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Influences of the size and dielectric properties of particles on electrorheological response

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The influences of the size and dielectric properties of particles on the electrorheological (ER) response are investigated by computer simulations. Our studies show that, in the case of two dimensions, the field-induced shear stress τ of one-component ER fluids can be expressed as $\tau \sim n\sigma^3$, where n is the concentration of particle number and σ stands for the diameter of particles. Compared with one-component ER fluids, τ is reduced for ER fluids consisting of two kinds of particles with different sizes. When small particles are in the minority, the reduction of the shear stress can be attibuted to two reasons: one is the weakening of chains, and the other is related to the shortening and thickening of chains. The latter can explain the reduction of the shear stress for two-component ER fluids, containing a few large particles . For ER fluids consisting of two kinds of particles with different dielectric properties, there is an approximately linear relation of τ to the particle concentration ratio. [S1063-651X(99)05103-X]

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I. INTRODUCTION

Electrorheological (ER) fluids are made by suspending particles in an insulating liquid whose dielectric constant or conductivity is mismatched in order to create a dipolar particle interaction in the presence of an ac or dc electric field. When an electric field is applied, the viscosity of these fluids increases dramatically. If the field exceeds a critical value, the ER fluids will turn into solids. The phenomenon is completely reversible and the time scale is of the order of milliseconds [1–9].

The particles in a liquid are polarized upon the application of an electric field. And a particle with a complex dielectric constant $\tilde{\epsilon}_p(\omega)$ in a liquid with a complex constant $\tilde{\epsilon}_f(\omega)$ has an induced dipole moment [2,5,10]

$$\mathbf{p} = 4\pi\,\mathrm{Re}[\tilde{\epsilon}_f(\omega)](\sigma/2)^3\beta\mathbf{E}_{\mathrm{loc}}, \tag{1}$$

where σ is the diameter of the particle. $\mathbf{E}_{\mathrm{loc}}$ is the local electric field and β is the effective polarizability which can be expressed as $\beta = [\tilde{\epsilon}_p(\omega) - \tilde{\epsilon}_f(\omega)]/[\tilde{\epsilon}_p(\omega) + 2\tilde{\epsilon}_f(\omega)]$ [7,9,10]. Equation (1) shows that the induced dipole moment is related to the size and dielectric properties of particles. And it can be known that the size and dielectric properties of particles have great influences on the characteristics of ER fluids.

Although the effect of particle size on ER fluids is significant [11–13], not much attention has been given to the subject and most existing theoretical prediction models do not predict a particle size effect in the ER response [11]. Very recently, Wu and Conrad studied experimentally ER fluids consisting of two kinds of particles with different sizes (6 and 100 μm). In this case, they found that the shear stress of the two-component ER fluid decreased and small particles filled in between large particles [11]. Several authors have studied the contribution of dielectric properties of particles to the shear stress for one-component ER fluids [7–9], but ER fluids consisting of two kinds of particles which have different dielectric properties have still not been studied.

In this paper, we investigate the general relations of the size and dielectric properties of particles to the ER response by molecular-dynamics simulations. We believe these relations would be useful to the application of ER fluids.

II. METHODS

The molecular-dynamics method, which is used to study ER fluids by many earlier researchers, has been proved to be an effective method [2,5,14,15]. It has been improved by using the local field approximation to consider the mutual polarization effects between particles [16]. The improved method can be used to evaluate the strength of ER effects [16,17]. Under the local field approximation, for the ith particle, $\mathbf{E}_{\mathrm{loc}}$ in Eq. (1) can be written as [16,17]

$$\mathbf{E}_{\mathrm{loc}} = \mathbf{E}_0 + \sum_{j \neq i} \mathbf{E}_i^j, \tag{2}$$

with

$$\mathbf{E}_i^j = \frac{1}{4\pi\,\mathrm{Re}[\tilde{\epsilon}_f(\omega)]}\,\frac{3\mathbf{e}_r(\mathbf{e}_r \cdot \mathbf{p}_j) - \mathbf{p}_j}{R_{ij}^3}, \tag{3}$$

where $\mathbf{E}_0$ is the external electric field, $\mathbf{R}_{ij}$ is the position vector between the ith and the jth sphere centers, $\mathbf{e}_r = \mathbf{R}_{ij}/R_{ij}$, and the sum is over all the particles including their period images within the cutoff radius r_c. We take the dipolar approximation and ignore the contribution from higher multipoles [16,17]. The electrostatic force experienced by the ith particle due to the jth particle is given by

