Title of the manuscript:

Stability behaviour of composite magnetorheological fluids by an induction method

Authors:

G.R. Iglesias[1*], Andrés Roldán[2], Luis Reyes[2], Laura Rodriguez-Arco[1] and J.D.G. Durán[1]

[1]*Department of Applied Physics, School of Science*

[2]*Department of Electronic and Technology in Computer*

University of Granada, 18071 Granada, Spain

* Corresponding author:

Guillermo R. Iglesias

Departamento de Física Aplicada

Facultad de Ciencias

Universidad de Granada

18071 Granada, Spain

Phone: +34-958 24 32 09

FAX: +34-958 24 32 14

E-mail: iglesias@ugr.es

HIGHLIGHTS

- Suspensions consisting of magnetic and non-magnetic particles (composite MR fluids) show enhanced colloidal stability against gravitational settling.

- The addition of non-magnetic particles also leads to an improvement of the magnetorheological response.

ABSTRACT

In this work we study the stability behaviour of composite magnetorheological (MR) fluids consisting of magnetic (iron) and non-magnetic (poly (methylmethacrylate), PMMA) particles dispersed in mineral oil. Because of the opacity of the suspensions, optical methods traditionally employed for evaluation of the gravitational settling in colloidal suspensions are not suitable for sedimentation follow-up in this case. For this reason, we use an alternative method based on the evaluation of the resonant frequency of the inductance of a thin coil surrounding the sample The movement of the coil along the height of the container at specified steps and time intervals allows obtaining information about the local volume fraction of particles inside the tube. The obtained successive profiles for the multi-component suspensions show a decrease of the iron particle settling and of the initial rate of settling as the PMMA volume fraction is increased. Finally, the increase of the PMMA concentration gives rise to an improvement of the rheological properties upon magnetic field application for a given concentration of iron. Both a strongrheological response and a good colloidal stability are essential for practical applications.

Keywords: stability; sedimentation profile; multi-component magnetic suspension; magnetic particles; non-magnetic particles; magnetorheological fluids.

1. INTRODUCTION

Magnetorheological (MR) fluids are suspensions of micron-sized particles of magnetic materials in liquid carriers. In the absence of an applied magnetic field, the rheological behavior of MR fluids is close to that of a Newtonian fluid. However, when an external magnetic field is applied, the rheological behavior changes to a plastic behavior with the appearance of a yield stress, whose magnitude increases with the intensity of the field. Such a change is known as the MR effect and it is due to the arrangement of the dispersed magnetic particles into chain-like aggregates aligned in the field direction (Bossis et al., 2002). The MR effect has many technological applications and allows the use of MR fluids as shock absorbers, dampers or clutches (Klingenberg, 2001, Jolly et al., 1999, Bombard et al., 2007). In most of these applications, a strong MR effect is achieved by using strong ferromagnetic metals as the materials of the dispersed phase of the fluid, such as iron, cobalt or nickel. However, these materials present a disadvantage concerning the colloidal stability of the fluids. More specifically, because of their high densities (i.e., close to 8 g/cm3), the dispersed particles experience strong gravitational settling with the formation of compact sediments at the bottom of the fluid container that are not easily redispersed. The consequence of an excessive gravitational settling is a decrease of the magnetorheological response because of the non-uniform distribution of the particles. Many methods have been proposed in the literature in order to reduce the extent of settling (Bombard et al., 2007, Fang et al., 2008, Lopez-Lopez et al., 2005, Shahrivar and de Vicente, 2013).

In some recent works it has been shown that the addition of non-magnetic particles to MR fluids gives rise to an unexpected increase of the MR effect (Ulicny et al., 2010, Klingenberg and Ulicny, 2011, Lopez-Lopez et al., 2008, Rodriguez-Arco et al., 2013b, Rodriguez-Arco et al., (2014). What is more, the addition of diamagnetic particles in these multi-component suspensions may also reduce gravitational settling of the magnetic particles due to the buoyancy of the non-magnetic particles, whose density is closer to that of the carrier fluid. For example, the use of non-magnetic particles in MR fluids as anti-settling agents provided with softer sediments once the magnetic particles settled down (Galindo-Gonzalez et al., 2005). In this study, the sedimentation of the magnetic particles was measured employing optical methods. However, due to the opacity of the suspensions, the use of these methods does not seem reliable

enough for sedimentation follow-up. The magnetic responsiveness of the magnetic particles, in turn, offers a number of possibilities for evaluating their settling. More specifically, when the magnetic particles settle down, the particle concentration along the fluid container changes, giving rise to changes of the suspension magnetic permeability. Using this principle, there exist some alternative methods for checking gravitational settling that are based on the measurement of the variation on the inductance of a coil surrounding the suspension as a function of time (Ngatu and Wereley, 2007, Chen and Chen, 2003, Gorodkin et al., 2000, Iglesias et al., 2011, Iglesias et al.,2007).

