CHARACTERIZATION OF THE PHYSICAL PARAMETERS IN A PROCESS OF MAGNETIC SEPARATION

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Abstract *In this work, a process of separation of oil from water with the use of magnetic microparticles has been investigated. In a first step, the motion of a drop composed by oil and magnetic microparticles is characterized by a scaling analysis. It is verified that the drift velocity of a magnetic drop scales with the square of the applied magnetic field when viscous forces dominate inertia forces. Experiments have also been carried out and the scalings verified. This was achieved by recording the motion of the drop along the direction of the applied field by using a digital video-camera, with the instantaneous velocity of the drop being determined by an image analysis technic. In a second step, a simple model is developed in order to predict the volume rate of oil that would be separated by magnetic action on the polarised particles of a magnetic fluid. A possible application of such study is on the remediation technology addressed to oil spills in natural environments.*

Key words: *magnetic emulsions, magnetic fluid, magnetic pressure coefficient, magnetic separation*

1. Introduction

Colloidal suspensions of fine particles of solid ferromagnetic material in a carrier liquid are known as magnetic fluids or ferrofluids (Kamiyama & Koike, 1992). Ferrohydrodynamics is concerned with the study of flows of these complex fluids. The motion of such fluids is strongly affected by the presence of an applied magnetic field due to an extra force acting on the fluid, the Kelvin force, associated to the bulk magnetization of the fluid. What differs ferrohydrodynamics from magnetohydrodynamics and electrohydrodynamics is the full absence of electric currents on the magnetic fluid, and the stronger strength of the forces acting on the flow.

In early stages of ferrohydrodynamics, the major motivation for the study of magnetic fluids was the possibility of controlling the motion of the fluid remotely, using magnetic fields. However, as more studies were carried on this subject, more interesting features related to these fluids were discovered. Zahn (1990), Shliomis & Morozov (1994) and, more recently, Felderhof (2001) have studied the flow of a magnetic fluid in an oscillating magnetic field in order to characterize the viscosity of a magnetic fluid as it flows. For steady magnetic fields or for fields with low frequencies of oscillation, the volume rate of the flow of a magnetic fluid is decreased, what can be associated to an increase of the viscosity of the magnetic fluid. On the other hand, if the frequency of the applied magnetic field is sufficiently high, the flow rate increases. This phenomenon is also called the magnetic pumping. The use of magnetic particles to stabilize high Reynolds number suspensions has also revealed to be effective. Several related works have been carried out in recent years, e.g. the experimental work of Hristov (1996) and the theoretical work of Sobral & Cunha (2003) on the stability of polarised fluidized beds and the work on collapsing bubbles in a magnetic fluid by Cunha et al. (2002). Rosensweig (1997) discuss a wide range of applications for magnetic fluids.

It should be noted, however, that there are still some open problems in ferrohydrodynamics, mainly those concerning the proposition of an evolution equation to the magnetization of a magnetic fluid. In fact, there is not enough knowledge on the microscopic behaviour of the magnetic particles that can support a definitive closed equation for the magnetization. In general, this equation is proposed based on the information available and, consequently, is subject to criticism based on individual feelings. A clear example of this is the recent work of Felderhof (2001), who studied three different equations for the magnetization, observing quite different behaviours concerning each of the equations on the prediction of magnetic pumping of magnetic fluids.

In this work, the focus is turned on a problem of magnetic separation, more specifically on the problem of separating oil from water. This has a major importance in petroleum industry, since when an accident occurs either during production, transportation or storage of oil, immediate measures must be put into practice to avoid major damages to the local environment. Under this perspective, the separation of oil form water using magnetic particles seems to be a promising method.

Investigations on the response of magnetic fluids to the applied field were carried out in this work, in order to provide some insight to evaluate the efficiency of magnetic separators. Under this perspective, we propose an estimation of the drift velocity for a drop of magnetic particles in a magnetic field by scaling arguments based on the governing equations of ferrohydrodynamics (Rosensweig, 1997). The relation between the applied field and the drift velocity for a viscous regime was validated by simple experiments carried out with a drop of magnetic fluid, composed of oil and magnetic microparticles, suspended in pure water. It was also investigated the magnetic induced deformation of the drop during its motion. It was verified that the drop changes its shape due to the local gradients of magnetic field which leads the drop to move faster than predicted by our theory. We develop a first order theory to predict the linear regime of deformation of the drop as a function of the applied field. A similar analysis, which evaluated the effect of the superficial stress on non-magnetic drops undergoing shear flows, was carried out by Taylor (1934).

Finally, we present a simple first order model for calculating the volume rate of magnetic fluids in a pipe flow as a function of the applied field and the magnetic properties of the ferromagnetic particles.

2. Governing Equations of Ferrohydrodynamics

In principle, all magnetic phenomena that arise in nature, what includes those related to ferrohydrodynamics, are described by the four Maxwell's equations (Rosensweig, 1997):

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}, \quad \nabla \cdot \mathbf{D_d} = \rho_q, \quad \nabla \times \mathbf{H} = \mathbf{J_f} + \frac{\partial \mathbf{D_d}}{\partial t}, \text{ and } \nabla \cdot \mathbf{B} = 0. \tag{1}$$

The first equation in eq.(1) is called the Faraday's law of induction and relates the electrical field $\mathbf{E}$ with the magnetic induction $\mathbf{B}$. The second equation in eq.(1) is the Gauss' law for electricity and indicates that the local flux of the displacement electric field $\mathbf{D_d}$ equals the local free electrical charge density ρ_q. The Ampère's law of magnetism with the Maxwell's corrections is presented as the third equation in eq.(1) and states that a magnetic field $\mathbf{H}$ can be either originated by a free current density $\mathbf{J_f}$ or by a time variation of the displacement electric field. The inexistence of magnetic monopoles in nature is translated by the Gauss' law of magnetism, the fourth equation in eq.(1), indicating that the magnetic induction is a solenoidal field. Full details concerning the definitions of the quantities appearing in eq.(1) can be found in Rosensweig (1997).

