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Particle size dependent rheological property in magnetic fluid



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ABSTRACT

The influence of the particle size on the rheological property of magnetic fluid was studied both by the experimental and computer simulation methods. Firstly, the magnetic fluids were prepared by dispersing Fe₃O₄ nanospheres with size varied from 40 nm to 100 nm and 200 nm in the solution. Then, the rheological properties were investigated and it was found that the relative magnetorheological effects increased with increasing the particle size. Finally, the molecular dynamic simulation was used to analyze the mechanical characteristics of the magnetic fluid and the chain-like model agreed well with the experimental result. The authentic chain-like structure observed by a microscope agreed with the simulation results. The three particles composed of the similar cluster nanostructure, thus they exhibited similar magnetic property. To this end, the unique assembling microstructures was the origination of the mechanical difference. And it was found that the higher MR (magnetorheological) effects of the large particle based magnetic fluid was originated from the stronger assembling microstructure under the applying magnetic field.

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1. Introduction

Magnetic fluids are a kind of smart materials which are prepared by dispersing the magnetic particles into the carrying fluids. Because of the magnetic dipolar–dipolar interaction among the magnetic particles, the magnetic fluids possessed typical MR effects, of which the viscosity could change reversibly and rapidly by tuning the external magnetic field [1–3]. According to the dispersing particles, the magnetic fluids could be simply divided into magnetorheological (MR) fluid and ferrofluid [4]. The magnetic saturation of the carbonyl iron particles is larger than the micro/nano-sized iron oxide, therefore, the relative MR effect in the MR fluid is higher than the ferrofluid. Due to their unique magnetic properties, the magnetic fluids have been attractive in a diverse range of applications, such as vacuum sealing, [5] magnetic resonance imaging, [6] intelligent sensors, [7] buffer solution in chips, [8,9] and drug delivery [10].

It is widely accepted that MR effect originates from the disorder to order transition of the particulate assembling under applying an external magnetic field [11]. Therefore, the magnetic property, shape, size, and inner-structure of the magnetic particles become the critical roles for affecting the rheological properties [12–15]. During the past decade, several groups performed intensive work

in this area to investigate the MR mechanism. Magnetic particles with various morphologies such as the cubic, [16] octahedral, [17,18] rod, [19,20] fiber [21] were applied for preparing MR fluids. Stronger assembling chains would be obtained for the anisotropic magnetic particles than the spherical ones, [22] because they have a larger contact surface. The larger friction force among the aggregated particles led to the higher viscosity. Moreover, the recent research indicated that the bundle wire-like aggregations composed of magnetic wires were more stable than the magnetic particles, thus wires based magnetic fluid presented better MR effect than the one composed of monodisperse particles [23]. To this end, the investigation of the particle structure dependent MR effects is favorable.

How does the particle size influence the mechanical properties of the magnetic fluid? This is a very fundamental question for understanding the MR mechanism [24]. Unfortunately, few work has been reported on this subject. The mostly used magnetic particles in MRF is commercial carbonyl iron, whose size is only tunable between several μm to tens of μm . If the size of the iron particles decreases to nano sized, they are very un-stable [25]. Therefore, other particles with tunable sizes are expected to solve this problem. Recently, Fe₃O₄ based magnetic fluids have attracted increasing interests since their better stability than the common carbonyl iron based MR fluid [26–29]. The magnetization of the Fe₃O₄ micro/nanospheres is sufficient for usage thus they could be considered as a proper substitution in MR materials. Various preparation method for Fe₃O₄ micro/nanospheres were reported and the size, shape, magnetic property of the Fe₃O₄ could be

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targetingly obtained [30–33]. Therefore, the Fe_3O_4 is not only an ideal candidate for understanding the MR mechanism but also attracting for its application.

In this work, the influence of the particle size on the rheological properties of the Fe_3O_4 magnetic fluids was both experimentally and theoretically studied. Firstly, the Fe_3O_4 nanospheres with different size were synthesized by using a modified solvothermal method [34]. Then, the rheological property of the Fe_3O_4 magnetic fluids thereof was tested and it was found that the MR effects increased with the particle size. Finally, the molecular dynamic simulation was used to analyze the mechanical characteristics of the magnetic fluid. The proposed chain-like model agreed well with the experimental result. This work is valuable for further understanding the origination of the MR effect.

2. Experimental

2.1. Chemicals

Diethylene glycol (DEG), ethylene glycol (EG), and polyacrylic acid (PAA) were purchased from Sigma-Aldrich. Iron(III)chloride hexahydrate ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$) and sodium acetate anhydrous (CH_3COONa , NaAC) were obtained from Sinopharm Chemical Reagent Co., Ltd. All chemicals were of analytical grade and used without further purification. Deionized water was used in these experiments.

