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An Experimental Study on the Effects of Temperature and Magnetic Field Strength on the Magnetorheological Fluid Stability and MR Effect

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In this study, stability and rheological properties of a suspension of carbonyl iron microparticles (CIMs) in silicone oil were investigated within a temperature range of 10 to 85 °C. The effect of adding two hydrophobic (stearic and palmitic) acids on the stability and magnetorheological effect of a suspension of CIMs in silicone oil was studied. According to the results, for preparing a stable and efficient magnetorheological (MR) fluid, additives should be utilized. Therefore, 3 wt% of stearic acid was added to the MR fluid which led to an enhancement of the fluid stability over 92% at 25 °C. By investigating shear stress variation due to the changes in shear rate for acid-based MR fluids, the maximum yield stress was obtained by fitting Bingham plastic rheological model at high shear rates. Based on the existing correlations of yield stress and either temperature or magnetic field strength, a new model was fitted to the experimental data to monitor the simultaneous effect of magnetic field strength and temperature on the maximum yield stress. The results demonstrated that as the magnetic field intensified or the temperature decreased, the maximum yield stress increased dramatically. In addition, when the MR fluid reached its magnetic saturation, the viscosity of fluid depended only on the shear rate.

Keywords

Bingham plastic model, magnetic field, magnetorheological fluid, stabilization, stearic acid, temperature effect.

Introduction

Smart materials are materials whose rheological properties change significantly when they are exposed to an external stimulus. In recent years, these materials have been the center of attention because of their high controllability in the presence of an external stimulus [1]. MR fluid is one of the most important smart materials which has been widely investigated in recent years thanks to its numerous advantages over other smart fluids, like their controllability by a magnetic field, acceptable stability, reversibility and simple preparation [2-5]. Since the discovery of MR fluids, they have been widely used in many fields. The application domain of MR fluids ranges from medical and civil engineering to oil and automobile industries. Some examples include linear shock absorbers, dampers, brakes, rotary clutches and control valves, surgical operations, cancer therapy and orthopedic knee braces [6].

The base fluid, magnetizable particles and additives are three main ingredients for preparation of MR fluids. Magnetizable particles have the highest influence on the MR effect and can be dispersed in the base fluid. Magnetizable particles need to be stabilized, so additives are used to enhance particles stability in the base fluid. Because of the density mismatch between the base fluid and heavy magnetizable particles, these particles sedimentation leads to a reduction in the stability and MR effect. In addition, the redistribution of magnetizable particles in the base fluid is difficult due to hard cake formation [7]. Therefore, in recent years, many researches have been conducted to improve the stability of these fluids [8]. Generally, the stabilization methods of MR fluids fall into six categories: coating magnetizable particles, using wire-like nanoparticles, using spherical nanoparticles, using stabilizer additives, using dense base fluid, and mechanical methods [9].

Different stabilizer additives have been used by many researchers to enhance the stability of MR fluids. However, unfortunately most of these stabilizers decrease the MR effect or increase the off-state viscosity of the MR fluid. In a relatively successful investigation, Premalatha *et al.* [10] enhanced the MR fluid stability up to more than 20% by adding 0.5 wt% of grease to a common MR fluid. In a similar study, Jiang *et al.* [8] added stearic acid as much as 3% of the mass of magnetizable particles to a suspension of iron nanowires and carbonyl iron microparticles in silicone oil. They reported that this MR fluid had more reliable MR effect and stability than common MR suspensions. Rankin *et al.* [11] suspended carbonyl iron microparticles in mineral oil and used grease to improve the suspension stability. They observed that grease, without much changing the MR effect, improved the stability of the MR fluid, significantly. López-López *et al.* [12] synthesized an MR fluid by dispersing carbonyl iron microparticles in kerosene. They used aluminum stearate to enhance the stability. The adsorption of aluminum stearate on the surface of particles contributed to the improvement of their distribution along the magnetic field lines and, thus, the magnetic interaction energy between the particles was enhanced and the aluminum stearate containing fluid got a larger MR effect. Aramaki *et al.* [13] showed that by increasing the carbon chain length of alcohols, the viscosity and shear stress of the micellar solution increased.

