An experimental study of particle effects on drop formation

Roy J. Furbank and Jeffrey F. Morris

School of Chemical and Biomolecular Engineering, Georgia Institute of Technology, Atlanta, Georgia 30332

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The formation of drops of particulate suspensions composed of spherical, neutrally buoyant, noncolloidal particles in a viscous liquid is examined experimentally. The suspensions are investigated over a range of particle volume fractions, \( \phi = n \pi d_p^3/6 \) where \( n \) is the particle number density and \( d_p \) is the particle diameter \( (d_p = 212-250 \, \mu m \) in most of the experiments), and for flow through three tubes of outer diameters \( d = 0.16, 0.32 \) and 0.64 cm; the corresponding inner diameters are 0.10, 0.22, and 0.53 cm. Drop formation in the dripping mode and the transition from dripping to jetting are investigated. In the dripping mode, the behavior of low-\( \phi \) suspensions \( (\phi \leq 0.10) \) is markedly different from that of higher-\( \phi \) suspensions \( (0.15 \leq \phi \leq 0.40) \), with a transition in the qualitative behavior over a relatively narrow range of concentrations. Pinch-off structures for low-\( \phi \) suspensions are similar to that of the pure liquid, consisting of a long slender liquid thread connecting a hemispherical cap of liquid at the orifice to the nearly spherical forming drop; the structures differ from those of pure liquids in that individual particles, or groups of particles, can be captured in the thread at rupture, leading to new modes of satellite drop formation. At higher \( \phi \), the presence of large numbers of particles in the thinning thread during necking results in thick conelike structures termed “spindles” owing to this labeling of structures of similar geometry observed in electrohydrodynamic sprays. Particles are found to substantially suppress the number of satellite drops at higher \( \phi \), but the few satellite drops produced are much larger than observed in pure liquid drop formation. The transition from dripping to jetting occurs at a smaller flow rate at finite \( \phi \), with much shorter coherent lengths after jetting than in the case of a pure liquid. The transition becomes less abrupt and more difficult to identify at the highest concentrations examined. © 2004 American Institute of Physics. [DOI: 10.1063/1.1691034]

I. INTRODUCTION

Drop formation is an area of research which has a rich and diverse history, yet continues to generate much interest today. A large majority of past and present effort in this area has involved systems where drops of a single, homogeneous liquid are formed into an immiscible fluid. In this work we seek to extend this base of knowledge by experimentally probing drop formation processes for liquids containing solid particles.

A. Background

Pure fluid drop formation has been extensively studied for over 100 years. A review by Eggers (1997) on the subject contained over 250 references with the earliest dated from the year 1686.

In contrast to the situation for pure fluids, the formation of drops from particle-laden liquids has received little attention despite its importance in a variety of applications which include spray drying, slurry fuel combustion, and certain applications of ink-jet printing. Of particular interest to this work is the growing desire to extend the range of traditional ink-jet printing technology to applications requiring the use of solid-laden inks, with examples in ceramics manufacture and textile printing. While some progress has been made in developing particle-laden inks for use in traditional ink-jet printers and designing printers that will jet more complex materials, little effort has been made to understand in detail the role the particles play in the overall process. Our goal in this work is to provide insight into this problem at a more fundamental level by examining drop formation from suspensions of solid spherical particles in a viscous liquid. We examine the dripping mode and the transition to the jetting mode.

The dripping mode is characterized by low flow rates where pendant drops are formed at the tip of the orifice. A common example of this is a dripping faucet. Pure fluid behavior in this mode has been extensively studied both experimentally and numerically. While the formation of drops had been studied for many years, focused examination on the bifurcation, or pinch-off, process is relatively recent. Peregrine et al. presented a detailed qualitative description of this process for water into air using high-speed photography. The authors highlighted an asymmetry of the developing structure which had not been previously noted. As the drop fell, a slender liquid column (also commonly referred to as a thread or filament) formed which connected the liquid at the orifice tip with that of the forming drop, with eventual rupture near the surface of the forming drop rather than at the middle of the thread. Following pinch-off, the thread was observed to retract due to the now unbalanced
surface tension and also to undergo a secondary rupture near the orifice to yield a satellite drop.

Shi et al. extended this work to more viscous liquids and examined the effect of viscosity on the shape of the forming drop near pinch-off. Increasing viscosity was found to lead to a lengthening of the thread at pinch-off as well as a smoothing in the transition from the thread to the surface of the drop. These authors also implemented the one-dimensional equations derived by Eggers and Dupont, and their numerical solutions qualitatively reproduce the pinch-off structures observed experimentally.

A group of recent studies by Basaran and co-workers has probed the pure fluid problem. Zhang and Basaran studied the formation of pendant drops of viscous liquid into air, by performing a wide range of experiments in which they were able to carefully follow the evolution of the forming drop with time. Among other results, the authors determined that increasing flow rate, orifice size, or viscosity led to longer thread lengths at pinch-off. Subsequently, Zhang numerically investigated both pendant drop formation into air and into a second immiscible liquid. Basaran and co-workers published a series of studies in which the region very near pinch-off was examined in detail before and after thread rupture both computationally and experimentally, showing that the interface of the forming drop can overturn prior to rupture.

The presence of this thread-like structure near pinch-off observed for pure fluids in the dripping mode is likely to play a critical role in particle-laden systems. We have been unable to find any published work aside from a French thesis for dripping-mode drop formation from suspensions. This thesis examined suspensions up to 23% solids primarily by image analysis, with a focus on the frequency of drop formation and coherent jet length in the dripping and jetting modes, respectively. We know of no work concerned with the structures formed near pinch-off for these systems. The work described here shows that the introduction of a new length scale to the problem, namely the particle diameter, fundamentally changes the evolution of the liquid thread near the pinch event: regardless of their size, the particles become significant relative to the narrowing thread at some point before pinch-off occurs. This problem thus presents a challenge to bulk flow simulation by continuum methods.

