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Capillary viscosimetry on ferrofluids

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Abstract

Experiments performed for different ferrofluids under shear flow have shown that an increase of the magnetic field strength applied to the sample yields an increase of the fluid's viscosity, the so called magnetoviscous effect. It has been shown that the magnitude of the effect is strongly related to the modification of the microstructure of ferrofluids and can be influenced by varying both the dipole–dipole interaction between the particles and the concentration of large particles within the fluid. This result has been further used to synthesize new ferrofluids which, on one hand, are more compatible for technical applications but, on the other hand, led to difficulties for the experimenters in measuring the viscous behavior in the presence of a magnetic field. To overcome this problem, a specially designed ferrofluid-compatible capillary viscometer has been developed. Within this paper, the experimental setup as well as experimental results concerning the investigation of the magnetoviscous effect in both diluted and concentrated cobalt-based ferrofluids are presented.

1. Introduction

The physical properties of ferrofluids, colloidal suspensions of magnetic nanoparticles, can be changed by means of moderate magnetic fields. Thanks to this, ferrofluids can be used to solve a wide variety of technical problems. The usage of magnetically controlled fluids requires a good knowledge not only about the basic properties of ferrofluids but also about their behavior in the presence of magnetic fields. Thus, effects such as field-induced changes of fluid viscosity are subjects of current research activities.

Previous experiments for ferrofluids under shear flow [1] have shown that an increase of magnetic field strength yields an increase of the fluid viscosity, the so called magnetoviscous effect, while increasing shear rate leads to a decrease of the viscosity. Combining rheological measurements with small angle neutron scattering investigations, a strong connection between structure formation in ferrofluids under the influence of a magnetic field and their macroscopical behavior has been established [2]. It has been pointed out that for the samples investigated, under the influence of a magnetic field, chains are formed within the fluids, contributing to the increase of the fluid's viscosity, while by increasing the shear rate, the viscosity gets reduced as a result of structure disruption.

While the model of rotational viscosity elaborated by Shliomis [4] describes well the behavior of highly diluted systems, former experiments [3] have shown that for commercial, concentrated fluids, the inter-particle interaction, which causes the formation of chain-like structures, has to be taken into account. Thus, an essential parameter for each fluid is the interaction parameter λ^* [5]:

$$\lambda^* = \frac{\mu_0 M_0^2 V}{24 k_B T} \left(\frac{d}{d+2s}\right)^3 \qquad (1)$$

where M_0 denotes the spontaneous magnetization of the magnetic material, d the mean magnetic diameter of the particles and V their respective volume. The thickness of the surfactant layer is denoted by s.

Defined as the ratio between the dipole–dipole energy of interaction between two neighboring particles and their thermal energy, λ^* describes the possibility that the particles contained in a ferrofluid interact and form chains. The formation of the chains can appear only for values of the interaction parameter larger than unity, i.e. if the interaction between the particles is strong enough to keep the particles together and overcome their thermal motion. By comparing the values of λ^* calculated for magnetite and cobalt particles it has been obtained that in the case of magnetite, the particles should be larger than 16 nm to contribute to the formation of structures, whereas 6.5 nm cobalt particles are already able to form chain-like structures.

While previous rheological experiments could be successfully performed using a cone-plate rheometer, new problems arise for the investigation of the highly concentrated Co ferrofluid samples. For example, by applying a magnetic field perpendicular to the free surface of a ferrofluid, fluid spikes can be formed, whereas the critical magnetization for the appearance of this instability is given in [6] as

$$M^2_{\text{critical}} = \frac{2}{\mu_0}\left(1+\frac{1}{\mu}\right)(\rho g \sigma)^{\frac{1}{2}} \tag{2}$$

with μ denoting the permeability, ρ the density and σ_s the surface tension of the ferrofluid. On increasing the magnetic field strength, the height of the spikes formed on the free surface of the ferrofluid varies, depending on the physical properties of the fluid, i.e. surface tension, magnetization etc. Therefore, the maximum magnetic field strength that can be applied to each fluid is limited by the appearance of a critical height of the spikes. The rheological investigations using a cone-plate rheometer [7] are possible only up to magnetic field strengths well below the value corresponding to the critical magnetization required for the appearance of the Rosensweig instability. Thus, for magnetite-based ferrofluids as well as for low concentration cobalt suspensions the measurements can be performed for a relatively wide range of magnetic field strengths, whereas for concentrated cobalt-based ferrofluids a viscometer without a free surface of the fluid has to be used. To overcome this problem, a specially designed capillary viscometer has been developed. The major advantage is that closed channel flows have no free surface in the test region. Since there is no free surface of ferrofluid in an applied magnetic field, the Rosensweig instability does not appear. In order to estimate the influence of the magnetic field on the viscosity of the concentrated cobalt ferrofluids as well as the consequences at the microstructural level, a dilute cobalt-based ferrofluid sample has been considered as a reference and it has been measured with both a capillary viscometer and a cone-plate rheometer.