Adding hydrophobic acids with more than 14 carbons to a common MR fluid resulted in significant enhancement in stability and MR effect [14]. The apparent viscosity of the MR suspensions grows under large magnetic field inductions. This elevation can lead to a significant development of viscose dissipation in MR fluids flows. A field-inductive yield stress of about 10^5 Pa was reported for MR fluids in large magnetic field strengths [15]. These fluids generate high energy at high shear rates which leads to a considerable increase in temperature inside the fluids and equipment [16]. Guerrero-Sanchez *et al.* [17] investigated the effect of temperature on the rheological behavior of dispersed iron oxide particles in an ionic liquid called 1-butyl-3-methyl imidazolium hexa fluorophosphate (BMI-PF6) within the temperature range of 25 to 76 °C. The obtained results were fitted using different models and it was concluded that the fluid followed the Bingham plastic model. This model is usually used for describing the behavior of MR fluids in the presence of a magnetic field. They also studied the effects of temperature and the intensity of magnetic field on the yield stress and demonstrated that the maximum yield stress of MR fluids was an exponential function of operational temperature. By suspending carbonyl iron particles into silicone oil, Claracq *et al.* [18] showed that the maximum yield stress had an exponential relation with the intensity of magnetic field at a constant temperature.

Arief and Mukhopadhyay [19] synthesized a ferrofluid composed of cobalt-nickel nanoclusters in castor oil and studied the effect of different temperatures (i.e. 25, 35, and 45 °C) on the rheological behavior of the synthesized ferrofluid. It was observed that as the temperature increased, the consistency parameter in the Herschel-Bulkley model was reduced but the yield stress did not change. There have been many attempts to produce a stable MR fluid with appropriate rheological properties [9, 20, 21]. Few researches have been conducted regarding the effect of temperature on the rheological properties of magnetorheological fluids and ferrofluids. In most cases, it has been observed that the shear stress of these fluids is reduced considerably as the temperature grows. Similarly, it has been shown that as the temperature increases, the viscosity of different base fluids decreases over 3 times in the absence of a magnetic field [22, 23]. Yet, it should be noted that, given the base fluid type, this change is variable between 0.5 to 3 times and the least change in the viscosity has been observed in the case of silicone oil [24].

In the present study, following the previous experimental studies [14, 25-27], the preparation of a stable and efficient magnetorheological fluid is investigated which not only is resistant to sedimentation, but also presents a reliable magnetorheological effect within a relatively wide temperature range in various seasons of the year and different thermal conditions. Thereafter, a model has been proposed for showing the dependence of the maximum yield stress on temperature and magnetic field intensity.

Experimental

In this paper, to prepare MR fluids, carbonyl iron particles (average density: 7.86×10^3 kg/m^3, CS grade, BASF, Germany) were dispersed in Polydimethylsiloxane (silicone oil, viscosity: 3.50×10^{-3} m^2/s, KCC, Korea). For the fluid stabilizing purpose, two hydrophobic acids, stearic acid (MIT, Malaysia) and palmitic acid (MERCK, Germany) were used. In all of the samples, the acid was first added to silicone oil and the mixture was stirred at 100 ° C in a water bath for 30 minutes to obtain a homogenous solution. Afterwards, the carbonyl iron particles were added to the sample which was then stirred for 30 minutes with an overhead stirrer (RZR, Heidolph, Germany, 2012) at 1000 rpm. Table 1 shows the specifications of the prepared MR fluids.

Table 1. Properties of the prepared MR fluids samples

Sample	Dispersed phase (wt%) (volume fraction %)	Continuous phase (wt%) (volume fraction %)	Additive (wt%) (volume fraction %)
MR1	Carbonyl iron (60) (15)	Silicone oil (40) (85)	No additive
MR2	Carbonyl iron (60) (15)	Silicone oil (37) (77)	Palmitic acid (3) (8)
MR3	Carbonyl iron (60) (15)	Silicone oil (37) (78)	Stearic acid (3) (7)
MR4	Carbonyl iron (60) (15)	Silicone oil (38) (80)	Stearic acid (2) (5)
MR5	Carbonyl iron (60) (15)	Silicone oil (39) (82)	Stearic acid (1) (3)

The spherical structure and the size of carbonyl iron particles were investigated by Scanning Electron Microscopy (SEM, MV2300, Tescan, Ltd., Czech Republic) with 15 kV operational voltage. The magnetic properties of carbonyl iron particles were measured by Vibrator Sample Magnetometer (VSM, MDKFD, Magnetic Danesh Pajoh Co. Ltd, Iran).

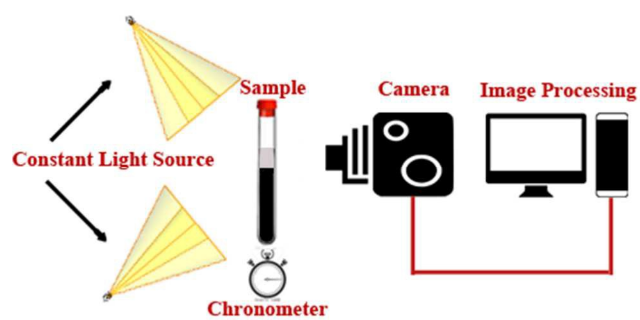

Figure 1. Schematic representation of the stability measurement system

In order to evaluate the stability, the prepared samples were placed in a water bath equipped with a thermal control system (Circulating Water Bath, PB 28 L, SPECTRALABTM, Instruments Pvt. Ltd., India) for over 700 hours at different temperatures (ranging from 10 to 85 °C). Then, the samples were photographed at certain intervals. Afterwards, the height and volume of each phase were determined through image processing, which represented a bi-phased suspension. Figure 1 shows the schematic image of stability determination system.