In most ferrohydrodynamics problems, however, there is no need to consider the four equations presented in eq.(1). Since the fluid is ferromagnetically responsive and no electrical field is applied, the electric quantities in eq.(1) are either null or negligible. This fact characterizes a magnetostatic regime and allows the magnetic problem arisen in ferrohydrodynamics to be described by only two simplified equations of the Maxwell's equations, eq.(1):

$$\nabla \cdot \mathbf{B} = 0, \tag{2}$$

and

$$\nabla \times \mathbf{H} = \mathbf{0}. \tag{3}$$

The magnetic inductance and the magnetic field intensity are related, in the vacuum, by

$$\mathbf{B}_o = \mu_o \mathbf{H}, \tag{4}$$

where $\mu_o = 4\pi \cdot 10^{-7} H/m$ is the vacuum magnetic permeability. In the presence of a magnetized material, eq.(4) should be written as

$$\mathbf{B} = \mu_0 (\mathbf{H} + \mathbf{M}). \tag{5}$$

The vector quantity $\mathbf{M}$ is called the magnetization of the material and represents the continuum effect of the intrinsic state of polarisation of the material in the presence of a magnetic field. It should be stressed out that the magnetization is not purely a property associated with the atoms or ions that compose the material, but also with the interactions among them. The strength of these interactions can define at least two different kinds of ferromagnetic materials (Rosensweig, 1997): soft materials, which are associated to very weak interactions, and non-linear materials, in which interactions are so strong that the magnetic dipole moment of the particles can be affected by the neighbouring magnetic particles. The nature of the these interactions in a magnetic fluid should be known since it must be taken into account when models are proposed for the local magnetization $\mathbf{M}$ of a magnetic fluid undergoing a flow. In this work, we focus on dilute magnetic fluid, that can be considered a soft material.

Besides the magnetic equations, hydrodynamic balance equations are needed for this hydrodynamic-magnetic coupled problem. We consider the continuity equation for incompressible flow

$$\nabla \cdot \mathbf{u} = 0, \tag{6}$$

where $\mathbf{u}$ represents the eulerian velocity field, and the Cauchy's equation for a continuum media

$$\rho \left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right) = \nabla \cdot \boldsymbol{\sigma} + \rho \mathbf{g}. \tag{7}$$

In this equations, ρ denotes the fluid density, $\boldsymbol{\sigma}$ denotes the stress tensor of the flow and $\mathbf{g}$ the gravity acceleration. Since this work deals with equivalent fluids that have magnetic properties, the stress tensor $\boldsymbol{\sigma}$ should be modified in order to consider magnetic effects on the flow. Thus, it can be written that

$$\boldsymbol{\sigma} = \boldsymbol{\sigma}^h + \boldsymbol{\sigma}^m, \tag{8}$$

where the part denoted by $\boldsymbol{\sigma}^h$ takes into account the hydrodynamic effects on the stress tensor and where $\boldsymbol{\sigma}^m$ accounts for the magnetic effects on the flow.

The usual Navier-Stokes linear stress tensor for an incompressible newtonian fluid is given by (e.g. Batchelor, 1967)

$$\boldsymbol{\sigma}^h = -p^*\mathbf{I} + 2\eta\mathbf{D}, \text{ with } \mathbf{D} = \frac{1}{2}\left(\nabla\mathbf{u} + \nabla\mathbf{u}^T\right). \tag{9}$$

In this equation, p^* is the mechanical pressure, η denotes the fluid viscosity and $\mathbf{D}$ is the rate of strain tensor, corresponding to the symmetric part of $\nabla\mathbf{u}$.

The magnetic contribution to $\boldsymbol{\sigma}$ comes from the Maxwell stress tensor, defined as

$$\boldsymbol{\sigma}^m = -p_m\mathbf{I} + \frac{1}{2}\left(\mathbf{BH} + \mathbf{HB}\right), \tag{10}$$

where the magnetic pressure of the applied field is defined as follows (Rosensweig, 1997):

$$p_m = \frac{1}{2}\mu_o\left(\mathbf{H}\cdot\mathbf{H}\right). \tag{11}$$

The Maxwell stress tensor is a symmetric tensor, since it is composed by an isotropic $-p_m\mathbf{I}$ and by the symmetric part of the tensor $\mathbf{BH}$. It should be noted that variations of magnetization with the specific volume of the magnetic fluid, usually called magnetostrictive effects (Kamiyama & Koike, 1992), are not important in dilute magnetic fluids and will not be considered in this work.

Taking the divergence of eqs. (9) and (10) and replacing the result in eq.(7), the linear momentum balance is finally written as

$$\rho\left(\frac{\partial\mathbf{u}}{\partial t} + \mathbf{u}\cdot\nabla\mathbf{u}\right) = -\nabla p + \eta\nabla^2\mathbf{u} + \mu_0\left[\mathbf{M}\cdot\nabla\mathbf{H} + \frac{1}{2}\mu_0\nabla\times\left(\mathbf{M}\times\mathbf{H}\right)\right], \tag{12}$$

where p denotes a modified pressure. What differs eq.(12) from the traditional Navier-Stokes equation is the Kelvin force term $\mu_0\mathbf{M}\cdot\nabla\mathbf{H}$ associated with magnetic field gradients and the magnetization of the fluid and the extra force term $\frac{1}{2}\mu_0\nabla\times(\mathbf{M}\times\mathbf{H})$ associated to the internal magnetic torques. The presence of the internal magnetic torques on the magnetic fluid, which are originated by the term $\mathbf{M}\times\mathbf{H}$, breakes the stress symmetry and imply that the angular momentum equation might also be considered. The equation for the angular momentum balance is written as:

$$\frac{\partial\mathbf{L}}{\partial t} + \mathbf{u}\cdot\nabla\mathbf{L} = \mu_0\mathbf{M}\times\mathbf{H} - \frac{1}{\tau_s}(\mathbf{L} - \mathcal{I}\boldsymbol{\Omega}) + \mathcal{D}\nabla^2\mathbf{L}. \tag{13}$$

