Numerical Study on Microstructures and Their Rheological Properties in Electrorheological Fluids

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We study the relation between microstructures of electrorheological (ER) fluids and their viscosity change by performing Brownian dynamics simulations of a model ER system both in a static state and in a simple steady shear. From large-scale three-dimensional simulations, it is found that (1) under no shear flow there are two principal phases in microstructural changes: first aggregation of particles into chains oriented along the field direction, and the subsequent slow coalescence of chains into columns, and (2) under a simple steady shear there are three stages in viscosity changes with increasing the field: Newtonian at a weak field, non-Newtonian at a moderate field, and Bingham plastic with yield stress at a high field.

(Received November 14, 2001; Accepted May 1, 2002)

Keywords: electrorheological fluid, brownian dynamics simulation, microstructural transition, shear viscosity, point dipole model

1. Introduction

Electrorheological (ER) fluids usually consist of fine dielectric particles suspended in a liquid of low dielectric constant. The large contrast of dielectric constant between the particles and the liquid makes the ER system easily polarizable in an electric field. When an electric field is applied to ER fluids, the induced particle dipoles create a long-range interaction that causes them to chain along the field direction and form complex column-like structures.

Moreover, the viscosity of ER fluids can be changed by an application of an electric field, which is called the ER effect. Indeed, in response to the electric field, ER fluids rapidly solidify, or at least increase their viscosity dramatically. Such a millisecond response of ER fluids is a subject of great interest because of many possible industrial applications, especially in the automotive and aerospace industries, for field-controllable fluids such as practical fast electro-mechanical actuators (e.g., fiber spinning clutches and active shock absorbers). It has been recognized that the drastic increase of the viscosity of ER fluids is due to the formation of particle chains that bridge the electrodes, and also their rheological properties are greatly affected by microstructure changes. Thus, the key to understanding and controlling the rheology of ER fluids lies in characterizing both microstructure formation kinetics and the response of the activated fluid to mechanical stresses, especially when flow is induced.

In the present work, to examine the relation between microstructure of ER fluids and their shear viscosity changes, we explore a model ER system, based on a point dipole picture without full hydrodynamic effects through the solvent such as many-body hydrodynamic interactions, lubrication force, and buoyancy effect. Although very approximate, this method is believed to capture most of the essential qualitative physics of ER fluids. Performing three-dimensional Brownian dynamics simulation of the model, we discuss the above subject, especially three aspects of ER fluids: microstructural changes, response times, and rheological properties. So far, each aspect has been discussed separately by using small scale simulations of respective different models. Thus, the present concern is to discuss all the three aspects from unified points of view by using large scale extensive simulations of a certain single model.

This paper is organized as follows. Section 2 describes the simulation model used here and numerical methods used to extract information on the system properties. Section 3 gives the simulation results and discussion thereof. Section 4 concludes the paper.

2. The Model

A model ER system studied in this paper is supposed to consist of \( N \) interacting spherical particles of dielectric constant \( \epsilon_p \) and diameter \( \sigma \), suspended in a fluid medium of dielectric constant \( \epsilon_i \) with \( \epsilon_i < \epsilon_p \) and viscosity \( \eta_r \). The model system is in a box with its volume \( V = L_x \times L_y \times L_z \), confined between two parallel electrodes at \( z = 0 \) and \( z = L_z \). An external electric field is applied along the \( z \)-direction. These particles are assumed to interact each other via short-range repulsive and point-dipole forces, and to be affected by the solvent through both Stokes drag and Brownian random forces. A constant shear strain with a shear rate \( \dot{\gamma} \) is also imposed along the \( x \) direction with a velocity field linearly varying along the \( z \) direction. We assume that the buoyancy neutralizes the gravity.

