RICE UNIVERSITY

Drop Breakup in Dilute Newtonian Emulsions under Steady Shear

by

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ABSTRACT

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High-speed video microscopy has been used to study drop breakup in dilute Newtonian emulsions under steady shear. Fundamental experimental studies on drop breakup have been limited to breakup in quiescent matrix or under pseudo-equilibrium conditions. This thesis represents the first direct visualization of drop breakup under steady shear at high capillary numbers (Ca).

The mechanisms of drop breakup depend on Ca and the viscosity ratio (λ). At $Ca-Ca_c$, drops are broken up via necking. At $Ca<2Ca_c$, drop breakup is caused by end pinching. At $Ca>2Ca_c$, the capillary instability is the dominant breakup mechanism.

For $Ca>2Ca_c$, breakup dynamics are strongly controlled by λ. For $0.1<\lambda<1$, drops with different initial sizes deform into threads with the same radius at breakup. The wavelength of the capillary instability is uniform along the length of a thread and from thread to thread. Fairly monodisperse dilute emulsions are obtained due to this size selection mechanism, with the average drop size being inversely proportional to the shear rate. For $1<\lambda<3.5$, the breakup mechanism is similar to that for $0.1<\lambda<1.0$, except that the satellite drops are substantially larger, resulting in polydisperse emulsions. For $\lambda<0.1$, the daughter drops are formed from long wavelength capillary instability and may break again. This induces collisions between drops, which in turn results in irregular drop re-breaking and
coalescence, producing polydisperse emulsions. This re-breaking mechanism has not been observed in previous studies in the literature.

Drops reach a pseudo-steady state before the capillary instability starts to grow. At this pseudo-steady state, the shear stress and the capillary pressure almost balance each other, determining a definite thread radius, which is independent of the initial drop size. We define a dimensionless thread number as the ratio of the two forces. The thread number is only a function of $\lambda$, and shows a minimum in $\lambda$. The measured thread number is in agreement with the slender body theory of Hinch and Acrivos (1980).