The terms on the left hand side of this equation represent the change of angular momentum $\mathbf{L}$ that a fluid particle undergoes on its motion seen by an observer convected with the fluid. The first term on the right hand side is the magnetic torque due to the local magnetic field $\mathbf{H}$ acting on a fluid particle with magnetization $\mathbf{M}$. The second term states the deviation of the angular momentum from the intrinsic angular momentum $\mathcal{I}\boldsymbol{\Omega}$, where $\mathcal{I}$ denotes the moment of inertia of the particle and $\boldsymbol{\Omega} = \frac{1}{2}\nabla\times\mathbf{u}$ its angular velocity. This change is associated to the characteristic magnetization relaxation time of the particle τ_s. The last term indicates that there may also be a diffusion of angular momentum, with $\mathcal{D}$ being the brownian diffusion coefficient defined by the brownian relaxation time τ_b and a characteristic length scale L as $\mathcal{D} = L^2/\tau_b$. Precise estimatives of τ_b can be found in Rosensweig (1997), but it can be assumed, in general, $\tau_b \sim \mathcal{O}(10^{-7}s)$.

Now, a relaxation equation for the magnetization $\mathbf{M}$ is needed. In this work, the magnetization equation proposed by Shliomis (Shliomis & Morozov, 1994), valid for dilute magnetic fluids with small magnetization is used:

$$\frac{\partial\mathbf{M}}{\partial t} + \mathbf{u}\cdot\nabla\mathbf{M} = \boldsymbol{\omega}\times\mathbf{M} - \frac{1}{\tau_s}(\mathbf{M} - \mathbf{M^o}), \tag{14}$$

where $\boldsymbol{\omega} = \boldsymbol{\Omega} + \frac{1}{\xi}\mathbf{M}\times\mathbf{H}$ is the effective angular velocity of the particle and $\xi = \mathcal{I}/(\tau_s\mu_0)$ represents an angular viscosity associated to the extra resistance to the rotation of the magnetic particles due to the local magnetic field. The vector quantity $\mathbf{M^o}$ is the equilibrium magnetization for a quiescent magnetic fluid. For a dilute magnetic fluid, the equilibrium magnetization is collinear with the applied field $\mathbf{H_o}$, $\mathbf{M^o} = M^o\frac{\mathbf{H_o}}{H_o}$, where M^o can be determined by the expression (Rosensweig, 1997)

$$M^o = \phi M_d \mathcal{L}(\alpha), \quad \text{with} \quad \alpha = \Theta H, \quad \text{and} \quad \Theta = \frac{3\chi_o}{\phi M_d}. \tag{15}$$

In this equation ϕ represents the particle volume fraction, M_d denotes the bulk magnetization of the particles, $\mathcal{L}(\alpha) = cotgh(\alpha) - \alpha^{-1}$ is the Langevin function and $\chi_o = M/H_o$ is the magnetic susceptibility based on the applied field H_o. It should be noted that the parameter α, usually called the energy ratio parameter, is evaluated in terms of the local field magnitude H. The first term on the right hand side of eq.(14) represents the change in the magnetization of a particle due to the local rotation of the fluid and to the internal torques, whereas the second states for the deviation from the equilibrium magnetization for quiescent magnetic fluid.

2.1 Dimensionless equations and simplifications of the governing equations

Equations (12), (13) and (14) can be made dimensionless for appropriated scales. Turning back to eq.(12), a typical velocity scale U is used as velocity scale, L as the length scale and H_o, the intensity of the applied field, as a typical scale for magnetic quantities. Convective time and pressure scales are L/U and ρU^2, respectively. After few algebraic manipulations, the dimensionless form of eq.(12) is found to be

$$\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} = -\nabla p + \frac{1}{Re} \nabla^2 \mathbf{u} + C_{pm} \left[\mathbf{M} \cdot \nabla \mathbf{H} + \frac{1}{2} \nabla \times (\mathbf{M} \times \mathbf{H}) \right], \tag{16}$$

where all quantities in this equations are now dimensionless. The dimensionless physical parameters appearing in the above equation are the Reynolds number and the magnetic pressure coefficient, respectively

$$Re = \frac{\rho U L}{\eta} \quad \text{and} \quad C_{pm} = \frac{\mu_o H_o^2}{\rho U^2}. \tag{17}$$

The Reynolds number measures the relative intensity of the inertial and viscous mechanisms of momentum transport, whereas the magnetic pressure coefficient states the importance of the magnetic pressure compared to the dynamical pressure of the flow, i.e. magnetic force relative to inertial force. It is clear that in the asymptotic limit of $C_{pm} << 1$, eq.(16) reduces to the well known Navier-Stokes equation for an incompressible fluid.

For the angular momentum equation, eq.(13), a typical scale for $\mathbf{L}$ is found from its classical definition from lagrangean mechanics, $\mathbf{L} = \boldsymbol{x} \times \rho \boldsymbol{v}$, where $\boldsymbol{x}$ states for the relative position and $\boldsymbol{v}$ for the velocity of an arbitrary particle. Consequently, a typical scale for $\mathbf{L}$ is simply $\rho U L$. A scale for the moment of inertia may also be found from its definition and may be chosen to be ρL^2. At last, the typical scale for the angular velocity of the particles is defined as U/L, while the the scales for the other quantities that appear in eq.(13) are kept the same as for eq.(12). The resulting dimensionless equation for the angular momentum is written as:

$$\hat{\omega} \left(\frac{\partial \mathbf{L}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{L} \right) = C_{pm} \hat{\omega} (\mathbf{M} \times \mathbf{H}) - (\mathbf{L} - \mathcal{I} \mathbf{\Omega}) + \frac{1}{Pe_m} \nabla^2 \mathbf{L}. \tag{18}$$

Note that all quantities in eq.(18) are dimensionless. The two new dimensionless parameters in eq.(18) are the magnetic dimensionless frequency $\hat{\omega}$ and the magnetic rotational Péclet number Pe_m. They are defined as

$$\hat{\omega} = \frac{\tau_s}{L/U} \quad \text{and} \quad Pe_m = \frac{\tau_b}{\tau_s} = \frac{L^2}{\mathcal{D} \tau_s}, \tag{19}$$

respectively. The magnetic dimensionless frequency is defined as the ratio between the magnetization relaxation time scale and the convective time scale. Since the latter should be, in general, much larger than the former, then $\hat{\omega} << 1$, what indicates that no important changes on angular momentum of fluid particles are due to convective effects. The magnetic rotational Péclet number will be the central point for some future simplifications, since it states the relation between the brownian and the magnetization relaxation time scales and, consequently, defines how fast magnetic particles should orientate themselves with the local magnetic field.

