Korea-Australia Rheology Journal
Vol. 13, No. 1, March 2001 pp. 19-27

Migration in concentrated suspension of spherical particles dispersed in polymer solution

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(Received March 21, 2001)

Abstract

In this symposium paper, the migration and hydrodynamic diffusion of non-colloidal, spherical particles suspended in polymer solutions are considered under Poiseuille or torsional flows. The migration phenomena in polymer solutions are compared with those in Newtonian fluids and the effect of fluid elasticity is discussed. The experimental results on particle migration in dilute polymer solution reveal that even a slight change in the rheological property of the dispersing medium can induce drastic differences in flow behavior and migration of particles, especially in dilute and semi-concentrated suspensions.

1. Introduction

The flow of solid suspension and particle motion in the concentrated suspension have been of great interest in the materials development and industrial processes such as high strength ceramics and reinforced polymer composites. There have been growing interests on the microstructure, hydrodynamic interaction and concentration distribution of particles in the suspension since the pioneering work of Leighton and Acrivos' analysis on shear induced particle migration (Leighton and Acrivos, 1987) and the introduction of MRI and LDV technique for velocity and concentration measurements (Abbot *et al.*, 1991; Altobelli *et al.*, 1991; Graham *et al.*, 1991; Phillips *et al.*, 1992; Chow *et al.*, 1993; Chow *et al.*, 1994; Mondy *et al.*, 1994; Hampton *et al.*, 1997, Koh *et al.*, 1994). Until now, most studies have been focused on suspensions dispersed in Newtonian fluids except a few cases (e.g. Tehrani *et al.*, 1996; Jefri and Zahed, 1990; Huang *et al.*, 1997, Kim *et al.*, 2000). However, in many practical cases, viscoelastic liquids are used as the dispersing medium. In this symposium paper, we have collected several results related to the migration of non-colloidal spherical particles in non-Newtonian media under various flow-field that have been done in our laboratory during the last several years. The results presented here are expected to give some insight on the migration and motion of spherical particles in suspensions under the influences of viscoelasticity of fluid.

This paper is organized as follows: In section II, we consider the tube flow of suspension in polymer solutions after a brief review of the migration phenomena in Newtonian fluids. In Section III, we describe the experimental results on the torsional flow of suspension in polymer solution between two parallel discs.

2. Migration of particles in the tube flow of suspension

2.1. Brief review of particle migration in Newtonian fluids

Researches on the migration of particles have been considered as an important issue in suspension rheology since the Segre and Silberberg's (1962) report on the inertial migration of non-interacting particles in a Poiseuille flow and Leighton and Acrivos' (1987) report on the hydrodynamic diffusion of spherical particles in concentrated suspensions following Gadala-Maria and Acrivos (1980).

As stated in the introduction, particle migration in concentrated suspension has been studied by MRI [Abbot *et al.* (1991); Altobelli *et al.* (1991); Chow *et al.* (1993); Graham *et al.* (1991); Mondy *et al.* (1994); Sinton and Chow (1994); Han et al. (1999)] and laser Doppler velocimetry [LDV, Koh *et al.* (1994)]. Using these techniques, they measured the particle concentration and velocity profiles in a density matched suspensions and reported velocity blunting and a non-uniform concentration profile: The magnitude of velocity blunting increased with the increase of either the particle concentration or the ratio of particle size to gap width; The particles moved toward the center of the channel, and when the particle loading is larger than 0.3, the particle concentration at the center approached the

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Hampton *et al.* (1997) have carried out very careful experiments using MRI and have reported that, in the cases of small particle Reynolds number, particles migrate to the low-shear-rate region in the center, and the migration results in the blunting of velocity profile. Surprisingly, however, when a/R (particle radius/tube radius) is 0.0656 and $\phi_0 = 0.1$, there was no detectable net radial migration of the particles to the center. They attributed this observation to the failure of the continuum behavior due to the finiteness of particle size. But this appears to be caused by the inertial effect. Considering that inertia tends to move particles toward the midway between the center and wall in the Poiseuille flow (Ho and Leal, 1974; Segre and Silberberg, 1962), and that particle-particle interaction tends to concentrate these particles at the center, these two effects appear to be balanced to give such a result. The flow of a highly filled suspension when the inertial effect is not negligible has been reported by Han *et al.* (1999) in our laboratory: Inertia may not be neglected even for the flow of concentrated suspensions when particle Reynolds number, Re_p, is larger than approximately 0.1. Also, once particles are concentrated at a certain position, particle-particle interaction tends to spread them out. This was also applied to the cases with low ϕ_0. Therefore, in the migration of particles in suspension, inertia as well as particle-particle interaction should be taken into account even in the cases of highly filled suspensions.

