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Drop formation in shear-thickening granular suspensions

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We study droplet formation in granular suspensions by systematically varying the volume fractions (φ) and particle diameters (d). For suspensions with water as the suspending liquid, we find three different regimes. For dilute suspensions (φ ≤ 45%), drop formation follows the predictions for inertial breakup and exhibits identical dynamics to that of pure water. The breakup is strongly asymmetrical in this case. Only for more concentrated suspensions (φ > 45%) does the presence of particles change the dynamics and two other regimes, a symmetrical inertial regime and a Bagnoldian regime, are uncovered. We construct and discuss a phase diagram that allows us to understand and predict the breakup behavior in granular suspensions.

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Drop formation is essential for many industrial applications and processes [1]. For Newtonian fluids, liquid neck breakup leading to drop formation is well understood: it is governed by a competition between capillary forces that drive the breakup and viscous and/or inertial forces slowing down the fluid flow in the neck [1]. The detailed understanding of the phenomenon relies on finding the similarity solutions for the shape of the fluid neck that connects the drop to the orifice, and that eventually breaks up in a finite time [1].

On the other hand, drop formation in non-Newtonian fluids, which is important in areas such as emulsification, inkjet printing, and agricultural spraying, is still ill understood in many cases [1–4] and continues to attract considerable attention [5–13]. For instance, theoretical analysis predicts that the drop breakup in shear thickening fluids proceeds faster than that in the Newtonian case due to the high elongational rates present in the fluid neck during the thinning [14,15]. To the contrary, experiments show that in shear thinning and very strongly shear thinning yield stress fluids, the breakup dynamics can be described completely by the equations for the breakup of simple fluids [6], whereas other experiments do report a signature of the shear thinning behavior [7,16,17]. A very recent paper shows that nonlocal rheology may play a role in these yield stress fluids [18].

Here we consider an important class of complex fluids: granular suspensions made of solid particles homogeneously dispersed in a simple Newtonian liquid. The viscosity of these suspensions can be simply increased by adding particles without affecting the density or the surface tension of the suspensions [19], making these systems excellently suited for a systematic study. Different questions then arise: what is the relation between shear and elongational rheology, does the size of the particles affect the breakup dynamics, and what is the role of the suspension concentration? To answer these questions we use a system whose rheology in shear flow is simple: if the suspensions are density matched they behave as Newtonian liquids at low shear rates, and show shear thickening at higher shear rates [20,21]. Recent studies show that during breakup different scenarios may occur [22]: the breakup may be viscocapillary but governed by either the solvent or the suspension viscosity [8–10]. Also new regimes are found that are dramatically different from the predictions for simple fluids [11–13]. In highly concentrated granular suspensions [11] the thinning of the neck was found to follow a power law versus time to breakup with an exponent of 2/3, which alone would be the inertial breakup; however with a counterintuitive symmetric breakup geometry. It was argued in [11] that the force balance takes place more locally here and that the capillary pressure exerted at the level of the individual particles protruding from the interface is balanced by the fluid inertia. Exponential thinning is also observed in colloidal and cornstarch suspensions: the thinning dynamics proceeds more slowly than that in the Newtonian case and the thinning neck becomes cylindrical [12,13]. The overall picture is still not clear and the different mechanisms at play deserve further systematic study.

In this paper, by systematically varying the particle diameters (d) and volume fractions (φ), we establish a phase diagram for suspension breakup. Three regimes for the drop breakup occur: an inertial regime identical to that of the solvent (water), a second inertial regime in which the breakup is still inertial but becomes up-down symmetric, and a third, which we refer to as a Bagnoldian regime, that likely corresponds to a shear-thickened state (with the emergence of significant normal stresses) of the material. The transitions between the different regimes are identified and possible criteria for the transitions between the different regimes are suggested.

The granular suspensions used in the experiments are prepared using poly(methyl methacrylate) (PMMA) particles (d = 1.3, 6, 10, 15 μm) with a density ρ ≈ 1.19 g/cm³ or polystyrene (PS) particles (d = 20, 40, 80, 140, 250, 500 μm) with ρ ≈ 1.05 g/cm³. To avoid sedimentation or creaming, we prepare density-matched suspensions by dispersing particles in pure water, in which the salt NaCl (purchased from Sigma Aldrich) is previously dissolved to adjust the density to that of the particles. The viscosity (η) of the suspensions is increased by simply increasing the volume fraction φ. Colloidal suspensions are prepared by dispersing 1.3 μm PMMA particles in density-matched mixed solvents, cyclohexyl bromide (CHB) and decalin, which has a similar viscosity to that of water (η ≈ 1 mPa s).

