Solid-fluid Duality and Bimodal Flows in a Packing of Soft, Slippery Particles

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We build a rheometer that allows simultaneous observations of stress response and internal grain movements in a packing of hydro-gel particles. Using cyclic shearing combined with static intervals, we demonstrate the solid-fluid duality at high volume fractions, and the relaxation of shear stress originated from stochastic grain-level movements inside the packing. Counterpart steady-state experiments reveals two modes of flows with a crossover shear rate consistent with the timescale of relaxation. Comparisons to harder, frictional particles lead to reconsiderations of the operative definition on solid-fluid transitions, in accommodating flows of the full spectrum of granular materials.

Dense packings of deformable grains are ubiquitous not only in a variety of industries but also in all biological systems including ourselves. However, their dynamics are still poorly understood in comparison to many established paradigms in granular dynamics such as collisional flows \([1–3]\) and fluid-mediated viscous flows \([4]\) in which grain deformations are unimportant, or various jammed systems in which particles move incrementally at quasi-static limits \([5–7]\). In a few two-dimensional experiments, stress relaxations and their relations with plastic rearrangements are resolved \([8–10]\); the force and pressure have been studied in depth at both microscopic and macroscopic levels \([11]\). However, the dynamics of dense grains in the context that particles are kept in elastic, enduring contacts still present considerable challenges: For instance, unlike molecular systems with thermal activities that provide their characteristic times, the intrinsic timescales for these dense macroscopic grains are yet to be identified, before specific dimensionless parameters can be defined in analogy to the Deborah number(s) for non-newtonian fluids.

In this Letter, we use hydro-gel particles as a generic expression of deformable grains at the frictionless limit in 3D, and study both the stress response and internal movements under shearing. In addition to the solid-fluid duality surveyed at a wide range of volume fractions, we have identified an empirical timescale that explains the crossover between two dynamical regimes for their flows at high densities. Further evidence calls for refining how we understand solid versus fluid, in accommodating flows formed by a broader class of materials.

Setup, the duality, and relaxation – Shown as in Fig. 1(a), our rheometer consists of two cones made with a stacking of acrylic structures forming a rough surface at the scale of our particles while imaging through the cones are allowed. The upper cone is suspended at a fixed height and rotating at programmable angular speed \(\Omega(t)\). Our hydro-gel particles have a nominal diameter \(d=1\text{cm}\) with a elastic moduli around 75kPa with a frictional coefficient less than 0.01 , and are stable in the timescale of days. Unless specified otherwise, particles are fully immersed in water with about 1.7 percent of PVP-360 (polyvinylpyrrolidion) to achieve a density match that prevents sedimentations. Frictions are negligible, as they are slippery either against the wall or among each other, with or without the fluid. Fig. 1(b) shows the typical cyclic driving imposed by a stepping motor, with the shear rate being \(\Omega(t)/2\tan\beta\) under the assumption of uniformity \([16]\). A static period \(\Delta_{\text{OFF}}\) is inserted between the steady shearing in opposite directions, with a sharp transition at the order of 10ms. We use six force sensors to support the container, from which the time-dependent torque and normal force are determined. The corresponding stresses as functions of time define the four characteristic stresses shown in Fig. 1(c): both the shear

![Image](https://example.com/image.png)

**FIG. 1.** (a) Schematics of setup, with a 3D construction of the roughened cones; (b) Shear rate of a cyclic driving with a waiting period \(\Delta_{\text{OFF}}\); (c) Response in torque and normal force that defines the plateau \((\sigma_{zz}^{(P)}, \sigma_{zz}^{(P)})\) and residue \((\sigma_{zz}^{(R)}, \sigma_{zz}^{(R)})\), at a volume fraction \(\approx 0.6\). The smooth curves represent best fits with exponentials to the average over multiple cycles.
Results are based on the average over 25 cycles, while the variations are comparable to the noise floor, with the exception of a barely detectable noise floor. We have also found a substantial time span for the relaxation of shear stress at each transition around the state of a solid that sustains a shear stress indefinitely. We have also found a substantial time span for the relaxation of shear stress at each transition around the state of a solid that sustains a shear stress indefinitely. Remarkably, unlike reported experiments with dense colloidal suspensions [12, 13], the residue $\sigma_{zz}^{(R)}$ here is robust and persist beyond our longest measurements [17]. This illustrates the solid-fluid duality: the packing is forced to flow (yield) when the shearing is on but at any instant, as soon as the driving is off, can also return to the state of a solid that sustains a shear stress indefinitely. We have also found a substantial time span for the relaxation of shear stress at each transition around 1s, well above that for the deceleration of our driving boundary.

In our experiments, we vary the volume fraction by changing the number of particles $N$, with a nominal volume fraction, $v_{1}/V_{total}$ in which $v_{1}$ stands for the volume of a single particle (under the approximation that $v_{1}$ is unchanged under squeezing) that is determined to the accuracy of percent. See Fig. 2 for effects on the four characteristic values and note the trends: as the volume fraction decreases, the drop of stress residues is notably steeper than that of the plateau values. Understandably, the stress residues should vanish before the hydrodynamic interactions. However, we have also run experiments during and after the removal of the interstitial fluid and found that, interestingly, these stresses show detectable difference by roughly a factor of two not only in flows but also in the residues, suggesting that the immersion might somehow change the effective elasticity and affects details of the packing, while the solid-fluid duality is robust.

