Flowing resistance and dilatancy of dense suspensions: lubrication and repulsion
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I. INTRODUCTION

Granular suspensions, emulsions, foams and blood share a similar microstructure: they are composed of individual particles immersed in a viscous fluid. Their flowing properties are pivotal in various practical issues, such as landslide mitigation, treatment of some cardiovascular diseases, fresh concrete settling or industrial food handling. A few decades of research have evidenced many of their rheological properties, but some great challenges still remain. The flowing behavior of immersed particles in dense configurations is one of them, as highlighted in many recent reviews [2, 6, 13–15, 21].

Many experimental and numerical studies measured the shear resistance to steady flow of dense suspensions. Most often, the particle concentration $\phi$ is selected prior to the experiment and would not vary. The material is then subjected to an imposed shear rate $\dot{\gamma}$ and the shear stress $\tau$ is measured, or vice-versa. Both methods highlight a flowing law in the form $\tau = \eta \dot{\gamma}$; with an effective viscosity $\eta$ that increases with $\phi$ and diverges at some maximum concentration $\phi_m$. Many data satisfy the empirical Krieger-Dougherty law, $\eta = \eta(1 - \phi/\phi_m)^{-n}$, $\eta$ being the viscosity of the surrounding liquid [3, 5, 10, 15, 21]. To date, the exponent $n$ has not been given any clear physical meaning, yet the values that fit the data vary only slightly around $n \approx 2$. By contrast, the maximum concentration $\phi_m$ has a clear physical meaning: in order for the material to be sheared, the particles need some free space to avoid each other. The maximum concentration of spherical, mono-dispersed and rigid particles can be deduced from geometrical analysis. However, such an estimate may be difficult for polydispersed, not perfectly rigid or non-spherical particles [2]. For dense systems, this uncertainty has a critical impact on the viscosity prediction due to the divergent form of $\eta(\phi)$. In practice, the knowledge of the particle concentration is not sufficient to accurately infer the viscosity of the material.

An alternative approach has been recently introduced to express the flowing law of dense granular materials without liquid [2, 4, 10, 13]. Experiments and simulations were not performed under constant volume conditions, but rather under constant normal confining stress $\sigma$, allowing the material to dilate or contract. This loading mode corresponds, for instance, to free surface flows down a slope. The behavior of the material can then be expressed by a friction law and a dilatancy law. To a first order approximation in $\dot{\gamma}$, they read:

$$\frac{\tau}{\sigma} \approx \mu_s + b \phi \dot{\gamma},$$  \hspace{1cm} (1)

$$\phi \approx \phi_s - c \dot{\gamma}.$$  \hspace{1cm} (2)

Here, $t$ is a time scale, discussed below, and $\mu_s$, $b$, $\phi_s$ and $c$ are numerical constants that depend on the nature of the grains, for instance their friction coefficient [1] and their possible cohesion [20]. This flowing behavior is similar to that of a Bingham fluid: it involves a critical shear stress $\tau_0 = \mu_s \sigma$ below which there is no flow, and an effective viscosity $\eta = b \sigma$ Unlike for a Bingham fluid, however, both the critical shear stress and the viscosity of granular matter depend on the normal stress $\sigma$. Besides, the viscosity is proportional to the inertial time $t_i = \sqrt{\frac{\tau_0}{\sigma}}$ built on the confining stress $\sigma$, the grain diameter $d$ and their mass $m$. Introducing this time scale in the friction law provides a robust and generic description of dense granular flows [2, 10]. Combining the friction law [13] and the dilatancy
law \(2\), one can express the flow resistance of dry grains as a function of the solid fraction with no singularity when approaching the maximum packing fraction, thereby avoiding the corresponding uncertainty:

\[
\frac{\tau}{\sigma} \approx \mu_s + \frac{b}{c}(\phi_s - \phi).
\]

(3)

