Flow of wet granular materials

N. Huang, G. Ovarlez, F. Bertrand, S. Rodts, P. Coussot, Daniel Bonn

1Laboratoire de Physique Statistique, UMR 8550 CNRS associated with University Paris 6 and Paris 7,
École Normale Supérieure, 24, rue Lhomond, 75231 Paris Cedex 05, France

2Laboratoire des Matériaux et Structures du Génie Civil, UMR 113 LCPC-ENPC-CNRS,
2, allée Kepler, 77420 Champs-sur-Marne, France and

3Van der Waals-Zeeman institute, University of Amsterdam,
Valckenierstraat 65, 1018 XE Amsterdam, the Netherlands

(Dated: August 15, 2018)

The transition from frictional to lubricated flow of a dense suspension of non-Brownian particles is studied. The pertinent parameter characterizing this transition is the Leighton number $Le = \frac{\gamma s}{\sigma}$, which represents the ratio of lubrication to frictional forces. The Leighton number $Le$ 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, as in the frictional regime; moreover the velocity profile has a single 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.

PACS numbers: 83.80.Hj,83.60.-a,47.55.Kf

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) 3,4,5,6,7,8.

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 for stresses smaller than a critical stress $\sigma_c$, the viscosity decreases with time; for $\sigma > \sigma_c$, the viscosity increases in time. If shear rate rather than the stress is imposed, below $\gamma_c$ the viscosity jumps from an infinite to a finite and low value at a critical shear rate $\gamma_c$, as observed previously for yield stress fluids and dry granular materials 9,10. Apart from a critical stress, the bifurcation also identifies a critical shear rate $\gamma_c = 0.4 \pm 0.1$ s$^{-1}$, beyond which steady flows are possible under an imposed stress. If shear rate rather than the stress is imposed, below $\gamma_c$...
the measured shear stress is almost independent of the shear rate, which is the hallmark of quasistatic granular (frictional) flow\textsuperscript{1,2}. For high shear rates ($\dot{\gamma} > \dot{\gamma}_c$), stress increases linearly with increasing shear rate, as for a viscous fluid (Fig. 1b)). These observations allow us to identify the transition between two different flow regimes.

To find out what determines the transition between the two regimes, we vary the viscosity of the interstitial fluid. As before, we determine the critical stress and critical shear rate are determined from the viscosity bifurcation. Fig. 2 summarizes the main results. The critical shear rate $\dot{\gamma}_c$ is inversely proportional to the viscosity of the interstitial fluid, whereas the critical shear stress $\sigma_c$ turns out to be constant (within experimental uncertainty). The former result is consistent with that of Prasad and Kytömaa\textsuperscript{4} and Ancey and Coussot\textsuperscript{5}, who only used two fluids of different viscosities.

We conclude from these data that the pertinent parameter characterizing the transition is the Leighton number $Le = \frac{\eta_c}{\eta}$, which represents the ratio of lubrication to frictional forces. Ancey and Coussot previously reported the inverse proportionality with respect to the frictional forces. By varying the lubrication forces, we conclude that the transition is entirely characterized by the Leighton number. The critical stress $\sigma_c$ does not vary significantly ($\sigma_c = 5 \pm 3$ Pa), so that $Le_c$ has the same order of magnitude for $\eta$, 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 roughly that of the low shear viscosity of the suspension multiplied by the critical shear rate. The Krieger-Dougherty model\textsuperscript{11} for hard spheres: $\eta = \eta_s (1 - \phi/\phi_m)^{-2.9\phi_m}$, gives $\sigma_c = \eta_c \approx 1$ Pa. This gives the correct order of magnitude of $\sigma_c$, but also explains why the critical stress remains constant: this follows from combining $\dot{\gamma}_c \propto 1/\eta$ and $\eta \propto \eta_s$\textsuperscript{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 first normal stress difference $N_1$ (neglecting the second normal stress difference) using a plate-plate geometry (diameter 40 mm) show that the normal and the viscous stresses are proportional in both flow regimes. For the plate-plate geometry, the critical shear rate is found to be very similar to what found in the Couette cell ($\dot{\gamma}_c = 0.5 \pm 0.2$ s$^{-1}$). Fig. 3 shows that the ratio $\sigma/N_1$ does not vary significantly with the shear rate $\sigma/N_1 = 0.39 \pm 0.15$ in both regimes, and over 4 decades in shear rate. Previous results in the ‘viscous’ regime are consistent with our findings\textsuperscript{2,4,13}, despite the fact that others\textsuperscript{5,14} have reported different results. Brady and Morris\textsuperscript{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 $\sigma/N_1$ to be equal in the frictional and lubricated regime as observed experimentally. This surprising observation is reinforced by the finding that the ratio $\sigma/N_1$ 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 $\theta$ 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 "MRI rheometer", which allows for a direct measurement of the local velocity distribution in a Couette geometry\textsuperscript{13}.

