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Rheological investigations of ferrofluids with a shear stress controlled rheometer

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Abstract
The appearance of field- and shear-dependent changes of viscosity—the magnetoviscous effect—is correlated to the formation of chains and structures of magnetic nanoparticles. Moreover, the formation of these structures leads to the appearance of viscoelastic effects or other non-Newtonian features in ferrofluids in the presence of a magnetic field. In order to describe these phenomena, different theoretical approaches have been developed which explain the mechanism of these effects with different assumptions. One point in which these models differ, and which has to be clarified, is the appearance of yield stress and its dependence on magnetic field strength. With this aim, a stress controlled rheometer has been designed to prove the existence of this very small field-dependent yield stress for ferrofluids. The results presented here show a dependence of the yield stress on the magnetic field strength as well as on the interparticle interaction and particle size distribution. Finally, yield stress experiments have been performed for different geometries of the shear cell in order to get more information about the microstructure formed by the magnetic particles.

1. Introduction

Previous experimental studies carried out with differently composed ferrofluids have shown that an increase in the magnetic field strength leads to strong changes in the viscosity. This phenomenon—the magnetoviscous effect—has been attributed to the formation of chain-like clusters due to strong interparticle interaction in the presence of a magnetic field. The hindrance of rotation of these clusters gives rise to an increase in viscosity under the influence of a magnetic field. In addition, the rupture of these clusters under the influence of the shear stress applied to the fluid is used as an explanation for the shear thinning [1, 2].

Over the last few years many theoretical studies have been performed to explain these effects, following different approaches. Some groups have based their investigation on microscopic assumptions [3–5], while others have based their studies on the mesoscopic or macroscopic level [6, 7]. In this context the models lead to different predictions and open up new questions, which are still waiting to be answered. A point which has to be clarified is the question of the appearance of yield stress in ferrofluids and its field dependence.

Earlier experiments carried out with different ferrofluid samples demonstrate a Newtonian behaviour in the absence of a magnetic field, and a non-Newtonian behaviour when a magnetic field is applied. Additionally, the flow curves of the ferrofluids tend to end up with a constant stress value—yield stress—when the flow curves are extrapolated to a shear rate of zero. An increase in the applied magnetic field increases this apparent yield stress. With a shear rate controlled rheometer—as has been used to study the magnetoviscous behaviour of ferrofluids [8]—the question of whether this extrapolated offset in stress is a real static yield stress cannot be answered. Clarification of this question requires stress controlled investigations, as discussed below.

2. Experimental data

2.1. Ferrofluid samples

Corresponding to the chain formation model [4], the magnetoviscous effect is described as a result of the formation of chain-like structures due to strong interparticle interaction under the influence of a magnetic field. The interparticle interaction can be quantified with a modified interaction parameter λ^* [9], which has been shown to determine the magnitude of the magnetoviscous effect

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

Here μ_0 denotes the vacuum permeability, M_0 the spontaneous magnetization of the magnetic material, V the volume of the magnetic particles, k_B the Boltzmann constant and T the absolute temperature. The mean diameter of the magnetic particles is denoted by d and the thickness of the surfactant layer is given by s. The parameter λ^* is thus simply the ratio of magnetic dipole interaction energy of two coated particles in contact to their thermal energy. Chain-like clusters can appear if the particle interaction is strong enough to overcome thermal mixing; i.e. for a modified interaction parameter λ^* larger than unity. By calculating the values of λ^* for magnetite and cobalt particles, one can predict that magnetite particles larger than 16 nm are able to form chains and structures, and in the case of cobalt the particles should be larger than 6.5 nm. Thus—besides the value of λ^*—the number of particles with diameters larger than the mentioned thresholds is of great importance for the magnetoviscous properties of a ferrofluid [9].

