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Study of a magnetorheological fluid submitted to a uniform magnetic field

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Abstract. In this work, the rheological and hyperfine properties of a magnetorheological fluid (MRF) under the action of a uniform external magnetic field are analysed. Powders of native mineral magnetite of micrometric particle size, after a pulverization process, form the solute of these fluids. The sizes of these samples are selected by sieving in order to obtain sizes of around 20µm and 45µm. The powders are characterized by means of Mössbauer spectroscopy to analyse their stoichiometry giving rise to a non-stoichiometric magnetite $Fe_{2.96}O_4$ in addition to a hematite component. Result of viscosity and shear stress in the low-speed regime were analysed using the Hershel Buckley method. In particular, the case of surface tension it decreases with the application of a uniform magnetic flux density, which is understood in terms of a phase separation due to the formation of mesoscopic structures, thus decreasing the cohesion force and increasing the adhesion force.

1. Introduction

Magnetic fluids or magnetic colloids are two-phase systems formed by a liquid (carrier) and a solid phase (ferromagnetic or ferrimagnetic powder), which act as dynamic fluids and they can be controlled by an external magnetic flux density. Magnetic fluids can be classified in ferrofluids (size nm), magnetorheological (MRF) fluids (µm size) and composite magnetic fluids. The solid or dust particles are coated by a tensoactive surfactant adhered to the liquid-solid interface to mediate the interaction between these phases by avoiding agglomeration by electrostatic interaction. On the other hand, thermal agitation holds particles in suspension by steric repulsion. Magnetic fluids form a new class of magnetic materials whose novelty lies in the way as they behave changing their properties under the influence of a magnetic fields without losing the character of fluid, changing rheological properties such as local density, viscosity, stress and surface tension.

Magnetic fluids are materials of paramount importance due to their potential for technological applications. They are used in impact (suspensions) or vibration (seismic) suspension systems [1], in optics for lens polishing or for improving lens capability [2], in microelectromechanical (MEMs) technology [3] and for mineral cleaning [4] among others.

In particular, magnetoviscosity and shear stress properties of MRF as well as the formation of mesoscopic structures under an external applied magnetic field is still a subject of interest and debate upon which we focus our attention in this work.

2. Experimental

Native magnetite was grinded up in a vibrating cylinder mill at 140rpm after which two set samples of sizes around 20µm labelled as T5 and 45µm labelled as T2 respectively, were obtained by sieving.

These values were then confirmed by applying statistical methods to micrographs taken by optical microscopy [5].

The magnetite powders were then covered with oleic acid and dissolved in SAE oil in percentages of 20%-30%-50% and 15%-22.5%-62.5% respectively. Mixtures were then mechanical stirred in a thermal bath at 56°C during periods of 0.5, 1, 1.5 and 2 hours.

Sample were characterized by means of room temperature Fe^{57} Mössbauer spectroscopy for stoichiometric purposes by taking into account spectral areas of the tetrahedral (A) and octahedral (B) crystallographic sites of magnetite. Surface tension was characterized by measuring the surface breaking stress made by an aluminium ring and viscosity was measured in a BrokField 115 viscometer in parallel plate mode. The measurements in the viscometer were established under two different modes. First, by fixing the value of the magnetic flux density and varying the speed of the spindle, and second by fixing the speed of spindle and varying the magnetic flux density.

3. Results and discussion

After fitting the room temperature Mössbauer spectrum of the native sample, two components, one ascribed to hematite (11%) and the other one to magnetite (89%), were evidenced as can be observed in Figure 1. As concerns to the magnetite component, the corresponding $Fe^{2.5+}$ y Fe^{3+} spectral areas allow an estimation of the degree of stoichiometry via the oxidation parameter (x) in the framework of a model of vacancies distributed in either tetrahedral or octahedral sites.
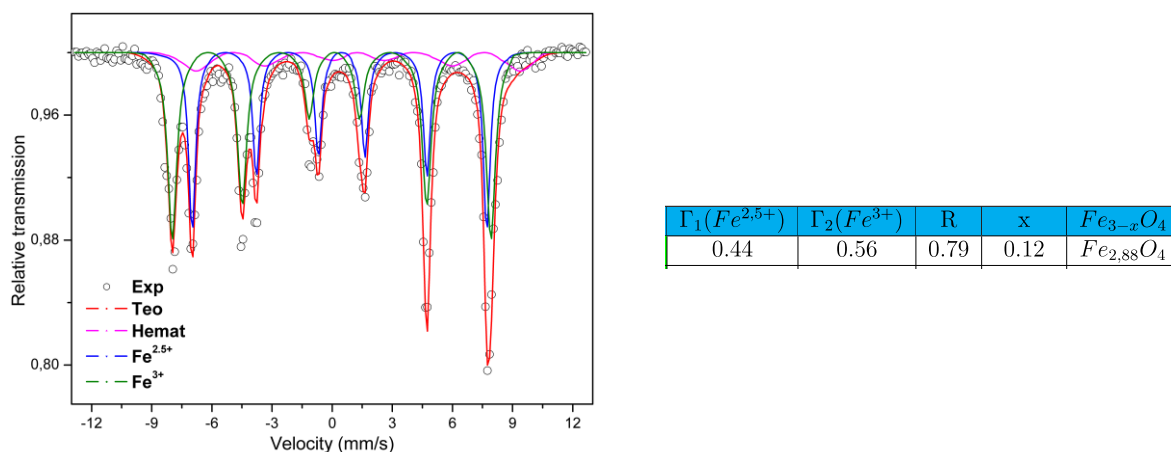