Now, the dimensionless version of the relaxation equation for the magnetization, eq.(14) can be obtained by using the same scales defined in the previous equation, namely

$$\frac{\partial \mathbf{M}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{M} = \mathbf{\Omega} \times \mathbf{M} + C_{pm} \hat{\omega} \frac{(\mathbf{M} \times \mathbf{H}) \times \mathbf{M}}{\mathcal{I}} - \frac{1}{\hat{\omega}} (\mathbf{M} - \mathbf{M^o}). \tag{20}$$

Note again that all quantities in eq.(20) are dimensionless. All dimensionless parameters appearing in this equation are defined in eq.(19).

If $\tau_s << \tau_b$, the local orientation of the magnetic particles with the magnetic field is almost instantaneous and the brownian effects that can change the angular momentum are very slow. In this case, $Pe_m >> 1$ and two direct consequences are observed: the diffusion term in eq.(18) plays no effective role on the angular momentum balance, as well as time variations following the particles, represented by the term $\partial\mathbf{L}/\partial t + \mathbf{u}\cdot\nabla\mathbf{L}$, are very weak. This can be interpreted as a quasi-steady state, on the scale of the flow, for the angular momentum.

Dilute magnetic fluids behave as described above, since no strong interactions among neighbouring particles are present, what allows the local magnetization to be instantaneously orientated on the direction of the local magnetic field. In the remainder of this work, it shall be considered that local magnetization is collinear with the local magnetization, what implies that

$$\mathbf{M}\times\mathbf{H} = \mathbf{0}. \tag{21}$$

Equation (21) leads to important simplifications on the governing equations. In eq.(16) the force term associated to the internal torques should vanish and the Kelvin force term may be simplified from $\mathbf{M}\cdot\nabla\mathbf{H}$ to $M\nabla H$, where $M = |\mathbf{M}|$ and $H = |\mathbf{H}|$. This implies that eq.(12) should assume the following dimensionless final version:

$$\frac{\partial\mathbf{u}}{\partial t} + \mathbf{u}\cdot\nabla\mathbf{u} = -\nabla p + \frac{1}{Re_L}\nabla^2\mathbf{u} + C_{pm}M\nabla H. \tag{22}$$

Eq.(18) may also be simplified for the case $\hat{\omega} << 1$ and $Pe_m >> 1$. The angular momentum equation, eq.(18), is then reduced to

$$\mathbf{L} = \mathcal{I}\mathbf{\Omega}, \tag{23}$$

what indicates that the local angular momentum of the particles are associated only to their local angular velocity.

Similarly, eq.(20) may be simplified based upon the same physical arguments as used to the angular momentum equation. Thus, the magnetization time variations following the particles, $\partial\mathbf{M}/\partial t + \mathbf{u}\cdot\nabla\mathbf{M}$, can be neglected for the limit $\hat{\omega} << 1$. However, it should be noted that the term $\mathbf{\Omega}\times\mathbf{M}$ may become of the same order of magnitude of $(\mathbf{M}-\mathbf{M^o})/\hat{\omega}$, what means that both should be kept on the final version of this equation. Then,

$$\hat{\omega}\mathbf{\Omega}\times\mathbf{M} = \mathbf{M} - \mathbf{M^o}. \tag{24}$$

For flows in which the rotational effects are very weak, however, a more simplified, but still consistent, model would be simply $\mathbf{M} = \mathbf{M^o}$.

3. The Motion of a Magnetic Drop

The motion of a drop of oil and magnetic microparticles is investigated under the light of ferrohydrodynamics. Thus, the governing equations determined on the previous section are used as the starting point to understand phenomenologically the mechanics involved in this process.

Consider a circular drop of oil of radius R immersed in a fluid of density ρ and shear viscosity η, as illustrated in fig. 1(a). Suppose also that magnetic spherical microparticles of radius a are also present on the drop in a $\phi = V_p/V$ volume concentration, where V_p denotes the volume occupied by the particles and V the total volume of the drop. The resulting suspension is now considered a magnetic drop of an equivalent fluid and may have its velocity $\mathbf{u}$ affected by an external applied field $\mathbf{H}$. Fig. 1(b) presents a real laboratory picture of this scenario.

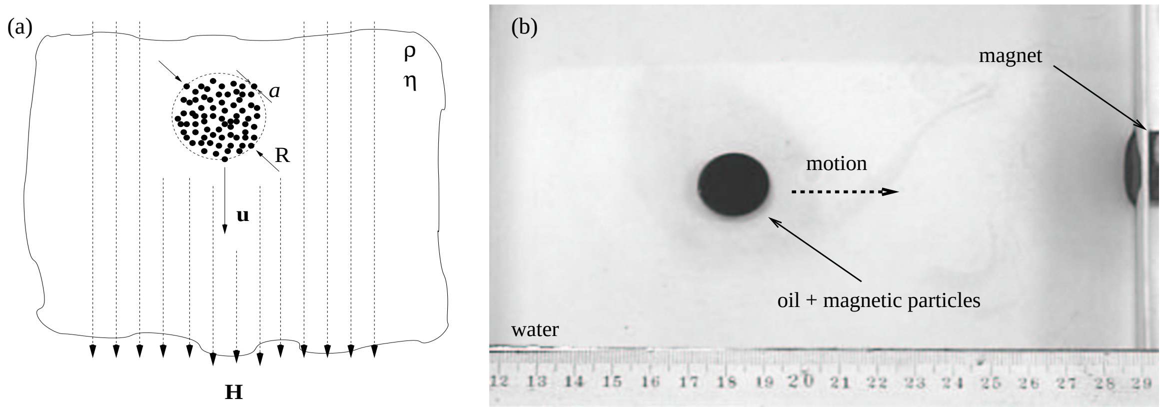

Figure 1: (a) Schematic of the problem of the capture of a drop composed of oil and magnetic microparticles suspended on water under the action of an external magnetic field. In (b), an image of the experimental setup.