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$$\mathbf{F}_{ij}=F_0\left(\frac{\sigma_i\sigma_j}{\sigma_0^2}\right)^3|\beta_i\beta_j|\frac{[3(\mathbf{p}_i^*\cdot\mathbf{e}_r)(\mathbf{p}_j^*\cdot\mathbf{e}_r)-\mathbf{p}_i^*\cdot\mathbf{p}_i^*]\mathbf{e}_r-[(\mathbf{p}_i^*\cdot\mathbf{e}_\theta)(\mathbf{p}_j^*\cdot\mathbf{e}_r)+(\mathbf{p}_i^*\cdot\mathbf{e}_r)(\mathbf{p}_j^*\cdot\mathbf{e}_\theta)]\mathbf{e}_\theta}{R_{ij}^{*4}}, \tag{4}$$

where $F_0=3\pi\,\mathrm{Re}[\tilde{\epsilon}_f(\omega)]\sigma_0^2E_0^2/16$. Here $\mathbf{p}_\alpha^*=\mathbf{p}_\alpha/\{\pi\,\mathrm{Re}[\tilde{\epsilon}_f(\omega)]\sigma_\alpha^3\beta_\alpha E_0/2\}$ $(\alpha=i,j)$. Also, σ_0 is the unit of length, which is chosen to be the maximum diameter of the particles in ER systems, and $R_{ij}^*=R_{ij}/\sigma_0$. The Brownian and colloid forces are left out for relatively strong electric field [15–17].

The hydrodynamic resistance acting on particles is simply treated as Stokes' drags [2,5,13–17]. To introduce hard spheres and hard walls into the simulation, we take a short-range repulsion between two particles as

$$\mathbf{F}_{ij}^{\mathrm{rep}}=F_0|\beta_i\beta_j|\exp\{-100[r_{ij}-(\sigma_i+\sigma_j)/2]\}\mathbf{e}_r \tag{5}$$

and the short-range repulsion between a particle and the electrodes as

$$\mathbf{F}_i^{\mathrm{wall}}=F_0|\beta_i\beta_j|\exp[-100(z_i-\sigma_i/2)] - \exp F_0|B_iB_j|\{-100[(L-z_i)-\sigma_i/2)]\}\mathbf{z}^0, \tag{6}$$

where $\mathbf{z}^0$ is the unit vector of the Z direction and L stands for the distance between the two parallel electrodes.

Ignoring the inertia terms [13,15–17], the equation of motion for the ith particle can be written as

$$\frac{d\mathbf{R}_i^*}{dt^*}=\left[\sum_{j\neq i}\mathbf{F}_{ij}^{\mathrm{el}*}+\sum_{j\neq i}\mathbf{F}_{ij}^{\mathrm{rep}*}+\mathbf{F}_i^{\mathrm{wall}}+\mathbf{V}^*(\mathbf{R}_i^*)\right]\Big/\ \sigma_i^*, \tag{7}$$

which has been made dimensionless by the following variables: force $\sim F_0$, length $\sim\sigma_0$, and time $\sim 16\eta/\mathrm{Re}[\tilde{\epsilon}_f(\omega)]E_0^2$, where η stands for the viscosity of the liquid. The electric force on each particle is the sum of dipolar forces exerted by other particles within the cutoff radius r_c, as well as images. The last term in Eq. (7) is the ambient flow velocity at $\mathbf{R}_i^*$. For steady flow, it can be represented by

$$\mathbf{V}^*(\mathbf{R}_i^*)=\dot{\gamma}^*z_i^*\mathbf{x}^0, \tag{8}$$

where $\dot{\gamma}^*$ is the shear flow rate and $\mathbf{x}^0$ is the unit vector of the X direction.

