Role of organic coating on carbonyl iron suspended particles in magnetorheological fluid

Department of Polymer Science and Engineering, Inha University, Incheon 402-751, Korea
M. S. Jhon
Department of Chemical Engineering, Carnegie Mellon University, Pittsburgh, Pennsylvania 15213

(Submitted on 9 November 2004; published online 17 May 2005)

Carbonyl iron (CI) has been widely used as a suspended particle in magnetorheological (MR) fluids. However, pristine CI-based MR fluids have several drawbacks, including severe sedimentation of the CI particles due to the large density difference with the carrier liquid, difficulties in redispersion after caking, abrasion of device surfaces during long-term operation, and rust of iron by oxidation. To overcome these shortcomings, we coated the CI particles with a poly(vinyl butyral) (PVB) shell. CI and CI-PVB particles were suspended in mineral oil and their MR characteristics were examined via a rotational rheometer in a parallel plate geometry equipped with a magnetic field supplier. Yield stress and flow response (shear stress and shear viscosity) were investigated at magnetic field strengths ranging from 0 to 343 kA/m. Although the MR properties, such as yield stress and shear viscosity of CI-PVB based MR fluids, changed slightly compared with those of the pristine CI based MR fluid, the dispersion qualities (e.g., sedimentation stability) were noticeably improved. © 2005 American Institute of Physics. [DOI: 10.1063/1.1853835]

I. INTRODUCTION

Magnetorheological (MR) fluids, suspensions of magnetic particles having a size on the order of a micron dispersed in a nonmagnetic medium, are regarded as one of the smart materials under the influence of an external magnetic field, they can be reversibly transformed from a fluidlike to a solidlike state within milliseconds by showing drastic changes in rheological properties (e.g., enhanced yield stress and viscoelasticity).1,2 Ferro and ferrimagnetic materials can be used as a dispersed phase of MR fluids since these particles are easily magnetized under an external magnetic field. MR fluids can exhibit a yield stress of 10–100 kPa within a reasonable magnetic field strength range. As compared to electrorheological fluids, MR fluids can be relatively easy to commercialize since a magnetic field is more stable than an electric field. However, MR fluid must have certain features including: noncorrosive, active over a broad temperature range, stable against settling, irreversible flocculation, chemical stability, and high magnetic saturation. Furthermore, it should have a large field-induced yield stress with small apparent viscosities in the absence of an applied magnetic field. Since its discovery, there have been a large number of studies on MR characteristics and their applications including active controllable dampers, torque transducers, and position controllers. The majority of research on MR fluids is focused on yield stress as a function of applied magnetic field strength for the optimum formulation in each specific application. However, the fundamental studies on the rheology of MR fluids are still lacking. Since most of the magnetic particles used in MR fluids have high densities and rigid structures, sedimentation and abrasion of the equipment becomes a concern. In order to resolve these issues, we synthesized composite particles with iron core particles covered by a poly(vinyl butyral) (PVB) (Mowital, Hoechst) shell. The PVB coating prevents the cake formation among the CI particles, reduces rusting of CI particles, and protects the device from abrasion. On the other hand, it is well-known that MR fluid has been used as an abrasive finishing (polishing) material,3 since CI used in MR fluids has enough hardness to abrade device. Therefore, analogously the coating of polymer4 (PVB) possessing low hardness can protect the device from abrasion. In this work, we investigated not only the rheological properties but also dispersion stability of MR fluids under applied magnetic fields.

II. EXPERIMENT

As a suspended particle in MR system, we used carbonyl iron (CI, standard CM grade, BASF, Germany), whose average particle size and density are 7 μm (polydispersity: 2.56 × 10−4) and 7.86 g/cm3, respectively. Magnetic particles were prepared in spherical shape with a PVB coating via a

FIG. 1. SEM image of CIPVB particle showing the increase in overall particle size and surface roughness. (Inset is the SEM image of pristine CI). Scale bars indicate 5 μm.
solvent evaporation technique. PVB was dissolved in chloroform, and the CI particles were transferred into this solution which was very quickly added into the deionized water containing stabilizer PVA, Dongyang Chem. Co., emulsifier (sodium dodecyl sulfate, SDS, Aldrich), and triblock copolymer (PEG15PPG53PEG15, Aldrich) in continuously agitated water. The dispersion medium was mixed with a mechanical stirrer at 400 rpm at the ambient condition. After 2 days, all of the chloroform was evaporated, leaving CI particles completely coated with PVB. These composite particles were washed with deionized water thrice to remove physically adsorbed reactant at the surface of the composites. To prepare the MR fluid, these composite particles were dispersed in a mineral oil (MO, Aldrich). In this paper, concentration of both the CI and CI-PVB composite particles in MO was set to be 20 vol %. MR characterizations were performed at 20 °C via a rotational rheometer (Physica MCR 300, Stuttgart, Germany) equipped with a magnetorheological device (MRD 180). A parallel-plate measuring system was made of nonmagnetic metal to prevent the occurrence of radial magnetic force components on the shaft of the measuring system. The magnetic field direction was set to be perpendicular to the flow direction.

