Rheology of dry, partially saturated and wet granular materials
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5. Rheology of suspensions

Abstract: The rheological characterization of suspensions and in general, complex fluids could be challenging because of the sensitive measurement conditions and procedures. Despite these complexities, because of its obvious importance in a wide range of industrial applications, rheology of suspensions has been an important research topic for many years.

This chapter aims to study the behaviour of dense suspensions of non-Brownian particles in confined geometry. We study the different regimes of suspension flows and the forces between the grains in a suspension in these regimes. We find that the local rheology presents a transition at low shear rate from a viscous to a shear-thickening behaviour with shear stresses proportional to the shear rate squared, as predicted by a scaling analysis.

5.1 Introduction to the rheology of complex fluids

Complex fluids are immensely important in our life, for industry, understanding certain biological processes, and so on. Such complex fluids are mostly granular materials, suspensions of particles such as colloids, polymers, or proteins in a solvent. The majority of these suspensions exhibit shear thinning: the faster the material flows, the smaller its resistance to flow, or apparent viscosity. If the system is sheared, the shear pulls the system over certain energy barriers that the system would not be able to cross without the applied shear; the viscosity consequently becomes small. Because of the generality of the shear-thinning phenomenon, it is interesting to note that exceptions to the rule exist. For certain concentrated suspensions of particles, shear thickening may be observed as an abrupt increase in the viscosity of the suspension at a certain shear rate [108].

The detailed mechanism of this shear-thickening phenomenon is still under debate [32, 108, 111]. For colloidal suspensions, the phenomenon is often attributed to the shear-induced formation of hydrodynamic clusters: in this case, the viscosity
increases continuously as a consequence of its dependence on particle configuration; this may be but is not necessarily accompanied by an order-disorder transition in the particle configuration.

For granular matter, studies of dry granular materials have evidenced considerable activity over the last decade. These works were motivated by the importance of dry granular materials in many practical and industrial situations [97]. So the number of studies on wet granular matter is almost negligible compared to that for dry. In the real world, however, we often see wet granular materials, such as beach sand and the cohesion induced by the liquid changes the mechanical properties of granular materials. The biggest effect that the liquid in granular media induces is the cohesion between grains and it is well known that this cohesion depends on the amount of liquid in the system. We discussed different regimes of liquid content in wet granular media in Chapter 1. Suspensions consisting rigid particles in a liquid and are in the category of the slurry regime of wet granular materials.

The rheological behaviour of concentrated suspensions and granular pastes has been dealt with mainly in three fields which consider different types of materials under various conditions (which might overlap in some cases): rheology of suspensions, physics of granular matter, and soil mechanics. The first field (rheology of suspensions) was developed by physicists somehow with extrapolating the approach of Einstein concerning dilute suspensions of hard spheres to concentrated systems [112]. Thus the viscosity of such systems is related to the viscosity of the interstitial fluid, \( \eta_0 \), the solid volume fraction \( \phi \), and the maximum packing fraction, \( \phi_m \), through various models [113, 114] such as the Krieger-Dougherty model or Zarraga model [111, 115].

Suspension rheology has seen considerable progress due to both experimental and numerical theoretical works which have helped in clarifying the relation between macroscopic stress and the microstructure. However, various disturbing effects during the measurements, such as wall slip, sedimentation, migration, and evaporation make concentrated suspensions and granular pastes precisely the most difficult systems to study with rheometers. The flow of granular materials has been an important topic of hot debate and viscosity, as the determinant parameter, is necessary for many applications to predict the resistance to flow [116].

5.2 Different regimes of sheared granular materials

An important topic in studies on dry granular media is the sustained (non-collisional) contacts in flow regimes. So, there is an identification which is correct in extreme cases, nearly jammed versus very dilute granular media, that led to the implicit reduction of granular rheology to two limiting situations: \textit{quasi-static} that is dominated by contact forces, and \textit{fast flows} in which grains interact only during binary collisions. This issue came into focus after experimental and numerical works provided evidence that Bagnolds scaling (1954) held in various types of dense flows.
Suspension rheology has also been considerable progresses, due to both experimental and numerical/theoretical works, which have helped clarifying the relation between macroscopic stress and the microstructure (Brady and Morris 1997). The non-zero values of the normal stress differences observed in both experiments and the numerical studies are a proof that particle-particle forces are involved [117,118].

