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MAGNETORHEOLOGY OF A MILLIMETRIC STEEL SPHERES SUSPENSION

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The magnetorheology of a "suspension" of millimetric-size steel spheres in oil has been studied. After a precise measurement of the magnetic properties of the particles we show that the yield stress is well characterized and that it can be predicted by a finite element method and a non-affine model where the chains break in the middle. Some experiments in microgravity confirm that the sedimentation does not alter our results and also shows that the magnetic stress decreases with the shear rate.

1 Introduction

The prediction of the strength of an electrorheological or a magnetorheological suspension as a function of the applied field relies on the prediction of its yield stress. Many models, based on the calculation of interparticle forces, can give the right order of magnitude in some cases and fail in other ones. For instance a full three-dimensional model taking into account multipolar interactions up to the 50^{th} order [1] is correct at low fileds but it will always predict a E^2 behavior which is not found experimentally in most cases due to the field limitation in the interparticle gap [2],[3]. In magnetorheological suspensions the situation can be easier to model since there is no charge transfer to deal with and calculations based on magnetic saturation of poles give the right order of magnitude for the yield stress [4],[5].In a magnetic material the permeability does not remain constant with the field and a precise estimation of the force between particles can only be obtained numerically by using finite elements[5],[6]. Even in this case some approximations are needed to avoid a full 3D calculation which would be very time consuming. The experimental situation is often not better as long as we want to characterize the particles (broad size distribution, existence of an oxide layer, difficulty to access to the bulk permeability of the particles, unknown structure, slippage on the walls[7]. In order to control most of the experimental parameters, we have used a suspension of monodisperse steel spheres with a diameter of one millimeter. Our aim in this paper is to examine the validity of the yield stress theories by comparing their predictions with the experimental results obtained on a reference "suspension". In a first section we shall describe our permeability measurements, then our yield stress measurements. As the sedimentation could be a factor of error in our experiments, we shall present in the last section some results obtained in microgravity.

2 Permeability of suspension

We have first measured the bulk permeability of the particles by using a rod of the same steel in an double coil system called hysteresimeter. As this steel presents a small hysteresis ,we have taken the average curve of magnetization. The curve is well fitted by a Frolisch- Kennelly law[8] with, as parameters, the initial permeability: $\mu_i=1+\chi_i$ and the saturation magnetization, Ms: $M=\chi_i H/(1+\frac{\chi_i}{M_s}H)$

A good agreement is obtained with M_s=1360 kA/m and μ_i=250. We have tested our finite element method and the consistency of these values by doing two other experiments one on the permeability of a chain of sphere and the other on the rupture force between two particles.

2.1 Permeability of a single chain of a steel spheres

First of all let us note that for such high permeabilities (μ_i*=250* for initial permeability and still μ_p*=90* for H=10kA/m), the Maxwell-Garnett theory for predicting the permeability, $\overline{\mu}$, of a suspension of spheres: $\frac{\overline{\mu}-1}{\overline{\mu}+2}=\Phi\cdot\beta$ with $\beta=\frac{\mu_p-1}{\mu_p+2}\approx1$ gives a large uncertainty Furthermore the suspension is not isotropic as it would supposed to be in this model. In order to get the average permeability of the suspension, that we need to calculate the average field: $H=H_0/\overline{\mu}(H)$, we better have to start with the permeability of a chain of spheres. We shall consider 6 spheres because it is the number of particles between the two walls of the cell. We have placed 6 spheres in a plastic tube and measured their permeability.

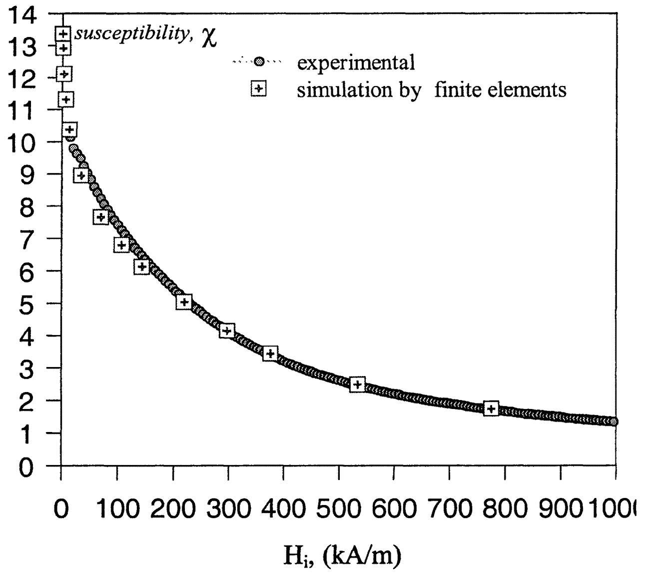

Fig.1 Comparison of the measured permeability of a chain of six spheres with the numerical predictions by finite elements using M_s=1360 kA/m and μ_i=250