Following such an approach for immersed particulate materials is quite promising. A significant step in this direction has been made by \[4\], in analyzing experimental immersed granular flow down an inclined plane. The flows were shown to satisfy a friction law similar to that of dry granular flows, see Eq. (1). In the limit of a negligible grain inertia, the time scale \(t\) of the friction law has been expressed from the new relevant physical parameters: the normal stress \(\sigma\) supported by the grains and the liquid viscosity \(\eta_0\). The viscous time \(t_v = 3\pi \eta_0 d^3\) thus replaces the inertial time suitable for dry grains. Beyond this robust scaling, however, some questions still remains. First, in the presence of a liquid, no dilatancy law similar to Eq. (2), and hence no regular expression of the flowing resistance such as Eq. (3) has been identified yet. Second, one can expect that the details of the interaction between close particles in a liquid should have a significant effect on their collective behavior. In particular, some surface repulsion may be set into action between close enough particles. One can expect that it should limitate the magnitude of the lubrication interaction \[13\], and that the collective behavior should therefore be different whether or not most pairs of neighbouring particles approach sufficiently to set this surface repulsion into action \[12\].

In this paper, we study the flowing behavior of a dense suspension whose grains interact by viscous lubrication and possible repulsion. The study is based on Soft Dynamics simulations of the plane shear, prescribing the shear rate and the normal stress. We show that the steady flows of this dense suspension can be described by a friction and a dilatancy law, whose coefficients are found to depend on a key parameter of the grains: the distance below which surfaces repel each other. A simple scenario explaining these non-trivial dependencies is proposed, supported by the analysis of the local force distribution.

The paper is organized as follow: the material under consideration and the simulation method are described in section II, the collective response to shear is studied in section III and the internal processes underpinning the collective behavior are discussed in section IV.

II. SIMULATION OF A MODEL DENSE SUSPENSION WITH THE SOFT DYNAMICS

The Soft Dynamics method is a Discrete Element Method which we specifically developed to simulate the dynamics of dense suspensions. In this section, we briefly present its main features, especially the modes of interaction accounted: grain elasticity, lubrication and surface repulsion. More details can be found in our previous works \[15\] \[19\].

Particles in a dense configuration interact with their close neighbours, with a typical surface-to-surface distance \(h\) much smaller than their own size \(d\). They interact by viscous lubrication, which then dominates the hydrodynamic forces [see for instance \[1\]]. Their surface may also interact directly with each other when they get close enough. This interaction can come from some electrostatic repulsion between surfactant molecules at the surface of bubbles or droplets (disjoining pressure). It may also reflect the size of surface asperities in the case of rough solid particles. In any case, under imposed normal stress \(\sigma\), some surface repulsion will generate a typical equilibrium gap \(h_0\) between neighbouring particles.

We chose to model this surface repulsion by a purely normal force in the form of a power law: \(F_{\text{dir}}^n = \sigma d^2 \left(\frac{h}{h_0}\right)^{-p}\), with \(p = 8\) as discussed in \[13\]. More generally, torques or tangential forces between surfaces could be included, as well as complex features such as solid friction, adhesion and interfacial visco-elasticity, depending on the nature of the particles under consideration. The normal and tangential lubrication force between both surfaces is expressed as \[13\]:

\[
F_{\text{vis}} = \frac{3\pi \eta_0 d^4}{2h^3} v_n, \quad F_{\text{t}} = \frac{\pi \eta_0 d^2}{h} v_t,
\]

(4)

where \(v_n\) and \(v_t\) are the normal and tangential relative surface velocities (which correspond to Poiseuille and Couette flow, respectively). Distance \(a \approx \sqrt{d(h + |\delta_n|)}\) is the lateral extension of the gap region that mediates most of the lubrication force. Here, \(\delta_n\), is the normal component of the surface deflection \(\vec{\delta}\) related elastically to the force: \(E_a \vec{\delta} = F_{\text{elas}} = F_{\text{vis}} + F_{\text{dir}}\), where \(E\) is essentially the Young’s modulus of the bulk material. A cutoff value for the gap \(h\) is introduced as a way to avoid long range hydrodynamic interactions which are irrelevant to dense configurations: the particles interact only when \(h < 0.4d\). Viscous and elastic torques are also included, as in \[17\].

