Discontinuous shear thickening in magnetorheological suspensions

G. Bossis[1], A.Ciffreo[1], Y.Grasselli[2], and O.Volkova[1]

[1]University of Nice Sophia-Antipolis, Institut de Physique de Nice,UMR7010,Parc
Valrose,06108 Nice Cedex 2
[2]Skema Bachelors,60 rue Dostoievski,BP085,06902 Sophia-Antipolis,France

ABSTRACT

The discontinuous shear thickening (DST) transition is studied in magnetorheological suspensions (MRS) with the advantage to control the transition with a magnetic field. We first show the difference of behavior between an usual MRS and this new kind of fluid. The critical stress increases with the value of the magnetic field and an explanation of this behavior is proposed. Some results obtained in microgravity during parabolic flights demonstrate the sensitivity of this transition to small changes of density.

INTRODUCTION

At high enough volume fraction some suspensions of microparticles show a discontinuous shear thickening (DST) which happens when the applied stress is large enough to overcome the repulsive barrier preventing the aggregation between particles[1-3] such as a ionic layer or a coating polymer. If the suspension is flowing under an applied stress then, above a critical one, the flow can stop suddenly which is quite undesirable in some industrial processes like the pumping of concrete. On the contrary, if this phenomenon can be controlled, it gives birth to an efficient two-state material for on-off applications in force or torque transmission. We recently demonstate[4-5] that such a fluid can be made using carbonyl iron particles and some superplastifier molecules used in cement industry[6]. In this presentation we shall first compare the usual behavior of a MRS with a typical volume fraction Φ =45% with the one of a very concentrated one at Φ=64%. This very high volume fraction is obtained thanks to the use of a superplastifier molecule called polyphosphonate polyoxyethlene(PPP). This molecule has a head made of two phosphonate groups (PO_3H^-) and an hydrophilic tail made of a polyoxyethylene chain. In a second part we shall present experiments made in microgravity during parabolic flights in order to check the effect of the sedimentation on the transition.

HIGH VERSUS MIDDLE VOLUME FRACTION AT IMPOSED STRESS

The experiments were performed with suspensions made of commercial carbonyl iron particles (CIP grade HQ from BASF) dispersed in ethylene glycol with the addition of the PPP molecule at a mass fraction of 0.2% relatively to the mass of the particles. The rheological curves are obtained in plate-plate geometry on a MCR502 from Anton Paar with serrated plates and a serrated plateau in order to prevent the slipping of the suspension on the walls. The stress was increased at a rate of 20Pa/min for which we do not observe a thixotropic behavior. The magnetic field is perpendicular to the plates and is applied with the help of a coil surrounding the sample. It is worth noting that in this configuration, the magnetic field inside the sample is lower than the external one: $H_i=H_e/\mu(H_i)$ because of the demagnetizing field. In Fig.(1) we have presented a set of curves showing the shear stress versus the shear rate for five different amplitudes of the magnetic field.

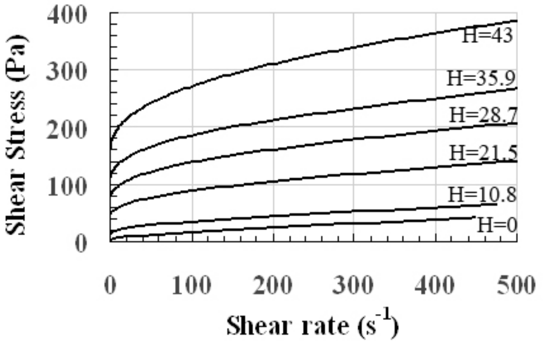

Figure 1. Volume fraction Φ=0.45; the magnetic field H is in kA/m

At the lowest values of the field the rheology follows approximately a Bingham law:

$$\tau = \tau_y + \eta_p \dot{\gamma} \quad (1)$$

with for H=0 a plastic viscosity η_p=0.080 Pa.s and a yield stress τ_y=8.9Pa. Then at higher field, the curve is better represented by a Herschel-Buckley law:

$$\tau = \tau_y + K\dot{\gamma}^n \quad ....(2)$$

With for instance for the largest field: H=43kA/m τ_y=114.1Pa , K=38.7, n=0.31. One can see that this behavior differs from the standard Bingham model where all the curves have the same plastic viscosity, meaning that the average stress induced by the field does not depend on the state of the flow, or in other words that the relative trajectories of the particles remain the same when we are above the yield stress. In order to emphasize this difference we can compare the differential viscosities,which is the slope of the curve, η_d, around a shear rate of 500s^{-1}. We have η_d= 0.08Pa.s for H=0 but η_d=0.19Pa.s for H=43kA/m., so a factor of 2.37 between these two fields.