Drops deform pseudo-affinely for $0.1 < \lambda < 1.0$, but deformation deviates from being pseudo-affine otherwise.
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<th>Symbol</th>
<th>Physical Meaning</th>
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<tr>
<td>$a$</td>
<td>Initial drop radius</td>
<td>μm</td>
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<td>$a_c$</td>
<td>Critical drop radius corresponding to $Ca_c$</td>
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<td>[-]</td>
</tr>
<tr>
<td>$C$</td>
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</tr>
<tr>
<td>$Ca$</td>
<td>Capillary number</td>
<td>[-]</td>
</tr>
<tr>
<td>$Ca_c$</td>
<td>Critical capillary number for breakup</td>
<td>[-]</td>
</tr>
<tr>
<td>$Ca_{t,c}$</td>
<td>Critical capillary number for coalescence</td>
<td>m</td>
</tr>
<tr>
<td>$D$</td>
<td>Deformation parameter, $D=(L-B)/(L+B)$</td>
<td>[-]</td>
</tr>
<tr>
<td>$D'$</td>
<td>Deformation parameter calculated from $L'$ and $B'$</td>
<td>[-]</td>
</tr>
<tr>
<td>$E$</td>
<td>Dimensionless strain rate, $E=\gamma \mu R_0/2\sigma$</td>
<td>[-]</td>
</tr>
<tr>
<td>$e_f$</td>
<td>Stretching efficiency</td>
<td>[-]</td>
</tr>
<tr>
<td>$F$</td>
<td>Driving force for film drainage</td>
<td>N</td>
</tr>
<tr>
<td>$f_{\text{max}}$</td>
<td>Maximum drop collision frequency (number of collision per 40ms)</td>
<td>times/40ms</td>
</tr>
<tr>
<td>$G$</td>
<td>Velocity gradient</td>
<td>m/s²</td>
</tr>
<tr>
<td>$G$</td>
<td>$Ca^{2/3}$</td>
<td>[-]</td>
</tr>
<tr>
<td>$g$</td>
<td>Standard acceleration of gravity, 9.807 m/s²</td>
<td>m/s²</td>
</tr>
<tr>
<td>$H$</td>
<td>Hamaker constant</td>
<td>J</td>
</tr>
<tr>
<td>$h$</td>
<td>Thickness of liquid film between two colliding drops</td>
<td>m</td>
</tr>
<tr>
<td>$h_{\text{crit}}$</td>
<td>Critical film thickness for coalescence</td>
<td>m</td>
</tr>
<tr>
<td>$h_o$</td>
<td>Initial film thickness between two colliding drops</td>
<td>m</td>
</tr>
<tr>
<td>$k$</td>
<td>Boltzman's constant, 1.38x10⁻²³ J/K</td>
<td>J/K</td>
</tr>
<tr>
<td>$k$</td>
<td>Dimensionless wavelength, $k=\omega/2R$</td>
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<tr>
<td>$L$</td>
<td>Length of deformed drop</td>
<td>m</td>
</tr>
<tr>
<td>$L'$</td>
<td>Drop length observed in velocity gradient direction, $L'=L\sin\theta$</td>
<td>m</td>
</tr>
<tr>
<td>$l$</td>
<td>Half-length of the thread, $l=L/2$</td>
<td>m</td>
</tr>
<tr>
<td>$\rho$</td>
<td>Polydispersity (standard deviation/mean)</td>
<td>%</td>
</tr>
<tr>
<td>$P$</td>
<td>Pressure</td>
<td>Pa</td>
</tr>
<tr>
<td>-----------</td>
<td>-------------------------------</td>
<td>---------------</td>
</tr>
<tr>
<td>$P_A$</td>
<td>Pressure of Fluid A</td>
<td>Pa</td>
</tr>
<tr>
<td>$P_B$</td>
<td>Pressure of Fluid B</td>
<td>Pa</td>
</tr>
<tr>
<td>$P_i$</td>
<td>Inner phase pressure</td>
<td>Pa</td>
</tr>
<tr>
<td>$q$</td>
<td>Growth rate of disturbances</td>
<td>s$^{-1}$</td>
</tr>
<tr>
<td>$R$</td>
<td>Thread radius</td>
<td>m</td>
</tr>
<tr>
<td>$R_o$</td>
<td>Initial thread radius</td>
<td>m</td>
</tr>
<tr>
<td>$R_b$</td>
<td>Thread radius at breakup</td>
<td>m</td>
</tr>
<tr>
<td>$r$</td>
<td>Radius of liquid film between two colliding drops</td>
<td>m</td>
</tr>
<tr>
<td>$r$</td>
<td>Radius of a pendant drop at position $z$</td>
<td>m</td>
</tr>
<tr>
<td>$r_1$ &amp; $r_2$</td>
<td>Principal radii of a pendant drop</td>
<td>m</td>
</tr>
<tr>
<td>$r_m$</td>
<td>Maximum radius of a pendant drop</td>
<td>m</td>
</tr>
<tr>
<td>$T$</td>
<td>Temperature</td>
<td>Degree</td>
</tr>
<tr>
<td>$Th$</td>
<td>Thread number</td>
<td>[-]</td>
</tr>
<tr>
<td>$t$</td>
<td>Time</td>
<td>s</td>
</tr>
<tr>
<td>$t_b$</td>
<td>Time to breakup (from onset of flow to the capillary instability just starts to grow)</td>
<td>s</td>
</tr>
<tr>
<td>$t_{drain}$</td>
<td>Drainage time</td>
<td>s</td>
</tr>
<tr>
<td>$t^*$</td>
<td>Grow time of disturbances from zero amplitude to breakup</td>
<td>s</td>
</tr>
<tr>
<td>$t_i$</td>
<td>Contact time of colliding drops</td>
<td>s</td>
</tr>
<tr>
<td>$u$</td>
<td>Velocity</td>