2.2. Experimental methods

The suspensions consisted of dry sieved poly(methyl methacrylate) (PMMA, $\rho = 1{,}188$ kg/m^3) spheres in a density matched Newtonian fluid. The average particle diameter was 550±50 micron. The density-matched fluid was prepared by mixing about the same amount of ethylene glycol ($\rho = 1{,}110$ kg/m^3) and glycerin ($\rho = 1{,}260$ kg/m^3). The viscosity of the Newtonian fluid was 140 mPa.s at 20 °C. As the polymer, reagent grade polyacrylamide (PAAm) with the molecular weight of 5-6 million (Aldrich Chem. Co., Catalog #18,127-7) was used as received. In the dilute limit, the Zimm relaxation time and the coil overlap concentration of the fluid determined from η_0 were 0.19 sec and 1,600 ppm, respectively. The polymer solution shows almost no shear-thinning phenomenon in our experimental range of 0 200 ppm as shown in Fig. 1.

The flow domain is a circular Teflon tube with the nominal diameter of 1/4 inch. The tube was placed horizontally and in the direction perpendicular to the main magnetic field direction of our MRI equipment. The entrance length was sufficiently long to have a fully developed flow (Nott and Brady, 1994). The detailed measuring technique by MRI is described in Han *et al.* (1999). We did not stop the flow when collecting the data because the recoil problem could be severe in the case of polymer solution as shown in Fig. 2.

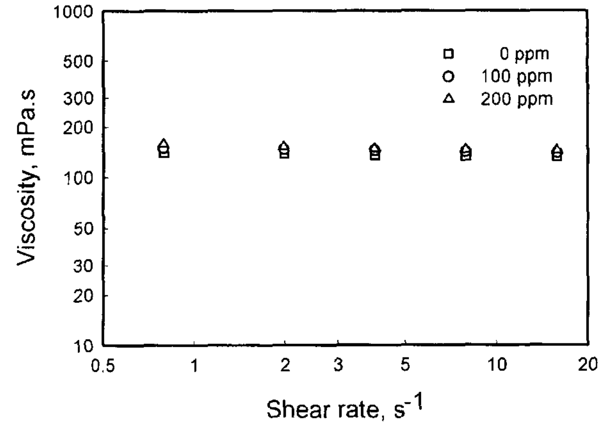


Fig. 1. Viscosity of polymer solutions. The viscosity changes less than 5% per decade.

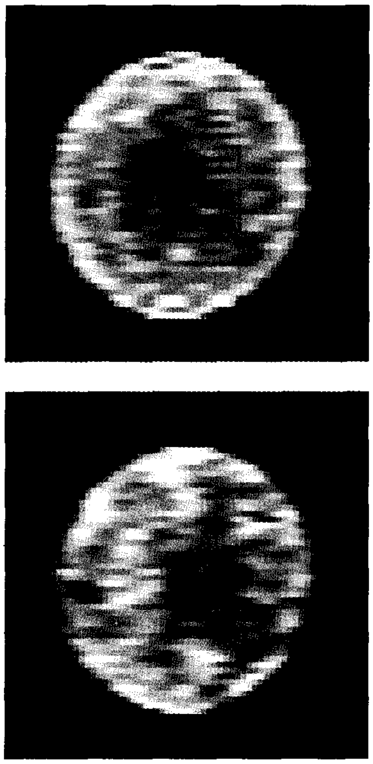


Fig. 2. Density images of particle loaded polymer solution flowing in a circular tube.
Upper: density acquired while the fluid flows; Lower density acquired when the flow stopped.

2.3. Particle migration in polymer solution

There are many combinations of suspensions of different particle loading and polymer concentration we performed the measurements. The particle loadings were 6, 25 and 40% and the polymer concentrations are 10, 50, 100 and

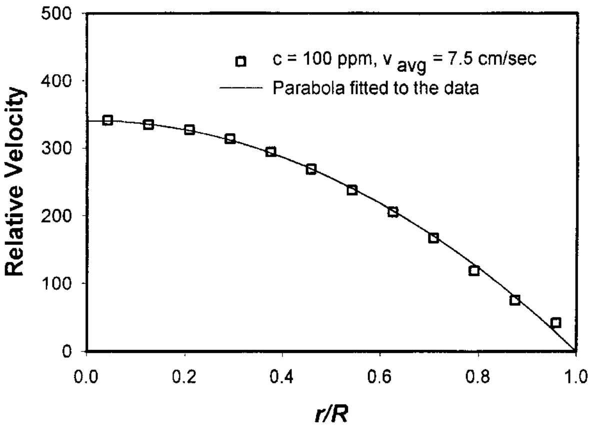


Fig. 3. Velocity profile for a flowing polymer solution when the flow rate is relatively large. The experimental data are perfectly fitted to the parabola with the same maximum velocity at the center. The velocity blunting that appears in the flow of suspension is not directly caused by the shear thinning of polymer solution. It is caused by the different particle-particle interaction in polymer solution.

