Rheology of dry, partially saturated and wet granular materials
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4. Flow irreversibility in granular suspensions

Abstract: Slow, viscous flows are perfectly time-reversible. However, recent experiments have shown that if a particle paste is subjected to slow oscillatory shear, the particle motion becomes irreversible above a certain critical deformation, the origin of the irreversibility being unclear. In this chapter by Magnetic Resonance Imaging to measure the homogeneity of the suspensions we show an irreversible migration of particles from high-into low shear rate regions. Second, we find that, above a critical deformation large normal forces appear in the suspensions pointing to frictional contacts between the particles. Such contacts also lead to irreversibility in the motion of the particles, and in addition give a quantitative criterion for the onset of irreversibility.

4.1 Introduction

One of the famous experiments of G. I. Taylor is the illustration of the reversibility of Stokes flow. In the experiment, a drop of ink is injected inside a viscous fluid in the gap between two concentric cylinders. When the inner cylinder is rotated several turns, the ink spot is no longer visible. However, when the same cylinder is rotated in reverse exactly the same number of turns, it reappears again. Also, theoretically, it is easily shown that the motion of an incompressible fluid at low Reynolds number, governed by the Stokes equation, is time-reversible.

When the suspended particles are small enough, the inertia of the fluid and particles are unimportant in determining their dynamics. We refer to these systems as Stokesian suspensions, as the fluid motion is governed by the Stokes equations. Unless the particles are very small, the effect of colloidal forces and Brownian motion can be discounted. The Stokes equation is obtained from the Navier-Stokes
Flow irreversibility in granular suspensions

Equation for incompressible fluids simply by omitting the non-linear term and the time derivative of the velocity field:

\[ \nabla p + \eta \nabla^2 u = 0 \]
\[ \nabla \cdot u = 0 \]  (4.1)

If we assume that the velocity field \( u(r) \) is a solution of the equation, with a corresponding pressure field \( p(r) \), \(-u(r)\) will also be a solution provided only that we reverse the sign of the pressure gradients, as well as that of the velocities, at every solid boundary. Eq. 4.1 is then again satisfied, since its two terms are replaced by their negatives and the boundary conditions are appropriately changed [5,89]. One may therefore anticipate that, at a low Reynolds number, every incompressible fluid should behave reversibly. It consequently came as a surprise that particles in suspensions of non-Brownian rigid particles can fail to return to their initial position under oscillatory shear [89,90]. In the experiments of references [89,90], rather concentrated suspensions were subjected to a large-amplitude oscillatory shear; one would then naively expect that after every oscillation all particles retrieve their initial position. However, in the experiments it was found that there exists a maximum strain value \( \gamma_{0m} \), above which the particle motion is irreversible. Understanding the mechanism that leads to this irreversible motion is currently the subject of debate [89–95].

In this chapter, we investigate in detail what happens if the same non-Brownian suspension that was used in [89,90] is again subjected to an oscillatory flow. These experiments unambiguously demonstrate that there is an irreversible migration of particles from high into low shear rate regions. We use Magnetic Resonance Imaging (MRI) [96] to measure the homogeneity of the suspensions. The experiments using by MRI techniques have been carried out in collaboration with the Statistical Physics Laboratory in Paris.

Second, above a given strain \( \gamma_{0c} \simeq \gamma_{0m} \) value [90], we find that large normal forces appear in the suspensions. Those forces are the signature of a transition between a viscous regime, in which the particle contacts are lubricated, and a collisional regime in which the contacts are frictional [97]. It is evident that frictional contacts will also lead to irreversibility in the motion of the particles [98]. In addition, the onset of frictional behaviour gives a criterion for the onset of irreversibility that agrees reasonably with both the experiments and the numerical simulations of [90].

4.2 Magnetic resonance imaging

Magnetic resonance imaging (MRI) is a technique to visualize the structures of a sample in detail and can provide nice colorful 3D pictures of it. Also NMR (nuclear magnetic resonance) is a similar technique for obtaining more detailed chemical information about molecules. It is known that most illustrations of the magnetic resonance imaging technique occur in the medical field. But it has different applications in chemical engineering, fluid mechanics and fluid transport in porous...
4.3 Flow irreversibility in granular suspensions under large amplitude oscillatory shear

Here, we investigate in detail what happens if a non-Brownian suspension subjected to an oscillatory shear flow. I have carried out the experiments by parallel plate geometry to find the transition from reversible to irreversible behaviour. These results show that above a critical deformation large normal forces appear in the suspensions pointing to frictional contacts between the particles.

