THE MICROSCOPIC MECHANISMS OF THE MAGNETOVISCOUS EFFECT IN FERROFLUIDS INVESTIGATED BY SMALL ANGLE NEUTRON SCATTERING

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Experimental studies made for different ferrofluid samples under the shear flow have shown that increasing the magnetic field strength yields an increase of the fluids viscosity, the so-called magnetoviscous effect, while increasing the shear rate leads to a decrease of the viscosity. The change of the viscosity with magnetic field strength can be theoretically explained as an effect of chain-like structure formation and, therefore, related to the modification of the microstructure of ferrofluids.

Using a specially designed rheometer, ferrofluids having different magnitude of the magnetoviscous effect were investigated by small angle neutron scattering (SANS). The obtained results show a good agreement with the qualitative model elaborated to explain the magnetoviscous effect.

Introduction. Rheological investigations of field induced changes of the viscosity of ferrofluids under the shear flow have shown that the magnitude of the magnetoviscous effect depends especially upon the concentration of large particles contained in ferrofluids [1]. While the classical theory of rotational viscosity [2] explains the magnetoviscous effect with the hindrance of the free rotation of particles in a shear flow by means of magnetic fields, the disagreement between the small content of large particles and the magnitude of the effect indicates a correlation of the magnetoviscous effect to structure formation in ferrofluids [3] due to the interaction between the magnetic particles. Therefore, the effect can be explained with chain-like structures formed in the fluids being aligned with the magnetic field. A shear flow applied to the fluid sample will give rise to a mechanical torque that diverts the chains from the initial direction, whilst a magnetic torque counteracts this misalignment [3]. An increase of the viscosity can be observed. Furthermore, with increasing the shear rate the relative change of viscosity diminishes (shear thinning). In this case the mechanical torque becomes dominant and the chains are broken.

Ferrofluids having a mean diameter of the particles of about 10 nm but with different size distribution of the particles and therefore with different particle-particle interaction have been investigated. The measured relative changes of the viscosity reveal a high magnitude for the fluid with strong interaction, as seen in Fig. 1, Ferrotec APG513A (7.2 vol.% magnetite in synthetic ester), whereas the fluid with a weak one, TOA (7.2 vol.% magnetite in transformer oil [4]) shows a low magnetoviscous effect (Fig. 2).

1. Experimental results. To point out the connection between the rheological behaviour of ferrofluids and their microstructure, not only rheological but also small angle neutron scattering investigations have been performed in a specially designed rheometer [5].

The SANS experiments were carried out in a magnetic field range from 0 to 160 kA/m, directed parallel to the neutron beam. Rheological measurements of both ferrofluids have shown no change of viscosity with the magnetic field strength

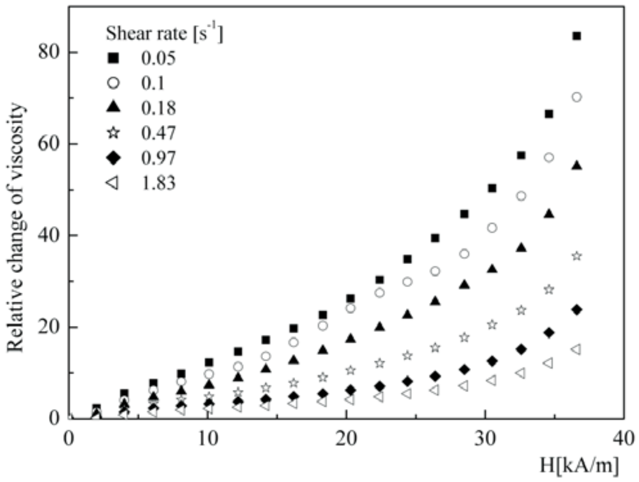

Fig. 1. Magnetoviscous effect in a ferrofluid sample with a strong particle-particle interaction (APG 513A).

