Properties and mechanism of ionic liquid/silicone oil based magnetorheological fluids

Yu Tong\textsuperscript{a,b}, Penghui Zhao\textsuperscript{a}, Xiaoguang Li\textsuperscript{a}, Ning Ma\textsuperscript{c}, Xufeng Dong\textsuperscript{a}, Chenguang Niu\textsuperscript{d,e}, Zhanjun Wu\textsuperscript{b} and Min Qi\textsuperscript{a}

\textsuperscript{a}School of Materials Science and Engineering, Dalian University of Technology, Dalian, People’s Republic of China; \textsuperscript{b}School of Aeronautics and Astronautics, Dalian University of Technology, Dalian, People’s Republic of China; \textsuperscript{c}School of Civil Engineering, Dalian University of Technology, Dalian, People’s Republic of China; \textsuperscript{d}School of Mechanical and Transportation Engineering, Taiyuan University of Technology, Taiyuan, People’s Republic of China; \textsuperscript{e}Taiyuan Tongze Heavy Industry Co, Ltd, Taiyuan, People’s Republic of China

ABSTRACT

A magnetorheological fluid (MRF) is a smart composite suspension composed of nonmagnetic liquid and soft magnetic particles. Carrier fluids can considerably influence the performance of MRFs; therefore, to investigate the effect of carrier fluids on MRFs, an SO/IL-MRF was prepared by mixing an ionic liquid (IL) with silicone oil (SO) in this study. Three types of MRF samples were prepared for experiments (pure SO, pure IL, and SO/IL). According to the experimental results, the SO/IL-MRF has better sedimentation stability than those based on pure SO and pure IL. Further, three methods were used to determine the shear yield stresses of the MRFs. The SO/IL-MRF achieved a higher shear yield stress than those of the other two because a network structure is formed between the ionic fragments and the molecular chains of the SO in the SO/IL-MRF. This increases the movement resistance of the particles in the carrier fluid, and it is unlike the mechanism of the IL-enhanced MRF. This work provides new ideas for improving the MRF performance.

CONTACT Xufeng Dong \textsuperscript{a} dongxf@dlut.edu.cn; Min Qi School of Materials Science and Engineering, Dalian University of Technology, Dalian 116024, People’s Republic of China

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1. Introduction

A magnetorheological fluid (MRF) is a smart composite suspension that comprises a nonmagnetic liquid and soft magnetic particles [1–4]. The rheology of the MRF can be modified from liquid to solid-like via the application of an external magnetic field [4–6]. Owing to this property, MRFs have potential applications in various scenarios. For example, Oh et al. reviewed the application of MRF actuators in multi-degree-of-freedom systems [7], and Lei et al. studied a seat suspension based on a compact variable stiffness and damping rotary magnetorheological damper [8]. In addition, MRF is used in polishing liquids for precision surfaces [9], magnetorheological clutches [10], magneto resistive micro-displacement sensors [11], etc.

The most common MRF combinations include silicone oil (SO) as a non-magnetic liquid and carbonyl iron powder (CIP) as soft magnetic particles with shear yield strengths up to 100 kPa [4,12]. However, this combination results in poor long-term stability because of the large difference in the densities of CIP and SO. Therefore, many studies have focused on improving the sedimentation stability of MRFs and on their magnetorheological properties [1]. Existing studies have investigated the morphology and composition of magnetic particles; for example, magnetic particle coating [13–17], surface modification [18], synthesis of new particle morphology [19–23], doping [24], ATRP [25,26], etc. Further, other studies have shown that MRF sedimentation stability can be improved using a carrier fluid with higher viscosity or density [27–29]. The results show that these methods can improve the settling stability of the magnetorheological fluid, and also improve the shear yield strength of the magnetorheological fluid. In fact, simultaneously improving the shear yield strength and settlement stability of MRFs has always been a problem to be solved by researchers. Visual observation is the most commonly used method to evaluate the settlement stability of MRF and was also the method used in this paper. This method is to inject the MRF into the cuvette and let it stand, and then calculate the sedimentation stability of the MRF by measuring the change of the liquid level. In addition, there are also methods to evaluate the sedimentation stability of MRF, such as the inductive method based on centrifugal sedimentation, the sedimentation potential method, and the light transmittance pulsation detection method [2].