200 ppm. Here we shall describe only some of the cases that are qualitatively different from case to case. Before examining the flow of suspension, we measured the velocity profile of polymer solution first to separate the velocity blunting due to the shear thinning of polymer solution from the one due to suspension flow. It was found that the velocity profile for polymer solution is well fitted to the parabolic form for the experimental ranges tested here (See Fig. 3). Therefore, velocity blunting observed in the suspension flow should not be directly due to the shear-thinning property of polymer solution.

Fig. 4 shows the images of particle distribution when $\phi_0 = 0.06$ for differing polymer concentration and particle Reynolds number. Here, the particle Reynolds number, Re_p, is defined using the radius of the particle, the average velocity of the suspension and the zero-shear viscosity as follows:

$$Re_p = \frac{a v_{avg} \rho}{\eta} \qquad (1)$$

In defining Re_p we assume that the particle velocity is the same as the fluid velocity. In all cases, particles have moved toward the center. Even when the polymer solution is extremely dilute, the elastic effect overwhelms the inertial effect which is very important in the case of Newtonian fluid (Han *et al.*, 1999). Fig. 5 shows the particle concentration and velocity profiles for several combinations of particle Reynolds number and polymer concentration while particle loading is fixed at 0.06. As the polymer concentration increases, the particle concentration at the tube axis becomes closer to the value of maximum packing fraction, 0.68. Also, as the polymer concentration increases, the velocity profile deviates from the parabolic profile severely.

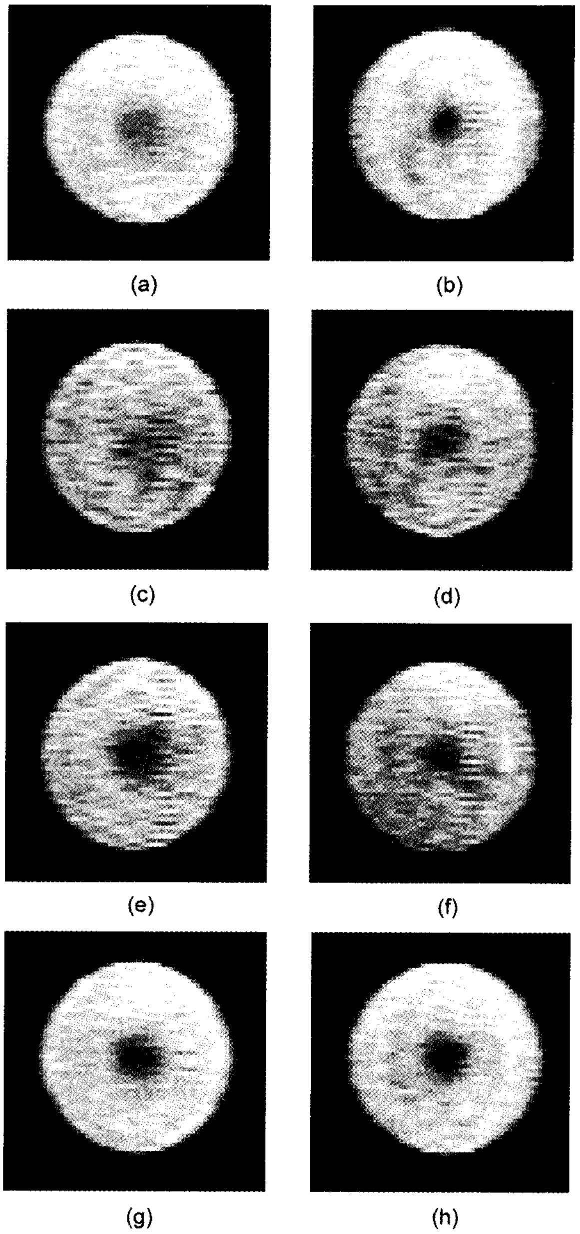


Fig. 4. Density images when $\phi_0 = 0.06$. The liquid is polymer solution.
(a) c = 10 ppm, $Re_p = 0.127$; (b) c = 10 ppm, $Re_p = 0.211$; (c) c = 50 ppm, $Re_p = 0.092$; (d) c = 50 ppm, $Re_p = 0.192$; (e) c = 100 ppm, $Re_p = 0.095$; (f) c = 100 ppm, $Re_p = 0.26$; (g) c = 200 ppm, $Re_p = 0.114$; (h) c = 200 ppm, $\mathrm{Re_p} = 0.23$.