2.2. Preparation of magnetic fluids with different Fe_3O_4 particle sizes

Monodisperse Fe_3O_4 nanospheres with different sizes were synthesized in a binary solvent system with diethylene glycol (DEG) and ethylene glycol (EG). Typically, $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (2.16 g), NaAC (8 g) and PAA (0.2 g) were dissolved in an 80 mL mixture of DEG and EG. After stirring for 30 min, the obtained yellow solution was transferred into a teflon-lined stainless-steel autoclave. The sealed reactor was heating at a temperature of 200 °C for 12 h. By cooling to room temperature, the obtained Fe_3O_4 nanospheres were washed by ethanol and deionized water for 5 times, respectively. Finally, about 0.6 g Fe_3O_4 nanospheres were achieved under drying in a vacuum oven. Here, the size of the Fe_3O_4 nanospheres was tunable by varying the DEG/EG ratio. The ratio of 64/16 leads to 40 nm, while the 60/20 and 50/30 leads to 100 and 200 nm, respectively. The relative Fe_3O_4 nanospheres based MRFs were prepared by dispersing the above powder into the carrying fluid. Thus the relative MRFs prepared by 40, 100, 200 nm nanospheres were defined as MRF-40, MRF-100 and MRF-200, respectively.

2.3. Characterization

The transmission electron microscopy (TEM) images were obtained on a JEM-2100F at an accelerating voltage of 200 kV. The samples which were diluted in absolute ethyl alcohol were deposited on a double-sided copper grid for TEM observation. X-ray diffraction (XRD) patterns of the samples were obtained with a Japan RigakuDMax- γ A rotating anode X-ray diffraction equipped with graphite with graphite monochromatized Cu K α radiation ($\lambda=1.54178 \text{ \AA}$). Infrared (IR) spectra in the wavenumber range 4000–400 cm^{-1} were recorded with a TENSOR Model 27 Fourier transition infrared (FT-IR) spectrometer using a KBr wafer. Thermogravimetric (TG) analysis was conducted on a DTG-60 H thermogravimetric instrument, samples were analyzed in alumina pans at a heating rate of 10 °C min^{-1} to 700 °C under the atmosphere of air flowing at 50 mL min^{-1} . The Au contents of the products were determined on an Optima 7300DV inductive

coupled plasma atomic emission spectrometer (ICP-AES). The UV-vis spectra were records on a TU-1901 spectrophotometer. A magnetic property measuring system (MPMS) vibrating sample magnetometer (VSM) (SQUID, Quantum Design Co., America) was used in studying the magnetic hysteresis loops of Fe_3O_4 nanospheres with different diameters.

2.4. Rheological properties of the magnetic fluid

The rheological properties of the Fe_3O_4 nanospheres based MRFs were investigated by using a commercial rheometer (Physica MCR 301, Anton Paar Co., Austria) with a magnetic field generator. 1 mL of the testing sample was placed on the plate in a uniform magnetic field. The magnetic field was applied perpendicular to the double parallel plates of the rheometer, thus the obtained particle chains were also perpendicular to the flow direction. The gap between the plates was kept at 1 mm. There were two types of measurements: magnetic flux density sweep tests and shear rate sweep tests. Pre-shearing and pre-structuring were carried out before the rheological tests. For the magnetic flux density sweep, the shear rate was set as 10 s^{-1} , 50 s^{-1} , 100 s^{-1} , 200 s^{-1} while the magnetic field density was varied from 0 mT to 160 mT at room temperature. On the other hand, the rheological curves were also obtained in shear rate sweep tests by changing the shear rate, while keeping the magnetic flux density as a constant.

3. Results and discussion

3.1. Characterization of Fe_3O_4 nanospheres with different sizes

In this work, the Fe_3O_4 nanospheres were synthesized by a modified bi-solvent solvothermal method. By varying the ratio of DEG/EG, the size of the Fe_3O_4 nanospheres was tunable. When the ratio was 64/16, 40 nm Fe_3O_4 nanospheres. As shown in Fig. 1, all the obtained Fe_3O_4 nanospheres was well dispersed without aggregations. Typically, these Fe_3O_4 nanospheres were composed of large amount of tiny nanocrystals, thus presented a cluster-like nanostructure, which agreed well with the previous report. The 100 nm and 200 nm Fe_3O_4 nanospheres also exhibited the similar secondary nanostructure. The Fe_3O_4 nanospheres have uniform spherical shape and size with narrow distribution enable them to be ideal candidate for preparing the MRF.

Fig. 2(a) showed the XRD diffraction pattern of the Fe_3O_4 nanospheres with 100 nm. The strong peaks in the pattern could be indexed to be the (220), (311), (400), (422), (511) and (440) crystal face in the cubic Fe_3O_4 (19-0629). No other peak was found in the XRD pattern, indicating the purity of the final product. Moreover, the broad nature of the peak also indicated the Fe_3O_4 nanospheres consisted of small nanograins, which agreed the TEM analysis. Here, the polyacrylic acid was used as the surfactant during the synthesis, thus it inevitably presented in the final nanospheres. Fig. 2(b) presented the typical FTIR spectrum of the Fe_3O_4 nanospheres. The two peaks located at 1550 and 1405 cm^{-1} were corresponded to the COO^- antisymmetrical vibration and COO^- symmetric vibration, indicated that large amounts of carboxylate groups were strongly coordinated to the iron cations [35]. Due to the presence of this water-soluble polymer, the as prepared Fe_3O_4 nanospheres could be dispersed into the water to form stable dispersion. The TG curve demonstrated that there was a large weigh loss between 200–300 °C in Fig. 2(c), which may be attributed to the residue PAA content in the Fe_3O_4 nanospheres. By calculation, the weight ratio of the PAA was about 10%.

The magnetic properties of the Fe_3O_4 nanospheres with different diameters were investigated by a MPMS VSM at room temperature. Fig. 3 shows the hysteresis loops of the samples. All

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