Also we use Magnetic Resonance Imaging (MRI) and Couette geometry to measure the homogeneity of the suspensions. These experiments unambiguously demonstrate that there is an irreversible migration of particles from high into low shear rate regions.

4.3.1 Experimental techniques

We studied the rheological properties of isodense granular suspensions composed of non-Brownian spherical particles immersed in a Newtonian fluid. The granular material was made of monodispersed polystyrene spheres (diameter \( d = 140 \pm 5\% \), density 1050 kg.m\(^{-3}\)) from Dynoseeds. We used a mixture of distilled water (19% in weight) and Triton (81% in weight) [90] as the interstitial fluid in order to get a
perfect density matching, as checked by centrifuging the samples: no sedimentation or creaming occurred.

The first studies and experiments on reversibility of non-Brownian suspensions have been carried out by applying a sinusoidally strain with a fixed frequency in a parallel plate geometry (with diameter 50 mm). This geometry enables us to measure the normal forces appear in the suspensions pointing to frictional contacts between the particles. Such contacts could show the irreversibility in the motion of the particles, and give a quantitative criterion for the onset of irreversibility. The sample with the liquid volume fraction ranged between 30% and 60% is subjected to oscillatory shear $\gamma(t) = \gamma_0 \sin \omega t$ at a fixed frequency 1 Hz.

Straining the system to strain amplitude $\gamma_0$ evolves the system forward in time; by reversing the flow in the next quarter cycle, we can check to see if the particles return to their initial positions.

Other experiments of classical rheology measurements were performed with Couette geometry (inner cylinder radius $R_i = 4.15$ cm, outer cylinder $R_0 = 6$ cm, height 11 cm) on a classical rheometer that controls the stress or the strain (Fig. 4.1 (b)). The volume fraction was fixed at 40% for all of the experiments discussed here, as in [89,90]. In the following, we define that the strain amplitude corresponds to the ratio of the maximum azimuthal translation of the inner cylinder and the size of the gap. It means that when the sample is subjected to oscillatory shear at a fixed frequency $\omega$ and a fixed strain $\gamma_0$, the time dependence of the strain $\gamma_0$, is given by $\gamma(t) = \gamma_0 \sin \omega t$. The strain amplitude $\gamma_0$ ranged between 0.5 and 3.5 and the frequency between 10 mHz and 1 Hz. Rough lateral surfaces are used in order to avoid wall slip effects. The order of magnitude of the roughness was the scale of one grain diameter. The largest Reynolds number we reached was then around 1.

The error in the measurement of the volume fraction is around 0.25% for MRI experiments, expected very close to the wall where the error is in the order of a percent.

4.4 Results

4.4.1 Viscosity and particle migration

Returning to the rheology experiments, during one experiment, the total accumulated strain experienced by the particles over a run is $\gamma = 4\gamma_0 \times n$, where $n$ is the number of cycles (because the strain is $\gamma_0$ in each quarter cycle) and in rescaled form is given by:

$$\gamma = 4\gamma_0 \times n \times (R_0 - R_i)/d,$$

where $d$ is the grain diameter. Performing oscillatory experiments at fixed strain and frequency, we observed a slow decrease of the complex viscosity (Figs. 4.2 and 4.3). Returning to the complex representation of the oscillatory motion, as an alternative to the complex shear modulus, we can define complex viscosity as:

$$\eta^* = \sqrt{G'\omega^2 + G''\omega^2}/\omega.$$
4.4. Results

Figure 4.1: Oscillatory experiments by using: (a) MRI technique to measure the local volume fraction and (b) Couette geometry to measure the complex viscosity.