The changes observed here should be caused by the changed particle motion in polymer solution due to the hydrodynamic interaction between particles in viscoelastic fluid. Apparent wall-slip was not observed when the particle loading is small.

Fig. 6 shows the particle-concentration and velocity profiles when the particle loading is 25%. Unlikely the case of small particle loading, the particle concentration decreases

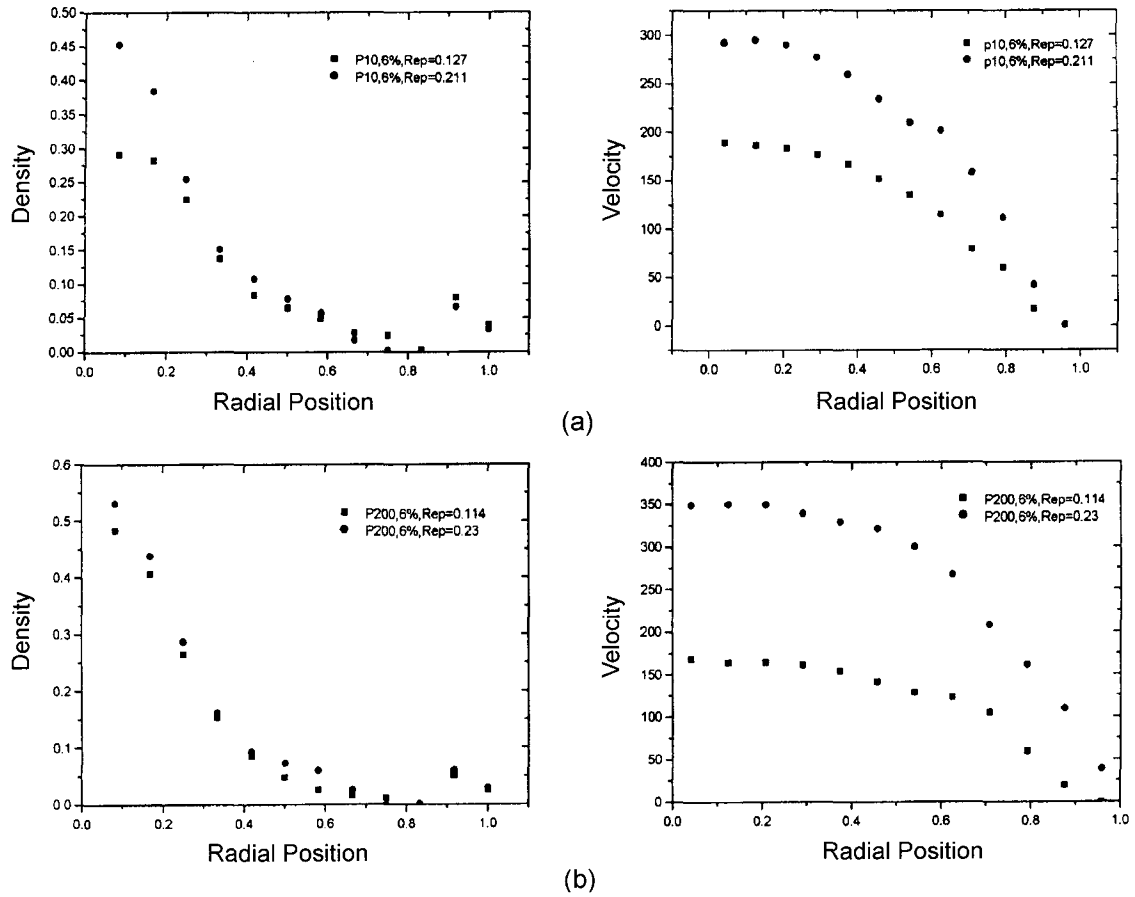


Fig. 5. Density and velocity profiles when $\phi_0 = 0.06$. The liquid is polymer solution.
(a) c = 10 ppm, $Re_p = 0.127$ and 0.211; (b) c = 200 ppm, $Re_p = 0.114$ and 0.23.

almost linearly from the center to the wall and the velocity profile is strongly blunted. But wall-slip is not still observed. With MRI experiments, unsteadiness cannot be probed and we have to examine the data assuming steady states. When we observed the flow, it was actually unsteady. Macroscopically, the flow was wavy as shown in Fig. 7. Some particles strived to stay away from the tube wall and to get into the particle core. In this case, the lift force due to the elasticity seemed to be large. When the particle loading is 40%, the particle distribution and velocity profile were almost the same as when the particle loading is 25%. But in this case, some extra particles are also observed between the wall and the particle core.