for high shear rates $\dot{\gamma} > 30\ s^{-1}$. According to the theoretical model, all chains are broken in this situation and the particles are homogeneously distributed in the fluid. Thus, the fluid can be assumed as a single particle system. Therefore, the high shear rate ($\dot{\gamma} = 200\ s^{-1}$) situation, i.e., the single particle system, has been considered as a reference. To eliminate the contribution of the small particles, the surfactant and the carrier liquid to the scattering, the reference was subtracted from the scattering patterns obtained for shear rates varied within a range from 0 to 200 s^{-1}. The difference scattering patterns obtained for the fluid with high magnetoviscous effect, APG513A, indicate a strong dependence of the scattered intensity on the shear rate and the magnetic field strength (Fig. 3).

For a high magnetic field strength and a low shear rate (Fig. 3*a*) the difference pattern is almost zero. Due to the high magnetic field, long segments of chains are formed, but their deviation from the initial direction is very small. Since the cross-section of the chains, as it can be seen by neutrons, have almost the same

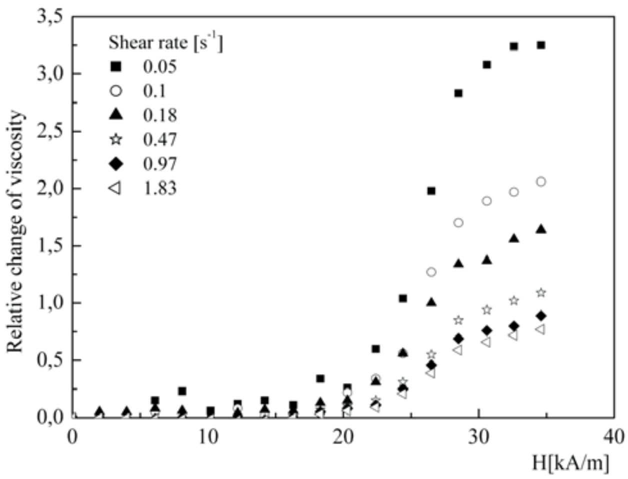

Fig. 2. Magnetoviscous effect in a ferrofluid sample with a weak particle-particle interaction (TOA).

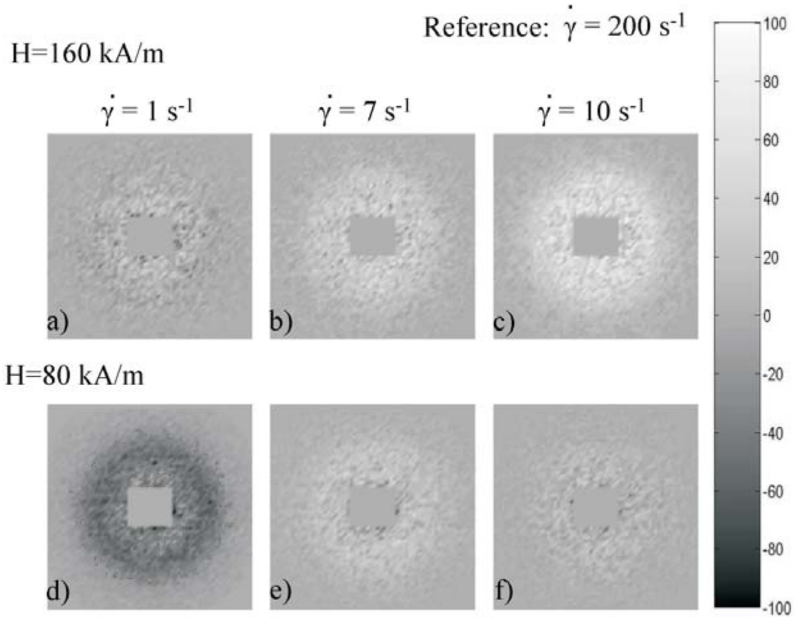

Fig. 3. Scattering patterns for APG 513A.

