Shear Thickening and Shear Induced Heterogeneities in Concentrated Particle Suspensions

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Abstract:

We study the behavior of dense suspensions of non-Brownian particles. We combine macroscopic rheometric experiments and local velocity and concentration measurements through MRI. In steady state, we find that the material is heterogeneous, while the local rheology presents a continuous transition from a viscous to a shear thickening regime (shear stresses proportional to the shear rate squared, as in dry granular materials) at low shear rate. The heterogeneity is shown to result from unexpectedly fast migration of grains during transient, which is attributed to the emergence of the granular rheology. This migration process is accompanied with macroscopic transient discontinuous shear thickening, which is thus not an intrinsic property of granular suspensions.

Mots clefs: Suspension, Shear thickening, Migration

1 Introduction

Cornstarch is not very complicated; it consists of irregular particles of starch of about 20 micrometers in size. When such particles are suspended in water however, the behavior of the suspension is very strange. If you walk fast on a pool filled with a concentrated cornstarch suspension you can actually walk over the water, but if you walk slowly you sink away. This transition between a liquid-like material at small (or slowly) applied stress and a solid material at high (or rapidly) applied stress is called shear thickening [1-6], and it is a phenomenon that is still incompletely understood. One may wonder why we cannot understand the behavior of this material, as it indeed appears extremely simple: just particles in water. Understanding shear thickening is an important issue in complex fluids with in addition a strong impact on energy consumption in industrial processes [1]. It is observed in dense colloidal suspensions [1, 2], where it has been related to the formation of dense clusters of particles [2-4]. In these systems, the competition between shear-induced cluster formation and Brownian motion that homogenizes the suspensions naturally determine a critical shear rate for the onset of shear thickening.

As Brownian motion is absent in suspensions of large particles, the sharp shear thickening transition observed in e.g. cornstarch suspensions [5] is a priori surprising. In fact, the conditions of emergence of shear thickening in non-Brownian suspensions remain ill-characterized: in some systems, thickening was observed at low shear rates [1, 5-7], while in others no shear thickening (only viscous behavior) is observed, even close to jamming [8-10]. An important effect of confinement on shear thickening of non-Brownian suspensions was recently evidenced [5], which shows that local observations are crucial to get a better insight into their intrinsic behavior.

In this paper, we study the emergence of shear thickening in model density-matched suspensions of non-Brownian particles in water. We use a wide gap Couette geometry to avoid confinement effects [5], and we access the intrinsic material behavior by measuring the local flow properties and particle concentration using Magnetic Resonance Imaging (MRI)

2 Materials and methods

We study dense suspensions of noncolloidal monodisperse spherical particles immersed in a Newtonian fluid. We use polystyrene beads (diameter 40 μm, polydispersity <5%, density 1.05 g.cm\textsuperscript{-3}) suspended in aqueous solutions of NaI to match the solvent and particle densities; the solution viscosity is 1 mPa s. The density
matching ensures that there are no gravity-induced contacts [11] and that the only source of normal stresses is shear [12, 13]. We focus on results obtained at a 59% mean volume fraction as experiments at other high volume fractions show similar features. The material behavior is studied with a wide-gap Couette rheometer (inner radius: 4.1 cm; outer radius: 6 cm; height of sheared fluid: 11 cm) inserted in a MRI scanner, allowing us to access local velocity and particle volume fraction profiles in the flowing sample [9, 14, 15]. Sandpaper is glued to the walls and there is no significant slip on the velocity profiles. The inner cylinder velocity is controlled, and we record the torque exerted by the material on the inner cylinder with a Bohlin rheometer.

3 Experimental results

3.1 Macroscopic and local observation

We first focus on the macroscopic behavior [Fig. 1(a)]. The torque T values measured during a slow ramp in rotational velocity \( \Omega \) (logarithmic ramp, 45 s/decade of shear) are shown as black circles. If we simply plot the necessary torque for rotating the cylinder as a function of its rotational speed, for low shear rates we observe that the torque is proportional to the rotation speed – a Newtonian behavior [9, 10]. Beyond a certain rotation rate, however, the torque jumps discontinuously to high values; i.e., we observe shear thickening that is similar to what happens for the cornstarch [5] system discussed in the introduction. If we subsequently go down in rotation rate we find that the system has completely changed: we descend on a completely different branch that is significantly below the original one. If we increase the shear (= rotation) rate again, we remain on this second branch; we can go up and down as long as we want but the system does not change anymore: there was an irreversible change in the system when we were on the first branch.