Now we are going to see what is the rheology at high volume fraction, for Φ=0.64 still with the same proportion of 0.2% of the PPP molecule. The result is shown in Fig.2 for five different fields.

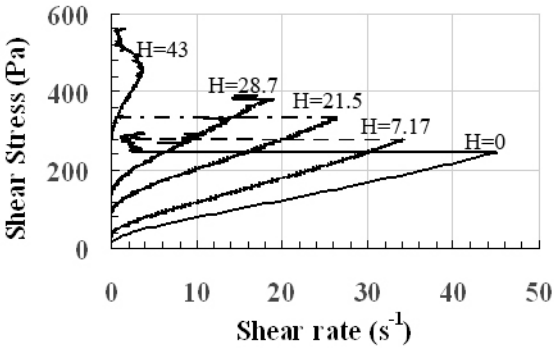

Figure 2. Volume fraction Φ=0.64; the magnetic field H is in kA/m

The first major difference is that now we have a discontinuous transition with a strong decrease of the shear rate. At H=0 this transition happens for a shear rate of 45 s^{-1} and a critical stress of 246Pa. When the field is increased the critical shear rate strongly decreases; for instance it is equal to 3.5 s^{-1} for a field H= 43kA/m and the critical stress is now σ_c=440Pa. We must emphasize that this increase of the critical stress with the field is not expected, because this is the stress (and not the shear rate) which controls the onset of the jamming transition. The explanation is that, above a critical stress, the repulsive force between two particles ,here due to the molecule PPP, no longer resists to the applied stress and the particles come into frictionnal contact, which triggers the transition. In our situation the applied stress is splitted in a magnetic and an hydrodynamic stress, but the total critical stress should remain the same. Nevertheless there is a different kind of action, since the magnetic force between two particles is mainly a radial force whereas the hydrodynamic force has both a radial and a tangential component. The radial component due to the magnetic force prevails on the hydrodynamic one when the field increases and the consequence is a more important entanglement of the polymer layers in the space between two particles especially at high volume fraction where the average gap between the particles becomes smaller. It appears as the increase of the differential viscosities with the magnetic field. For example we have η_d= 5.86 Pa.s for H=0 and 41.5 Pa.s for H=43kA/m, that is to say an increase by a factor of 7.08. For the same fields the increase of η_d was only by a factor of 2.37 at Φ=0.45. If it is the tangential hydrodynamic stress which must reach a critical value to sweep the polymer out of the gap between the particles, then the yield stress which characterizes the effect of the field does not intervene and the value of the hydrodynamic stress (as given by η_d times the critical shear rate) appears to be approximately constant around 200Pa.

Until now the experiments were performed at imposed stress, we are now going to see what happens at imposed shear rate. In order to reduce the role of the sedimentation which could greatly modifies the jamming transition, we have made some experiments in microgravity during parabolic flights.

JAMMING TRANSITION AT IMPOSED VELOCITY IN MICROGRAVITY

When, in the imposed stress mode, the shear rate decreases during a ramp of stress, it means that, if we are in the imposed velocity mode, we shall have a jump of stress when we reach the critical shear rate. That is actually what we observe and the jump of stress is so strong that it overcomes the maximum torque of the rheometer of 0.3N.m. On the other hand in the imposed stress mode, if we increase the stress well above the critical one, the particles are then expelled out of the suspending fluid[7] with, as a consequence, an artificial decrease of the viscosity. In order to avoid these two drawbacks, we have designed a homemade rheometer based on the high torque rheometer (10N.m) developped by CAD company for the rheology of concrete. The cell, represented in fig.3 is closed by a lip seal in order to avoid the leakage of the suspension at high stress.

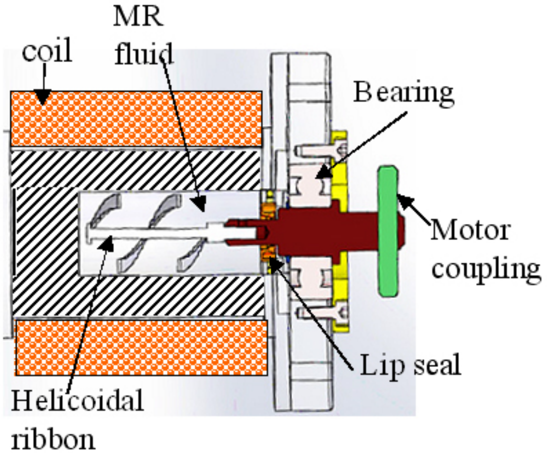

Figure 3. Sketch of the cell installed on the high torque rheometer

The geometries we used are either a double helicoidal ribbon[8] or a vane geometry in order to prevent the slippage of the suspension on the wall. Furthermore the outer cylinder was serrated for the same reason.

