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Shear Thickening and Migration in Granular Suspensions

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We study the emergence of shear thickening in dense suspensions of non-Brownian particles. We combine local velocity and concentration measurements using magnetic resonance imaging with macroscopic rheometry experiments. In steady state, we observe that the material is heterogeneous, and we find that the local rheology presents a continuous transition at low shear rate from a viscous to a shear thickening, Bagnoldian, behavior with shear stresses proportional to the shear rate squared, as predicted by a scaling analysis. We show that the heterogeneity results from an unexpectedly fast migration of grains, which we attribute to the emergence of the Bagnoldian rheology. The migration process is observed to be accompanied by macroscopic transient discontinuous shear thickening, which is consequently not an intrinsic property of granular suspensions.

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The realm of complex fluids encompasses biological liquids such as blood, many liquid foodstuffs, building materials, glasses and plastics, crude oil, etc. Despite their importance, the most basic, quintessential question about the flow of these fluids has remained unanswered: it is generally impossible to predict their flow resistance, and it is even unclear why most fluids shear thin, whereas only some shear thicken. Understanding shear thickening, i.e., the increase of the apparent viscosity of materials with increasing flow rate, is thus 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 [2,3], where it has been related to the formation of dense clusters of particles [2–4]. The viscosity rise with the shear rate is then usually reversible (it is a steady-state property), continuous, and is sharper at higher volume fractions [2–4]. For colloids, the competition between shear-induced cluster formation and Brownian motion that homogenizes the suspensions then naturally determines a critical shear rate for the onset of shear thickening.

As Brownian motion is absent in pastes made of large particles, the sharp shear thickening transition observed in, for instance, cornstarch suspensions [5] is highly surprising. In fact, the conditions of emergence of shear thickening in non-Brownian suspensions remains 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 whatsoever, even close to jamming [8–10]. Up to now it is thus impossible to predict whether a given system will shear thicken or not.

In systems for which it is observed, a more pronounced shear thickening [6,7] is observed near jamming, similarly to colloidal suspensions, and is attributed to aggregation of hydroclusters into a percolating network [4,11]. However, one should question whether these observations of sharp and discontinuous shear thickening reflect an intrinsic (local, steady-state) property of materials. For example, an important effect of confinement on shear thickening was recently evidenced in both colloidal [12] and noncolloidal [5] suspensions. In channel flows [12], the macroscopic shear thickened state was shown to form a plug flow and was not observed in large channels, which shows that local observations are crucial to get a better insight into shear thickening.

In this Letter, we address these puzzles by studying the emergence of shear thickening in the simplest of systems: model density-matched suspensions of non-Brownian particles in water. We use a wide-gap Couette geometry to avoid confinement effects, and we access the intrinsic material behavior by measuring the local flow properties and particle concentration using magnetic resonance imaging. In steady state, we show that the material is heterogeneous, and that the local rheology presents a continuous transition from a viscous to a shear thickening, Bagnoldian, behavior (shear stresses proportional to the shear rate squared) at any fixed volume fraction, as predicted by a scaling analysis. The heterogeneity is shown to result from an unexpectedly fast migration of grains during the transient, which is attributed to the emergence of the Bagnoldian rheology. The migration process is accompanied by macroscopic transient discontinuous shear thickening, which is thus not an intrinsic property of granular suspensions.

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−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 [13] and that the only source of normal stresses is shear [14,15]. 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 magnetic resonance imaging scanner, allowing us to access local velocity and particle volume fraction profiles in the flowing sample [9,16,17]. Sandpaper is glued to the walls and there is no significant change in open symbols, implying that the change in \( \phi(R) \) occurs over a very short time interval, corresponding to a small total strain of order 100. Such a rapid migration is a puzzle as it is not predicted by classical theories [9].

**Constitutive behavior.**—We now analyze the steady-state behavior. We first note that density and velocity profiles are in steady state whenever the torque is. Moreover, while \( \phi(R) \) is independent in steady state, the dimensionless velocity profiles \( V(R, \Omega)/V(R_\text{f}, \Omega) \) measured at various \( \Omega \) do not superpose, implying that the local behavior is not simply viscous [9,18]. 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 a density threshold of \( \phi(R) > \phi_m = 60.5\% \) above which the material is jammed [9].

