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Flow of Wet Granular Materials

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The transition from frictional to lubricated flows of a dense suspension of non-Brownian particles is studied. The pertinent parameter characterizing this transition is the Leighton number $Le = \frac{\eta_s \dot{\gamma}}{\sigma}$, the ratio of lubrication to frictional forces. Le defines a critical shear rate below which no steady flow without localization exists. In the frictional regime the shear flow is localized. The lubricated regime is not simply viscous: the ratio of shear to normal stresses remains constant and the velocity profile has a universal form in both frictional and lubricated regimes. Finally, a discrepancy between local and global measurements of viscosity is identified, which suggests inhomogeneity of the material under flow.

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In recent years, the flow behavior of granular matter has been the subject of considerable controversy. Simple questions such as whether viscosity can be properly defined for granular systems are still the subject of hot debate [1,2]. A notion of viscosity is necessary for many applications to predict the resistance to flow. Perhaps even more crucial, in view of its importance in geophysics and civil engineering, is the resistance to flow of wet granular materials. Nevertheless, the number of studies on wet granular matter is almost negligible compared to that for sand. The usual picture is that for an interstitial fluid at low viscosity the material behaves similarly to dry granular systems (with frictional contacts between the grains), and for an interstitial fluid at high viscosity the material behaves similarly to viscously dominated systems (with lubricated contacts between the grains) [3–6]. If this picture were true, it is of importance to investigate what parameters determine the frictional to viscous transition, and whether a viscosity can be defined.

The purpose of this Letter is to answer some of these questions. We find that the transition between frictional and lubricated flow regimes is characterized by a viscosity bifurcation [7,8] and is governed by the Leighton number $Le = \frac{\eta_s \dot{\gamma}}{\sigma}$, with η_s the interstitial fluid viscosity, $\dot{\gamma}$ the shear rate, and σ the total shear stress. For a given interstitial fluid, Le fixes the critical shear rate $\dot{\gamma}_c$ at which the transition between the two regimes occurs. In the frictional regime, no steady flow exists without localization. We also find that the viscous regime is similar to the granular flow regime, posing the same problem for the definition of the viscosity [1,2].

We study the rheological behavior of a paste (a dense suspension) composed of non-Brownian spherical particles immersed in a Newtonian fluid of the same density at a volume fraction ϕ of 60%. The particles are monodisperse polystyrene beads (diameter 0.29 ± 0.03 mm, density 1.04 g cm⁻³). To avoid sedimentation or creaming effects we match the density of the interstitial fluid

with the density of the particles. Adequately salted water ($\eta_s = 1$ mPa s) and Rhodorsil 10646 silicone oil ($\eta_s = 2300$ mPa s) are perfectly density matched. The oil is miscible with water: we mix it with salted water in order to have an isodense liquid, whose viscosity varies between 1 and 2300 mPa s. These experiments are completed with pastes prepared with silicone oils of variable viscosities [9]. The rheology is done with a vane-in-cup geometry on a commercial rheometer that imposes either stress or shear rate. The vane geometry we use is equivalent to a cylinder (diameter 16 mm, height 52 mm) with a rough lateral surface on the scale of the beads, reducing the slipping of granular materials [5]. For the same reason, the inside of the cup (diameter 26 mm) is covered with the granular particles using double-sided adhesive tape.

In the first set of experiments, we use a 20 mPa s oil as the interstitial fluid. Figure 1(a) shows the evolution of viscosity with time for different applied stresses. We find that for stresses smaller than a critical stress $\sigma_c = 5 \pm 3$ Pa the viscosity of the sample increases in time until the flow

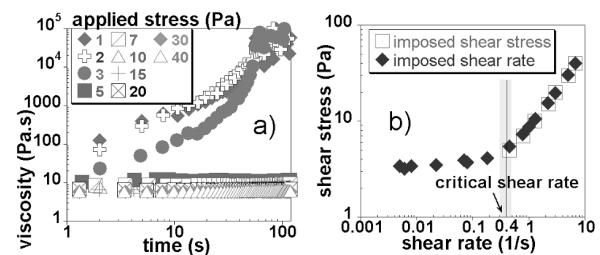


FIG. 1. (a) Viscosity bifurcation: under an imposed stress the viscosity either grows in time or decreases; therefore the steady-state viscosity jumps to infinity at a critical stress. This allows us to define both the critical stress and the critical shear rate. Before each experiment, the material is presheared during 30 s at 30 s⁻¹ to obtain a reproducible initial state. (b) Flow curve (shear stress versus shear rate) for 20 mPa s silicone oil, both at an imposed macroscopic shear stress and shear rate. The shaded area is the statistical error bar.