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2\(\gamma/\rho\) face tension changes upon varying the volume fractions. The surface tension does change [19,20,23], but this should be irrelevant as stated in the introduction, neither the density nor the surface tension changes upon varying the volume fractions. The scale bar is 1 mm. (b) Left: prefactor \(\alpha\) from fitting the thinning curves in (a). Right: asymmetry coefficient. Inset: Reynolds number Re vs \(\varphi\).

FIG. 1. (Color online) (a) Thinning dynamics 2\(R_{\text{min}}\) vs \((t_0 - t)\) for water and 40 \(\mu\)m suspensions with \(\varphi\) varying from 45\% to 59\%. Continuous lines: power law fit with 2\(R_{\text{min}} = \alpha(t_0 - t)^{2/3}\) [Fig. 1(a)]; \(\alpha\) is indistinguishable between the two methods of fitting and is shown in Fig. 1(b). Note that for low volume fractions this prefactor is similar to that for water (0.27 mm/s\(^{2/3}\)) but drops to a smaller value for \(\varphi \geq 50\%\). Further, there is a major, visual change in the shape of the interface at breakup at different volume fractions \(\varphi\): for small \(\varphi\) the breakup is strongly asymmetric, whereas for large \(\varphi\) it becomes symmetric [Fig. 1(a) inset] and this happens at a larger scale instead of a single-particle level. The asymmetry coefficient defined in [6,26] shows a clear and sharp transition from asymmetric to symmetric breakup around \(\varphi \approx 45\%\) [Fig. 1(b)]. Here the observation of the 2\(3\) law thinning dynamics together with the symmetric breakup is in agreement with the observations in [11].

The main difference between concentrated and dilute suspensions may be that for the former particles are more likely to deform the interface during the breakup, as also observed for shear flow due to dilatancy [27]. As discussed in the introduction, the local force balance at the level of the individual particles leads to the scaling 2\(R_{\text{min}} \sim (\Lambda \gamma d/\rho R_0)^{1/3}(t_0 - t)^{2/3}\) [11], which resembles the usual inertial scaling but includes an extra factor \((\Lambda \gamma d/R_0)^{1/3}\) with \(R_0\) the syringe radius, and \(\Lambda\) an additional constant that accounts for the characteristics of the particles at the interface such as the contact angle and the depth of immersion. This is then likely to be the cause of the changed prefactor \(\alpha\) in Fig. 1(b), allowing us to calculate the constant \(\Lambda\); as an example, for \(\varphi \approx 59\%, \Lambda \approx 0.114\), close to the reported value [11]. In fact, all the data with \(\varphi \geq 50\%\) collapse on a single master curve at \(\Lambda \approx 0.1\) if \(R_{\text{min}}/(R_0 \Lambda^{1/3})\) is plotted vs \((t_0 - t)/(\rho R_0^2/\gamma d)^{1/3}\) (Fig. 2), suggesting that the breakup in all concentrated suspensions falls in the symmetric inertial regime.

As stated in the introduction, neither the density nor the surface tension changes upon varying the volume fractions. The viscosity does change [19,20,23], but this should be irrelevant for the inertial breakup dynamics. Consequently, the viscous forces are considered to be negligible not only in the inertial
but also in the symmetric inertial regimes, as viscosity is absent in both scalings. To examine whether the viscous forces can indeed be neglected, we estimate the ratio of the inertial to the viscous forces for the last instant before breakup: the Reynolds number can also be estimated as $\text{Re} = \rho v_t R_{\text{min}}/\eta$; here $v_t = d(2R_{\text{min}})/dt$ is the neck thinning velocity, and $\eta$ is obtained from shear rheology. We take an average velocity in the final stages (the last few milliseconds) of the thinning curves to estimate $\text{Re}$. This Reynolds number can also be estimated as $\text{Re} = (R_{\text{min}}/l_v)^{1/2}$ with the viscous length scale given by $l_v = \eta^2/\rho \gamma$; viscous forces should dominate the dynamics when $R_{\text{min}}$ is smaller than $l_v$. As shown in Fig. 1(b) inset, the estimated $\text{Re}$ decreases with increasing $\phi$ (increasing $\eta$). Thus, for high concentrations, for which $\text{Re} < 1$ and despite the fact that for a substantial part of the thinning dynamics the neck radius $R_{\text{min}}$ is smaller than the viscous length scale $l_v$, the thinning does not happen at constant speed, but rather as $2R_{\text{min}} \propto (t_0 - t)^{2/3}$. For Newtonian fluids, if $R_{\text{min}} < l_v$, one would, expect $2R_{\text{min}} \propto (t_0 - t)$; the thinning happens at constant speed.