The motion of \( i \)th particle having mass \( m \) at time \( t \) and position \( \mathbf{r}_i(t) = x_i \hat{x} + y_i \hat{y} + z_i \hat{z} \) is described by the following classical equation of motion:

\[
\frac{d^2 \mathbf{r}_i}{dt^2} = \mathbf{F}_i - 3\pi \sigma \eta_r \left( \frac{d\mathbf{r}_i}{dt} - \dot{\gamma} z_i \hat{x} \right) + \mathbf{R}_i \tag{1}
\]

where \( \mathbf{F}_i \) is a total force acting on the \( i \)th particle, the second term is the Stokes drag, and the last term is Brownian random force. Under an applied electric field denoted as \( \mathbf{E}_{\text{ext}} = E_0 \hat{z} \), each particle has an induced dipole moment of \( \mathbf{p} = \alpha \epsilon_i (\sigma / \epsilon_p)^3 \mathbf{E}_{\text{loc}} \), where \( \alpha = (\epsilon_p - \epsilon_i) / (\epsilon_p + 2 \epsilon_i) \) and \( \mathbf{E}_{\text{loc}} \) is the local electric field. In the followings, we consider the case \( \alpha \ll 1 \), and thus \( \mathbf{E}_{\text{loc}} = \mathbf{E}_{\text{ext}} \). Under this situation, the

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dipolar force acting on the $i$th particle due to the $j$th particle at $r_j$ is given by

$$F_{ij}^d = \frac{3p^2}{\epsilon r_{ij}^3} \left[(1 - 3\cos^2\theta_{ij})\hat{r} - \sin(2\theta_{ij})\hat{\theta}\right] \tag{2}$$

where $p = |p|$, $r_{ij} = r_i - r_j$ with $r_{ij} = |r_{ij}|$, $\theta_{ij}$ is the angle between $r_{ij}$ and the $z$-axis, $\hat{r} = r_{ij}/r_{ij}$, and $\hat{\theta} = \hat{r} \times (\hat{r} \times \hat{z})/|\hat{r} \times (\hat{r} \times \hat{z})|$. To simulate the hard spheres and the hard sphere-wall interactions, we introduce an exponential short-range repulsive force between particles $i$ and $j$.

$$F_{ij}^{\text{rep}} = \frac{3p^2}{\epsilon \xi_c^3} \exp[-100(r_{ij}/\sigma - 1)]\hat{r} \tag{3}$$

and between particle $i$ and the two electrodes

$$F_{ij}^{\text{wall}} = \left\{ \frac{3p^2}{\epsilon \xi_c^3} \exp[-100(z_i/\sigma - 0.5)]\hat{z} - \frac{3p^2}{\epsilon (L_z - z_i)^3} \exp[-100((L_z - z_i)/\sigma - 0.5)]\hat{z} \right\} \tag{4}$$

Now the total force in eq. (1) is given by

$$F_i = \sum_{j(\neq i)} (F_{ij}^d + F_{ij}^{\text{rep}}) + \sum_j F_{ij}^{\text{kin}} + F_{i}^{\text{wall}} \tag{5}$$

where $F_{ij}^{\text{kin}}$ is the summation force on the particle $i$ due to an infinite number of images of particle $j$ reflected about the electrodes. The Brownian force $R_i$ is determined independently by a normal distribution with $\langle R_i(\alpha(t)) \rangle_{\epsilon n} = 0$ and $\langle R_i,\alpha(t)R_j,\beta(t) \rangle_{\epsilon n} = 6\pi \sigma \eta k_B T \delta_{ij} \delta_{\alpha\beta} \delta(t)$, where $R_i,\alpha$ is the $\alpha$-component of $R_i$, $\langle \cdot \cdot \cdot \rangle_{\epsilon n}$ denotes an ensemble average, $k_B$ is the Boltzmann’s constant, and $T$ is the absolute temperature.

To study the parametric properties of many different ER fluids, we here introduce scale units: the unit of time as $t_0 = 3\pi \sigma \eta^{-1}(kbT)$, length as $r_0 = \sigma$, force as $F_0 = p^2/(\epsilon \sigma^4)$, Brownian random force as $R_0 = k_BT/\sigma$, stress as $\sigma_0 = k_BT/\sigma^3$, and viscosity as $\eta_0 = t_0\sigma_0$. By using these units, eq. (1) can be rewritten in the dimensionless form as

$$A \frac{d^2r_i}{dt^2} = QF_i - \frac{dr_i}{dt} + 8P\epsilon z_i\hat{z} + R_i. \tag{6}$$

For most real parameters of ER fluids, the magnitude of $A = mk_BT/(3\pi \eta \sigma^2)^2$ in eq. (6) is very small ($\sim 10^{-10}$), so in the following simulations we neglect this inertial effect. Thus, we have two dimensionless parameters, $Q = p^2/(\epsilon \sigma^4 k_BT)$ and $Pe = 3\pi \eta \sigma^3 \gamma/(8k_BT)$, where $Q$ characterizes the ratio of the dipolar force to the Brownian force, which also indicate the strength of the electric field, and $Pe$ represents the ratio of the shear force to the Brownian force, called as Peclet number. This means that many different ER fluids with the same values of $Q$ and $Pe$ belong to the same class of fluids.