To understand and explain the migration of particles and change of velocity profile in the flow of suspension, we need to know the particle-particle interaction in viscoelastic fluid. This field is now in infant years. For example, there are discrepancies among experiments even in the case of sedimentation of a single particle in Boger fluids. Now the different extensional nature of different Boger fluids is known to be responsible for the difference (Solomon and Muller, 1996). But still we need more studies on the particle-particle interaction in non-Newtonian fluids to understand the particle migration and rheological behavior of suspensions in non-Newtonian fluids.

3. Migration of particles in torsional flows

3.1. Migration of spherical particles in Newtonian fluid

In a torsional flow between two parallel discs, a single particle does not tend to move along the radial direction in Newtonian fluid. Also, in a concentrated suspension, particles do not migrate along the radial direction (Chow *et al.*, 1994). This observation is contrary to the case of Couette or Poiseuille flows in which particles migrate toward the region of smaller shear rate when the particle Reynolds number is vanishingly small. Recently Krishnan and Leighton (1996) reported a very interesting result on migration of particles in Newtonian liquid. They observed that, in bidisperse suspension, larger particles move toward the edge of the discs in torsional flow of the concentrated suspension in Newtonian fluid. Based on the observation, they argued that there is one more mechanism of particle

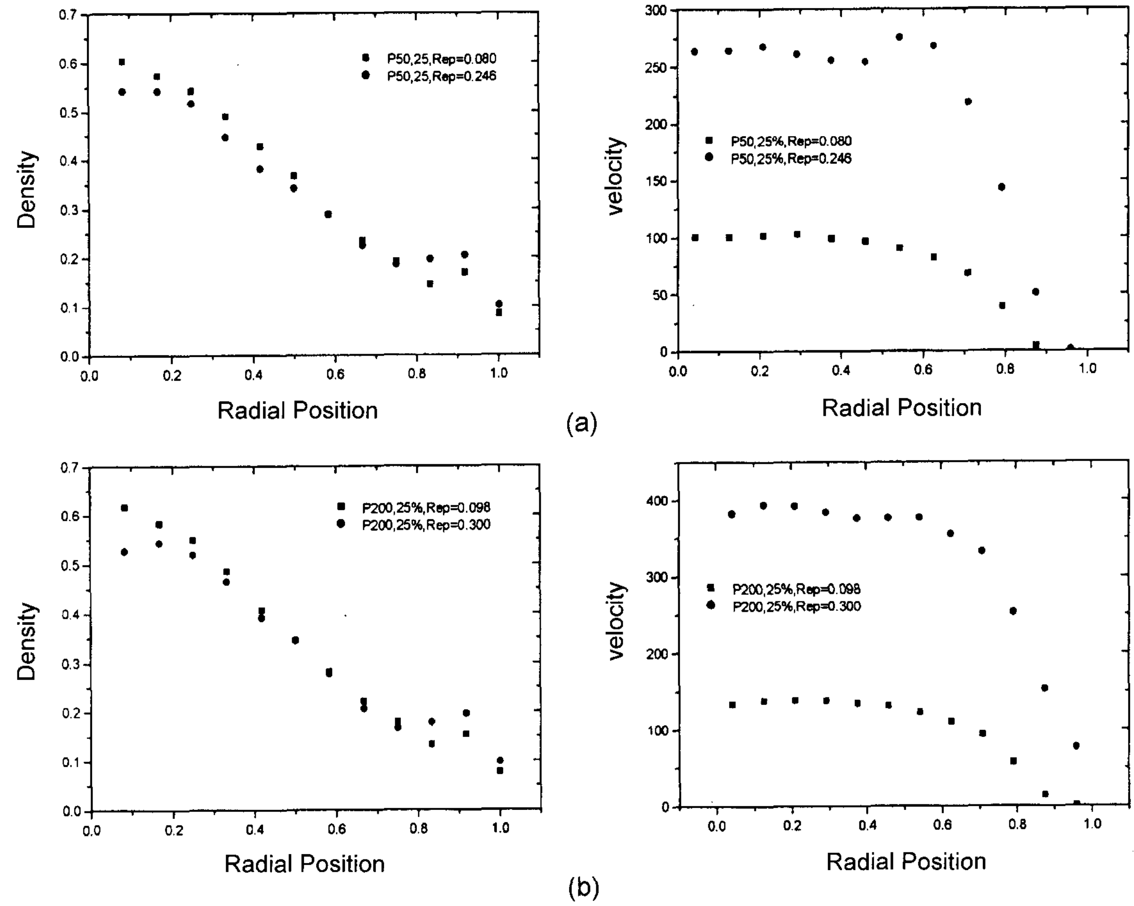


Fig. 6. Density and velocity profiles when $\phi_0 = 0.25$. The liquid is polymer solution.
(a) c = 50 ppm, $Re_p = 0.080$ and 0.246; (b) c = 200 ppm, $Re_p = 0.098$ and 0.300.