is halted altogether, which corresponds to an infinite steady-state viscosity [10]. For a stress only slightly above σ_c , the viscosity decreases with time; for long times, it reaches a (low) steady-state value η_0 . This implies that at the critical stress the steady-state viscosity jumps from an infinite to a finite and low value at σ_c . This behavior implies the existence of a bifurcation of the viscosity, which abruptly passes from a flowing state above σ_c to a jammed state below σ_c , as observed previously for yield stress fluids and dry granular materials [7,8]. Apart from a critical stress, the bifurcation also identifies a critical shear rate $\dot{\gamma}_c = 0.4 \pm 0.1 \text{ s}^{-1}$, beyond which steady flows are possible under an imposed stress. If shear rate rather than the stress is imposed, below $\dot{\gamma}_c$ the measured shear stress is almost independent of the shear rate, which is the hallmark of quasistatic granular (frictional) flow [1,5]. For high shear rates ($\dot{\gamma} > \dot{\gamma}_c$), stress increases linearly with increasing shear rate, as for a viscous fluid [Fig. 1(b)]. These observations allow us to identify the transition between two different flow regimes.

To find out what determines the transition, we vary the viscosity of the interstitial fluid and determine the critical stress and shear rate from the viscosity bifurcation. Figure 2 summarizes the main results. $\dot{\gamma}_c$ is inversely proportional to the viscosity of the interstitial fluid, whereas the critical shear stress σ_c turns out to be constant (within experimental uncertainty). The former result is consistent with that of [4,5], who used only two fluids of different viscosities.

We conclude that the pertinent parameter characterizing the transition is the Leighton number $Le = \frac{\eta_s \dot{\gamma}_c}{\sigma}$, which represents the ratio of lubrication to frictional forces. Ancy and Coussot previously reported the inverse proportionality with respect to the frictional forces [5,6]. By varying the lubrication forces, we conclude that the transition is entirely characterized by the Leighton number. The critical stress σ_c does not vary significantly ($\sigma_c = 5 \pm 3 \text{ Pa}$), so that Le_c has the same order of magnitude for η_s varying from 10^{-3} to 2.3 Pa s [$Le_c \approx (7 \pm 5) \times 10^{-4}$]. The order of magnitude of the critical stress is that of the low shear viscosity of the suspension multiplied by $\dot{\gamma}_c$. The Krieger-Dougherty model [11] for hard spheres, namely,

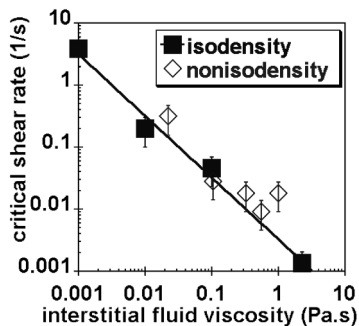


FIG. 2. Critical shear rate versus interstitial fluid viscosity, for polystyrene beads in a Newtonian liquid. The line is a power-law fit to the isodensity values with a slope of -0.99 ± 0.10 .

$\eta = \eta_s(1 - \phi/\phi_m)^{-2.5\phi_m}$, gives $\sigma_c = \eta\dot{\gamma}_c \approx 1 \text{ Pa}$. This gives the correct order of magnitude of σ_c but also explains why the critical stress remains constant: this follows from combining $\dot{\gamma}_c \propto 1/\eta_s$ and $\eta \propto \eta_s$ [12].

All of the rheometric experiments agree with the hypothesis that the low shear, low interstitial fluid viscosity regime is frictional, in the sense that the material behaves similarly to dry granular matter. The remaining question is whether the second regime is viscous. Surprisingly, measurements of the normal stress N_1 using a plate-plate geometry (diameter 40 mm) show that normal and viscous stresses are proportional in both flow regimes. For the plate-plate geometry, $\dot{\gamma}_c$ is found to be very similar to that found in the Couette cell ($\dot{\gamma}_c = 0.5 \pm 0.2 \text{ s}^{-1}$). Figure 3 shows that the ratio σ/N_1 does not vary significantly with the shear rate: $\sigma/N_1 = 0.39 \pm 0.15$ in both regimes and over four decades in shear rate. Previous results in the “viscous” regime are consistent with our findings [3,4,13], despite the fact that others [5] have reported different results. Brady and Morris [14] have shown theoretically that a shear stress and a pressure proportional to shear rate may result from hydrodynamic interactions between the particles; however, in order to generate normal stress *differences*, hard sphere contacts must be incorporated. As a consequence, there is no reason for the ratio σ/N_1 to be equal in the frictional and the lubricated regime as observed experimentally. This surprising observation is reinforced by the finding that the ratio σ/N_1 is equal to the internal static friction coefficient of the dry granular material $\mu_s = 0.40 \pm 0.04$ (as obtained from the slope angle θ of a heap of dry beads with $\mu_s = \tan\theta$). We conclude that, in the lubricated regime, the material has a number of characteristics of dry granular materials, while it simultaneously dissipates viscously in the interstitial fluid (as follows from $\eta \propto \eta_s$).