As the flow rate is elevated sufficiently, the flow at the orifice undergoes a transition from dripping to jetting. The transition from dripping to jetting has been experimentally examined by Scheele and Meister, who measured jetting velocities in liquid–liquid systems and developed a correlation for predicting the onset of jetting and the resulting drop size based on a force balance at the orifice tip. Richards et al. developed a numerical model based on a volume-of-fluid method and were able to accurately reproduce the data of Scheele and Meister both before and after the transition. Still more recently Clanet and Lasheras considered the transition from dripping to jetting for water into air. They experimentally determined the jetting transition for different orifice sizes and presented an analysis for estimating this transition in the inviscid limit.

Following this transition, jetting ensues. The initial study of jetting behavior for pure liquids dates to Rayleigh and his linear stability analysis for the inviscid case. The breakup of a liquid column to drops by capillary forces is, as a consequence, now a classic problem in hydrodynamic stability. More recent works are detailed in reviews by Bogy and Eggers. In contrast to the dripping mode, the jetting mode has been considered for such complex fluids as polymeric solutions, magneto-rheological fluids, and a few simpler particle-laden systems. The works on both polymer solutions and magneto-rheological fluids reveal an inhibition of jet breakup, relative to the pure fluid, and the development of a “beads-on-string” structure where forming drops are connected by long slender liquid filaments. In contrast to these systems, the work of Ogg and Schetz describes slurries composed of water and glass beads jetting into air and shows that the particles introduce nonaxisymmetric disturbances and lead to irregular breakup.

B. Problem description

The problem considered here is that of a neutrally buoyant suspension of particle volume fraction \( \phi \) and particle diameter \( d_p \) flowing through a capillary tube with outer diameter \( d \) at a constant flow rate \( Q \) (Fig. 1); note that \( \phi = n \pi d_p^2/6 \) where \( n \) is the particle number density (number of particles per unit volume). The suspending liquid has density \( \rho \), viscosity \( \mu \), and surface tension \( \sigma \). The drop forms in the direction of gravity and fully wets the outer edge of the capillary orifice. For sufficiently small \( Q \), drop formation occurs in the dripping mode and drops are formed directly at the tip of the capillary tube. Increasing \( Q \) eventually leads to a transition from the dripping mode to the jetting mode in which drops are formed away from the orifice at the end of a stable column of suspension.

Figure 1 illustrates the situation for a suspension of \( \phi = 0.05 \) in the dripping mode and introduces the notation used in this study. Initially, the drop grows essentially statically at a constant flow rate \( Q \) (Fig. 1); note that \( \phi = n \pi d_p^2/6 \) where \( n \) is the particle number density (number of particles per unit volume). The suspending liquid has density \( \rho \), viscosity \( \mu \), and surface tension \( \sigma \). The drop forms in the direction of gravity and fully wets the outer edge of the capillary orifice. For sufficiently small \( Q \), drop formation occurs in the dripping mode and drops are formed directly at the tip of the capillary tube. Increasing \( Q \) eventually leads to a transition from the dripping mode to the jetting mode in which drops are formed away from the orifice at the end of a stable column of suspension.

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forming drop would remain attached to the tip of the capillary tube. Once the weight of the forming drop exceeds some critical value it begins to fall [Fig. 1(b)] and the width of the liquid column between the orifice exit and the drop begins to rapidly narrow. This process of rapid radial thinning is referred to as necking and continues until a time very near that shown in Fig. 1(c), the last image captured before the drop detached. This configuration is termed the pinch-off structure. The time over which necking occurs, \( t_n \), is short relative to \( \pi \), the time between drops; \( t_n/\pi \approx 0.06 \) in Fig. 1. Throughout the process \( L \) denotes the total length of liquid measured from the orifice outlet which is continuously attached to the capillary tip and the slightly subjective quantity \( L_N \) is the length of the slender thread-like region. A brief time after the instant captured in Fig. 1(c) the thread ruptures and the primary drop (diameter \( D \)) is formed [Fig. 1(d)], potentially accompanied by some number of smaller secondary, or satellite, drops.

The dimensionless parameters characterizing the problem include the Bond (Bo), capillary (Ca), and Reynolds (Re) numbers in the absence of particles:

\[
Bo = \frac{\rho g d^2}{\sigma}, \quad Ca = \frac{U \mu}{\sigma}, \quad \text{and} \quad Re = \frac{\rho U d}{\mu},
\]

where \( U = 4Q/\pi d^2 \) is the mean velocity at the orifice outlet. The Weber number (We = Ca Re) is commonly used in describing the process in the jetting mode. The particles, in the case of monodisperse and neutrally buoyant suspensions upon which we focus in this work, introduce two further dimensionless parameters: the particle volume fraction \( \phi \) and the ratio of the orifice diameter to particle diameter \( d/d_p \).

The particles are sufficiently large that Brownian motion is negligible (the Péclet number is infinite). Colloidal forces are also of no relevance, as the particles are over 200 microns in most of the experiments.

In this paper we experimentally examine drop formation from particulate suspensions in the dripping mode and the transition to jetting, with the goal of highlighting the effects introduced by the particles. In Sec. II we present a description of the experimental setup and an outline of the experimental parameters investigated. In Sec. III we present the results of particle-laden drop formation experiments in the dripping and jetting modes, and in the transition between the two. A discussion of these results and the key particulate effects is presented in Sec. IV, and is followed by our conclusions in Sec. V.

