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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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FIG. 1. (Color online) (a) Thinning dynamics $2R_{\text{min}}$ vs $(t_0 - t)$ for water and $40 \, \mu m$ suspensions with $\varphi$ varying from 45% to 59%. Continuous lines: power law fit with $2R_{\text{min}} = \alpha(t_0 - t)^{2/3}$. Inset: two images showing the asymmetric and symmetric breakup, respectively. The scale bar is $1 \, \text{mm}$. (b) Left: prefactor $\alpha$ from fitting the thinning curves in (a). Right: asymmetry coefficient. Inset: Reynolds number Re vs $\varphi$.

To study the dynamics of drop detachment for the granular suspensions in air, we use a high-speed video camera (Phantom V7 at 10000 frames/s). The controlled release of the drops is achieved by using a syringe pump to set a low drop emission rate. To prevent possible jamming in the syringe tip, two kinds of syringes are used, with radii $R_0 \approx 1 \, \text{mm}$ for $d \leq 40 \, \mu m$ particles and $2.25 \, \text{mm}$ for $d \geq 80 \, \mu m$ particles. The rheological measurements are carried out using a cone-plate geometry with a $50 \, \text{mm}$ diameter cone and a $4^\circ$ angle on a Physica MCR 300 rheometer for the smallest particles. For the larger particles we use a $5 \, \text{mm}$ gap Couette cell or a plate-plate geometry with a $50 \, \text{mm}$ diameter cone and a $4^\circ$ angle. The controlled release of the drops is achieved by using a syringe pump to set a low drop emission rate.

We first discuss the results for the $40 \, \mu m$ PS beads [20,23]. Figure 1(a) shows the temporal variation of the minimum neck diameter $2R_{\text{min}}$ for different volume fractions $\varphi$. For comparison, we also plot the thinning curve for water which follows the inertial-capillary prediction [1,24]: $2R_{\text{min}} = 0.7(\gamma/\rho)^{1/3}(t_0 - t)^{2/3}$, where $\gamma$ is the surface tension, $\rho$ is the density, and $t_0$ is the breakup time. For dilute suspensions ($\varphi \leq 45\%$), the dynamics is indistinguishable from that of pure water [Fig. 1(a)] with an asymmetric breakup [Fig. 1(a) inset], showing that the particles have no influence on the breakup dynamics [25]. To the contrary, the neck thinning curves for concentrated suspensions ($\varphi \geq 50\%$) deviate from the inertial prediction in that the prefactor is different, but the power of $2/3$ in time seems to be conserved [Fig. 1(a)]. To quantify the prefactor, we fit the thinning curves close to the breakup time $t_0$. For a fitting range $0 < (t_0 - t) < 2 \, \text{ms}$, the data are linear when plotted as $2R_{\text{min}}^3$ vs $(t_0 - t)$, allowing evaluation of the slope. We also fit directly using $2R_{\text{min}} = \alpha(t_0 - t)^{2/3}$ [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 ($0.27 \, \text{mm}^2/\text{ms}^{2/3}$) 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 $2R_{\text{min}} \sim (\Lambda \gamma d/\rho R_0)^{1/3}(t_0 - t)^{2/3}$ [11], which resembles the usual inertial scaling but includes an extra factor $(\Lambda d/R_0)^{1/3}$ 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 $R_{\text{min}}/(\rho R_0 \Lambda^{1/3})$ 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 Re\(\approx\) \(\frac{\rho v_i R_{min}}{\eta}\); here \(v_i = \frac{\partial (2R_{min})}{\partial t}\) 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 Re. This Reynolds number can also be estimated as Re\(=\) \(\left(\frac{R_{min}}{l_v}\right)^{1/2}\) with the viscous length scale given by \(l_v = \eta^2/\rho \gamma\); viscous forces should dominate the dynamics when Re\(\approx\) \(\frac{Re_{min}}{l_v}\) is smaller than \(l_v\). As shown in Fig. 1(b) inset, the estimated Re decreases with increasing \(\phi\) (increasing \(\eta\)). Thus, for high concentrations, for which Re < 1 and despite the fact that for a substantial part of the thinning dynamics the neck radius \(R_{min}\) is smaller than the viscous length \(l_v\), the thinning does not happen at constant speed, but rather as \(2R_{min} \propto (t_0 - t)^{2/3}\). For Newtonian fluids, if \(R_{min} \ll l_v\), one would expect \(2R_{min} \propto (t_0 - t)\): the thinning happens at constant speed.