In order to get more information about the influence of interparticle interaction and volume concentration of sufficiently large particles on the yield stress, ferrofluids of different compositions have been investigated. For the experiments two commercial fluids of the same type, Ferrotec APG513A (7.2 vol% magnetite in synthetic ester, dynamic viscosity $\eta_{(T=20\,^\circ\mathrm{C})} = 170$ mPa·s), but with different content of particles larger than 16 nm, and one cobalt based ferrofluid, Co220_02 (2.83 vol% cobalt in L9 oil, dynamic viscosity $\eta_{(T=20\,^\circ\mathrm{C})} = 65$ mPa s) prepared by Matoussevitch at Forschungszentrum Karlsruhe GmbH, were used. The particles in the first magnetite based ferrofluid (sample 1) have a mean diameter of 10 nm, but this fluid has a broad particle size distribution compared to sample 2. A small number of particles in sample 1 with a volume concentration $\phi = 0.8$ vol% have a mean diameter larger than about 16 nm and can contribute to the clusters. The interaction parameter for this fraction of particles is $\lambda^* = 2.87$, considering a thickness of the surfactant layer of 2 nm. Sample 2 contains only small particles with a mean diameter $d = 10$ nm; for this sample a modified interaction parameter $\lambda^* = 0.5$ has been calculated. In the cobalt based ferrofluid (sample 3) all particles are large enough to form chain-like structures. The mean diameter of the particles is $d = 8$ nm and the modified interaction parameter amounts to $\lambda^* = 2.18$.

2.2. *Investigations of the yield stress in ferrofluids*

The yield stress experiments were carried out with a stress controlled rheometer [10] in a magnetic field range from 0 to 80 kA m^{-1}, and the shear stress was varied within the range from 0 to 1 Pa. The measuring cell is a cone-plate arrangement (opening angle 3°, cone diameter 70 mm, mean gap distance $d_{\mathrm{mean}} = 1.75$ mm) combined with a Couette region. Starting from zero, the stress is increased stepwise until some movement of the fluid can be observed. Repeating this procedure for different magnetic field strengths, the dependence of the yield stress on magnetic field strength can be obtained.

In figures 1(a) and (b), the dependence of the yield stress on magnetic field strength for different interparticle interactions and volume concentration of cluster-forming particles for the differently composed ferrofluids is presented. For samples 1 and 3 (figure 1(a)), the yield stress grows with the square of magnetic field strength. This observation is in accordance with the usual description of the magnetoviscous effect. The particles in the fluid interact and link together to form chains and other structures. When the mechanical stress imposed on the fluid exceeds a certain critical magnitude, these structures break up and the fluid flows. The highest value of stress for which no flow is observed is defined as the yield stress. As seen in figure 1(b), for sample 2 no yield stress could be detected even for the highest magnetic field strength applied. Due to the fact that this fluid contains only small particles, no significant chain and structure formation should appear. The experimental results for yield stress with the magnetite based ferrofluids (samples 1 and 2) agree with the presumption that there is a correlation—as was proved earlier by experimental as well as theoretical results for the magnetoviscous effect [1]—between the interparticle interaction of large particles and the yield stress.

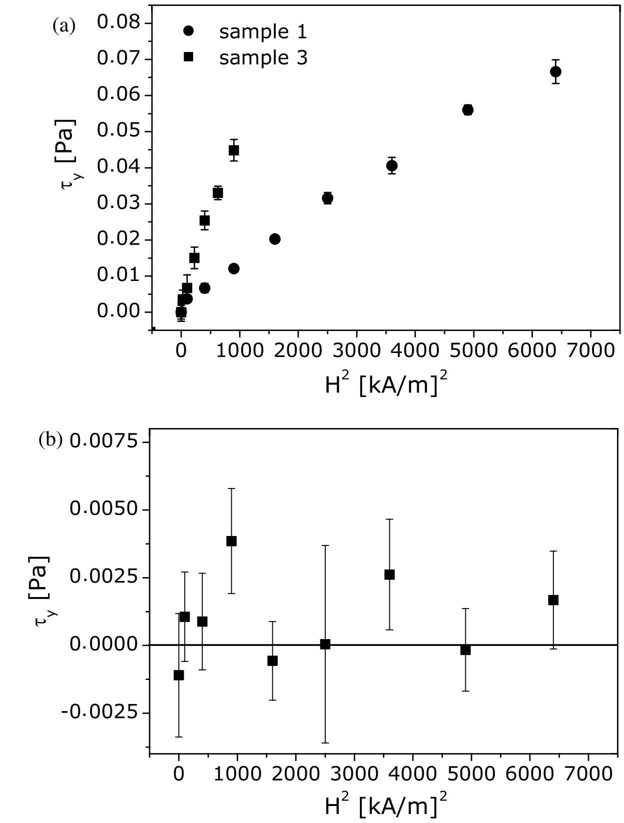

Figure 1. (a) The yield stress for fluids containing structure-forming particles. Interpretation is given in the text. (b) The yield stress for sample 2 containing no structure-forming particles. For this ferrofluid no yield stress could be observed.