To investigate the rheological properties of MR suspensions, a rotational plate-plate rheometer (MCR300, Anton-Paar, Germany) connected to a magnetorheological device (MRD 180, Physica, Germany), which applies a homogeneous magnetic field perpendicular to the fluid movement to the samples, was used. To study the effect of temperature on the rheological properties of stable MR suspensions, the viscosity and the shear stress of samples were measured at shear rates of 0.01 to 1000 s^{-1} within the temperature range of 10 – 85 $^{\circ}$C. MCR 300 was connected to a JULABO F25 temperature control unit with a circulator head and a cooling machine and an electronic proportional temperature control that regulates the supplied heat to the liquid. This system has been designed to heat/cool the liquid of bath tank. Various temperatures (i.e., 10, 25, 40, 55, 70 and 85 °C) were maintained with an accuracy of ±0.01 °C in the measurements for this study. A uniform magnetic field of up to 234 kA/m with a gap distance of 1 mm was applied to all of the samples. To ensure the reproducibility of the obtained data, all of the experiments were performed twice and the average values of the results are reported in all figures.

Results and Discussion

Characterization of Magnetizable Particles

The spherical structure of carbonyl iron microparticles was evaluated by SEM. Through image processing, it was found that the size range of particles was from 1 to 6 µm and the average size of particles was 2.7 µm. Figure 2 shows the spherical structure of the carbonyl iron microparticles. As can be seen from Figure 2 (b), more than 50% of the carbonyl iron particles have diameters of 2-3 µm.

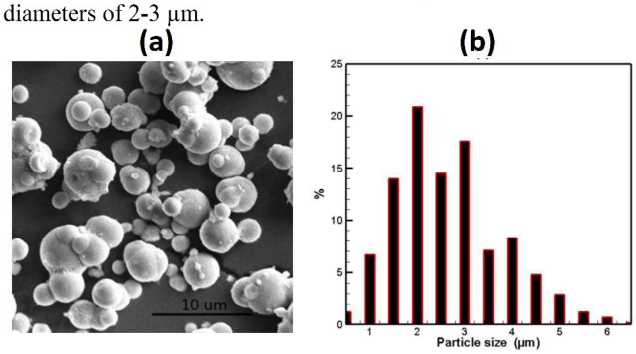

Figure 2. Carbonyl iron characterization (a) SEM image and (b) particle size distribution

The magnetic properties of carbonyl iron particles were investigated using VSM. The obtained results are shown in Figure 3. As can be seen in the figure, the magnetizability curve shows only a small amount of the magnetic hysteresis. Due to the high magnetic saturation of carbonyl iron particles, they are the best candidate for the preparation of industrial MR fluids. The high magnetic permeability of carbonyl particles with a magnetic saturation of 165 emu/g was observed in the 8 kOe field.

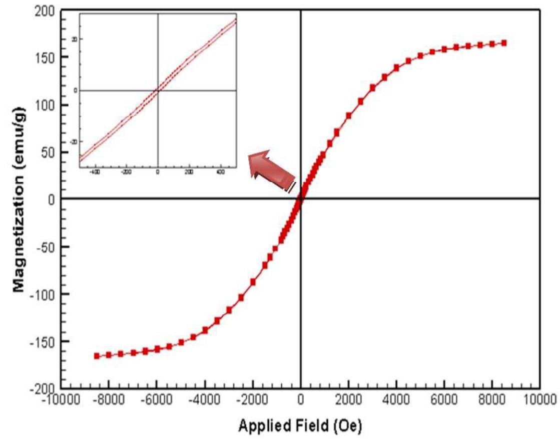

Figure 3. Magnetization curve (VSM) of carbonyl iron particles

Stability of the MR fluids

As it has been mentioned, the sedimentation percentage is defined as the ratio of the clear liquid to the total suspension volume as follows:

$$\text{Sedimentation ratio}(\%) = \frac{\text{Volume of supernatant liquid}}{\text{Volume of total suspension}} \times 100\ \% \quad (1)$$

In order to investigate the effect of additives on the stability of MR suspensions at different temperatures, the samples were kept in a fixed place for over a month and photographed within certain time intervals. Figure 4 demonstrates that using additives is inevitable to achieve a stable MR fluid. As can be seen from Figure 4, adding 3 wt% of each of the mentioned acids to the fluid increases the stability of fluid. However, the amount of stability enhancement was 1.5 times for palmitic acid and over 8 times for stearic acid. On the other hand, since the stability of the samples containing even lower percentages of stearic acid was remarkable compared to the stability of the sample containing palmitic acid, it seemed that stearic acid was a more promising additive for preparing a stable MR fluid.

