



Shear induced organization of particles in non-colloidal suspensions in steady shear flow



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ABSTRACT

A non-colloidal particle suspension is studied in shear, and it is found that shear thinning takes place at a much higher Peclet number than in colloidal suspensions. In a steady shear-rate flow, the suspension viscosity increases and reaches a steady value when a critical strain is achieved. This critical strain is independent of the shear rate, and decreases with increasing particle concentration. It is proposed that the development of a shear-induced structure of particles, which grows when the shear rate decreases, is account for the increase of viscosity. The shear thinning in a non-colloidal suspension may not be due to Brownian motion, in contrast to colloidal suspensions.

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1. Introduction

Examples of suspensions can be found in many engineering applications, in sediment, paints, inks, biological fluids, aerosols, emulsion, to name a few. Their non-Newtonian character is determined by both the particle microstructure and the rheological behavior of the suspending liquid. The particle microstructure changes during shear, which causes the viscosity to be shear thinning at low and shear thickening at high shear rates, if the suspending liquid is Newtonian [1,2]. This behavior has been well studied for colloidal suspensions with suspended nano-particles in a Newtonian fluid (Newtonian suspension). The Peclet number, $Pe = \dot{\gamma}l^2/D$, is often used as a dimensionless shear rate in these studies, where l is a characteristic length, $D = kT/6\pi\eta_0 a$ is the self-diffusivity of a particle, a is a particle average radius. It is generally accepted that a colloidal suspension shear thins when $Pe \ll 1$. At low Pe number, the equilibrium structure of particles due to Brownian motion is distorted by the flow, which causes shear thinning of the viscosity [2]. When $Pe \gg 1$, the viscosity is considered to be independent with the shear rate, particles form a pairwise fore-aft asymmetric microstructure and hydrodynamic interactions between particles dominate the flow behavior. The first and second normal stress differences, N_1 and N_2 are both negative at high Pe number [3,4]. In the recent study, it is found that the frictional contact between particles caused by roughness can

influence the suspension viscosity and normal stress differences [5], as well as lead to discontinuous shear thickening [6]. If the solvent is viscoelastic, the non-Newtonian behavior of the suspension is affected by the particle microstructure. The onset of shear thinning in the viscosity and the normal stress differences N_1 and N_2 shifts with increasing particle volume fraction [7–9]. While shear thinning behavior of colloidal suspension has been well studied, it is found that in non-colloidal Newtonian suspensions (with particles diameter $2a > 1 \mu\text{m}$ for which Brownian motion is negligible), shear thinning also occurs at $Pe \gg 1$ [3,4,10]. It may be that the Pe defined for suspensions with nano-particles in assessing the shear thinning behavior is not appropriate for non-colloidal particles, or that other mechanisms may be responsible for the shear thinning behavior of non-colloidal suspensions – this also has been suggested by Tanner [11].

Ordering of colloidal particles in suspension has been intensively studied both in static state and in shear flow (see for example, [12–17]). Self-organization of particles in non-colloidal suspension in an oscillatory shear flow has also been studied. Corte et al. [18] showed a short-term organization of particles into a reversible absorbing state to avoid further collision between particles in oscillatory shear flow. It has also been found that in a long-term small amplitude oscillatory shear flow, particles organize into a “crystal-like” structure, which causes an increase in the apparent complex viscosity at long-term [19–22]. Lin et al. [22] pointed out that the ordering of non-colloidal particles in oscillatory shear flow is somewhat similar to the crystallization of colloidal particles induced by Brownian motion.

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In this paper, by focusing on the shear thinning behavior of non-colloidal Newtonian suspensions, we propose that its shear thinning is caused by the formation of a particle structure that diminishes with increasing shear rate, different to colloidal suspensions. Our findings and experimental data are expected to be helpful in the constitutive modeling of non-colloidal suspensions.

2. Experiments

The suspension for our rheological experiments is made up of hollow glass spheres (Dantec Dynamics), of average diameter of 3.35 μm , in silicone oil (KF-96-1000, Shin-Etsu). The particles are polydisperse in sizes, with diameters distributed from approximately 1–7 μm [22]. The glass sphere density is 1.1 g/cm^3 . The viscosity and density of the silicone oil is 1.0 Pa s and 0.97 g/cm^3 , respectively. The silicone oil is found to be Newtonian in the shear rate range examined (Fig. 1(a)). Due to the high viscosity of the silicone oil used, the suspension is stable and no observable particle sedimentation is noted during the experiment. The suspensions are made and kept in storage for a day before they are re-homogenized and tested. Measurements are carried out by a control-stress HAAKE MARS III rotational rheometer (Thermo Scientific). The cone-plate geometry with cone diameter of 35 mm is employed for suspensions. The cone angle is 1.984° and it is truncated at 104 μm from the vertex. The cone with the diameter of 60 mm is used for pure silicone oil. Three volume fractions, ϕ , of 30%, 40% and 50% are investigated in this study. The experimental temperature is kept at 25 °C.