In addition, our experiments show that the change of regimes is observed to happen around Re\(\approx\) 1. This is surprising since in classical theory for Newtonian fluids, for 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 2\(R_{min,i}\) \(\ll\) 2\(R_{min,u}\) with the critical neck diameters 2\(R_{min,i}\) and 2\(R_{min,u}\) 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 \(\frac{\partial (2R_{min})}{\partial t}\), as shown in Fig. 3(b). Upon increasing the volume fraction, 2\(R_{min,i}\) decreases from 0.19 to 0.09 mm, while 2\(R_{min,u}\) increases from 1 to 1.4 mm. The temporal dynamics in this regime can be fitted well by the exponential scaling \(2R_{min} = A \exp{(t_0 - t)/\tau}\), 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 countered by the elongational viscosity of the suspensions
and the tensile stress in the neck is usually associated with an
increase of the first normal stress difference \( \tau_{zz} - \tau_{rr} \), with \( \tau_{zz} \) and \( \tau_{rr} \) 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 \( \mu \text{m} \) suspensions with \( \varphi \geq 55\% \)) collapse on a single master curve if \( 2R_{\min}/A \) is plotted vs \((t_0 - t)/\tau\) 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: \( \tau \propto d^2 \) [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 solutions [4]; here either secondary instabilities set in or dilution effects become important before a true asymptotic regime can be uncovered.

Combining the results for all different systems (systematically varying the particle diameters \( d \) and volume fractions \( \varphi \)), a phase diagram for drop formation in aqueous granular suspensions is constructed (Fig. 4). Three regimes are shown in the diagram: the inertial, the symmetrical inertial, and the exponential regime. Interestingly, the transitions between the different regimes are observed to happen as a function of both the volume fractions \( \varphi \) and the particle diameters \( d \) (Fig. 4).

To understand the transitions, we start by noting that all suspensions at low volume fractions (\( \varphi \leq 45\% \)) follow the inertial regime for water. As the volume fraction \( \varphi \) is increased (thus increasing the suspension viscosity), the particles start
to affect the breakup and we observe the symmetric inertial regime. As shown in Fig. 4, the transition between these two regimes happens at \( \varphi \approx 45\% \) (which coincides with our estimate of \( \text{Re} \approx 1\)); 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 \( \mu \text{m} \) particles; even for \( \varphi > 45\% \), 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 \( \Delta P \propto \gamma/d \) 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 \( R_0 \) (\( R_0 \approx 2.25 \text{ mm syringe is used for 500} \mu \text{m particles} \)) becomes of order unity, and since \( (\lambda y d/\rho R_0)^{1/3} \) is of the order of the usual inertial prefactor \( \alpha \) (0.27 \text{ mm/ms}^2/\text{3}), 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 \( d \sim R_0 \) 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 \( \mu \text{m} \) 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 \( N_1 \); here \( N_1 = \tau_{11} - \tau_{22} \) with \( \tau_{11} \) and \( \tau_{22} \) being the normal stress components along and perpendicular to the shear direction, respectively. Figure 5(a) shows that the viscosity of 6 \( \mu \text{m} \) particle suspensions is constant before shear-thickening, in agreement with [23]. The corresponding \( N_1 \) curves are plotted in Fig. 5(b). We measured a significant positive \( N_1 \) in systems with \( \varphi \geq 55\% \); for \( \varphi < 55\% \) cases, we did not observe positive normal stresses even at shear rates as high as 3000 s\(^{-1}\). A plot on a log-log scale shows that \( N_1 \) scales with shear rate squared \( \gamma^2 \) [Fig. 5(b) inset]; the quadratic scaling suggests a Bagnoldian behavior, for which the shear and normal stresses are both proportional to the shear rate squared as observed in dry granular materials [20,30,31]; particle inertia may contribute to the emergence of the nonzero normal force [20,30]. We fit the \( N_1 \) curves (positive part) with \( N_1 = \Psi_1 \gamma^2 \), where \( \Psi_1 \) is the first normal stress coefficient [Fig. 5(c)]. This enables us to compare the characteristic time \( \tau \) for the exponential thinning with the relaxation time \( \Psi_1/\eta \), measured in the shear rheology with \( \eta \) being the suspension viscosity. We find that for the non-Brownian (6 and 10 \( \mu \text{m} \)) suspensions, the ratio of time scales is constant, i.e., \( \Psi_1/\eta \) varies linearly with \( \tau \): \( \Psi_1/\eta \approx 0.1 \tau \) [Fig. 5(d)]. For the Brownian suspensions, this measurement is difficult due to the rapid evaporation of the more volatile of the two solvents, and the suspension properties significantly change over time. The relation found nevertheless suggests a direct link between the time scales in elongational and shear flows. In [13], where an exponential
thinning behavior was also observed in cornstarch suspensions, a critical shear rate \( \gamma_N \) is defined as the point where \( N_1 \) starts to deviate from zero. In our experiments, if \( \gamma_N \) 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 \( \gamma_N \): \( \tau \sim 3/\gamma_N \) [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 \( St = \rho d^2 \gamma / \eta_0 \) 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 \( \tau_\nu = \rho d^2 / \eta_0 \) [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 \( St = \tau_\nu \dot{\varepsilon} \approx 2 \times 10^{-3} \) 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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