The material and flow being heterogeneous, macroscopic torque measurements \( T(\Omega) \) are not sufficient to infer the intrinsic constitutive behavior, in particular, the stress-strain rate relationship in the shear thickening regime. This intrinsic behavior can, however, be obtained using our local measurements. The key point [9] is that the steady-state density profile \( \phi(R) \) is independent of \( \Omega \); a change of variables can then be performed between radius \( R \) and \( \phi(R) \). In addition, the stress profile is prescribed by momentum balance \( \tau(R) = T/(2\pi HR^2) \) while the local shear rate can be extracted from the velocity profile \( V(R) \) via \( \dot{\gamma}(R) = Rd(V(R)/dR). A local stress-strain rate curve \( \tau(\dot{\gamma}, \phi) \)---at fixed and well-defined density \( \phi --- is then obtained by collecting all measurements of local stress \( \tau(R) \) and shear rate \( \dot{\gamma}(R) \) for a fixed \( R \) and varying \( \Omega \).

The results of this local analysis [Fig. 2(a)] show that, for a fixed volume fraction \( \phi \), a clear transition...
explains the existence of a crossover between the two simple

FIG. 2 (color online). (a) Local shear stress versus local shear
rate measured for various local volume fractions when varying
the inner cylinder rotational velocity from 0.1 to 50 rpm (from
right to left: φ = 56.8%, 57.6%, 58.1%, 58.5%, 58.8%, 59%, 59.3%, 59.5%, 59.7%, 59.8%, 60%). The full lines are γ scaling;
the dotted lines are γ² scaling. (b) Critical shear rate for the
γ/γ² transition versus volume fraction.

from a τ ∝ γ (Newtonian) to a τ ∝ γ² (Bagnoldian) regime
occurs at a critical shear rate γc(φ) [Fig. 2(b)]. Such
a transition has been predicted to be a generic property of
noncolloidal suspensions on the basis of theoretical dimen-
sional arguments [22,23]. The γ² scaling signals a regime
where particle inertia dominates over viscous forces [8,23],
leading to a behavior analogous to that of dry granular
materials (it need not be associated with collision-
dominated flows as Bagnold suggested [24]). It is par-
cularly striking that inertial scaling arises in our dense,
highly damped, suspension, with particles of size of only
~10 μm. Moreover, the critical shear rate γc (i) is rather
low (of order 1 s⁻¹), (ii) vanishes almost linearly as the
volume fraction tends to φc = 60.5%, which (iii) is iden-
tical—with the experimental accuracy—to the threshold
φm at which the material jams.

Viscous-inertial transition.—To understand what con-
trols these scaling regimes, following [22,23], we write
Newton’s equations for a set of rigid particles. For the
particles’ centers of mass rᵢ, these read
\[ md²rᵢ/dt² = \sum_i F_{ij} + F_{ij}^{-\text{visc}}, \]
where \( F_{ij} \) denote rigid contact forces and \( F_{ij}^{-\text{visc}} \)
hydrodynamic forces which we suppose linear in terms
of all the velocities entering into the problem. No other force
is supposed to be involved. The key of the analysis is to
remark that rigid forces \( F_{ij} \) do not introduce, by definition,
any force or length scale [22,23]. Two limiting cases can
then be identified: “viscous” (V) when viscous forces are
dominant over grain inertia
\[ 0 = \sum_i F_{ij} + F_{ij}^{-\text{visc}} \]
and “inertial” (I) when grain inertia is dominant
\[ md²rᵢ/dt² = \sum_i F_{ij}. \]
Both expressions verify scale invariance by a change
of time and force units [23], guaranteeing \( F_{ij} \propto γ \) in (V)
and \( F_{ij} \propto γ² \) in (I), with identical scaling with γ for all com-
ponents of the stress tensor as rigorously shown in [23]. The
full problem then reduces to (V) [(I) at low (high) γ, which
explains the existence of a crossover between the two simple
scaling regimes \( τ ∝ γ \) (viscous) and \( τ ∝ γ² \) (inertial).