<td>m/s</td>
</tr>
<tr>
<td>$u^*$</td>
<td>Velocity at infinity distance</td>
<td>m/s</td>
</tr>
<tr>
<td>$u_i$</td>
<td>Velocity of inner fluid</td>
<td>m/s</td>
</tr>
<tr>
<td>$u_x$</td>
<td>Velocity in $x$ direction</td>
<td>m/s</td>
</tr>
<tr>
<td>$u_y$</td>
<td>Velocity in $y$ direction</td>
<td>m/s</td>
</tr>
<tr>
<td>$V$</td>
<td>Volume</td>
<td>m$^3$</td>
</tr>
<tr>
<td>$V_m$</td>
<td>Volume of liquid cylinder corresponding to one wavelength long</td>
<td>m$^3$</td>
</tr>
<tr>
<td>$V_f$</td>
<td>Volume of final drop</td>
<td>m$^3$</td>
</tr>
<tr>
<td>$W$</td>
<td>Width of deformed drop along the vorticity direction</td>
<td>m</td>
</tr>
<tr>
<td>$x$</td>
<td>Coordinates along the flow direction</td>
<td>m</td>
</tr>
<tr>
<td>$x$</td>
<td>Wavenumber, $x=2\pi R/\omega$</td>
<td>[-]</td>
</tr>
<tr>
<td>$x_m$</td>
<td>Wavenumber of fastest growing disturbance</td>
<td>[-]</td>
</tr>
<tr>
<td>$x_{opt}$</td>
<td>Wavenumber of dominant disturbance</td>
<td>[-]</td>
</tr>
<tr>
<td>$y$</td>
<td>Coordinates along the velocity gradient direction</td>
<td>m</td>
</tr>
<tr>
<td>$z$</td>
<td>Coordinates along the vorticity direction</td>
<td>m</td>
</tr>
<tr>
<td>$z$</td>
<td>Vertical position</td>
<td>m</td>
</tr>
<tr>
<td>$z_{ma}$</td>
<td>Vertical position of maximum radius of a pendant drop</td>
<td>m</td>
</tr>
<tr>
<td>Parameter</td>
<td>Description</td>
<td>SI Unit</td>
</tr>
<tr>
<td>-----------</td>
<td>-------------</td>
<td>---------</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>Parameter to characterize flow type, $\alpha=0$ corresponds to simple shear flow</td>
<td>[-]</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>Amplitude of disturbances</td>
<td>m</td>
</tr>
<tr>
<td>$\alpha_0$</td>
<td>Initial amplitude of disturbances</td>
<td>m</td>
</tr>
<tr>
<td>$\alpha_m$</td>
<td>Minimum amplitude of disturbances</td>
<td>m</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>Strain, $\gamma = \dot{\gamma} \times t$</td>
<td>[-]</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>Viscosity ratio</td>
<td>[-]</td>
</tr>
<tr>
<td>$\mu$</td>
<td>Outer phase viscosity</td>
<td>Pa.s</td>
</tr>
<tr>
<td>$\mu_i$</td>
<td>Inner phase viscosity</td>
<td>Pa.s</td>
</tr>
<tr>
<td>$\theta$</td>
<td>Drop orientation angle, angle between the principal axis of the drop and the velocity gradient direction</td>
<td>Angular degree</td>
</tr>
<tr>
<td>$\theta_0$</td>
<td>Initial drop orientation angle</td>
<td>Angular degree</td>
</tr>
<tr>
<td>$\rho_A$</td>
<td>Density of fluid A</td>
<td>kg/m$^3$</td>
</tr>
<tr>
<td>$\rho_B$</td>
<td>Density of fluid B</td>
<td>kg/m$^3$</td>
</tr>
<tr>
<td>$\sigma$</td>
<td>Interfacial tension</td>
<td>N/m</td>
</tr>
<tr>
<td>$\sigma_{\omega}$</td>
<td>Standard deviation of $\omega$</td>
<td>m</td>
</tr>
<tr>
<td>$\sigma_{\alpha f}$</td>
<td>Standard deviation of $\alpha_f$</td>
<td>m</td>
</tr>
<tr>
<td>$\sigma_{R_b}$</td>
<td>Standard deviation of $R_b$</td>
<td>m</td>
</tr>
<tr>
<td>$\omega$</td>
<td>Wavelength of disturbances</td>
<td>m</td>
</tr>
<tr>
<td>$\Gamma$</td>
<td>Velocity gradient tensor</td>
<td>s$^{-1}$</td>
</tr>
<tr>
<td>$\Omega$</td>
<td>Dimensionless growth rate of disturbances</td>
<td>[-]</td>
</tr>
<tr>
<td>$\Omega_m$</td>
<td>Fastest growth rate at a fixed $\lambda$</td>
<td>[-]</td>
</tr>
<tr>
<td>$\dot{\gamma}$</td>
<td>Shear rate</td>
<td>s$^{-1}$</td>
</tr>
<tr>
<td>$\dot{\epsilon}$</td>
<td>Stretching rate</td>
<td>s$^{-1}$</td>
</tr>
<tr>
<td>$\dot{\gamma}_{c.c}$</td>
<td>Critical shear rate above which coalescence and breakup may coexist</td>
<td>s$^{-1}$</td>
</tr>
<tr>
<td>$\Delta V_x$</td>
<td>Relative velocity of two colliding drops in x direction</td>
<td>m/s</td>
</tr>
<tr>
<td>$\Delta y$</td>
<td>Distance between two drops in y direction</td>
<td>m</td>
</tr>
<tr>
<td>$\Delta y_f$</td>
<td>Final distance between two drops in y direction</td>
<td>m</td>
</tr>
<tr>
<td>$\Delta y_0$</td>
<td>Initial distance between two drops in y direction</td>
<td>m</td>
</tr>
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</table>
CHAPTER 1 INTRODUCTION

