Scalings for Droplet Sizes in Shear-Driven Breakup: Non-Microfluidic Ways to Monodisperse Emulsions

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Abstract: We review studies of a drop of viscous liquid, suspended in another liquid, and undergoing breakup in an impulsively started shear flow. Stokes flow conditions as well as the effects of inertia are reported. They reveal a universal scaling for the fragments, which allows one to use sheared emulsions to produce monodispersity as an alternative to microfluidic devices.

Keyword: Drop deformation, drop size distribution, numerical methods.

1 Introduction

Drop breakup is important for a wide range of engineering and biomedical applications including production and processing of emulsions and immiscible polymer blends, aerosols, and drug delivery systems. The application of shear and inertia to a premixed emulsion of various drop sizes is one technique for the production of monodisperse droplets. Experimental, theoretical and numerical studies of drop breakup in imposed flows are reviewed in Rallison (1984); Bentley and Leal (1986); Stone (1994); Basaran (2002); Guido and Greco (2004); Cristini and Tan (2004). Criteria for breakup are investigated experimentally and analytically in, for example, Bentley and Leal (1986); Navot (1999); Blawzdziewicz, Cristini, and Loewenberg (2002, 2003). The distribution of drop fragments resulting from breakup in shear flow is studied in Cristini, Blawzdziewicz, Loewenberg, and Collins (2003); Schmitt, Leal-Calderon, and Bibette (2003), among others. The drop size distribution resulting from breakup events determines the mechanical properties and rheology of such mixtures. These investigations are therefore key to understanding the microstructure of immiscible blend systems in materials processing.

1.1 Physical Parameters for Stokes Flow

A drop of viscosity \( \mu_d \) and density \( \rho_d \) is suspended in another liquid of viscosity \( \mu_m \) and density \( \rho_m \). At time \( t = 0 \), the drop is spherical with radius \( a \). Another option is to place a drop in the liquid that already flows at an imposed constant shear rate \( \dot{\gamma} \). For \( t > 0 \), the ambient fluid undergoes simple shear in the x-z plane with velocity

\[
\dot{x} = \frac{\dot{\gamma}}{2} (S + A)x, \quad S = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad A = \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix},
\]

(1)

where \( x = \begin{pmatrix} x \\ z \end{pmatrix} \). The symmetric matrix \( S \) generates elongation along \( x = z \) and the antisymmetric matrix \( A \) generates rotation.

We take the densities of the drop and matrix liquids to be the same (\( \rho_d = \rho_m \)) and focus on the competition between viscous force, capillary force and inertia. The viscosity ratio of the drop to matrix liquids is denoted

\[
\lambda = \frac{\mu_d}{\mu_m}.
\]

(2)

The time scales \( \tau_\gamma \) for drop stretching in the flow, and \( \tau_\sigma \) for drop shape relaxation by surface tension are given by

\[
\tau_\gamma = \frac{1}{\dot{\gamma}}, \quad \tau_\sigma = \frac{\mu_m a}{\sigma}.
\]

(3)

The ratio of these time scales defines the capillary number

\[
Ca = \frac{\mu_m \dot{\gamma} a}{\sigma},
\]

(4)

where \( \sigma \) is the surface tension.
1.2 **Critical Condition for Stokes Flow**

Initially, the drop is stretched by viscous shear stress in the external flow. Shear stress is proportional to the viscosity multiplied by the rate of elongation. Continuity of shear stress across the drop/matrix interface therefore yields

\[ \lambda \times \{ \text{rate of elongation in the drop} \} \sim \{ \text{rate of elongation in the matrix} \}. \]  

(5)

The decomposition (1) for simple shear in the matrix fluid shows that

\[ \{ \text{rate of elongation in the matrix} \} \sim \{ \text{rate of rotation in the matrix} \} \]  

(6)

In addition, velocity is continuous across the interface; thus,

\[ \{ \text{rate of rotation in the matrix} \} \sim \{ \text{rate of rotation in the drop} \} \]  

(7)

Retracing the steps back to (5),

\[ \lambda \times \{ \text{rate of elongation in the drop} \} \sim \{ \text{rate of rotation in the drop} \}. \]  

(8)

This shows heuristically that when the drop is very viscous (\( \lambda \gg 1 \)), the rate of elongation in the drop is small compared to the rate of rotation in the drop; i.e., the drop can not break. The critical viscosity ratio \( \lambda^* \) beyond which the drop does not break for Stokes flow is roughly 3.

