Structure formation in cobalt-based ferrofluids, investigated by small angle neutron scattering

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Previous rheological experiments, performed for magnetite as well as for cobalt based ferrofluids, have evidenced strong magnetoviscous effects that could be correlated with structural changes within the fluid samples in shear flow and under the influence of a magnetic field.
In the last years, a lot of experimental and theoretical studies have been performed in order to explain the microscopic mechanisms of the magnetoviscous effect. As a result of these efforts a model, based on numerical and experimental data, has been established, which is able to explain the observed behavior [1]. Chain formation of magnetic particles with strong particle-particle interaction as well as structure destruction by means of shear influence are the essential processes for the understanding of the magnetoviscous phenomena.

Experimental setup

In order to observe the chain formation process, a special rheometer has been designed [2]. The rheometer allows rheological measurements as well as the investigation of the microstructure of ferrofluids using small angle neutron scattering (SANS), performed under the same experimental conditions.
For the SANS experiments two configurations, one with the magnetic field oriented parallel to the neutron beam and the other with 10 degree between the magnetic field direction and the neutron beam, have been used. Both, rheological and SANS investigation have been carried out for magnetic fields varied between 0 and 160 kA/m, applied perpendicular to the vorticity of the flow. The shear rates have been varied within the range from 0 up to 200 s^{-1}.

Experimental results

Scattering as well as rheological experiments were performed for three different cobalt based ferrofluids, manufactured by N. Matoussevitch and H. Bönnemann (Forschungszentrum Karlsruhe) [3]. The concentration of the magnetic material was varied in the range from 0.35 vol.% up to 3.25 vol.% in order to observe its influence on the magnitude of the magnetoviscous effect.
To eliminate the influence of the carrier liquid, surfactant and small particles to the scattering, a reference data set, $\dot{\gamma}=0\ s^{-1}$, has been subtracted from the scattering patterns obtained for various shear rates.
Depending on the parameters of the investigated ferrofluids, strong dependency of the resulting difference scattering patterns on the magnetic field strength and shear rate were observed. These results are in a good agreement with the rheological data and with the chain formation model developed to explain the mechanism of the magnetoviscous effect.
According to the qualitative model presented in [2], for low magnetic field strengths the chains formed are short and hence their projection on the detector is also small. An anisotropy of the difference scattering pictures appears only for high deviations of the chains. For stronger magnetic fields, the chains become longer; their projection, which can be detected by neutrons, increases with increasing shear rate. The scattered intensity as well as the anisotropy grows relative to the reference.

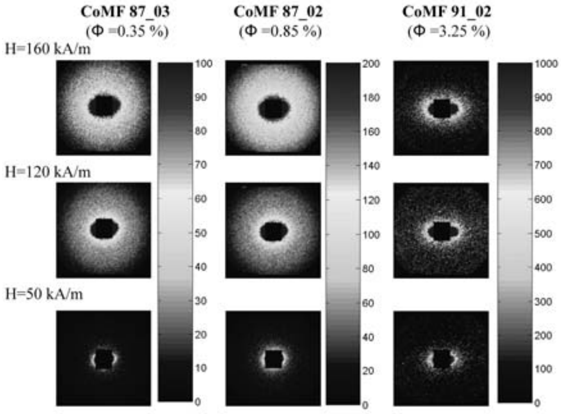

Figure 1: Comparison between the difference scattering patterns obtained at shear rate $\dot{\gamma} = 200\ s^{-1}$. The three cobalt based ferrofluids have the same mean particle diameter (10 nm) but different concentrations of the magnetic material. The magnetic field is orientated parallel to the neutron beam.

This behaviour can be observed in both cases for the magnetic field parallel to the neutron beam (fig.1) and for 10° between the magnetic field and neutron beam (fig. 2).

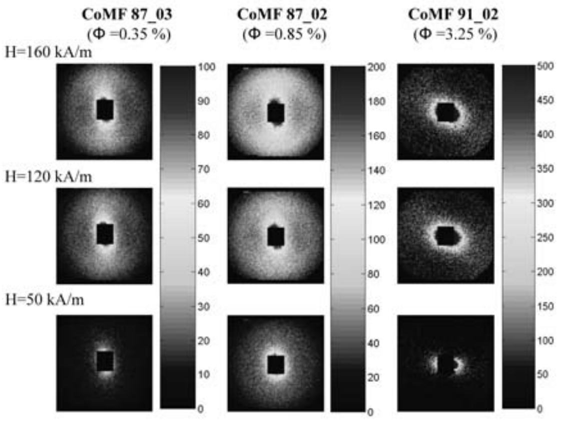

Figure 2: Comparison between the difference scattering patterns for the case with 10 degree between the magnetic field and the neutron beam and for shear rate $\dot{\gamma} = 200\ s^{-1}$.

In the first case (fig. 1), the anisotropy appears only due to the deviation of the chain-like structures under the combined effects of shear rate and magnetic field. In the second case (fig. 2), by rotating the rheometer in the horizontal plane (10° between the magnetic field and neutron beam), even in the static case a vertical anisotropy of the scattering patterns can be observed in the presence of a magnetic field, indicating the existence of chains. Furthermore, applying a shear rate, the chains are deviated from the direction of the magnetic field. Thus a deviation of the anisotropy from the vertical direction can be noticed (fig. 2).
The influence of the concentration of the cobalt particles on the structure formation within the ferrofluid samples, i.e. on the magnitude of the magnetoviscous effect, will be discussed. Thus, the SANS results will be compared with the rheological data obtained for the investigated ferrofluids. Additionally, the experimental results will be compared with two different theoretical approaches, the chain formation theory [4] as well as molecular dynamic simulations [5].

Acknowledgments

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