These puzzling observations raise the following questions: What happens in the flow? What is the distinction between the two regimes? To address these questions, we carried out experiments with a velocity controlled

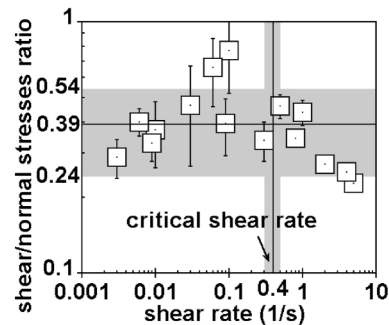


FIG. 3. Shear/normal stresses ratio σ/N_1 as a function of the shear rate for polystyrene beads in 20 mPa.s silicone oil (solid volume fraction: 58%), at imposed shear rate. Both plates are covered with sand paper to avoid wall slip. The shaded area is the statistical error bar. The horizontal line appears off center because of the logarithmic scale.

“magnetic resonance imaging (MRI) rheometer,” which allows for a direct measurement of the local velocity distribution in a Couette geometry [15]. The main results from these MRI measurements are that the velocity profiles are roughly exponential as in dry granular materials [16], and that they occupy only a small fraction of the gap at low rotation rates, i.e., in the frictional regime: we observe shear localization. However, if the rotation frequency is increased, a surprising behavior is observed: contrary to what happens for dry granular matter, the higher the rotation rate, the larger the fraction of the paste that is sheared (Fig. 4). In the lubricated regime, beyond $\dot{\gamma}_c = 0.4 \pm 0.1 \text{ s}^{-1}$, the whole sample is sheared ($\dot{\gamma}_c$ measured with the MRI is the same as $\dot{\gamma}_c$ found in the rheology) [17].

In the frictional regime, the behavior is different only in the sense that a smaller fraction of the material is sheared. In this regime the reduced velocity $V(R)/V_i$ (V_i being the velocity of the rotating inner cylinder) can be collapsed onto the same universal curve when plotted as a function of the rescaled coordinate $(R - R_i)/d_c(V_i)$ (Fig. 5). The length $d_c(V_i)$ simply gives the extent of the material that is sheared. In addition, we find that this universal rescaling of the velocity profile also applies to dry granular materials: two velocity profiles from [1,18] fall along the same universal curve (Fig. 5), underlining the universality of the roughly exponential velocity profiles. The inset of Fig. 5 gives the extent of the sheared region d_c as a function of the rotation rate. Starting from low V_i , d_c increases and eventually fills the whole gap for $V_i > V_c$ [19].

This last observation provides a natural explanation for the macroscopic rheology data. In the first (frictional) regime, no steady flow without localization can be

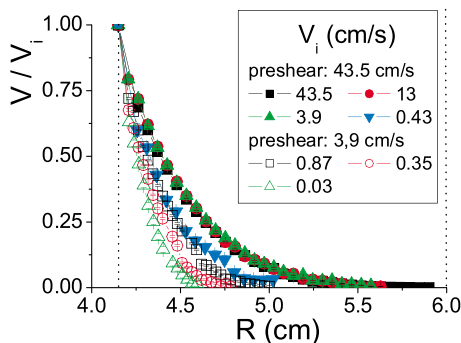


FIG. 4 (color). Velocity rescaled with the velocity of the inner cylinder (inner cylinder radius $R_i = 4.15 \text{ cm}$, outer cylinder radius $R_e = 6 \text{ cm}$, height 11 cm ; we again use the 20 mPa s Rhodorsil silicone oil). For technical reasons, V_i can be varied either between 0.01 and 3.91 cm/s or between 0.43 and 43.5 cm/s . As in the macroscopic rheology experiments, we preshear the material at a rotational speed $V_i = 43.5 \text{ cm/s}$ during 30 s . For the low velocity setup, we preshear the material at the maximum possible rotational speed V_i , i.e., 3.91 cm/s . The rough inner cylinder is driven at a rotational velocity V_i ranging between 0.013 and 43.5 cm/s , yielding overall shear rates between 0.006 and 20.9 s^{-1} , the critical shear rate $\dot{\gamma}_c$ being $0.4 \pm 0.1 \text{ s}^{-1}$ (or $V_c = 1.04 \pm 0.26 \text{ cm/s}$).

achieved at imposed shear rates: there is a coexistence between a sheared and an unsheared region, which results in a constant stress not unlike stress plateaux and the corresponding shear bands observed for certain surfactant and polymer solutions [20]. When the sheared region has invaded the gap, there is no longer coexistence and the stress increases again with increasing shear rate. The transition between the frictional and lubricated regimes then happens at the end of the coexistence.

