Shear Flow and Relaxation of Soft Granular Particles at Controlled Volumes

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We study the responses of fluid-immersed hydrogel spheres to both steady shearing and cyclic cessations. Particles are slippery, deformable, and density-matched with interstitial fluid between geometrically roughed cones, allowing a survey of phenomena with volume fractions ranging from 0.26 to 0.71. At high volume fractions, measured steady-state flow curves reveal a nontrivial trend towards the dynamic yield stress, calling for an extension of the paradigmatic theories in granular-fluid flows. We observe the timescales of relaxation upon shear cessations and discuss their possible interplay with an imposed shearing. All assessments are accompanied by evidence from simultaneous refractive index-matched imaging that captures the particle rearrangements in the interior.

Soft-matter systems, in many circumstances, exhibit solid-fluid duality. They can be driven to flow steadily and accommodate indefinite amount of shear strain. Meanwhile, the same systems may exhibit a solidity with a resistance to shear, namely, a yield stress. Monitoring the development of yield stress has been a commonly used criterion for the emergence of solidity over the change of controlling parameters including, but not limited to, the density or temperature of suspensions 1,2. The solidity is sometimes presented in the form of a residual stress upon the cessation of driving. Stress residue has been a widely studied subject in glassy systems such as synthetic clays or microgel consisting of highly deformable droplets 3,4 or in numerical studies where particle deformations do not play an active role 5, both revealing a wealth of phenomena involving relaxations at different scales.

Granular materials, packed at high densities, present interesting examples showing such duality. In the past decades, studies using shear flows have established dimensionless numbers and their combinations that successfully capture behaviors of dry grains and particle-fluid mixtures for a wide range of volume fractions φ up to jamming points 4,6. Pointed out in Ref. 6, prior experiments and numerical studies are mostly stress-controlled, whereas the current paradigm in theories has assumed Newtonian viscosities, i.e., constituent relations in which the stress varies linearly with the shear rate with a φ-dependent prefactor to describe the slow limits of these flows. Interestingly, it seems that such important constituent assumption has not been tested against volume-controlled experiments so far. In addition, hard-sphere theories would inevitably lose their prediction power above jamming (notably with the random-close-packing φc ≈ 0.635 beyond which flows are strictly prohibited), as a direct consequence of the divergence in constituent coefficients. However, as an everyday experience, it is not uncommon that particles of finite rigidities can be driven to yield and flow with φ > φc. How we understand the development of yield stress over the change of volume fractions and driving rates remain profound challenges up to date. One objective of this work is to offer clues from our experiments, combining steady shear and cyclic cessations at controlled volumes, to address these issues.

Setup and steady shearing — Shown as in Fig. 1a, our particles fill the space formed by two cones that are geometrically roughed at the scale of the particle with diameter d=1cm, surrounded by a smooth glass container as the sidewall with an inner diameter 2R=23cm. The upper cone is set to rotate at a fixed height and with an angular speed Ω that is driven by a programmable stepping motor, at a spatial resolution of 10−4 rotation. Six independent force sensors are used with the specially designed base to determine the complete forces and moments that are required to resist the shearing and keep the system stationary, providing key information about the torque and normal force as functions of time, with a cut-off at around 40ms in response. The gap between the rotating boundary and the sidewall is 2mm, and that between the two tips of the cones is 1mm, such that no particles can go in except with fluid going through with negligible resistance. The nominal volume fraction is defined as VF= Nψ/Vtotal in which N = O(105) is determined by particle counting, while Vtotal and the mean particle volume v1 are determined separately by Archimedes method, to an accuracy of percent. Data discussed in this work are based on hydrogel particles that are commercially available 11. Using a standard rheometer, we have confirmed that the response of these hydrogel spheres to normal compression is well described by a Hertzian contact with the elastic moduli in O(10¹) kPa. Meanwhile, we have determined that the friction coefficients between the hydrogel surfaces and with the sidewalls are less than 0.01, by other independent tests. For all data discussed in this paper, particles are fully immersed in water with about 1.7 percent of PVP-360 (polyvinylpyrrolidon) added to achieve density match that prevents sedimentation. For several representative values of VF, Fig. 1(b) shows the measured steady-state shear stress, σS ≡ 3 < torque > /2πR³.
plotted against the effective shear rate $\dot{\gamma} \equiv \alpha \Omega / 2 \pi$ in which $\alpha = \pi / \tan \beta \approx 13.7$ and $< ... >$ stands for the average over $O(10^2)$ in accumulated strain. The same data are presented in linear and logarithmic plots, respectively.