This formalism now helps us understand why the critical
shear rate γc(φ) can be so low and vanishes precisely at φm.
In the viscous (V) and inertial (I) regimes, the stresses are,
respectively, of the form \( τ = η₀γ Σ_φ(φ) \) and \( τ = ρd²γ²Σ_φ(φ) \).
Numerical simulations [25] indicate that Σ_φ(φ) and \( Σ_φ(φ) \) should diverge at the same (jamming)
packing fraction \( φ_m \) and read \( Σ_φ(φ) ∝ (φ_m - φ)^{-α_φ} \),
\( Σ_φ(φ) ∝ (φ_m - φ)^{-α_φ} \), where ρ and d are the particle
density and diameter, and η₀ is the interstitial fluid viscosity.
The crossover between the viscous and inertial regimes
is found by equating the two expressions for the stress, finally
leading to \( γ_c(φ) ∝ (η₀/ρd²)(φ_m - φ)^{α_φ-2α_φ} \). Together
with the values \( α_I = 2, α_φ = 1 \) proposed in the literature
[26,27], this equation explains, as observed, that \( γ_c(φ) \)
vanishes (i) linearly with φ, (ii) at the jamming packing
fraction \( φ_m \). Moreover, the crossover stress verifies \( τ_c(φ) ∝ (η₀/ρd²)(φ_m - φ)^{α_φ-2α_φ} \), which, together with the same
values of \( α_I, α_φ \) as above, suggests that \( τ_c \) should indeed be
independent of volume fraction. Although our stress mea-
surements are not sufficiently accurate to assert that
\( τ_c(φ) ∝ \) const, we then note in Fig. 2(a) that, indeed, in
the experiments \( τ_c \) does not vary much. We finally conclude
that it is the difference in singular behavior of the inertial
and viscous stresses \( ρd²γ²Σ_φ(φ) \) and \( η₀γΣ_φ(φ) \) at the
approach of jamming (i.e., when \( φ → φ_m \) which leads to
the linear vanishing of \( γ_φ(φ) \), and hence permits this
transition to take place at low strain rates.

Accelerated shear-induced migration.—We now show
that this transition explains the sudden migration associ-
ated with the macroscopically observed transient discon-
tinuous shear thickening [Fig. 1(a)]. In viscous
suspensions, shear-induced migration is usually thought
as negligible when small particles are involved. Indeed,
the typical strain scale for migration is very large:
it is expected to be rate independent and to scale as \( ∝ (R_o - R_i)^2/a^2 \) [9,19–21], leading to an expected strain of
order 50 000 [9], more than 500 times higher than what we
observe here at the onset of shear thickening. Our observa-
tions may be understood as a strong enhancement of
migration kinetics in the inertial regime.

Within the framework of two-phase models, migration
is driven by gradients of internal normal stresses within
the particle network Σ_{ii} (not the total stress) and requires the
fluid to filter through the granular phase to compensate for
the local changes of packing fraction [20,21]. This filtration
process exerts an average hydrodynamic drag \( ∝ U \) on the
particle network, with \( U \) the average filtration velocity.
The balance between these two effects controls the migration-filtration rate, leading to \( U ∝ ∇Σ_{ii} \). When
injected in a mass conservation equation \( ∂ρ/∂t = -∇(ρU) \),
this leads to a diffusion equation for the particle density ρ
[20,21]. The local particulate stress \( Σ_{ii} \) entering this analy-
sis is expected to display local viscous or inertial scaling
over very short strain scales [22,25,26] compared to those of
the migration process. If \( Σ_{ii} ∝ γ \) in the whole system,
the time scale of migration scales as 1/γ and migration is
controlled by a (large) rate-independent strain scale, which
is the classical result [19–21]. Strikingly, the same analysis
performed in the inertial regime now yields an unexpected $1/\gamma$ strain scale for migration: this explains why migration is much accelerated and manifests itself abruptly when entering the inertial regime.

To confirm this analysis, we have studied the migration kinetics at constant $\Omega$’s, starting each time from a homogeneous state. We define the critical strain $\Gamma_{\text{mig}}(\Omega)$ as the macroscopic strain above which the instantaneous volume fraction profile matches the steady state one within experimental uncertainty (0.2%). Figure 1(c) shows $\Gamma_{\text{mig}}(\Omega)$ versus $\Omega$: it decreases strongly with $\Omega$. Although the exact kinetics results from a complex history (as both $\gamma$ and $\phi$ change locally in time), the asymptotic $1/\gamma$ decay predicted by the above scaling analysis in the Bagnoldian regime is roughly consistent with our observations [see line in Fig. 1(c)]. Migration theories based on normal stresses [20,21] are thus shown here to be more generally applicable than diffusive theories [19].

Macroscopic shear thickening.—To summarize, we propose the following scenario: (i) the intrinsic behavior of dense noncolloidal suspension presents a continuous transition at low strain rates from a viscous to a shear thickening, Bagnoldian, rheology characterized by shear stresses $\propto \gamma^2$; (ii) in the Bagnoldian 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.

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