3.1 Scaling arguments for the velocity of the drop

From eq.(12), one observes that there might be a regime of the above described flow where viscous forces may balance magnetic forces. This regime is significant for slow motions of a neutrally buoyant magnetic drop induced by a magnetic field. We shall propose typical scales for these forces and analyse the regime when viscous forces balance magnetic forces.

A typical scale for viscous force per unit of volume, from eq.(12), is given by

$$|\eta \nabla^2 \mathbf{u}| \sim \eta \frac{U}{R^2}. \tag{25}$$

In analogy, the scale for the magnetic force per unit of volume is obtained from the simplified Kelvin force term $\mu_o M \nabla H$ on eq.(12). Thus,

$$|\mu_o M \nabla H| \sim \mu_o M \frac{H}{R} \sim \mu_o \chi^s \phi \frac{H^2}{R}, \tag{26}$$

where a characteristic scale for the magnetization, based on the saturation susceptibility of the magnetic particles χ^s (Rosensweig,1997) is used, $M \sim \chi^s \phi H$.

When viscous force balance magnetic forces, it follows that the drift velocity U scales as

$$U \sim \frac{\mu_o}{\eta} \chi^s \phi R H^2. \tag{27}$$

Equation (27) predicts a drift velocity scaling with the square of the applied field and linearly with the saturation susceptibility and the volume fraction of the magnetic particles.

Now, a typical scale for the drift velocity is obtained from the ferrohydrodynamic Bernoulli equation (Rosensweig, 1997), say $U_c \sim H_o\sqrt{\mu_o/\rho}$. Therefore, the dimensionless scaling is written as:

$$\frac{U}{U_c} \sim Re_m \chi^s \phi \hat{H}^2, \tag{28}$$

where $\hat{H} = H/H_o$ is the dimensionless absolute value of the applied magnetic field and the parameter Re_m, the magnetic Reynolds number, that compares viscous effects with magnetic effects, is defined as

$$Re_m = \frac{\sqrt{\rho \mu_o} R H_o}{\eta}. \tag{29}$$

Note that the aggregate radius R may be written in terms of the particle volume fraction ϕ and of the particle radius a. From the definition of ϕ, it comes that $R \sim a\sqrt[3]{N/\phi}$, where N denotes the number of particles on the aggregate.

In order to validate the predicted scalings, a simple experiment was carried out. Using the set up shown in fig. 1(b), the motion of a magnetic drop was recorded with a digital camera with acquisition frequency of 30Hz. By means of image analysis techniques, the instantaneous velocity of the aggregate was determined, as well as the shape of the drop as the time evolve. In addition, the magnetic field, created by a permanent magnet, was measured with a digital gaussmeter with resolution of $10^{-3} mT$. The self-consistent magnetic field generated by the magnetization of the magnetic fluid is much smaller than the applied magnetic field and may be neglected. This is usually valid for materials with small magnetic susceptibility (Felderhof, 2001).

The magnetic particles used on the experiment were magnetite nanoparticles immersed in styrene-divinylbenzene spherical micron-sized polymer templates. More details on the chemical preparation and physical characterization of such particles can be found in Rabelo et al. (2001) and Morais et al. (2001), respectively.

The experiments were carried out at conditions that $Re_m = 760$ and under the action of a magnetic field generated by a permanent magnet with profile shown in fig. 2(a). Figure 2(b) shows the dimensionless velocity of the magnetic drop as a function of the magnetic parameter $Re_m \hat{H}^2$. It can be seen that the scaling proposed for the drift velocity is in excellent agreement with the values measured for small and moderate intensities of the applied field. This indicates that for $Re_m \hat{H}^2 \leq 70$, the dependence of the drifting velocity with the applied field is quadratic and obey a law such as

$$U/U_c = \mathcal{C}_1 Re_m \hat{H}^2, \tag{30}$$

where $\mathcal{C}_1$ is a constant that should take into account the physical properties of the system such as ϕ and χ^s. Our results led to $\mathcal{C}_1 = 9 \cdot 10^{-5}$.

For higher applied field intensities, however, the velocity no longer follows the predictions of eq.(28). It is verified that the actual drift velocity of the magnetic drop is higher than that predicted by $U/U_c = \mathcal{C}_1 Re_m \hat{H}^2$, as it can be