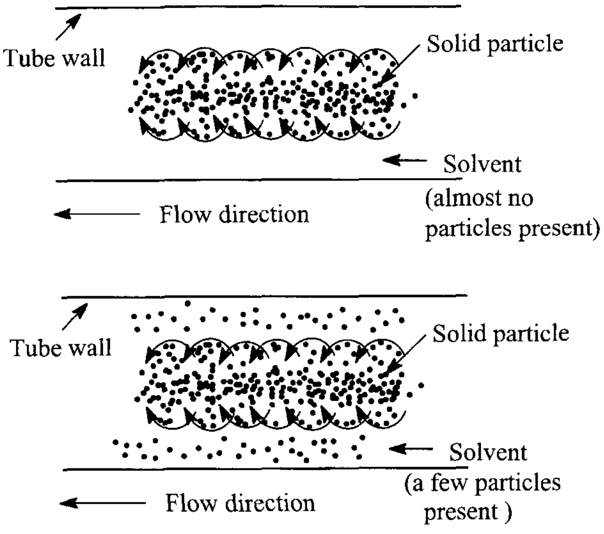


Fig. 7. Unsrteady characteristics during the flow of particle loaded polymer solutions.
Upper: particle loading = 25%; Lower: particle loading = 40%

migration in curved streamline flow in addition to the well known driving force of viscosity gradient and concentration gradient. They showed that in curved streamline flow, the net flux of particles along the radial direction is given by the following formula.

$$N_r = K_r\dot{\gamma}a\phi^2\frac{a}{r} - K_\sigma\phi^2a^2\frac{d\dot{\gamma}}{dr} - D\dot{\gamma}a^2\frac{\partial\phi_s}{\partial r}$$

$$= (K_r - K_\sigma)\frac{\Omega}{h}a^2\phi^2 - D\dot{\gamma}a^2\frac{\partial\phi_s}{\partial r} \tag{2}$$

In the above equation the term that contains the K_r represents the flux due to curvature of streamline. In the case of suspensions in Newtonian fluids, $K_r = K_\sigma$ is expected since no radial migration is observed. This finding explains some discrepancies that have been reported in the literature, especially on the migration of bidisperse suspension in the Couette geometry.

In the case of suspensions in Non-Newtonian fluid, there has been no experimental study on particle migration under torsional flow of Boger fluids. Only the single-particle cases have been reported in the literature by the Prieve group (Karis *et al.*, 1984; Prieve *et al.*, 1985; Choi *et al.*, 1987). It has been observed that the particle tends to move radially outward when the radial position of the particle is initially larger than a critical value, r_c. If the initial position is smaller than r_c, the particle tends to move radially inward. But theoretically it is predicted that the particle moves toward the center of disc in a second order fluid (Chan and Leal, 1977; Brunn, 1980; Choi, 1991). This discrepancy has not been resolved yet.

3.2. Experimental method

In this study, we performed a series of experiments on the migration of spherical particles suspended in Boger fluids. Particle loading was in the range of 10-50 %. To expedite the migration speed we used a rather concentrated polymer solution than used in the tube flow experiment. But the rheological behavior was qualitatively the same as the highly dilute solution. In other words, the concentrated solutions did not show appreciable shear thinning as shown in Fig. 8.

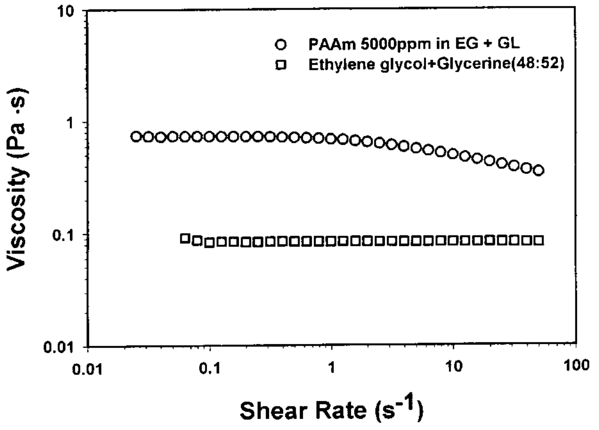


Fig. 8. Viscosity of 5000 ppm solution used in torsinal flow studies. Even in this rather concentrated solution, viscosity is almost constant and shear thinning is not severe.