II. EXPERIMENT

A. Apparatus

Experiments were performed using the apparatus depicted in Fig. 2. Suspension was driven from the reservoir of a drop delivery vessel through a short (3–5 cm) stainless steel capillary tube using a Harvard Apparatus PHD 2000 syringe pump (accuracy of imposed flow rate \( \pm 1\% \)), employing silicone oil (polymethylsiloxane) as a driving fluid in order that particle migration effects not lead to maldistribution in the syringe or tubing upstream of the reservoir. The silicone oil is less dense than and immiscible with the suspending liquid, providing a particle-free driving fluid; a stable interface between the suspension and silicone oil within the reservoir was always observed. The ends of the capillary tubes were electrolytically cut and polished (Valco Instruments), with outer diameters 0.16, 0.32, 0.64 cm and inner diameters 0.10, 0.22, 0.53 cm. The ratio of capillary length, \( l \), to inner diameter varies in the range of \( l/d_i \) = 5.7–50, and both experiments\(^{39}\) and suspension flow models\(^{24,40}\) suggest that particle migration toward the tube axis should be a minor influence except perhaps at the highest \( \phi \) and largest \( l/d_i \) studied.

The evolution of the forming drops at the capillary tip was captured photographically using one of two CCD cameras: a Cooke SensiCam camera with a frame rate of 30 frames per second (fps), and a Kodak MotionCorder Analyzer, Model 1000 with variable frame rates up to 10,000 fps (the highest rate used was 250 fps in the work described here). Back-lighting was provided by a 300 W bulb diffused through translucent Lexan of 0.64 cm thickness.

The observation chamber was an open-top glass box filled with silicone oil (\( \rho = 0.95 \text{ g/cm}^3 \), \( \mu = 19 \text{ cP} \)) or left empty. In the latter case the chamber served only to isolate the forming drop from air currents in the laboratory. The drop delivery vessel consisted of a 7.6 cm diameter Lexan cylinder (the reservoir) with a removable aluminum bottom into which the capillary was pressed. When assembled, the two end caps of the cylinder were parallel and the capillary tube was normal to both. To ensure that the capillary tube was parallel to gravity, a Newport tilt platform was used and a level was placed in the center of the upper end piece of the cylinder to precisely position the entire drop delivery vessel. The tilt platform was mounted to an aluminum frame (not shown in Fig. 2) and the drop delivery vessel was clamped to the platform. Both delivery vessel and observation chamber were fixed to a vibration-damped table (Newport).

B. Suspensions

Suspensions were prepared such that the particles were neutrally buoyant. The liquid viscosity was chosen large enough to examine the pinch-off region in detail without
requiring extremely high camera frame rates (rates greater than 250 fps reduce the resolution), but not so large that the stretching and subsequent pinch-off occurred over too great a distance.

Spherical poly(methyl methacrylate) (PMMA) particles were suspended in a mixture of 23% (wt.) ZnCl₂/water solution and UCON 90,000 (polyalkylene glycol; Dow Chemical). The composition of the final mixture was 30% (wt.) UCON with properties ρ = 1.184 ± 0.002 g/cm³, μ = 400 ± 20 cP, and σ = 49 ± 3 dyne/cm by drop weight and 47 ± 2 dyne/cm by the du Nuoy ring. Particles sieved to dₚ ≈ 212–250 μm were used in the majority of experiments performed; a few experiments used particles with dₚ < 106 μm.

C. Experimental parameters

The experimental parameters examined are presented in Table I. We conducted experiments where drops of suspension of various particle fraction were formed into both ambient silicone oil and air using three different size orifices and two different particle sizes. The range of flow rates encompassed both dripping and jetting behavior with Re < 1 throughout and Re < 1 before the onset of jetting. Drops formed into silicone oil provide primarily qualitative information and allow visualization of the suspended particles, while quantitative results were obtained for drop formation into air.

III. RESULTS

In this section experimental results of particulate effects on drop formation for suspensions of various φ are presented. The results are divided into three areas: development of the liquid thread during necking and the subsequent pinch-off structures formed, the formation of the primary drop and satellite drops after pinch-off, and transition from the dripping mode of drop formation to jetting.

A. Development of liquid thread and pinch-off structure

Here, we address the motion of the forming drop after surface tension forces holding it to the tip of the orifice have been exceeded and the drop falls under its own weight. We examine particle motions in the developing liquid thread for a low-φ suspension, followed by consideration of the pinch-off structures of various suspensions. The above-described work uses silicone oil as the ambient fluid. Lastly, we describe the evolution of the liquid thread through pinch-off for a more concentrated suspension flowing into air.

1. Particle motions during necking

Figures 3 and 4 illustrate the photographic results from a typical experiment in the vicinity of the pinching time for pure suspending liquid and a suspension of φ = 0.05 (dₚ = 212–250 μm), respectively, for the same conditions (d = 0.32 cm, Q = 1.8 cm³/min, Re = 0.04). The time between frames is 1/30 s. In both instances the drops are formed into silicone oil which slows the necking process (relative to flow into air) and allows for a direct visualization of the particles.

In the case of the pure liquid (Fig. 3) a thread is formed as the drop falls away from the orifice. This liquid thread continues to stretch, eventually rupturing near the trailing edge of the forming drop. This result is similar to that observed by numerous researchers of pure liquid drop formation.8–11

The particle-laden case is illustrated in Fig. 4. The particles are initially randomly and uniformly distributed throughout the liquid as it flows through the orifice and into the hanging drop; the relative uniformity is illustrated by the first image, Fig. 4(a). The early stage of necking occurs in a fashion similar to that observed for the pure liquid as may be seen from the similarity of the shapes in the two earliest photographs, (a) and (b), from each of Figs. 3 and 4. However, the presence of the particles and their distribution within the forming thread introduce novel features not seen.

![Fig. 3. Evolution of the liquid thread up to and slightly beyond pinch-off for φ = 0, d = 0.32 cm into silicone oil with Q = 1.8 cm³/min (Re = 0.04). Time between frames is 1/30 s.](image-url)
in pure liquids as the thread progresses toward rupture. Depending on the random distribution of particles in the liquid when necking begins, many possibilities exist.