In addition, our experiments show that the change of regimes is observed to happen around $\text{Re} \approx 1$. This is surprising since in classical theory for Newtonian fluids, for $\text{Re} \approx 1$ the drop breakup should follow viscous-inertial-capillary dynamics which is linear in time and asymmetric in breakup geometry [26]. The exact significance of this result is not obvious at present and may suggest that the mechanism behind the symmetric inertial regime works only when viscous forces are important even though they do not enter explicitly into the force balance.

Measurements on systems with different particle diameters show the generality of the symmetric inertial regime; typical data for the thinning of the neck in 6 $\mu$m suspensions show that, similarly to what happens for the 40 $\mu$m particles, as $\phi$ is increased ($50\% < \phi < 55\%$), the thinning curves deviate from that of water and again fall in this regime. However, for these small particles, yet another regime exists. If volume fraction is increased beyond $\phi \sim 55\%$, the breakup dynamics shows a very different behavior. As shown in Fig. 3(a), before the final breakup, the thinning dynamics slows down for $2R_{\text{min,l}} \leq 2R_{\text{min}} \leq 2R_{\text{min,u}}$ with the critical neck diameters $2R_{\text{min,l}}$ and $2R_{\text{min,u}}$ being the lower and the upper end of this regime, respectively. The positions of these diameters (inflection points) can be obtained from the time dependence of the thinning velocity $d(2R_{\text{min}})/dt$, as shown in Fig. 3(b). Upon increasing the volume fraction, $2R_{\text{min,l}}$ decreases from 0.19 to 0.09 mm, while $2R_{\text{min,u}}$ increases from 1 to 1.4 mm. The temporal dynamics in this regime can be fitted well by the exponential scaling $2R_{\text{min}} = A \exp [(t_0 - t)/\tau]$, where $A$ is the prefactor, $\tau$ is the exponential thinning time, and $t_0$ is the breakup time. Contrary to the above two regimes, the neck geometry becomes cylindrical during the exponential thinning [Fig. 3(a) inset]. The cylindrical shape is lost near the end of the
breakup process where the thinning process accelerates and the
symmetry is broken as observed before [13]. After the breakup,
the upper neck recoils, suggesting that the neck supports
a strong tensile stress. All of these features (exponential
thinning and cylindrical neck geometry) are consistent with
the observations in [12,13]. The capillary force is mainly
counteracted by the elongational viscosity of the suspensions
in elongational flows, which are prepared in a mixed solvent of
cyclohexyl bromide and decalin. For large particle (250 and 500
μm suspensions, which are prepared in a mixed solvent of
two regimes happens at ϕ = 45% (which coincides with our
estimate of Re ≈ 1); the fluctuations near the boundary are
mainly due to the viscosity variations in different particle
suspensions due to slight density mismatches that occur if
the laboratory temperature varies. The asymmetric-symmetric
transition is not observed for the 500 μm particles: even for
ϕ > 45%, the breakup still falls in the inertial regime. In
agreement with the arguments of [11], this happens because the
pressure induced by the particles becomes small; the Laplace
equation ΔP ≈ γ/d shows that the larger the particle, the
smaller the pressure at the scale of the particle. For our case,
if the ratio of the particle diameter d and syringe radius R0
(R0 ≈ 2.25 mm syringe is used for 500 μm particles) becomes
of order unity, and since (Δγ d/ρ R0)^1/3 is of the order of the
usual inertial prefactor α (0.27 mm/ms^2), the equations for
the two regimes become identical; in this case, the local force
balance at the level of individual particles can be neglected.
The condition d ~ R0 then roughly defines the transition
between the two regimes as a function of the particle diameter.

For the smallest particles at the highest concentrations,
the breakup behavior also depends on the tensile stress in
the thinning neck. Note that the exponential regime is also
observed for a colloidal suspension of 1.3 μm particles
(Fig. 4), showing that the thermal (Brownian) fluctuations of
the particles do not change the breakup dynamics qualitatively;
this is nontrivial since in some cases the breakup can be altered
by fluctuation forces [28,29].