For \( \lambda < \lambda^* \), there is a critical capillary number, above which the drop breaks [Rallison (1984); Khismatullin, Renardy, and Cristini (2003)]:

\[ Ca^* \sim 1. \]  

(9)

Critical conditions are shown in Fig. 1: \( Ca^* \to \infty \) at both \( \lambda = 0 \) and \( \lambda = \lambda^* \). We define a dimensionless time \( \tilde{t} = t/\tau_\gamma \), and radius \( \tilde{a} = a/a^* \), where \( a^* \) denotes the radius corresponding to \( Ca^* = \mu_m^* {\dot{a}}^* / \sigma \). (9) gives a simple relationship between generated droplet size and imposed flow rate. The larger the flow rate, the smaller the droplet size.

Accurate critical capillary numbers are intrinsically difficult to obtain due to the divergence of the time \( \tilde{t}_0 \) required to reach stationary state [Blawdziewicz, Cristini, and Loewenberg (2002)],

\[ \tilde{t}_0 \sim (1 - \overline{\alpha})^{-1/2}. \]  

(10)

Experimental data (inset of Fig. 1) have large scatter due to the uncertainty of determining whether a drop is gradually breaking or gradually attaining a stationary configuration at shear rates close to the critical value. The same difficulty applies to numerical computations.

2 **Numerical methods**

The two main approaches to simulating drop deformation are interface tracking and interface capturing.

2.1 **Interface tracking methods**

Interface tracking methods include boundary-integral methods [Pozrikidis (1992); Prosperetti and Oguz (1997); Coulliette and Pozrikidis (1998); Cristini, Blawdziewicz, and Loewenberg (1998); Zinchenko, Rother, and Davis (1999); Cristini, Blawdziewicz, and Loewenberg (2001); Hou, Lowengrub, and Shelley (2001); Pozrikidis (2002); Ye, Matar, de Ortiz, and Hewitt (2003)], finite-element methods [Wilkes, Phillips, and Basaran (1999); Hooper, Cristini, Shakya, Lowengrub, Macosko, and Derby (2001); Hooper, de Almeida,
Scalings for Droplet Sizes in Shear-Driven Breakup

Figure 2: Adaptive 3-D boundary-integral simulation of drop breakup in shear flow. The simulation is continued past the transition by reconnecting the computational mesh [Cristini, Blawzdziewicz, and Loewenberg (1998, 2001)]. Macosko, and Derby (2001); Notz, Chen, and Basaran (2001)], and immersed-boundary methods [Nobari, Jan, and Tryggvason (1996); Tryggvason, Bunner, Esmaeili, Juric, Al-Rawahi, Tauber, Han, Nas, and Jan (2001); Shin and Juric (2002); Esmaeili (2005)]. These are 'sharp-interface' methods; the computational mesh elements lay in part or fully on the fluid/fluid interfaces. Such methods are very accurate for simulating the onset of breakup and coalescence transitions but have difficulties in simulating through and past the transitions.

In boundary-integral methods, the flow equations are mapped from the immiscible fluid domains to the sharp interfaces separating them, thus reducing the dimensionality of the problem (the computational mesh discretizes only the interface). In finite-element methods, the fluid domains are discretized by a volume mesh and thus the dimensionality is not reduced. Both these approaches lead to accurate and efficient solution of the flow equations because the interface is part of the computational mesh and the equations and interface boundary conditions are posed exactly. In immersed-boundary methods, the interfacial forces are calculated on a surface mesh distinct from the computational volume mesh where the flow equations are solved; thus, in addition, interpolation onto the volume mesh is needed. A three-dimensional boundary integral simulation is shown in Fig. 2. The first three frames show the evolution of a drop towards breakup and the formation of a thinning liquid thread separating two large daughter drops. The labels report the dimensionless time from breakup. The calculated drop shapes (computational mesh) compare well with experimental data (solid contour), demonstrating the high accuracy of the numerical method. These simulations use an adaptive triangulated mesh [Cristini, Blawzdziewicz, and Loewenberg (2001)].