Equation (6) without inertial term is integrated with a time step $\leq 0.01/Q$ using Euler’s method. A system of $N = 2 \times 10^3$ particles in a box with $L_x = L_y = L_z = 20\sigma$ is simulated along many trajectories in the space $(Q, Pe)$. At each state point, averages are taken over $10^4$ time steps as well as 20 independent simulation runs. The particle volume fraction is 0.13, enough low to make the present dipolar model valid.

Periodic boundary conditions are imposed in the $x$ and $y$ directions, reflecting sheared boundary conditions in the $z$ direction.

During simulations, microstructures in the static state are probed by the following three order parameters:

$$c_1 = \frac{1}{N} \sum_{i=1}^{N} \exp(i\hat{b}_j \cdot r_i) \tag{7}$$

where $j = 1, 2, 3$ and the three reciprocal lattice vectors of a body-centered tetragonal (bct) lattice are $b_1 = (2\pi/\sigma)(2\epsilon/\sqrt{6} - \hat{z})$, $b_2 = (2\pi/\sigma)(2\epsilon/\sqrt{6} - \hat{z})$, and $b_3 = (4\pi/\sigma)\hat{z}$. In these order parameters, $c_3$ characterizes the formation of chains along the $z$ direction, while $c_1$ and $c_2$ characterize the structure of particle arrangement in the $x$-$y$ plane.

To study directly the relation between variation of particle interactions and different structures of the suspension, we ignore the hydrodynamic contribution to the viscosity.

3. Simulation Results

First, we discuss the final steady microstructures of the system without shears ($Pe = 0$) as a function of the parameter $Q$ after the long-time simulations from the initial disordered state shown in Fig. 1. Typical steady state structures for three values of $Q$ are shown in Fig. 2. From these results and further simulations with calculations of order parameters, it is found that this system exhibits two phase transitions as

Fig. 1 Initial random configuration of the system considered here.
the applied electric field increases. That is, in the absence of shears, there are two critical values of $Q$, associated with two critical electric fields. When $Q < Q_{c1} (\sim 40$ in the simulation), the system is a liquid with no long-range order, i.e., $c_i \leq 0.1$ for $i = 1, 2, 3$, as is shown in Fig. 2(a). When $Q_{c1} < Q < Q_{c2} (\sim 150)$, the system begins to form chains between two electrodes aligned along the field direction, but the distribution of these chains is random, i.e., $c_3 \geq 0.9$, but $c_1, c_2 \leq 0.3$, as is shown in Fig. 2(b). As $Q$ exceeds $Q_{c2}$, the system is a solid, where an ideal structure is a bct lattice, i.e., $c_3 \geq 0.9$ and $c_1, c_2 \simeq 0.8$, as is shown in Fig. 2(c). Although we cannot see the difference between Fig. 2(b) and Fig. 2(c) clearly, we have confirmed that the structure in Fig. 2(c) has the bct symmetry by calculating the order parameters $c_i$ (see the discussion on Fig. 3 below). The qualitatively similar behaviors have been obtained in previous small scale simulations.4–6)

A crucial feature of the ER response is the time required to reach the final state. In Fig. 3 we show the time evolution of three order parameters at $Pe = 0$ for $Q = 200 (> Q_{c2})$. The system is characterized by two time scales: the order parameter $c_3$ rapidly reaches the maximum value 0.95, while $c_1$ and $c_2$ slowly increase up to the maximum value 0.85. This means that following the rapid chain formation until $t < t_1 (\sim 10^{-4} t_0)$, the chains aggregate into the bct lattice structure slowly up to $t \simeq t_2 (\sim t_0)$. We have also found that the chain formation time $t_1$ is inversely proportional to $Q$.