Two features are noteworthy for these flow curves. (1) The linear plots show that, even though the two dense cases ($VF=0.71$ and 0.62) bear the appearance of Bingham clays (linearity with an offset) for data at high shear rates, their data below $1s^{-1}$ show clear deviations from the linear relationship (represented by dashed lines) that would extrapolate to a false yield stress. On the other hand, one might hope to fit these data with Herschel-Bulkley (HB) models $\sigma - \sigma_y \sim \dot{\gamma}^n$ with a non-integer exponent; it is obvious, however, that HB model cannot fit those data above $1s^{-1}$ simultaneously. (2) The logarithmic plots of Fig. 1(b) show the trend of flow curves over a wide range of $VF$. With particles being neutrally-buoyant, data with $VF$ below 0.44 exhibit a power-law dependence that shares the exponent $\approx 1.7$ which might be specific to the geometry [12], whereas the fact that the two sets of data at the dilute limit ($VF=0$) appear insensitive to the eight-fold change of fluid viscosity suggests the dominance of inertia in these limiting cases. Above 0.44, the flow curves exhibit transitional behaviors with no identifiable Newtonian plateaus; the effects of inertia and viscosity are comparable in the fluid dynamics as indicated by the calculation of Reynolds number (to be discussed shortly below). Further increase in $VF$ illustrates the development of yield stress: It is not our intention to identify a critical value of $VF$. Nevertheless, we believe that the flow curves as shown with our selections of $VF$ (from 0.49 up to 0.71) do reveal a generic trend, that should serve as a starting point for extending the current paradigm of theories [7–10] — toward the proper description of “jammed flows” in which a yield stress (rather than that associated with a Newtonian viscosity) dominates the force in response to shearing.

In addition, we measure the normal stress $\sigma_N \equiv < F_z > / \pi R^2$. At high $VF$s and slow shearing, the rate dependence is qualitatively similar to that of $\sigma_S$. In Fig. 1(c), we plot the stress ratios against three dimensionless variables. Given the density matched at $\rho$ and fluid viscosity $\eta=8mPa\cdot s$, the particle Reynolds number is defined as $Re_p \equiv \rho(d/2)^2 \dot{\gamma} / \eta$, for measuring the inertial stress relative to the viscous stress around individual particles, and can be seen as a non-dimensionlized substitute for $\dot{\gamma}$ in Fig. 1(b) above. The numerical values as displayed on the graph indicate that our selections of shear rate for experiments with $VF=0.49$ and above make these experiments in the transitional zone where inertia and viscosity are comparable in the hydrodynamic interactions. However, both hydrodynamic forces are small in relation to stress created by direct contacts, as shown by the calculation of the other two dimensionless numbers: $J \equiv \eta \dot{\gamma} / \sigma_N$ measuring the viscous stress against the total pressure, and $I^2 \equiv \rho(d/2)^2 \dot{\gamma} / \sigma_N$ assessing the role of inertia in creating the stress — see Ref. [8, 9] and typical values on the two abscissas here. Both representations of the stress ratio as a function of driving rate show a clear distinction between data for $VF=0.49$ and those for higher-$VF$, and we anticipate that perhaps a suitable combination of $J$ and $I^2$ (as discussed in Ref. [10]) might give even better collapse on the data for $VF=0.53$ and above. The stress ratio

FIG. 1. (a) Vertical cross-section of the setup, definition of symbols, and an angled view of the cone structures. (b) Shear stress $\sigma_S$ as functions of the shear rate $\dot{\gamma}$. Values of $VF$ are shown by slanted numbers on the right. Data are time-averaged over strain accumulations over 100 for each rate, plotted in linear and logarithmic scales, respectively. Values of $\sigma_S^{(d)}$ are also adapted from Fig. 2d for $VF=0.71$ and 0.62, to indicate lower bounds of yield stress. In the logarithmic plots, data with pure fluids ($VF=0$) are displayed with crosses for the standard PVP solution, and with plus signs for water, even though they are overlapped. (c) Stress ratios $\sigma_S / \sigma_N$ plotted against three dimensionless parameters as defined in main texts.