5.2.1 Particle-particle contacts in suspensions

Suspension flows can be classified in these three regimes: quasi-static, viscous and inertia. Reynolds (1885) showed that a saturated non-Brownian suspension in a random close packed (RCP) configuration must dilate under shear to sustain continued deformation. This phenomenon is called Reynolds dilatation. At extremely low shear rates and under the application of a normal force, the system would be in the quasi-static regime is dominated by short range frictional interactions between the particles resulting from extended contact. As the shear rate is increased, the interstitial fluid plays an important role and at sufficiently high shear rates, fluid viscosity will govern the behaviour of the mixture. This viscous regime was first studied systematically by Bagnold in 1954, but later studies demonstrated the inaccuracy of Bagnold’s theory. Several theoretical studies (Leighton and Acrivos 1987; Brady and Bossis 1985 [101,119]) have suggested that normal stress cannot be generated in Newtonian suspensions in the absence of particle contact [120].

Actually Bagnold succeeded to use kinetic theory for the scaling between stress and shear rate ($\sigma \sim \dot{\gamma}^2$) first observed in dense granular materials [121]. At still higher values of shear rate, the flow enters the inertial regime which is dominated by collisional interactions between the particles and was first discovered by Bagnold in 1954. The notion that particle-particle contacts may contribute to stress in a wide range of situations, while their presence is not easily seen via macroscopic scaling relations, challenges a simplifying view that is consistently found in the literature. In colloidal suspensions, a transition from shear-thinning to shear-thickening can be seen at low Peclet numbers, $Pe = \eta \dot{\gamma}R^3/kT$, where $R$ is the grains diameter and $\eta$ is the suspending fluid viscosity [34, 97,122]. We have discussed this regime in Chapter 1.

For non-Brownian suspensions (typically, $R > 10 \mu m$ and $Pe > 10^3$), the main types of macroscopic behaviour such as, shear-thinning, viscous and shear-thickening are often interpreted as the dominance of one specific form of interactions: contact forces, viscous forces or collisions. Figure 5.1 shows scaling of these forces between the grains. The scale of contact forces is given by:

$$F_{cont} \sim \mu pR^2,$$

where $\mu$ is the friction coefficient and $p$ is the pressure between the grains. When the grains come into contact, the roughness of the particles preserves a thin layer of fluid of thickness $\epsilon$ which prevents the divergence of the amplitude of lubrication forces. The scale of viscous forces is thus estimated as:
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Figure 5.1: Schematic picture of a suspension and dominant interactions [97].

\[ F_{\text{visc}} \sim \eta R^3 \dot{\gamma} / \epsilon. \]  

(5.2)

Also the collision forces could be estimated as:

\[ F_{\text{colli}} \sim m \dot{\gamma}^2 R, \]  

(5.3)

where \( m \) is the average particle mass.

Determining the transition between these regimes is one of the important debates in investigating the behaviour of the materials. Dimensionless parameters can provide criteria to analyse and specify the boundary conditions. To determine the boundaries between these regimes, we can mention two dimensionless parameters that are constructed from the above estimates: the ratio of viscous force to the contact force which is called the Leighton number \( Le = \frac{\eta R \dot{\gamma}}{\mu \epsilon p} \), and the ratio of collisional force to the viscous force that is the Bagnold number \( Ba = m \dot{\gamma} \epsilon / \eta R^2 \) [97, 116]. In the frictional regime, no steady flow exists without localization. So the scaling and this analysis would suggest that how increasing the shear rate and other parameters affects the dominant forces in the suspension and the transition between different regimes.

5.3 Experimental methods

We studied dense suspensions of non-colloidal spherical particles immersed in a Newtonian fluid. The suspensions were made of spherical polystyrene beads from Dynoseeds with diameter 20 \( \mu \)m, polydispersity < 5%, and density 1050 kg.m\(^{-3}\), suspended in aqueous solutions of NaCl to match the densities of the solvent and
the particle. When the densities of the particles and the fluid are not matched, buoyancy effects appear and density matching ensures that there are no gravity induced contacts and sedimentations. Also we used a little surfactant to change the wetting of the particles. We varied the volume fraction of the particles, \( \phi = V_g/V_t \), defined as the volume of grains \( V_g \) on the total volume \( V_t \), from 56% to 58%.

The experiments were carried out with cup-and-plate geometry on a commercial rheometer. The cup-plate geometry is equivalent to plate-plate geometry with lateral surfaces. It enabled us to undertake the experiments in a confinement system with a constant volume of the sample. The diameter of the plate was 50 cm and the two plates had rough surfaces by sandpaper to minimize the wall slip effect (Fig. 5.2 (a)). Another advantage of this geometry is that we can measure the normal stresses. The rheometer measures a torque \( T \) and a rotation rate \( \Omega \), which are related to the stress and shear rate at the edge of the sample by \( \sigma = 12T/\pi D^3 \) and \( \dot{\gamma} = \pi D\Omega/d \), where \( D \) is the plate diameter and \( d \) is the gap between the plates.