In the simulation, we assume that an electric field is applied instantaneously at $t=0$. Then we integrate the motion equation of each particle at each step. The field-induced part of the shear stress can be evaluated according to the relative positions of the particles [16–18]:

$$\tau(t)=\frac{1}{V}\sum_{i,j(\text{all pairs})}\langle\tau_{ij}\rangle, \tag{9a}$$

$$\tau_{ij}=F_{ij}^x(z_j-z_i), \tag{9b}$$

where V is the volume of the simulation cell and the F_{ij}^x is the summation of the x component of the electrostatic and short-range repulsive force experienced by the ith particle due to the jth particle. The scale of the shear stress is $\tau_0=3\pi\,\mathrm{Re}(\tilde{\epsilon}_f)E_0^2/16$.

III. RESULTS AND DISCUSSION

To simplify the problem, we consider a two-dimensional square cell (20×20) which is confined between the two parallel electrodes at $Y=0$ and $Y=20$. The cell is in the presence of a dc electric field. Then β is given as [8]

$$\beta=\frac{\gamma_p-\gamma_f}{\gamma_p+2\gamma_f}=\frac{\gamma_{pf}-1}{\gamma_{pf}+2},$$

where γ_p and γ_f are the conductivities of the particles and liquid, respectively, and $\gamma_{pf}=\gamma_p/\gamma_f$, where γ_{pf} is the relative conductivity.

A. Effect of particle size on electrorheological response

ER fluids consisting of the identical particles with the same diameter and dielectric properties, are taken into account. First we investigate the effect of particle size on the ER response. It is assumed that $\beta\approx1$, i.e., $\gamma_{pf}\gg1$. In our simulation, five initial configurations of particles are used for each case and the area fractions are limited between 0.13 and 0.40 as that used generally [19,20]. The steady shear flow rate is chosen to be 0.2. The σ^* is taken between 0.5 and 1, since the dipolar approximation will fall down for an ER system composed of particles whose sizes differ too much.

Figure 1 gives the simulation result of one-component ER fluids when the reduced particle diameter changes from 0.5 to 1. The particle concentrations are taken as 0.45, 0.375, and 0.3, respectively. It can be seen that the reduced shear stress τ^* is proportional to the cube of reduced particle diameter σ^* when the particle concentration n is the same. Figure 2

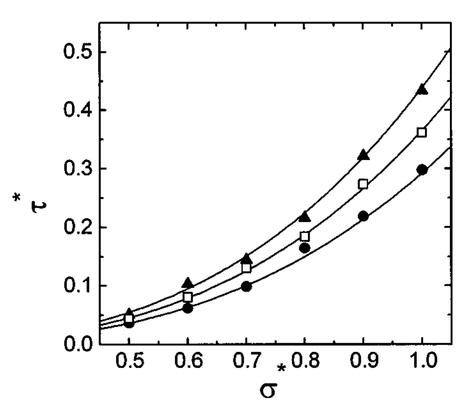

FIG. 1. The reduced shear stress of one-component ER fluids versus reduced diameter of particles. The particle concentrations: $n=0.45$ (solid triangle), 0.375 (open square), and 0.3 (solid circle). The solid lines are plots of Eq. (10).

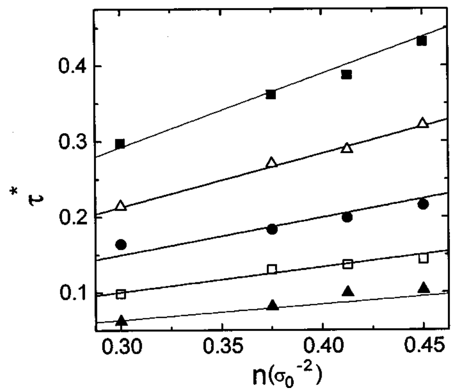

FIG. 2. The reduced shear stress of one-component ER fluids versus particle concentration. The reduced diameters of particles: $\sigma^* = 1.0$ (solid square), 0.9 (open triangle), 0.8 (solid circle), 0.7 (open square), and 0.6 (solid triangle). The solid lines are plots of Eq. (10).