The novelty of Soft Dynamics is to simulate the evolution of the elastic deflection \(\vec{\delta}\) and of the twist angle for each pair of neighbouring particles and to take into account how they, in turn, affect the lubrication flow in the
FIG. 1: (Color online) Plane shear of a dense particulate material immersed in a viscous liquid. (a) 5000 spheres, with a slightly polydisperse distribution \((d \pm 20\%)\), are subjected to an imposed shear rate \(\dot{\gamma}\) (imposed through Lees-Edwards boundary conditions) and a fixed normal stress \(\sigma\) in a fully periodic domain. To impose the normal stress, the target normal stress is set equal to \(\sigma\) at \(t = 0\) and the height \(H\) of the simulation cell is set to evolve according to \(M \dot{H} = \sigma - \sigma_{pp}\), where \(\sigma_{pp}\) is the current normal stress and where \(M\) is a mass fixed at a value that corresponds to about one monolayer of particles. Observational normal stress fluctuations are small (less than 1%). (b) Interaction between two close elastic particles in a viscous fluid (see main text for detail). (c) Regimes of viscous (green color) and elastic (red) behaviours depending on the shear rate \(\dot{\gamma}\). Observed normal stress fluctuations are small (less than 1%).

TABLE I: Range of parameter investigated. The unit length, stress and time are the particle diameter \(d = 1\), the normal stress \(\sigma = 1\) and the viscous time \(t_v = 3\pi \frac{d}{\eta} = 1\). The particle mass is set low so that the inertial time is smaller than the viscous time, \(t_i = \frac{\sqrt{\pi d}}{\eta} \approx 0.1 t_v\). The value of the stiffness number \(\kappa\) corresponds either to solid grains subjected to high normal stress \((E \approx 10^5 \text{ Pa}, \sigma \approx 10^6 \text{ Pa})\), or to the softer living cells, droplets or bubbles under lower stress \((E \approx 10^{1-5} \text{ Pa}, \sigma \approx 10^{1-2} \text{ Pa})\). The range of shear rates investigated corresponds to \(10 \text{ s}^{-1} < \dot{\gamma} < 4 \times 10^3 \text{ s}^{-1}\) for a viscosity close to that of water \((\eta = 10^{-3} \text{ Pa.s})\) and an applied normal stress \(10^2 \text{ Pa} < \sigma < 4 \times 10^4 \text{ Pa}\), or for more viscous liquid \((\eta = 10 \text{ Pa.s})\) and a normal stress \(10^6 \text{ Pa} < \sigma < 4 \times 10^8 \text{ Pa}\).

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inter-particle gap. As a result, the elastic, viscous and direct force/torque are balanced at the particle surfaces at all times. Correspondingly, the contributions to the stress \(\sigma = \frac{1}{V} \sum \vec{F} \otimes \vec{t}\), (where \(V\) is the volume of the sample, \(\vec{F}\) the force transmitted between a pair of particles and \(\vec{t}\) their center-to-center vector) arising from normal and tangential viscous forces as well as direct forces can be isolated: \(\sigma = \sigma^{\text{dir}} + \sigma^{\text{vis}} + \sigma^{\text{el}}\).
shear rate $\dot{\gamma}$, velocity $\dot{\gamma}$ and static counterpart. Although we do not observe jamming, the system exhibits two different behaviours (Fig. 1c), always flows at low shear rates, and we measure an effective, dynamic friction coefficient, which may differ from its

\[ \sigma \]

impose the normal stress $(\text{this test are plotted on Fig. 1c: small red crosses correspond to situations where the particles deform and the force...)}$

\[ \gamma_{d} \]

Steady flow versus elastic response. Because we impose the shear rate $\dot{\gamma}$ and allow for material dilatancy (we impose the normal stress $\sigma$ rather than prescribing the volume), there is no jamming in the usual sense: the system always flows at low shear rates, and we measure an effective, dynamic friction coefficient, which may differ from its static counterpart. Although we do not observe jamming, the system exhibits two different behaviours (Fig. 1d), depending on the equilibrium gap $h_{0}$ and on the shear rate $\dot{\gamma}$. For low shear rates or large equilibrium gaps, the shear stress first increases and eventually reaches a steady value, which is similar to a viscous response. Conversely, for smaller equilibrium gaps or higher shear rates, the shear stress increases indefinitely, which is rather similar to an elastic response.