Figure 3 shows an application of the finite-element method to high-Reynolds-number satellite production from a jet [Notz, Chen, and Basaran (2001)]. Cross-sections of the computed evolution in time (top) compare well with experimental data (bottom). The computational accuracy allows the authors to recover features of the evolutions such as capillary waves (a-c), overturning of the top and bottom of the satellite in (f), spade-shaped profiles (g, h), and spawning of a subsatellite.

Figure 3: From Figs. 2 and 3 of Notz, Chen, and Basaran (2001). Finite-element simulation (top) of satellite formation and dynamics from a jet compared to an experiment (bottom). Near breakup and coalescence transitions, sharp-interface models break down because of the formation of singularities in flow variables [Hou, Lowengrub, and Shelley (1997)], and complex ad hoc cut-and-connect algorithms have been employed [Mansour and Lundgren (1990); Tryggvason, Bunner, Esmaeili, Juric, Al-Rawahi, Tauber, Han, Nas, and Jan (2001); Cristini, Blawzdziewicz, and Loewenberg (2001)] to change the topology of the meshes and continue simulating through
a transition. A method to automatically reconnect sharp interfaces has been recently developed by Shin and Juric (2002). Nobari, Jan, and Tryggvason (1996) reconnected the interfaces as their separation fell under a prescribed value but noticed that the flow can depend sensitively on the time at which the interface reconnections are performed. Reconnection conditions based on asymptotic theories for liquid thread pinch-off and film drainage are also used to extrapolate to the instant of breakup or coalescence [Keller and Miksis (1983); Eggers and Dupont (1994); Eggers (1995); Blawzdziewicz, Cristini, and Loewenberg (1997); Lister and Stone (1998)]. For instance, the simulation of Fig. 2 (bottom) employs a cut-and-connect mesh algorithm after the asymptotically linear thinning of the liquid thread [Lister and Stone (1998)] has been established. This allows simulations to be continued past the transition while preserving accurate information on breakup time and fragment sizes.

2.2 Interface-capturing methods

Simulations through breakup using interface-capturing methods do not require mesh cut-and-connect operations because the mesh elements do not lay on the interface, but rather the interface evolves through the mesh. Such methods include the lattice-Boltzmann and lattice-gas [Rothman and Zaleski (1997); Chen and Doolen (1998); Nourgaliev, Dinh, Theofanous, and Joseph (2003); Sankaranarayanan, Kevrekidis, Sundaresan, Lu, and Tryggvason (2003); Watanabe and Ebihara (2003)], constrained-interpolation-profile [Yabe, Xiao, and Utsumi (2001)], level-set [Osher and Fedkiw (2001); Jimenez, Sussman, and Ohta (2005); Hogea, Murray, and Sethian (2005)], volume-of-fluid [Scardovelli and Zaleski (1999)], coupled level-set and volume-of-fluid [Sussman and Puckett (2000)] and partial-miscibility-model and phase-field methods [Lowengrub, Goodman, Lee, Longmire, Shelley, and Truskinovsky (1999); Anderson, McFadden, and Wheeler (1998); Yabe, Xiao, and Utsumi (2001); Jaqcmín (1999); Badalassi, Ceniceros, and Banerjee (2003); Chen (2002); Yue, Feng, Liu, and Shen (2004, 2005)]. The fluid discontinuities (e.g., density, viscosity) are smoothed and the surface tension force is distributed over a thin layer near the interface to become a volume force (surface tension being the limit as the layer approaches zero thickness). Interface-capturing methods are then ideal for simulating breakup and coalescence in immiscible two-fluid systems (and the effect of surfactants) and for three or more liquid components, and can be especially powerful for micro channel design. Lattice-Boltzmann methods are based on a particle distribution function and on averaging to capture the macroscopic behavior. The constrained-interpolation-profile, level-set and volume-of-fluid methods describe the macro scale directly and use auxiliary functions advected by the flow (e.g., level-set, volume fraction, and color functions) to mark the different fluid domains.

