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Yield behavior of magnetorheological suspensions

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Abstract

The rheology of suspensions containing magnetic particles of micrometer size is strongly modified by the application of a magnetic field. We first describe a model experiment where the suspension is made of spheres of millimeter size and we show that the experimental dependence of the yield stress is well predicted by a non-affine model where the chains of particles break in the middle. Then we compare these predictions with some experimental results obtained on suspensions of carbonyl iron; it is shown that this model does not apply in this case. We propose a mechanism where after some friction on the walls the aggregates begin to rotate and break at a strain smaller than unity, due to the interactions with other aggregates.

Keywords: Magnetorheological suspensions; Magnetic particles; Yield stress

1. Introduction

The prediction of the strength of 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 3D model taking into account multipolar interactions up to the 50th order [1] is correct at low fields but it will always predict a H^2 behavior which is not found experimentally in most cases due to the field limitation in the interparticle gap [2,3]. In magnetorheological suspensions, calculations based on magnetic saturation of poles between two particles inside a chain give the right order of magnitude for the yield stress [4]. 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 for the characterization of the particles: broad size distribution, existence of an oxide layer, difficulty to access to the bulk permeability of the particles, unknown structure. In order to control most of the experimental parameters, we have used a suspension of monodisperse steel spheres with a diameter of 1 mm. Our first aim was 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 these results obtained from this reference suspension. In a second section we shall give some results concerning yield stresses on a real suspension made of carbonyl iron particles of micrometer size in oil and we shall compare the two situations.

2. Experiments on steel spheres

The yield stress of a model suspension of steel spheres (1 mm diameter) has been studied in the parallel plate geometry of the Carri-Med CSL-100 rheometer. In order to avoid the particles to slip on the walls, a layer of spheres was glued on each plate of the measuring cell. The number of spheres fixed on the plates was adjusted as a function of each studied volume fraction. The gap

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between the two plates was 6 mm. Different volume fractions $\Phi = 0.07$, 0.15, 0.25, 0.37, 0.52 were studied over a quite large range of applied fields. We have then compared the experimental results with finite elements calculations based on the calculation of forces between two particles located inside an infinite chain. The unit cell contains two particles with periodic boundary conditions in the direction of the field (z-axis) and the average field is fixed on the side of the cell whose location is given by the volume fraction. The shear force between two particles is deduced from the radial one:

$$F_s = F_r^* \sin(\theta)$$

and we suppose that the particles follow the strain given by $\gamma = \tan(\theta)$; this is the standard model of affine motion. 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 of the chain and the corresponding stress is the yield stress. If we know the interparticle shearing force F_s between two particles, then we can find the shear stress as

$$\tau = \frac{N_c}{S} F_s = \frac{\Phi}{(2/3)\pi a^2} F_s,$$

where Φ is the volume fraction of solid particles, a the radius of a particle and N_c/S the number of chains per unit surface. The only input of the model is the permeability of the bulk steel as a function of the field which has been independently measured [7]. It can be fitted by the Frohlisch–Kennelly law with

$$M = \chi_i H / \left(1 + \frac{\chi_i}{M_s} H\right)$$

as parameters, the initial permeability $\mu_i = 1 + \chi_i = 250$ and the saturation magnetization $M_s = 1360$ (kA/m).

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), with the experimental one. This comparison is shown in Fig. 1 for the volume fraction $\Phi = 0.15$ as a function of the internal average field H. The upper solid curve corresponds to the assumption that all the particles in the six particles chain have an affine motion; in this case all the particles separate at the same time and the separation between two adjacent particles remains the same everywhere in the chain. On the contrary, for the two other curves we impose either two separations in a chain of six particles or a unique separation in the middle of the chain. We see that the predicted values are strongly dependent on the assumption concerning the way the particles separate and that the predicted stress is lower for the case where they break only at the middle. The experimental values are represented by the triangles

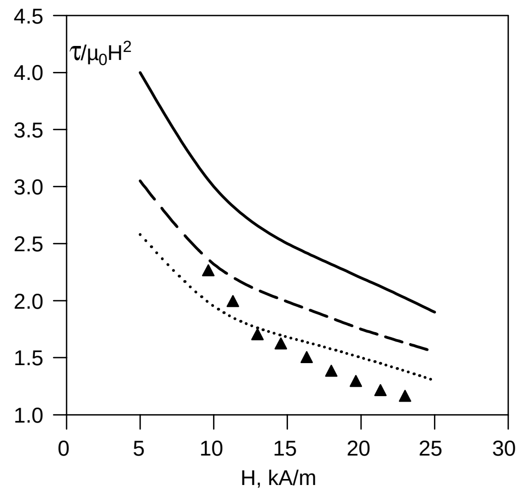

Fig. 1. Normalized yield stress versus internal magnetic field for a volume fraction $\Phi = 15\%$ suspension of millimetric steel spheres: (▲) experimental; (—) calculated by FEM equal gap between spheres (affine model); (- - -) calculated by FEM rupture, each two spheres; (· · ·) calculated by FEM rupture, each three spheres.

and are normalized by the average field

$$H = H_0/\bar{\mu}(H).$$

The agreement with the scheme where the chains break at the middle corresponds to what we observe in the cell. Actually, the affine motion of the chains gives an overestimation of the yield stress because, for the same strain, the gaps are distributed among each pair of particles, hence the distance between two particles does not decrease as quickly with the strain as in the case of one single gap.