![Figure 1](image_url)

Fig. 1: a) Torque vs. rotational velocity: increasing ramp (black squares) and stationary state (empty circles); the line is a viscous law. b) Volume fraction vs. radius, for various rotational velocities, when performing an increasing velocity ramp on a 59% suspension. c) Dimensionless velocity vs. radius (empty symbols: before shear induced migration, filled symbols: various rotational velocities after shear induced migration). d)
Critical strain needed to induce full migration in a 59% suspension as a function of the inner cylinder velocity; the dotted line indicates the $\gamma^2$ transition on the 59% suspension. The line is a $1/\gamma$ scaling.

In order to see what happens we measure velocity profiles $V(R)$ and concentration profiles $\varphi(R)$ with the help of the MRI. The velocity and the concentration profiles show that the system is well behaved and the particle concentration remains homogeneous [Fig. 1(b)], if we remain below the discontinuous jump in the torque on the first branch – there is no jammed region within the gap [5]. The velocity profile $V(R)$ closely matches that of a Newtonian fluid [Fig. 1(c)] [9, 16], consistently with the linear behavior of the torque. However as soon as we go beyond the critical rotation rate, instead of going to zero continuously over the entire gap, the velocity profile again goes to zero at some finite distance from the inner cylinder. The concentration profile measurements show that this happens because a strong gradient in particle concentration develops: the system shows particle migration [9, 17-19]. The result of the migration is that the particle concentration becomes smaller near the inner cylinder and larger near the outer cylinder. The latter observation also explains the velocity profile: a region develops that is so concentrated in particles that it has a yield stress, and so again this part of the system is subjected to a shear stress but does not flow. The migration of particles from the high shear rate to the low shear rate region in the flow is a classical effect that is usually explained by the fact that a particle in the center of the gap will on average undergo more collisions from the particles close to the inner cylinder that from those closer to the outer cylinder, simply because the velocity gradient is larger close to the inner cylinder [9]. Thus, the irreversible change in the system that we observe is due to the migration of the particles. If we subsequently ramp the rotation rate up and down there is no further change in the system, in line with the observation that once the migration has occurred we remain on the second branch, which has then become reversible. However, it is particularly striking here that the change of $\varphi(R)$ occurs over a very short time interval, corresponding to a small total strain of order 100 [Fig. 1(c)]. Such a rapid migration is a puzzle as it is not predicted by classical theories [9].

### 3.2 Constitutive law

We now analyze the behavior in steady state. The material and flow being heterogeneous, macroscopic torque measurements are indeed insufficient to infer the intrinsic constitutive behavior of our system at constant volume fraction: this justifies the use of local measurements. From these measurements, we first note that density and velocity profiles are in steady state whenever the torque is. Moreover, while the density profile $\varphi(R)$ is essentially $\Omega$-independent in steady state, the dimensionless velocity profiles $V(R)/V(R_i)$ do not superpose (they become more and more curved with increasing $\Omega$), implying that the local behavior cannot be simply Newtonian [9, 16]. Finally, the flow is always strictly localized: for all $\Omega$, there is a jammed region beyond a critical radius $R_m=5.7\text{cm}$; this corresponds to density threshold $\varphi(R) > \varphi_c \approx 60.5\%$ above which the material is jammed, consistent with previous observations [9]. Using the fact that the steady-state density profile $\varphi(R)$ is independent of $\Omega$, a change of variables can be then performed between radius $R$ and $\varphi(R)$. In addition, the stress profile is prescribed by momentum balance $\sigma(R) = T/2\pi HR^2$ while the local shear rate can be extracted from the velocity profile $V(R)$ via $\dot{\gamma}(R) = R d(V(R)/dR$. A stress/strain-rate curve $\sigma(\dot{\gamma}, \varphi)$ - at fixed and well-defined density $\varphi$ - is then obtained by collection all measurements of local stress $\sigma(R)$ and local shear rate $\dot{\gamma}(R)$ for a fixed $R$ and varying $\Omega$. An example of such a flow curve is shown in Fig. 2(a), where we now clearly observe two regimes: one at low shear rates where the stress is proportional to the shear rate, as it would be for a Newtonian liquid, and a second one that clearly shows a stronger dependence of stress on shear rate, i.e., we observe shear thickening for the homogeneous system. The dependence of stress on shear rate appears to be well described by a quadratic, i.e., Bagnold-like dependence [18-20]. The transition from viscous and Bagnoldian regime occurs at a critical shear rate $\dot{\gamma}_c(\varphi)$ [Fig. 2(b)]. Such transition has been predicted to be a generic property of noncolloidal suspension on the basis of theoretical dimensional arguments [22, 23]. The $\dot{\gamma}^2$ scaling signals a regime where particle inertia dominates over viscous forces [8, 21], leading to a behavior analogous to that of dry granular materials (it need not be associated with collision dominated flows.
as Bagnold suggested [22]). It is particularly striking that inertial scaling arises in our dense, highly damped, suspension, with particles of size of only \( \approx 20 \mu m \). Moreover, the critical shear rate \( \dot{\gamma}_c (\phi) \) (i) is rather low (of order \( 1s^{-1} \)), (ii) vanishes almost linearly as the volume fraction tends to \( \phi \approx 60.5\% \), which (iii) is identical – within the experimental accuracy – to the threshold \( \phi_m \) at which the material jams.