In Fig. 4, we reproduce a simulation of drop breakup in shear flow using a volume-of-fluid method [Li, Renardy, and Renardy (2000); Renardy, Cristini, and Li (2002); Renardy and Cristini (2001a,b)]. The drop is strongly stretched in the supercritical flow leading to rupture into numerous fragments of alternating sizes (see for comparison, Cristini, Guido, Alfani, Blawzdziewicz, and Loewenberg (2003); Tan, Cristini, and Lee (2006)). The diffuse-interface (phase-field) approach is based on free-energy functionals and uses chemical diffusion in narrow transition layers between the different fluid components as a physical mechanism to smooth flow discontinuities and to yield smooth evolution through breakup and coalescence. The singularity which arises at a pinch-off [Papageorgiou (1995); Eggers (1995); Lister and Stone (1998)] affects the resolution of the subsequent small drops.

Figure 4: 3-D volume-of-fluid simulation of drop breakup in simple shear, view along the velocity gradient. Fig. 16 of Li, Renardy, and Renardy (2000).
In Renardy and Renardy (2002), a parabolic reconstruction of the interface in the surface tension (PROST) force for volume-of-fluid methods is formulated. The algorithm achieves convergence with spatial refinements in repeated drop breakup simulations. The main features of PROST are that it reconstructs the interface with a least square fit of a quadratic interface to the values of the volume fraction function for each interface cell, and calculates the curvature directly from the quadratic surface at cell centers. This avoids numerical differentiation of the discontinuous volume fraction function. The PROST algorithm is extended to the Giesekus constitutive law for viscoelastic liquids in Khismatullin, Renardy, and Cristini (2006), where the experimental data of Guido, Simeone, and Greco (2003) are simulated.

Figure 5 (a)-(c) is a breakup simulation for Stokes flow with PROST, and the close agreement with the boundary integral simulation (d) illustrates the accuracy of this sharp-interface volume-of-fluid method.

Figure 6 shows a breakup simulation with inertia. The drop evolves to an ellipsoidal shape, then to a dumbbell with the central portion continuing to stretch and thin. There is vortical motion inside the bulbs which detach to form the first daughter drops. When the mother drop is large enough, the ends of the remaining portion retracts slightly due to surface tension, then bulb up and end-pinching repeats. If the capillary number is sufficiently high, end-pinching leaves behind a long cylindrical thread, where capillary wave breakups are observed. This results in a distribution of large satellite droplets, interspersed with small droplets.

2.3 Adaptive Mesh

Accurate numerical simulations require the computational mesh to resolve both the macro (e.g., droplet size, channel geometry) and micro scales where pinch-off or coalescence occurs; for example, local interface curvature, separation between interfaces, surfactant distributions, and relevant stresses and forces. Adaptive mesh algorithms greatly increase accuracy and computational efficiency in boundary-integral [Cristini, Blawzdziewicz, and Loewenberg (2001)], finite element [Wilkes, Phillips, and Basaran (1999); Hooper, Cristini, Shakya, Lowengrub, Macosko, and Derby (2001); Cristini, Hooper, Macosko, Simeone, and Guido (2002)], immersed boundary [Tryggvason, Bunner, Esmaili, Juric, Al-Rawahi, Tauber, Han, Nas, and Jan (2001)], and interface capturing [Agresar, Linderman, Tryggvason, and Powell (1998); Sussman, Almgren, Bell, Colella, Howell, and Welcome (1999); Provatas, Goldenfeld, and Dantzig (1999); Ubbink and Issa (1999); Ceniceros and Hou (2001); Jeong, Goldenfeld, and
Figure 6: Numerical simulation of fragmentation. $Re = 12, Ca = 0.175 = 1.14Ca^*, \lambda = 1$. $t = 0, 1, 9, 19, 21, 22, 22, 22, 24, 24, 24, 24, 24$. The VOF-PROST algorithm is used.

Dantzig (2001); Jeong, Dantzig, and Goldenfeld (2003); Ginzburg and Wittum (2001); Zheng, Roach, and Ismagilov (2003) methods. Figure 7 shows novel unstructured adaptive meshes in level-set simulations of Anderson, Zheng, and Cristini (2005); Zheng, Lowengrub, Anderson, and Cristini (2005). Their algorithm automatically imposes a mesh element size proportional to the distance from the interface. As the interfaces deform, approach or pinch-off, the mesh dynamically maintains accurate resolution of the flow near the interface. The 3-D simulation accurately describes drop breakup during retraction of a previously elongated drop.