**Self rearrangement of grains during $\Delta_{OFF}$ — Internal imaging allows us to monitor particle movements deep inside the packing.** Using techniques developed previously [14, 15], this is achieved by dying the particles with a small amount of Nyle Blue, setting a 1mm-thick horizontal laser sheet (635nm in wavelength) that goes through the small gap between the two cones at the mid-height, and taking video images through the bottom of our rheometer. Fig 3(a) shows a typical fluorescent image of the particles with the excitation light filtered. Time-elapsed sequences reveal that, at high volume fractions, these particles undergo substantial movements that last considerably longer than the deceleration of the driving boundary but return to quiescence well within 2s. Demo movies are available online [18].

These rearrangements appear quite inhomogeneous, and are at a scale smaller or at most comparable to the grain size. We characterize the accumulation of rearrangement by subtractions of the final frame recorded in each waiting period from each images, and calculate the total intensity, averaged over multiple cycles, as functions of the elapsed time $t-t_{1}$, as shown in Fig. 3(b-c). The decays over time are surprisingly similar between the cases of $v_{1}/V_{total}$=0.71 and 0.62, and the dependence on shear rate is only within a transient of 0.4s or less. Despite the interference of image noise, we determine that the characteristic time for these stochastic self rearrangements is at the order of 1s that is insensitive to the packing density.

At lower volume fractions, grains gradually lose their...
contacts, and the case nVF=0.53 reaches a marginal state as described in Fig. 2: images reveals that the higher shear rate (1.10μs⁻¹) induces considerable collisions and some of the clustered particles continue to rearrange during ΔOFF to relax the stress, but the lower shear rate (0.14μs⁻¹) does not – images show no recognizable rearrangements during the waiting period but a small drift that is quickly damped (by the fluid between these nearly close-packed particles).

Two regimes of elastic flows, a crossover – We also perform counter-part experiments with the conventional long steady shearing, but with a wider range of volume fractions and shear rates. Interestingly, at high volume fractions dominated by elastic contacts, Fig. 4(a) shows a crossover at τ_c⁻¹ ≈ 1s⁻¹, above which these mean stresses exhibit a Bingham-type linear dependence on the shear rate. This characteristic shear rate is noteworthy, as it reveals a time τ_c ≈ 1s that matches both the typical timescale of self rearrangements we have derived from internal imaging and of the relaxation of shear stress towards its residue. We believe that the matching is not a coincidence but implies a separation of two regimes of dynamics: (1) the slow mode, in which the local rearrangements are almost complete or comparable with the macroscopic straining – the stress thus created reflects a balance with the thoroughness of adjustments among the grains, or (2) the fast mode, in which the accumulation of strain is much faster than the relaxation and the increase of stress with shear rate reflects primarily the hydrodynamics.

Even though a complete explanation on this empirical timescale τ_c demands further work, we believe that the behaviors are generic for elastic packings of particles at the frictionless limit under shear. Furthermore, based on dimensional analyses, one can define

\[ S_{PG} \approx \rho d^2 \dot{\gamma}^2 / E \]

in which \( E \) stands for elastic modulus of the particles, with a numerical factor taking into account the packing fraction. This measures the spatial density of kinetic energy imposed by the driving, as particles are forced to overcome each other, against that of the elastic response. We see it as assessing the importance of softness of these packed grains at shear rate \( \dot{\gamma} \). From this, we anticipate that the time distribution of local rearrangements, unless divergent, should scale inversely with \( \sqrt{E} \) and so does the characteristic time \( \tau_c \). We will revisit this assumption after reviewing further experimental information. Our steady-state experiments are also performed in a much wider range of volume fractions than that of Fig. 2: the gradual transition towards the dilute-regime suspension dynamics is shown in Fig. 4(b).[20]

Comparisons to harder materials – To connect with more “traditional” granular materials, we also run experiments using silicon-gel particles of the same size, which has a Young’s modulus roughly 20 times that of our hydro-gel particles. They are also immersed in fluid (with NaCl) at a matching density, with detectable friction. Firstly, Fig. 5(a) reveals that, at a relatively low nVF (≈0.45) compared to the hydro-gel counterparts, the packing of silicon-gel particles already exhibits a substantial resistance to shear, with a dramatic growth in stress: at increments of volume fraction by merely 4 percent, the mean values already exceed those for hydro-gel particles with nVF as high as 0.71. In addition, these curves show no signs on a scaling of \( \tau_c \) in shear rates or a recognizable crossover. Moreover, the fluctuations are large and comparable to the mean. This aspect is presented more intuitively by Fig. 5(b) as we compare the responses in cyclic experiments: unlike hydro-gel particles showing well defined plateaus and residues, the response with silicon-gel particles bears a poor resemblance to the stepwise shear rate as the signals fluctuate strongly. In addition, the residues \( \sigma^{(R)}_{xz} \) are substantial but have a considerable distribution that is comparable to their mean. Lastly, Fig. 5(c) characterizes an important contrast in fluctuations: the shear stress and normal
stress are strongly correlated for Si-G, whereas no recognizable correlations are found for Hydro-G in all cases even at nVF as high as 0.71 at which grains are in tight contacts.

