Effect of storage and ball milling on the sedimentation and rheology of a novel magnetorheological fluid

H B Cheng¹, P Hou¹, Q J Zhang¹ and N M Wereley²

¹State Key Laboratory of Advanced Technology for Materials Synthesis and Processing, Wuhan University of Technology, Luoshi Road 122, Wuhan, 430070, P.R.China

²Department of Aerospace Engineering, University of Maryland, 3180 Martin Hall, College Park, MD 20742, USA

E-mail: zhangqj@whut.edu.cn

Abstract. In this study, the sedimentation stability and rheology of a new magnetorheological fluid formulation were experimentally investigated before and after being stored for 24 months, as well as after being milled. The results reveal that, after undergoing 24 months of steady storage, the off-state viscosity and field-induced yield stress of the MR fluid remained constant, and that its flowability and redispersibility were maintained. After being subjected to ball milling for 2 hours at 200 rpm and 300 rpm, the field-induced yield stress was also maintained.

1. Introduction
Magnetorheological (MR) fluids are suspensions composed of magnetizable particles, nonmagnetic liquid and additives, and show a unique ability to undergo rapid, nearly completely reversible, significant changes in their apparent viscosity upon application of an external magnetic field [1]. Typically, this change is manifested by the development of a yield stress that monotonically increases with applied magnetic field. This yield stress controllability has stimulated considerable research in developing controllable shock absorbers, clutches, engine mounts, alternators, power steering pumps, control valves, and artificial joints. However, MR fluids are thermodynamically unstable systems which tend to settle and may even form a hard “cake” at the bottom of its container in time, which limits their application in systems where the MR fluid remains quiescent for long intervals. Therefore, investigating the long-term stability of MR fluids is very important for civil engineering [2] and single use systems like energy absorbers for crashworthiness systems such as helicopter seats. In this work, we focus on how off-state viscosity and field-induced yield stresses of a novel MR fluid are affected by long-term storage and by ball milling (simulating in use shear thickening), to evaluate the stability and durability of the MR fluid.

2. Experimental
The samples used in this study were (1) freshly mixed MR fluid (MRF-140WUT, Wuhan University of Technology, China)[3], which is composed of carbonyl iron 82 wt%, silicone oil 15 wt% and additives 3 wt%, (2) MR fluid after being stored for 24 months, and (3) MR fluids after ball milling.
Ball milling was used to simulate the durability, or in-use shear thickening, of the MR fluid. The process was carried out under vacuum conditions, using a stainless steel pot (height 7 cm, ID 7.5 cm) and stainless steel balls (diameter 6 mm and 10 mm), the weight ratio of balls to MR fluid was 5; The rotation rate of the pot was 200 and 300 rpm, respectively. Magnetorheological characterization was carried out using a parallel-plate advanced rheometric expansion system (ARES, TA Co.) equipped with a magnetic field supplier at room temperature.

3. Results and discussion

3.1. Dispersion Stability

The samples of MR fluid were transferred to 10 ml graduated test tubes and placed in still storage. The volume of the supernatant liquid can be obtained by observing the phase boundary between the supernatant liquid and the concentrated suspension, until reaching an asymptotic value. The sedimentation ratio is defined as:

\[
\text{Sedimentation Ratio} (%) = \frac{\text{volume of the supernatant liquid}}{\text{volume of the entire suspension}} \times 100\%
\]  

(1)

Based on experimental observations, sedimentation ratios were calculated according to Eq. (1), and plotted as a function of time in figure 1. Sedimentation ratio is inversely proportional to the dispersion stability of the MR fluids. Figure 1a shows the dispersion stabilities of the fresh MR fluid (■ #0) and the redispersed MR fluid following 24 months of storage (■ #1). Figure 1b shows the dispersion stability of the stored MR fluid after ball milling for 2 hours at 200 rpm (■ #2) and 300 rpm (● #3), respectively. In figure 1, sample #1, #2 and #3 were made according the procedures as follows: at first the stored MR fluid was stirred to create a homogeneous suspension as sample #1, and then portions of sample #1 were milled with a ball mill for 2 hours at the rotation rate of 200 and 300 rpm, respectively, thus obtaining sample #2 and #3. From figure 1a, the sedimentation ratio of the novel MR fluid was not affected by storage, and both the fresh and stored MR fluids exhibited nearly identical sedimentation behavior. The segregation phenomenon occurred slowly, and reached equilibrium after about 105 days. The sedimentation ratio increased at the beginning correspondingly fast, and then slowly, finally kept constant, the maximum is about 11%. Moreover, the novel MR fluid can be easily redispersed after 24 months steady storage through a weak stirring force (300 rpm×10 minutes). Comparing the sedimentation ratios of the MR fluid before and after ball milling (samples #1, #2 and #3 in figure 1b), it was found that the sedimentation ratio decreased by about 77% after milling, and extent of the reduction increased with the rotation rate of the pot. These effects are the result of non-spherical particles and superfines produced after milling (see fig. 3 below), and are analogous to methods of improving MR fluid stability of by introducing non-spherical particles [4] and/or nanoparticles [5].