At a time very shortly following that of Fig. 4(b), resistance to further narrowing by the two particles in the center of the liquid thread induces a bulging of the thread. Particles located below this point are completely swept into the forming drop before rupture while those above it are retracted towards the orifice—with a single exception. Surprisingly, the two particles located directly above the central pair move in opposite directions [Figs. 4(c)–4(d)]. The particle slightly closer to the orifice actually returns to join the liquid at the tip of the orifice while the lower particle remains in nearly the same location, and is eventually incorporated into the satellite drop seen in Fig. 4(e). This sequence is only one illustrative example for a low-\(\phi\) suspension as the thread width approaches that of the particle size.

2. Pinch-off structure

The presence of particles in the liquid leads to pinch-off structures which are qualitatively different from those seen in the pure liquid. At low particle concentrations (up to \(\phi \approx 0.10\)) the macroscopic behavior of the structure near pinching is very similar to that of the pure liquid, as illustrated by Fig. 5 where again drops are formed into silicone oil. Notably, the shape of the forming drop is that of the “needle-sphere” configuration typical for pure liquids \(^8–11\) in which a slender, needle-like liquid thread connects the liquid at the tip of the capillary orifice to the nearly spherical drop. The subsequent rupture occurs near the point of attachment of the thread to the drop. Although the overall pinch-off shape up to moderate \(\phi\) is similar to that of the pure liquid, the presence of the particles leads to fundamentally different behavior simply because individual or groups of particles may be captured in the thread during necking. In this event, more common at higher \(\phi\) but observed even at \(\phi = 0.02\), single particles or groups of particles resist stretching and prevent further thinning of the thread. This leads to the generation of satellite drops not observed for pure liquid, as illustrated by Fig. 4 in the preceding subsection.

As the particle concentration is increased beyond \(\phi \approx 0.10\), as illustrated in Fig. 6 for drops into silicone oil, larger numbers of particles are present in the liquid thread during the early stages of necking. As necking proceeds, these particles are forced out of the thinning thread back up towards the capillary tip and down into the forming drop. These particle motions create new structures, which are thicker and more cone-shaped than observed for the pure liquid; these structures at the ends of the liquid thread result in a more gradual transition from the thread to the liquid at the capillary tip and at the tail of the forming drop. We term these structures “spindles” due to this descriptive term used for similar structures observed in electrohydrodynamic sprays.\(^{41}\) These spindles are also similar to particle structures observed by Schaflinger and Machu\(^{42}\) and Nicolas\(^{43}\) who have examined dense suspensions falling under gravity into the suspending liquid and hence in the absence of interfacial tension. These authors describe particle tails developing behind the falling drops. The spindle structures we observe here become more pronounced with increasing \(\phi\), and the length of the region over which final necking occurs is much smaller than that observed for the pure liquid.
shorter than that observed for pure liquid and lower $\phi$ ($L_N/L = 0.6$ for $\phi = 0.10$ and decreases to $L_N/L = 0.3$ for $\phi = 0.25$). In addition, at higher $\phi$ pinch-off occurs higher on the thread away from the edge of the forming drop, the shape of which has become more pear-like than spherical.

Pinch-off from two additional orifice diameters $d = 0.16$ and $0.64$ cm was also examined using the same suspension of $d_p = 212 - 250$ $\mu$m. In both instances, the overall trends with increasing $\phi$ are similar to that presented above for the $0.32$ cm orifice. The number of particles captured in the neck at pinch-off does, however, depend strongly on the orifice size. The smallest orifice is characterized by very short necks with particles rarely captured in the neck. As $d$ increases, the pinch-off length and number of particles in the neck both increase.

3. Evolution of the thread at higher $\phi$

In addition to qualitatively altering the shape of the structure at pinch-off, the particles also lead to much different behavior of the developing thread as the drop falls away from the tip of the capillary. In contrast to the results presented above, those presented here are for drops forming into air. The forming drops are thus dark in the photographs.

In Fig. 7, the evolution of the thread up to and beyond rupture for the pure liquid and a suspension of $\phi = 0.20$ are compared. The time between consecutive frames is 1/250 s. In both instances, the overall trends with increasing $\phi$ are similar to that presented above for the $0.32$ cm orifice. The number of particles captured in the neck at pinch-off does, however, depend strongly on the orifice size. The smallest orifice is characterized by very short necks with particles rarely captured in the neck. As $d$ increases, the pinch-off length and number of particles in the neck both increase.

B. Drop formation and satellite drop generation

Following pinch-off of the liquid thread, a primary drop is formed. This formation may be accompanied by the subsequent break-up of the retracting thread into smaller, satellite drops. In order to assess the influence of suspended particles on this process, measurements were made of the primary drop size as well as the number and size of satellites formed as a function of $\phi$ for a range of $Q$ sufficiently low that all are in the dripping mode. The results are presented in Figs. 8–11.

1. Drop size

Figures 8 and 9 illustrate how the primary drop size changes with $\phi$ for two orifice sizes, $d = 0.16$ and $0.32$ cm.
For each flow rate the dimensionless drop size \((D/d)\) is plotted against \(\phi\). The drop size was determined using the MATLAB image analysis toolbox to detect the edge of the drop from photographs and, assuming axisymmetry, the volume was computed as
\[
V_{\text{drop}} = \int \pi r(z)^2 dz
\]
where \(r\) is the detected radius of the drop and \(z\) is in the direction of gravity; the effective diameter was determined by solving \(V_{\text{drop}} = (1/6) \pi D^3\) for the diameter. This approach was required to assess the size of nonspherical drops prevalent at the imaging location for higher \(\phi\); the drops relax to spherical as they fall. The diameters of the pure liquid drops measured here agree well with those predicted by the correlation developed by Scheele and Meister, being consistently slightly (from 1–6%) smaller than predicted. This is consistent with results reported by Zhang and Basaran and serves to validate our technique.

At a low flow rate, \(Q=0.5\) and \(1\ \text{cm}^3/\text{min}\), little effect of \(\phi\) on drop size is observed for either the \(d=0.16\) cm (Fig. 8) or \(d=0.32\) cm (Fig. 9) orifice. Although there appears to

![Graph showing dimensionless primary drop size vs. \(\phi\) for different flow rates.](image)

**FIG. 8.** Dimensionless primary drop size \((D/d)\) for drops forming into ambient air as a function of \(\phi\) for \(d=0.16\) cm, \(d_p=212–250\ \mu\text{m}\).