The magnetization curve of this chain was obtained in the hysteresimeter and in the vibrating sample magnetometer (VSM). Figure 1 shows the susceptibility $\chi_{ch}=\mu_{ch}-1$ of a chain versus the magnetic field H_i inside the chain. The magnetic field inside and outside the chain are related by $H_i = \dfrac{H}{1+N_d \cdot \chi_{ch}(H_i)}$ with N_d=0.017 for the equivalent length/diameter cylinder [8]. The crosses are the predictions of a finite element calculation using as input the permeability of bulk steel fitted with the Frohlish-Kennely law. It is quite interesting to note that the initial susceptibility of a chain is an order of magnitude less than the one of a bulk steel. This is related to the fact that for a given average internal field H_i, the real field can be very large in the region of contact of pairs of particles inside the chain which greatly decreases the permeability. On one hand it is a good test of our numerical method and on the other hand it also allows to calculate the average permeability of the suspension by stating that the magnetic field H_{ch} acting on the chain and average field H in the suspension are equal. Then knowing the magnetic moment of a chain, $m_{ch}(H)$, the magnetization of the whole system of chains is: $M=\chi(H)H=N_{ch}\, m_{ch}/V = (3\Phi/2\, \pi\, a^2)\, m_{ch}$ where Φ is the volume fraction of the suspension. The average permeability of the suspension is represented in Fig.2a.

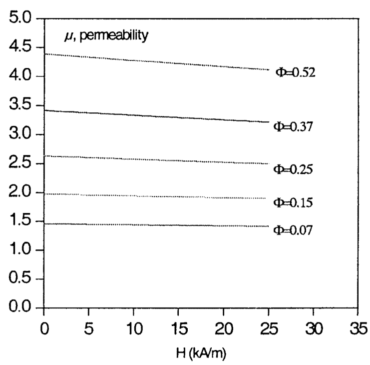

Fig. 2a: Permeability of the suspension for different volume fractions versus the field inside the suspension

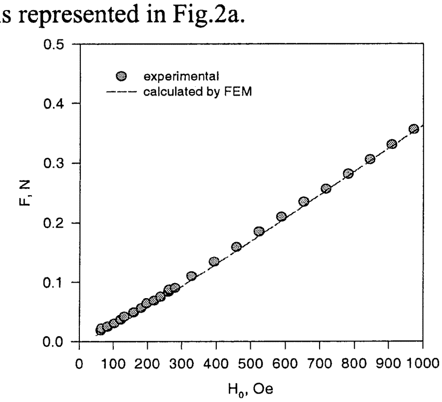

Fig.2b. Rupture force F(Newton) between two steel spheres versus external field

The relation between the applied field H_0 and the field inside the suspension H is obtained by taking into account the permeability of the suspension and the demagnetization factor of the experimental cell.

We have also calculated the rupture force between two spheres and measured it by attaching one sphere to a wire below the plate of an electronic balance and pulling on the other for different intensities of the magnetic field. The comparison between the experiment and the numerical prediction of the rupture force is also very good as can be seen in Fig. 2b.

3 Yield stress of the system of steel spheres

Two methods are often used for the experimental definition of the yield stress:

1) Measurements of the stress as a function of the strain. In this case we progressively increase the applied stress (or strain) and we record $\tau(\gamma)$ up to strains of a few unities. The stress corresponding to a plateau of the stress-strain plot will define the yield stress τ^y

2) We can also find the yield stress with the help of the flow curves. Here we obtain the applied shear stress as a function of the shear rate $\tau(\dot{\gamma})$. Then by the extrapolation of the curve towards $\dot{\gamma}\rightarrow 0$ we can obtain the yield stress τ^y.

