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Oscillatory squeeze flow of suspensions of magnetic polymerized chains

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Abstract

We report a rheological study of suspensions of non-Brownian chain-like magnetic particles in the presence of magnetic fields. These particles have been synthesized using spherical iron particles by linking them with a polymer and are called polymerized chains. We have shown that, in oscillatory squeeze mode, the suspensions of such chain-like particles develop yield stress several times higher than that of conventional magnetorheological fluids based on spherical iron particles. This is explained in terms of solid friction between polymerized chains, which form entangled aggregates in the presence of a magnetic field. For the suspension of spherical particles, the squeezing force increases with the magnetic field intensity at low magnetic fields, but decreases dramatically at higher fields because of cavitation or air entrainment. Such a decrease in transmitted force does not take place in suspensions of polymerized chains, at least for fields smaller than 30 kA m^{-1}, which could make these suspensions preferable for application in squeeze-film dampers.

1. Introduction

Magnetorheological damping devices have been broadly developed during the last decade and nowadays they are appearing on the market. Most of them work in a squeezed mode, where a magnetorheological (MR) fluid is squeezed by two moving surfaces and offers some viscoelastic resistance to the motion of these surfaces. The viscoelastic response of MR fluids can be controlled by magnetic fields. A way to improve MR damping devices is to use novel MR fluids with enhanced magnetorheological properties. With this aim, we propose to use suspensions of magnetic fibers, obtained by chemical linking of micron sized iron particles. From now on, we will call these fibers polymerized chains. The synthesis and characterization of these polymerized chains are described in [1]. Due to the presence of thick polymer layers, the magnetization of the polymerized chains is two to three times smaller than the magnetization of the spherical iron particles. Surprisingly, in shear flow, the suspensions of polymerized chains develop nearly the same yield stress as the suspensions of spherical iron particles.

Many studies have been carried out on shear and squeeze rheology of classical MR fluids, composed of spherical magnetic particles [2–6]. However, to our knowledge, there is no rheological study of MR suspensions based on non-spherical magnetic particles. In the present work we are interested in analyzing the squeeze rheology of a suspension of polymerized chains. Finally, we compare the rheological properties of these novel MR fluids with the properties of the classical MR fluids.

2. Experiments and data analysis

In our experiments we used two kinds of MR fluids. They were suspensions of magnetic microparticles in ethylene glycol (Prolabo) stabilized by a silica gel, Aerosil 300. The particle shape was the only difference between these two MR fluids. The first MR fluid had spherical carbonyl iron particles (supplied by BASF) with diameter ranging between 0.5 and 3 μm. The second one had ellipsoid-like particles composed

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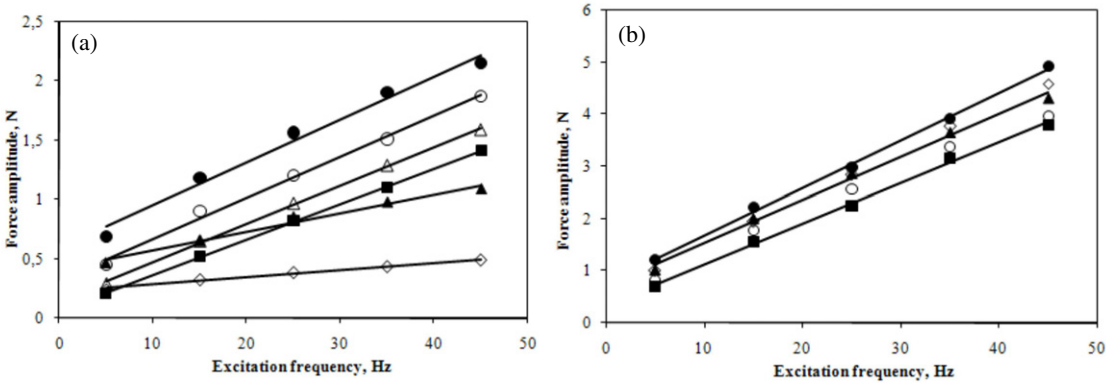

