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# Control of discontinuous shear thickening with the help of a magnetic field

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## Résumé :

*Des suspensions très concentrées de particules de fer carbonyle dans l'eau peuvent être obtenues grâce à l'utilisation de superplastifiants utilisés dans l'industrie du ciment. Ces suspensions ont un simple comportement rhéoépaississant quand on augmente le taux de cisaillement en l'absence de champ magnétique. Quand l'expérience est menée à taux de cisaillement constant on peut déclencher la transition de blocage en appliquant un faible champ magnétique. On observe alors un très grand accroissement de la contrainte pour un champ magnétique critique dont la valeur décroît en raison inverse du taux de cisaillement appliqué. En utilisant un viscosimètre à fort couple on montre qu'on peut obtenir dans le régime de blocage une contrainte constante supérieure à 200kPa pour de faibles champs magnétiques. On comparera cette transition à celles observées dans d'autres suspensions de particules non magnétiques où on a utilisé le même superplastifiant. L'importance des contraintes obtenues et le fait de pouvoir contrôler la transition de blocage avec de faibles champs permet d'envisager de nouvelles applications des fluides magnétorhéologiques.*

## Abstract : (16 gras)

*Highly concentrated suspensions of iron particles in water can be obtained thanks to the use of superplastifier molecules which are currently used in concrete industry. These suspensions are usually shear thickening when the shear rate is increased in the absence of magnetic field. When the experiment is conducted at constant shear rate the jamming transition can be triggered by the application of a small magnetic field. We then observe a huge increase of the stress for a critical critical shear stress whose value decreases as the applied shear rate is increased. Using a high torque viscosimeter we show that in the jamming domain a constant stress higher than 150kPa can be obtained for low magnetic fields. We shall compare this transition to the one which are observed in other suspensions of non magnetic particles where we have used the same superplastifier. The fact that high stress can be triggered by low magnetic fields opens the way to new applications*

**Mots clefs : rhéoépaississement ; particules magnétiques ; suspensions**

## 1 Introduction

Shear thickening occurs frequently in suspensions of non colloidal particles and its origin lies essentially in the formation of clusters of particles. These clusters are usually formed progressively when the hydrodynamic stress generated by the shear flow dominates the short range repulsive forces between particles. The prerequisite is that, at rest, the particles are prevented from aggregation either by the presence of an ionic double layer or more often by a polymer or a surfactant adsorbed on the surface of particles. At some point, when the applied shear stress overcomes a certain limit, the surface of the particles can come in frictional contact, either due to the expulsion of the adsorbed layer from the contact zone or due to the forced interpenetration of the polymer layers. The propagation of the network of frictional contacts in the entire cell causes a "dynamic jamming" which is called discontinuous shear thickening (DST); this phenomenon was well reproduced and analysed by numerical simulation [1]. In a rheogram where a ramp of stress is imposed a sudden decrease of the shear rate will be observed or if it is an imposed ramp of shear rate; the transition will appear as a sudden jump of stress. A well known suspension which shows this kind of behavior is the cornstarch suspension [2],[3] but many other systems based on silica [4],[5], gypsum [6], poly-methylmethacrylate [7], calcium carbonate [8],[9] etc.. also present the DST phenomenon. We have recently shown [10] that suspensions of carbonyl iron, currently used in the formulation of magnetorheological fluids, can also present a very strong DST transition and above all that this transition can be triggered with a magnetic field. The high volume fraction necessary to observe this transition was obtained by coating the iron particles with a superplasticizer currently used in cement industry. In this paper we shall briefly compare the DST transition observed with this same molecule in suspensions of calcium carbonate and iron in water, then we shall present some experimental results illustrating how the DST transition can be triggered by a magnetic field both in stress imposed and shear rate imposed mode.