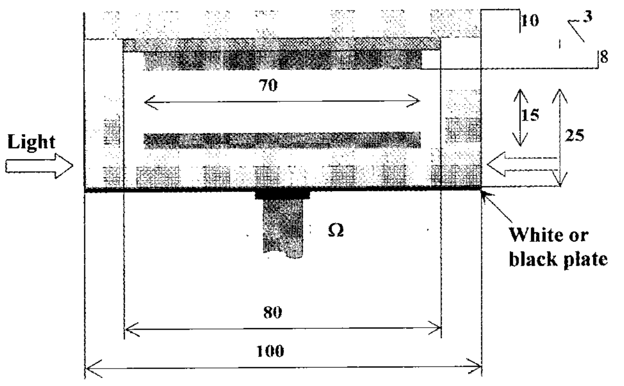


Fig. 9. Schematic diagram (side view) of the experimental apparatus for torsional flows.

As was done by Prieve group and Krishnan and Leighton, we also investigated the migration of particles directly by setting up a parallel plate system that is shown in Fig. 9. The plates were machined from transparent Plexiglas. With this set-up, the alignment and the gap distance were always ensured. At the bottom of the lower plate we attach an opaque polyacetal plate. Depending upon the color of the suspended particle, a sheet of black or white paper was inserted between the lower Plexiglas plate and polyacetal plate for better observation. The shear rate was controlled between 5-30 sec^{-1}. The diameter of plate was 70 mm and the gap distance was fixed at 2 mm.

In Fig. 10, the change of particle concentration as time is shown when the particle loading is 30%, polymer concentration is 5000 ppm and the shear rate is 10 sec^{-1}. At the start, the particle concentration is uniform. At t = 20 min, many concentric cells begin to form. As time passes, the time evolution of concentration profile is very complicated. But eventually almost all the particles migrated toward the edge. Also the particle distribution appears continuously distributed along the radial direction. We observed that the profile evolution varied from run to run due to slightly different initial distribution that was unavoidable in loading the suspension in the gap. However, after long time shearing the concentration profile converged to almost the same profile. In Fig. 11, we have shown the final distribution of particles for differing particle loading and shear rate. In the figure we may observe that particle distribution is strongly dependent upon shear rate and particle loading. This result is quite different from the cases of suspensions in Newtonian fluid. When the shear rate is less than 20 sec^{-1}, many concentric cells are observed. From this result, we can surmise that there are radial motions and the streamlines are not simple circles. Savins and Metzner (1970) already observed the existence of many radial cells, induced by the inertial effect. Until now, we have not been able to reach a definite conclusion whether this is an instability problem, a different constitutive behavior of polymeric suspension or just a distribution of particles while keeping the circular streamline during the flow. The last case cannot be excluded because even the migration of a single particle is dependent upon the initial position. When the shear rate is 30 sec^{-1}, almost all the particles move toward the edge of the plates eventually.

When the particle loading is 50%, we have not observed visibly strong particle migration in polymer solution as shown in Fig. 12. In this range of particle loading, it was not be easy to observe migration with our apparatus unless the migration is strong enough. Another point is that we did not observe the aggregation of particles. Until this point, our tentative conclusion is that particles does not get aggregated or chained in the dilute solution under the torsional flow with the shear rate less than 30 sec^{-1} as in the