Emulsions are stable dispersions of one liquid in another immiscible liquid. They are ubiquitous in the agriculture, food, medicine, cosmetics and paints industries. Drop size control is crucial in controlling the physical properties of emulsions, such as their viscosity, stability, turbidity, transport properties, etc. (Becher, 1965; Mason, 1999). For example, a perfluorocarbon emulsion is used as a red blood cell substitute, where only droplets with a specific size are well tolerated. In addition, the side effects decrease dramatically as the drop size distribution becomes narrower. To manufacture a monodisperse emulsion with a targeted drop size presents a great challenge (Spahn and Pasch, 2001).

Emulsions are usually formed by dispersing drops of one liquid in another in a mixer, which results in polydisperse emulsions. The average drop size and size distribution of emulsions are controlled by drop breakup and coalescence under shear. Drop breakup in industrial emulsification process is extremely complicated because of the complex flow, the multitude of fluid properties, and the interactions between drops of different sizes. An empirical approach that is often taken involves phenomenologically correlating the drop size and distribution with the processing parameters and the physical properties of the emulsions for a specific mixer. However, it is difficult to transpose the results to other emulsion systems or to other mixers.

Fundamental studies of drop deformation and breakup were pioneered by Taylor (1932, 1934, & 1964), who investigated the deformation and breakup of a single Newtonian drop in a Newtonian matrix in well-defined flow fields, such as simple shear flow and plane hyperbolic flow. Most fundamental studies thereafter followed Taylor's approach, and many
valuable results have been obtained such as the steady drop shape at small deformation (Rumscheidt and Mason, 1961; Guido and Villone, 1998), the critical conditions for breakup (Grace 1982; Bentley and Leal, 1986), breakup of threads in a quiescent matrix (Tomotika, 1935; Rumscheidt and Mason, 1962; Stone et al., 1986 & 1989), and quasi-equilibrium breakup (Torza and Mason, 1972). However, a vital piece of puzzle determining the final drop size and distribution is still missing, because not much is understood about how drops break up under flow (Stone 1994; Janssen, 1997).