Figure 1. Squeezing force amplitude versus excitation frequency for the suspension of spherical iron particles (a) and for the suspension of polymerized chains (b), at 15% volume fraction. The displacement amplitude is $\Delta h = 50\ \mu$m; the initial gap is $h_0 = 0.5$ mm. Different curves correspond to the different magnetic field intensities. (a) ■ —0 kA m^{-1}, △—3.3 kA m^{-1}, ○—6.5 kA m^{-1}, ●—10.9 kA m^{-1}, ▲—14.1 kA m^{-1}, ◊—17.4 kA m^{-1}. (b) ■ —0 kA m^{-1}, ○—6.5 kA m^{-1}, ▲—14.1 kA m^{-1}, ◊—17.4 kA m^{-1}, ●—27.2 kA m^{-1}.

of the same spherical iron particles linked by a polymer (ethylene glycol methacrylate phosphate) [1]. The average particle length and diameter were about 100 μm and 15 μm respectively. Both types of suspension were prepared at 5 and 15% particle volume fractions. The sedimentation tests showed that all MR fluids remained well dispersed for at least 30 min, a time sufficient to carry out the rheological measurements—shear and squeeze tests.

The oscillatory squeeze test was performed with an RAC815A viscoanalyzer (Metravib Instruments). A MR fluid was confined between two parallel non-magnetic disks. The upper disk, driven by the viscoanalyzer, oscillated in the vertical direction with imposed amplitude of displacement and frequency. The MR fluid squeezed film created a viscoelastic response, which was transmitted to a force transducer connected to the fixed lower disk. The experiments consisted of measuring the squeezing force amplitude and the phase angle between the periodic force and the displacement signals in response to the imposed harmonic oscillations of the upper disk. In our experiments, the excitation frequencies f were 5, 15, 25, 35 and 45 Hz, the displacement amplitude Δh of the upper disk was 50 μm, the initial gap between the disks was $h_0 = 0.5$ mm and the upper disk radius was $R = 8.5$ mm. These measurements were carried out both in the absence and in the presence of a uniform magnetic field (0–32 kA m^{-1}) created by a solenoid placed around the disks.

All the results of the squeeze test are presented in the form of squeezing force amplitude versus frequency dependences, like the one shown in figure 1. For both MR fluids, all these dependences are perfectly approximated by a linear function $F_0 = F_Y + Af$ giving some nonzero force amplitude when interpolated to zero frequency. Furthermore, the phase lag angle between force and displacement signals lies between 85° and 90° for any measurement, which is characteristic of a weak elastic response. Such linear behavior of the squeezing force amplitude possessing a yield value F_Y depending on the magnetic field strength allows us to assume a Bingham model of MR fluids in squeeze flow: $\tau = \tau_Y \mathrm{sgn}(\dot\gamma) + \eta\dot\gamma$, with τ_Y and η being the MR fluid yield stress and viscosity respectively. To extract the yield stress and the viscosity from these data, we must consider a fluid mechanics problem of a Bingham fluid flow between two parallel disks. A good first order approximation to this problem is given in the classical work of Covey and Stanmore [7]. Using the lubrication approximation, they obtained the following expression for the squeezing force F as a function of the upper disk speed $\dot h$:

$$F = -\frac{\pi\tau_Y R^3}{h}\mathrm{sgn}(\dot h) - \frac{3\pi\eta\dot h R^4}{2h^3}. \qquad (1)$$

This formula, derived for weak Bingham numbers $Bn = \frac{\tau_Y h}{\eta\dot h} \ll 1$, can be applied safely for a very wide range of Bingham numbers upon introduction of appropriate correction factors in both terms of (1) (see [8, 9]). We estimated that formula (1) can be applied to our experiments with a 14% error, acceptable for the semi-quantitative analysis.