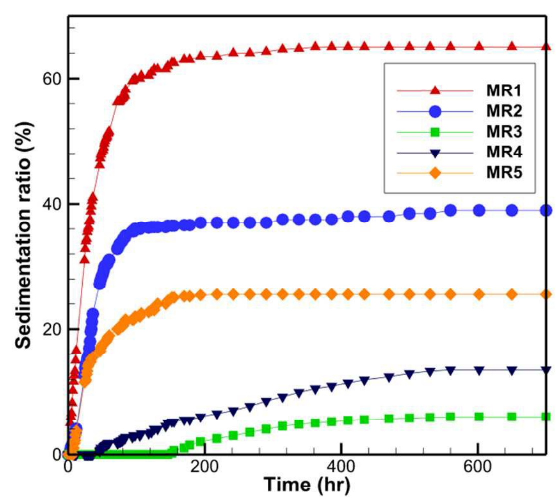

Figure 4. Sedimentation ratio versus time at 25 ° C

The effect of temperature on the stability of samples was investigated. Figure 5 shows the stability curve of the samples within the temperature range of 10–40 °C. As can be seen from Figure 5 (a), the stability of the additive-free sample was almost constant at various temperatures and it was independent from operational temperatures because the viscosity of base fluid and magnetizable particles were independent from temperature. Because of the longer carbon chain length of stearic acid compared to that of palmitic acid, when using stearic acid as an additive, changing the temperature has more effect on the formation of the gel–like structure in silicone oil [13] and therefore it influences the instability amount and rate to a greater extent. It is also evident that at all temperatures, the stability of the sample containing stearic acid was more than that of the sample with palmitic acid. The comparison between the diagrams in Figure 5 (b) shows that as the wt% of stearic acid increases, the effect of temperature on the stability becomes more significant. Similarly, the only sample that has a stability of more than 90%, over one month, was the one containing 3 wt% of stearic acid (MR3).

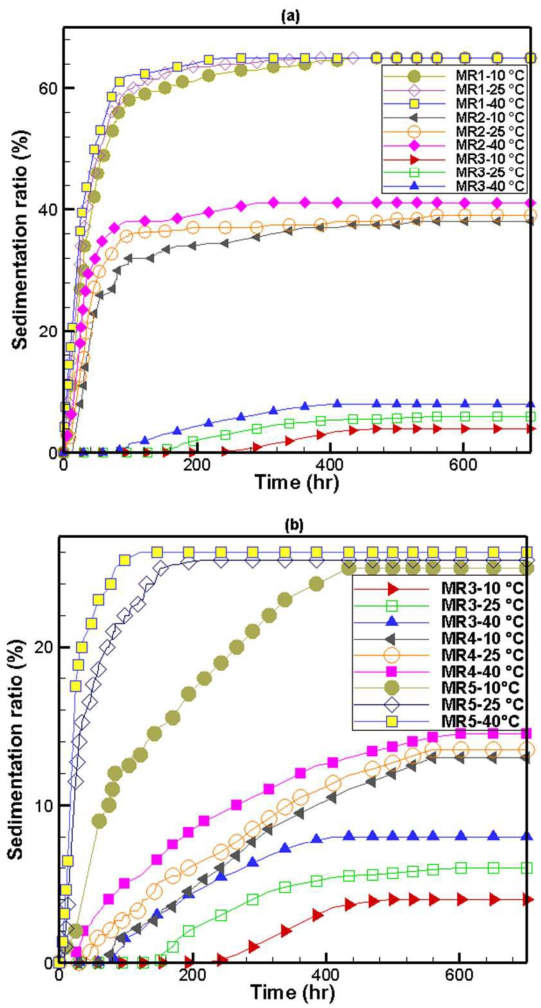

Figure 5. Sedimentation ratio curves for MR fluids; (a) with and without acid-additives and (b) with different wt% of stearic acid at different temperatures

Further investigations showed that at temperatures higher than 55 °C samples became unstable and they reached their maximum level of instability in less than 12 hours. It can be concluded from Figure 5(a) that at higher temperatures, the additive-free sample was affected much less than the other samples due to the independence of the base fluid from temperature.