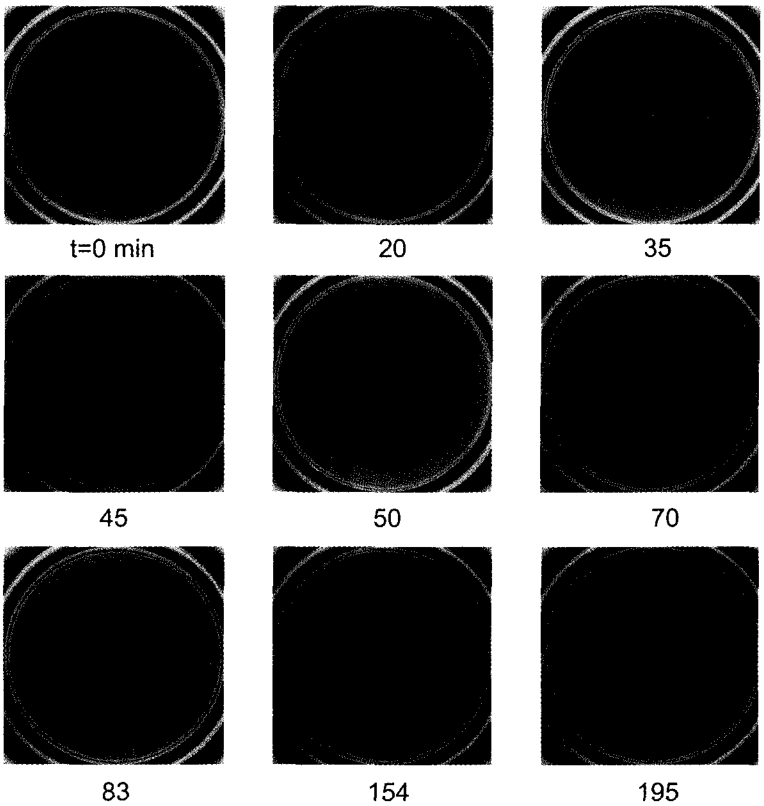


Fig. 10. Migration of spherical particles in polymer solution under a torsional flow when the particle loading is 30%, polymer concentration is 5000 ppm and the shear rate is 10 sec^{-1}. There are more particles in the lighter area than in the dark area.

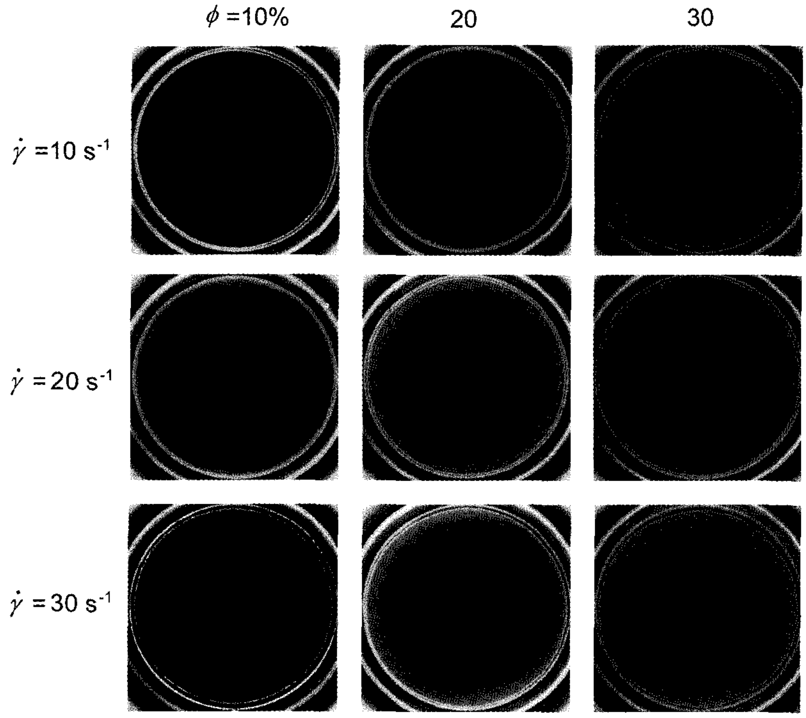
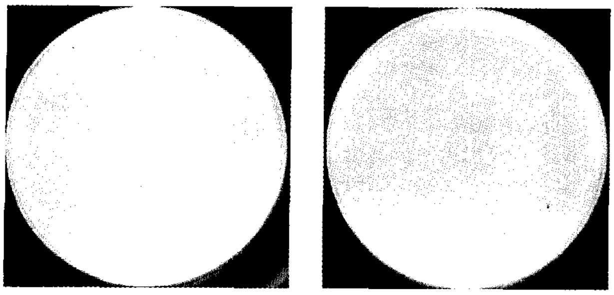


Fig. 11. Particle distribution after long time shearing. ϕ_0 = 10-30%; c = 5000 ppm; $\dot{\gamma}$ = 10-30 sec^{-1}; Gap = 20 mm. There are more particles in the lighter area than in the dark area.


t=0

5 hr

Fig. 12. Migration of spherical particles in polymer solution under a torsional flow when the particle loading is 50%, polymer concentration is 5000 ppm and the shear rate is 30 $\sec^{-1}$. No migration is observed in this case.

case of suspensions in Newtonian fluid. But we have to be very cautious in saying that a polymer solution is dilute and flow is weak.

This research is now in the beginning stage, hence the result is only qualitative rather than quantitative. But the results on particle migration in dilute polymer solution obtained so far reveal that even a slight change in the rheological property of the dispersing medium can induce drastic differences in flow behavior and migration of particles, especially in semi-concentrated suspensions.

Acknowledgments

The author wish to acknowledge the financial support from the Applied Rheology Center (Project Number: 2000 G0101), Korea University (ERC supported by KOSEF).

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