Figure 7: Adaptive unstructured meshes of tetrahedra maintain computational accuracy during simulations (data from Zheng, Lowengrub, Anderson, and Cristini (2005)). Some tetrahedra may appear skewed as a result of projecting the 3-D mesh onto the plane of the figure.

3 Fragment size distribution for Stokes flow

Fragments may be categorized roughly into three types. The first 'daughters' produced by the primary breakup event have the largest volumes. In Fig. 5, these roughly split the mother drop at criticality. Next comes the neck region between the dumbbells. Capillary force breaks this region into smaller but still substantial 'satellites'; see, for instance, the middle satellite of the last frame of Fig. 6. The third category is comprised of the smallest fragments which lie between the satellites and make up only a few percent of the mother drop volume. Figures 8 and 9 show experimental photographs of fragments produced by breakup in shear. The fragments are found to scale with the critical size $a^*$, independent of the parent drop radius $a_0$, so that an appropriate dimensionless daughter radius is $\bar{a_d} = a_d/a^*$. This scaling has also been observed in experiments of drop breakup in shear flow [Marks (1998); Cristini, Guido, Alfani, Blawzdziewicz, and Loewenberg (2003)], and in numerical simulations of drop breakup in stochastic flows [Cristini, Blawzdziewicz, Loewenberg, and...
Collins (2003)] and shear flows with inertia [Renardy and Cristini (2001a); Renardy, Cristini, and Li (2002); Renardy and Cristini (2001b)].

Figure 8: Fig. 2 of Cristini, Guido, Alfani, Bławzdziwicz, and Loewenberg (2003). Sequence of breaking drop in shear flow \( \lambda = 1.02, \bar{\eta}=1.17 \); time \( t \) as indicated, predicted values shown in parentheses. Images from experiment; profiles from numerical simulation with boundary integral method up to pinch-off event. Top-view of drop, in direction parallel to velocity gradient.

Figure 8 shows a fluid pair with viscosity ratio \( \lambda = 1.02 \) in Stokes flow just above criticality. Numerical simulations with a boundary integral method are plotted together. In figure 9, the mother drop size is larger so that more droplets are created. The first daughter drops attain a constant value \( \bar{a}_d \) for sufficiently super-critical \( \bar{\eta}_0 \). In the figures, \( \bar{a}_d \sim 0.90 \) for \( \bar{\eta}_0 > 1.4 \); smaller parent drops have insufficient volume to produce daughters of this size. Experiments at other fluid properties show the same qualitative behavior of daughter drops that saturate to a specific percentage of the critical drop size. After the first daughter drops separate, a ‘neck’ is left behind which breaks up further, either by end-pinching or capillary-wave instability [Stone and Leal (1989)]. The larger satellite drops scale with the critical size drop, just as in the case of the first daughters. For small satellite drops, the external flow is unimportant. Their sizes are determined by the width of the neck produced by the primary pinch-off event. For \( \bar{\eta}_0 >> 1 \), the neck width is found to scale with \( a^* \). In particular, the dimensionless

\[
\pi r_n^2 l_n = \bar{\eta}_0 - 2\bar{\eta}_d, \quad (11)
\]

where \( \bar{\eta}_0 - 2\bar{\eta}_d \) is the excess volume of the parent drop. For large \( \bar{\eta}_0 \), \( r_n \) approaches a fixed value, and satellites produced by the breakup of the neck therefore scale with the critical drop size \( a^* \). Thus, all drop fragments scale with the critical drop size.

Cumulative size distributions of drop fragments obtained from Stokes flow experiments are shown in Fig. 10 [Cristini, Guido, Alfani, Bławzdziwicz, and Loewenberg (2003)]. The vertical axis shows the fraction of