2. Experimental setup

In this section the main features of the capillary viscometer developed for measurements with highly concentrated cobalt-based fluids will be outlined. The viscometer (see figure 1) works on the basis of a constant velocity piston driving ferrofluid from the reservoir through the capillary. By employing a differential pressure transducer, the pressure drop along the capillary is measured. For a constant flow rate, and considering the geometrical parameters of the capillary, the viscosity η of the fluid can be calculated using the Hagen–Poiseuille law [8]:

$$\dot{V} = \frac{\pi \Delta p R^4}{8\eta L} \Leftrightarrow \eta = \frac{\pi \Delta p R^4}{8\dot{V} L} \tag{3}$$

with $\dot{V}$ denoting the flow rate and Δp the pressure drop; R and L are the radius and the length of the capillary respectively.

To derive the viscosity relation, the following necessary assumptions have been made. First, there is no slip at the walls (i.e. the fluid velocity at the wall is zero). Second, the fluid streamlines are parallel to the wall and there is no radial or circumferential flow. Last, the hydrostatic pressure is

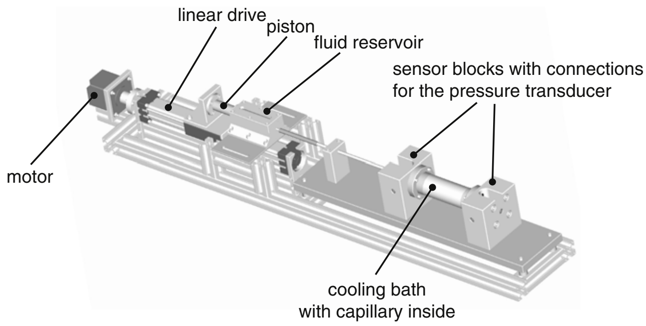

Figure 1. Technical 3D drawing of the capillary viscometer designed for the investigation of highly concentrated Co ferrofluids.

uniform across any radial section of the capillary and the fluid is incompressible, with a viscosity independent of pressure.

For Newtonian fluids the measured volumetric flow rate $\dot{V}$ is related to the shear rate $\dot{\gamma}$ by the expression

$$\dot{\gamma} = \frac{4\dot{V}}{\pi}\frac{r}{R^4}. \tag{4}$$

For the experimental setup shown in figure 1, the wall shear rate, calculated with equation (4) for $r = R$, can be varied by changing the velocity of the piston that drives the ferrofluid from the reservoir to the capillary. The piston is connected to a stepping motor through a linear drive system ensuring a precise driving of the piston. The low shear rate regime can be achieved by mounting an additional 1:512 planetary gear. Thus, for a radius of the capillary of 1.2 mm, a wall shear rate range between 10^{-6} and 10^3 s^{-1} can be covered.

Homogeneous magnetic fields up to about 25 kA m^{-1} are created by a system of four coils in a Fanselau arrangement. The magnetic field can be orientated parallel as well as perpendicular to the flow. By measuring the pressure drop along the capillary with and without applied magnetic field, information about field-induced changes of viscosity are obtained.

3. Ferrofluid samples

Focusing on the class of ferrofluids having a strong interaction parameter, three cobalt-based ferrofluids, Co87_02, Co87_03 (10 nm cobalt particles coated with an aluminum oxide shell, stabilized with korantin SH and suspended in kerosene) and Co_91 (3.25 vol% 10 nm cobalt particles coated with an aluminum oxide shell, stabilized with korantin SH and suspended in L9 oil), supplied by Matoussevitch and Boennemann (Mülheim/Karlsruhe), have been investigated.

The samples Co87_02 (0.85 vol% magnetic material) and Co87_03 (0.35 vol%) originate from the same fluid batch, via simple dilution. This means that they have the advantage of an identical mean particle diameter and the same particle size distribution.