Let the upper disk oscillate with a frequency f such that the MR fluid film thickness varies as $h = h_0 + \Delta h\cos(2\pi f t)$ and, therefore, the upper disk velocity is $\dot h = -2\pi\Delta h f\sin(2\pi f t)$. Substituting these expressions into equation (1) and taking into account that $h \ll h_0$, we arrive at the final expression for the squeezing force amplitude as a function of the excitation frequency:

$$F_0 = \frac{\pi\tau_Y R^3}{h_0} - \frac{3\pi^2\eta\Delta h R^4}{h_0^3} f = F_Y + Af. \qquad (2)$$

The first term in (2) represents the yield force F_Y corresponding to the dynamic yield stress and the second one corresponds to the Newtonian term of the Bingham law. So, once the values of F_Y and A were obtained by a linear fit of the experimental data, we calculated the yield stress τ_Y and the viscosity η from formula (2) by putting the first term equal to F_Y and the second one to (Af).

To validate our experimental technique, we made measurements on two calibrated Newtonian liquids and found that the viscosity measured by the oscillatory squeeze test corresponded to the one obtained using standard rheological measurements.

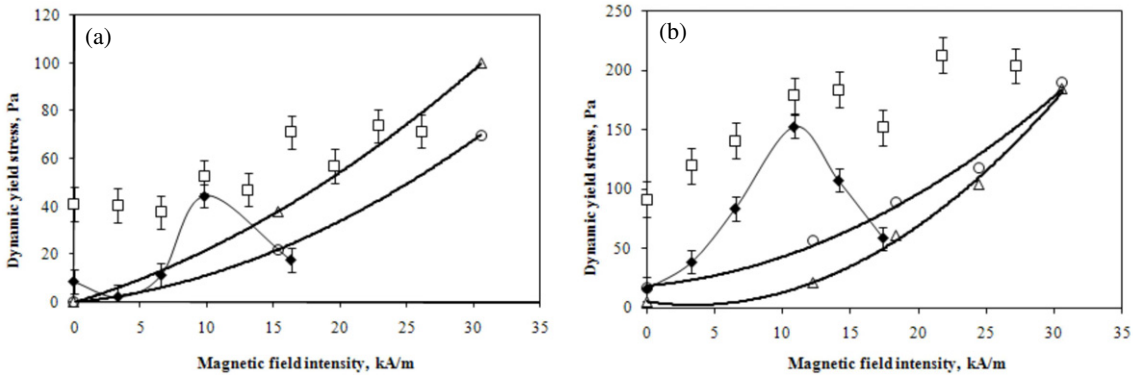

Figure 2. Field dependence of the dynamic yield stress of suspensions of spherical particles and polymerized chains at 5% (a) and 15% volume fraction (b): comparison between shear and squeeze rheometry. In squeeze tests, the displacement amplitude is $\Delta h = 50$ μm; the initial gap is $h_0 = 0.5$ mm. In both figures, the symbols ♦ correspond to squeeze flow of the suspension of spherical particles; □—squeeze, polymerized chains; △—shear flow, spherical particles; ○—shear flow, polymerized chains. Reproduced with the kind permission from Springer Science and Business Media: Kuzhir P, Lopez-Lopez M T, Vertelov G, Pradille Ch and Bossis G 2008 *Rheologica Acta* **47** 179–87.

3. Results and discussion

The force versus excitation frequency curves of the squeeze rheometry, being somewhat analogous to the flow curves of the shear rheometry, are presented in figures 1(a) and (b) for the suspensions of spherical particles and polymerized chains, at the same volume fraction of 15%. As stated above, all these curves are linear and suggest a Bingham behavior of the MR fluids in the experimental range of parameters. As observed in figure 1, the squeeze flow curves are shifted upward with increasing magnetic field, signifying a viscosity increase of MR fluid. For the suspension of spherical particles this tendency reverses at a critical value of the magnetic field ($H \approx 14$ kA m^{-1}), and a strong decrease in force is observed for higher magnetic fields (figure 1(a)). On the other hand, such force decrease is not observed for the suspension of polymerized chains even at higher fields (figure 1(b)). This phenomenon is connected with cavitation and will be discussed in detail below.