This peculiar collective response can be understood from the microscopic relaxation time at the particle scale. As discussed in [17], each pair of particles behaves like a non-linear Maxwell analogue. The prescribed shear rate tends to separate their centers at a typical velocity $\dot{\gamma}d$. If the shear rate is low enough with respect to the characteristic time of the equivalent Maxwell element, the particle surfaces have enough time to separate from each other (starting from their initial gap of order $h_{0}$) and the material tends to flow in a viscous manner. By contrast, if the flow is faster, many pairs of particle surfaces do not have enough time to get apart, hence the particles themselves deform and build large elastic forces. The material thus tends to respond elastically.

We tested this simple picture by simulating the separation of only two particles at a constant center-to-center velocity $\dot{\gamma}d$, from an initial surface-to-surface distance $h = h_{0}$ up to the cutoff distance $h = 0.4d$. The results of this test are plotted on Fig. 4: small red crosses correspond to situations where the particles deform and the force

FIG. 2: (Color online) Mechanical behaviour of the simulated suspension as a function of the equilibrium gap: $h_{0}/d = 0.1$ ($\times$), 0.08 ($\triangledown$), 0.06 ($\Diamond$), 0.05 ($\square$), 0.03 ($\triangle$), 0.02 ($\bullet$), 0.01 ($\bullet$), 0.008 ($\blacksquare$), 0.006 ($\blacktriangle$). (a) Friction law: shear stress $\tau$ versus shear rate $\dot{\gamma}$ for each value of $h_{0}$. (b) Corresponding slopes $b$ (log-log plot in inset) and (c) $y$-intercepts $\mu_{s}$ of the linear fits $\tau \approx \mu_{s}(h_{0}) + b(h_{0})\dot{\gamma}$.

(d) Dilatancy law: solid fraction $\phi$ versus shear rate $\dot{\gamma}$ for each value of $h_{0}$. (e) Corresponding slopes $c$ (log-log plot in inset) and (f) $y$-intercepts $\phi_{s}$ of the linear fits $\phi \approx \phi_{s}(h_{0}) - c(h_{0})\dot{\gamma}$.

III. COLLECTIVE RESPONSE TO SHEAR: IMPORTANCE OF SURFACE REPULSION

The numerical experiment consists in subjecting a sample of particles interacting as described in Section II to a shear, prescribing the normal stress $\sigma$ and the shear rate $\dot{\gamma}$ (Fig. 1a,b). Both the normal stress $\sigma$ and the Young’s modulus $E$ of the grains are fixed in all the simulations presented here. A value of the equilibrium gap $h_{0}$ is selected prior each run and is kept constant throughout the run. For each value of $h_{0}$, a series of runs with different values of the shear rate are performed. The range of parameters under investigation is presented in Table I. Each test starts from a loose configuration with no interaction or velocity. Both the normal stress and the shear rate are applied at the same time (see caption of Figure 1) and the stress within the simulation cell is measured as indicated above. Under the action of the normal stress, the simulation cell contracts, thus bringing the grains closer to each other. As they start interacting, the shear stress rises.

In the remaining of this work, we will describe and rationalize the observed behaviour with various degrees of detail, from macroscopic response to microstructure properties. The macroscopic behaviour is shown by Fig. 1 (conditions for obtaining a steady flow), Fig. 2 (shear stress and dilatancy) and Fig. 3 (shear stress as a function of volume fraction). The relative importance of each type of force (lubrication versus direct forces due to surface repulsion) is shown by Fig. 3 and Fig. 4 (dominance of viscous versus direct forces). Microscopic data are presented through two types of statistics in Fig. 5 for each type of force (contributions by neighbour rank and angular distribution of contributions).
data to shed light on this apparent loss. We shall therefore now examine local stress, $\tau/\sigma$ (which is the limit of constant, it is anticipated that all forces (including surface repulsion) should scale linearly with the shear rate for shear stress is dominated by surface repulsion or by lubrication. They contribute equally when the shear rate is given by $\dot{\gamma}$, to predict the flow behaviour of such immersed grains.