III. RESULTS AND DISCUSSION

The surface of the synthesized CI-PVB particles becomes rugged as a result of the PVB coating, as shown in the scanning electron microscope (SEM) image (Fig. 1). The particle size of CI-PVB, measured by dynamic light scattering (ELS 8000, Otsuka, Japan), is 7.18 µm (polydispersity: 4.82×10⁻¹) which is slightly larger than that of CI particles due to the capsulation of PVB. After PVB coating on CI, particle size distribution became broader than that of the pristine CI. In general, monodisperse particle suspension made the better MR fluid. But, both CI and CI-PVB particles do not have monodispersity. The density of the CI-PVB at 30 °C was measured, to be 3.2 g/cm³ via a pycnometer, which is about half of the density of pristine CI. The PVB coating on CI particles can improve the sedimentation and redispersion since the PVB coating prevents the cake formation among CI particles, reduces rust for CI particles, and protects the device from abrasion. Figure 2 shows the magnetization curves for the powder form of CI and CI-PVB. Hysteresis behavior of the CI and CI-PVB powders is quite similar, but the magnetic saturation of CI is larger than that of CI-PVB. Ideally, this means that a MR suspension based on CI powder will have a stronger MR effect than CI-PVB powder if the magnetization was the only determining factor for the MR effect. Rheological behavior of the MR suspensions was investigated under steady shear flow and a static magnetic field. Figures 3(a) and 3(b) show shear stress and shear viscosity as a function of shear rate for both the CI and the CI-PVB based MR fluids under four different external magnetic field strengths ranging from 0 to 343 kA/m. With the increase in magnetic field strength, the shear stresses leveled off for the entire shear rate region. Both MR fluids exhibited the Bingham plastic behavior in the absence of the magnetic field, implying the formation of the stable chain structures of magnetized particles. Both fluids exhibit non-Newtonian behavior in the absence of the magnetic field, probably due...
to the remnant magnetization of the magnetic particles. Yield stress can be obtained by extrapolating the shear stress values to the zero shear rate. The yield stresses at 343 kA/m are 15.5 kPa for the CI suspension and 10.5 kPa for the CI-PVB suspension, respectively. The off-state viscosity of CI-PVB suspension is slightly higher than that of the CI suspension. In the MR device design, it is important to know the minimum "active fluid volume (AFV)" $V$, which is proportional to the ratio of fluid viscosity without an applied magnetic field, $\eta_{\text{off}}(\eta_{\text{pl}})$, to the square of the yield stress under an applied magnetic field, $\tau_y^2$.

$$V = k \left( \frac{\eta_{\text{pl}}}{\tau_y^2} \right) \lambda W_m \text{ with } \eta_{\text{pl}} = \lim_{\gamma \to \infty} \frac{\partial \tau}{\partial \gamma}$$

Here, $k$ is a constant and $V$ can be regarded as the necessary AFV in order to achieve the desired control ratio $\lambda$ at a required controllable mechanical power level $W_m$. For a shear flow, $k=1$. The MR fluid having small $V$ is known to have better performance with both higher yield stress and lower off-state viscosity in the MR fluid. Using Eq. (1), the minimum AFV can be obtained under 343 kA/m. Off-state-plastic viscosities were 0.8 Pa s for the CI and 1.2 Pa s for the CI-PVB, respectively. The plastic viscosity was determined at an infinite value of shear rate. The ratio $\eta_{\text{pl}}/\tau_y^2$ was $3.33 \times 10^{-9}$ in CI and $1.08 \times 10^{-8}$ in CI-PVB, respectively. It can be noticed that considerably small ratio of $\eta_{\text{pl}}/\tau_y^2$ was attained by pristine CI suspension as compared to the CI-PVB suspension. Yield behaviors with increasing shear rate were consistent for all magnetic field strengths tested implying that particle cluster structures constructed within fluid under the applied magnetic field are continuously broken and reformed under the shear rate of 100 s$^{-1}$. The shear thinning behaviors observed in the shear viscosity stem from the changes in internal structures under shear deformation. In other words, particle structures developed under external magnetic fields remained same until a shear rate of 100 s$^{-1}$. Figure 4 shows the sedimentation ratio as a function of time for both the CI and the CI-PVB in MO media, in which the settling of the macroscopic phase boundary between the concentrated suspension and the supernatant liquid was observed. The MR fluids with and without the PVB coating on the CI were set in a static condition until it reached the asymptotic values. The decreased sedimentation ratio clearly demonstrated the role of CI-PVB in sedimentation, showing that the CI-PVB suspension has a better stability than the CI suspension.

IV. CONCLUSIONS

Two MR fluids with or without PVB coating on CI particles were prepared and the MR behaviors and sedimentation characteristics of these fluids were examined. The PVB coated CI particle not only improves sedimentation behavior with a decreased particle density but also maintains yield behaviors with constant shear stresses in a broad range of shear rates. In addition, we also applied the active fluid volume concept to interpret our MR fluid systems.

ACKNOWLEDGMENT

This study was supported by research grants from KOSEF through the Applied Rheology Center, Korea. This work was also partially supported by the Advanced Environmentally-benign Structural Materials R&D Center, Korea (2004).