Figure 2 shows the magnetization curves for Co87_02 (black squares) and for Co87_03 (open circles) normalized to their saturation magnetization. Whereas the concentration of magnetic material is evaluated from the saturation

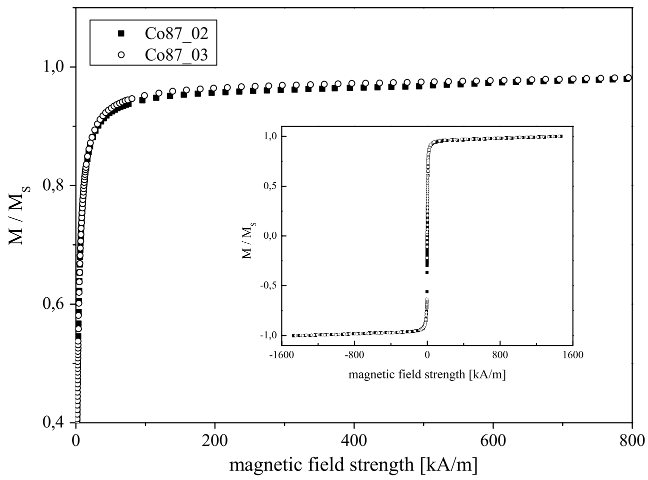

Figure 2. Magnetization curves for Co87_02 ($\phi = 0.85$ vol%, $\chi_i = 2.93$, $M_s = 12.53$ kA m^{-1}) and for Co87_03 ($\phi = 0.35$ vol%, $\chi_i = 1.10$, $M_s = 5.08$ kA m^{-1}) normalized to the corresponding saturation magnetization.

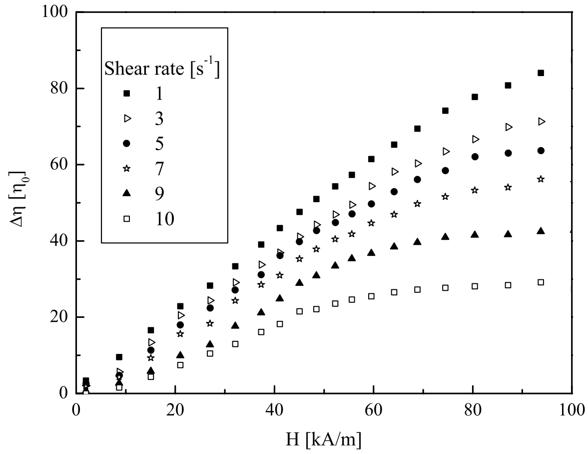

Figure 3. Magnetoviscous effect for Co87_03 for shear rates between 1 and 10 s^{-1} measured with a cone-plate rheometer.

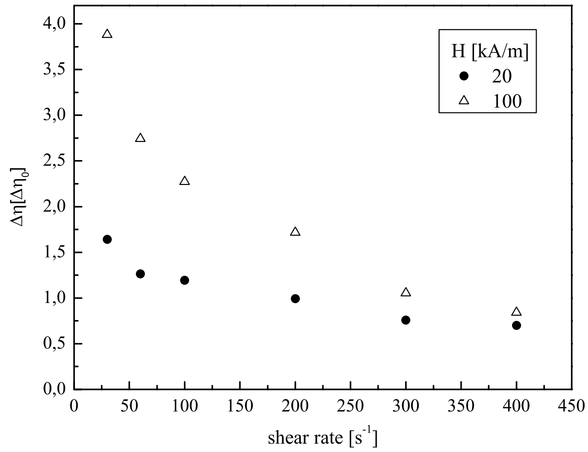

Figure 4. Magnetoviscous effect for Co87_03 for shear rates between 30 and 400 s^{-1} measured with a cone-plate rheometer.

magnetization and the mean particle diameter from the initial susceptibility, the shape of the magnetization curve between these two regions is determined by the particle size distribution. The identical shapes of the curves confirms that no difference in the particle size distribution functions for these two fluids appears. Therefore, a variation of the magnitude of the magnetoviscous effect for these two fluids should appear only due to the difference in the volume fraction of the cobalt particles.

Additionally, since the particle sizes are distributed in the range from 7 nm to about 16 nm [9], all particles can contribute to chain formation and therefore strongly influence the magnitude of the magnetoviscous effect. The corresponding interaction parameter, calculated for a particle diameter of 10 nm and a thickness of the surfactant of 2 nm, is $\lambda^* = 5.26$.