Finally, we discuss the rheological properties of the model under simple shears, especially the shear viscosity change as a function of parameters $Pe$ and $Q$. In the present simulation, we apply a shear with different shear rate to the system which is already exposed to a fixed electric field. As is discussed below, the viscosity is found to increase with the field through three stages. In a weak field ($Q < Q'_{c1} \sim 50$), the system remains Newtonian, as is shown. In Fig. 4, although viscosity increases with the field. At this stage, the particles drift along the flow almost independently. In a moderate field ($Q'_{c1} < Q < Q'_{c2} \sim 180$), the system becomes non-Newtonian, i.e., nonlinear relation between viscosity and shear rate, as is shown in Fig. 5. The viscosity is also shown to be a decreasing function of the shear rate $Pe$. In this stage, the system has tilted and broken chains moving with the shear flow, although deformed chains are well separated, as is shown in Fig. 6. We have also observed the reconnection of these broken chains. These breaking and rejoining behavior of deformed chains has been known to result in the fluctuation of the internal stress over time. Such a fluctuation is understood as follows: A break in one chain generates some decrease in stress. On the other hand, when the broken chains rejoin, a relatively stable structure lasts for a while, leading
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Fig. 4 Effective viscosity as a function of $Pe$ at $Q = 10$ and 30.

![Graph showing effective viscosity vs. log10(Pe) for Q = 10 and Q = 30.]

Fig. 5 Effective viscosity as a function of $Pe$ at $Q = 80$ and 120.

![Graph showing effective viscosity vs. log10(Pe) for Q = 80 and Q = 120.]

to the stress increase. Thus, the viscosity is calculated by averaging over some time steps for $Q > Q'_c$. In a high field ($Q > Q''_c$), the system becomes Bingham plastic with yield stress, as is shown in Fig. 7. This means that the particles do not move when the applied stress is below the yield stress. In Fig. 7 the yield shear stress, $\tau_{yield}$, is plotted as a function of $Pe$ at $Q = 200$, which is obtained from simulation data of the stress-strain characteristic. The static yield stress $\tau^*_{yield}$ is obtained from Fig. 7 by extrapolating the curve to $Pe = 0$. We estimate $\tau^*_{yield}/\tau_0 \simeq 290$ for $Q = 200$. We have also found that the static yield stress is proportional to $Q^{2.8}$. In this stage, we can see the condensed phase of fragmented chains, as is shown in Fig. 8, where breaking and rejoining phenomena occur frequently.

4. Conclusion

We have performed large-scale three-dimensional Brownian dynamics simulations of the model ER system under applied electric fields with and without shears. We have examined the...

Fig. 7 Yield shear stress $\tau_{yield}$ as a function of $Pe$ at $Q = 200$.

![Graph showing yield shear stress vs. Pe for Q = 200.]

Fig. 8 Snapshot of configuration under a simple shear, projected on the $x$-$z$ plane at $Q = 200$. Particles only within $8\sigma \leq y \leq 12\sigma$ are plotted to avoid the overlap of particles projected.

![Snapshot showing configuration under simple shear at Q = 200.]

Fig. 6 Snapshot of configuration under a simple shear, projected on the $x$-$z$ plane at $Q = 100$. Particles only within $8\sigma \leq y \leq 12\sigma$ are plotted to avoid the overlap of particles projected.
relationship between microstructures of ER fluids and their rheological properties. It has been found (1) two principal phases in structural changes under no shear flow, and (2) three stages in viscosity changes with the field under a simple steady shear flow.

Now, we briefly discuss the reliability of the present simulation studies. For a real ER system, such as alumina particles in petroleum oil, one has $\epsilon_f \sim 2$, $\epsilon_p \sim 10$, $\eta_f \sim 0.01 \text{ Pa} \cdot \text{s}$, $\sigma \sim 1 \mu\text{m}$, $T \sim 300 \text{ K}$, and the mass density of particles $\rho \sim 1 \text{ g/cm}^2$. In this case, one can estimate $t_0 \sim 24 \text{ s}$ and $\tau_0 \sim 4 \times 10^{-3} \text{ Pa}$. Thus, the chain formation time $t_1$ is of the order of a millisecond, while the bct lattice formation time $t_2$ is of the order of minutes. As for the critical electric field, $E_{c_1}$ associated with $Q_{c_1} = 40$ is of the order of a few kV/mm. The static yield stress is also estimated to be a few Pa for $Q = 200$. These simulation results estimated are in good range for experiments.

Thus, it would be very interesting to see if the present findings hold in real experiments. However, further studies to relate many effects ignored here with real situations are needed in order to simulate real ER fluids reliably. An improvement in this direction is now under way.

Acknowledgements

The authors thank Prof. M. Tokuyama for valuable discussions.

REFERENCES