![Graph showing average number of satellite drops vs. \(\phi\) for different flow rates.](image)

**FIG. 10.** The average number of satellite drops formed as a function of \(\phi\) for drops forming into ambient air for \(d=0.32\) cm, \(d_p=212–250\ \mu\text{m}\).

![Graph showing total volume of satellite drops vs. \(\phi\) for different flow rates.](image)

**FIG. 11.** The total volume of satellite drops formed for (a) \(\phi=0\), (b) 0.02, (c) 0.15, and (d) 0.25 into ambient air for \(d=0.32\) cm, \(d_p=212–250\ \mu\text{m}\), \(Q=3.5\ \text{cm}^3/\text{min}\) (Re=0.07). The abscissa represents the number of primary drop events observed over the course of an experiment (for example, for \(\phi=0\) slightly less than 80 primary drops were observed).
be a slight decrease with $\phi$ for the smaller orifice and a slight increase for the larger these changes are small compared with the changes at higher flow rates. For $Q = 3$ and 5 cm$^3$/min, both orifices show a marked decrease in drop size with increasing $\phi$. The results indicate that the decay of drop size with $\phi$ depends upon the orifice size or its ratio to the particle size: the decay begins at higher $\phi$ for the larger ($d = 0.32$ cm) orifice and is less pronounced than in the case of the small orifice, while decay begins essentially at $\phi = 0$ for the smaller ($d = 0.16$ cm), with the largest decrease occurring as $\phi$ is increased from 0.10 to 0.20. Accompanying this increase in concentration is a change in behavior to one in which a higher flow rate no longer corresponds with a larger drop size; this is counter to the observed trend for the pure liquid and lower concentrations.

2. Satellite drop formation

An unanticipated result of these experiments is the inhibition of satellite drop formation with increasing $\phi$. As presented in Fig. 5 and remarked upon in Sec. III A, the presence of particles in the neck at the time of thread rupture could be expected to lead to the generation of more satellite drops than that of the pure liquid. This does, in fact, seem to be true for suspensions of very small $\phi$ (Fig. 10), where a slight increase in the number of satellite drops formed is observed for some flow rates. The behavior at high $\phi$ is markedly different. As the particle concentration increases, satellite drop generation falls off rapidly.

Although Fig. 10 indicates that the average number of satellites decreases quite dramatically with $\phi$, it tells us nothing of the size. The size information is presented in Fig. 11 which contains plots of the total volume of satellite drops formed for each pinch-off event observed during the course of an experiment; satellite drop volume was assessed by the direct measurement of diameter after the drops became spherical. The pure liquid forms a nearly constant volume of satellite drops ($=0.5$ mm$^3$) for each primary drop. This behavior is changed quite radically by the addition of even a small particle fraction, as seen for the results at $\phi = 0.02$ where the satellite-drop volume becomes scattered. As $\phi$ is increased, the instances in which even a single satellite drop is formed become relatively few, but these few are much larger than observed for a pure liquid.

The manner in which satellite formation is affected by suspended particles is illustrated in Fig. 12, which compares the evolution of the suspension column and the subsequent thread rupture for two drop formation events for a concentrated suspension of $\phi = 0.40$. In the first case, pinch-off occurs near the middle of the thread with two long spindles eventually retracting without the generation of satellite drops. This case occurs most frequently and results from final necking and pinching in a single location, usually near the center of the thread. The other case is the result of nearly simultaneous pinching in two separate locations near the ends of the thread. This rare dual pinching leads to the formation of a large, concentrated satellite drop.

C. Transition from dripping to jetting

To study particulate effects on the transition from dripping to jetting, the pinching event was studied for suspensions of varying $\phi$ and orifices of different diameter, $d$, over a range of increasing flow rates. In each experiment the initial flow rate was such that dripping was observed. This rate was increased slowly over the course of additional experiments until jetting was observed. The thread length was measured as a function of time in each experiment and used in identifying the transition. All results presented in this section are for drops forming into ambient air.

Typical time series illustrating $L/d$ are presented in Fig. 13. The first graph [Fig. 13(a)] shows the results for the pure liquid. At early times, $t < 3$ s, drops are formed in the dripping mode with a constant period. At $t = 3$ s the flow rate is increased a small amount (from 12.7 cm$^3$/min to 12.8 cm$^3$/min) and the system undergoes an abrupt transition and forms a column of liquid stable to $\sim 50d$; in Fig. 13(a) $L/d = 15$ represents the field of view of the camera and breakup of the pure liquid jet occurs much farther downstream (the position was determined in independent experiments) by the familiar capillary instability. Although other researchers have reported an intermediate oscillatory regime between periodic dripping and jetting, this was not observed in our experiments, apparently due to our larger liquid viscosity.

Also shown in Fig. 13 are results illustrating $L/d$ near the transition for suspensions of $\phi = 0.05$ and 0.20. In the case of $\phi = 0.05$, shown in Fig. 13(b), the behavior is quite similar to that of the pure liquid in the sense that an abrupt lengthening of the thread is observed due to a small increase in flow rate, with a similar flow rate at transition. What is different, however, is that the accompanying lengthening of the thread is much less for the suspension: $L/d \approx 10$ for $\phi = 0.05$ compared with $L/d \approx 50$ for the pure liquid. The behavior of the more concentrated suspension ($\phi = 0.20$) is shown in Fig. 13(c). In this case a transition is evident when $Q$ is increased from 11 to 12 cm$^3$/min, although the behavior prior to transition is different from that of either the pure
liquid or the low-\(\phi\) suspension. It appears that prior to the onset of jetting the suspension goes through an intermediate mode where the minimum value of \(L/d\) fluctuates. Lengthening of the liquid column attached to the tip of the orifice occurs when the liquid thread following rupture is unable to retract fully before a new drop begins to form. Following the transition the corresponding lengthening of the thread is lower still than the \(\phi = 0.05\) suspension.