4. Rheological investigations

As mentioned in section 1, in order to estimate the influence of the magnetic field on the viscosity of the concentrated cobalt ferrofluids, a dilute ferrofluid sample, Co87_03, has been considered as a reference and it has been measured with both a capillary viscometer and a cone-plate rheometer.

In figure 3 the increase of the viscosity due to the magnetic field influence is presented. The measurements have been performed with a cone-plate rheometer [7] with a magnetic field oriented perpendicularly to the direction of the vorticity of the flow. In order to observe the flow behavior at magnetic field strength values close to those corresponding to the saturation of magnetization, the magnetic field range has been extended up to 100 kA m^{-1}.

It can be seen that an increase of the magnetic field strength leads to a strong rise of the viscosity, especially at low shear rates. In [2] it has been shown that this behavior can be correlated with the formation of chains within the fluid while the shear thinning, shown in figures 3 and 4, is evidence of disruption of chains in the flow. Peculiar to the Co87_03 fluid is its behavior at high shear rates (see figure 4). In contrast to the magnetite-based ferrofluid treated in [2], the cobalt-based ferrofluid shows, at shear rates up to about 400 s^{-1}, a magnitude of the magnetoviscous effect which cannot be neglected. The shear stability even in the high shear rate regime indicates the presence of chain-like structures which are not completely destroyed.

Compared with the cone-plate rheometer, the capillary viscometer has the disadvantage of a variable shear rate that depends on the radial position of the fluid element. Additionally, the flow profile is symmetrical with respect to the axis of the capillary. Thus, assuming that under the influence of the magnetic field chain-like structures are formed within the ferrofluid samples, it is expected that they will behave differently from the structures formed in the cone-plate system. As a result, the magnitudes of the magnetoviscous effect obtained with the two systems, the capillary viscometer and the cone-plate rheometer, cannot be compared directly. Therefore, in order to estimate the magnitude of the magnetoviscous effect measured with the capillary viscometer, an effective shear rate for the capillary flow has been defined. For this, the

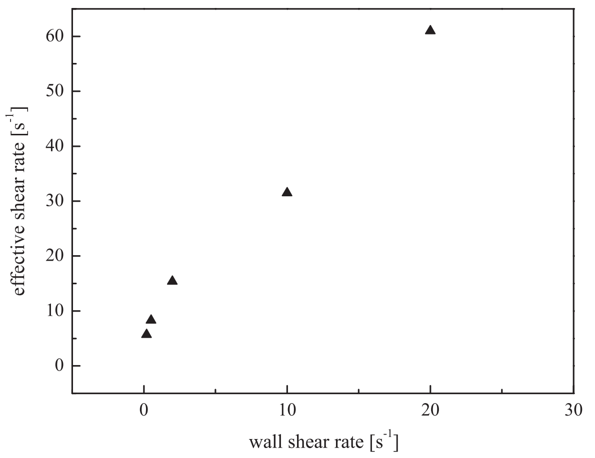

Figure 5. Effective shear rate (cone-plate geometry) corresponding to the wall shear rate for the capillary viscometer.

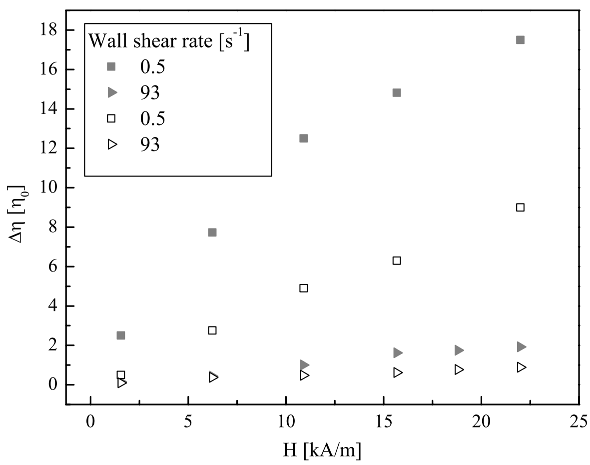

Figure 6. Magnetoviscous effect for Co87_02 (closed symbols) and Co87_03 (open symbols) measured with the capillary viscometer.

