



Preparation and characterization of carbonyl iron/strontium hexaferrite magnetorheological fluids



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ABSTRACT

The heat transfer oil-based magnetorheological fluid (MRF) was prepared using oleic acid-modified micron carbonyl iron powder as a magnetic dispersed phase and strontium hexaferrite (SrFe₁₂O₁₉) nanoparticles as an additive. The sedimentation stability of MRFs was studied. The results indicated that the stability of MRFs was improved remarkably by adding SrFe₁₂O₁₉ nanoparticles and the sedimentation ratio was only 0.88 in 20 days when the content of nanoparticles reached 10 wt%. The rheological properties were characterized by a HAAKE rheometer without a magnetic field and a capillary rheometer with and without a magnetic field. The effects of SrFe₁₂O₁₉ nanoparticles, the temperature, and magnetic field strength were investigated. In addition, the rheological properties could be predicted well using the improved Herschel–Bulkley model, even under a magnetic field. A theoretical model was also proposed to predict the yield stress based on the microstructure of the MRF under an applied magnetic field.

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Introduction

Magnetic suspensions, as a class of smart magnetic materials, can be divided into two broad categories (Vékás, 2008): (a) magnetic fluid or ferrofluid (FF): a suspension of magnetic nanoparticles (1–15 nm) in a nonmagnetic liquid and (b) magnetorheological fluid (MRF): a suspension of magnetic micron-sized particles (1–20 μm) in a carrier liquid (Klingenberg, 2001; Odenbach, 2003). Because of the significant difference in particle size, FF and MRF exhibit distinctive rheological behaviors in an applied magnetic field. FF shows small changes in viscosity and does not develop a yield stress under application of high magnetic field, which is limited to those where only low magnetoviscous response are required (Vékás, Bica, & Avdeev, 2007). However, MRF is capable of being reversibly changed from a liquid fluid to a viscoelastic solid and the viscosity changes by several orders of magnitude with the aid of a magnetic field within milliseconds. This phenomenon is also

referred to as the magnetorheological effect (Bossis, Khuzir, Lacin, & Volkova, 2003; Park, Fang, & Choi, 2010). MRFs possess outstanding rheological characteristics, such as high yield stress and shear viscosity, which can be altered by tuning an external magnetic field. Therefore, they have a wide range of technological applications, including semi-active damping devices (Bica, 2002), magnetorheological finishing (Sidpara & Jain, 2012), and mechanical seal and aerospace materials (Odenbach, 2003). MRFs also exhibit potential applications in biotechnology (Scilingo, Bicchi, De Rossi, & Scotto, 2000) and medicine, such as drug delivery and cancer therapy (Häfeli, 2004; Liu, Flores, & Sheng, 2001).

A good MRF should have certain features, including high magnetic saturation, stability against settling and aggregation, corrosion resistance, and temperature stability (Claracq, Sarrazin, & Montfort, 2004). High magnetic saturation mainly depends on the dispersed magnetic particles in MRF. Carbonyl iron particles with relatively high saturation magnetization (2.1 T/212.7 emu/g) and low cost (Vékás, 2008) are commonly used over other alloy and oxide powders. The temperature stability of MRF is closely related to the carrier liquid. Typically, petroleum-based oils, silicone, mineral oils, polyesters, polyethers, and water are employed as carrier

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Nomenclature

a	lengths of transparent supernatant, cm
b	lengths of lower turbid liquid, cm
C	capillary constant
E_A	activation energy, J/mol
f_p	electrostatic force between particles, N
H	local field strength, mT
H_0	external magnetic field strength, mT
k'	inertial energy correction factor
k	consistency coefficient, $\text{N s}^n/\text{m}^2$
L	capillary length, cm
m	magnetic moment, A m^2
N	the number of chains per unit area
n	shear-thinning factor
ΔP	pressure drop in a capillary, MPa
Q	flow rate in the capillary, m^3/s
R	radius of capillary, mm
R_0	distance between two adjacent micron-particles, μm
r	particle radius of the carbonyl iron powder, μm
S	area of polar plates, m^2
t	time, s
V	fluid volume between polar plates, m^3

Greek letters

γ	angle between vertical direction and magnetic chain, rad
$\dot{\gamma}$	shear rate, s^{-1}
δ	particle radius of the $\text{SrFe}_{12}\text{O}_{19}$ nanoparticles, μm
η	viscosity, mPa s
μ_0	vacuum permeability, $4\pi \times 10^{-7} \text{ N/A}^2$
τ	shear stress, Pa
τ_0	yield stress, Pa
τ_{H_0}	yield stress under an applied magnetic field, Pa
χ	susceptibility of magnetic particles

