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Magnetic separation of ferrofluids

S. Thurm*, S. Odenbach

ZARM, University of Bremen, Am Fallturm, 28359 Bremen, Germany

Abstract

The size distribution of nanoscalic magnetic particles in ferrofluids is of fundamental significance if the fluid is subjected to shear flow under the influence of an applied magnetic field. By magnetic separation it is possible to divide a ferrofluid into two fractions which differ in mean particle size. During the separation process large particles diffuse to regions which are located in the stronger field, whereas this effect is very weak for smaller particles. After the experiment there are two different fractions: one fraction with lower, and another fraction with higher percentage of bigger particles. Rheological investigations after the separation process show the important effect of bigger particles on the magnetoviscous behavior of the fluid. It has been found that the fraction with a high percentage of big particles shows a strong, field dependent increase of viscosity, which is also dependent on the shear rate. The other fraction shows just a weak increase of viscosity, and the shear-rate dependence becomes smaller. (C) 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

The appearance of field-dependent increase in viscosity in ferrofluids which are subjected to shear flow under the influence of an applied magnetic field perpendicular to the vorticity of the flow was first observed by McTague [1] in 1969, and theoretically described by Shliomis [2] in 1972. This effect can be interpreted as follows: the shear-flow induced rotation of particles moves the magnetic moment of the particle out of the direction of the external magnetic field. This causes a magnetic torque, which acts against the mechanical rotation generated by the flow. The hindrance of free particle rotation leads finally to a macroscopic increase of fluid viscosity.

Elemental for the field-dependent increase in viscosity, the so-called rotational viscosity, is the kind of relaxation process of the magnetic moment inside the particle. Two different processes in principle are possible. Either the magnetic moment rotates freely inside the particle (the so-called Néel relaxation), or the magnetic moment is fixed within the particles body (the so-called Brownian relaxation). The critical values for the appearance of each process are the corresponding relaxation times—the fastest process dominates in the system. There are two basic assumptions in Shliomis theory: no interactions exist between particles, and only magnetic hard particles exist in the system. Both assumptions do not pertain to commercial ferrofluids. Although the quota of magnetic hard, single particles is very small in those ferrofluids, experiments have shown a much stronger effect on viscosity than theoretically expected. Thus a proof, whether these few large particles or agglomerates can induce the observed effects, is required.

The aim of this project is a controlled separation of magnetic hard particles, to find out detailed information about volume concentration of these particles and inner structure of ferrofluids under the influence of stress and magnetic field. Similar experiments had been performed by Odenbach [7]—magnetic separation due to a field gradient around a thin current leading wire. In the field of HGMS, e.g. Gerber [4] developed a generalized theory, which in principle can be adapted to the experiment described here. The important influence of

^{*}Corresponding author. Fax: +421-218-2521.

E-mail address: thurm@zarm.uni-bremen.de (S. Thurm).

bigger particles on the magnetoviscous effect was qualitatively shown by Odenbach and Raj [8].

2. Numerical simulation

The development of a separation device assumes information about required magnetic field strength, field geometry, and corresponding diffusion times. Calculations of diffusion of nanoscalic magnetic particles in an appropriate carrier liquid due to a magnetic filed gradient can be found in Refs. [3,4,7]. Starting from the continuity equation (1) for the magnetic component:

$$\frac{\partial c}{\partial t} + \nabla \cdot \vec{J} = \frac{\partial c}{\partial t} + \nabla \cdot (\vec{J}_{\rm D} + \vec{J}_{\rm F}) = 0, \tag{1}$$

where *c* is the volume concentration of the magnetic particles, \vec{J} the particle flux and *t* the time. The particle flux can be divided into two parts: the diffusion part $\vec{J}_{\rm D} = -D\nabla c$ and the magnetic force part $\vec{J}_{\rm F} = \vec{v}c$. The diffusion coefficient *D* is calculated from the Nernst-Einstein equation, D = ukT, where *u* is the particle mobility, *k* the Boltzmann constant and *T* the absolute temperature. One can use Stoke's approximation for spherical particles $u = 1/(6\pi\eta r)$, with *r* as particle radius and η as viscosity. The drift velocity \vec{v} can be expressed as $\vec{v} = u\vec{F}$, where \vec{F} is the force acting on a particle. If gravitational forces are neglected, Eq. (1) can finally be written as

$$\frac{\partial c}{\partial t} = \frac{1}{6\pi\eta r} \left[kT(\nabla^2 c) - \nabla \cdot (\mu_0 V_{\text{mag}} M_0 \nabla H c) \right].$$
(2)

The first term on the right-hand side of Eq. (2) is the concentration-gradient compensational part (generally called diffusion), the second part is the forced diffusion part due to magnetic forces. It is assumed that no interparticle interaction exists. Fig. 1 shows numerical concentration profiles for different particle diameters after a separation time of 1 week. The calculation shows



Fig. 1. Numerical calculated time-dependent concentration profiles for three different particle diameters after a separation time of 1 week. Bigger particles move significantly faster than smaller particles

that bigger particles have a much stronger separation effect than smaller particles. This is the main advantage of magnetic separation: due to the high selectivity the separation acts mainly on those particles which play an important role for the magnetoviscous effect.

The diffusion effects after a separation time of 1 week are quite weak. Thus we have to deal with experimental times in the range of many weeks, which in turn means that the requirements for long-term stability are extremely sharp.

3. Experimental setup

Fig. 2 shows a sketch of the experimental setup. The maximal strength of magnetic field gradient ∇H is greater than 10^7 A/m^2 . The trapezoid-shaped container has a volume of up to 300 ml.

During the separation process three coils measure the time-developing concentration profile over the height of the container, which is temperature controlled to ensure that no temperature-driven mass flow in the ferrofluid appears. The fluid which is used for the experiments is APG513A from Ferrofluidics, with a saturation magnetization of 400 G and a viscosity of 120 mPa s.

4. Results

Fig. 3 shows results from shear measurements with a rheometer under the influence of an applied magnetic field. The fluid was exposed to a magnetic field gradient of about 10^7 A/m^2 for a time period of 2 weeks. Shown is the dependence of increasing viscosity versus applied magnetic field, and the variable is the shear rate. The upper three lines represent the lower fraction, the other three lines the upper fraction. The remarkable increase of viscosity in the lower fraction emphasizes the strong influence of bigger particles and agglomerates on the magnetoviscous effect. Of importance is also the



Fig. 2. Sketch of a principle design of the separation device. The fluid container is shaped like a trapezoid, and the strength of magnetic field increases downwards.



Fig. 3. Investigations with a rheometer show a strong sheardependent effect in the fraction with larger particles and agglomerates, whereas there is only a week and shearindependent effect in the other fraction.

influence of higher shear rates on the viscosity. The prediction of cluster and chain formation in suspensions with sufficiently big particles [5,6], which can be destroyed by high shear rates, is confirmed in this experiment.

The upper fraction with a smaller mean particle diameter shows very small effects compared with the lower fraction. In addition to that the shear dependence vanishes. These effects are expected in the case that most of the particles in this fraction behave according to the Néel-relaxation mechanism.

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