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Journal of Magnetism and Magnetic Materials 289 (2005) 303–306
Journal of magnetism and magnetic materials
www.elsevier.com/locate/jmmm

Microstructure and rheology of ferrofluids

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Available online 7 December 2004

Abstract

Experimental studies made for different ferrofluid samples under shear flow have shown that increasing the magnetic field strength yields an increase of the fluids viscosity, the so-called magnetoviscous effect, while increasing shear rate leads to a decrease of the magnitude of the viscosity (shear thinning). The change of the viscosity with magnetic field strength is theoretically explained as an effect of chain-like structure formation in ferrofluids whereas its magnitude depends on the particle–particle interaction. Both effects, the shear thinning and the magnetoviscous effect, can therefore be related to the microstructure and microstructure dynamics of ferrofluids. Using a specially designed rheometer, ferrofluids having different magnitude of the magnetoviscous effect were investigated by small-angle neutron scattering. Correlated to the structure formation in the fluid, the scattered intensity shows a variation with the magnetic field and shear rate only in the case of the fluids with a high magnetoviscous effect. The presented results show that there is a strong connection between the rheological behaviour of ferrofluids and their microstructure.

PACS: 75.50.Mn; 83.60.Fg; 83.60.Rs

Keywords: Ferrofluids; Magnetoviscous effect; Microstructure; Small-angle neutron scattering; Scattering patterns

1. Introduction

Rheological investigations of field-induced changes of the viscosity of ferrofluids under 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]. Therefore, the effect can be explained with chain-like structures formed in the fluids and 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 shear rate the relative change of viscosity decreases (shear thinning). In this case the mechanical torque becomes dominant and the chains are broken.

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0304-8853/$ - see front matter © 2004 Published by Elsevier B.V.
doi:10.1016/j.jmmm.2004.11.086

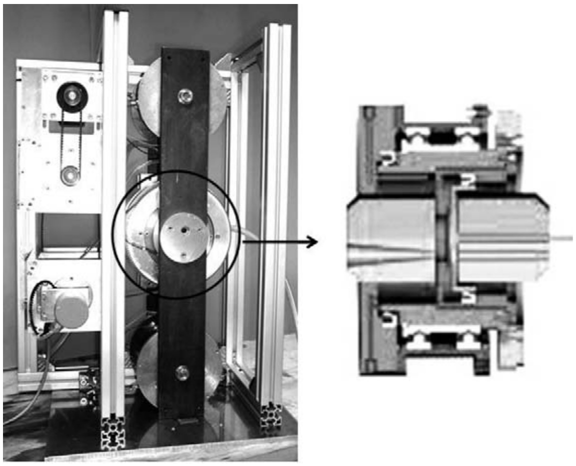

Fig. 1. Photo of the experimental set-up for SANS.

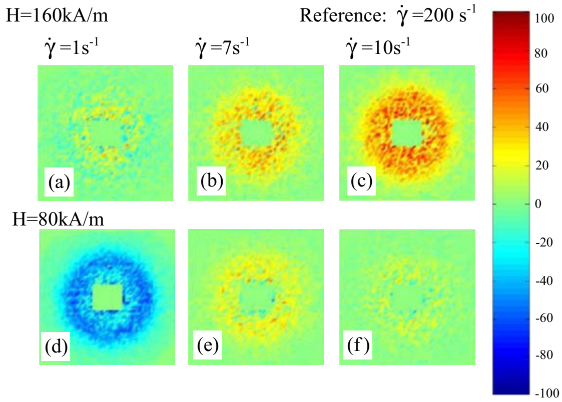

Fig. 2. Scattering patterns for APG 513A.

To prove the correlation between the macroscopical behaviour and the microstructure, measurements using the small-angle neutron scattering technique (SANS) have been performed. For this procedure a special rheometer was designed (see Fig. 1). The SANS experiments were carried out in a magnetic field range from 0 to 160 kA/m, directed parallel to the neutron beam and shear rates were varied within the range from 0 up to 200 s^{-1}.

The most important feature of the rheometer is that it allows SANS measurements in the same geometry and under the same conditions, i.e. magnetic field strengths and shear rates, as the rheological experiments. Rheological as well as SANS measurements with ferrofluids having different dipole–dipole interaction between the magnetic particles and therefore different magnitudes of the magnetoviscous effect have been carried out.

2. Experimental results

Three ferrofluids, Ferrotec APG513A (7.2 vol% magnetite in synthetic ester, dynamic viscosity $\eta_{(T=20^\circ)} = 128$ mPa), TOA UT (7.2 vol% magnetite in petroleum, $\eta_{(T=20)} = 20$ mPa) prepared by D. Bica at UT Timisoara [4] and Co85 (0.3 vol% Co in L9, $\eta_{(T=20)} = 65$ mPa) prepared by N. Matoussevitch at MPI Mühlheim [5–7] were investigated.

To obtain information only about modifications of the structure, one has to eliminate the contribution of the small particles, surfactant and carrier liquid to the scattering. The high shear rate situation, $\dot{\gamma} = 200\ s^{-1}$, when no change of the viscosity with the magnetic field appears was considered as a reference for the magnetite-based ferrofluids (Fig. 2); all chains are broken and the particles are homogeneously distributed in the fluid. By subtracting the reference from the scattering patterns obtained for various shear rates, the results give information only about modifications of the structure.

For a high magnetic field strength and a low shear rate (Fig. 2a), chains formed in fluid are long but orientated close to the field direction. Thus, the situation is similar to the static case and therefore the difference pattern is almost zero. With increasing shear rate (Fig. 2b and c), the deviation of the chains from the magnetic field direction becomes larger and therefore their projection, as seen by neutrons, increases, leading to a strong change in the difference scattering pattern. For lower magnetic field strengths and low shear rate (Fig. 2d), 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 modify compared to Fig. 2a. The chains formed are shorter but their alignment with the field is weaker. Therefore, the difference between the scattering patterns and the reference becomes non-zero (Fig. 2d). An increase of the shear rate (Fig. 2e) will force a higher deviation of the chains but also their break up. Therefore, the pattern changes again and the difference to the reference vanishes with increasing $\dot{\gamma}$ (Fig. 2e and f).

In the case of the fluid with very low magnetoviscous effect, TOA, no structure formation occurs and thus the difference scattering patterns show no modification with shear rate. However, a variation with magnetic field strength can be observed (see Fig. 3); with increasing magnetic field strength, the magnetic moments of the particles are oriented closer to the field direction, inducing an additional contribution of the magnetic scattering.

The cobalt-based ferrofluid shows a high magnetoviscous effect, comparable to the one for the sample APG513A. Rheological investigations have shown that the magnetoviscous effect is still present even for high shear rates. Thus, the situation without shear flow has been considered as a new reference. The obtained scattering patterns have shown an extremely strong