In our previous work, we found that the shear yield strength of CIP-based magnetorheological fluids can be improved using denser ionic liquids (ILs) [30,31]. Subsequent experiments revealed that using denser ILs did not improve the sedimentation stability of the IL-based MRF, and this was attributed to the particles being more likely to aggregate in ILs, which would affect the stability of the MRF.

Consider that mixing different carrier fluids is also a way to get a new carrier fluid. In this work, in order to preserve the high shear yield strength of IL-based MRF while being able to improve the sedimentation stability of MRF, we performed MRF prepared by mixing IL and commonly used SO with similar viscosity. This mixed MRF was analyzed to determine if it shows improvements in sedimentation stability.
2. Experimental

2.1. Preparation of MRFs

MRFs were prepared using typical mechanical agitation preparation procedures. The materials for preparing MRFs included SO (Beijing Hangping Silicone Co., Ltd.; 0.5 Pa s), 1-octyl-3-methylimidazolium tetrafluoroborate (IL; Linzhou Keneng Materials Technology Co., Ltd.; 0.44 Pa s), and CIP (CN-type from BASF with a particle size of 6.5–8.0 μm); the CIP content was 20% by volume. MRFs with pure SO, pure IL, and a 1:1 volume fraction of a mixture of SO and IL were prepared; they were referred to as SO-MRF, IL-MRF, and SO/IL-MRF, respectively. All reagents were of analytical grade and did not require further purification.

2.2. Evaluation of MRFs

The static observation method was used to test the sedimentation stability of the three MRF samples. To this end, the prepared MRF samples were placed in a 5 mL cuvette and left to stand at 25°C for observation. The clear liquid level was recorded, and the sedimentation rates of the three MRF samples were calculated.

The magnetorheological properties of the MRFs were measured using a Physica MCR301 rheometer (Anton Paar, Austria); the test gap was 1 mm. The shear yield strengths and viscosities of the MRFs were tested in the steady-state shear mode; their dynamic moduli were tested in the oscillatory shear mode. The test parameters reported in Ref [31] were used without changes. The curve of shear stress increased linearly with a shear rate between 10 and 100 s⁻¹ under different magnetic fields. The yield stress curves of MRFs with magnetic field changes at a shear rate of 1 s⁻¹ were tested. The variation curves of the MRF moduli with shear

![Figure 1](imageurl)  
**Figure 1.** (a) Sedimentation photos of the three MRFs after 6 days; (b) Curves of the sedimentation rates of the three MRFs as a function of time.
strains of 0.01–100\% and applied magnetic field strengths of 0–436 kA·m$^{-1}$ were tested using the oscillating shear mode. All tests were performed at 25°C.

3. Results and discussions

Figure 1 shows the sedimentation properties of the three MRFs. The SO/IL-MRF shows excellent sedimentation stability: the sedimentation rate is 5.1\%, which is lower than those of SO-MRF (35.6\%) and IL-MRF (40.0\%). The viscosity of the selected IL is slightly lower than that of SO, and therefore, the settleability of the IL-MRF is expected to be slightly poor. However, the MRF achieves better settling stability after mixing the two carrier fluids, and therefore, zero-field viscosities of the three MRFs are tested to identify a cause for the better settling stability.

Figure 2 shows that the viscosities of the three MRFs; the viscosity of the IL-MRF is slightly lower than that of the SO-MRF. However, the SO/IL-MRF has a higher zero-field viscosity than that of SO, and it can be attributed to the strong van der Waals force (inductive and dispersion forces) formed between the IL with strong polarity and the SO. The van der Waals force forms a network structure in the carrier fluid mixed with IL and SO, and this increases the motion resistance of particles in the carrier fluid, which increases its dynamic viscosity. A schematic of the relationship between the carrier fluid and particles in the three MRFs is presented in Figure 3.

![Figure 2. Curves of zero-field viscosity versus the shear rates for the three MRFs.](image-url)
Figure 3. Schematic of the relationship between carrier fluid and particles in the three MRFs.

Figure 4. Curve of shear stress as a function of shear rate under different magnetic field strengths for (a) SO-MRF, (b) IL-MRF and (c) SO/IL-MRF; (d) Curves of shear yield stress as a function of magnetic field strength for the three MRFs fitted by the Bingham model.
Thus, the SO/IL-MRF has higher zero-field viscosity, which increases its shear yield stress. The shear yield stress of SO/IL-MRF would be between those of IL-MRF and SO-MRF without the above-mentioned mechanism. To confirm this, the shear stress curves of the three MRFs as a function of the shear rate at different magnetic field strengths were tested.