**Fig. 2:** a) Local shear stress vs. local shear rate measured for various local volume fractions when varying the inner cylinder rotational velocity from 0.1 to 50rpm. The full lines are \( \dot{\gamma} \) scaling; the dotted line are \( \dot{\gamma}^2 \) scaling. b) Critical shear rate for the \( \dot{\gamma} / \dot{\gamma}^2 \) transition vs. volume fraction

### 3.3 Analysis

To understand what controls these scaling regimes, we must, following [20, 21], write Newton’s equations for a set of rigid particles. For the particles’ centers of mass \( r_i \) they read: 

\[
\frac{\partial^2 r_i}{\partial t^2} = \sum_j F_{ij} + F_{i}^{\text{visc}}
\]

where \( F_{ij} \) denote rigid contact forces and \( F_{i}^{\text{visc}} \) hydrodynamic forces which are supposed linear in terms of all the velocities entering the problem. No other force is supposed to be involved. The key of the analysis is to remark that if \( F_{ij} \) are rigid forces, they do not introduce, by definition, any force or length scale. We then consider the two limit problems: “viscous” (V) when viscous forces are dominant over grain inertia 

\[
0 = \sum_j F_{ij} + F_{i}^{\text{visc}}
\]

and “inertial” (I) when grain inertia is dominant 

\[
\frac{\partial^2 r_i}{\partial t^2} = \sum_j F_{ij}
\]

which both verify exact scale invariance by a change of time and force units [21], guaranteeing \( \dot{\gamma} \) in (V) and \( \dot{\gamma}^2 \) in (I), with identical scaling for all components of the stress tensor. The full problem will then reduce to (V) (resp. (I)) at low (resp. high) \( \dot{\gamma} \), which explains the existence of a crossover between the two simple scaling regimes \( \sigma \propto \dot{\gamma} \) (viscous) and \( \sigma \propto \dot{\gamma}^2 \) (inertial).

This formalism helps us understanding why the critical shear rate \( \dot{\gamma}_c (\phi) \) is so low and vanishes precisely at \( \phi_m \). In the viscous (V) and inertial (I) regimes, the stresses read respectively \( \sigma = \sum_{\nu} (\phi) \dot{\gamma} \) and \( \sigma = \sum_{\nu} (\phi) \dot{\gamma}^2 \). Numerical simulations [23] indicate that \( \sum_{\nu} (\phi) \) and \( \sum_{\nu} (\phi) \dot{\gamma} \) should diverge at the same (jamming) packing fraction \( \phi_m \) and read \( \sum_{\nu} (\phi) \approx (\phi_m - \phi)^{-\alpha_V} \), \( \sum_{\nu} (\phi) \approx (\phi_m - \phi)^{-\alpha_I} \). The cross-over between the viscous and inertial regimes is found by equating the two expressions for stress in either regime, finally leading to \( \dot{\gamma}(\phi) \approx (\phi_m - \phi)^{\alpha_I - \alpha_V} \), i.e. \( \dot{\gamma}_c (\phi) \) vanishes precisely at the jamming packing fraction.
as observed. Moreover, the linear vanishing of \( \dot{\gamma}_c(\varphi) \) means \( \alpha_i - \alpha_v = 1 \), which is consistent with the values \( \alpha_i = 2 \), \( \alpha_v = 1 \) proposed in the literature [24, 25]. We add that, the stress level at the cross-over verifies \( \sigma_\alpha(\varphi) \approx (\varphi_m - \varphi)^{\alpha_i - 2\alpha_v} \); we then note that the critical stress [see Fig. 2(a)] does not vary much, even though our stress measurements are not sufficiently accurate to assert that \( \sigma_\alpha(\varphi) \approx \text{const} \), which is again consistent with \( \alpha_i = 2 \), \( \alpha_v = 1 \). We finally conclude that it is the difference in singular behavior of the inertial and viscous behaviors at the approach of jamming which leads to the linear vanishing of \( \dot{\gamma}_c(\varphi) \), and hence permits this transition to take place at low strain rates.