$\Gamma_1(Fe^{2,5+})$	$\Gamma_2(Fe^{3+})$	R	x	$Fe_{3-x}O_4$
0.44	0.56	0.79	0.12	$Fe_{2,88}O_4$

Figure 1. Room temperature Fe^{57} Mössbauer spectrum with its respective fit. The corresponding line widths (□) of tetrahedral and octahedral sites, the ratio of the spectral areas (*R*) and oxidation (*x*) parameters are included.

To do that, we employ the following equation [6], from which the oxidation parameter *x* was extracted and where the ratio $R = A(Fe_B^{2.5+})/A(Fe_A^{3+})$, obtained from the fitting process, was used,

$$R(\phi_B, x) = \frac{2-6x}{1.06(1-(1-\phi_B)x)+(6-\phi_B)x} \tag{1}$$

In equation (1), ϕ_B represents the fractional concentration of vacancies in *B* sites, which ranges between 0 and 1. However, as it has been already shown [6], the oxidation parameter *x* is practically insensitive to ϕ_B and differences are only noticed within a 0.8% of precision. In our case, the best estimate for *x* is 0.12 giving rise to a non-stoichiometric magnetite with formula $Fe_{2.88}O_4$. The effects of the hematite on the native sample is also reflected in the magnetic properties, more concretely, coercive forces and saturation values are given by: $Hc_{T2}=132.75Oe$, $Hc_{T5}=144.5Oe$, $Ms_{T2}=81.59emu/g$, $Ms_{T5}=77.53emu/g$ [5].

To analyse the rheological properties, it must be stressed that the viscosity of the MRF is established according to the conditions of laminar flow and its value must be in principle independent of the method used to measure it. However, the rheological information implies using a methodology that depends in practice upon some variables such as the container and spindle. The corresponding results are shown in Figure 2 were a sweep of low shear speeds by instrumentals limitations was considered.

The behaviour of the viscosity is ruled by the magnetic properties of the hydrodynamic volume of the magnetite, showing a special rearrangement or rotation of its magnetic moments to become aligned according to the magnetic flux density consistent with the Hershel-Buckley model as endorsed by the linear fitting shown in Figure 2. In this way, more resistance to the laminar movement is expected.
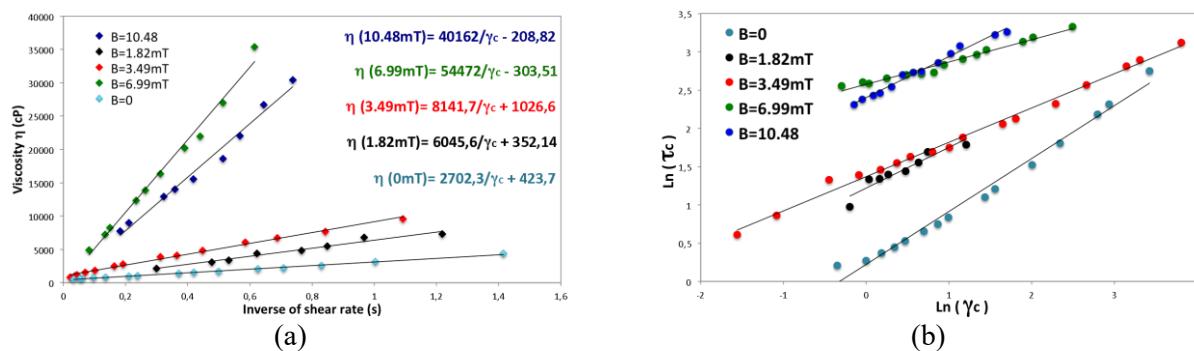

Figure 2. Behaviour of viscosity (a) and shear stress (b) for different values of the magnetic flux density and shear rate for percentages 20% of magnetite with size T2.