Comparing the field-induced yield stress for sample 1 and sample 3 in figure 1(a), it can be observed that the magnitude of the yield stress of the cobalt based ferrofluid is much higher than for the magnetite based ferrofluid, despite the slightly stronger modified interaction parameter λ^*. This leads us to the conclusion that the yield stress does not only depend on

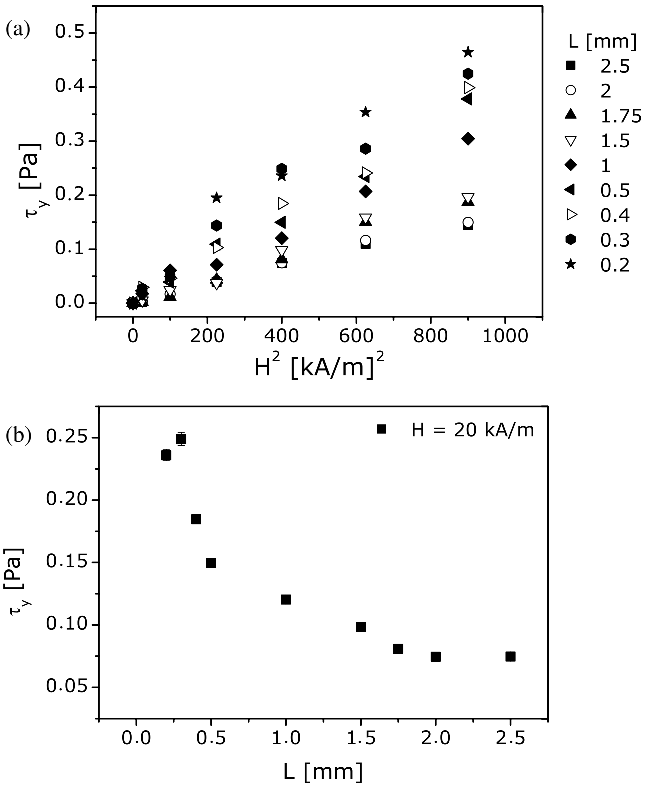

Figure 2. (a) The yield stress for the ferrofluid sample 3 as a function of the square of the applied magnetic field for various distances between the walls in plate–plate geometry. With a decrease in the gap distance, the yield stress increases significantly. (b) The yield stress for the ferrofluid sample 3 as a function of gap distance L for a magnetic field strength of 20 kA m^{-1}.

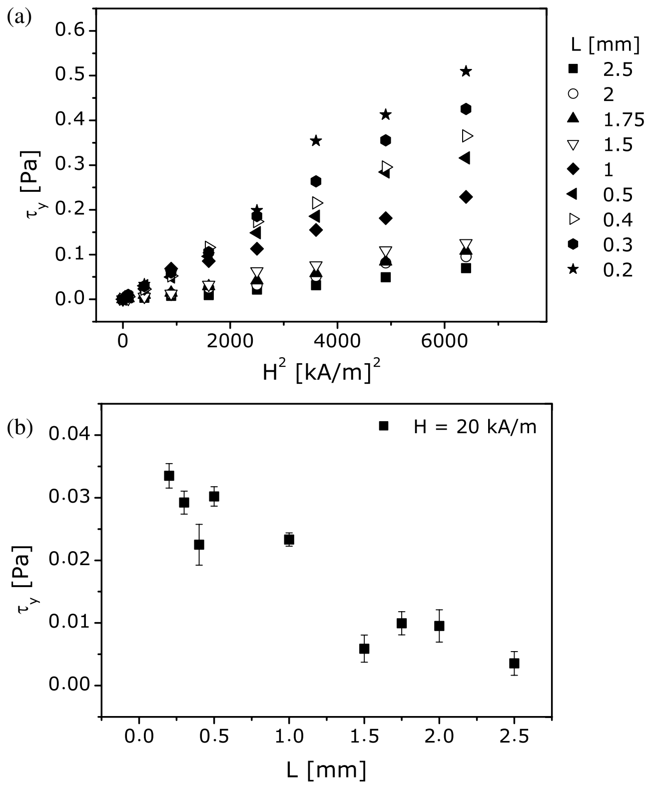

Figure 3. (a) The yield stress is obtained for sample 1 as a function of the square of the strength of the applied magnetic field for various distances between the walls in plate–plate geometry. With a decrease in the gap distance L, the yield stress increases significantly. (b) The yield stress for the ferrofluid sample 1 as a function of gap distance L for a magnetic field strength of 20 kA m^{-1}.