## 2 DST in aqueous suspension of iron and calcium carbonate

The superplasticizer molecule was made of a polyoxyethylene chain of 44 units with an attached head consisting of two phosphonate groups that we shall call PPP44. The counterions of the phosphonate group  $\text{PO}_3\text{H}$  are sodium ions. The adsorption of this molecule on the surface of calcium carbonate or of iron is due to interactions between ions of opposite charges, namely  $\text{PO}_3\text{H}^-$  with positively charged calcium or iron atoms on the surfaces of the particles. The particles used were respectively calcium carbonate from Omya with an average diameter of  $5.5\mu\text{m}$  and carbonyl iron (grade HS from BASF) with average diameter  $1.2\mu\text{m}$  and the suspending fluid was water. In both cases the mass fraction of the superplasticizer PPP44 was 0.2% of the mass of particles; for  $\text{CaCO}_3$  it corresponds to the plateau of adsorption as measured by total organic carbon. For iron particles in water we do not observe an adsorption plateau but the rheology did not change at higher mass fraction of PPP44. The experiments were made in plate-plate geometry with sand paper stucken on the plates to avoid slipping. In fig.1 is reported the stress versus shear rate during a ramp of imposed shear stress at the rate of  $20\text{Pa}/\text{mn}$  for  $\text{CaCO}_3$  and iron particles. In both cases we have a strong DST with a sudden and large decrease of the shear rate. Besides this common point, we see that the PPP44 molecule is less efficient to reduce the yield stress, since it is  $25\text{ Pa}$  for iron at a volume fraction of 62% compared to something below  $1\text{Pa}$  at 68% for  $\text{CaCO}_3$ . We must nevertheless keep in mind that the iron particles are spherical and that their polydispersity is smaller than with  $\text{CaCO}_3$  which contributes to lower the volume fraction under which it can flow without dilatancy. An other difference is that the domain of

shear thickening appears only above 20 s<sup>-1</sup> with iron suspension whereas the CaCO<sub>3</sub> suspension shear thickens from the beginning.

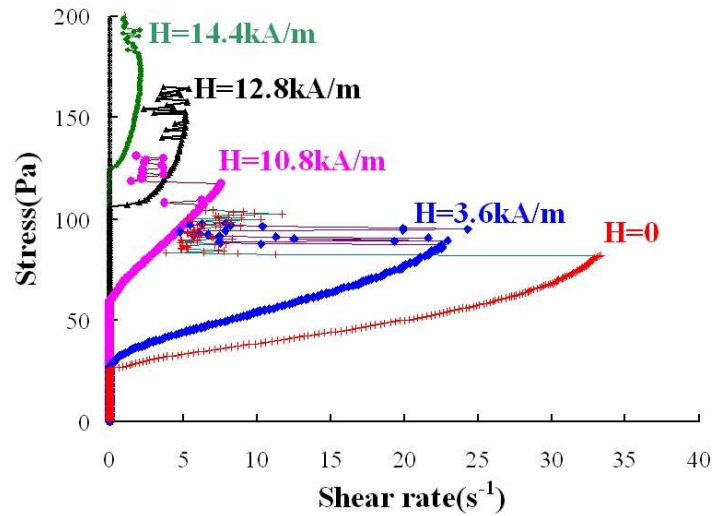
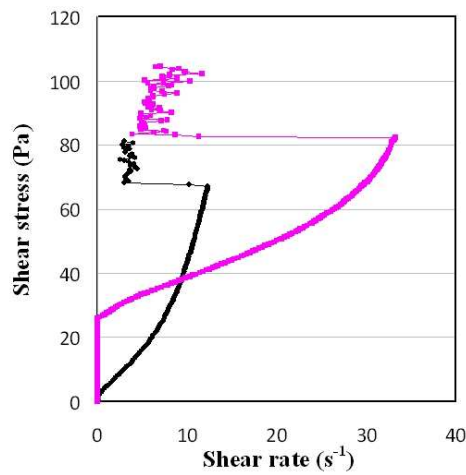


FIG.1 Ramp of stress for suspension of CaCO<sub>3</sub> in water at  $\Phi=68\%$  (black dots) and iron in water at  $\Phi=62\%$  (purple squares)

### 3 Influence of magnetic field on the DST

The application of a magnetic field on a suspension of magnetic particles results in the magnetorheological effect, that is to say, it increases the yield stress. In our experiment the magnetic field is perpendicular to the plane of the disks We can actually see in fig.2 that, increasing the magnetic field, we increase the yield stress. As the fields we have used are quite low (the maximum field of 14.4kA/m corresponds to an induction in vacuum of 0.018 Tesla) the yield stress does not increase a lot but nevertheless, the effect on the critical shear rate of the DST transition is very important since it decreases from 33 s<sup>-1</sup> to 2 s<sup>-1</sup>. It means that with a low field we are able to control the DST transition on the entire range of shear rate. An other significative result is that the critical stress is not constant, as it should if we consider that the DST transition corresponds to the applied stress which is needed to overcome the repulsive force produced by the adsorbed polyelectrolyte [11],[12],[9].