In this work we study the stability against settling of multi-component suspensions of magnetic and non-magnetic particles using a an alternative method based on the evaluation of the resonant frequency of a parallel circuit formed by a capacitor and an inductance. We provide with a description of the prototype employed for this purpose. We also present results for a MR fluid with the same volume fraction of magnetic particles. Finally we show results on the dynamic rheological properties of all the suspensions.

2. Experimental

2.1. Materials

Carbonyl iron powder (HS quality) from BASF was used as the magnetic material. It consisted of polydisperse spherical particles with a median particle size of d50 = 2.3 μm (see figure 1a) (density = 7.5 g/cm3). The non-magnetic particles were poly(methylmethacrylate), PMMA, microspheres (figure 1b), which were purchased from Microbeads (Spheromers CA10). The average diameter of the particles was 9.9±0.4 μm (density = 1.2 g/cm3). Figures 1a and 1b show electron microscopy pictures of both types of particles. The base carrier fluid for all the suspensions was mineral oil (Sigma Aldrich); its viscosity at 25 °C was 0.028 ± 0.001 Pa•s. In one of the suspension prepared the oil carried also contains in solution 6.15 g/L of aluminum stearate (AlSt) surfactant (Sigma Aldrich).
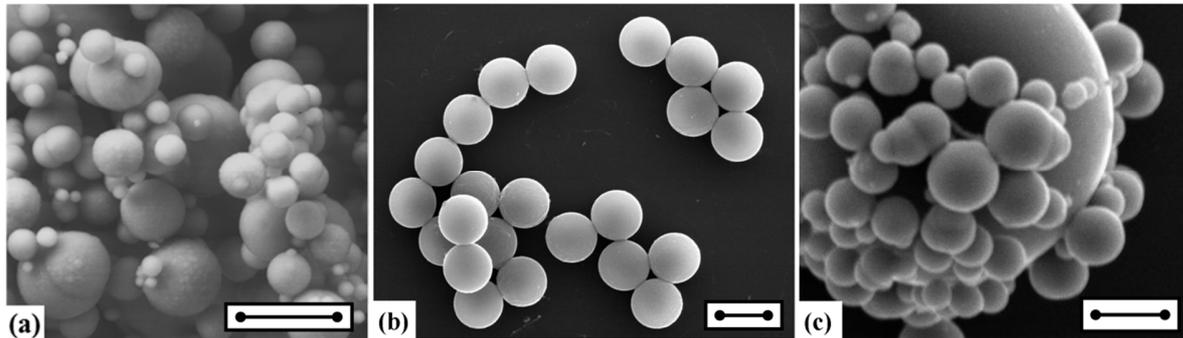
Figure 1. Scanning electron microscopy pictures of iron (a) and PMMA (b) particles, and of a non-magnetic-core-magnetic-shell composite consisting of a PMMA particle (core) coated by an irregular shell of iron particles (c). Bar lengths: (a) 2 μm; (b) 10 μm; (c) 2 μm.

2.2. Suspension preparation

We first of all prepared a MR fluid consisting of 10 vol % of iron microparticles in pure mineral oil or, alternatively a solution of AlSt in oil was employed in this step. For this purpose, appropriate amounts of the solid powder were dispersed in the liquid carrier and the suspension was mechanically stirred until complete homogenization. In addition we prepared three multi-component suspensions that consisted of iron and PMMA particles in mineral oil using the same procedure. The volume fraction of iron was 10 vol % for all of them and the volume fraction of PMMA ranged from 10 to 30 vol %. All the samples were redispersed using a vortex mixer before the rheologic/stability tests.