From Figure 2, η is the viscosity, γ_c shear rate, τ_c shear stress the slopes represent the effort required to move the MRF at each speed and the cut-off point indicates the limit of Newtonian behaviour. As is observed, slopes increase with the magnetic flux density until a critical value of the field of B=6.99mT, from which phase the separation occurs by gravitational and magnetic effects due to the formation of mesoscopic structures, so the information recorded does not belong to a uniform MRF. The shear stress curves are given in the Figure 3, which are in agreement with those obtained by J. M. Linke and S. Odenbach [7].
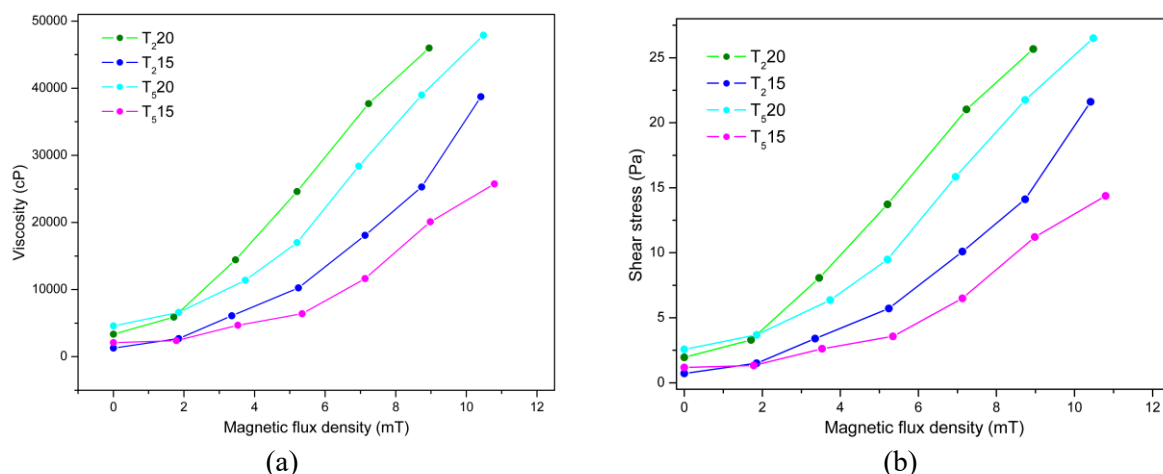

Figure 3. Dependence of viscosity (a) and shear stress (b) with the magnetic flux density at a constant shear rate and for two different percentages of magnetite (15% and 20%).

On the other hand, by varying the magnetic flux density and fixing the speed of the spindle at 2rpm we obtain the results shown in Figure 3. As can be observed, magnetoviscosity is evidenced through

the remarkable variations of viscosity as the magnetic flux density increases, which is attributed to the formation mesoscopic structures. Our results are consistent with those found by E. V. Korobko, *et. al.* [8] for shear stress and R. P. Pant, *et. al.* [9] for viscosity.

The increase of viscosity and shear stress with magnetic flux density can be explained as due to the long-range magnetic dipole interaction between agglomerates causing a spatial rearrangement of solute in liquid carrier. Moreover, the magnetic dipole moment μ of the agglomerate is proportional to the hydrodynamic volume given by $\mu = V\chi B/\mu_0$, where V is the hydrodynamic volume, χ the magnetic susceptibility and μ_0 magnetic permeability [9,10].

Rotation and rearrangement of the magnetic dipole moments are ruled by the surface functionality provided by the electric character of the oleic acid layer around the aggregate; which can in turn gives rise to some preferred orientations of the magnetic moments and changes in surface tension T_σ.