Similar observations had been made for suspensions under steady state flow long ago [101], and were interpreted as shear-induced particle migration: the particles have a tendency to move from high to low particle pressure regions due to the unequal frequency of collisions: if a shear rate gradient exists in the sample there will, on average, be more collisions on the high shear rate side than on the low shear rate side of the particle, causing a slow drift towards the low shear rate side [101]. Recently, a viscosity decrease, concomitant with a non-uniform volume fraction inside the gap of a Couette cell after shearing for a long time at a constant rate, was also observed directly using MRI [102]. Using the same MRI technique we can also measure the local volume fraction in an oscillatory flow.

In the classical rheology experiments, the migration, evident as a slow decrease in the measured viscosity, is very slow, necessitating total deformations of several tens of thousands [103–106]. Note that we did not observe a plateau for long periods of times (Figs. 4.2 (a) and 4.3 (a)), as expected, if only migration occurs. However, since the experiments were very slow, the latter was very likely due to sedimentation or creaming; we very carefully matched the density at a given temperature, but small changes in the temperature of the laboratory will cause a density mismatch that is visible over very long times periods as a slow change in the viscosity.

Whether the apparent viscosity goes up or down depends on the precise geometry of the experimental setup; in general the apparent viscosity increases in time (a detailed investigation of a similar system was published recently [107]). However if there is a dead volume into which the particles may cream or sediment, the viscosity may go down. The dead volume is present in our experiment; however we
Figure 4.2: The suspension is subjected to an oscillatory shear flow in Couette geometry for different strain amplitudes $\gamma_0$ from 0.05 to 3.5 at frequency $f = 0.05$ Hz. (a) The paste has a volume fraction $\phi = 40\%$. Time evolution of the complex viscosity, (b) Rescaled complex viscosity $\eta^* = \sqrt{G''^2 + G'^2}/\omega$ as a function of the total deformation $\gamma = 4\gamma_0 \times n \times (R_0 - R_i)/d$. The curves are rescaled by the asymptotic value for the highest total deformation.

cannot rule out that the slow decrease is also partly due to slow migration. More recently, a second, and faster type of particle migration was observed in a system similar to the one discussed here; the fast migration there was attributed to the normal stresses [101–103].

Also we carried out the MRI experiments with the same Couette geometry as for the rheology (Fig. 4.1 (b)). The inner cylinder was driven by a motor that allowed us to adjust the amplitude and frequency of the oscillation. We performed the oscillations outside of the scanner and subsequently gently introduce the Couette geometry into the MRI apparatus to verify the homogeneity of the sample [96]. The measured MRI signal is proportional to the density of protons. By choosing a suitable frequency, the MRI scanner only detects the protons inside the liquid (not inside the polystyrene beads). We were then able to measure the local volume
4.4. Results

The suspension is subjected to an oscillatory shear flow in a Couette geometry for different strain amplitudes $\gamma_0$ from 0.05 to 3.5 and frequencies. The paste has a volume fraction $\phi = 40\%$. (a) Time evolution of the rescaled complex viscosity. (b) Rescaled complex viscosity $\eta^* = \sqrt{G' + G''}/\omega$ as a function of the total deformation $\gamma = 4\gamma_0 \times n \times (R_0 - R_i)/d$. The curves are rescaled by the asymptotic value for the highest total deformation.

**Figure 4.3:** The suspension is subjected to an oscillatory shear flow in a Couette geometry for different strain amplitudes $\gamma_0$ from 0.05 to 3.5 and frequencies. The paste has a volume fraction $\phi = 40\%$. (a) Time evolution of the rescaled complex viscosity. (b) Rescaled complex viscosity $\eta^* = \sqrt{G' + G''}/\omega$ as a function of the total deformation $\gamma = 4\gamma_0 \times n \times (R_0 - R_i)/d$. The curves are rescaled by the asymptotic value for the highest total deformation.

The measured density profiles show that the particle concentration is homogeneous initially, but after a long time of shearing the volume fractions is significantly lower close to the moving inner cylinder, and increases roughly linearly with the distance from the axis (Fig. 4.4). This confirms the idea that the viscosity decreases with time inside the gap due to particle migration. The migration automatically generates a local variation of the viscosity, from which the observed global decrease of the viscosity results.