In the industrial applications of MR fluids, high stability of fluids is highly desired. Therefore, the sample containing 3 wt% stearic acid was chosen for the rest of studies. The obtained results show that further increase in stearic acid will lead to undesirable elevation of the fluid viscosity without significant enhancement in the stability of fluid.

Rheometry Analysis

The most important feature of MR fluids is their resistance against movement when exposed to a magnetic field. The majority of MR suspensions have high viscosity in the absence of a magnetic field which is undesirable in most industrial applications. Figure 6 shows the curve of viscosity in terms of shear rate in the absence of a magnetic field for the additive-free MR fluid (MR1) and MR fluids with 3 wt% additives (MR2 and MR3). As can be seen from Figure 6, both palmitic (MR2) and stearic (MR3) acids increased the viscosity of fluid in the absence of a magnetic field. Since the viscosity elevation was not very significant in both palmitic and stearic acids and the stability of the sample containing stearic acid was far more than that of the sample containing palmitic acid (see Figure 4), the sample containing 3 wt% stearic acid (MR3) was chosen as the sample with a proper off-state viscosity which possesses a good stability, for the subsequent experiments.

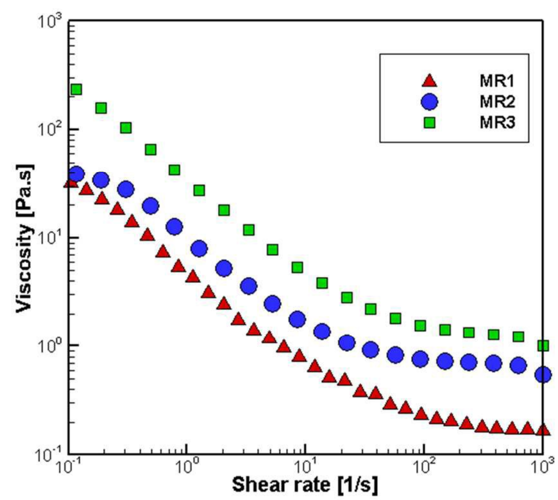

Figure 6. MR fluids viscosity versus shear rate (MR1, MR2 and MR3) in the absence of a magnetic field at 25 ° C

To investigate the circumstance of improving the viscosity of the MR fluid by adding stearic acid, the changes of viscosity are reported in Figure 7 in terms of shear rate for the base fluid (the sample without additives and magnetizable particles), the sample containing the base fluid and additives, and for MR3 at 25 ° C. As can be seen from Figure 7, the base fluid has a semi-Newtonian behavior with constant viscosity. On the other hand, by adding stearic acid, the viscosity developed significantly as a result of formation of a gel-like structure in silicone oil [8]. The comparison of the diagrams in Figure 7 shows that MR3 viscosity enhancement is largely due to the formation of a gel-like structure of stearic acid in silicone oil.

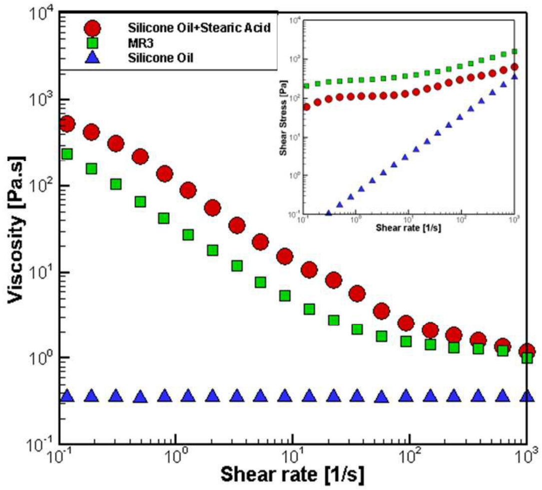

Figure 7. Variation of viscosity versus shear rate in the absence of a magnetic field at 25 ° C. The inset figure exhibits the shear stress versus shear rate in the absence of a magnetic field at 25 ° C.

To investigate this unexpected behavior, a falling film of MR3 with and without carbonyl iron particles has been examined. Figure 8 depicts the photos of MR3 (a) in comparison to the paste-like solid mixture of stearic acid in silicone oil (b), (particle-free sample), at 25 °C which was poured on two inclined surfaces with the same inclination after 1 second from pouring. As can be seen in Figure 8, MR3 easily slipped on the inclined surface but the mixture of stearic acid and silicone oil could not move on the surface. These observations may be due to the fact that stearic acid and silicone oil form a paste-like solid at 25 °C which prevents the movement of mixture. On the other hand, our observations showed that when carbonyl iron was added to this mixture, at higher temperatures, the spherical particles easily filled the constructed holes and prevented the paste-like solid creation even at lower temperature. This confirmed the higher viscosity of stearic acid and silicone oil mixture in comparison to that of MR3 (see Figure 7).