The flow curves of squeeze rheometry for 5% volume fraction MR suspensions are similar to the flow curves at 15% and are not presented here for brevity.

The main result of our study—the magnetic field effect on the dynamic yield stress of both types of suspension—is presented in figure 2. In this figure, we compare at the same time the magnetorheological effect for both types of MR fluids and the yield stress obtained by means of two different methods: shear rheometry [1] and squeeze rheometry (present study).

In shear rheometry we observe a parabolic-like growth of the dynamic yield stress with the magnetic field intensity for both types of MR fluids. We note that the dynamic yield stress of the dilute suspension ($\phi = 5\%$) of spherical particles is higher than that of the equivalent suspension of polymerized chains. For the most concentrated suspension ($\phi = 15\%$), it is the suspension of chain particles that offers the highest yield stress. However, for both concentrations, the yield stress of the suspension of spherical particles experiences a more rapid growth with the magnetic field. The higher yield stress of the concentrated suspension of polymerized chains ($\phi = 15\%$) is associated with a significant initial yield stress $\tau_Y \approx 20$ Pa in the absence of magnetic field. This yield stress can be explained by a solid friction between the chains in shear deformation that can appear even at apparently low chain volume fractions [10]. In contrast, there is no significant yield stress for the suspension of spherical particles at zero magnetic field.

In squeeze rheometry we observe a rather high dynamic yield stress for suspensions of polymerized chains in the absence of magnetic field (45 Pa for $\phi = 5\%$ and 80 Pa for $\phi = 15\%$). This dynamic yield stress is much higher than the one observed in shear rheometry. This suggests that the resistance of chain particle suspensions to squeeze deformation is much higher than to shear deformation. Similar to shear rheometry, we also observe an increase of the dynamic yield stress of both MR suspensions with the magnetic field, meaning field-induced structuring. For both volume fractions (5 and 15%), the suspensions of polymerized chains show higher values of τ_Y than the suspensions of spherical iron particles. However, at magnetic fields $H < 10$ kA m^{-1}, the yield stress τ_Y of the suspensions of spherical particles increases much faster with the field than that of the suspension of polymerized chains. We explain this by the higher magnetization of the spherical particles as compared to the polymerized chains, which contain a lot of polymer [1]. At higher fields, we observe a dramatic decrease of the dynamic yield stress of the suspensions of spherical particles, which is not the case for the suspensions of chain particles. To explain these observations we shall consider the squeeze flow of both suspensions.

3.1. Suspension of spherical particles (figure 3(a))

In the presence of the magnetic field, spherical iron particles become magnetized, attract each other and form chain-like aggregates extended in the direction of the magnetic field. The MR fluid becomes a Bingham one and, according to [9, 11], the squeeze flow contains two distinct regions—a shear zone and a central plug zone, as depicted in figure 3(a). In the plug zone, the radial component of the fluid velocity varies only in the radial direction, suggesting purely extensional

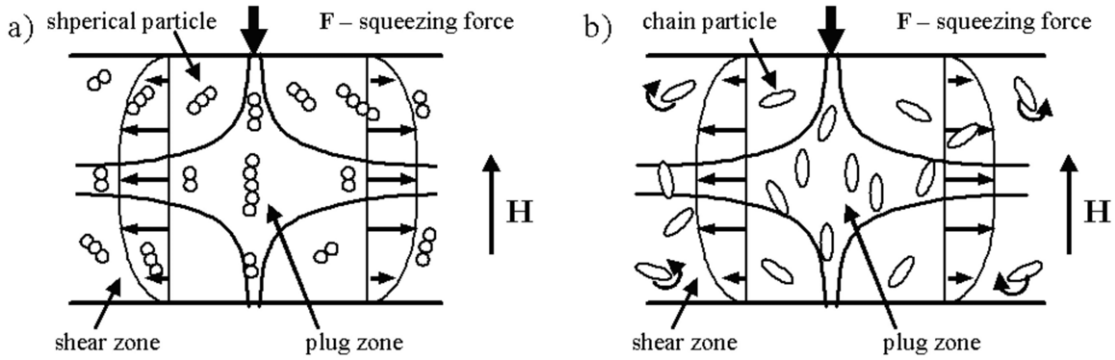

Figure 3. Squeeze flow of the suspension of spherical iron particles (a) and of the suspension of polymerized chains (b) in the presence of a magnetic field. Two flow zones coexist—a shear zone near the walls and a central plug zone of purely extensional flow.