2.3. Stability measurements

The stability study was based in the so called "induction method" where the key element is a coil sensor surrounds the sample and the associated dependence between its inductance L (Ngatu and Wereley, 2007, Chen and Chen, 2003, Gorodkin et al., 2000, Iglesias et al., 2011, Iglesias et al., 2007) and the volume fraction ϕ of magnetizable particles inside the sample. The definition of the inductance of a solenoid is given by:

$$L = \mu_0 \mu_r \frac{N^2 A}{\ell} \qquad (1)$$

where N is the number of turn, A is the cross-sectional area, l is the solenoid length, μ_0 is magnetic permeability of vacuum and μ_r is the relative permeability of the magnetic particle. Instead of measure the inductance value, we measure the resonant frequency of a parallel circuit formed by L and capacitor C given by:

$$f = \frac{1}{2\pi\sqrt{LC}} \qquad (2)$$

Where the relationship between the frequency and spherical particle concentration is given by:

$$\mu_r = \mu_{rm} \frac{1 + 2\beta\phi}{1 - \beta\phi}; \quad \beta = \frac{\mu_{rp} - \mu_{rm}}{\mu_{rp} + 2\mu_{rm}} \qquad (3)$$

where μ_{rp} is the magnetic permeability of the particles and μ_m is the magnetic permeability of themedium and β is the magnetic contrast factor. In the case of iron particles, $\mu_{rp} >> \mu_{rm}$, $\beta \approx 1$:

$$\mu_r = \mu_{rm} \frac{1 + 2\phi}{1 - \phi} \qquad (4)$$

With this latter expression and eqs. (1) and (2), we can define the instantaneous frequency as the radio between the initial frequency f_0 and the frequency at any time $f(t)$, this frequency gives us the concentration of solid particles:

$$f(t) \propto \sqrt{\frac{1 - \phi(t)}{1 + 2\phi(t)}} \qquad (5)$$

$$f_r(t) = \frac{f(t=0)}{f(t)} = \sqrt{\frac{\left[1+2\phi(t)\right](1-\phi_0)}{[1+2\phi_0](1-\phi(t))}} \qquad (6)$$

where ϕ_0 is the initial concentration of the suspension after preparation (i.e., the particles are homogeneously distributed in the suspension and the volume fraction is uniform). Considering a fixed position at a certain time, the expression (6) could be re-write including the position z ($z = 0$ corresponds to the bottom of the test tube):

$$f_r(t,z) \equiv \frac{f_0}{f(t,z)} \sqrt{\frac{[1+2\phi(t,z)](1-\phi_0)}{[(1+2\phi_0)][1-\phi(t,z)]}} \qquad (7)$$

Therefore, by measuring the resonant frequency $f(t, z)$ of the coil at position z and time t we were able to calculate the volume fraction of magnetic solids at every position and time. This was a direct evaluation of the stability of the suspension, with the advantage of being performed in real time. The calibration of the system was conducted by measuring the resonant frequency using a test tube filled with mineral oil and thus, with no particles inside (f_0) and the frequency (f) with different concentrations of solids in the range of volume fractions of interest.

The successive profiles obtained when the sensor coil swept vertically along the tube at specified steps and time intervals gave a rapid view of the settling state of the sample. For this purpose, we designed and built an automatic system able to scan the sample at specified steps and time intervals providing information on the local volume fraction of particles. The system is based on a microcontroller with a standard USB output connected to the computer, where the users can storage data of the total time of the measurement, steps, frequency, concentration, positions of the coil, etc. Figure 2 shows the 3D layout of the mechanical design of the device (a), a detailed sketch of the sensor coil (b) and a photograph of the device prototype (c).
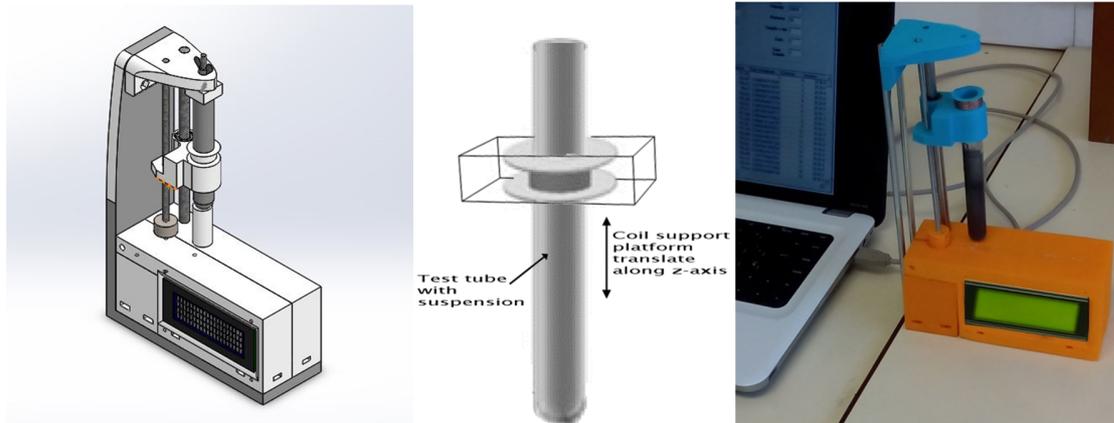
Figure 2.a) 3D layout of the mechanical design of the device. b) Sketch of the sensor coil. c)Photograph of the prototype. The sensor coil scans the whole height of the test tube and the resonant frequency of a parallel circuit is measured. This frequency is correlated with the volume fraction of magnetic particles at each height in real time, providing with a view of the particle settling.

