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Viscosity and sedimentation behaviors of the magnetorheological suspensions with oleic acid/dimer acid as surfactants



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ABSTRACT

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Keywords: Magnetorheological fluid Viscosity Sedimentation Surfactant This work deals with the role of polar interactions on the viscosity and sedimentation behaviors of magnetorheological suspensions with micro-sized magnetic particles dispersed in oil carriers. The oleic acid and dimer acid were employed to make an adjustment of the hydrophobicity of iron particles, in the interest of performing a comparative evaluation of the contributions of the surface polarity. The viscosity tests show that the adsorbed surfactant layer may impose a hindrance to the movement of iron particles in the oil medium. The polar attractions between dimer acid covered particles gave rise to a considerable increase in viscosity, indicating flocculation structure developed in the suspensions. The observed plateau-like region in the vicinity of 0.1 s^{-1} for MRF containing dimer acid is possibly due to the flocculation provoked by the carboxylic polar attraction, in which the structure is stable against fragmentation. Moreover, a quick recovery of the viscosity and a higher viscosity-temperature index also suggest the existence of particle-particle polar interaction in the suspensions containing dimer acid. The sedimentation measurements reveal that the steric repulsion of oleic acid plays a limited role in the stability of suspensions only if a large quantity of surfactant was used. The sedimentation results observed in the dimer acid covered particles confirm that loose and open flocculation was formed and enhanced sedimentation stability.

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1. Introduction

Magnetorheological fluids (MRFs) are suspensions of micronsized particles dispersed in nonmagnetic carrying fluids that have the ability to undergo rapid and significant changes in their dynamic properties when an external magnetic field is applied [1,2]. Due to their tunable rheological properties, MRFs have drawn considerable interest in the scientific and commercial applications such as semi-active shock absorbers, seals, dampers, torque transfer devices [3–5]. However, the significant mismatch between iron particle density and surrounding fluid density makes the magnetorheological suspension susceptible to long term stability. To avoid irreversible particle aggregation, the surface of the magnetic particles is typically sterically stabilized by using appropriate surfactants as one prevailing method. An understanding of the effects of these surfactants on the rheology, sedimentation, redispersibility, and durability of MRF is a prerequisite for the effective utilization of these materials in various applications, as the presence of adsorbed chemicals not only can impose a non-zero gap between the magnetic particles, but also can change the charge of particles, enhance lubrication, or change the initial viscosity of MRF to produce a stable suspension [6,7].

Traditionally, surfactant-based synthesis routes were developed to prepare magnetic nanoparticles. In these methods [8–11], surfactants coordinated their polar end groups on the nanoparticle surface. Hence, the hydrophobic carbon tails of the organic chemicals were pointed outwards from the surface of synthesized magnetic particles. The hydrophobic alkyl chains on the surface of the synthesized particles produced stable suspensions only in non-polar organic solvents. As for MRF with micro-scale iron particles, the use of surfactant alone seems to play a limited role in promoting the sedimentation property in the polar carrier. Lopez-Lopez et al. [12,13] examined the effect of adding aluminum stearate to a suspension of iron and kerosene and observed this surfactant improved the redispersibility of particles which was consistent with the claim of other researchers [14,15] but reduced stability. They also examined the stability of MR fluids after the addition of oleic acid by tracking changes in the optical absorbance against time [16]. The authors demonstrated that low concentrations (< 1 vol%) of oleic acid have little effect on the stability properties of the suspension. However, as the concentration of oleic acid increases (> 1 vol%) an improvement in the stability can be observed. Besides, the introduction of different surfactants (e.g., 1-dedecylamine, sodium dodecyl sulfate) into MRFs has been found to obtain well-dispersed suspensions and enhance the

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redispersibility [17,18] which provides for a soft sediment once the magnetic particles settle out.

Instead of direct adding into the suspensions, the metallic soap surfactants existing in the grease, such as aluminum, or lithium used as a thickener, are capable of conferring MRF with desired stability. Premalatha et al. [19] studied the effect of additives fraction (grease) on the stability of a magnetorheological fluid. Based on their examination, the sedimentation of the fluid will be less significant at higher percent of the grease. They also observed that the magnetorheological fluid has non-Newtonian behavior in the absence of a magnetic field.

Electrostatic stabilization procedure with ionic surfactant can be used to prepare such water based colloidal fluids. The formation of an electric double layer or adsorption layers of surfactants on the magnetic particle surface enhances the stability of lyophobic colloids in aqueous carriers [7,20,21].