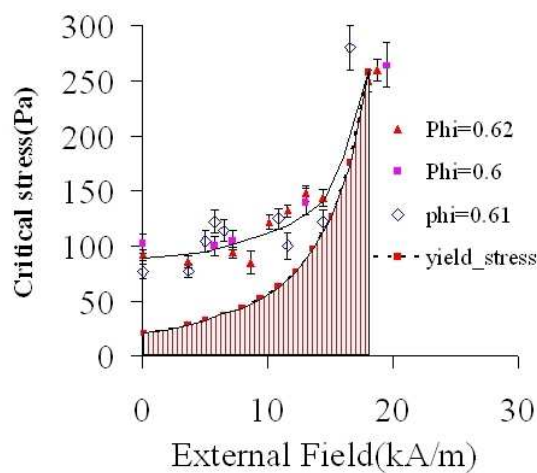


FIG.3 Critical stress versus magnetic field at three volumes fractions

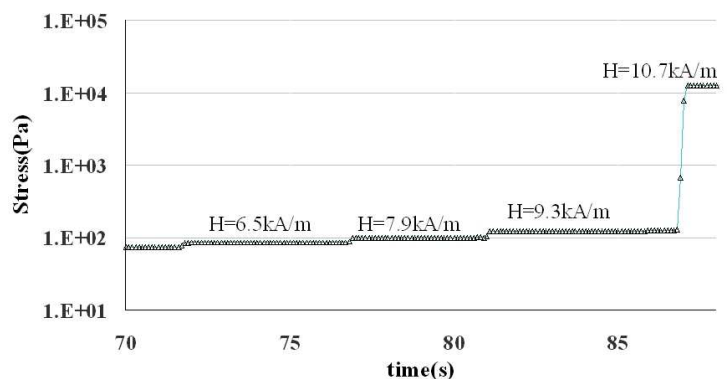


FIG.4 Increase of field step by step at a constant shear rate of 10s<sup>-1</sup>

If we look to the evolution of the critical stress with the applied field, which is plotted in fig.3, we see that it does not depend significantly on the volume fraction, but that it increases quite strongly with the magnetic field, especially for the highest magnetic field at a shear rate close to zero. The dotted red line is a fit of the yield stress, which is the magnetic stress plus the zero field yield stress coming from residual attractive forces:  $\sigma_Y(H)$ . The upper solid line is to guide the eye through the experimental points. The difference between the two lines is the hydrodynamic stress at the critical shear rate:  $\sigma_h^c$ . The important point is that the angular dependence of the hydrodynamic stress is different from the one of the magnetic stress: the hydrodynamic stress is maximum along the compression axis, at  $45^\circ$  from the perpendicular of the walls, whereas the magnetic force is maximum when the particles are aligned with the field, that is to say at  $0^\circ$ . A chain of forces on the compression axis will be much more efficient to block the rotation of the aggregates in the shear flow than a chain of forces perpendicular to the walls since the magnetic pressure on the walls will likely not prevent the aggregate to slip and deform. Although this effect is difficult to estimate we believe that it can explain why the magnetic stress needed to obtain the jamming transition at shear rate close to zero is larger than the hydrodynamic stress needed for this same transition at high shear rate.

#### 4 DST in shear imposed mode

Up to now, we have presented results obtained when we impose the stress. If it is the shear rate which is imposed, then, when the shear rate will be larger than the critical one, we shall have a jump of stress. In the case where the shear rate is kept constant, if we increase the field, we shall trigger the transition and for the same reason we should observe a jump of stress. That is actually what we see in Fig.4 where the imposed shear rate is  $10 \text{ s}^{-1}$  and the field is increased step by step, each step of  $1.4 \text{ kA/m}$ . At a critical field we observe a huge jump of stress which overcomes the capacity of our rheometer and more often the suspension begins to be expelled out of the gap between the disks. In order to see what happens at higher stress we have used a viscosimeter built by the CAD company for measuring the viscosity of concrete. Its maximum torque of about  $8 \text{ N.m}$ -that is to say approximately 30 times the one of rheometers- is limited by a mechanical safe guard.