2.4. Rheological measurements

The dynamic rheological properties were measured at 25 °C using a Haake MARS III (Thermo Fisher Scientific, USA) rheometer with parallel plate geometry (diameter of 3.5 cm, surface roughness of about 0.5 mm, and distance between the plates of 400 μm).The measuring protocol was as follows: (i) pre-shear: application of a shear rate of 150 s^{-1}for 30 s; (ii) waiting time: sample at rest for 2 minutes; (iii) amplitude/frequency sweep. All the samples were first subjected to an amplitude sweep at a fixed frequency of 1 Hz. The sinusoidal stress at each step was applied for a time large enough to allow 8 periods of oscillation. Both types of measurements were performed for different values of the magnetic field strength.

3. Results and discussion

3.1 Stability profiles evaluation

The multi-component suspensions showed a clear reduction of the settling extent and rate. We first of all measured the evolution of the volume fraction along the height of the container as a function of time. The MR fluid without PMMA, i.e., only 10 vol % of iron particles and pure oil,

showed a strong reduction of the volume fraction near the top of the tube even at the beginning of the experiment (figure 3a). For this sample, the experiment ended up (i.e., after 24 h) with all the solids deposited at the bottom of the container where the volume fraction was almost 23% higher than at the top of the tube, because of the settling of the particles. Secondly, in figure 3b the sedimentation profile of a MRF containing a stabilizing surfactant (aluminum stearate, AlSt) are shown. As observed, in this figure 3b the beginning of the sedimentation is delayed in comparison with the Fe/pure oil MRF, as it will be better analyzed below in figure 4. In addition, the concentration of settled particles in the bottom of the container increase up to 28% (i.e. after 24 h), likely because the surfactant avoids the formation of loose big iron aggregates in this region. For the multi-component suspensions the profiles clearly changed and much longer times were needed to achieve a significant reduction in volume fraction in the upper half of the tube (figure 3 c-d-e). After 24 h, for example, the increase of the volume fraction at the bottom for the sample with 30 vol % of PMMA was of about 2%, while it was between 23% and 28% for the both samples without non-magnetic particles. These results suggested that increasing concentrations of PMMA prevented particles from fast sedimentation.
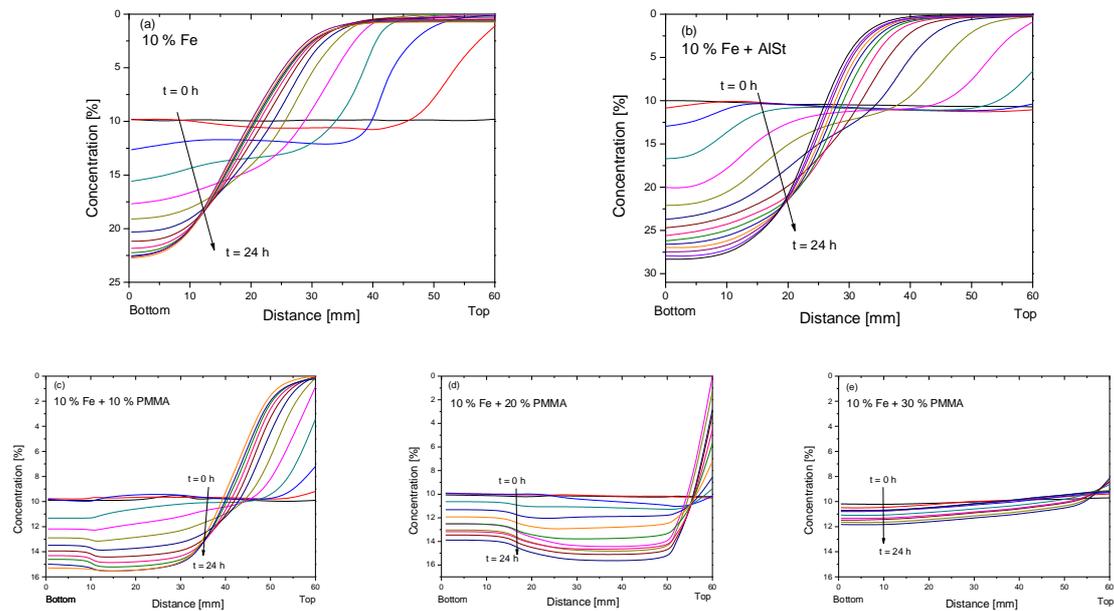
Figure 3. Iron volume fraction profile along the suspension container as a function of time for 10 % iron suspensions and the following additives: a) without additives; b) AlSt surfactant; c)10 % PMMA particles; d) 20 % PMMA; e) 30 % PMMA.