liquids. Nevertheless, because of the large density mismatch between the magnetic particles (in the range of $4.0\text{--}8.0 \text{ g/cm}^3$) and the carrier fluids (about 1.0 g/cm^3), MRFs are susceptible to serious sedimentation, directly restricting their applications and producibility. As a result, a wide variety of methods have been adopted to improve the stability of MRF: (i) addition of surfactants or polymers (e.g., oleic acid, poly(methyl methacrylate)) (Choi, Park, Cho, & Choi, 2006; Dang, Ooi, Fales, & Strove, 2000), (ii) addition of thickening agents (e.g., organoclays, carbon fibers) (López-López, Vertelov, Bossis, Kuzhir, & Durán, 2007; López-López, Gómez-Ramírez, Durán, & González-Caballero, 2008), (iii) addition of magnetic nanoparticles (Rosenfeld, Wereley, Radhakrishnan, & Sudarshan, 2002; Viota, Gonzalez-Caballero, Duran, & Delgado, 2007; Wereley et al., 2006), and (iv) use of ionic liquids as carriers (Guerrero-Sanchez, Lara-Ceniceros, Jimenez-Regalado, Rasa, & Schubert, 2007). The first solution has been demonstrated to prevent aggregation by means of steric repulsion in many studies. However, the introduction of nonmagnetic coatings could reduce the magnetic properties of suspension, which could inevitably degrade the magnetorheological response at the same time (Pu & Jiang, 2005). Thixotropic networks can be formed to alleviate the setting problem of the suspensions by adding thickening agents. However, some thickening agents may expand to make the suspension thicken irreversibly after continuous abrasion, which causes an increase in off-state viscosity and a decrease in performance. Compared with the above methods, adding the appropriate type

and amount of ultrafine particles or magnetic nanoparticles is an effective way to resolve the stability problem in MRF.

Soft magnetic materials, such as Fe_3O_4 and Fe, are widely used as dispersed phases (Rosenfeld et al., 2002; Viota et al., 2007). Surprisingly, the addition of hard magnetic additives has attracted relatively little attention. Hard magnetic strontium hexaferrite ($\text{SrFe}_{12}\text{O}_{19}$) nanoparticles, with moderate saturation magnetization, coercivity, and Curie temperature (Pullar, 2012; Veverka, Pollert, Závěta, Vassure, & Duguet, 2008), are thought to improve the magnetic response of MRF when compared with soft magnetic nanoparticles. Moreover, $\text{SrFe}_{12}\text{O}_{19}$ nanoparticles have good stability against oxidation.

In this study, $\text{SrFe}_{12}\text{O}_{19}$ nanoparticles were synthesized using chemical co-precipitation, and were chosen as dispersed particles to prepare MRFs. The effects of $\text{SrFe}_{12}\text{O}_{19}$ nanoparticles on the rheological properties and stability of MRF were investigated. An improved Herschel–Bulkley (H–B) model was proposed to predict the yield stress and shear stress based on the microstructure of the MRF under a magnetic field.

Experimental

Materials

Ferric chloride hexahydrate ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$), strontium chloride hexahydrate ($\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$), sodium hydroxide (NaOH), sodium chloride (NaCl), potassium chloride (KCl), ethanol, and oleic acid ($\text{C}_{18}\text{H}_{34}\text{O}_2$) were all analytical grade reagents and purchased from the SCRC (Sinopharm Chemical Reagent Co., Ltd., Shanghai, China). Carbonyl iron powder (purity 99.50%) was purchased from Chengdu Sagawell Technology Co., Ltd., China. Achid general heat transfer oil 320 which contains synthetic hydrocarbon, dispersant and high-temperature antioxidant additives was industrial grade and purchased from Shanghai Fortune Lubricating Oil Co., Ltd., China. Deionized water was used throughout the experiment.

Preparation of $\text{SrFe}_{12}\text{O}_{19}$ nanoparticles

According to our previous research (Lu, Hong, & Li, 2011), the precursor of $\text{SrFe}_{12}\text{O}_{19}$ was prepared by co-precipitation using $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ and $\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$ as the starting materials and NaOH as a precipitant. $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ and $\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$ with a molar ratio of 9.23:1 (which was proposed by Hessien, Rashad, & El-Barawy, 2008) were dissolved in deionized water. NaOH aqueous solution (3 M) was heated to 80°C in a three-neck flask. The metal salt solutions were added to the NaOH solution under vigorous mechanical stirring (1000 rpm) to form a brownish precipitate. The reaction was carried out at 80°C for 0.5 h. After cooling naturally to room temperature, the precipitate was filtered and washed five times with deionized water and three times with ethanol. It was dried at 100°C overnight. The as-dried precursor was ground with a 1:1 mass ratio of NaCl and KCl. The mixture was calcined in a muffle furnace at 850°C for 2 h. The product was washed with a dilute HCl solution to remove the impurity (SrCO_3), and washed several times with deionized water and ethanol. The product was dried at 60°C for 12 h and ground to obtain $\text{SrFe}_{12}\text{O}_{19}$ nanoparticles.

Surface modification of carbonyl iron powder

Carbonyl iron powder has a strong tendency to aggregate because of its high surface energy. It is easily oxidized because of the absorption of water and oxygen when exposed to air. Therefore, it is necessary to modify the microparticles with surfactants.

Oleic acid was used to modify the surface of the carbonyl iron powder. Oleic acid (calculated by 10 wt% of carbonyl iron powder)

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