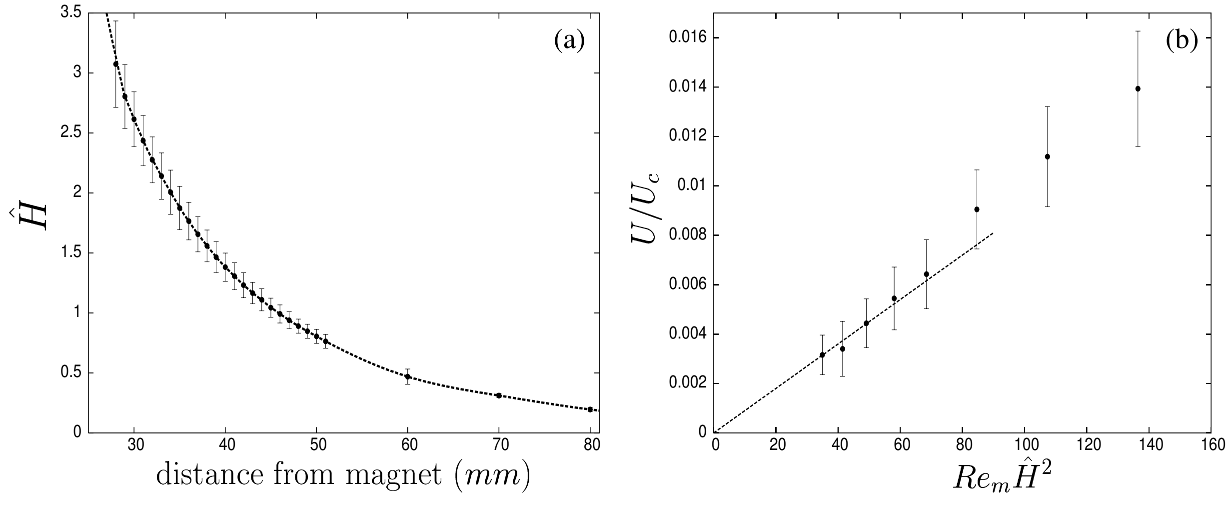

Figure 2: Dimensionless magnetic inductance profile behaviour with the distance from the magnet (a) and dimensionless drift velocity (b). The dashed line in (b) represents the prediction given by eq.(28). The values used to determine Re_m were $\rho = 10^3 kg/m^3$, $\eta = 10^{-3} Pa \cdot s$, $B_o = 4.69 \cdot 10^{-3} T$ and $R = 5.8 \cdot 10^{-3} m$ and $\mu_o = 4\pi \cdot 10^{-7} H/m$.

seen in fig. 2(b). The prediction of the proposed theory is no longer valid for this region since one of the hypothesis on which the scaling on eq.(28) was based fails: the drop deforms and can no longer be modelled by a circular cylinder. Nevertheless, the super-linear behaviour observed in the experimental data of fig. 2(b) is in accordance with the model picture as follows. The local gradients of magnetic fields cause an internal motion of the magnetic particles on the drop towards the higher field intensity regions. The drop starts to assume a shape of a slender body and the viscous resistance to its motion is reduced. This causes the magnetic forces to be more effective, what allows the drop to move faster.

3.2 Scaling arguments for the deformation of the drop

Motivated by the results of the previous section, we shall analyse more carefully the deformation that the drop undergoes while it moves towards the magnet, since it was observed that its shape changed significantly as it approaches the magnet. The history of deformation of the magnetic drop can be evaluated by following closely the analysis developed by Taylor (1934), who analyzed the deformation of a non-magnetic drop under shear flows. The drop is approximated instantaneously by an ellipsis with major and minor axes $\mathcal{A}$ and $\mathcal{B}$, respectively. Taylor (1934) proposed that the deformation of the drop may be calculated as

$$\mathbb{D} = \frac{\mathcal{A} - \mathcal{B}}{\mathcal{A} + \mathcal{B}}. \tag{31}$$

In our experiments, $\mathbb{D}$ could be calculated on each frame obtained from the images of the motion of the drop. It can be seen from eq.(31) that $\mathbb{D} \to 0$ when the shape of the drop is close to a perfect circle, and $\mathbb{D} \to 1$ when the drop has the shape of a rod. This method is efficient for drops that do not deformate in excess, since, in this case, the values of $\mathbb{D}$ would not differ significantly from 1. The studies developed here correspond to a first order theory, which may be applied for small deformation regimes.

A few arguments may clarify the behaviour of the deformation of the drop with respect to the applied magnetic field. If one considers the total magnetic force scale f_m on the scale of the drop to be given by $f_m \sim \mu_o H^2 R^2$, a typical scale for the shear stress caused by magnetic forces should be given by

$$\frac{f_m}{area} \sim \frac{\mu_o}{\pi R^2} H^2, \tag{32}$$

where $area = \pi R^2$ is the total area of a circular drop. Now, the shear stress is balanced by the viscous shear stress, say $\eta\dot{\gamma}$, where $\dot{\gamma}$ is the rate of strain of the flow and may be defined as $\mathbb{D}/(R/U)$, with R/U being a time scale of the deformation and with U defined now as $U \sim H\sqrt{\mu_o/\rho}$. The first order approximation for $\mathbb{D}$ is expressed by

$$\mathbb{D} \sim \frac{\sqrt{\rho\mu_o} R H}{\pi\eta} \sim \frac{1}{\pi} Re_m \hat{H} + \mathcal{O}(Re_m^2). \tag{33}$$

It is thus expected that the deformation of the magnetic drop would change linearly with the applied field at $\mathcal{O}(Re_m)$. This result was validated by our experimental data obtained under the same conditions as for the drift velocity analysis.

The results are shown in fig. 3(a) and some shots of the experiment are presented in fig. 3(b).

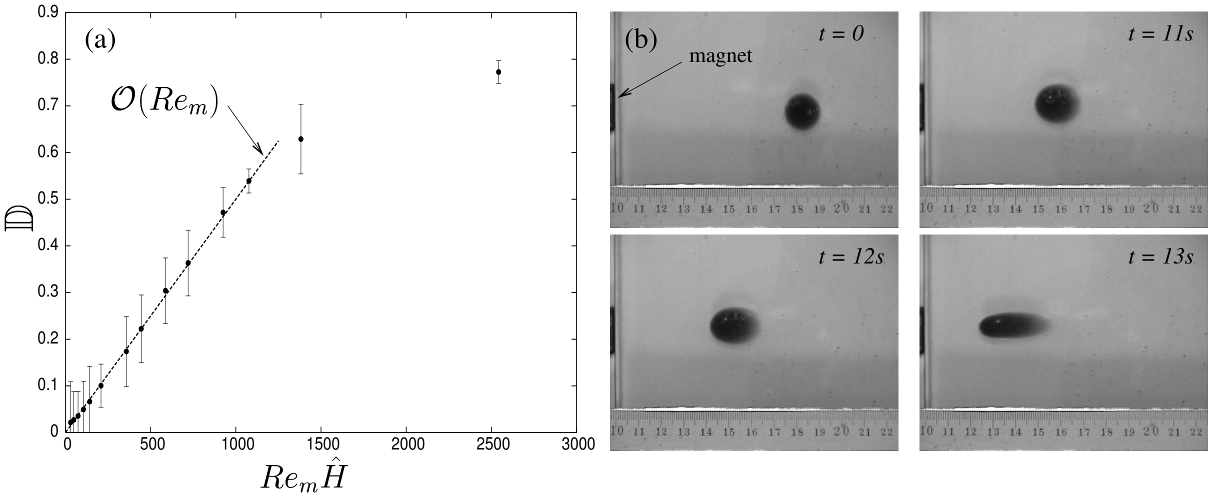

Figure 3: Deformation of a magnetic drop as a function of the local applied field (a). In (b), a typical sequence of the motion of the drop under the action of an applied magnetic field.