For a clearer picture of the settling profiles we can select just two specific positions of the sensing coil and to display the ϕ(t) trends (Figure 4). At the top position (Figure 4a) a fast and strong reduction in ϕ, connected to gravitational settling, was observed for the samples without PMMA. In Figure 4a (container top) we can observe that in the case of the iron/AlSt sample, the settling rate is slower than in both the Fe/pure oil and the Fe/10% PMMA suspensions. On the contrary, as mentioned before, the surfactant favors the formation of the most concentrated sediment at the bottom region (see figure 4b). The AlSt addition seems to induce the formation of a packed sediment of non-aggregated particles. For the Fe/PMMA suspensions, the variation in the particle concentration is progressively reduced (both in the top and bottom positions) as the PMMA volume fraction is increased, being almost negligible for the sample with 30 vol % of

PMMA.A more quantitative figure can be obtaining by estimating the initial settling rate $d\phi/dt$ from the concentration vs. time curves in figure 4a (top), as shown in Table 1.

Table 1.Initial settling rate as obtained from data in Figure 4(a)

Sample (10% Fe)	Settling rate (h^{-1})
+AlSt	1.53
+ 0% PMMA	4.48
+ 10% PMMA	1.19
+ 20% PMMA	0.53
+ 30% PMMA	0.014

As observed, the estimated settling rate was negligible for the largest PMMA concentrations. The profiles at the bottom position were consistent with those of the top, showing an increase in the concentration of particles at the lowermost section of the tube coming from the upper regions (Figure 4b).
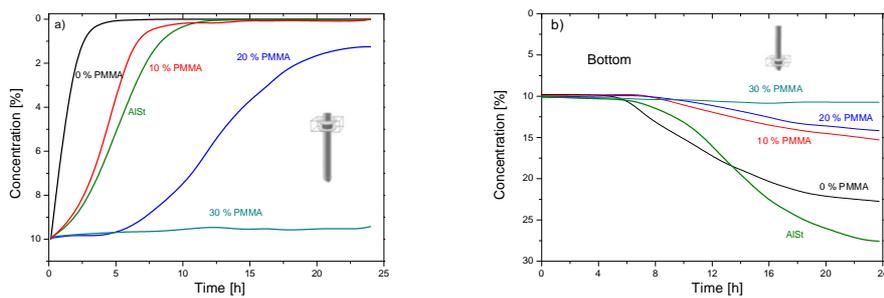
Figure 4. Time evolution of the volume fraction for all the samples studied at two positions of the container: a) top, b) bottom.

3.2 Dynamic Rheological Properties

All the samples showed a yielding behavior which became stronger with the concentration of PMMA particles. The yielding behavior is manifested by a transition from an elastic behavior (i.e., solid-like) to a viscous behavior (i.e., liquid-like) at some critical value of the stress amplitude, σ_0. The elastic and the viscous contributions to the stress response are quantified by the storage (G'), and loss (G'') moduli, respectively. At low values of the imposed oscillatory stress both moduli remained almost linear, and thus, independent of the stress amplitude for all the samples (figure 3a). This region of the G'-G'' vs. σ_0 graph is the so-called viscoelastic linear region (VLR). At higher values of σ_0, G' exhibited a strong decrease. In the case of G'', there was a slight increase followed by an abrupt decrease similar to the one of G'. At some critical value of σ_0 the curves of G' and G'' crossed, and thus, the rheological behaviour changed from being that of an elastic solid to a viscous liquid behavior (figure 3a). Such a transition in the dynamic behavior is connected to the yielding behavior of magnetorheological fluids upon magnetic field application. From the microscopic point of view, it is associated with the breakage of the magnetic field-induced structures inside the suspension because of the shear forces (Parthasarathy and Klingenberg, 1995). The critical value of σ_0 at which the crossing between G' and G'' would be equivalent to the yield stress in steady state measurements, and similarly to what happens to the yield stress, the critical amplitude increased with the strength of the applied magnetic field for all the samples (figure 3b). Interestingly, the transition to the viscous regime shifted to higher values of the stress amplitude when the volume fraction of PMMA particles in the suspensions was increased (figure 3b). This result suggested that the particle structures became stronger with the addition of PMMA particles, which is in good agreement with the steady state results reported previously for these suspensions (Rodriguez-