3. Experiments on carbonyl iron suspensions

In the case of carbonyl iron particles of micrometer size, we can identify two different stresses: a frictional yield stress, which corresponds to the beginning of a slip of the aggregates on the wall, we call it τ^s, and a Bingham yield stress which is obtained from the fit of the curve by a Bingham law: $\tau = \eta_0 \dot{\gamma} + \tau^y$. The frictional yield stress can be obtained either from an extrapolation (on a logarithmic scale for the shear rate) of the stress versus shear rate curve or from a direct measurement of the yield stress from a stress versus strain experiment. We have verified that the results do not depend on the way it is done: for instance, we obtain the same curves in a plate–plate geometry and two different gaps of 100 and 300 μm. We have also used different materials (iron, glass, stainless steel) and either a plate–plate or a cone–plate geometry. All the results are plotted for the

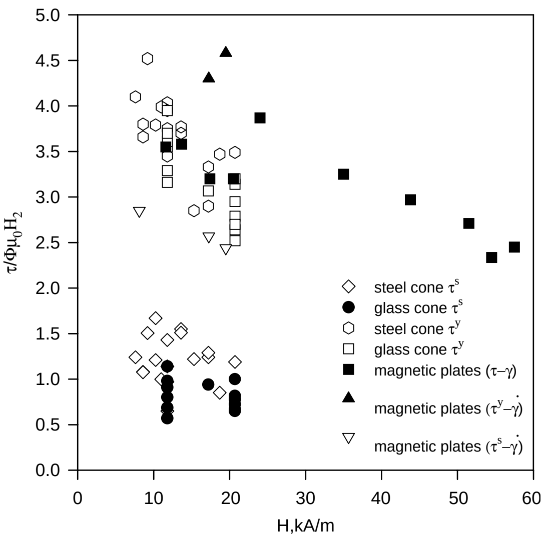

Fig. 2. Field dependence of the frictional stress, τ^s, and of the stress obtained from a Bingham fit, τ^y, for $\Phi = 5\%$ suspension of carbonyl iron particles in various geometries. All the values τ^y are obtained from a Bingham fit. The quantities τ^s are obtained either from a stress–strain $(\tau - \gamma)$ or from an extrapolation to zero shear rate in logarithmic coordinates.

normalized stress $\tau/\Phi\mu_0 H^2$ versus the internal field inside the suspension. We first note that the frictional stress is about three times lower than the Bingham yield stress. Also its value is about the same for glass or stainless steel, but much higher for iron plates. Actually, in this latter case the particles are attracted by the iron plates and blocked inside microcavities. The second important point is that, despite some scatter between the results, the experimental values are much smaller than the one obtained with the standard model, cf. Fig. 1 (the theoretical curves should be divided by the volume fraction which is 0.15 in order to compare with Fig. 2).

Furthermore, the magnetization saturation of carbonyl iron is underestimated by taking the one measured on steel

The fact that the affine model overestimates the Bingham yield stress is not surprising: if we suppose that the chains break in the middle, then the critical strain needed to separate two particles in order to pass the maximum of the restoring force becomes increasingly small as the number of particles inside the gap increases. Since the yield stress is proportional to the product of the radial force between two spheres times the critical strain, the yield stress should decrease as the gap increases, but this is not found experimentally.

4. Discussion

From these observations one should conclude that the aggregates do not break but begin to slip on the wall, then they begin to rotate and no longer touch the plates. At some critical strain the hydrodynamic shear force will break the aggregates. If we suppose that the aggregates are linear chains which do not interact between each other, this critical strain, γ_c can be predicted, and the Bingham yield stress based on this model is given by [8]

$$\tau^y/(\mu_0 H^2 \Phi) = 9 f_\Gamma \gamma_c / 8(1 + \gamma_c^2)^2.$$

f_Γ as well as γ_c depend on the permeability of the particles, which depend on the field. For high permeabilities, typical values are, respectively, $f_\Gamma = 2$ and $\gamma_c = 6$. Due to the dependence on γ_c^{-3}, if γ_c is larger than unity, this model predicts values which are much lower than the experimental one. But, if we consider interactions between aggregates, a critical strain larger than one is not realistic since the aggregates will attract each other and break before reaching an angle 45°. Experimentally, stress–strain experiment gives a value of γ_c between 0.2 and 0.5, so we recover the right order of magnitude for the yield stress.

In conclusion we can say that, except in some special cases where the forces between the particles and the wall can be larger than the interparticle magnetic force, the aggregates slip on the walls when the stress is increased and begin to rotate before breaking. This scheme together with chain interactions can account for the observed behavior.

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