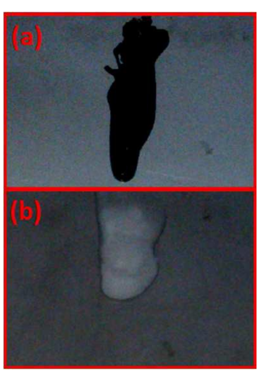

Figure 8. Movements of (a) MR3 and (b) paste-like solid created by the addition of 3 wt% of stearic acid to silicone oil after 1 second on two inclined planes

The effect of temperature on the rheological properties of MR fluids at the temperatures of 10, 25, 40, 55, 70 and 85 °C was evaluated. The changes of shear stress in terms of shear rate for MR3 at 10, 40 and 70 °C are provided as some typical examples in Figure 9. The results showed that when a constant magnetic field was applied to the MR fluid, the shear stress of the fluid was highly dependent on the shear rate and temperature.

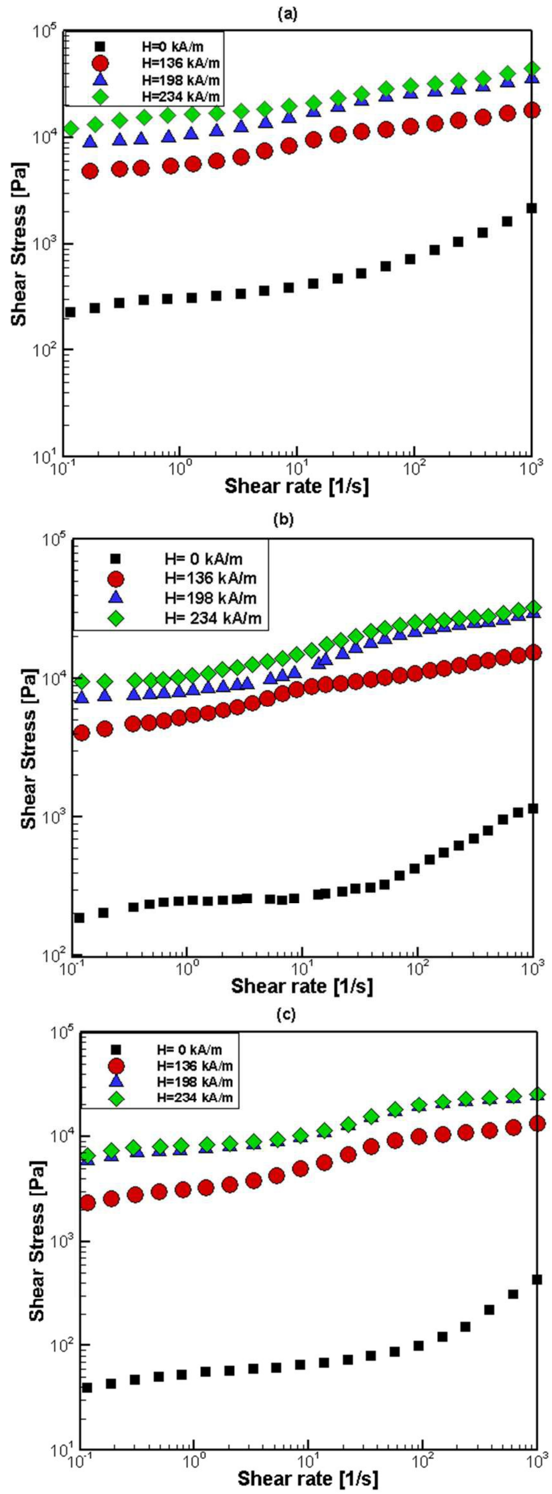

Figure 9. Shear stress versus shear rate for MR3 at temperature; (a) 10 °C, (b) 40 °C and (c) 70 °C

The results showed that when the suspension reached its magnetic saturation, the shear stress of fluid did not change with temperature and intensity of magnetic field and it was only a function of shear rate. Since the further development of magnetic field over 234 kA/m has no significant effect on the shear and yield stress of the prepared MR fluids, no further growth was applied to the magnetic field strength.