Figure 1. Sedimentation ratio versus time. (a): ■ #0 fresh MR fluid; ■ #1 redispersed MR fluid after 24 months of storage; (b): ■ #2 milled 2 hours at 200 rpm; ● #3 milled 2 hours at 300 rpm

3.2. Rheological properties analysis
3.2.1. Off-state viscosity. Figure 2a shows the off-state viscosity of fresh MR fluid (☐ #0) and after 24 months of storage (■ #1). Both MR fluid samples exhibited typical shear thinning behavior, decreasing monotonically with shear rate. The difference between the two viscosity curves is small. Even though the MR fluid was placed in static storage for 24 months, its off-state viscosity remained constant after being redispersed. This result indicates that this MR fluid is stable for long-term storage and no irreversible sedimentation and agglomeration takes place during the storage process. Figure 2b shows the off-state viscosities of MR fluids (#2 and #3) after milling at different rotation rates as a function of shear rate. Compared with that of the original sample free of milled (#1), the viscosities averagely increase by 230%, whether the rotation rate is 200rpm or 300rpm. That is to say, ball milling will increase the viscosity of MR fluids, which is in agreement with the results of the above stability test and results of Ulicny [6]; it is also similar to the so-called in-use-shear-thickening [8]. Increased viscosity may be due to the increase in aspect ratio of the particles [8], because some magnetic particles were volumetrically reduced after milling, and also partly due to the increase of

Figure 2. Off-state viscosity versus shear rate. (a): ☐ #0 fresh MR fluid; ■ #1 redispersed MR fluid after 24 months of storage; (b): ☐ #2 milled 2 hours at 200 rpm; ● #3 milled 2 hours at 300 rpm

Figure 3. SEM for different ball milling rates. (a): unmilled; (b): milled 2 hours at 200 rpm; (c): milled 2 hours at 300 rpm. Scale bars indicate 5µm (a), and 10 µm (b and c), respectively.

Figure 4. Yield stress versus field strength. (a): ☐ #0 fresh MR fluid; ■ #1 redispersed MR fluid after 24 months of storage; (b): ☐ #2 milled 2 hours at 200 rpm; ● #3 milled 2 hours at 300 rpm
superfine particulates spalled from the surface of the carbonyl iron particles after milling. Since even a small quantity of carbonyl iron particles are replaced with superfinest, the off-state viscosity would be increase [7]. To validate this hypothesis, photomicrographs of the particles in MR fluid before and after milling were observed in an SEM—Hitachi S-4800 FESEM—and shown in figure 3 (a-c). It was found that the larger particles were at first worn down and some non-spherical abrasion particles and superfine particles appeared after milling (see figure 3 b and c). Ulicny [6] obtained similar results in a durability test of an MR fan clutch. Interestingly, this MR fluid did not become too dense to flow, but presented better flowability even after being milled 2 hours at 300 rpm. That is to say, the MR fluid did not fail after being subjected to severe wear; the yield stress measurements below support this conclusion.

3.2.2. Field-induced yield stress.

The field-induced yield stress of the MR fluid samples were tested with magnetic flux density ranging from 0 to 0.6T, at the shear rate of 1 s\(^{-1}\) and shown in figure 4. Figure 4a shows the relationship of magnetic flux density and the field-induced yield stresses of the MR fluid before and after 24 months of storage. Interestingly, the yield stress of the MR fluid is nearly identical for the fresh and stored MR fluids. Together with the above demonstration of the stability of MR fluid viscosity before and after 24 months of storage, we conclude that the MR fluid has very stable properties even after being stored for 24 months and redispersed. Figure 4b shows the field-induced yield stresses of MR fluid samples before and after milled increase. Under weak magnetic field (lower than 0.45T); the yield stresses of the three samples exhibit similar trend of increasing yield stress with magnetic field. The results may imply that the field-induced shear stress of the MR fluids is not very sensitive to the shape of particles under weak field, or the amount of broken particles is not enough to induce large change of particles-chain in MR fluids under weak field. For magnetic flux density greater than 0.5T, yield stress of the two milled samples #2 and #3 exceeded that of the unmilled sample #1. This behavior is analogous to the results observed when adding nanometer lithium magnesium silicate into MR fluids [9]; it further validates that one cause of increasing viscosity is the superfine particles produced in the milling process.

4. Conclusions

The novel MR fluid formulated here was proven empirically to have a high level of stability. There was measurable change in neither the viscosity nor yield stress between freshly mixed MR fluid and fluids subjected to 24 months of quiescent storage. In addition, there was no measurable worsening of the field-induced yield stress after ball milling at 300 rpm for 2 hours, although viscosity increased because of the increase of weight fraction of superfines and non-spherical as the result of milling.

References

[1] Ginder J M, 1998, Behavior of magnetorheological fluids. *MRS Bull.*, **23** 26
[2] Dyke S J, Spencer B F, Jr, Sain M K and Carlson J D. 1998, *Smart Materials and Structures*, **7** 693-703.
[3] Cheng H B et al. 2006 *China Pat.*, ZL200610124728.0
[4] Pu H T, Jiang F J, Yang Z L, Yan B and Liao X, 2006, *Journal of Applied Polymer Science*, **102**, 1653-1657.
[5] Ngatu G T and Wereley N. 2007 *2007IEEE Transactions on Magnetics*, **43(6)** 2474-2476
[6] Ulicny J C, Balogh M P, Potter N M and Waldo R A. 2007, *Materials Science and Engineering A.* **443** 16–24
[7] Carlson, J.D. 2002. *Journal of Intelligent Material Systems and Structures*, **13(7–8)** 431–435.
[8] Kwon T M, Jhon M S and Choi H J. 1998, *Journal of Molecular Liquids.*, **75** 115-126.
[9] Cheng H B, Zhang J, Feng J, Guan J G, Zhang Q J and Qu W L. 2006, *Journal of functional material* **7(37)** 1166-68