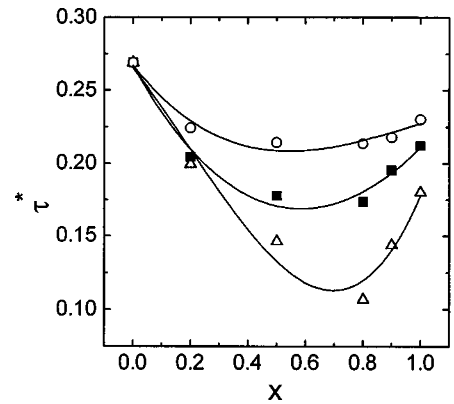

FIG. 3. The reduced shear stress of the ER fluids consisting of particles with two kinds of sizes versus the concentration ratio x of small particles. The area fraction is 0.236. The ratios of particle sizes: $\sigma_1^*/\sigma_2^* = 1/0.8$ (open circle); 1/0.7 (solid square), and 1/0.6 (open triangle). The solid lines are guides to the eye.

plots the reduced shear stress as a function of the particle concentration for the one-component ER fluids with the reduced particle diameters of 1.0, 0.9, 0.80, 0.7, and 0.6. It shows that τ^* is proportional to n. Thus, we obtain the relation of the reduced field-induced shear stress to the reduced particle diameter and concentration for one-component ER fluids as

$$\tau^* = kn\sigma^{*3}, \tag{10}$$

where the coefficient k depends on the effective polarizability β and shear flow rate $\dot{\gamma}$. In the present case, $k = 0.9733$.

This relation indicated in Eq. (10) is reasonable. It can be explained as follows: In steady flow, the particles in ER fluids will form oblique chains in the balance between the hydrodynamic and electrostatic forces [1,3,6]. We consider two systems which have the same particle concentration but different particle size. The particle diameters are σ_1 and σ_2 in the two systems, respectively. It is assumed that the arrangements of particles are the same in these two systems, i.e., that the numbers of chains are the same and shapes of chains are similar for the two systems. Now we compare the contributioins to the shear stress τ from corresponding particle couples, taken from the corresponding chains in the respective systems. The ratio between the contributions to the shear stress from the two couples can be calculated by $\tau_{ij}^1/\tau_{ij}^2 = F_{ij}^{x1}\Delta y_{ij}^1/F_{ij}^{x2}\Delta y_{ij}^2$, where τ_{ij}^α is the contribution to the shear stress from the couple of the ith and the jth particles; α is used to distinguish the two systems ($\alpha = 1$ or 2). Here $F_{ij}^{x^\alpha}$ stands for the x component of the force between the ith and jth particles and Δy_{ij}^α is the y component of the displacement between the ith and jth particles. We have $\Delta y_{ij}^1/\Delta y_{ij}^2 = \sigma_1/\sigma_2$. According to Eq. (4),

$$\frac{F_{ij}^{x1}}{F_{ij}^{x2}} = \frac{(\sigma_1)^6/(R_{ij}^{*1})^4}{(\sigma_2)^6/(R_{ij}^{*2})^4} = \frac{(\sigma_1)^2}{(\sigma_2)^2},$$

where $R_{ij}^{*\alpha}$ is the reduced distance between the ith and jth particles. Then we have $\tau_{ij}^1/\tau_{ij}^2 = (\sigma_1/\sigma_2)^3$. By using Eq. (9) in the case of two dimensions, the ratio of the shear stresses, $\tau_1/\tau_2 = [\Sigma_{i,j(\text{all pairs})}\tau_{ij}^1]/[\Sigma_{i,j(\text{all pairs})}\tau_{ij}^2] \approx (\sigma_1/\sigma_2)^3$, i.e., $\tau \sim \sigma^3$. Because τ is a statistic result, produced by all particles, it can be easily understood that $\tau \sim n$. Proceeding as before, we obtain $\tau \sim n\sigma^3$.