5.4 Results

5.4.1 Shear stress behaviour and constitutive equation

A stress-strain curve \( \sigma(\dot{\gamma}, \phi) \), at a fixed volume fraction is obtained by collecting the measurements of stress and shear rate for different size of the gaps (Fig. 5.2 (b)). The results show that at a critical shear rate \( \dot{\gamma}_c \), a transition from a Newtonian regime to an inertial regime occurs. Figure 5.3 shows the critical shear rate and critical shear stress as a function of volume fraction. As we explained about viscous and inertial regimes in section 5.2.1, the scaling for the viscous (Newtonian) regime shows that \( \sigma \sim \dot{\gamma} \) and in the inertial regime the shear stress is proportional to the shear rate squared; \( \sigma \sim \dot{\gamma}^2 \). The \( \dot{\gamma}^2 \) scaling identifies a regime where particle...
inertia dominates over viscous forces [97, 103]. Here we investigate the viscous-inertial transition (the mechanism that controls these scaling regimes) and the forces acting on the grains in the wet granular media.

The forces between a set of particles immersed in a viscous interstitial fluid introduce hydrodynamic interactions between grains and the contact between rigid grains. The equation of motion of particles’ center of mass $r_i$, governing a granular system is Newton’s equation, which in the absence of external forces is given by:

$$m \frac{d^2 r_i}{dt^2} = \sum_j F_{ij} + F_{visc}^{ij},$$ \hspace{1cm} (5.4)

where $m$ is the mass of particles and $F_{ij}$ and $F_{visc}^{ij}$ are the rigid contact forces and hydrodynamic forces, respectively. The rigid forces $F_{ij}$ cannot introduce by definition any force or length scale and to perform scaling analysis, we need to separate the two limiting cases when either inertial or viscous terms dominate.

The first condition specifies a viscous limiting case that occur when viscous forces are dominant over grain inertia: $\sum_j F_{ij} + F_{visc}^{ij} = 0$ and inertial regime can be identified when grain inertia is dominant: $md^2 r_i/dt^2 = \sum_j F_{ij}$. Both expressions imply that in “viscous” regime $F_{ij} \sim \dot{\gamma}$ and in “inertial” regime $F_{ij} \sim \dot{\gamma}^2$ [97, 103, 123, 124]. This interpretation shows the existence of a crossover between the two simple scaling regimes at low shear rate $\sigma \sim \dot{\gamma}$ (viscous) and high shear rate $\sigma \sim \dot{\gamma}^2$ (inertial). Figure 5.4 shows the shear stress versus shear rate measured for various volume fractions. This figure also shows a dependence of critical shear rate on volume fraction.

Also the scale invariant formalism which lead to an exact scaling analysis of viscous and inertial forces helps to understand why the critical shear rate $\dot{\gamma}_c$ can be so low at high volume fraction and vanishes precisely at $\phi_m$ [97]. So with the form of
5.4. Results

Figure 5.4: Shear stress versus shear rate measured for various volume fractions. Diameter of the beads is 20 µm

stresses related to the viscous and inertial regimes, \( \dot{\gamma}_c \) vanishes (i) linearly with \( \phi \), (ii) at the jamming packing fraction \( \phi_m \).

Figure 5.5 shows the behaviour of critical shear rate of dense suspensions of non-Brownian particles versus volume fraction by macroscopic rheometric experiments [103]. According to this Ref. the crossover between the viscous and inertial regimes which is found by equating the two expressions for the stress, finally leading to \( \dot{\gamma}_c(\phi) \sim (\eta_0/\rho R^2)(\phi_m - \phi) \) and \( \sigma_c \sim \eta_0^2/\rho R^2 \), where \( \rho \) and \( R \) are the particle density and diameter, and \( \eta_0 \) is the interstitial fluid viscosity. These equations and the results of Fig. 5.5 show that \( \dot{\gamma}_c \) vanishes precisely at the jamming packing fraction \( \phi_m \).

5.4.2 Viscosity of suspension

The notion of viscosity of granular systems and suspensions is necessary for many applications to predict the resistance to flow. Figure 5.6 (a) shows the evolution of viscosity curves for suspensions versus shear rate for different sizes of the gap. At low shear rates, a relatively Newtonian behaviour is observed. We therefore observe a clear and well-defined transition from the Newtonian to the shear-thickening regime beyond a critical shear rate \( \dot{\gamma}_c \) that is in addition similar to previous results. Shear-thickening is a category of non-Newtonian fluid behaviour in which the viscosity \( \eta \) defined by \( \eta = \sigma/\dot{\gamma} \) increases as a function of shear rate \( \dot{\gamma} \) or shear stress \( \sigma \) over some parameter range.