(i) Normal viscous forces (particle approach or separation) contribute much more than tangential viscous forces (sliding). That is consistent with Eq. (4) and with the dynamics of few particles in [19]. (ii) The viscous shear stress is approximately proportional to the shear rate, $\tau^{vis} \approx b^{vis} \dot{\gamma}$. Note that the corresponding slope varies roughly like $b^{vis} \approx b^{vis} \propto (h_0/d)^{-2/3}$, just as the overall slope $b$. The slope $b^{vis}$ for sliding depends more weakly on the equilibrium gap $h_0$. (iii) The contribution of surface repulsion, $\tau^{dir}$, as opposed to that of viscous forces, does not vanish at small $\dot{\gamma}$ under such imposed normal stress conditions, see figure 4c; by contrast, if the volume was kept constant, it is anticipated that all forces (including surface repulsion) should scale linearly with the shear rate for small shear rates, see [12]. Hence, surface repulsion alone is responsible for the finite frictional flow threshold $\mu_s$ (which is the limit of $\tau/\sigma$ at small velocities). For illustration purposes, Figure 4 shows whether the macroscopic shear stress is dominated by surface repulsion or by lubrication. They contribute equally when the shear rate is given by $\dot{\gamma}^{CL} = \frac{\mu_s}{\sigma^{CL}} \dot{\gamma}^{CL}$. This quantity happens to increase almost linearly with the equilibrium gap $h_0$. (iv) There is a tendency for the contributions of surface repulsion to the normal stress, $\sigma^{dir}$ (and to a weaker extent to the shear stress, $\tau^{dir}$), to decrease with the applied shear rate $\dot{\gamma}$.

Among the above results, points (i), (ii) and (iii) are qualitatively not very surprising. By contrast, point (iv) does not have any straightforward interpretation in terms of macroscopic arguments. We shall therefore now examine local data to shed light on this apparent loss of contact.
FIG. 4: (Color online) Contribution of viscous lubrication and surface repulsion to the stress. Symbols denote the equilibrium gap: \( h_0/d = 0.1 (\times), 0.08 (\triangledown), 0.06 (\circ), 0.05 (\square), 0.03 (\triangle), 0.02 (\circlearrowleft), 0.01 (\bullet), 0.008 (\blacksquare), 0.006 (\blacktriangle). \) Shear stress supported by (a) all viscous forces (normal and tangential: \( \tau^\text{vis} \)), (b) normal viscous forces (\( \tau^\text{vis}_n \)) and (c) tangential viscous forces (\( \tau^\text{vis}_t \)). (d–f) Corresponding slopes \( b^\text{vis} \) from the linear fits \( \tau^\text{vis} \sim b^\text{vis} \dot{\gamma}_t \) (insets: log-log plots). (g,h) Shear stress \( \tau^\text{dir} \) and normal stress \( \sigma^\text{dir} \) supported by the surface repulsion forces.

FIG. 5: (Color online) Microstructure of the steady flow. The statistical indicators presented here are averaged over 50 snapshots during a shear deformation of 5. Results correspond to one material (with \( h_0 = 0.02d \)) subjected to different shear rates \( \dot{\gamma}_t = 0.04 (\square), 0.1 (\circlearrowleft) \) and 0.4 (\blacktriangle). In graphs (a–d), \( N = 1 \) corresponds to an average over the closest gap of each particle, \( N = 2 \) to the second closest gaps, etc. (a) Average gap \( \bar{h} \) for each value of \( N \). (b) Cumulative distribution (in terms of \( N \)) of contributions to shear stress from surface repulsion forces. (c) Similar cumulative distribution for normal viscous forces. (d) Same distribution for tangential viscous forces. (e–h) Angular distribution \( d\tau/d\beta \) of the shear stress mediated by pairs of grains with orientation \( \beta \) for each type of force.
IV. INTERNAL DYNAMICS AND RELATIVE GRAIN MOTION

In order to understand the apparent loss of contact between grains when the deformation rate is increased, let us examine the simulation data from the point of view of the surface-to-surface distance between neighboring grains.

Unexpected stress contribution as a function of surface-to-surface distance. In order to test the influence of the surface-to-surface distance \( h \), we have constructed several averaged quantities displayed in Figures 5a–d. To do that, we determined the closest neighbour of each grain (in terms of \( h \)) at all times. We used the corresponding pairs of grains to compute several averaged quantities labeled with \( N = 1 \). We then determined the neighbour of each grain with the next smallest value of the gap \( h \) and used it to compute values for \( N = 2 \), and so on.