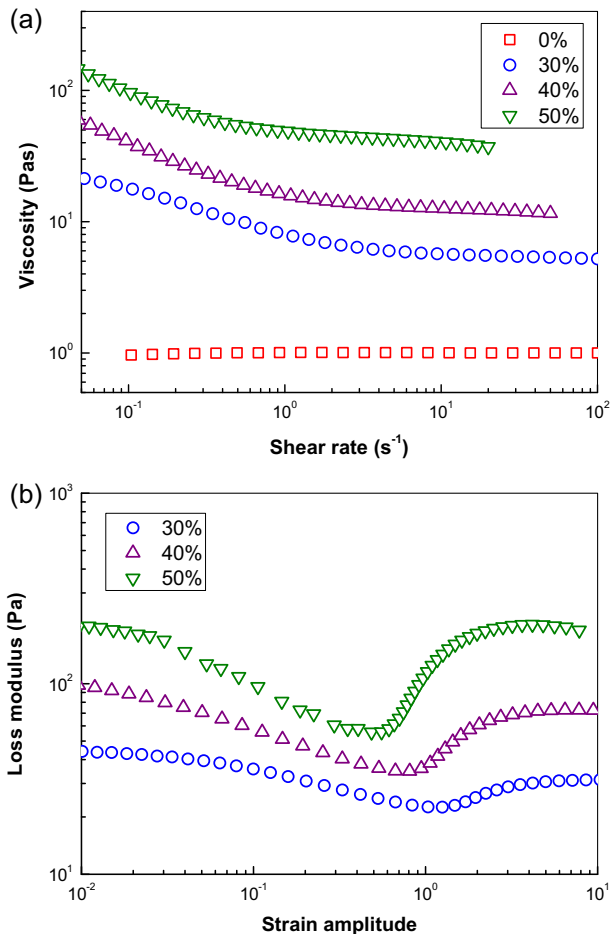


Fig. 1. (a) Suspension viscosity at various volume fractions of particles; (b) G'' as a function of the strain amplitude, in stress-amplitude sweep test.

The suspension viscosity, Fig. 1(a), is obtained by applying step increments in shear rate (using control-rate mode of the rheometer). At each step the data is generated when $(\Delta\tau/\tau)/\Delta t \leq 0.15\%$, where τ is the shear stress and $\Delta t = 1$ s. The maximum shear time for each step is 30 s (in each step, if a steady state is not reached in 30 s, the data point at 30 s is adopted). The amplitude sweep in oscillatory shear experiment is carried out using a control-stress mode, the responded strain amplitude, γ , is used to scale the results, as shown in Fig. 1(b). Evolution of the suspension viscosity in steady shear-rate flow is measured using a control-rate mode. In this mode, a transition in shear rate can be observed before the target shear rate value is reached, as shown in Fig. 2. The transition time is around 10 s for all shear rates. Before the steady shear-rate experiment and stress-amplitude sweep experiment, a pre-shearing with the shear rate of 10 s⁻¹ for a shearing time of 45 s is performed in order for the sample to achieve a well-defined initial state.

3. Results and discussion

As shown in Fig. 1(a), for our non-colloidal suspensions, shear thinning can be observed at low shear rate range, which is very common for Newtonian non-colloidal suspensions. For micron-sized particles, it is often considered that Brownian motion are negligible, and thus shear thinning should not have been taken place. The micromechanics at this level are governed by Stokes equations, implying linearity and reversibility – all the stress components are expected to be linear with the shear rate leading to a constant viscosity. For colloidal suspensions, shear thinning behavior occurs at $Pe \leq 1$, because of the distortion of the equilibrium structure of particles due to Brownian motion. For our non-colloidal suspensions, the Peclet number estimated is of $O(10^3)$ where shear thinning occurs (for example, at shear rate of 0.1 s⁻¹ in Fig. 1, considering the particle size in our experiment, $Pe \approx 2000$). Therefore, a mechanism different to those in much-studied colloidal suspensions, may be responsible for the observed shear thinning.

In the shear thinning range with a steady shear rate, a transition behavior in the viscosity can be seen, as shown in Fig. 3. The viscosity increases with strain (time \times shear rate) before reaching a steady state. The critical strain, γ_c , for the viscosity to reach a steady state can be estimated from the experimental data, and tabulated in Table 1. Note that the transition behavior of the viscosity can be influenced by the transition of the shear rate caused by the rheometer itself (as shown in Fig. 2). As this is obvious at high

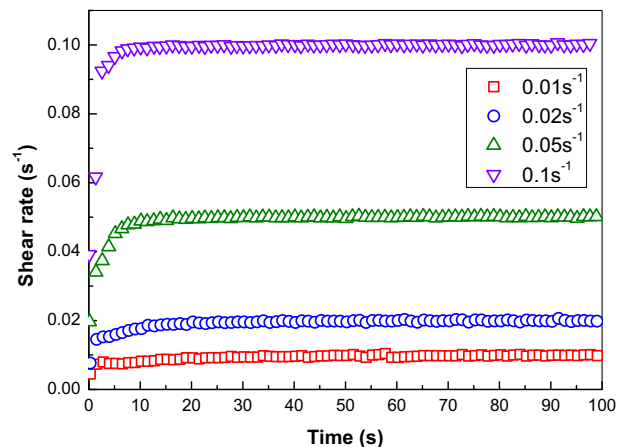


Fig. 2. Transition in shear rate in a steady shear rate measurement on a 50% suspension in the control-rate mode of the rheometer.

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