Shear-thickening is generally interpreted as the consequence of dilatancy. The results show that for the onset of thickening the smaller gaps leads to the lower
shear rate at which thickening occurs. At a certain shear rate, a very abrupt increase in viscosity is observed, this critical shear rate increases with increasing gap (Fig. 5.6 (b)). Dependency of the critical shear rate for the onset of shear-thickening on the gap of the geometry, can be explained by the tendency of the sheared system to dilate which is a result of collisions between the grains.

It is tempting to see whether the shear-thickening phenomenon itself can be due to the confinement: if the sample is confined in such a way that the grains cannot roll over each other, this could in principle lead to an abrupt jamming of the system. To check this effect, in the rheometer, instead of setting the gap size for a given experiment, one can impose the normal stress and make the gap size vary in order to reach the desired value of the normal stress and we can see the dependence of the gap variation on shear rate. Increase in the gap, allowing the system to dilate [108,125].

## 5.5 Normal stresses

The principal information obtaine from the normal stress measurements. The normal stresses are reminiscent of the Reynolds dilatancy of dry granular matter: when sheared, it will dilate in the normal direction of the velocity gradient. Dilatancy is a direct consequence of collisions between the grains: to accommodate the flow, the grains have to roll over each other in the gradient direction, and hence the material will tend to dilate in this direction. However, in our system, the grains are confined, both between the plates and in the solvent.

The latter provides a confining pressure that is mainly due to the surface tension of the solvent, making it impossible to remove grains from the suspension. As suggested by Cates et al. [17], the confinement pressure associated with this should
be on the order of the surface tension over the grain size, $P_c = \gamma/R$, of the same order of magnitude as the typical normal stresses measured in the experiments near the onset of shear thickening.

In the parallel plate geometry, the upward force on the rheometer is measured and the normal stress $\tau_N$ is obtained by dividing this normal force by the plate cross-sectional area. The normal stress $\tau_N$ is shown in Fig. 5.7 as a functions of shear rate $\dot{\gamma}$ for the measurements in different volume fractions. At low shear rate there is no normal force, and the Newtonian regime is dominant on the behaviour of the suspensions. But beyond a critical shear rate, a sudden large normal force emerges. For this shear-thickening suspensions, we found positive normal stresses, meaning the sample is pushing against the rheometer plates, in agreement with other measurements of shear-thickening [108, 125–127]. When dilation of the granular shear flows is prevented by confinement, shear is instead accompanied by normal forces against the walls.

Hoffman (1982) argued that discontinuous shear-thickening will occur in dense suspensions whenever the particles segregate into layers but are constrained from rotating as groups below the onset stress. While Wagner showed that in this cases the layering is not necessary, because the onset is determined by a point where the shear stress is large enough to shear particles in such a way to cause dilation [125].

5.6 Conclusion

We investigated the rheology of dense suspensions of non-Brownian particles in confined system. When a concentrated colloidal suspension is sheared, particles organize. They exhibit an anisotropic microstructure, which in its turn modifies the flow properties of the suspension. At low shear rate, this coupling is stable, and leads to permanent ordering of the colloids under shear, for example, along
planes parallel to the shear direction. When the shear rate becomes high enough, the coupling may become instable. In that case, transient structures develop and lead to a increasing of the suspension viscosity. Indeed, anisotropic microstructure of the particles leads to imbalance of the bulk stress, and thus to nonzero normal stress differences.

Here, we observed a sharp shear-thickening transition at a critical shear rate $\dot{\gamma}_c$ from a viscous to a inertial regime with shear stress proportional to the shear rate squared $\sigma \sim \dot{\gamma}^2$, as predicted by a scaling analysis. This critical shear rate decreases with increasing the volume fraction and vanishes at the jamming packing fraction $\phi_m$.

Shear-thickening can then be interpreted as the consequence of dilatancy which is a direct result of collisions between the grains. In our system the grains are confined, and to accommodate the flow the grains have to roll over each other and hence the material will tend to dilate. For the onset of thickening we observed that the small gaps of the rheometer, leads to the lower shear rate at which thickening occurs.

We have studied the mechanism of transition between different regimes in suspensions and shear-thickening in these sytems, but some details remain unresolved and this problem warrant further experiments. To investigate possible effects of the frictional stress, heterogenity, etc. We intend to carry out more measurements for a wide range of experimental conditions.

**Figure 5.7:** Normal stress measurements ($\tau_N$) vs. shear rate for different volume fractions of a sample of 20 µm beads.