The overall behavior of MR fluids followed Herschel-Bulkley model [28], but the determination of Herschel-Bulkley parameters at very low shear rates, which has a significant effect on the model prediction, is difficult. Therefore, some researchers [28-31] have fitted Bingham plastic model to their experimental results at high shear rates and have reported the maximum yield stress. Bingham plastic model is defined as [32]:

$$\begin{cases} \tau = \tau_0 + \eta \dot{\gamma} & \tau \geq \tau_0 \\ \dot{\gamma} = 0 & \tau < \tau_0 \end{cases} \quad (2)$$

Where τ and τ_0 are shear and yield stress and $\dot{\gamma}$ and η denote shear rate and shear viscosity, respectively. Figure 9 illustrates that the behavior of fluid obeys Bingham plastic model at high shear rates. Therefore, Bingham plastic model was fitted on the data at high shear rates using the non-linear least squares method to calculate the maximum yield stress. In Table 2, the amount of maximum yield stress for MR3 is provided in various magnetic field strengths and at different temperatures.

Table 2. The yield stress (τ_0) and shear viscosity (η) obtained from Bingham plastic model

Temperature, T [°C]	Magnetic Field Strength, H [kA/m]	Maximum Yield Stress, τ_0 [kPa]	Shear Viscosity, η [Pa.s]
10	0	0.82	1.31
	136	13	4.38
	198	26	9.44
	234	31	13.21
25	0	0.81	0.81
	136	12	4.02
	198	24	7.43
	234	27	7.87
40	0	0.74	0.41
	136	11	3.65
	198	22	6.82
	234	25	7.54
55	0	0.34	0.38
	136	10	3.34
	198	21	4.57
	234	23	4.78
70	0	0.08	0.35
	136	10	3.26
	198	20	3.42
	234	22	3.57
85	0	0.07	0.19
	136	9	1.48
	198	19	1.94
	234	22	2.26

Figure 10 shows the maximum yield stress in terms of magnetic field strength in each examined temperature. As can be seen from Figure 10, the maximum yield stress at 234 kA/m decreases from 31 to 22 kPa by increasing the temperature from 10 to 85 °C. It is also evident that by increasing temperature, the maximum yield stress is decreased which is consistent with the results of other researchers [17, 33].

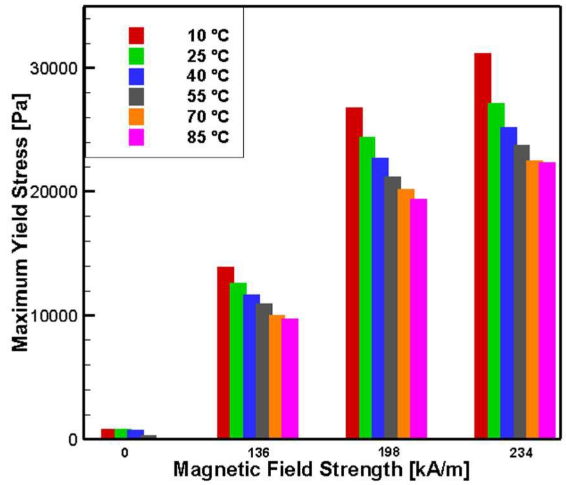

Figure 10. The maximum yield stress versus magnetic field strength at various temperatures

In addition to our previous studies [28], other researches [31, 34-36] have also shown that there are various relations between the maximum yield stress and the magnetic field strength at a constant temperature. In one of these papers, Piao *et al.* [31] developed the relation of yield stress and magnetic field strength as follows:

$$\tau_0 \propto H^n \tanh\left(\sqrt{H}\right) \quad (3)$$

Where n, τ_0 and H denote the equation exponent, the maximum yield stress and the magnetic field strength, respectively. On the other hand, a deeper study of Figure 10 shows that both temperature and magnetic field strength have significant effects on the maximum yield stress. To introduce a relation between the maximum yield stress and temperature, one would be addressed to the Arrhenius relationship between viscosity and temperature:

$$\eta = A \exp\left(E_a / RT\right) \quad (4)$$

Where R is the universal gas constant, T is temperature (K), A is constant of the equation and E_a is activation energy. As the concept of maximum yield stress is similar to the viscosity and Arrhenius equation relates the viscosity to the temperature [33], in this study, the maximum yield stress has been related to temperature with an exponential function. Sahin *et al.* [33] suggested a new model for the dependency of yield stress on temperature which is consistent with our results. In their model, yield stress has an exponential relation with temperature:

$$\tau_0 \propto H^{1.3} \exp\left(-0.005T\right) \quad (5)$$

Based on the foregoing, the following equation is used by the incorporation of the mentioned correlations to model the dependency of maximum yield stress on magnetic field strength and temperature:

$$\tau_0 = \alpha \times H^n \tanh\left(\sqrt{H}\right) \exp\left(-\beta T\right) \quad (6)$$

Where α and β are the equation constants which are obtained by fitting the data of Table 2 for the maximum yield stress. Therefore, relation (5) is changed to:

$$\tau_0 = 65.25 \times H^{1.41} \tanh\left(\sqrt{H}\right) \exp\left(-5.38 \times 10^{-3}\, T\right) \quad (7)$$

Relation (7) shows the less dependency of yield stress on temperature in comparison to that of the magnetic field strength[33] which is also obvious in Figure 10. A deeper evaluation of relation (7) reveals that as the temperature increases, the maximum yield stress decreases, confirming the results achieved by other researchers [19, 22, 37]. This observation may be due to the fact that as the temperature increases, the gel structure of the suspension becomes weaker and the fluid resistance against movement will be decreased.