Figure 9: As in Fig. 8 with \( \bar{\eta} = 1.38 \). Side-view of drop, in direction parallel to vorticity. Flow stopped in last frame.
daughter drops of size less than or equal to the number plotted on the horizontal axis. The distributions for each experiment show two distinct daughter drops and three size classes of satellite drops (1) \(0.4 < \overline{a} < 0.8\), (2) \(0.10 \leq \overline{a} \leq 0.3\), and (3) \(\overline{a} \leq 0.1\). The number of satellites within each class is denoted by \(N_i (i = 1, 2, 3)\). The largest (class 1) satellites are clearly distinguishable. The number of class 1 and 2 satellites are approximately equal, consistent with the alternating periodic sequence of large and small satellites that is typical of jet breakup. Accordingly, class 2 satellites are defined as the \(N_2\) largest subclass-1 fragments, where \(N_2 = N_1 + 1\). The smallest (class 3) fragments account for only \(\approx 0.1\%\) of the parent drop volume. Figure 11 replots the same information as Fig. 10, but shows separately the cumulative distribution of the primary daughter drops and the class 1 and 2 satellites. For large \(\overline{a}_0\), the distribution of large satellites shows a tendency to become independent of the parent drop size, consistent with the scaling arguments discussed earlier. Zhao and Goveas (2001) observed that a narrower size distribution of fragments is produced by drop breakup in viscoelastic, rather than Newtonian fluids.

Methods for producing controlled micro-sized droplets mostly rely on the use of surfactants and complex fluids in a variety of flows such as shear (see section 5 below), co-flowing streams [Umbanhowar, Prasad, and Weitz (2000)] and extrusion flow [Kobayashi, Yasuno, Iwamoto, Shono, Satoh, and Nakajima (2002)]. Even without the use of surfactants, Cristini, Guido, Alfani, Blawzdziewicz, and Loewenberg (2003) demonstrated that nearly bi-disperse emulsions of large numbers of microscopic droplets of controlled sizes and generation times can be achieved; see Figs. 10 and 11. The two sizes alternate, perhaps because of the asymmetrical evolution of the drop interface near the pinch-off region into cones with different angles during the final stages of pinch-off [Lister and Stone (1998)].

The scaling for the first daughter drops and neck fragments aid in interpreting the experimental data of Section 4.6 of Marks (1998). His Fig. 4.6.1.a, reproduced in Fig. 12 gives five sample histograms of daughter drop sizes, placed in four bins. The 1.0 bin, 0.75 bin, 0.50 bin and 0.25 bin represent \(0.75 < \text{Ca}_d/\text{Ca}^*\), \(0.50 < \text{Ca}_d/\text{Ca}^* < 0.75\), \(0.25 < \text{Ca}_d/\text{Ca}^* < 0.50\), \(\text{Ca}_d/\text{Ca}^* < 0.25\), respectively. The first daughter drops fall into the 1.0 bin. The neck fragments mostly fall into the 0.75 bin, while tiny drops fall into the other two bins. Beyond the production of the two largest daughters, no other drops of this order of magnitude are produced because the rest of the drops come from the elongated neck. The trend, therefore, is that as the mother capillary number
increases, the volume fraction in the 0.75 bin increases and eventually dominates. The histogram at $K_i = 1.0$ denotes the fact that just above criticality, most of the drop goes into the first daughter drops in the 1.0 bin. Our results suggest that just two daughter drops fall into the 1.0 bin. These graphs represent typical histograms and were chosen to show a range of $K_i$’s.

4 Inertia-induced breakup

Inertia is measured by the Reynolds number

\[ Re = \frac{\rho \dot{\gamma} a^2}{\mu} \]  

There are three trends for the overall effect of inertia [Li, Renardy, and Renardy (2000); Renardy and Cristini (2001a)]. First, inertia rotates the drop, so that at higher Reynolds numbers, the steady states are more aligned toward the vertical than in Stokes flow and therefore the drop experiences greater shear. Secondly, in Stokes flow, the flow inside the drop consists of a single vortical swirl. With increased inertia, the velocity field bifurcates, with additional swirls. Thirdly, the length of the drop in steady states just below breakup shortens as inertia increases. The symmetry across the mid-plane of the steady state, evident in Stokes flow, is lost.

For large Reynolds numbers, the Reynolds stress is of order $\rho |\dot{\mathbf{v}}|^2$ where the speed is $\dot{\gamma} a$. When this inertial stress reaches the magnitude of capillary stress, which is of order $\sigma / a$, the drop breaks: $\rho \dot{\gamma} a^2 \sim \sigma / a$. Division by $\rho \dot{\gamma}$ yields the order of magnitude of the critical Reynolds number, above which the drop breaks:

\[ Re \sim 1 / Ca. \]  

In fact, the ratio of inertial to capillary forces is the Weber number

\[ We = Re Ca. \]  

In the inviscid limit, the critical Weber number is approximately 3.3, which is consistent with the scaling of (14).