Quantitative results for the transition experiments are presented in Figs. 14 and 15 in which the dimensionless thread length \((L/d)_{\text{min}}\) is plotted as a function of \(Q\) for all \(\phi\) studied. The quantity \((L/d)_{\text{min}}\) represents the coherent length of the liquid or mixture attached to the orifice, as it is an average of the minimum values of \(L/d\) observed over all drop formation events in an experiment (the minima in the \(L/d\) of the time series). For example, in Fig. 13(a), \((L/d)_{\text{min}} = 1.8\) for \(\phi = 0\) before the transition. For \(\phi = 0.05\), \((L/d)_{\text{min}}\) decreases greatly relative to the pure liquid, from \(\sim 90\) to \(\sim 11\) for the smaller orifice \((d = 0.16\) cm) and from \(\sim 50\) to \(\sim 9\) for the larger \((d = 0.32\) cm). The data points corresponding to \((L/d)_{\text{min}}\) at rates above transition for \(\phi = 0\) in both Figs. 14 and 15 are omitted so that the effects of increasing \(\phi\) can be presented more clearly.

Although the coherent jet lengths following transition are substantially shorter, Figs. 14 and 15 show that low-\(\phi\) suspensions otherwise behave similarly to the pure liquid. Prior to transition, suspensions at both \(\phi = 0.05\) and 0.10 have \((L/d)_{\text{min}}\) very similar to the pure liquid. These mixtures also display sharp transitions at flow rates very nearly the...
same as that observed for the pure liquid. As ϕ is further increased two trends are apparent: first, the lengthening of the liquid column leading to transition occurs more gradually and, second, it begins at progressively lower \( Q \).

Smoothing of the transition at higher ϕ is evident for both orifices examined, and is most pronounced for the smaller orifice (Fig. 14). Here \( (L/d)_{min} \) for \( ϕ = 0.20 \) and 0.30 gradually increases from 2 to nearly 8 with no obvious abrupt transition, although the column of liquid stable to \(~8d\) at the largest volume fraction is indicative of jet-like behavior. Hence, the most we can safely say about these two concentrated suspensions is that between \( Q = 3 \text{ cm}^3/\text{min} \) and \( 9 \text{ cm}^3/\text{min} \) the behavior changes gradually from dripping to jetting. The two highest ϕ for the larger orifice also display smoothing of the transition.

The onset of earlier jetting is clearly evident for \( ϕ = 0.20 \) in the \( d = 0.32 \) cm orifice, as shown in Fig. 15. In this case an abrupt lengthening is seen when the flow rate is increased from 11 to 12 cm/\text{min}. Although the difference between the transition \( Q \) for \( ϕ = 0 \) and 0.20 is small (12.7 and 12.0 cm/\text{min}, respectively), jetting is induced by the addition of particles at a flow rate for which periodic dripping is observed for pure liquid.

Experiments on the jet transition were also performed using smaller particles (\( d_p < 106 \mu \text{m} \)) and the results are presented in Fig. 16. Only \( ϕ = 0.05 \) and 0.10 were examined in these experiments, whose purpose was to compare the transition behavior—both the transition flow rate and the jet length attained just above the transition flow rate—of small- and large-particle suspensions. In Fig. 16 it is illustrated that the coherent jet length in the form \( (L/d)_{min} \) for the smaller particles was nearly twice that of the larger particles, but only about one-half the length of the pure liquid jet. The flow rate at transition for the smaller particles is the same as that for the pure liquid and the larger particle suspensions for \( ϕ = 0.05 \); for \( ϕ = 0.10 \) with sub-106 \( \mu \text{m} \) particles, the transition occurs at a slightly lower flow rate than in the pure liquid.

In addition to the transition to jetting from dripping, the reverse case of slowing flow rate to transition from jetting to dripping was also examined in a limited set of experiments. Since the abrupt nature of the transition disappears at elevated \( ϕ \), only suspensions of \( ϕ = 0.05 \) for \( d_p = 212–250 \mu \text{m} \) and \( d_p < 106 \mu \text{m} \) were examined in flow through the \( d = 0.32 \) cm orifice. All of the cases examined displayed hysteresis, as the flow rate \( Q_+ \) for a transition to jetting differed from the flow rate \( Q_- \) for transition to dripping from jetting. For pure liquid and the small particle suspension, \( Q_+ = 13 \text{ cm}^3/\text{min} \) and \( Q_- = 10 \text{ cm}^3/\text{min} \), while for the larger particles, \( Q_+ = 13 \text{ cm}^3/\text{min} \) and \( Q_- = 11 \text{ cm}^3/\text{min} \). The larger particles appear to reduce hysteretic effects, perhaps due to the large fluctuations induced.

Two other, more qualitative, results were obtained in examining the process once the transition from dripping to jetting was complete: at large \( ϕ \) the jet developed a “swinging” side-to-side motion and the wetting of the orifice changed with increasing \( ϕ \). Figures 17 and 18 show the jet structures for various \( ϕ \) for both \( d = 0.16 \) and 0.32 cm. The swinging motion is most pronounced for the smaller orifice and is evident in Fig. 17(e), which illustrates the jet structure for \( ϕ = 0.30 \). Swinging is observed for the highest concentrations and flow rates, and seems to develop due to unbalanced forces in the retraction after breakup. This behavior results from asymmetric particle configurations in the suspension above and below the pinch location, which then interact in a nonaxisymmetric fashion with the unbalanced surface tension. Although the swinging motion develops at high \( Q \), the asymmetric retraction following pinch-off is observed at low \( Q \) while pendant drops are still forming (Fig. 19). However, the time between drops is sufficiently large that all lateral motion induced following pinch-off of a drop subsides prior to the development of the subsequent drop. The second observation is that the shape of the mixture near the orifice changes with increasing \( ϕ \). Specifically, the film wetting the cut face of the capillary tube becomes thinner under high flow rate and elevated \( ϕ \), appearing to almost dewet, as illustrated in Figs. 17 and 18.
IV. DISCUSSION