Arco et al., 2013b,Rodriguez-Arco et al., 2014). The formation of such composites also led to an increase of the viscosity and the shear stress in the absence of field, which also explains the yielding behavior observed in the dynamic measurements of the present work. Indeed, the critical stress amplitudes were non-negligible at zero field, specially for the suspension with the highest concentration of PMMA particles (Figure 3b). The second mechanism appeared when the formation of non-magnetic-core-magnetic-shell composites was hindered because of the addition of a surfactant. In this second case, the increase of the yield stress was attributed to an enhancement of the rotational diffusion of the aggregates of magnetic particles due to collisions with the non-magnetic particles under shear (Rodriguez-Arco et al.,2014). The reality is that both mechanisms are likely present and contribute to enhance the MR properties in multi-component suspensions.
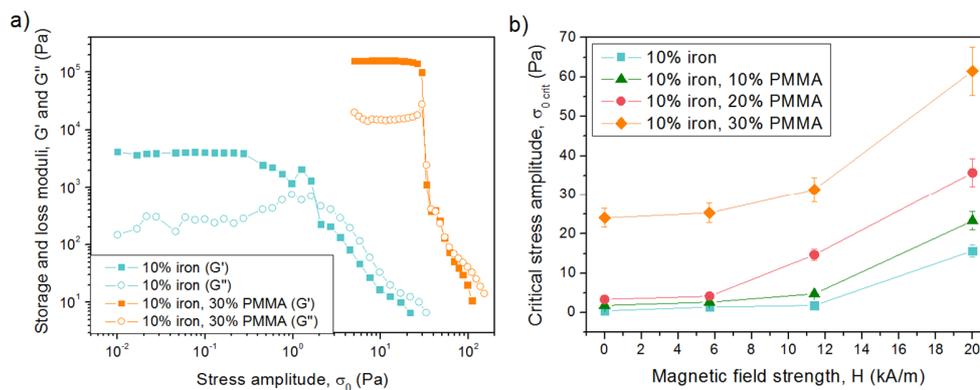
Figure 5. a) Amplitude sweeps for suspensions of 10 vol % of iron and PMMA volume fractions of 0 vol % and 30 vol % (indicated in the graphs). The magnetic field strength was 11.4 kA/m (similar results were obtained for other strengths, not shown here for brevity). Both suspensions showed a yielding behavior as shown by the crossing of the storage modulus, G', and the loss modulus, G'', at a critical value of the stress amplitude, $\sigma_{0\,crit}$. Both G' and G'' were higher for the suspension with PMMA particles. **b)** Critical stress amplitude, $\sigma_{0\,crit}$, as a function of the magnetic field strength for all the samples. As observed, $\sigma_{0\,crit}$ increased with the PMMA concentration because of the reinforcement of the particle structures. It also increas13ed with the magnetic field strength.