λ^*, but also on the concentration of large particles, which are the ones contributing to the formation of chain-like structures. The number of strongly interacting particles in sample 3 is 3.5 times higher than in sample 1, and this is reflected in the stronger yield stress effect observed in sample 3 which exceeds the effect in sample 1 also by a factor about 3.5 for $H = 30$ kA m^{-1}.

These results demonstrate that the magnitude of the yield stress depends strongly on the interparticle interaction as well as on the number of large particles, which are able to form chains and structures. Finally, a variation of the geometry of the shear cell can be used to obtain information on the microstructure formed by the magnetic particles and its dependence on magnetic field strength. For the experiments we investigated samples 1 and 3, which have shown yield stress effects before. A plate–plate configuration was used and the gap L varied from 0.2 to 2.5 mm. The stress applied to the fluid in the measuring cell—as mentioned above—was increased stepwise from zero until a flow could be observed. This procedure was repeated for different magnetic field strengths. In figure 2(a) the yield stress for sample 3 is plotted versus the square of the applied magnetic field for different gap distances L. For each gap, the yield stress grows quadratically with the magnetic field strength independent of the value of L. With a decrease of the gap distance, the yield stress increases significantly. As an example, the results of the yield stress as a function of gap distance L for $H = 20$ kA m^{-1} applied to the sample are shown in figure 2(b). The yield stress increase about 3.3 times when the gap L decreases from 2.5 to 0.2 mm. As mentioned before, the appearance of the yield stress can be considered as an effect of particle–particle interactions within the ferrofluid sample. With an applied magnetic field, the magnetic moments of the particles try to align along the magnetic field lines; particles interact and form chain-like clusters. A combination of magnetic field strength and gap distance L determines the change of the chains and structures, which lead to a specific magnitude of the yield stress. In figure 2(b) it can be seen that the rate at which the yield stress changes with the gap distance decreases as the gap distance increases. The influence of the gap distance on the yield stress is stronger with smaller gaps. For very large gap distances, the influence of the walls on the microstructure is negligible.

In figures 3(a) and (b), the dependence of the yield stress on magnetic field strength and gap distance L for the magnetite based ferrofluid (sample 1) is presented. For each gap distance, the yield stress—as for sample 3 (figure 2(a))—is proportional to the square of the applied magnetic field strength. A decrease in the gap distance leads to an increase of the yield stress. As an example, the results for the yield stress as a function of gap distance at $H = 20$ kA m^{-1} applied to the sample are given in figure 3(b). The yield stress increases about nine times when the gap L is decreased from 2.5 to 0.2 mm.

One finds that the cobalt based ferrofluid (sample 3) tend to show a finite constant limit of yield stress for large gap distances while for the magnetite based ferrofluid (sample 1) yield stress vanishes for large geometric dimensions. This effect can be again associated with the large number of chain-forming particles which are able to provide gap-spanning structures for the cobalt based fluid for large geometries.

3. Discussion

The rheological investigations presented here have shown that the yield stress is proportional to the square of applied magnetic field strength. Moreover in the experiments with the magnetite based ferrofluids (samples 1 and 2), the dependence of the yield stress on interparticle interaction has been confirmed. Comparing sample 1 and sample 3, which have the same interaction parameter but different concentration of large particles, it can be observed that the yield stress of the cobalt based ferrofluid (sample 3) is much stronger than for the magnetite based ferrofluid (sample 1). The results verify that there is a strong connection between the yield stress and the number of large particles, which are the ones able to form the chain-like structures.

Finally, a particular combination of magnetic field strength and gap distance L leads to a specific magnitude of the yield stress. An increase in the gap distance L leads to a strong decrease in yield stress; i.e. an influence of the macroscopic distance of the walls of the shear cell on the microstructure formed by the magnetic particles can be observed. Comparing sample 1 and sample 3, keeping the magnetic field constant, it has been found that the relative change of the yield stress of sample 1 with an increase in L is much stronger than for sample 3, leading to a tendency of vanishing yield stress for large gaps in sample 1. The higher number of structure-forming particles leads in contrast in sample 3 to a finite static yield stress limit.

Future investigations will focus on the specific interaction between the particles or structures and the walls, followed by a comparison between the experimental and the theoretical results based on different assumptions. For these both material and roughness of the walls will be varied. Furthermore, investigations concerning a variation of the interaction parameter by a further change of size and material of the magnetic particles will be performed.

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