In the preceding section, our goal was to present with limited interpretation the results of many experiments to illustrate the pinch-off behavior of particle-laden liquids in both the dripping (or pendant drop) mode and the transition to jetting. Here, we discuss the experimental results with a focus on elucidating the effects introduced by the particles, with the primary goal of gathering certain ideas which provide insight to general features of particulate influence on the drop formation processes we have studied. Critical to this goal is an examination of the drop formation process, both for dilute suspensions, where the process occurs in a manner similar to that of the pure liquid with some notable differences, and for more concentrated suspensions, where the process as a whole is different. We will also address the inadequacy of a continuum description of the mixture during drop formation due to finite-size effects of individual particles. Lastly, we will consider the effects induced by the particles upon the behavior of the liquid jet.

A. Particle fraction dependence in the dripping mode: Low- and high-\(\phi\) behavior

Although the drop formation process in the dripping mode continuously changes with increasing \(\phi\), it is illustrative to examine the two extreme \(\phi\) studied here. Consider the behavior exhibited by the \(\phi=0.05\) suspension presented in Fig. 4. Although initially the particles are apparently uniformly distributed in the forming drop, the determining factor in how the eventual drop is formed is the location of the particles within the thread once necking begins. Just before necking begins, the particles have a certain configuration and it is this configuration which ultimately determines whether particles are captured in the neck at thread rupture. If particles are captured in the liquid thread there are two consequences. First, the thread will be unable to attain the same length as in the case of the pure liquid, because continued uniform thinning of the stretching thread beyond the particle size is impossible. Second, the breakup of the thread following initial rupture will occur differently than is observed in secondary breakup of a pure liquid thread. These two effects at low \(\phi\) can be interpreted as particles causing a destabilization of the liquid thread.

For \(\phi=0.40\), as shown in Fig. 12, the process proceeds differently. Just prior to necking, there are many more particles present than in the low-\(\phi\) case. The motions of these particles are restricted within the developing thread by their close proximity to neighbors. This contrasts with the low-\(\phi\) case, where particles are nearly free to move within the thread until the thread width has thinned to a few particle diameters. The close packing of particles at \(\phi=0.40\) results in a resistance to necking, making it difficult for the particles to rearrange and accommodate further stretching. We observe that for thread rupture to occur a particle-free cross-section of the thread must develop, and as noted above, the location of rupture is dependent on the distribution of particles when necking begins and can occur anywhere along the length of the thread. It appears most likely the rupture results at a position determined by existing (as opposed to dynamically induced) fluctuations in \(\phi\) which become more pronounced relative to the mean—with consequently large material property fluctuations—as the thread diameter becomes small. Here, we assume uncorrelated number density fluctuations, such that the fluctuation in number \(N\) within a volume scales as \(\delta N/N \sim N^{-1/2}\). Following thread rupture the large number of particles present in the top and bottom spindles resist necking to the small scales necessary for secondary rupture and the accompanying generation of satellite drops. Both this and the initial resistance to necking lead us to conclude that large \(\phi\) provides a stabilizing effect on the liquid thread, whereas small \(\phi\) induces large fluctuations which are destabilizing. The cross-over is in the vicinity of \(\phi=0.10\) to 0.15 for the situations probed by our work.

B. Continuum breakdown

One typical technique employed in modeling the flow of suspensions is to neglect the discreteness of the solid particles and account for the particulate phase through an increased effective mixture viscosity, such as the well-known
Krieger form \( \eta_s(\phi) = \mu (1 - \phi/\phi_{\text{max}})^{-1.82} \) with \( \mu \) the suspending liquid viscosity and \( \phi_{\text{max}} \) an empirically determined maximum packing fraction. We find, not unexpectedly, that the continuum approach breaks down at small thread diameters and, more surprisingly, that the use of an effective viscosity does a very poor job of explaining the \( L/d \) behavior at pinch-off in the dripping mode.

Comparing our results for pinch-off structures with those of Shi et al.,\(^9\) we observe a similarity between our concentrated suspensions (Fig. 6) and their high viscosity pure liquid (glycerin, \( \mu = 12 \) Pa). Specifically, these authors noted a change in pinch-off structure with increasing viscosity similar to that observed here due to increasing \( \phi \): the structures changed from needle-and-sphere configurations to the pear-shaped drops described previously for high-\( \phi \) suspensions. Motivated by this similarity, a suspending liquid mixture of the same three components (in different proportions than described in Sec. II) was prepared such that the viscosity was \( \mu \approx 10 \) Pa. This viscosity is nearly equivalent to the effective viscosity obtained using the Krieger empirical correlation for the \( \phi = 0.25 \) suspension in the \( \mu = 4 \) Pa suspending liquid used in our other experiments, assuming \( \phi_{\text{max}} = 0.64 \). Drops were formed into silicone oil using this more viscous pure liquid and compared with the results from the previous, particle-laden, experiments (Fig. 20).

From Fig. 20 it is clear that the effective viscosity of the suspension does not determine the behavior. At pinching neither the overall thread length attained nor the structure are at all similar. The higher viscosity pure liquid has a much longer thread length and a structure more similar to that of the pure liquid lower viscosity mixture than the concentrated suspensions. Liquid viscosity is apparently governing the thread length at pinch-off. The change in thread length caused by an increasing solid fraction from \( \phi = 0 \) to \( \phi = 0.25 \) is negligible compared with the change from increasing the viscosity of the pure liquid.

