds number, the earlier work by Karnis *et al.* (1966), having failed to detect any variations in concentration.

Our experiments on the transient viscosity increase and its subsequent long-time decrease also enabled us to infer the values of the shear-induced diffusion coefficients which are important in determining the particle concentration profile that is established in shear flows. These diffusion coefficients were measured for the two directions that are of primary significance in parallel shear flows: the direction normal to the plane of shear; and that parallel to gradients in fluid velocity within the plane of shear. For such flows, migrations in the direction of fluid motion are dominated by convective diffusion and, as a result, their magnitude is governed by diffusion along gradients in fluid velocity (i.e. normal to the direction of fluid motion within the plane of shear).

4.2. *Theoretical work*

As described in §3, we provided evidence that, in sufficiently concentrated suspensions, non-colloidal spheres will come into actual physical contact as a result of surface roughness. These same interactions were also shown to lead to a shear-induced diffusive drift both normal to the plane of shear and parallel to gradients of fluid velocity within the plane of shear, as well as to a drift from regions of high to low shear stress.

These shear-induced particle drifts represent a new explanation for a number of other phenomena such as the blunting of velocity profiles in the flow of suspensions through tubes. It must be emphasized that the drifts identified here are viscous in nature; i.e. they persist at zero Reynolds number, unlike well-known sources of particle migrations such as the Segré–Silberberg effect for flow through tubes (Ho & Leal 1974) which are caused by small inertial effects and thus are proportional to the Reynolds number. As a consequence, in low-Reynolds-number flows of concentrated suspensions, they are likely to represent the dominant source of drift across streamlines.

4.3. *Suggestions for future work*

The measurements and drift calculations that have been described here represent only the beginning of the investigation into the flow behaviour of concentrated suspensions in terms of particle drift and diffusion. Of immediate importance for future work, however, is the detailed study of the concentration profile induced by the flow of concentrated suspensions through tubes, channels, Couette shear flows, and other flow geometries. Such direct measurement experiments are necessary to determine the relative importance of the various sources of drift identified here. In addition, some effort should be made to study in a more rigorous fashion irreversible interparticle interactions in concentrated suspensions, perhaps using a numerical approach similar to that employed by Bossis & Brady (1984) in determining accurately their impact on the rheology of suspensions and on the drift of particles across streamlines.

This work was supported in part by the Department of Energy, under grant Nos 80-ER10659 and 85-ER13328, by the National Science Foundation under grant No. CPE 81-17299, by a National Science Foundation fellowship to D.T.L. and by NATO Research Grant No. 538.83.

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