- $\alpha = \pi / \tan \beta \approx 13.7$ and $< ... >$ stands for the average over $O(10^2)$ in accumulated strain.
- $\alpha = \Omega / 2 \pi$
- $\dot{\gamma} \equiv \alpha \Omega / 2 \pi$
- $\rho(d/2)^2 \dot{\gamma} / \eta$
- $\eta \dot{\gamma} / \sigma_N$
- $\sigma_N \equiv < F_z > / \pi R^2$
- $Re_p \equiv \rho(d/2)^2 \dot{\gamma} / \eta$
- $J \equiv \eta \dot{\gamma} / \sigma_N$
- $I^2 \equiv \rho(d/2)^2 \dot{\gamma} / \sigma_N$
- $\eta=8mPa\cdot s$
\[ \frac{\sigma_S}{\sigma_N} \] here are generally between 0.1 and 0.2, substantially lower than reported values (around 0.3). Whether such difference suggests effects from particle deformation, or depends somewhat on flow profiles in different experimental systems, demands further work to clarify.

**Cyclic shearing, residual stress, and relaxations**—To probe the intrinsic response of the packing without sustained boundary movements, we perform experiments with cyclic shear cessations (CSC). Our CSC is defined by two characteristic times \( \Delta_{On} \) and \( \Delta_{Off} \) plus a shear rate \( \dot{\gamma}(\text{p}) \) imposed alternatively in two directions, as shown in Fig. 2(a). By precision optical measurements, we have verified that the rotating boundary reaches a complete stop well within 10ms upon each stopping (\( t_{\text{Stop},2n} \) and \( t_{\text{Stop},2n+1} \)). Fig. 2(b) shows the typical response in torque and normal force that can be converted to shear and normal stress, respectively, in analogy to that defined in steady-state experiments. The nearly periodic patterns define four characteristic values: two plateaus \( (\sigma_S^{(P)}, s_N^{(P)}) \) and two residues \( (\sigma_S^{(R)}, s_N^{(R)}) \). Results from experiments at five different volume fractions are displayed in Fig. 2(c-d).

Measuring the true yield stress \( \sigma_y \) as a function of VF requires experiments at the zero-speed limit and can be hard to achieve. However, in Fig. 2(d) we have shown the estimated trend, based on that (1) \( \sigma_y \) must be bounded between measured values of \( \sigma_S^{(P)} \) and \( \sigma_S^{(R)} \), and that (2) analogous experiments using molded hydrogels with higher elastic moduli (not shown) reveals, as VF approaches 0.55+ from above, both values exhibit a steep drop below the noise level. In the case of VF=0.53, only \( \sigma_S^{(P)} \) is barely detectable and is not shown on the graph.

In addition, we monitor the stress response with a long stall after the CSC sequence, in order to identify timescales involved in the intrinsic response – see Fig. 2(a). Firstly, in the initial stage up to \( t = 10^3 \text{s} \), the stress ratio \( \frac{\sigma_S}{\sigma_N} \) shows a significant reduction of anisotropy, going from its steady-state value (around 0.2, Fig. 1I) toward 0.05 — further evidence from our internal imaging (to be discussed with Fig. 4) reveals that this is mainly due to the rearrangement of particles. Secondly, at timescales around \( 10^2 \text{s} \) and up, both \( \sigma_S \) and \( \sigma_N \) decay proportionally with time, with their ratio showing no significant changes – our examination of images also reveal no identifiable particle movements over such long time.
One plausible explanation for such isotropic decay of stress is a result of the internal evolution of the hydrogel itself over time. To test this hypothesis, we place single hydrogel particles under a standard rheometer (Anton Parr MCR-302) and monitor their response to static compression as time evolves. The protocol and results are presented in Fig. 3(b), with data from three randomly selected particles. In each run, the particle is compressed at \( d - c_0 \) that matches the initial condition \( F(0^+) = 0.06N \) which is about 10 times of \( \delta^2 \sigma_N \) to account for the load-bearing particles inside a static packing. We confirm that (1) the hydrogel maintains its strength up the timescale of \( 10^2 \)s, so that our choice of \( 10^{\Delta g} \), in CSC experiments are justified; and that (2) hydrogel particles do weaken significantly at the timescales of \( 10^3 \)s and above. The observation explains the proportional decays in Fig. 3(a) beyond \( 10^2 \)s, in which the difference between particles seems to average out such that the reduction of stress appears isotropic.