As sedimentation can also change the conditions of the jamming transition, we have used the opportunity of parabolic flights offered by the CNES to realize experiments in microgravity to see in which extent the pressure due to gravity can play a role on the jamming transition[9]. A comparison of two experiments made one after the other in an interval of approximately 1mn is represented in Fig.4. In this experiment a ramp of shear rate between 0 and 1.6 s^{-1} was applied during 2s then the shear rate was kept constant. The ramp was applied just at the beginning of the microgravity period which is of 22s. A field of 19.2kA/m was applied at the same time. The resulting curve is the one with the solid line which grows up to 170kPa and then decreases slowly with time.

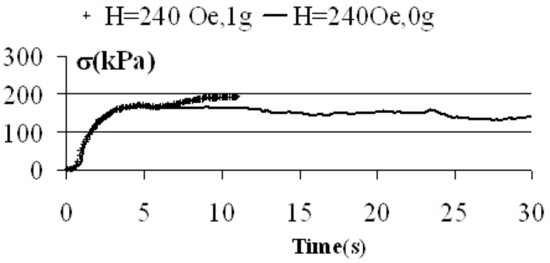

Figure 4. Vane geometry. Shear stress versus time during a ramp of shear rate. The upper curve is in microgravity, the lower one in 1g. H=19.2kA/m (240Oe)

The second curve with crosses was recorded in the same conditions immediately after but in normal gravity between two parabolas. We see that the rising of the stress is the same but, once the maximum the stress is reached, it continues to rise and the experiment stops because the maximum torque was reached. It appears that in microgravity the final stress is lower than in normal one. This effect was confirmed in several experiments.

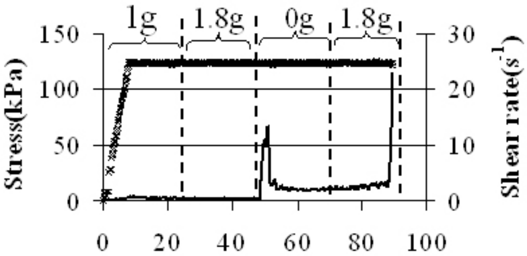

Fig.5 Lower curve: stress versus time, Upper curve (right scale): shear rate versus time. The dotted lines separate the different values of gravity during a parabolic flight. Field amplitude: H=17.6kA/m turned on at the start of the 0g period.

An other demonstration of the effect of the gravity on the triggering of the jamming transition is illustrated in Fig.5. In this experiment we have used the helicoidal geometry of diameter 24mm and the shear rate was kept constant at a value of 24 s^{-1-} (corresponding to a rotational velocity of 20rpm for a gap of 1mm) after a rising ramp of 8s, a field of 17.6 kA/m (220Oe) was applied just at the beginning of the microgravity period and kept constant. We see a peak of stress at a value of 60kPa when the field is turned on but followed by a quick decrease towards a plateau of 10kPa , then an abrupt increase which overcomes the maximum torque of the motor and triggers its shutdown. This sudden increase occurs during the period of hypergravity (1.8g) and this is not a coincidence because it was observed several times. It means that, despite the rotation of the geometry at 20rpm which prevents the sedimentation, the larger pressure due to the extra weight of a column of particles of height h: $\Delta P=138$ Pa ($\Delta P=\rho\ \Delta g\ h$ with h=1mm, $\rho=7.8g/cm3$, $\Delta g=1.8x9.81$) is enough to trigger the transition.

CONCLUSION

The possibility to modulate the jamming transition in a supension of iron microparticles by the application of a magnetic field is an opportunity to get a better understanding of this phenomenon. The increase of the differential viscosity with the applied magnetic field before the jamming transition indicates that the layers of polymer adsorbed on the particles interpenetrate each other more and more. The hypothesis that it is the tangential hydrodynamic stress which is responsible for the jamming transition and not the total stress, can explain the increase of the critical stress with the magnetic field even if additional experiments are needed to confirm this point. Finally, experiments made in microgravity emphasize the sensitivity of the jamming transition to the gravity force.

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