Apart from the electrostatic repulsion, the internal colloidal interactions between particles, such as van der Waals, polar or acid-base interactions, can determine the state of dispersion (homo-dispersed or heteroaggregated), and in consequence, the rheology as well as stability of MRFs. Park et al. obtain MRFs consisting of carboxyl iron particles dispersed in an emulsion made up of oil and water. The stabilization agent is Tween 80. The stabilization was achieved through polar attraction between hydrophilic-treated carboxyl iron and water emulsion in continuous phase which resulting in dense iron particles' adsorption around oil droplets thereby increasing their stability [22]. As in the medium of silicone or hydrocarbon oils, the particle-particle polar interactions could dominate over the particle-carrier interactions while electrostatic interactions will be negligible for little surface charge acquired by iron particles. Interestingly, Pacull and his coworkers [23] studied the magnetorheological and interfacial thermodynamics properties of two silica-coated magnetite nanoparticles in oil media. One of them is hydrophilic, while the other one is hydrophobic. It is suggested that the non-negligible interfacial interactions and the attractive van der Waals induce aggregates that could make the field-induced chains weaker. There has, however, been scarce work on investigating the effects of polar interactions on the rheological and sedimentation behaviors of MRF consisting of micro-sized iron particles despite its importance for the success of applications.

The aim of this research work was to probe and understand the possibility of modulating the surface polarity of iron particles, and the influence on the off-state viscosity as well as sedimentation properties of MRF by addition of surfactants. To address the issue, the control of surface functional groups on the suspending particles is required to obtain two kinds of iron particles differing in their hydrophobicity.

The oleic acid, traditional surfactant to stabilize particles in nonpolar carrier is employed to confer the lipophilic property to particles [8,10,16,24,25]. Dimer acid, usually dimeric fatty acid is a complex mixture of ingredients, produced from the use of unsaturated fatty acids (oleic acid and linoleum acid) and obtained by mutual polymerization. Its main components contain two end carboxylic acid groups that can improve the hydrophilic property of materials. For this reason, dimer acid is employed to modify the surface property of iron particles.

2. Experimental section

2.1. Materials

Soft magnetic carbonyl iron (OM quality, supplied by BASF) was used in the preparation of the MRFs. The mean particles size diameter and density of iron particles were approximately $3.5 \ \mu m$



Fig. 1. Molecular structure of the dibasic acid.

and 7.8 g/cm³, respectively. Methyl silicone oil (0.35 Pa · s at 25 °C, the viscosity is high enough to against the sedimentation in the rheological tests) and mineral oil (0.06 Pa · s at 25 °C) were used as carried liquids. Dimer acid (DA, Technical quality) was purchased from ATUREX Company in China. The dibasic acid (C36H64O4, molecular weight: 560.91) content is about 80~85%, while the monomer and trimer acids content is less than 20%. The molecular structure of the main component in dimer acid is shown in Fig. 1. Oleic acid (OA, analytical quality) was used as surfactants. Iso-propyl alcohol was solvent.

2.2. Preparation of the MR suspensions

The iron particles were dried overnight (> 12 h) in a vacuum to remove physical adsorbed water on the surfaces. The dried powders were immersed in the mixture of isopropyl alcohol and surfactant, and then stirred by ultrasonic dispersion (30 min) and mechanical ball-milling (300 rpm, six hours) to allow the adsorption of dimer acid on the iron particles. After surface modification, the dried iron powder was added to the silicone oil and the resulting mixture was stirred by ultrasonic dispersion and high-speed mechanical ball-milling for a long time (12 h) to ensure the required homogeneity. The detailed preparation process of MR suspensions and schematic representation of adsorption process are shown in Fig. 2. The final concentrated suspension contained approximately 20 vol% of iron particles and was labeled as CI/OA or CI/DA suspension. The dilute MR suspensions containing 2.5 g/L iron particles and mineral oil were prepared by the mechanical shaking and sonication. The concentration of particles is low enough to avoid multiple scattering and attenuation of the incident beam. The concentration of surfactants ranged from 0.5 to 8 g/L.

2.3. Characterization

The morphology of the iron particles was characterized by scanning electron microscopy (SEM). As shown in Fig. 3(a), the iron particles are spherical and polydisperse. The contact angles were measured with the sessile-drop method using a goniometer and image-analysis software (KRüSS DSA 100). The dried iron particles were pressed into a tablet with a thickness of 1 mm and diameter of 10 mm, and then placed on the sample table (with load of 10 kN for three minutes). The contact angle was obtained by the observation of a sessile drop of water on the solid substrate. The CI/DA particles are hydrophilic with a contact angle of 0° for the infiltration of water drop. The contact angle measured on oleic acid covered particle substrates was significantly different to dimer acid covered ones, with water contact angle of 134° as shown in (b), which indicates the lipophilic property for the OA adsorption laver.

The microstructure of diluted MRF suspensions (2.5 g/L) was

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