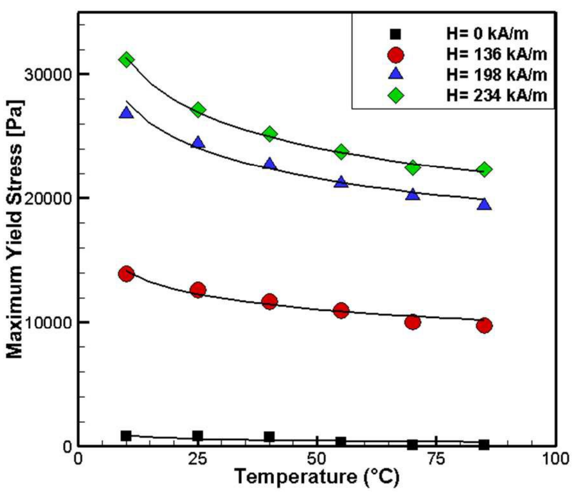

Figure 11. Maximum yield stress versus temperature at different magnetic field strength

Figure 11 shows the the maximum yield stress curve in terms of temperature in each strength of magnetic field which is consistent with the results of other researchers [17, 33]. As can be seen from Figure 11, by increasing temperature, the effect of temperature on the maximum yield stress becomes less. However, the stability of MR3 enhanced over 95, 92 and 88% at temperatures of 10, 25, 40 °C, respectively. This means that the effect of temperature on the stability and rheological properties of MR fluids is of secondary importance in comparison to the effect of magnetic field strength.

Conclusions

In the present study, the rheological and stability properties of the suspensions of carbonyl iron microparticles in silicone oil were evaluated at various temperatures. The results showed that adding 3 wt% of stearic acid to the MR fluid resulted in 92% stability enhancement of the suspension even over a period of a month. Also, the stability of this sample was eight times more than that of common MR fluids which is a remarkable achievement in the area of magnetorheology.

The evaluation of rheological behavior of MR fluids showed that the highly stable sample, i.e. the suspension of carbonyl iron microparticles in silicone oil stabilized with 3 wt% stearic acid (MR3), demonstrated a relatively low off-state viscosity and high yield stress. Furthermore, the maximum yield stress of this MR fluid which was as high as 27 kPa (in 234 kA/m magnetic field strength) at 25 °C, was noticeably more than the yield stress of common MR fluids.

By applying Arrhenius analogy for the relation of yield stress and temperature and also based on the existing correlation of maximum yield stress and magnetic field strength, we suggested a new correlation. It was observed that as the magnetic field intensified, the maximum yield stress was enhanced dramatically. On the other hand, as the temperature increased, the viscosity and maximum yield stress decreased. Such a significant increase in the yield stress was not observed in the case of decreasing temperature. The investigation of the type and weight fraction of MR fluid additives and also particle polydispersity on the MR effect and MR fluid stability can be the subject of the future research areas.

Nomenclature

α	Constant (Eq. 6)	$Pa.m^n.A^{-n}$
β	Constant (Eq. 6)	K^{-1}
η	Shear viscosity	Pa.s
τ_0	Maximum yield stress	Pa
A	Arrhenius constant	Pa.s
E_a	Activation energy	$J.mol^{-1}$
H	Magnetic field strength	$A.m^{-1}$
n	Constant (Eq. 3)	-
R	Universal gas constant	$J.(mol.K)^{-1}$
T	Temperature	K

Notes and references

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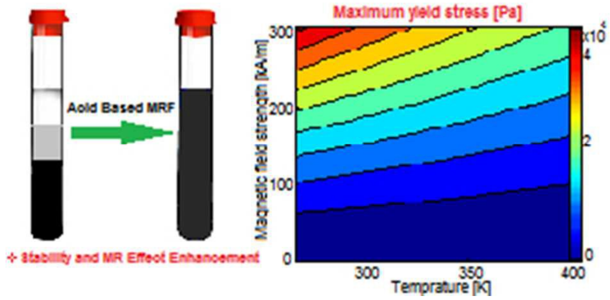

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