The goal of this thesis is to experimentally investigate the transient breakup of drops in Newtonian emulsions in a simple shear flow. The approach taken here is to visually follow the deformation and breakup of drops in simple shear flow using high-speed photography. We focus on the following questions:

What are the mechanisms by which a drop breaks up in shear flow? How does the drop breakup mechanism affect the drop size and distribution? What can this teach us about making monodisperse emulsions?

Our work provides the first experimental verification of existing theoretical analysis on drop breakup in shear flow (Hinch and Acrivos, 1980; Khakhar and Ottino, 1987).

We will review the current understanding of this subject in Chapter 2, followed by a description of our experimental techniques in Chapter 3. We report our experimental results in two subsequent chapters: Chapter 4 focuses on drop breakup mechanisms, while Chapter 5 focuses on the final drop size and distribution. Conclusions and future work are presented in Chapter 6.
Chapter 2 Review of Drop Deformation, Breakup, and Coalescence in Simple Shear Flow

This chapter focuses on reviewing the studies of the deformation and breakup of a single drop in Newtonian systems in simple shear flow since Taylor’s pioneering work in the 1930s (1932, 1934). Acrivos (1983), Rallison (1984), Stone (1994), Janssen (1997), and Ottino et al. (2000) have all done major reviews of this topic. In section 2.8, we will also give a brief review about drop coalescence. This thesis is not an extensive investigation of drop coalescence by any means. However, in some cases at $\lambda<0.1$ (as we shall see in Chapter 4) we observe the daughter drops from the capillary instability may collide with each other and coalesce. Thus, we discuss some issues in coalescence that are directly related to our experimental observations. Recently, Chesters (1991) reviewed the existing theories on flow-driven drop collision and coalescence, and subsequent reviews by Janssen (1997) and Ottino et al. (2000) extending Chesters’ work have appeared. The reader can refer to these papers for more details.

2.1 Problem Description

Figure 2-1 shows the typical deformation and breakup of a drop in a shear flow. An initially spherical drop of radius $a$ and viscosity $\mu_i$, is suspended in another immiscible liquid of viscosity $\mu$. Under shear, the drop is deformed into an ellipsoid. When the shear rate is below a critical value, the drop is able to maintain a steady shape. As the shear rate increases, the steady shape of the drop becomes increasingly elongated. When the shear rate exceeds a critical value, the drop is unable to maintain a steady shape, and undergoes a transient
deformation process which ends with it breaking up into smaller drops.

In most cases, the drop size is sufficiently small, so the Reynolds number of the drop is small and the inertia is negligible with respect to the viscous shear stress. Furthermore, the density of the two liquids is usually comparable so that the buoyancy effects can be neglected. The fluid motion is then governed by the Stokes equation and the continuity equation (Stone, 1994),

\[ \nabla \cdot u = 0 \]  
\[ \nabla \cdot u_i = 0 \]  
\[ \nabla^2 u = \nabla P \]  
\[ \nabla^2 u_i = \nabla P_i \]

where \( u \) is the velocity, \( P \) is the dynamic pressure of the outer phase, and the subscript \( i \) denotes inner phase properties.

The deformation and breakup of drops depends only on the capillary number (\( Ca \)), the viscosity ratio (\( \lambda \)) and the type of flow. The viscosity ratio is the ratio of the viscosity of the drop to that of the matrix. The capillary number characterizes the ratio of the viscous shear stress of the external fluid (\( \mu \dot{\gamma} \)) to the capillary pressure (\( \sigma/\alpha \)),
\[ Ca = \frac{\mu \dot{\gamma} a}{\sigma} \]  

(2-5)

where \( \dot{\gamma} \) is the shear rate, and \( \sigma \) is the interfacial tension. The viscous stress tends to deform the drop, while the capillary pressure resists the deformation. When the capillary number is low, the capillary pressure is dominant, so that the drop remains nearly spherical. When \( Ca \) is high enough, the shear stress is able to overcome the capillary pressure and leads to the breakup of the drop. The critical value at which the drop is unable to maintain a steady shape is called the critical capillary number, \( Ca_c \).