In retrospect, one may interpret the packing of our hydro-gel particles, when at rest, as forming a “weak” granular solid that is featured by an almost unique residue in shear stress reflecting only the prior direction of straining. This packing can be driven into a relatively smooth fluidic state, that is featured by re-arrangements at the local scale with a sharply defined \( \sigma_{xx}(\gamma) \) for every shear rate \( \gamma \) but with a separation into two regimes by \( \tau_c^{-1} \). On the other hand, the flows of silicon-gel particles, featured by strong fluctuations of macroscopic forces, favor the pictures of percolating force chains (shear jammed, [5]): the highly inhomogeneous spatial patterns might challenge the concept of “\( \sigma_{xx} \)” for a system of our size, but well explains the strong correlation between the fluctuations in torque and normal force— as both can be dominated by just a few strongest chains [7] that extend from the upper boundary to the bottom but can change catastrophically upon a minute movement of the boundary. We may coin this state of flow as a quasi-solid. In addition, while it may be up to future work to decide whether these silicon-gel particles present a crossover at higher shear rates, it is clear that, as we switch off the shear rate, the widely distributed residues \( \sigma_{zz}(R) \) are sensitively reflecting the state of force network upon the stopping. We interpret the static state thus created as a “strong” solid.

We believe that a finite friction coefficient \( \mu \), combined with a higher elastic modulus, is responsible for the dramatic changes between silicon-gel and hydro-gel particles, and we propose that the dimensionless pair, \( S_{PG} \) and \( \mu \), is likely the minimal set of parameters governing the generic behavior of deformable grains under shear, at each packing density. Probing whether a small but finite \( \mu \) would make the time distribution of grain rearrangements change significantly, or even diverge, might be an interesting starting point.

To summarize, we combine stress measurement and internal imaging in studying sheared packings of soft grains, exploring the solid-fluid duality with a wide range of volume fractions. We have identified an empirical timescale based on grain-level rearrangements that explains the separation of two regimes of the flow of packed deformable grains at the frictionless limit – also as a legitimate test ground for theories. Comparisons to behaviors with harder and frictional particles that are commonly used in prior experiments provoke thoughts on refining the definition of solid versus fluid, and provide clues on establishing a framework for accommodating flows beyond prior attentions, especially when enduring, elastic contacts are taken into account.

![Figure 5](image_url) FIG. 5. Comparisons using silicon-gel (Si-G) versus hydro-gel (Hydro-G) particles. (a) Shear stress measured in the long uni-directional experiments, as in Fig. 4, as functions of shear rate. For Si-G, the nVF is presented in terms of a characteristic volume fraction \( \phi_0 \), and the root-mean-squared fluctuations are indicated by bars on the symbols (representing the mean). For Hydro-G, volume fractions are indicated by slanted numbers, and rms fluctuations are comparable to the size of the symbols. (b) Responses in cyclic shearing experiment defined in Fig. 1. For Si-G, note the fluctuating values of \( \sigma_{zz}(R) \) with \( \delta \) representing the typical variation, and circles highlighting dips in \( \sigma_{zz}(t) \) upon each reversal of the boundary movement. (c) Correlation between the fluctuations of the two stress components \( \sigma_{xx} \) versus \( \sigma_{zz} \), during long uni-directional shearing (both at 1.75s \(^{-1} \) with signals smoothed over 0.1s to filter the acoustic noises). The correlation coefficients are presented as \( r^2 \) on the graphs.
[16] By tracking tracer images (to be published elsewhere) we have verified that, despite the inhomogeneous rearrangements, the time-averaged velocity of grains is roughly linear with respect to $z$ so that the average strain is reasonably uniform.
[17] We have established that the residues in shear stress are insensitive to the shear rate during the steady driving and to the accumulated strain (as long as being greater than unity), and are robust against deliberate reversals of the driving equivalent to at least -1/25 in strain upon the stopping at a high volume fraction. Our longest records also reveal that a residue in shear stress can persist beyond 24 hours, except for a relatively small decrease reflecting a much slower thermal dynamic aging of the material. See our Online Supplements below.
[20] These experiments also provide baselines for assessing the maximal possible contribution of fluid on the shear stress in the slow mode of the high-VF (elastic) flows, from extrapolations of the data at high shear rates. Also note that all values of $\sigma_{xz}(0)$ we obtain are above the residues in the cyclic experiments. The values do go down as the shear rate decreases, but the logarithmic plot in Fig. 4(b) reveals that they do not approach the cluster of residues. This suggests that the slow mode still has a fundamental difference from a static packing (solid).