It is worth while stressing that the total deformation these samples have undergone is strictly zero, showing directly that the migration process is irreversible, even for lower strains. This is therefore a possible explanation for the observation of irreversibility in the previous experiments.
We found that the amount of migration and hence the value of the complex viscosity depends only on the total strain (see Figs. 4.2 (b) and 4.3 (b)). For different oscillation frequencies, the viscosity values collapse on to a single curve when plotted as a function of the strain. This means that migration even occurs for the lowest strain amplitudes. In Figs. 4.2 (b) and 4.3 (b), the complex viscosity curves are rescaled by the asymptotic value for the highest total deformation; as already mentioned, the observed small differences are likely to be due to residual sedimentation or creaming effects [107]. The conclusion from these experiments is that even for small deformations under $\gamma_0$, the shear-induced diffusion results in an irreversible flow for long-time scales. It is worthwhile underlining that the migration happens in the radial direction, whereas the particle trajectories observed in [89,90] were determined in the plane perpendicular to this direction; thus we cannot directly compare the two data sets.

In addition, although it may very well be that for small amplitude oscillations, the time over which the migration occurs becomes so large that in practice it is not visible, this does not account for the relatively well defined critical strain beyond which the flows were irreversible in the earlier experiments [89,90]. There should be another mechanism that probably acts simultaneously with the migration.

**Figure 4.4:** Density profile of the granular suspension (volume fraction $\phi = 40\%$) in the gap of the MRI Couette cell at different time. The open symbols correspond to the initial density profile and the dashed line is a guide for the eye. Circles are taken at $t = 30$ minutes and square at $t = 60$ minutes. The frequency is $51.5$ mHz and the amplitude of the deformation is fixed at $\gamma_0 = 15$. 

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**Chapter 4. Flow irreversibility in granular suspensions**
4.4.2 Critical strain for irreversibility

The transition from reversible to irreversible behaviour is fundamental to statistical physics.

![Graphs of shear stress and normal force vs. strain](image)

**Figure 4.5:** (a) Shear stress measurements as a function of the applied strain $\gamma_0$, (b) Normal forces measurements as a function of the applied strain $\gamma_0$ at a given frequency $f = 1 \text{ Hz}$ in parallel plate geometry, (gap is fixed at 2 mm) with rough surfaces.

To further investigate this, we looked in more detail at the rheological behaviour of the system. The deviation from Newtonian behaviour of Stokesian suspensions has been measured by study of normal stress and the phenomenon of shear-induced particle migration in suspensions is also closely related to normal stress. From a fundamental perspective, normal stresses in a non-colloidal Stokesian suspension are worthy of study because they are the most important non-Newtonian characteristic it exhibits.

To investigate possible normal stresses in the sample and because it is extremely difficult to measure normal forces in Couette geometry, oscillatory measurements were taken in a circular 50 mm diameter plate-plate geometry. The rheology measured in the Couette cell is very similar to that in the plate-plate geometry. However, the geometry of the measurement cell will influence both the migration and the strain at which the normal forces will occur. For the former, the gap size of the Couette cell determines the stress gradient and thus the migration speed [101]. For the latter, the confinement is known to influence the transition between the two regimes [108].

The result for the normal force as a function of the applied strain is shown in Fig. 4.5(b): at low strains there is no normal force, but beyond a rather well-defined critical strain, all of a sudden a large normal force emerges. This figure also shows that that migration is extremely rapid by increasing the volume fraction. The order of magnitude of the normal stress becomes exactly that of the viscous stress; $N = F_N/A \approx 0.05/(2.5 \times 10^{-2})^2 \text{ Pa} \approx 100 \text{ Pa}$ (A is the surface area of...
the plate) whereas the viscous stress is also around 100 Pa. The fact that the two stresses become very similar clearly indicates that beyond the critical strain, we are dealing with a frictional system. However, since our measurements are inherently non-stationary, it is difficult to say something quantitative about the friction coefficient.

The emergence of the normal stresses is therefore the hallmark of frictional behaviour; indeed theory predicts a crossover from lubricated (viscous) behaviour to collisional (frictional) behaviour upon increasing the strain rate [97]. This is also what happens in our experiment: at fixed frequency, an increase in the strain amplitude also implies an increase in the strain rate. We therefore observe a clear and well-defined transition, at a critical strain $\gamma_{0c}$ that is, in addition, similar to that in the earlier experiments of [89, 90]. It appears evident that if the interactions between the grains are frictional, their motion can no longer be reversible, as is also confirmed by simulations on systems similar to the one studied here [98].