Figure 3(a) shows that the deformation of the drop varies linearly with the applied field even for $Re_m\hat{H}$ close to 1200. We propose the deformation of the drop as being described by the simple relation

$$\mathbb{D} = \mathcal{C}_2 Re_m \hat{H}, \tag{34}$$

where $\mathcal{C}_2$ is a constant. Our measurements led to $\mathcal{C}_2 = 5 \cdot 10^{-4}$. For values of $Re_m\hat{H} > 1200$, approximately, the deformation of the is no longer linear with respect to $\hat{H}$ and the $\mathcal{O}(Re_m)$ theory is no longer valid. Fig. 3(b) illustrates the time evolution of the drop shape for a typical run of our experiments. Significant deformations of the drop may occur for short times when the applied field intensity is high, so that a higher frequency of acquisition should be more adequate for a detailed analysis under this condition.

4. An Asymptotic Solution for the Flow Rate of a Magnetic Fluid in a Pipe Flow

Once the capture process of a magnetic drop under the action of a magnetic field is understood, we shall evaluate the flow rate that would be obtained for a magnetic fluid flow under the action of a axial magnetic field. The flow of magnetic fluids under the action of axial fields has been subject of several studies developed by Zahn (1990), Shiliomis & Morozov (1994) and Felderhof (2001). In all these works, the central problem is to define adequate hypothesis that would linearize this problem, which is strongly non-linear. In this work, we shall propose a linearized theory which allows the definition of a closed expression for the volume rate $\mathcal{Q}$ in terms of the applied field. We understand these studies as a first starting point to predict the rate of water-oil obtained in a process involving magnetic separation.

Suppose a unidirectional axisymmetric flow of a magnetic fluid in a pipe of dimensionless radius $\mathcal{R}^* = 1$ with a constant pressure gradient $-\mathcal{G}$, with $\mathcal{G} > 0$, so that, by the continuity equation, eq.(6), it is valid to assume $\mathbf{u} = u(r)\mathbf{e}_z$. Under this condition, eq.(12) reduces simply to:

$$\mathcal{G} + \frac{1}{Re}\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial u}{\partial r}\right) + C_{pm}M\frac{\partial H}{\partial z} = 0. \tag{35}$$

The parameter Re is the radius based Reynolds number, defined as in eq.(17), with $L = \mathcal{R}$. The magnetization M is determined by eq.(14) and, since $\mathbf{\Omega} = -\frac{1}{2}\frac{\partial u}{\partial r}\mathbf{e}_\theta$, the M_θ component of the magnetization vanishes and the following equations for the other components are obtained:

$$M_z\frac{\partial u}{\partial r} = -\frac{2}{\hat{\omega}}(M_r - M_r^o), \quad \text{and} \quad M_r\frac{\partial u}{\partial r} = \frac{2}{\hat{\omega}}(M_z - M_z^o). \tag{36}$$

As a first approximation, the changes on the magnetization due to the flow and to the applied field can be considered as a small deviation from the equilibrium states M_r^o and M_z^o, that is $M_r \approx M_r^o + Mr'$ and $M_z \approx M_z^o + Mz'$, with the disturbances Mr' and Mz' being small. Retaining only linear terms in eq.(36) and using that $M_r^o = 0$, since $\mathbf{H_o} = H_o\mathbf{e}_z$, it follows that

$$M_r = -\frac{1}{2}\hat{\omega}M_z^o\frac{\partial u}{\partial r}, \quad \text{and} \quad M_z = M_z^o. \tag{37}$$

This means that the radial magnetization will be changed slightly by the flow whereas the axial magnetization remains constant. This assumption allows a significant simplification on the absolute value of the dimensionless magnetization that appears in eq.(22), namely

$$M \approx M_z^o \left[1 + \frac{1}{8}\hat{\omega}^2 \left(\frac{\partial u}{\partial r} \right)^2 \right]. \tag{38}$$

Consequently, eq.(35) takes the form:

$$\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial u}{\partial r} \right) + \mathcal{G}Re + C_{pm}ReM_z^o\frac{\partial H}{\partial z} + \frac{1}{8}\hat{\omega}^2 C_{pm}ReM_z^o\frac{\partial H}{\partial z}\left(\frac{\partial u}{\partial r} \right)^2 = 0. \tag{39}$$

As M_z^o and $\partial H/\partial z$ are functions of z only, eq.(39) can be written as

$$\frac{\partial^2 u}{\partial r^2} + \frac{1}{r}\frac{\partial u}{\partial r} + \mathcal{T}(z) + \epsilon\mathcal{F}(z)\left(\frac{\partial u}{\partial r} \right)^2 = 0, \tag{40}$$

with

$$\mathcal{T}(z) = \mathcal{G}Re + C_{pm}ReM_z^o\frac{\partial H}{\partial z} \quad \text{and} \quad \mathcal{F}(z) = C_{pm}ReM_z^o\frac{\partial H}{\partial z}, \tag{41}$$

and where $\epsilon = \hat{\omega}^2/8$ is a small parameter. The order of eq.(40) can be reduced if the substitution $v(r) = \partial u(r)/\partial r$ is introduced. This leads to a first order weakly non-linear ordinary differential equation in $v(r)$. Since $\epsilon << 1$, a regular asymptotic solution may be applied by expanding $v(r)$ like

$$v(r) = v_o(r) + \epsilon v_1(r) + \epsilon^2 v_2(r) + \cdots \tag{42}$$

The boundary condition for $v(r)$ comes from the symmetry of $u(r)$ with respect to $r = 0$, i.e. $v_o(0) = v_1(0) = v_2(0) = 0$. The following equations are obtained for v_o, v_1 and v_2 respectively:

$$\frac{\partial v_o}{\partial r} + \frac{v_o}{r} + \mathcal{T}(z) = 0, \quad \frac{\partial v_1}{\partial r} + \frac{v_1}{r} + v_o^2\mathcal{F}(z) = 0 \quad \text{and} \quad \frac{\partial v_2}{\partial r} + \frac{v_2}{r} + 2v_o v_1\mathcal{F}(z) = 0. \tag{43}$$