flow [9]. The nature of the flow and the MR microstructure are different in both zones and are determined by a competition between hydrodynamic and magnetic interparticle forces. Such competition is described by the Mason number, which scales as $\mathrm{Mn}_{\mathrm{shear}} \propto \frac{\eta_0 \dot{\gamma}}{\mu_0 H^2}$ for shear flow and, by analogy, $\mathrm{Mn}_{\mathrm{ext}} \propto \frac{\eta_0 \dot{\epsilon}}{\mu_0 H^2}$ for extensional flow. Here, $\eta_0 = 0.32$ Pa s is the viscosity of the dispersing liquid (ethylene glycol with Aerosil 300 silica particles); $\mu_0 = 4 \times 10^{-7}$ H m^{-1} is the magnetic permeability of vacuum. In complex squeeze flow, shear rate $\dot{\gamma}$ and extension rate $\dot{\epsilon}$ fields are not homogeneous and their characteristic values scale as [12]: $\dot{\gamma} \propto \frac{\dot{h}R}{h_0^2} = \frac{2\pi \Delta h f R}{h_0^2}$ and $\dot{\epsilon} \propto \frac{\dot{h}}{h_0} = \frac{2\pi \Delta h f}{h_0}$. For the given range of experimental parameters, we estimated the order of magnitude of the Mason numbers in the squeeze flow of the MR suspension: $10^{-2} < \mathrm{Mn}_{\mathrm{shear}} < 10$ and $10^{-3} < \mathrm{Mn}_{\mathrm{ext}} < 1$. These relatively low values of the Mason numbers indicate that, in our experiments, magnetic interaction between particles either dominates over hydrodynamic interaction or is of the same order of magnitude, except for $\mathrm{Mn}_{\mathrm{shear}} > 1$. This suggests that, in squeeze flow, the MR fluid aggregates resist hydrodynamic forces and are displaced with the flow, being more or less aligned with the magnetic field.

3.2. Suspension of polymerized chains (figure 3(b))

In the presence of magnetic field, polymerized chains become aligned with the field and, as for spherical particles, can form aggregates containing a few chain particles. These aggregates are expected to be much longer than the aggregates of spherical particles. Such big aggregates cannot sustain large shear forces and, whereas the aggregates of spherical particles are completely destroyed at Mason numbers of the order of unity [13], the aggregates of polymerized chains should be destroyed at $\mathrm{Mn}_{\mathrm{shear}} \ll 1$. Therefore, we expect no aggregates in the shear zone of the squeeze flow. However, there should be some remarkable increase of the shear viscosity because the magnetic field alters the orientation distribution of the chain particles and makes them spend more time perpendicular to the flow. In the plug zone, in the absence of magnetic field, chain particles align with the flow, maximizing the extensional viscosity of the suspension. At low extensional Mason numbers, the magnetic field can maintain the chain particles perpendicular to the flow and, in contrast to shear flow, the extensional viscosity in the plug zone is decreased. However, in our experiments, in most cases the yield force F_{Y} is several times smaller than the squeezing force amplitude F_0 and the shear zone must dominate over the plug zone. Thus, the resulting effect is a field-induced increase of the viscous dissipation in squeeze flow.