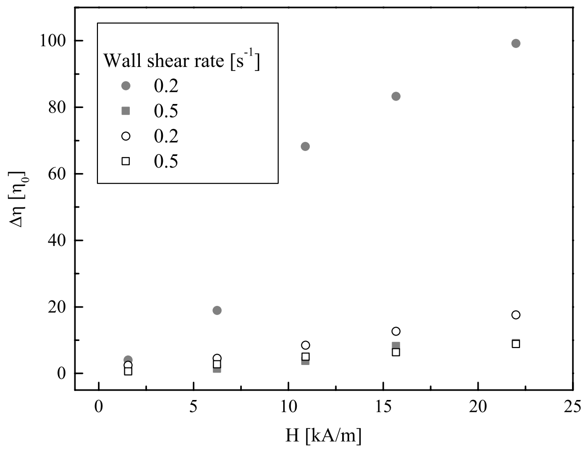

Figure 7. Magnetoviscous effect for Co91 (closed symbols) and Co87_03 (open symbols) measured with the capillary viscometer.

relative increase of the viscosity measured with the cone-plate rheometer has been plotted as a function of the shear rate, for one magnetic field strength. Further, the field-induced changes of the viscosity measured with the capillary viscometer have been plotted in the same diagram so that they fit to the curve obtained for cone-plate rheometer. The corresponding shear rates, i.e. the shear rates which in a cone-plate geometry would correspond to the magnitudes of the magnetoviscous effect as measured with the capillary viscometer, have been considered as effective shear rates.

In figure 5 the relationship between the effective shear rates and wall shear rates in the capillary is presented. As can be seen, there is no linear dependence between these two types of shear rate, which might be connected to the non-Newtonian behavior of ferrofluids. A detailed study of this dependence with various magnetic field strengths as well as in the absence of the magnetic field is foreseen.

Using the capillary viscometer presented in section 2, the ferrofluid samples have been investigated for a wall shear rate range between 1 and 100 s^{-1}. Magnetic field strengths up to 25 kA m^{-1} orientated parallel to the flow direction have been applied to the samples. Thus, the magnetic field is always perpendicular to the vorticity of the flow, therefore ensuring a maximum magnetic field-induced change of the viscosity [4].

As mentioned in section 3, the Co87_03 and Co87_02 samples differ only in concentration of the magnetic material. The ratio of their concentrations can be found again as a factor of proportionality between the magnitudes of the magnetoviscous effect. As it can be seen in figure 6, a concentration of the cobalt particles about twice larger than that in Co87_03 leads to a magnetoviscous effect which is about two times stronger in the Co87_02 sample.

For Co91, the most concentrated cobalt-based ferrofluid studied within this work, it can be seen that an increase of the concentration of about 10 times compared to the Co87_03 sample produces field-induced changes of the viscosity which are, for the lowest shear rate presented here (figure 7), only about five times larger. Additionally, a strong shear thinning can be observed for Co91; the magnitude of the magnetoviscous effect reduces to values comparable to those obtained for Co87_03. In order to understand this behavior it has to be taken into account that the viscosity of the carrier liquid used for Co91 is about twenty times larger than that for Co87_03. A high viscosity, i.e. strong viscous forces acting on the particles and on the structures formed within the fluid, limits the maximal length of the chains [1]. This could be an explanation for the relatively low increase of the viscosity as well as for the strong shear thinning of the Co91 sample, compared to the Co87_03 ferrofluid.

5. Conclusions

The experimental results presented in the previous section have shown that the capillary viscometer can be successfully used for the investigation of the magnetoviscous effect in both diluted and concentrated cobalt-based ferrofluids. Effects like shear thinning as well as the shear stability at high values of the shear rate have been confirmed not only for the low concentration sample, Co87_03, but also for the more concentrated ones. The comparison between the fluids originating from the same batch, Co87_02 and Co87_03, supports the finding from the small angle neutron scattering experiments [2] according to which the length and the orientation of the chains formed within these fluids remain the same for comparable shear rate and magnetic field strength. The only difference between these samples is the different concentrations of the magnetic material and, in the presence of a magnetic field, the different numbers of chains.

In future, the modularity of the capillary viscometer will allow a variation of the angle between the orientation of the magnetic field and the direction of the flow. This will provide additional information concerning the anisotropy of the magnetoviscous effect and will enable a comparison between experimental data and the results of non-equilibrium computer simulations leading altogether to a better understanding of the magnetoviscous effect in concentrated ferrofluids.

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