size as those in a single particle system, the difference between this situation and the reference scattering pattern is merely given by a lower concentration of the scattering centres, due to the structure formation in the ferrofluid sample. With increasing the shear rate (Fig. 3b, c), the deviation of the chains from the magnetic field direction becomes larger and therefore their projection increases, leading to a change in the difference scattering pattern. For lower magnetic field strengths and low shear rate (Fig. 3d) the influence of the magnetic torque on the chains as well as of the mechanic one that diverts the chains from the field direction will change if compared to Fig. 3a. The chains formed are shorter but their alignment with the field is weaker than for the strong field situation. Therefore, the difference between the scattering patterns and the reference becomes non zero (Fig. 3d). An increase of the shear rate (Fig. 3e) will force a higher deviation of the chains but also their break up. Thus, the pattern changes again and the difference to the reference vanishes with increasing the shear rate (Fig. 3e, f).

In the case of the fluid with low magnetoviscous effect, TOA, no significant structure formation occurs and thus the difference scattering patterns show no modification with the shear rate (Fig. 4). The slight modification of the difference scattering patterns with the magnetic field strength that can be observed is most likely only an orientation effect of the magnetic moments of single particles.

For a low magnetic field strength the magnetic moments of the particles are randomly distributed in the ferrofluid sample. For a higher magnetic field strength, the magnetic moments of the single particles tend to be orientated closer to the direction of the magnetic field. This orientation process induces a change of the resulting magnetic moment of the sample an, therefore, a marginal modification of the difference scattering patterns with the magnetic field strength can be observed.

It was shown above, qualitatively, that a direct connection between the strength of the magnetoviscous effect and the structure formation in ferrofluids which can be observed using small angle neutron scattering can be established.

These results were confirmed by recent experiments performed for cobalt based ferrofluids. Here the size of the particles as well as the concentration of

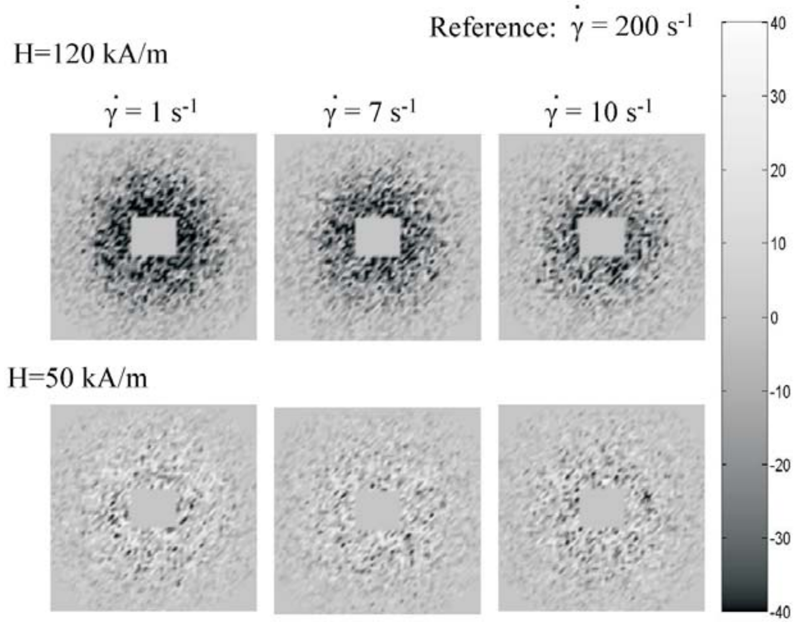

Fig. 4. Scattering patterns for TOA.

the magnetic material was changed, in order to facilitate formation of various structures and therefore different magnitude of the magnetoviscous effect. Due to the high magnetic moments of the Co particles, a strong dependency of the scattering patterns on the magnetic field strength and shear rate was observed. Additional indications of chain formation as well as of deviation of chains due to the shear flow were obtained by varying the direction of the applied magnetic field relative to the neutron beam.

The presentation will contain a brief introduction of the experimental setup followed by experimental results from rheological as well as SANS investigations for different ferrofluids. The information obtained from the data evaluation will be compared with numerical simulations [6, 7].

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