Hydrodynamic diffusivity of spherical particles in polymer solution

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Abstract In this research experiments were performed to examine the hydrodynamic diffusion of spherical particles in a highly filled suspension. The suspension consisted of nearly monodisperse polymethylmethacrylate spheres in a density matched polymer solution. The polymer solution was prepared by dissolving 0–700 ppm of polyacrylamide in a mixture of ethylenglycol and glycerine. The polymer solution did not show appreciable shear thinning. The particle loading was varied from 30 to 55%. The hydrodynamic diffusivity was estimated by measuring the time-dependent viscosity when the suspension was subjected to a circular Couette flow with an air bubble trapped under the rotor of the Couette apparatus. The results show that the dimensionless diffusivity ($D/\gamma a^2$) of particles in polymer solution is not proportional to shear rate ($\gamma$), as in the case of a Newtonian fluid, but that it decreases with increasing shear rate. The diffusivity also decreases with increasing polymer concentration. It is suggested that the elongational thickening behaviour and the increased lubrication force due to the first normal stress difference may be responsible for the reduction of diffusivity in the polymer solution.

Key words Couette flow · Particle-particle interaction · First normal stress difference · Elongational thickening

Introduction

The flow of a solid suspension and particle motion in the concentrated suspension have been of great interest in materials development and industrial processes such as high strength ceramics and reinforced polymer composites. There has been growing interest in the microstructure, hydrodynamic interaction and concentration distribution of particles in the suspension since the pioneering work of Leighton and Acrivos (1987) on shear induced particle migration and the introduction of the MRI 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, 1994; Mondy et al. 1994; Hampton et al. 1997). Until now, most studies have been focused on suspensions dispersed in Newtonian fluids except for a few cases (e.g. Tehrani et al. 1996; Jefri and Zahed 1990). However, in many practical cases, viscoelastic liquids are used as the dispersing medium. In this research, in order to understand the effect of elasticity on the migration of particles in viscoelastic fluids, the viscosity of a suspension of non-Brownian spherical particles in dilute polymer solutions was investigated as a function of time in inhomogeneous shear flows.

Gadala-Maria and Acrivos (1980) first reported that the viscosity of concentrated suspensions decreased slowly and eventually reached a steady value when a Couette viscometer was used with an air bubble trapped under the bob. The suspensions were made from Newtonian fluids and contained non-Brownian spherical particles. But they were not able to give a proper
explanation for the phenomenon. Leighton and Acrivos (1987) repeated the experiment with the same geometry and they observed that the viscosity increased first and then decreased to an asymptotic value as Gadala-Maria and Acrivos had reported. Leighton and Acrivos showed that the increase or decrease in viscosity was caused by the migration of particles in radial or axial directions in the Couette geometry. They also showed that the particle migration could be modeled by a diffusion process and the diffusivity was proportional to $\dot{\gamma}a^2$. Here $\dot{\gamma}$ is shear rate and $a$ is the radius of particle. They called this phenomenon hydrodynamic diffusion. They argued that the hydrodynamic diffusion was due to irreversible collision in the shear field between imperfectly spherical particles. After this pioneering report, the particle migration in concentrated suspension has been studied systematically adopting new experimental techniques such as magnetic resonance imaging (MRI) and laser Doppler velocimetry (Koh et al. 1994). These techniques can measure particle concentration and velocity profiles in flowing suspensions non-invasively. In particular Chow et al. (1994) used the MRI technique to study the migration in a Couette geometry and confirmed the correctness of the Leighton and Acrivos hydrodynamic diffusion model by showing that the concentration gradient developed in the Couette cell along the axial direction.

To the authors’ knowledge there has been no report on hydrodynamic diffusion in a non-Newtonian dispersing medium. In this study we have followed Leighton and Acrivos’ work by using dilute polymer solutions as the dispersing medium. We measured the viscosity in the same type of Couette cell and determined the hydrodynamic diffusivity by assuming that the migration in polymer solution was governed qualitatively by the same mechanism as in Newtonian liquids. This study is a continuation of the previous work (Han and Kim 1996) and aims to understand particle migration in viscoelastic fluids. The results show that hydrodynamic diffusivity in polymer solutions is smaller than in Newtonian liquids. This could be explained by the altered particle-particle interaction due to the elasticity of polymer solutions.

### Theory of hydrodynamic diffusion

Leighton and Acrivos (1987) assumed that the migration of spherical particles along the axial direction in a Couette apparatus could be expressed as hydrodynamic diffusion. Here we assume that the migration of spherical particles in a viscoelastic liquid can also be expressed as hydrodynamic diffusion. The validity of this assumption will be justified later in this paper. For the sake of clarity, we will briefly outline the theory of hydrodynamic diffusion by Leighton and Acrivos. Let us consider a Couette apparatus filled with a suspension. The initial concentration is uniform at $\phi_0$. The lower edge of the rotor is recessed and the lower part of the rotor traps an air bubble which doesn’t exert stresses on the suspension underneath. We will call this space the reservoir (see Fig. 1). When the rotor begins to rotate from a stationary rest state, some particles move into the reservoir where the shear rate is lower than in the gap. The reservoir concentration then increases and this induces back diffusion towards the gap due to the concentration difference. A pseudo-steady state will be established when these two diffusive fluxes are balanced. The time that is required to migrate to the reservoir should be much shorter than the time for diffusion along the gap length since the gap length is large compared to the gap distance. This means that we may regard the diffusion process proceeding under a pseudo-steady state that varies slowly with time. But as the concentration in the reservoir increases the concentration at the base of gap, $\phi^*$, also increases.

The equation governing the diffusion process can be written as follows:

$$\frac{\partial \phi}{\partial t} = D \frac{\partial }{\partial z} \left( \frac{\partial \phi}{\partial z} \right), \quad 0 < z < h.$$  \hspace{1cm} (1)

In the above equation $\phi$ is the volume fraction of particles, $t$ is time, $z$ is the axial coordinate from the lower edge of the gap, $h$ is the length of the gap and $D$ is the hydrodynamic diffusivity. The initial condition is written as follows:

$$\phi|_{t=0} = \begin{cases} \phi_0, & 0 < z < h \\ \phi^*, & z = 0 \end{cases}. \hspace{1cm} (2)$$

Here we assume a uniform initial concentration in the gap except at the base of the gap. The boundary

![Fig. 1 Schematic diagram of the Couette cell. $R_o = 21$ mm; $R_i = 20.04, 18.4$ and $15.2$ mm for the rotors MV1, MV2 and MV3, respectively. $L = 60$ mm. The spacing from the bottom of the cup to the lower edge of the rotor is 18.5 mm.](image-url)