### 3.4 Accelerated migration

We now show that this transition also explains the sudden migration which was seen to be associated with the macroscopic sharp shear-thickening [Fig. 1(a)]. In viscous suspensions, shear-induced migration is usually neglected when small particles are involved: the typical strain scale for migration is indeed expected to be rate independent and to scale as \( \propto (R_e - R_j)^2 / a^3 \) [9, 17-19]; a review of the literature [9] would then lead to an expected strain of order 50000, more than 500 times higher than what we observe at the onset of shear thickening. By taking into account the inertial regime, we are able to understand this puzzling observation. In the framework of two phase models, migration is driven by gradients of internal normal stresses within the particle network \( \sum p \) (not the total stress), and that it requires the fluid to filter through the granular phase in order to compensate for the local changes in packing fraction [18]. This filtration process exerts, on the particle network, an average hydrodynamic drag \( \propto U \), with \( U \) the average filtration velocity. In two phase models, the mechanical balance between these two effects controls the migration/filtration rate, leading to \( \sum p U \). When injected in a mass conservation equation \( \partial \rho / \partial t = -\nabla (\rho U) \), this constitutive relation leads to a diffusion equation for the particle density \( \rho \) (rigorous derivation is found in [18]). The scaling with the shear rate \( \dot{\gamma} \) of the timescale of migration is then basically the inverse of that of \( \sum p \). Two behaviors can then be predicted. At low strain rate, in the viscous regime \( \sum p \propto \dot{\gamma} \), the timescale of migration should scale with \( 1 / \dot{\gamma} \); migration process is thus controlled by a constant strain scale; this is the classical result for viscous suspensions [17, 19]. On the other hand, at higher strain rates, when the particle network stress enters the \( \propto \dot{\gamma}^2 \) regime, this analysis yields a \( 1 / \dot{\gamma} \) strain scale: this would thus explain why migration is accelerated and may be quite sudden when increasing \( \dot{\gamma} \) and entering the inertial regime.

To confirm this analysis, we have studied in more detail the kinetics of the phenomenon. Starting from a homogeneous suspension, we have applied a given macroscopic shear rate \( \dot{\gamma}_{\text{macro}} \) (constant \( \Omega \)) to the system, and we have measured the volume fraction profile evolution in time. In Fig. 1d, we plot the macroscopic strain scale for inducing full migration for various \( \dot{\gamma}_{\text{macro}} \). We observe that this strain scale may be as high as 2000 at low rotational velocity (here, 2 rpm) and that it decreases strongly with the rotational velocity, down to less than 50 at 20 rpm. We also find that it decreases qualitatively as the inverse shear rate as proposed. This result constitutes a strong experimental validation of migration theories based on normal stresses [18], which prove here to be more generally applicable than diffusive theories [17]. Note however that the exact kinetics results from a complex history: as both \( \dot{\gamma} \) and \( \varphi \) change locally in time [Fig. 1] and as the transition shear rate depends strongly on \( \varphi \) [Fig. 2], the flow regime (and thus migration kinetics) may vary both in space and time. This may explain why very rapid migration was previously observed in a suspension that had apparently a purely viscous behavior in its heterogeneous state [9].
4 Conclusion

To summarize, we propose the following scenario: (i) the intrinsic behavior of dense non-colloidal suspension presents a continuous transition at low strain rates from a viscous to a shear thickening, granular, rheology characterized by shear stresses $\sigma \propto \dot{\gamma}^2$; (ii) in the granular regime, a very fast particle migration then occurs towards low shear zones; (iii) the interplay between flow and migration shows up as a sharp shear thickening of the transient macroscopic stress.

The steady-state behavior $\sigma \propto \dot{\gamma}^2$ differs from previous observations [7] in a similar system (spheres in a Newtonian fluid). This is explained by our results, which tend to show that macroscopic data (as those of [7]) are obtained on heterogeneous systems and cannot be directly related to the local constitutive behavior of suspensions.

Let us finally note that our mechanism may also be at work in Brownian suspensions, in competition with or as an alternative to hydrodynamic clustering [2]. It is compatible with the reversibility of the shear thickening transition usually observed in Brownian suspensions: migration is indeed expected to be reversible due to the osmotic pressure. It is therefore particularly striking that our mechanism leads to a constant critical shear stress, exactly what is observed for colloids [2].

References