Renardy and Cristini (2001b) and Renardy, Cristini, and Li (2002) focus on the case of equal viscosities and densities for the drop and matrix liquids, and investigate neck breakup by keeping the flow strength the same and increasing the parent drop size. For example, when fluid properties and external flow are fixed at $Re = 391Ca^2$, the first daughter drops are approximately 50% to 60% of the critical drop volume. The volumes of the neck drops are 10% to 15% of the critical drop volume. Each main drop is followed by one or more small drops, or ‘moons’.

Figure 13 shows evolution at $Re = 15, Ca = 0.196 = 1.27Ca^*$, where the first daughter drop volume is 54% of the critical volume. The volumes of the main neck fragments again lie between 10% to 17% of the critical drop volume. Each main drop is followed by one or more small drops, or ‘moons’.

Figure 14 shows mother drops indicated by circles. The first daughter drops (indicated by stars) approach 54% of the critical volume (radius $a_d \approx 0.81a^*$) as the mother
Asterisks denote first daughter drop data; mother drop varies. Circles represent mother drop data. Properties and flow strength fixed, while the radius of the discretized mesh are not included.

At the instant when the first daughter drops pinch off, the neck is elongated to roughly its maximum length, and cylindrical in a central region, tapering off to pencil tips toward the ends. Renardy, Cristini, and Li (2002) tabulates the ‘effective’ capillary number for the cylindrical neck $Ca_n$. This is calculated from the length $L$ of the neck (projected onto the x-axis) just after the first daughter drops detach, and the volume in the neck. An effective radius $r_n$ is calculated for this, assuming the neck is cylindrical, and of length $L$, or $\pi r_n^2 L = (4/3) \pi a_0^3 - 2(4/3) \pi a_d^3$, so that

$$r_n = \left[ \frac{4a_0^3(1 - 2(a_d/a_0)^3)}{3L} \right]^{1/2},$$

where $a_d$ is the first daughter drop radius, and

$$Ca_n = Ca(r_n/a_0).$$

At $Re = 15$, the drops which result from the neck have capillary numbers $Ca_{D\text{neck}}$ between 0.08 and 0.09, and if $Ca_{D\text{neck}} = F Ca_n$, then $F$ ranges from 2.4 to 2.8. If the breakup were a capillary breakup of a viscous jet in a quiescent liquid as considered in Tjahjadi, Stone, and Ottino (1992), then

$$Ca_{D\text{neck}} = F Ca_n, \quad F = 2.$$
Here, however, we have repeated end-pinching, which produces larger drops.

Figure 15 shows the volume fraction relative to the mother drop volume, $V_d/V_a = (Ca_d/Ca^*)^3/(Ca_a/Ca^*)^3$, for each $Ca_d/Ca^*$. There are always tiny drops between the main fragments which have not been included in the graphs. The trend, as the mother drop size increases, is the growth in drops of size roughly half that of the mother drop. The first daughter drops remain as the largest drops, at roughly 0.8 of the mother drop volume, and there are no other drops of that magnitude in the simulations reported.

The scaling for the drops As the capillary number increases along the parabola shown in Fig. 14, the first daughters saturate to a specific percentage of the critical drop size. The neck radius determines the size of the subsequent drops because they are of the same order of magnitude. As the capillary number increases to infinity, the ratio of daughter drop radii to mother drop radius decreases to zero, $a_d/a_0 \to 0$ and equation (16) shows that the effective neck radius satisfies

$$r_n \sim L^{-1/2} a_0^{3/2}.$$  \hspace{1cm} (19)

The capillary numbers covered in Renardy, Cristini, and Li (2002) are not sufficiently large for this scaling to apply, because the ratio $a_d/a_0$ is not negligible. Therefore, in our regime, $r_n$ is influenced by the two competing effects in equation (16): $L$ increases with increasing $Ca$, and the numerator $1 - 2(a_d/a_0)^3$ also increases.