To understand the breakup behavior in the exponential
regime, we correlate our observations with the shear rheology
through measurements of the first normal stress difference N1;
here N1 = τ_{zz} - τ_{rr}, with τ_{zz} and τ_{rr} being the normal stress components along the
axis and radius of the neck, respectively. In agreement with
this idea, we find that all the thinning curves (for 1.3, 6, and
10 μm suspensions with ϕ ≥ 55%) collapse on a single master
curve if 2R_{min}/A is plotted vs (t_0 - t)/τ in semilogarithmic
coordinates [Fig. 3(c)]. This suggests that the exponential
thinning model for this regime captures all the experimental
data. The characteristic time of the exponential thinning shows
a quadratic dependence on the particle diameter: τ ∝ d^2
[Fig. 3(d)].

An interesting question in this regime is that the exponential
thinning behavior cannot continue infinitely and must cease
at some finite time (when 2R_{min} = 2R_{min,1}) before the final
breakup. What happens close to breakup after exponential
thinning has been studied rather extensively for polymer
suspensions [12], where an exponential thinning behavior cannot continue infinitely and must cease
to affect the breakup and we observe the symmetric inertial
regime. As shown in Fig. 4, the transition between these
two regimes happens at ϕ ≈ 45% (which coincides with our
estimate of Re ≈ 1); the fluctuations near the boundary are
mainly due to the viscosity variations in different particle
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thinning behavior was also observed in cornstarch suspensions, a critical shear rate $\dot{\gamma}_N$ is defined as the point where $N_1$ starts to deviate from zero. In our experiments, if $\dot{\gamma}_N$ is defined in a similar way: the minimum point where $N_1$ starts to increase on increasing the shear rate [Fig. 5(c)], we also find a one-to-one correlation between the thinning time $\tau$ and $\dot{\gamma}_N$: $\tau \sim 3/\dot{\gamma}_N$ [Fig. 5(d)] in agreement with the results in [13] albeit for a different suspension. All these observations point to a direct link between the elongational and shear rheology of suspensions with similar microscopic mechanisms regarding the emergence of the positive normal stresses. Therefore, we interpret the exponential regime regarding the drop breakup behavior as a granular Bagnoldian regime. The interesting negative part of the $N_1$ curves in Fig. 5(b) may be due to the solvent being sucked into the granular packing that is dilating due to the shear [32]; however, its study is beyond the scope of this work and will be pursued elsewhere.

For predicting the onset of shear thickening a critical Stokes number $St = \rho d^2 \dot{\gamma}/\eta_0$ is sometimes used, with $\eta_0$ the solvent viscosity [33]. In our elongational experiments (drop formation), we can estimate this from the elongation rate $\dot{\varepsilon} = (-2/R_{\text{min}})(dR_{\text{min}}/dt) = 2/\tau$ and the viscous time $\tau_v = \rho d^2/\eta_0$ [using $\tau \propto d^2$ as shown in Fig. 3(d)]; the latter determines the time scale for the interstitial fluid passing through the space between particles. The Stokes number for the emergence of the Bagnoldian regime then turns out to be $St = \tau_v \dot{\varepsilon} \approx 2 \times 10^{-3}$ for $\varphi = 55\%$, but gradually becomes somewhat smaller ($\sim 1 \times 10^{-3}$) when the volume fraction increases up to $\varphi = 59\%$, so that the Stokes number varies over a factor of 2. The phase diagram (Fig. 4) then indicates that $St \sim (1-2) \times 10^{-3}$ gives the transition between the Bagnoldian regime and the symmetric inertial regime.

Compared with the Stokes number for the onset of shear thickening in the rheology, the onset shear rate for shear thickening gives Stokes numbers that are much smaller and vary over an order of magnitude when the volume fractions are changed: $0.3 \times 10^{-4} < St < 5.4 \times 10^{-4}$. Such small Stokes numbers for the onset of thickening are also found in other shear thickening systems; this is surprising since if the thickening were due to a viscous-to-inertial transition, one would expect the Stokes number to be of order unity. One interpretation of these small Stokes numbers is that they result from short-range interactions between particles that give the onset of thickening [20,34–37].

In conclusion, we studied drop formation in aqueous granular suspensions. Systematically changing the volume fractions ($\varphi$) and the particle diameters ($d$), we established a phase diagram that allows us to understand and predict the breakup behavior. It turns out that the behavior is very different from that of Newtonian fluids; which is important since many suspension flows can have an important elongational part, and it is this part that is characterized by our experiments. In addition, the results here allow the finite-time singularity that occurs at the breakup to be tailored in a controlled fashion.
One pertinent example is that the asymmetric inertial breakup invariably leads to the formation of satellite droplets. These are in fact suppressed in the symmetric inertial regime, which could find applications, e.g., in spraying and inkjet printing, where such satellites are unwanted.

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