3.3. Cavitation

An oscillatory squeeze flow consists of periodic compression and traction of the fluid film. Under traction, when the plates move apart, an inward radial flow occurs in the fluid film, provoking a pressure decrease inside the film. A pressure decrease can induce a vapor or gaseous cavitation and even an air entrainment into the fluid film and the formation of a bubbly fluid mixture. This is exactly what happens in squeeze-film dampers, affecting dramatically their load capacity [14]. The same phenomena are observed in traction of viscous fluids and adhesive materials [15]. In our case, cavitation/air entrainment must occur at some critical magnetic field corresponding to a critical traction force, at which the pressure decreases below a cavitation pressure. The dramatic decrease in the squeezing force amplitude of the suspension of spherical particles can be explained by fluid film inflation with air bubbles sucked from the ambient air or formed by evaporation of the liquid at overcritical magnetic fields. We do not have a clear explanation of why this does not happen to the suspension of polymerized chains. We can suppose that, under magnetic field, chain particles form much more deformable structures than spherical particles. This filament structure, spanning the gap, could better transmit the force between the upper and the lower disk, and prevent cavitation from provoking a rupture between the fluid and the disks.

4. Conclusions

The present work is devoted to the effect of the magnetic field on the yield stress of MR suspensions of polymerized chains, in oscillatory squeeze flow. Such flow appears to be rather complex, combining regions of both shear and extensional deformation. The dynamic yield stress of MR suspensions

is extracted from the extrapolation of force versus excitation frequency curves, using the lubrication approximation. Both in the presence and in the absence of magnetic field, chain particle suspensions offer much higher yield stress than spherical particle suspensions under squeeze. This is explained in terms of a solid friction between chain particles, which form more intricate aggregates. Due to the stronger magnetization of spherical iron particles, the yield stress of the suspension of spherical particles experiences a more rapid growth with the magnetic field than that of the suspension of polymerized chains. An abrupt decrease in squeezing force amplitude with increasing magnetic field is observed for the suspension of spherical iron particles at magnetic fields higher than 14 kA m^{-1}. Surprisingly, this phenomenon, attributed to cavitation or air entrainment, does not occur in the suspension of polymerized chains, at least at magnetic fields $H < 30$ kA m^{-1}. This could make suspensions of elongated magnetic particles preferable for use in squeeze-film dampers.

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References

[1] López-López M T, Vertelov G, Kuzhir P, Bossis G and Duran J D G 2007 *J. Mater. Chem.* **17** 3839
[2] Bossis G, Volkova O, Lacis S and Meunier A 2002 *Ferrofluids* ed S Odenbach (Berlin: Springer) p 202
[3] Tang X, Wang X J, Li W H and Zhang P Q 1998 Electro-rheological fluids, magneto-rheological suspensions and their applications *Proc. 6th Int. Conf.* ed M Nakano and K Koyama (Singapore: World Scientific) p 870
[4] Bashtovoi V, Kabachnikov D and Bossis G 2002 *J. Intell. Mater. Syst. Struct.* **13** 515
[5] See H 2003 *Rheol. Acta* **42** 86–92
[6] Viera S L, Ciocanel C, Kulkarni P, Agrawal A and Naganathan N 2003 *Int. J. Veh. Des.* **33** 36–49
[7] Covey G H and Stanmore B R 1981 *J. Non-Newton. Fluid Mech.* **8** 249
[8] Matsoukas A and Mitsoulis E 2003 *J. Non-Newton. Fluid Mech.* **109** 231
[9] Engmann J, Servais C and Burbridge A S 2005 *J. Non-Newton. Fluid Mech.* **132** 1
[10] Servais C, Manson J-A E and Toll S 1999 *J. Rheol.* **43** 991
[11] Scott J R 1931 *Trans. Inst. Rubber Ind.* **7** 169
[12] Macosko Ch W 1994 *Rheology Principles, Measurements, and Applications* (New York: Wiley–VCH) pp 270–4, 297–303
[13] Volkova O, Cutillas S and Bossis G 1999 *Phys. Rev. Lett.* **82** 233
[14] Diaz S and San Andrés L 2001 *J. Tribol.* **123** 5
[15] Gay C and Leibler L 1999 *Phys. Rev. Lett.* **82** 936