Journal of Magnetism and Magnetic Materials 324 (2012) 424–429

Contents lists available at SciVerse ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Viscosity and dispersion state of magnetic suspensions

Jihoon Jeon, Sangkyun Koo*

Department of Industrial Chemistry, Sangmyung University, Seoul 110-743, Republic of Korea

ARTICLE INFO

Article history:
Received 13 June 2011
Received in revised form
29 July 2011
Available online 22 August 2011

Keywords:
Magnetic suspension
Intrinsic viscosity
Yield stress
Aggregate
Dispersion state

ABSTRACT

We investigate the viscosity behavior of a magnetic suspension in which magnetic particles are dispersed in a mixture of polyacrylic liquids. The size of magnetite particles is nearly 300 nm and the volume fraction of the magnetic particles is in the range of 0.003–0.03. The particle concentration dependence of the suspension viscosity yields the intrinsic viscosity $[\eta]$, which varies from 25.6 at 5 s^{-1} to 5.1 at 400 s^{-1}. The yield stress and the infinite shear viscosity of the suspension increase non-linearly as the particle concentration ϕ increases. We examine the effect of process conditions such as milling time and amount of dispersant on the viscosity behavior of the suspension. As milling time elapses, yield stress and low shear viscosity decrease and then reach constant values while the infinite shear viscosity remains constant. When oleic acid is added as a dispersant, the yield stress and low shear viscosity of the suspension show minimum values as the amount of oleic acid increases. These results agree with experimental results of sedimentation tests, which enable us to estimate the aggregate size of magnetic suspension. The yield stress and the low shear viscosity of the magnetic suspension are found to be useful in evaluating the dispersion state of the magnetic suspension.

1. Introduction

Magnetic particles have recently drawn much attention due to their novel applications in biomedical materials such as targeted drug delivery and contrast agent for magnetic resonance imaging, in addition to their conventional applications for magnetic recording media and printing inks. The magnetic particles have been also considered as a main ingredient for optical materials with controllable microstructure [1–4]. In most cases the magnetic particles are utilized in the form of colloidal dispersion, and it is necessarily important to obtain well dispersed state of the magnetic particles in vehicle liquid. This requires proper characterization of the dispersion state. One of the characterization methods is rheological measurement. Although the rheological analysis is an indirect assessment of dispersion state it has some advantages over optical methods such as light scattering, which are commonly used. The rheological measurement does not require dilution of original liquid vehicle and it is applicable to opaque materials, where light can neither be transmitted nor be reflected. Moreover, in situ measurement is possible. Due to these advantages, rheological methods have been widely utilized to characterize dispersions containing a variety of particles depending on applications.

For magnetic particles Smith and Bruce [5] determined the intrinsic viscosities of magnetic suspensions consisting of acicular ferrite (γ-Fe_2O_3) particles and ethylene glycol. Kwon et al. [6] investigated the effect of particle shape and Peclet number (*Pe*), which is a measure of advection relative to diffusion terms of the flow, on the viscosity of the magnetic suspensions for various shapes of the magnetic particles. These studies focused on understanding the effect of particle shapes that are needle-like or plate-like on the suspension viscosity. For the applications such as magneto-rheology and magnetic fluid, major research interests have been directed to elucidating the magnetic interaction among magnetic particles or the effect of applied magnetic field on the viscosity behavior of the magnetic suspension [7–10].

The present study is concerned with understanding the relation between the viscosity behavior and dispersion state of magnetic suspensions. We deal with magnetic suspension in which magnetite particles of 300 nm in diameter are suspended in a mixture of polyacrylic liquids. This magnetic suspension is formulated to form a solid thin film, in which the magnetite particles are aligned regularly in the polymeric medium. Fig. 1, as an example, shows a cross-sectional image of a magnetic particulate film in which magnetic particles align along the external magnetic field normal to the surface of the film. The spatial distribution of the magnetic particles is controllable with external magnetic field and the physical properties of the particulate film vary accordingly. The overall performance of the magnetic particulate film is much influenced by the dispersion state of the magnetic particles. We carry out rheological analysis of magnetic suspensions to examine microstructure of the suspensions, and the analysis is practically important in determining the optimum manufacturing conditions for the magnetic suspensions.

Specifically we investigate the particle concentration and shear dependence of the viscosity of the magnetic suspension

* Corresponding author. Tel.: +82 2 2287 5338; fax: +82 2 2287 0070.
E-mail address: skkoo@smu.ac.kr (S. Koo).

0304-8853/$ - see front matter © 2011 Elsevier B.V. All rights reserved.
doi:10.1016/j.jmmm.2011.08.025

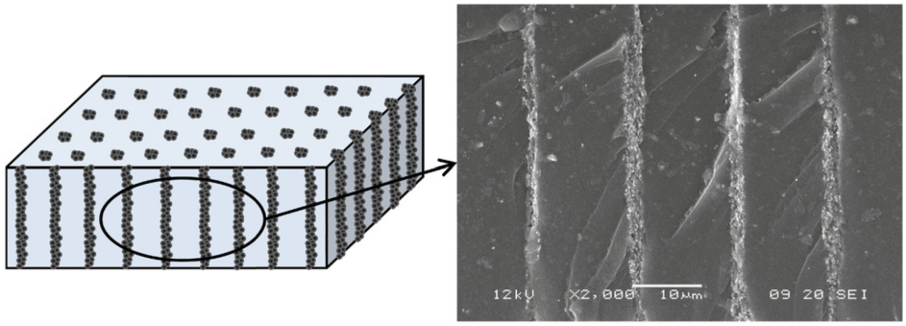

Fig. 1. Cross-sectional view of a magnetic particulate film where magnetic particles are aligned in the direction of an external magnetic field.

and see how the viscosity behavior changes with manufacturing conditions such as milling time and amount of dispersant. The viscosity behavior is compared with the theoretical estimations and the experimental results of sedimentation test, which measure average sedimentation velocity of dispersed particles and thus enable us to estimate the average size of settling aggregates. Finally we will show that the viscosity measurement is quite useful to evaluate the dispersion state of the dispersion.

