Effects of nonmagnetic interparticle forces on magnetorheological fluids

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Abstract. Effects of nonmagnetic interparticle forces on the on- and off-state behavior of MR fluids are investigated experimentally and with particle-level simulations. Suspensions of iron particles in an aliphatic oil are modified by surface-active species. The modifications significantly alter the off-state properties, but have little impact on the field-induced stresses. Simulations show similar behavior. Off-state rheological properties are strongly influenced by van der Waals forces and modifications of the short-range repulsive forces. Field-induced stresses are less sensitive to the nonmagnetic forces.

1. Introduction

Much research has illustrated that the field-induced behavior of magnetorheological (MR) fluids can be explained largely in terms of magnetostatic forces and their competition with hydrodynamic forces. However, other properties, such as off-state viscosity, sedimentation, redispersability, and durability, are strongly influenced by forces other than magnetic forces. Off-state viscosity of particulate suspensions in general depends strongly on colloidal forces [1]. Sedimentation can be controlled by various additives, some of which may act by inducing colloidal interactions between particles [2, 3]. van der Waals forces, as well as remnant magnetization, can influence the stability and redispersability of suspensions [4]. Ulicny et al. [5] found that surface active additives significantly reduced the off-state drag in an MR clutch, and improved the durability of MR fluids. Surfactants can also help to disperse particles [6]. These observations are consistent with the added chemicals altering the nonmagnetic interparticle forces.

Here we investigate the effects of nonmagnetic interparticle forces on the on- and off-state rheological properties of MR suspensions. Specifically, we experimentally investigate the effects of surface-active additives on rheological properties. We also employ particle-level simulations to investigate the effects of van der Waals forces and modifications of short-range repulsive forces on rheological properties.

2. Experimental

The MR fluids were prepared using two iron powders, with average diameters of 2 and 8 µm (BASF HS and CM, respectively). The base liquid was Mobil SHF 21—a polyalphaolefin (PAO) of molecular weight of approximately 280. Three MR suspensions were prepared, each composed of 30 ± 1 vol% iron particles in PAO. One was composed of only the small diameter (HS) particles, one composed of only the large diameter (CM) particles, and one composed of a bimodal mixture of equal masses of each type of particle. These suspensions are referred to as the “nontreated” suspensions.

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A second set of MR fluids were prepared by modifying aliquots of the MR fluids described above. To a portion of each of the fluids, lithium 12-hydroxy stearate (LiHS; City Chemical) and zinc-o,o-di-n-butylphosphorodithiolate (ZBPD; Flexsys) were added, in ratios of $3.47 \times 10^{-3}$ g LiHS/g MR fluid and $2.78 \times 10^{-4}$ g ZBPD/g MR fluid. The resulting suspensions are referred to as “treated” suspensions.

Off-state rheological properties (no applied magnetic field) were measured using a Haake RS150 rheometer with serrated parallel disks. The shear stress was measured as a function of shear rate, as the shear rate was first increased from 0 to 400 s$^{-1}$, and then decreased from 400 to 0 s$^{-1}$. Results were reproducible after the first such shear rate “loop.” Rheological properties in the presence of a magnetic field were measured using an Anton Paar MCR501 magnetic rheometer. After placing the sample between parallel disks, the shear stress was measured as a function of shear rate, with the shear rate increased in steps from 0 to 400 s$^{-1}$. Experiments were repeated for various magnetic field strengths. Shear stress-shear rate data were subsequently fit with a Bingham model to extract the yield stress and plastic viscosity.

### 3. Simulations

Particle-level simulations were used to investigate effects of various forces on MR fluid behavior. We used the point-dipole simulation method that we have employed previously [7, 8] modified by adding van der Waals forces, and altering the short-range repulsive forces to account surface modifications.

van der Waals forces are represented by the near-field form [9] with a small separation cutoff $h_{\text{min}}$.

$$F_{ij}^{\text{vdw}} = \begin{cases} \frac{A}{24} \sigma_{ij} \ell \frac{e_r}{r_{ij}} & \text{for } h_{ij} > h_{\text{min}}, \\ \frac{A}{24} \sigma_{i} \ell \frac{e_r}{h_{\text{min}}} & \text{for } h_{ij} \leq h_{\text{min}}, \end{cases}$$

where $A$ is the Hamaker coefficient, $\sigma$ is the particle diameter, and $h_{ij} = r_{ij} - \sigma$ is the separation between the surfaces of spheres $i$ and $j$. Here we use a cutoff of $h_{\text{min}}/\sigma = 10^{-3}$. Short-range repulsive forces are modeled with the short-ranged function

$$F_{ij}^{\text{rep}} = -F_0 \exp [-\kappa (h_{ij} - h^*)/\sigma] e_r,$$

where

$$F_0 = \begin{cases} \frac{3}{10} \mu_0 \sigma^2 \beta_M^2 H_0^2 & \text{for linear magnetization}, \\ \frac{\pi}{12} \mu_0 \sigma^2 M_s^2 & \text{for saturated magnetization}, \end{cases}$$

is the magnitude of the magnetostatic force, $\kappa = 10^3$ for all results reported here, and $h^*$ represents a steric barrier. We include $h^*$ to model the effect of adsorbed or grafted species (i.e., $h^* \approx 2\ell$, where $\ell$ is the thickness of a densely grafted layer). Analogous expressions are employed to describe interactions between the particles and the bounding surfaces.