In addition to stronger structures, the addition of PMMA resulted in an increase of friction forces between the particles inside the structures. The strength of the particle structures against the oscillatory stimulus can be alternatively quantified by the value of G' in the VLR. As mentioned before, in this region, the suspension behaves as an elastic solid and as a result, G' is independent of the stress amplitude. Such behavior is due to an elastic, homogeneous, deformation of the particle structures inside the suspension. The addition of PMMA to the suspensions of iron particles resulted in an increase of the strength of the structures, because the values of G' in the VLR increased with the volume fraction of PMMA (figure 4a). Such an increase was also obtained in the case of G'', which implies that the addition of PMMA particles also increased the dissipation of energy upon the oscillatory stimulus (figure 4b)(Halsey et al., 1992). This could be due to the formation of the non-magnetic-core–magnetic-shell composites described above. More specifically, the irregular distribution of iron particles around the PMMA core (see figure 1c) must have increased the roughness of the surface of the composites in comparison to iron particles, which would result in an increase of friction between the composites inside the aggregates.
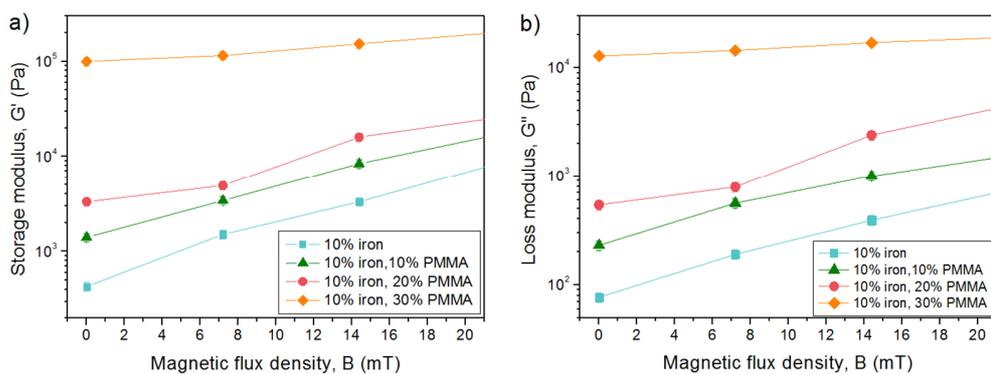
Figure 6. Storage, G', **(a)** and loss, G'', **(b)** moduli in the viscoelastic linear region (VLR) as a function of the applied magnetic flux density for all the samples. Both moduli increased with the magnetic field strength and with the volume fraction of PMMA. The increase of G' was

connected to a reinforcement of the field-induced particle structures, while an increase of G''
revealed a stronger friction between the particles inside the field-induced aggregates.

CONCLUSIONS

In this work an inductance-based method has been performed for the evaluation of the stability
and sedimentation profile of suspensions of magnetic particles. We have found that the addition
of polymerparticles (PMMA) to the formulation leads to an enhancement of the colloidal
stability against gravitational settling in the suspensions as compared with that in MR fluids only
containing iron particles and pure oil. Even more, the addition of PMMA particles improves the
stability in comparison with a MR fluid stabilized by surfactant addition.

In particular, both the extent of the magnetic particle settling and its initial rate were reduced
when the volume fraction of PMMA particles was larger than 10 vol % for a given fixed volume
fraction of magnetic particles of 10 vol %. In contrast to other shear-thickening agents usually
employed to reduce settling, the addition of non-magnetic particles also led to an improvement
of the magnetorheological response of the suspensions. More specifically the transition from an
elastic behavior to a viscous behavior took place at higher values of the stress amplitude when
the non-magnetic particle volume fraction was increased for a fixed concentration of magnetic
particles. In addition, both the storage and the loss moduli in the viscoelastic linear region
increased with the volume fraction of non-magnetic particles. The enhancement of the dynamic
properties can be explained by the combination of an enhancement of the suspension magnetic
properties and an increase of the rotational diffusion of the aggregates of magnetic particles. The
double benefit of having a better colloidal stability and a strong magnetorheological response in
multi-component suspensions is very desirable from the point of view of the technological

applications of magnetic suspensions. Finally note that non-magnetic particles are less dense than magnetic ones, which reduces the final weight of the suspensions and of the devices in which they are implemented.

ACKNOWLEDGEMENTS

REFERENCES

BOMBARD, A. J. F., KNOBEL, M. & ALCANTARA, M. R. 2007. Phosphate coating on the surface of carbonyl iron powder and its effect in magnetorheological suspensions. International *Journal of Modern Physics B*, 21, 4858-4867.

BOSSIS, G., VOLKOVA, O., LACIS, S. & MEUNIER, A. 2002. Magnetorheology: Fluids,structures and rheology. *Ferrofluids: Magnetically Controllable Fluids and Their Applications*,594, 202-230.

CHEN, L. S. & CHEN, D. Y. 2003. Permalloy inductor based instrument that measures the sedimentation constant of magnetorheological fluids. *Review of Scientific Instruments*, 74,3566-3568.

FANG, F. F., YANG, M. S. & CHOI, H. J. 2008. Novel Magnetic Composite Particles of Carbonyl Iron Embedded in Polystyrene and Their Magnetorheological Characteristics. *Ieee Transactions on Magnetics*, 44, 4533-4536.

GALINDO-GONZALEZ, C., DE VICENTE, J., RAMOS-TEJADA, M. M., LOPEZ-LOPEZ,M. T., GONZALEZCABALLERO,F. & DURAN, J. D. G. 2005. Preparation and sedimentation behavior in magnetic fields of magnetite-covered clay particles. *Langmuir*, 21, 4410-4419.