ER fluids consisting of particles with two different sizes were studied too. We take the diameter ratio of the two kinds of particles as 1.0/0.6, 1.0/0.7, and 1.0/0.8. The area fraction is kept constant at 0.236. Figure 3 shows plots of the reduced shear stress versus the concentration ratio of small particles. The shear stresses of the two-component ER systems are smaller than that of the corresponding one-component systems. The ER systems composed of larger particles give a higher shear stress than those composed of smaller ones. Our simulation agrees with the experimental results of Wu and Conrad [11]. It can be seen from Fig. 3, also, that the more the sizes of the two kinds of particles differ, the more the shear stress of the ER fluids decreases.

Figure 4 makes a comparison between the structures of one-component and two-component ER fluids under an external electric field and steady flow. As is often seen, the arrangements of particles of the one-component ER fluids is chain like [see Fig. 4(a)]. But for two-component ER fluids composed of particles with different sizes, the particles prefer to form thick and short chains or clusters, rather than form individual chains [see Fig. 4(b)].

To study the influence of components on the breakings of chains, we consider three kinds of chains. The first one consists of identical particles. The second kind of chain is almost similar to the first one, but a smaller particle replaces one of the particles in the chain. The third kind of chain comes from the first one, but a larger particle replaces one of the particles in the chain. Figure 5 shows the breaking in these three kinds of chains. For a homogeneous chain consisting of identical particles, the breaking position is random [see Fig. 5(a)]. A smaller particle embedded in a homogeneous chain makes the chain weaken; so the breaking appears at the place between two different particles, as shown in Fig. 5(b). A larger particle, embedded in a homogeneous chain, strengthens the chain. It follows that the breaking point prefers to the place between identical particles, rather than that between the larger particle and a smaller one [see Fig. 5(c)].

To sum up, the structures of chains or clusters have im

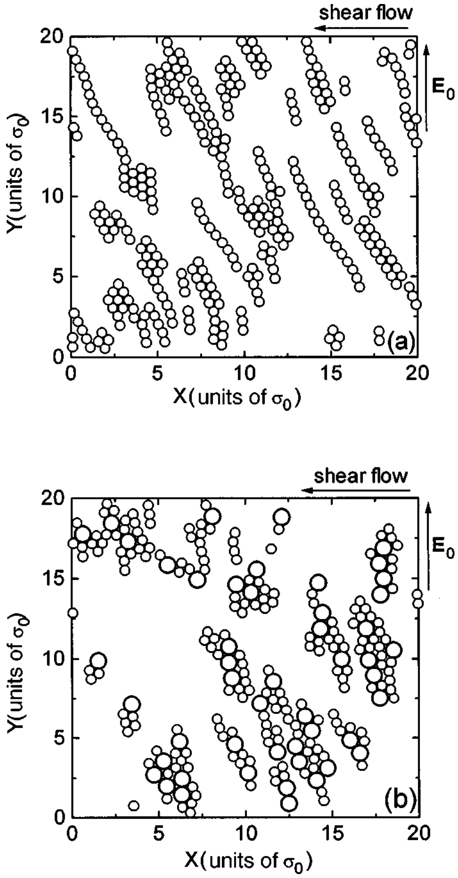

FIG. 4. The structures of ER fluids in steady shear. $\dot{\gamma}^* = 0.2$. (a) One-component ER fluid; (b) two-component ER fluid. The ratio of the diameters of two kinds of particles is $\sigma_1^*/\sigma_2^* = 1/0.6$ and the concentration ratio of large particles is 0.2.

portant effects on the ER response [11], and individual longer chains contribute to the shear stress more than thicker and shorter chains, and the clusters [21,22]. In addition, the smaller substituting particles weaken the chains and the larger ones strengthen the chains. Thus, it turns out that a decrease of the shear stress, for two-component ER fluids, containing a few smaller particles, can be attributed to two reasons: one is the weakening of the chains, and the other is related to the structures of the short and thick chains or clusters. The latter can explain the reduction of the shear stress for two-component ER fluids containing a few larger particles.