Evidence from internal imaging – Fluorescent internal imaging allows us to monitor the rearrangement of particles during relaxations \[ \text{[14].} \] Notably, transient rearrangements have been predicted in numerical studies with parameters for microgels \[ \text{[8], whereas our experiments present a visual demonstrations but in the context of granular flows. Here, we stain these hydrogel particles with Nyle Blue, and illuminate them at the mid-height by a 1mm-thick laser sheet at a wavelength 635nm \text{— see Fig. 4(a) showing a close-up of the bandpassed image (at 656±10nm). For high-VF experiments, the accumulated displacements of every particle after each stopping are much smaller than \( d \).} \] We therefore define \( D_\infty(t) \) as the image at time \( t \), subtracted by some reference frame much later at which movements between adjacent frames are already below pixel noises. The two images as shown in Fig. 4(b) illustrate the rapid but measurable decay of particle movements. We also calculate the phase-averaged intensity, by integrating \( D_\infty(t) \) over the field of view and averaging over multiple cycles — see Fig. 4(c). Interestingly, the time evolutions are surprisingly similar between the cases for \( VF = 0.71 \) and \( VF = 0.62 \). Nevertheless, there are subtle differences over the change of \( \dot{\gamma}(P) \) at each VF — such differences provide a vivid illustration for the prior studies with microgel that the outcome of relaxations also depends upon rates of the preshearing \[ \text{[3].} \] Scenarios change dramatically as VF decreases, with video clips available online \[ \text{[16].} \] At values of VF as low as 0.44, particles become well separated such that the motion last much longer due to the lack of damping: the total displacements of each particles can easily exceed \( 1d \) and, for this reason, \( D_\infty(t) \) is no longer a good indicator of the differential movements.

Role of relaxations on steady-state stress— Transient measurements have provided us additional clues in understanding steady states. In a steady shearing, the inverse of the shear rate \( \dot{\gamma} \) defines a time, which can be seen as the maximum for local configurations to stay intact. Therefore, the fact that flow curves deviate from linear extrapolation of Bingham model at shear rate around \( 1s^{-1} \) and below (Fig. 1b with \( VF = 0.71 \) and 0.62) suggests additional mechanism of relaxation at play if contacts are maintained longer such characteristic time — and this indeed coincides with our observation with Fig. 3(a), showing significant reduction of stress from \( 10^0 \)s and up, and with simultaneous evidence from imaging (Fig. 4) revealing prominent rearrangements within \( 1s \). Additionally, the long-time trend in Fig. 3(a), combined with our supporting experiments using single particles (Fig. 3b), have demonstrated substantial effects from the evolution of the materials, at timescales of \( 10^3 \)s and up. These in turn explain why these flow curves for seem unable to reach a flat “quasi-static” limit even at shear rates as slow as \( 1/10^2 \)s (Fig. 1b, in logarithmic scales).

Concluding remarks — Using hydrogel particles that are soft and slippery, we study their response to shearing in a wide range of volume fractions. At high volume fractions at which rigid particles are expected to jam, the steady flow curves of the soft-particle packing reveal a non-trivial trend toward the dynamic yield stress,
whose true values can be bounded by supplemental experiments with cyclic cessations. The flow curves also convey messages on the competition between shearing and relaxations in which both particle rearrangements and material evolution are involved. While clear separations on different timescales of relaxation would require further work, the use of hydrogel particles provides easily accessible data as a stepping stone toward extending the current paradigm in granular-fluid flows – to the regime in which direct, enduring contacts between particles dominate the mean stress.

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