The main results from these MRI measurements are that the velocity profiles are roughly exponential in dry granular materials\textsuperscript{17}, 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. How-
FIG. 3: Shear/normal stresses ratio $\sigma/N_i$ 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 shadow zone is the statistical error bar. The horizontal line appears off center because of the logarithmic scale.

FIG. 4: Velocity rescaled with the velocity of the inner cylinder (inner cylinder radius $R_i = 4.15$ cm, outer cylinder radius $R_c = 6$ 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 $V_i = 43.5$ cm/s during 30 s. For the low velocity setup, we preshear the material at the maximum possible rotational speed $V_c$, 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 s$^{-1}$ and 20.9 s$^{-1}$, the critical shear rate $\gamma_c$ being 0.4 ± 0.1 s$^{-1}$ (or $V_c = 1.04$ ± 0.26 cm/s).

Moreover, 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 $\gamma_c = 0.4 ± 0.1$ s$^{-1}$, the whole sample is sheared ($\gamma_c$ measured with the MRI is the same as $\gamma_c$ found in the rheology) [17].

In the frictional regime, the behavior is only different 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] and [18] fall along the same universal curve (Fig. 5), underlining the universality of the roughly exponential velocity profiles which goes beyond granular pastes: The inset to 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 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 $\sigma_i$ 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 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. The possible reasons for this is 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 is then: 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 $\sigma \propto \sigma_i/R^2$. 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 paper, 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.

We thank F. Chevoir and F. da Cruz for their data, and A. Goriely for a critical reading of the manuscript.

[1] GDR MiDi, Eur. Phys. J. E 14, 341-365 (2004).
[2] H.M. Jaeger, S.R. Nagel, and R.P. Behringer, Rev. Mod. Phys. 68, 1259-1273 (1996).
[3] R.A. Bagnold, Proc. R. Soc. Lond. 225, 49 (1954).
[4] D. Prasad, H.K. Kytömaa, Int. J. Multiphase Flow 21, 775 (1995).
[5] C. Ancey, P. Coussot, C.R. Acad. Sci. Paris 327, Série II b, 515 (1999); P. Coussot, C. Ancey, *Rheophyse des pâtes et des suspensions*, EDP Sciences, Paris (1999).
[6] P. Coussot, C. Ancey, Phys. Rev. E 59, 4445 (1999).
[7] P. Coussot, Q. D. Nguyen, H.T. Huynh, D. Bonn, Phys. Rev. Lett. 88, 175501 (2002).
[8] F. da Cruz, F. Chevoir, D. Bonn, P. Coussot, Phys. Rev. E. 66, 051305 (2002).
[9] The density of these oils is 0.96 g.cm$^{-3}$. Due to the small density difference we are at a volume fraction of 58%; this small difference turns out to be unimportant; in addition sedimentation effects are small due to the fact that we are at high solid loadings.
[10] The apparent saturation at a finite value of the viscosity is due to the finite resolution of the position transducer of the rheometer.
[11] R.G. Larson, *The structure and rheology of complex fluids*, Oxford Univ. Press (1999).
[12] $Lc \sigma$ does of course depend on other parameters of the system such as the solid volume fraction $\phi$. $\sigma_c$ goes to zero at $\phi \approx 45 \%$. Below this value of $\phi$, the low shear rate 'frictional' regime disappears.
[13] M.L. Hunt, R. Zenit, C.S. Campbell, C.E. Brennen, J. Fluid Mech. 452, 1 (2002).
[14] J.F. Brady, J.F. Morris, J. Fluid Mech. 348, 103 (1997).
[15] Measurements are performed on the oil phase. Cf. J.S. Raynaud et al., J. Rheol. 46, 709 (2002) and S. Rodts et al., C. R. Chimie 7 (2004) for a description of the setup.
[16] D.M. Mueth et al., Nature 406, 385-389 (2000).
[17] This behavior is similar in plate-plate experiments for
dense suspensions. Cf. C. Barentin, E. Azanza, B. Pouligny, Europhys. Lett., 66, 139-145 (2004).
[18] F. Da Cruz, Ph.D thesis, ENPC (2004).
[19] $d_c$ is slightly smaller for a 9 rpm preshear. This is likely to be a sedimentation effect, since resuspension is more efficient at 100 rpm. However, this does not affect the generality of our results; rheometric experiments performed in both conditions show very little difference.
[20] C.Y. David Lu, P.D. Olmsted, and R.C. Ball, Phys. Rev. Lett. 84, 642 (2000).
[21] This result is usual with granular media which often presents great inhomogeneities, but not a priori evident for granular pastes, as these can dilate much less than granular matter due to the interstitial fluid. We also verified that the second conclusion is not due to a size difference between the rheology and MRI Couette cells: rheology done with the MRI Couette cell shows results that are quantitatively similar to those of Fig. 11.