Solving this system of differential equations, and imposing the respective boundary condition for each equation, we obtain

$$v(r) = -\frac{1}{2}r\mathcal{T}(z) - \frac{\epsilon}{16}r^3\mathcal{F}(z)\mathcal{T}^2(z) - \frac{\epsilon^2}{96}r^5\mathcal{F}^2(z)\mathcal{T}^3(z) + \cdots \tag{44}$$

Since $v(r) = \partial u(r)/\partial r$, if eq.(44) is integrated with respect to r and the non-slip boundary condition at the wall of the tube, i.e. $u(1) = 0$, is imposed, we obtain the following expression for the velocity profile $u(r)$:

$$u(r) = \frac{1}{4}\mathcal{T}(z)\left(1 - r^2\right) + \frac{\epsilon}{64}\mathcal{F}(z)\mathcal{T}^2(z)\left(1 - r^4\right) + \frac{\epsilon^2}{576}\mathcal{F}^2(z)\mathcal{T}^3(z)\left(1 - r^6\right) + \cdots \tag{45}$$

The volume rate of $\mathcal{Q}$ can be found if the velocity profile is integrated. Then, it follows:

$$\mathcal{Q} = 2\pi\int_0^1 u(r)rdr = \frac{\pi}{8}\mathcal{T}(z) + \frac{\pi\epsilon}{96}\mathcal{F}(z)\mathcal{T}^2(z) + \frac{\pi\epsilon^2}{768}\mathcal{F}^2(z)\mathcal{T}^3(z) + \cdots \tag{46}$$

If we keep the level of description up to ϵ^2, this is the last result one can achieve if the magnetic field is not solved explicitly. We can, however, propose a closed solution for this equation, based only on the values of the magnetic field in the extremities $z = 0$ and $z = \ell$ of the tube, by holding only the linear terms in $\partial H/\partial z$ in eq.(46). From the definitions of $\mathcal{T}(z)$ and $\mathcal{F}(z)$ in eq.(41), the $\mathcal{O}(\epsilon)$ term admits simplifications to keep strictly the linear dependence on the magnetic field gradient. This allows one to obtain a closed solution for $\mathcal{Q}$, but may reduce its range of validity in term of the gradient of the applied field. Thus, expanding the term corresponding to the square of $\mathcal{T}(z)$ and keeping only the linear terms in the magnetic field gradient, it may be written that:

$$\mathcal{F}(z)\mathcal{T}^2(z) = \left(C_{pm}ReM_z^o\frac{\partial H}{\partial z} \right)\left(\mathcal{G}Re + C_{pm}ReM_z^o\frac{\partial H}{\partial z} \right)^2 \sim \left(C_{pm}ReM_z^o\frac{\partial H}{\partial z} \right)(\mathcal{G}Re)^2 + \mathcal{O}\left[\left(\frac{\partial H}{\partial z} \right)^2 \right]. \tag{47}$$

Isolating the magnetic field gradient in eq.(46), and from the definition of equilibrium magnetization, eq.(15), on its dimensionless version, it follows:

$$\left(\mathcal{Q} - \frac{\pi}{8}\mathcal{G}Re\right)\left(\frac{\pi}{8}C_{pm}Re + \epsilon\frac{\pi}{96}\mathcal{G}^2Re^2C_{pm}\right)^{-1} = M_s^*\left(cotgh(\Theta^*H) - \frac{1}{\Theta^*H}\right)\frac{\partial H}{\partial z}, \quad (48)$$

where $\Theta^* = H_o\Theta$ and $M_s^* = M_s/H_o$. Integrating the left side of this equation with respect to z, from $z = 0$ to $z = \ell$, and the right side with respect to H, from the value of the applied field in $z = 0$, H_o, to the value of the applied field in $z = \ell$, H_ℓ, the expression for the flow rate is finally obtained as:

$$\mathcal{Q} = \frac{\pi}{8}\mathcal{G}Re\left[1 + \frac{M_s^*}{\Theta^*\ell}C_{pm}\left(\frac{1}{\mathcal{G}} + \frac{\epsilon}{12}Re^2\mathcal{G}\right)\ln\left|\frac{H_o sinh(\Theta^*H_\ell)}{H_\ell sinh(\Theta^*H_o)}\right|\right], \quad (49)$$

This result shows that the flow of a magnetic fluid in a tube in the presence of a applied field is equivalent to a Poiseuille flow with a non-newtonian correction associated to the magnetization of the magnetic fluid. If the above equation is written in dimensional variables, one can define an effective viscosity $\bar{\eta}$ of the magnetic fluid given by

$$\bar{\eta} = \eta\left[1 + \frac{M_s}{\Theta\mathcal{G}\ell}\left(\mu_o + \epsilon\frac{\mathcal{R}^2\mathcal{G}^2}{12\eta}\right)\ln\left|\frac{H_{zo}sinh(\Theta H_{z\ell})}{H_{z\ell}sinh(\Theta H_{zo})}\right|\right]^{-1}. \quad (50)$$

Note that now all quantities appearing in this equation are dimensional.

Equation (49) may capture the first non-linear effect due to the changes of magnetization of the magnetic fluid submitted to weak magnetic field gradients in the flow.

5. Concluding Remarks

In this work, we have studied the physical parameters involved in a process of magnetic separation and we have proposed a theory for the drift velocity of a magnetic drop in the viscous regime and the deformation of a magnetic drop for small and moderate values of applied field. In addition, we found a closed expression for the flow rate of magnetic fluid in a tube in the presence of a magnetic field.

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