The pinch-off structures formed are also not similar. The concentrated suspension pinch-off structure is pear shaped, while the equivalent viscosity pure liquid retains the needle-sphere structure. (This may appear to be a contradiction of the results of Shi et al.,\(^9\) but is, in fact, not because the drops formed here were into silicone oil rather than air as in the cited work.) This indicates that it is not simply a contribution to the effective shear viscosity of the mixture made by the particles which governs the pinch-off shape of the suspension. The discrete size of the particles as they are forced out of the liquid thread during necking apparently also plays a role due to the clustering of particles in regions at either end of the thread. The clustered particles are unable to rapidly integrate into the liquid cap at the orifice or into the forming drop. This causes the formation of the spindles and thus alters the geometry at pinch-off.

Both of these observations seem to preclude the use of standard effective viscosity descriptions in modeling particle-laden drop flows in accounting for the complete process. However, it may be possible to break the process down into two stages for the more concentrated suspensions where the use of an effective viscosity may be warranted in the first stage. These two stages will be referred to here as initial necking and final necking. Initial necking occurs while the thread width is large relative to the particle size. During this stage, the concentration of particles within a segment of thread apparently remains nearly uniform and the main role of the particles seems to be resisting further thinning and stretching of the thread. Final necking begins once the thread width has thinned to several particle diameters and a significant fluctuation in the concentration is sampled. Once this happens a particle-free region develops and thread rupture rapidly occurs. In this portion of the process, the substantial literature on pure liquid drop formation should be applicable.

### C. Particle effects on jetting transition

The presence of particles affects the transition from dripping to jetting, leading to drastically shorter jets and, at higher \( \phi \), smoothing the abrupt nature of the transition with respect to flow rate. In liquids, the onset of jetting is characterized by an abrupt and large increase in length of the coherent column of fluid connected to the orifice and is easily identified visually. Once particles are introduced, identifying...
this transition becomes more complicated. Eventually, at sufficiently elevated $\phi$, the process does not exhibit an identifiable transition but gradually changes from dripping to jetting over a range of flow rates. The introduction of particles thus appears to eliminate the critical nature of the parameter (the flow rate for a given system) associated with the transition in a pure liquid.

The particles within the forming jet appear to have two conflicting effects. The first of these is that the particles within the jet create disturbances to the free surface, inducing earlier breakup. The second effect is that large numbers of particles in the jet resist thinning of the liquid column. Based on the results presented in Figs. 14 and 15, it appears that the first effect is dominant and is responsible for the much shorter jet lengths observed for the suspensions. The second, stabilizing, effect would seem to induce earlier jetting of highly concentrated suspensions because the large numbers of particles present are unable to rearrange themselves to allow pinch-off before the next forming drop over-runs the first. This may explain, for low flow rates, the observed increase in $(L/d)_{\text{min}}$ relative to the pure liquid for the highest $\phi$ studied. A consequence of increased $(L/d)_{\text{min}}$ at low flow rates is the decrease in drop size for large $\phi$ in Figs. 8 and 9. From Figs. 14 and 15 it is evident that the concentrations exhibiting this decrease in drop size (0.20 and 0.30 for the smaller orifice and 0.40 for the larger one) correspond to those with longer $(L/d)_{\text{min}}$.

The consideration of particle rearrangement within the liquid column seems to be critical to understanding the process. The ease with which the particles can accomplish this rearrangement determines how the jet transition occurs. Particle rearrangement should be easiest for small particles relative to the orifice (large $d/d_p$) and low $\phi$. In fact as the particles become progressively smaller, the behavior of the pure liquid is expected to be asymptotically recovered, an assertion supported by the experiments performed using smaller particles. Here we recall that the jet length increased relative to the larger particle suspensions. At the other extreme (small $d/d_p$) individual particles take up a greater percentage of the liquid cross-section and rearrangement is more difficult. In all of this discussion, it should be noted that rearrangement is within a liquid column where the motions encounter resistance by a flexible surface and not a rigid boundary. Clear evidence of surface deformation by the particles is seen in the work we have presented. It is to be expected that particle and drop scale will play a strong role in this aspect of the problem, as capillary forces become stronger as the scale is reduced.

V. CONCLUSIONS

This work has experimentally examined the formation of drops of suspension composed of a viscous liquid and spherical, neutrally buoyant, noncolloidal particles. The presence of the particles introduces a number of effects which make the process distinct from pure liquid drop formation.

In the dripping mode the pinch-off structures for the particle-laden systems are qualitatively different from the pure liquid case. For low concentration suspensions (up to $\phi=0.10$) the macroscopic behavior is similar to that of the pure liquid in that the structure is that of the well-known needle-sphere configuration familiar in the study of pure liquid drops.\(^8,9\) although either individual or groups of particles can become trapped in the forming neck and produce satellite drops which would otherwise not be present. As $\phi$ is increased the pinch-off structures change: the particles form thick cone-like structures (termed “spindles” here) both near the orifice and the trailing edge of the forming drop. This change in structure is not explained simply as the result of an increase in the effective viscosity of the mixture.

During necking, particles in the thread resist this further thinning and, depending on the number of particles present, this resistance can have either a destabilizing or a stabilizing effect on the thread relative to the case of the pure liquid. The presence of a small number of particles in the thread introduce regions within the thread which cannot thin beyond the size of an individual particle and lead to earlier rupture. A large number of particles in the thread tend to have a stabilizing effect both before and after pinch-off. In this case there are sufficient particles present in the thread that their individual motions are restricted and the necking of the thread is slowed. These two conflicting particulate effects provide an explanation for the observed changes in satellite drop generation and jetting transition with increasing $\phi$. The stabilizing effect of high $\phi$ suspensions leads to suppression of satellite drops and earlier jetting while the destabilizing effect is responsible for reducing the coherent jet lengths.

Finally, the observation that the process appears to occur in two stages may be useful in future attempts to model this behavior. In the initial stages of necking, where the thread width is much greater than individual particles, the use of an effective viscosity description of the process may be able to capture the observed resistance to thinning. In the second stage, the suspending fluid properties appear to be critical.

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