B. Effect of the dielectric properties of particles on the electrorheological response

Our study is limited to ER fluids composed of two kinds of particles which have the same size but different conductivity. The shear flow rate $\dot{\gamma}$ and reduced particle diameter σ^* are chosen to be 0.2 and 1, respectively. γ_{pf}^1 is taken as 18, and γ_{pf}^2 is taken as 4, 6, 8, or 12 corresponding to the range of $|\beta|^2$ from 0.25 to 0.73. Here the relative conductivity of the ith kind of particles is expressed as γ_{pf}^i ($i = 1,2$). The concentration ratio of particles with small conductivity is varied from zero to 1. And the total concentration of particles was always kept constant at 0.375. The simulation results are given in Fig. 6. It shows that the shear stress has an approximately linear relation to the concentration ratio of particles:

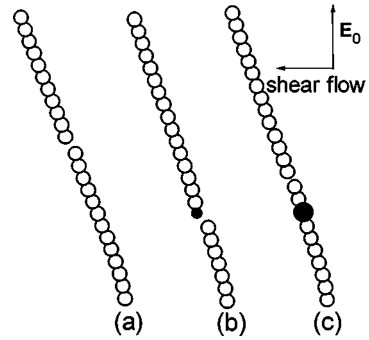

FIG. 5. Breaking of chains: (a) the chain consisting of identical particles, (b) chain containing a smaller particle, and (c) chain containing a larger particle.

$$\tau^* = \tau_1^* + (\tau_2^* - \tau_1^*)x, \tag{11}$$

where τ_1^* is the reduced shear stress for the ER system consisting of particles with relative conductivity γ_{pf}^1, and τ_2^* represents that for the ER system with relative conductivity γ_{pf}^2. Here x stands for the concentration ratio of particles with relative conductivity γ_{pf}^2.

We also investigated the structures of ER fluids composed of two kinds of particles with different conductivities, under an external electric field and steady flow. We find that the arrangements of particles are thin and long-chain-like for this kind of two-component ER fluid. The arrangements are similar to those for corresponding one-component ER fluids. This is just the reason why the shear stress of these two-component ER fluids has an approximately linear relation to the particle concentration ratio, such as Eq. (11).

IV. CONCLUSION

This work shows that molecular-dynamics simulation is an effective method to study the electrorheological response. We obtain two expressions: one is the relation of the shear stress to the particle size and concentration for one-

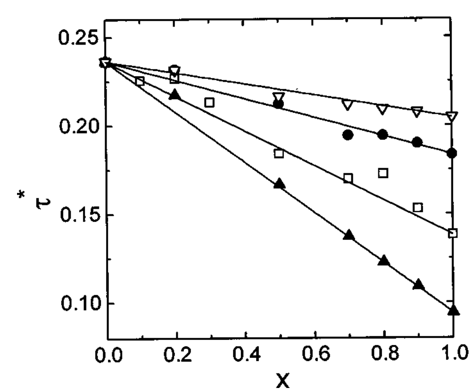

FIG. 6. The reduced shear stress of ER fluids consisting of two kinds of particles with the different conductivities versus the concentration ratio x of particles with small conductivity. The ratio of relative conductivities: $\gamma_{pf}^1/\gamma_{pf}^2 = 18{:}12$ (open triangle), 18:8 (solid circle), 18:6 (open square), and 18:4 (solid triangle). The solid lines are plots of Eq. (11).

component ER fluids; the other is the relation of the shear stress to the concentration ratio for two components ER fluids composed of two kinds of particles with different dielectric properties. They are useful to the application of ER fluids.

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