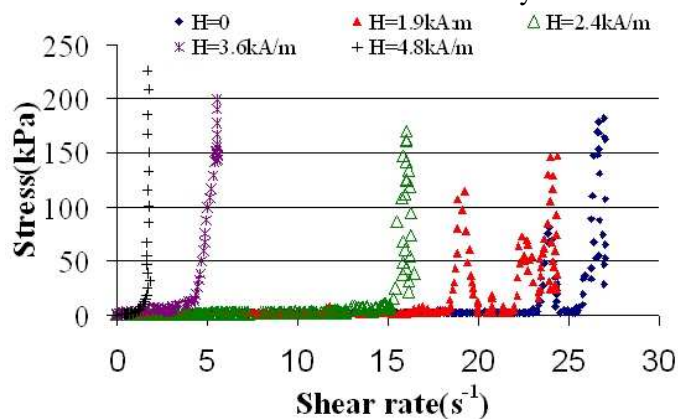


FIG.5 Ramp of shear rate at different values of the field. Closed Couette cell with a double helix;  $\Phi=61\%$

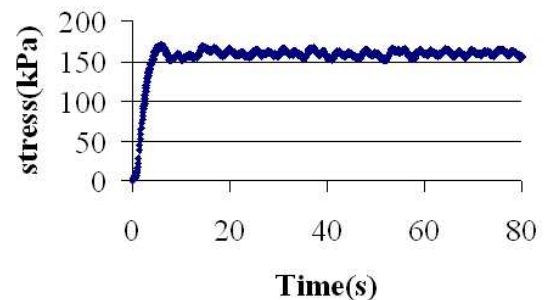


FIG.6 Constant shear rate of  $12.2 \text{ s}^{-1}$  from 7s to 80s. Magnetic field  $H=8.2 \text{ kA/m}$ ;  $\Phi=61\%$

The cell is a horizontal Couette cell with the internal cylinder replaced either by a double helicoidal ribbon or a vane geometry in order to prevent slipping of the suspension; also the external cylinder has regular stripes of depth  $0.2 \text{ mm}$ . Around the cylindrical cell a coil allows to impose a magnetic field which is then directed along the rotation axis. The stress for a ramp of shear rate at different constant magnetic field is represented in Fig.5. Still, despite the high torque limit, for the larger field the stress overcomes the security torque and the motor stops. It means that the stress developed by the jamming

transition can be larger than 200kPa . The other important thing to note is that the critical shear rate decreases as the field is increased, ranging from  $27 \text{ s}^{-1}$  in the absence of the field to  $2 \text{ s}^{-1}$  for a field of 4.8kA/m. This behavior was expected and mimics the one obtained in stress imposed mode (cf Fig.2) but the values of the critical shear rate differ. That is due to the different geometry: plate-plate geometry versus Couette cell with a double helix, and also to the fact that the magnetic field is now parallel to the vorticity instead of being parallel to the shear rate gradient. For the lowest field , the maximum stress is below the one which triggers the mechanical security and we observe oscillations of the stress between a low and a high value. This observation was already done on silica spheres [4] but with a much smaller amplitude. At higher field, we observe, both with vane geometry and double helix geometry, that, instead to have an abrupt transition during a ramp of shear rate (like in Fig.5), we have a progressive increase of stress which stabilizes at a high value, in the jamming domain, without returning back to the low stress regime. This is the case shown in Fig.6 where the shear rate is increased from 0 to  $12 \text{ s}^{-1}$  in 7s and kept constant after. The stress remains constant at a value of 150kPa : the suspension flows in the jammed state which is maintained thanks to the magnetic field. Although we do not have a clear explanation for this behavior , this situation is quite promising for applications since a high torque can be maintained with a very low field. For comparison, to obtain a maximum yield stress between 50kPa and 80kPa in usual magnetorheological fluid, a magnetic field fifty times higher than the one applied in Fig.6 is needed.

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