2. Material and methods

We use magnetite particles (Fe_3O_4) obtained from Saehan Media Company (grade SMT-01S) in Korea. The average diameter of the particles is about 300 nm and the specific surface area and magnetic saturation are 8.5 m^2/g and 85 emu/g, respectively. Fig. 2 shows a photographic image of the magnetite particles by a scanning electron microscope and it is seen that the size of the magnetite particles is nearly monodisperse. The magnetite particles are dispersed in a mixture of acrylates. We used a polyester acrylate (EBECRYL 810) with molecular weight (M_w) of 1000. The viscosity of this oligomer is 500 cps at 25 °C. Acrylic monomers such as HDDA (1,6-hexanediol diacrylate) and TPGDA (tripropylene glycol diacrylate) were added to the oligomer to yield the mixture of acrylates. All these acrylic oligomer and monomers were supplied by SK Cytec Corporation. As a dispersant we used oleic acid produced by Samchun Chemicals Corporation.

As the continuum phase we prepared a mixture of acrylic liquids consisting of a polyester based oligomer and monomers, which are HDDA and TPGDA. These were mixed in the weight ratio of oligomer:HDDA:TPGDA = 3:3:4. The viscosity of acrylic solution is around 20 cps at 25 °C, which is suitable for the ball mill process. Mechanical dispersion of magnetite particles proceeds stepwise. We took few grams of magnetite particles depending on the particle concentrations ranging from 1 to 3 volumetric percent. The magnetite particles are wetted with 10–15 ml of acrylic liquid with spatula for about 5 min. Then these wetted particles are diluted with the acrylic liquid and vigorously stirred with a mechanical homogenizer (Model HG-150, WiseTIS Corporation). The stirring is carried out at 8000 rpm for 3 min to yield a slurry state of the magnetic mixture. This slurry is treated with a ball mill (Model Wisemix BML, Daihan Company) that is loaded with a cylindrical high density polyethylene bottle (*D* 60 mm and *H* 130 an). This bottle is filled with the magnetic slurry and zirconia beads of 3 . Tdiameter. The volumetric ratio of the magnetic slurry and the zirconia beads is fitted to be unity and the rotating rate of the ball mill shaft is 300 rpm. After 3 h milling, we finally obtain a magnetic suspension.

For characterization of magnetic suspension we assess the viscosity and sedimentation behavior. In order to measure the viscosity behavior, we use a cone and plate type viscometer (Model LVDV-II, Brookfield Co.) implemented with the cone (Model CP-40 and CP-52) with a tilting angle of 4° from the plate plane. The shear rate range is from 5 s^{-1} to 400 s^{-1}. The temperature is kept at 25 °C ($\pm$0.1 °C) by chiller circulating the plate. The viscosity is measured after pre-shearing at 5 s^{-1} for 60 s.

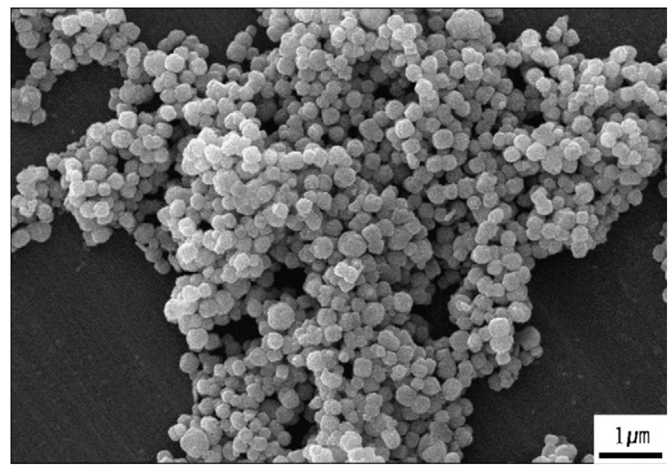

Fig. 2. Photograph of magnetite particles by scanning electronic microscopy.

The sedimentation behavior is analyzed using a cylindrical tube filled with magnetic suspension at 25 °C. As time progresses the dispersed magnetic particles and the aggregates start to settle, forming an interface between clear liquid and settling sediment. We measured the position of the interface as a function of time. This device is designed to keep the temperature at 25 °C ($\pm$0.1 °C) during the sedimentation process.

3. Results and discussion

Microstructure of suspension plays a major role in determining viscosity of the suspension. For low concentrations, at which particle interactions are negligible, the concentration dependence of the suspension viscosity can be described by the well-known Einstein equation $\eta = \eta_0(1+2.5\phi)$, where η and η_0 are the viscosity of suspension and fluid, respectively, and ϕ is the volume fraction of the suspended particles. At high concentrations, the viscosity increases non-linearly with concentration due to particle interactions such as hydrodynamic and Brownian interaction among particles. When the Peclet number *Pe*, given by $a^2\Gamma/D_0$ with a being the particle radius, D_0 the particle self-diffusivity, and Γ the shear rate of the flow, is much larger than unity, the hydrodynamic interaction contributes dominantly to the suspension viscosity. As the particle concentration increases, the distance between particles reduces and thus the hydrodynamic interaction becomes strong. Since the calculation of the hydrodynamic interaction, however,