The ratio of hydrodynamic to field-induced forces defines a dimensionless shear rate, $\tilde{\gamma} = 3\pi \eta_c \dot{\gamma}/F_0$, which is set to $10^{-3}$ for the results presented here. The dimensionless shear rate is related to the Mason number by $\tilde{\gamma} = 32\text{Mn}$. The ratio of van der Waals and field-induced forces is

$$\tilde{F}_0^{\text{vdw}} = \frac{A/24\sigma}{F_0} = \begin{cases} \frac{2A}{9\pi \mu_0 \sigma^3 \beta_M^2 H_0^2} & \text{linear magnetization}, \\ \frac{2A}{2\pi \mu_0 \sigma^3 M_s^2} & \text{saturated magnetization}. \end{cases}$$

Typical values for $\tilde{F}_0^{\text{vdw}}$ are estimated using the parameter values $A = 50 \times 10^{-20}$ J [9], $\mu_0 M_s = 2$ T, and $\beta_M = 1$. For $\sigma = 1.0 \mu$m, $F_0^{\text{vdw}}$ is roughly $1.0 \times 10^{-7}$ for large magnetic field strengths (saturated magnetization) and increases with decreasing field strength; for a flux density of 0.1 T, $F_0^{\text{vdw}} \approx 4 \times 10^{-6}$. Although these values for $\tilde{F}_0^{\text{vdw}}$ are much less than 1, we show below that van der Waals forces can significantly impact the field-induced stresses in this range.
Simulations were performed by randomly placing 150 spheres in a periodic simulation cell of dimensions \( (L_x, L_y, L_z) = (10\sigma, 5\sigma, 5\sigma) \), corresponding to a particle volume fraction of \( \phi = 0.314 \). Shear flow is simulated to a strain of 5. The shear stress, calculated in a conventional manner \([8]\), becomes steady after a strain of 1. Results reported here are averaged over the strain interval \( \gamma = 1–5 \), and over 5 different initial configurations for each set of simulation parameters.

4. Results

The field-induced yield stress is plotted as a function of flux density in Fig. 1, for both the nontreated and treated suspensions. The surface treatments have little effect on the field-dependent yield stress. This is in contrast to results for thicker coatings where the field-induced stress is significantly decreased by the coating \([10, 11, 12]\).

![Figure 1. Yield stress vs flux density for various monodisperse and bimodal suspensions (30 vol%), with and without surface treatments (ZBPD + LiHS).](image)

Off-state rheological properties were also measured for these systems. Shear stress-shear rate data were fit with the Bingham equation. The yield stresses and plastic viscosities obtained from these fits are presented in Figs. 2 and 3, respectively. The surface treatments significantly reduce the off-state yield stress as well as the plastic viscosity.

![Figure 2. Off-state yield stress for various nontreated and treated suspensions.](image)

![Figure 3. Off-state plastic viscosity for various nontreated and treated suspensions.](image)

Simulation results for the on-state properties are illustrated in Fig. 4, where the average dimensionless shear stress, \( \langle \tilde{\tau} \rangle = \langle \tau \sigma^2/F_0 \rangle \), is plotted as a function of \( \tilde{F}_{0,\text{vdw}} \) for several values of \( h^* \). The results for \( h^* = 0 \) show that the field-induced stress is independent of \( \tilde{F}_{0,\text{vdw}} \) for small \( \tilde{F}_{0,\text{vdw}} \), and then increases with \( \tilde{F}_{0,\text{vdw}} \) for larger values. For \( \tilde{F}_{0,\text{vdw}} = 1.5 \times 10^{-6} \), the stress is 26% larger than in the absence of van der Waals forces. Thus, van der Waals forces significantly impact the rheology for small values of \( \tilde{F}_{0,\text{vdw}} \); however, values of \( \tilde{F}_{0,\text{vdw}} \gtrsim 10^{-6} \) are typically only achieved for relatively small magnetic field strengths or small particle diameters. The effect of steric barrier thickness \( h^* \) on the stress is also illustrated in Fig. 4 (for \( \tilde{F}_{0,\text{vdw}} = 10^{-6} \)). Increasing \( h^* \) causes the stress to decrease. For \( h^*/\sigma = 10^{-3} \),...
the field-induced stress is reduced to that obtained in the absence of van der Waals forces. A decrease in stress with increasing $h^*$ is expected, since larger interparticle separations result in weaker van der Waals forces. As mentioned above, such trends are observed experimentally, where sufficiently thick coatings can cause a decrease in the field-induced stresses [10, 11, 12].

![Figure 4. Field-induced shear stress vs $F_0^\text{vdw}$ for several values of $h^*$.](image)

![Figure 5. Off-state shear stress vs $F_0^\text{vdw}$ for several values of $h^*$.](image)

The stress is much more sensitive to van der Waals forces and the steric barrier in the off state, as illustrated in Fig. 5 where the shear stress is plotted as a function of $F_0^\text{vdw}$ for two values of $h^*$ (for the off state, $F_0^\text{vdw} \equiv A/24\sigma F_0$ is scaled by the repulsive force magnitude $F_0$). In contrast to the field-induced stress, the off-state stress (at low shear rates) approaches zero as $F_0^\text{vdw} \to 0$. The effect of the barrier is more pronounced as well, as the stress is reduced by 88% as $h^*/\sigma$ is increased from 0 to $10^{-3}$ (for $F_0^\text{vdw} = 1.2 \times 10^{-6}$).

5. Conclusion

Effects of nonmagnetic interparticle forces on the on- and off-state behavior of MR fluids are investigated experimentally and with particle-level simulations. Surface modifications significantly alter the off-state properties, but have little impact on the field-induced stresses. Simulations show similar behavior. Off-state rheological properties are strongly influenced by van der Waals forces and modifications of the short-range repulsive forces. Field-induced stresses are less sensitive to the nonmagnetic forces.

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