The effects of surface tension allow us to consider whether the MRF can be used in cleaning native minerals that may be with unwanted material on the surface of others like as coal [4]. The results of the surface tension measurements are shown in Figure 4.
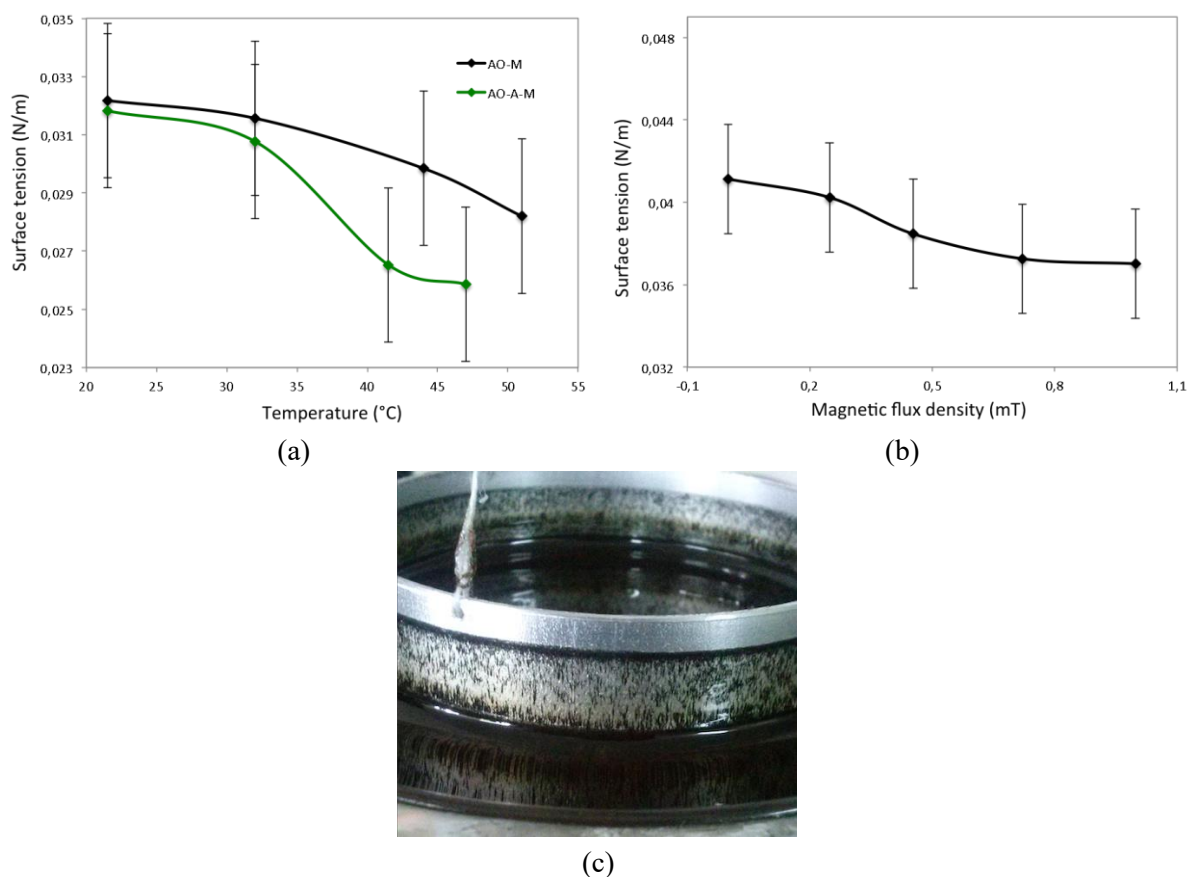

(a)

(b)

(c)

Figure 4. Behavior of surface tension as function of temperature for the liquid residue of oleic acid (OA) – magnetite (M) and oleic acid (OA) – oil (O) – magnetite (M) (a), and dependence with the magnetic flux density (b). At the bottom the formation of mesoscopic structures due to the field is visualized for percentage 20% of magnetite with size T5 (c).

Figure 4 shows also the formation of mesoscopic columnar structures and of fibrous-type for which a magnetostatic interaction between them, as a consequence of the radial asymmetry and the diameters involved, is expected [10]. Additionally, magnetite powders are not physically bound to each other due steric repulsion as a consequence of the oleic acid molecules, which are coordinated and attached of the Fe atoms of the bi-dentate surface formed by covalent oxygen bonds. Besides, the oxidation

parameter favours bonding by increasing adhesion to the molecule. For concentrations of oleic acid higher than the approximate equivalent of two monolayers there is an appreciable steric repulsion as it has been reported elsewhere [10-12].

4. Conclusions

Results of the rheological properties of the MRF show characteristics that are strongly dependent on the concentration and size of magnetite, temperature and the magnetic flux density. This MRF was characterized from its nature of micrometric sized aggregates and non-uniform geometry, which exhibit a pseudoplastic behaviour consistent with the Herschey Bulkley model.

The study of the MRF, gives information of our understanding about how this fluid reacts under certain circumstances or how to control some reaction, as it would be the case of damping systems by effects of instability by an applied field; or in transport systems where magneto-viscosity play an importance role.

It can be established also that the effective surface of the magnetic powders increases with size reduction allowing a better colloidal stability by lowering the gravitational energy $E_g = \Delta\rho V_{T_5} gL$ as compared to the thermal one $E_T = kT$ [10].

Finally, in MRF, its knowledge about the rheological behaviour under an applied magnetic flux density is currently incomplete and models of constitutive equations have not been provided still in general form [13].

Acknowledgements

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