The results in this Ref. show that a manybody suspension of near hard spheres in Stokes flow loses reversibility as a result of particle interactions. The observed tendency toward hydroclusters and perhaps solid contact leading to efficient stress transmission is thus seen to be a natural consequence of the highly anisotropic structure which causes the particle phase to generate normal stresses.

We studied the dependence of this critical strain $\gamma_{0c}$ on the volume fraction (Fig. 4.6). The data are in reasonable agreement with the earlier findings of Pine et al. using numerical simulations that accounted for the experimental finding of [90] and shows the descending behavior of critical strain with increasing volume fraction. However, in our case, $\gamma_{0c}$ appears to diverge around a volume fraction of 20%, whereas the data of Pine et al. [90] $\gamma_{0c}$ diverge for a somewhat lower volume fraction.

![Figure 4.6: Critical strain beyond which the normal forces appear as a function of the volume fraction for the experimental measurements.](image-url)
We propose the following explanation for the dependence of the critical deformation on the volume fraction. By considering the existence of a frictional regime, where dynamic contacts are formed, the confinement pressure should play an important role [108, 109]. 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. [110], the confinement pressure associated with this should be on the order of the surface tension over the grain size. We then expect that the critical strain $\gamma_{0c}$ would correspond to the balance between viscous stress and confinement pressure [103, 107–109] and consequently verify \[ P_{\text{conf}} = \eta(\phi)\gamma_{0c} = \eta(\phi)\gamma_{0c}\omega. \]

Here, the confinement pressure is due to the fact that the beads are trapped in the liquid, leading to $P_{\text{conf}} \sim \gamma/R$, with $\gamma \simeq 20$ mN/m the surface tension of the liquid and $R$ the radius of the particles. This directly leads to an estimate for the critical strain $\gamma_{0c} \sim 200/\left(1.4\pi \times \eta(\phi)\right)$ as a function of the volume fraction, that is in good agreement with our experimental data (Fig. 4.6), without any adjustable parameters.

4.5 Conclusion

We have shown by a combination of MRI experiments and classical rheology what the characteristics are of the irreversible behavior in an oscillatory flow of granular suspension.

For a low $Re$ non-brownian suspension, however, reversing the boundary motion is equivalent to reversing time. Thus, we have an opportunity to test the limits of reversibility experimentally: we could undertake a quantitative version of Taylor’s experiment and measure the degree to which particles return to their initial positions. Here we reported a study of reversibility, which is made by straining a viscous suspension periodically in circular Couette flow and plate-plate geometries. Even if the total deformation undergone by the samples is zero, during the repeated deformations particles migrate from the rotating inner cylinder to the stationary outer cylinder. The migration depends on the accumulated deformation and thus may not account for the observation of a relatively well-defined critical strain for irreversibility in earlier experiments by Pine et al. [90]. So we present experimental measurements of the normal stresses in sheared suspensions. This was achieved by applying a sinusoidally strain with a fixed frequency, and using a parallel plate geometry. The experiments show that the interactions in the system can lead to either reversible or irreversible motion, depending on the amount of deformation that is imposed on the suspension. We found that there is a strain dependent threshold which particles do not return to their starting configurations after some cycles. What can account for the existence of such a critical strain is the onset of frictional behavior, as evidenced by the sudden increase of normal stresses at a critical strain
that strongly depends on volume fraction. An important conclusion of the work was that migration is extremely rapid as $\phi \to \phi_{max}$. The emergence of normal forces strongly suggests that dynamical contacts are formed in the suspension.

In the absence of nonhydrodynamic particle interactions, the shear stress is linear in the shear rate, yielding a Newtonian shear viscosity. Normal stress can arise only if there is anisotropy in the microstructure and irreversibility occurs only above a well-defined threshold strain amplitude, which depends strongly on the concentration of the suspension. This should be the signature of the formation of dynamical clusters.