The yield stress of a model suspension of steel spheres (*1mm* and *2mm* diameter) has been studied in the parallel plate geometry of the Carri-Med CSL-100 rheometer. In order to avoid any kind of slipping, a layer of spheres was glued on to each plate of the measuring cell. The number of spheres fixed on the plates has been adjusted as a function of each studied volume fraction. The two methods of yield stress measurements were used and they gave the same (within 5% of error interval) values of the yield stress. Figure 3 shows a characteristic flow curve obtained for a volume fraction Φ=15% in an external field H=31(kA/m). The dots connected by a line represent the values of the flow curve (shear stress versus shear rate) obtained by a slow increase of stress and the single big point corresponds to the yield stress obtained with the stress-strain measurements. We see that there is a good agreement between the extrapolation of the flow curve at zero shear rate and the stress-strain value. The hysteresis between the charge curve (increasing the stress) and the discharge curve (decreasing the stress) could be due to sedimentation since, when the structures are breaking, we can suppose that each sphere can fall a little bit. Actually the cell is transparent and we do not observe a sedimentation because the magnetic forces are much higher than the weight of the particles. This hysteresis is often observed even with colloidal suspensions and is due to a change of structure with the shear[8];we shall confirm this interpretation in the next section.

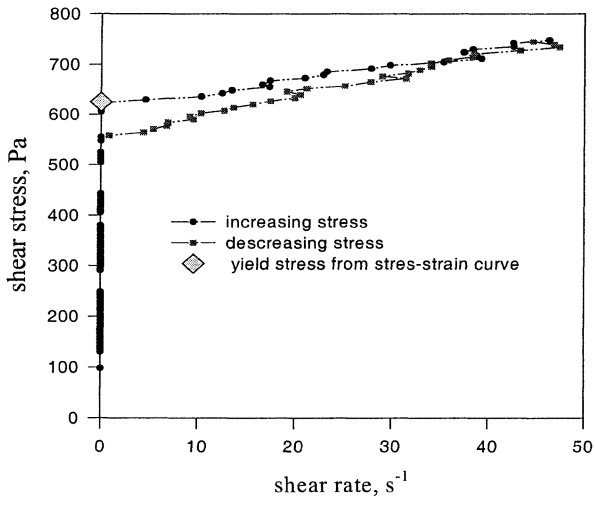

Figure 3. Flow curve for Φ=15% suspension and the yield stress found from stress-strain curve

Different volume fractions:Φ=0.07; 0.15; 0.25; 0.37; 0.52 were studied over a quite large range of applied fields.

Stress-strain measurements were also carried out in order to obtain the yield stress and simultaneously, the critical strain. The measurements were made in stress control mode and in the situation of equilibrium. That means that we set the stress and we measure the strain; then we increase the stress again only after the stabilization of the strain. It is quite easy to find the yield stress from the stress-strain curves. On the contrary defining the critical strain is more difficult since it changes with the position inside the cell: at the centre of the plates the strain is always zero, it increases with the distance from the centre of the plates and reaches its maximum at the extremity *(r=R)*. The other difficulty in the critical strain definition comes from the fact that the chains, which have been broken once, re-form themselves with the other pieces of broken chains. We have compared the experimental results with the finite elements calculations using the standard cubic network of chains. If we know the interparticle shearing force F_x between two particles, then we can find the shear stress as: $\tau=\frac{N_c}{S}F_x=\frac{\Phi}{(2/3)\pi a^2}F_x$ where Φ is the volume fraction of solid particles and N_c/S the number of chains per unit surface. The projection of the interparticle force on the shear direction increases with the strain, passes through a maximum and then falls down. The maximum of the force corresponds to the rupture and the corresponding stress is the yield stress. On the other hand the model is based on an initial structure made of a cubic lattice of simple chains which is affinely strained. The only input is the permeability of the bulk steel as a function of the field.

The test of the chain model is done by comparing the dimensionless calculated and measured yield stress, $\tau/\mu_0 H^2$, (where H is the field inside the suspension) to the experimental one. This comparison is shown in Figure 4 for the volume fraction Φ=0.15 as a function of the internal average field H. The upper solid curve corresponds to the assumption that all the particles in the 6 particles chain are separating at the same time. On the contrary in the lower curve (long-dash line on the graph) we impose that the chain breaks only at its center. We see that the predicted values are strongly dependent on the assumption concerning the way the particles separate.

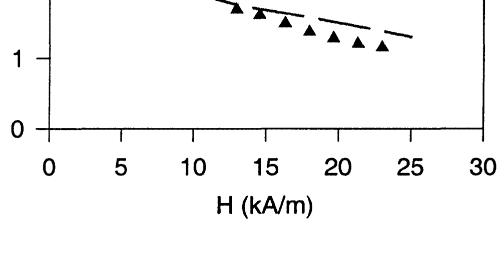

Figure 4. Normalised yield stress as a field function for Φ=15%

The experimental values are represented by the triangles and are normalized by the average field $H = H_0/\overline{\mu}(H)$. The agreement with the scheme where the chains break at the middle correspond to what we observe in the cell. Actually the affine motion of the chains gives an overestimation of the yield stress because, for a same strain the gaps are distributed among each pair of particles ,hence the distance between two particles does not decrease as quickly as in the case of one single gap.