GALINDO-GONZALEZ, C., IGLESIAS, G. R., GONZALEZ-CABALLERO, F. & DURAN, J.D.G. 2007. Stability of concentrated aqueous clay-magnetite suspensions. *Colloids and Surfaces a-Physicochemical and Engineering Aspects*, 306, 150-157.

GORODKIN, S. R., KORDONSKI, W. I., MEDVEDEVA, E. V., NOVIKOVA, Z. A.,SHOREY, A. B. & JACOBS, S. D.2000. A method and device for measurement of a sedimentation constant of magnetorheological fluids. *Review of Scientific Instruments*, 71,2476-2480.

HALSEY, T. C., MARTIN, J. E. & ADOLF, D. 1992. RHEOLOGY OFELECTRORHEOLOGICAL FLUIDS. *Physical Review Letters*, 68, 1519-1522.

IGLESIAS, G. R., LOPEZ-LOPEZ, M. T., DELGADO, A. V. & DURAN, J. D. G. 2011.Description and performance of a fully automatic device for the study of the sedimentation of magnetic suspensions. *Review of Scientific Instruments*, 82.

IGLESIAS, G. R., RUIZ-MORON, L. F., INSA MONESMA, J., DURAN, J. D. G. & DELGADO, A. V. 2007. An experimental method for the measurement of the stability of concentrated magnetic fluids. *Journal of Colloid and Interface Science*, 311, 475-480.

JOLLY, M. R., BENDER, J. W. & CARLSON, J. D. 1999. Properties and applications of commercial magnetorheological fluids. *Journal of Intelligent Material Systems and Structures*, 10, 5-13.

KLINGENBERG, D. J. 2001. Magnetorheology: Applications and challenges. *Aiche Journal*, 47,246-249.

KLINGENBERG, D. J. & ULICNY, J. C. 2011. Enhancing Magnetorheology. *International Journal of Modern Physics B*, 25, 911-917.

LOPEZ-LOPEZ, M. T., DE VICENTE, J., GONZALEZ-CABALLERO, F. & DURAN, J. D. G.2005. Stability of magnetizable colloidal suspensions by addition of oleic acid and silica nanoparticles. *Colloids and Surfaces a-Physicochemical and Engineering Aspects*, 264, 75-81.

LOPEZ-LOPEZ, M. T., GOMEZ-RAMIREZ, A., DURAN, J. D. G. & GONZALEZ-CABALLERO, F. 2008. Preparation and characterization of iron-based magnetorheological fluids stabilized by addition of organoclay particles. *Langmuir*, 24, 7076-7084.

NGATU, G. T. & WERELEY, N. M. 2007. Viscometric and sedimentation characterization of bidisperse magnetorheological fluids. *IEEE Transactions on Magnetics*, 43, 2474-2476.

PARTHASARATHY, M. & KLINGENBERG, D. J. 1995. A Microstructural investigation of the nonlinear response of electro rheological suspensions .2. Oscillatory Shear-Flow. *Rheologica Acta*, 34, 430-439.

RODRIGUEZ-ARCO, L., LOPEZ-LOPEZ, M. T., KUZHIR, P., BOSSIS, G. & DURAN, J. D. G. 2013a. Optimizing the Magnetic Response of Suspensions by Tailoring the Spatial Distribution of the Particle Magnetic Material. *Acs Applied Materials & Interfaces*, 5, 12143-2147.

RODRIGUEZ-ARCO, L., LOPEZ-LOPEZ, M. T., KUZHIR, P. & DURAN, J. D. G. 2013b. Steady state rheological behaviour of multi-component magnetic suspensions. *Soft Matter*, 9, 5726-5737.

RODRIGUEZ-ARCO, L., LOPEZ-LOPEZ, M. T., KUZHIR, P. & GONZALEZ-CABALLERO,F. 2014. How nonmagnetic particles intensify rotational diffusion in magnetorheological fluids. *Physical Review* E, 90.

SHAHRIVAR, K. & DE VICENTE, J. 2013. Thermoresponsive polymer-based magneto-rheological (MR)composites as a bridge between MR fluids and MR elastomers. *Soft Matter*, 9,11451-11456.

ULICNY, J. C., SNAVELY, K. S., GOLDEN, M. A. & KLINGENBERG, D. J. 2010.Enhancing magnetorheology with nonmagnetizable particles. *Applied Physics Letters*, 96.