The interesting question is then whether a viscosity can be defined for the material, in the lubricated regime at least, that completely characterizes its resistance to flow. If we suppose our material is a continuum medium in stationary flow, momentum conservation leads to $\sigma(R) = \sigma_i R_i^2/R^2$ (where σ_i is the total shear stress on the inner cylinder), independently of the constitutive equation of the material: the stress varies within the gap. The shear rate is the spatial derivative of the velocity profile. Therefore it varies within the gap and a single MRI experiment allows one to recalculate a complete stress-shear rate curve. The result is shown in Fig. 6 for different rotation velocities. There are two surprises: first, the data are not consistent between different MRI experiments, and second, they are inconsistent with the macroscopic rheology data [21]. We are therefore forced to conclude that the velocity profiles are not consistent with the macroscopic rheometric data, both when there is an unsheared region in the material and when the material is entirely sheared. This implies that either the material is inhomogeneous or that no simple constitutive equation relating shear stress to shear rate exists for the material (as in dry granular materials [1]).

The question then is this: What happens in the lubricated regime? We can reasonably assume that, even if force chains are present in the sheared system, momentum conservation is likely to hold, at least on average; therefore

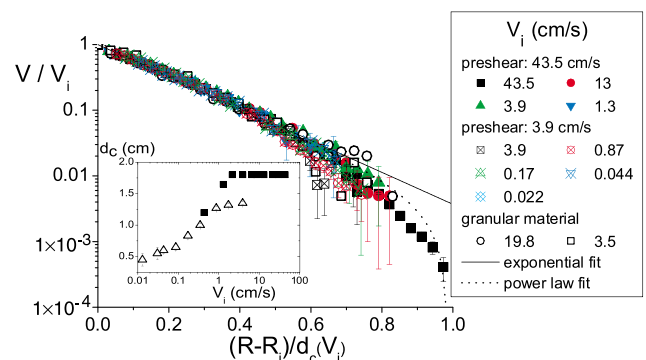


FIG. 5 (color). Velocity profiles measured in a Couette geometry. We plot V/V_i versus $(R - R_i)/d_c(V_i)$, where R_i is the inner cylinder radius and d_c an adjustable parameter; we also plot two velocity profiles extracted from [18] for the shearing of a dry granular material (1 mm mustard grains) in a Couette geometry ($R_i = 3 \text{ cm}$, $R_e = 6 \text{ cm}$). The line is an exponential fit: $V/V_i = e^{-5.6(R - R_i)/d_c(V_i)}$; the dotted line is for a power-law fluid fit with exponent $n = 0.13$. Inset: d_c versus V_i for a 3.91 cm/s preshear (open triangles) and a 43.5 cm/s preshear (squares).

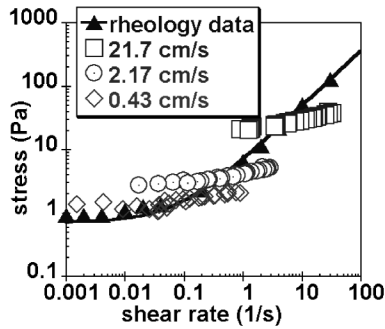


FIG. 6. Stress versus shear rate calculated from the velocity profiles for different rotational velocities and measured macroscopically [same data as Fig. 1(b)].

$\sigma \propto \sigma_i/R^2$. If we combine this with the roughly exponential decay of the velocity profile and calculate a local viscosity from the ratio of the two, it follows that this local viscosity is small near the moving inner cylinder and increases with increasing distance from the moving wall. This is, in fact, easy to imagine, if the particle concentration is slightly smaller near the wall and increases with the radius. The expected concentration profile can easily be evaluated with a Krieger-Dougherty-like model for the dependence of the viscosity on the volume fraction; it is outside of the scope of this Letter, but we plan to measure the concentration profile with the MRI and compare them to what is expected from the velocity profiles.

In conclusion, we have studied the transition between the frictional and lubricated flows of a dense paste. We find that the bifurcation of viscosity completely and unambiguously characterizes the transition between jammed and flowing states under imposed stress and between localized flows and homogeneous flows without localization under imposed shear rate. The bifurcation gives both the critical stress and critical shear rate, the critical shear rate being inversely proportional to the fluid viscosity. We have shown that the critical Leighton number, which follows from the critical shear rate and stress and from the solvent viscosity, completely fixes the transition between the two regimes. In the “frictional” regime, the shear flow is localized, with a shear zone increasing with the macroscopic shear rate unlike what happens for dry granular flows. Moreover, the “lubricated” regime is not simply viscous: the ratio of normal to shear stresses remains constant; the velocity profiles are roughly exponential and can be rescaled in a universal way in the two regimes. Finally, we show that either the material is inhomogeneous or that no simple constitutive relation exists in the lubricated regime; it is in any case impossible to define a macroscopic viscosity that does not depend on the measurement system or the geometry, both in the frictional and in the lubricated regime.

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