In these experiments with millimetric spheres the liquid plays a role in the viscous dissipation since the stress increases with the shear rate as can be seen in Fig.3. It is interesting to know if the hypothesis that the magnetic stress remains constant when we increase the shear rate (Bingham model) is a good one or not. We have checked this point by doing the same experiments in microgravity (parabolic flights with 20s of microgravity) on a gas of steel spheres.

4 Rheology of magnetic spheres in microgravity

The measurements were carried out with imposed ramp of velocity on the viscosimeter HAAKE-20 in the parallel plates geometry both in normal gravity and in microgravity for different values of the magnetic field. In the results plotted in Fig.5 we see that we have two regions. In the first part for shear rates between 0 and $200s^{-1}$ we see a decrease of the stress when the shear rate is increasing. We can say that, once the chains have been broken the average distance between the particles is increasing and, as the magnetic interparticle force decreases with the distance, we observe this decrease of stress, which is not compensated by an increase of viscous dissipation since we have no liquid. Actually there is a small dissipation due to the inelasticity of the collisions between the steel spheres.

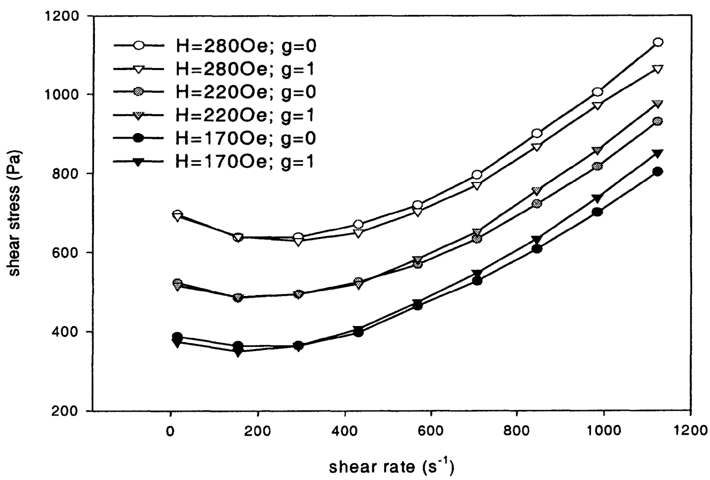

Fig.5. Comparison between the results obtained in microgravity (g=0) and in normal gravity (g=1)

This collisional dissipation dominates at higher shear rates and gives the strong increase of shear stress in the second part of the graph. We have verified that the sedimentation was not the cause of this anomalous behavior (decrease of stress when increasing the shear rate) by comparing experiments done in microgravity and in normal gravity. We can see that the difference between normal gravity and microgravity is very small below $400s^{-1}$ which confirms that sedimentation does not play a role in our experimental conditions, so we can trust the yield stress values we have measured as well as the existence of a true hysteresis as shown in Fig.3

Aknowledgements

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References.

1. H.J.H Clercx, and G. Bossis "Many-body electrostatic interactions in electrorheological fluids" *Phys.Rev E* 48, 2721 (1993)
2. L.C.Davis "Ground state of an electrorheological fluid", Phys Rev A, **46**,R719(1992)
3. N.Felici,J.N.Foulc, and P. Atten, Electrorheological Fluids, edited by R.Tao and G.D.Roy (World Scientific,Singapore, p 139-(1994)
4. J.M. Ginder and L.C. Davis "Shear stresses in magnetorheological fluids:role of magnetic saturation", *Appl. Phys.Lett.*, **65**,3410 (1994);
5. J.M. Ginder , L.C. Davis and L.D.Elie. 1995 "Rheology of magnetorheological fluids:Models and measurements" Proc, 5th International Conf. Electro-Rheological Fluids, Magneto-rheological Suspensions and associated Technology, Univ. of Sheffield, UK, July 1995, 10-14
6. S.Lacis, O.Volkova, G.Bossis, in Modelling of Material Processing- Proc. Int. Colloq. (University of Latvia,Riga, p 122,1999)
7. O.Volkova, PhD Thesis, University of Nice, (1998)
8. D. Jiles, Introduction to magnetism and magnetic materials, Chapman&Hall, London, (1995)
9. 4. G.Bossis, E.Lemaire, O.Volkova, H Clercx, Yield stress in MR and ER fluids: a comparison between microscopic and macroscopic structural models, *J. of Rheology*, **41** 687 (1997)