Figure 5 displays the average \( h \) as a function of rank \( N \); it ranges from high \( h \approx h_0 \) for \( N = 1 \) to high \( h \approx 0.4d \) (interaction cutoff). The cumulative distribution displayed on Figure 5 shows that the shear stress arising from surface repulsion, \( \tau^{\text{dir}} \), is mediated almost totally by the first two closest neighbours, suggesting the existence of force chains as in dry granular flows. By contrast, Figures 5c–d show that as many as 10 neighbours contribute significantly to the viscous shear stress. That is rather counter-intuitive when considering that viscous forces vary very strongly with the gap \( h \) (see Eq. 3) and that the latter varies by one order of magnitude between the first and the tenth neighbour (see Figure 5).

Angular distribution of stress contributions and relative grain motion. The surprisingly comparable viscous contribution from each of the first ten neighbours of each grain to the macroscopic shear stress indicates that mutual velocities and orientations of pairs of grains compensate for their gap variability. Therefore, we examined the angular distribution of the pairs of grains that contribute to the macroscopic shear stress. The angular distribution of the total shear stress, shown on Figure 5e, seems rather complex. But the decomposition into surface repulsion, normal and tangential force contributions, see Figures 5f–h, suggests a very simple microscopic scenario for each typical pair of neighbouring particles.

A pair of particles first approach each other around \( \beta \approx 3\pi/4 \), (the angle \( \beta \) is defined in the diagram of Figure 5). As a result, for pairs oriented in this direction, surface repulsion and normal viscous forces both resist macroscopic shear and thus contribute positively, see Figures 5f–g. Once they have approached, both particles then slide around each other when \( \beta \approx \pi/2 \). At such an orientation normal forces (both surface repulsion and viscous forces) do not contribute to the macroscopic shear stress, while tangential shear forces do contribute to it (see Figure 5h). When both particles depart, typically around \( \beta \approx \pi/4 \), surface repulsion helps this motion, hence the (weak) negative contribution in Figure 5f, while normal viscous forces again resist macroscopic shear, see Figure 5k. Such a scenario suggests that at low shear rates, the first few closest neighbours of a given particle are neither approaching nor separating, but rather sliding, maintaining their surface-to-surface distance around \( h \approx h_0 \). Their contribution to \( \tau^{\text{vis}} \) is then expected to be smaller than that of the next few neighbours, consistently with the lower slope in Fig. 5 for \( N < 5 \). Conversely, because they are currently sliding, their contribution to \( \tau^{\text{vis}} \) is higher than that of the next few neighbours, see Fig. 5 for \( N < 4 \).

With the above microscopic scenario, the loss of contact observed in Figures 5k–h can now be understood as follows. While the normal stress \( \sigma \) tends to compress pairs of grains of all orientations, the shear stress \( \tau \) is compressive in some directions (around \( \beta \approx 3\pi/4 \) in the notations of Fig. 5) and tensile in other directions (\( \beta \approx \pi/4 \)). As the shear rate is increased, one might naively think that all velocities and viscous forces are increased and that typical particle trajectories should not be altered. But in fact, the present imposed normal stress conditions imply that the ratio of the shear stress and normal stress \( \tau/\sigma \) increases when the shear rate is increased. Hence, tensile forces within particle pairs become more frequent as compared to compressive forces. That explains why the average surface-to-surface distance increases with velocity (as observed in Fig. 5) and why the stress supported by direct forces decreases at high shear rate in Figures 5i–h. This loss of contact is more significant for small values of the equilibrium gap \( h_0 \).

V. CONCLUSION

The flowing behavior of the present model dense suspension appears to satisfy a friction law and a dilatancy law, similar to those of dry grains. Likewise, the shear to normal stress ratio developed in steady flow are a regular function of the solid fraction (see Eq. 3), with no singularity at maximum packing. The coefficients involved in these laws depend on the surface repulsion through the equilibrium gap \( h_0 \) (see Figure 3).

A closer look at the microscopic data in the simulation has revealed that a simple scenario (see Fig. 5h) captures much of the microscopic distribution of the overall shear stress. In particular, it appears (see Figures 5c–d) that in such a dense, lubricated system, most forces transmitted to neighbouring particles are comparable: locally, particle pairs appear to exchange an imposed force rather than to move at an imposed relative velocity: in a sense, the lubrication film extends each particle in such a way that it is in effective mechanical contact with its neighbours, thus
building a much more robust force network than in a dry granular material.