For large capillary numbers, the effect of surface tension is initially small and the drop stretches following the simple shear flow, elongates, until eventually, the cross-section of the drop evolves to a circular shape, length scales are reached at which surface tension becomes important, and pinching begins. This pinching begins when an effective radius is approximately the critical radius:

$$r_e \approx r_c, \quad r_c = a_0 \frac{Ca}{Ca^*}.$$  \hspace{1cm} (20)

This effective radius might be interpreted either as the effective neck radius for an extremely elongated case, but more appropriately for our simulations, as an average radius for a less elongated shape, where $\pi r_e^2 L_T$ equals the original drop volume, with $L_T$ being the total length. When pinch-off occurs, the total length of the drop is of order

$$L_T \sim r_c^{-2} a_0^3 \sim a_0 \left(\frac{Ca}{Ca^*}\right)^2.$$  \hspace{1cm} (21)
Note that along the parabola of Fig. 14, \( Re = K Ca^2 \), and equation (21) gives

\[ L_T \sim Re. \]  

(22)

5 Complex Fluids

Emulsification is typically promoted by surfactants which decrease surface tension on the droplet interfaces [Stone and Leal (1990); Milliken, Stone, and Leal (1993); Milliken and Leal (1994); Pawar and Stebe (1996); Eggleton, Pawar, and Stebe (1999); Li and Pozrikidis (1997); Siegel (1999); Maldarelli and Huang (1996)]. Monodispersed emulsions of large numbers of droplets with controlled sizes are generated using the tip-streaming phenomenon due to redistribution of surfactants to localized end caps on the drop interface [Bruijn (1993); Eggleton, Tsai, and Stebe (2001)]. Experimental studies show that the use of complex fluids can lead to a dramatic alteration of the rupturing phenomenon Utracki and Shi (1992). Mason, Bibette, and Weitz (1996); Mason and Bibette (1997) produced monodisperse emulsions in a narrow-gap shear cell with a viscoelastic medium; Fig. 16 shows their size distribution.

Figure 16: Fig. 8 of Mabille, Schmitt, Gorria, Calderon, Faye, Deminière, and Bibette (2000). Size distribution of an emulsion obtained by shearing at 2500 s\(^{-1}\), a premixed emulsion whose elasticity is mainly provided by the continuous phase: \( C = 30\%; \Psi = 40\%; D[4,3] = 5.5\mu m; U = 19.3\% \). Zhao and Goveas (2001) study a dispersed Newtonian phase in a Newtonian medium and a viscoelastic medium under the same shear rates. Initially, the emulsion contains mother drops of various sizes. During satellite formation, the viscoelastic medium produces cylinders of more uniform radii than the Newtonian medium. The cylinders break up due to capillary waves and result in more uniform daughter drops; see Fig. 17.

Figure 17: From Figs. 3 and 4 of Zhao and Goveas (2001). Left: (a) Presheared Newtonian emulsion containing 2 wt \% silicone oil in glycerol with 2 wt \% TWEEN-80, which corresponds to a viscosity ratio of 0.453. (b) Newtonian emulsion under shear. Drops of different sizes are deformed into cylinders which have different widths at breakup. The resulting daughter drops are nonuniform along the length of the cylinder. Right: (a) Presheared viscoelastic emulsion containing 2 wt \% silicone oil in an aqueous solution of 12 wt \% PVP and 2 wt \% SDS, which corresponds to a viscosity ratio of 0.482. (b) Viscoelastic emulsion under shear, showing that cylinders have similar widths at breakup. The resulting daughter drops are very uniform.

6 Conclusions

Theoretical, numerical and experimental investigations of drop breakup in sheared emulsions were reviewed. The magnitude of flow and fluid properties are important in determining the drop size distribution. The examination of universal scalings for this type of flow leads
to the idea that sheared emulsions can be used as an alternative to microfluidics for the production of controlled monodisperse droplets. Drop sizes can be optimized with the use of direct numerical simulations. Open challenges to be addressed include the study of the effect of complex fluids such as viscoelastic liquids to achieve desired drop sizes. Finally, for non-dilute emulsions, effects of coalescence events need to be taken into account [Lappa (2005a,b)].

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References


Scalings for Droplet Sizes in Shear-Driven Breakup


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