The shear yield stress curves of the three MRFs versus the magnetic field strength are obtained by fitting the Bingham model; the results are presented in Figure 4. Although the Bingham model is not the best model for describing the MRF shear yield stress [32], because its fitting is relatively simple and can meet the requirements for comparing different MRF performances, the Bingham model was used here for fitting. Figure 4d shows that the shear yield stress of SO/IL-MRF is higher than that of SO-MRF. However, it is slightly lower than that of IL-MRF at higher magnetic field strengths. Ionic fragments in the IL form an ionic layer on the particle surface, which improves the interaction between particles and enhances the shear yield stress of the IL-MRF; this was explained in our previous work [31]. It remains difficult to explain why the shear yield stress of SO/IL-MRFs is lower than that of IL-MRFs; since the difference is small, it can be attributed to the insufficient accuracy of the model chosen for fitting. As shown in Figures 4a–c, the fitted line is slightly higher than that of the test data. Thus, the accuracy of determining the yield stress of a sample using this method depends on the rheometer resolution.

Figure 5. Curves of absolute value of shear yield stress as a function of magnetic field strength for three MRFs at a shear rate of 1 s$^{-1}$. 

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The yield stress determined with a rheometer that can detect very small shear rates is lower than that determined with an older rheometer that does not have a very high resolution. The two commonly used methods for determining shear yield stresses of viscoelastic liquids were tested to determine the shear yield stresses of the three MRFs.

The absolute value of shear yield stresses of the MRFs are approximated by testing the curve of stress versus magnetic field strength at a shear rate of 1 s⁻¹, as shown in Figure 5. The shear yield stress of the SO/IL-MRF is higher than that of the IL-MRF, as expected. However, it is uncertain whether this method is more accurate. Storage modulus $G'$ represents the elastic portion of the material under measurement, and loss modulus $G''$ represents the viscous portion of the material under measurement. Within a certain deformation range, both moduli remained constant, regardless of the set amplitude and frequency. The strain range with reversible elastic deformation is known as the linear viscoelastic region (LVE). The end point of the LVE is reached when the two moduli are no longer constant; this point is

Figure 6. Dependence of the dynamic shear modulus (storage modulus $G'$ and loss modulus $G''$) under different magnetic field strengths of (a) SO-MRF, (b) IL-MRF and (c) SO/IL-MRF on the shear stress at a constant angular frequency of 10 rad·s⁻¹; (d) The curve of the yield point of the three MRFs as a function of the magnetic field strength.
called the yield point, from which the structure of the sample becomes irreversibly destroyed. Based on this, the curves of $G'$ and $G''$ for the three MRFs as a function of shear stress under different magnetic field intensities are tested, as shown in Figure 6. Figure 6d shows a plot of the yield stress as a function of the magnetic field strength for the three MRFs. This plot achieves very similar values as that in Figure 5, and SO/IL-MRF exhibits a higher yield stress; this proves that the previously suspected enhancement mechanism is correct.

Figure 7 shows that SO/IL-MRF has a higher $G'$, which implies its internal structure is more stable. The reason for this stability cannot be the same as that of the IL-MRF. Thus, the special ‘structure’ formed in the SO/IL-MRF can be considered the key reason for its performance improvement.

4. Conclusions

SO/IL-MRFs were prepared by mixing ILs with SO. The experimental results showed that SO/IL-MRF had better sedimentation stability than MRF based on pure SO and pure IL, and its sedimentation rate was only 5.1% during the test; this was lower than those of SO-MRF (35.6%) and IL-MRF (40.0%). Further, it was shown that the SO/IL-MRF has a higher shear yield stress by comparing the results obtained using three different methods to determine the shear yield stress of the MRF. The improved performance of SO/IL-MRF is attributed to the formation of stronger van der Waals forces (especially the inductive force and dispersion force) between the IL with strong polarity and SO, which forms a network structure
that increases the movement of particles in the carrier fluid resistance. This is unlike why pure ILs enhance MRF performance. Thus, this study provides a new idea for improving MRF performance.

In the future, the proportions and types of ILs will be further adjusted to obtain MRFs with better performance.

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