![Diagram of flow fields](image)

(a) Simple shear flow  
(b) Plane Hyperbolic flow

Figure 2-2 Flow fields of simple shear flow and plane hyperbolic flow and drop deformation in the two flows (after Hinch and Acivos, 1980)

The flow types that have been most often studied are plane hyperbolic flow and simple shear flow. The velocity of a simple shear flow is

\[ u_x = G y \]  

(2-6)

\[ u_y = 0 \]  

(2-7)

where \( u_x \) and \( u_y \) are the velocity components along the \( x \) and \( y \) direction and \( G \) is the velocity gradient. Figure 2-2(a) shows the flow field of a simple shear flow and the typical drop deformation therein. Simple shear flow consists of equal amounts of vorticity and rate of
strain, and the direction of maximum elongation is oriented along $45^\circ$ toward the flow direction. As a result of the vorticity, the drop rotates as it elongates in simple shear flow. Plane hyperbolic flow, or two-dimensional extensional flow (see Figure 2-2 (b)) is

$$u_x = G x$$  \hspace{1cm} (2-8)

$$u_y = -G y$$  \hspace{1cm} (2-9)

which is a pure straining motion with no vorticity. In plane hyperbolic flow, a drop elongates along the $x$ direction, and thins in the $y$ direction, without rotating.

Both flows were first studied by Taylor (1934), who invented a four-roll mill to generate a plane hyperbolic flow and a parallel band apparatus to generate a simple shear flow. The four-roll mill has been extensively used by other researchers, e.g. Rumscheidt and Mason (1961) and Grace (1971), to study hyperbolic flow. Other apparatuses including: a Couette cylinder device (Torza et al., 1972; Grace, 1971), a cone-and-plate fixture (Tsakalos, 1998) and a linear shear cell (Guido and Villone, 1998; Zhao & Goveas, 2001) have been used to generate simple shear flow. Bentley and Leal (1986a and 1986b) have utilized a computer controlled four-roll mill to study drop deformation and breakup in a generalized two-dimensional linear flow,

$$\vec{u}(x) = \Gamma x$$  \hspace{1cm} (2-10)

where

$$\Gamma = \frac{\dot{\gamma}}{2} \begin{pmatrix} 1+\alpha & 1-\alpha & 0 \\ 1-\alpha & -1-\alpha & 0 \\ 0 & 0 & 0 \end{pmatrix}$$  \hspace{1cm} (2-11)

The ratio of vorticity to strain rate in these flows is given by $(1-\alpha) / (1+\alpha)$. $\alpha = 0$ corresponds to simple shear flow, while $\alpha = 1$ corresponds to plane hyperbolic flow. Bentley and Leal's apparatus can generate a flow with $\alpha = 0.2 \text{ to } 1.0$. 
2.2 Equilibrium/Quasi-Equilibrium Drop Deformation ($Ca < Ca_c$, $Ca \sim Ca_c$)

Most early experimental works about drop deformation and breakup (Taylor, 1934; Bartok and Mason, 1959; Rumscheidt and Mason, 1961; Torza et al., 1972; Grace, 1971) have focused on the steady shape of the drop at small deformation and the critical conditions ($Ca_c$) for breakup.

2.2.1 Steady drop shape ($Ca < Ca_c$)

The steady ellipsoidal shape of a drop in simple shear flow is shown in Figure 2-3. The three axes of the drop are $L$, $B$, and $W$. The angle between the velocity gradient direction ($y$ direction) and the principal drop axis $L$ is denoted as $\theta$.

![Figure 2-3 Steady ellipsoidal shape of a drop in simple shear flow (modified from Guido and Villone, 1998)](image)

Taylor (1932, 1934) developed a small deformation theory for slightly deformed drops ($Ca \ll 1$) by solving the Stokes equations inside and outside the drop, subject to the following boundary conditions,

1. continuity of velocity across the interface,
2. continuity of the shear stress across the interface,
3. equality of the normal stress difference across the interface and the interfacial pressure.
The theory predicted the steady state of a slightly deformed drop as a function of $Ca$ and $\lambda$,

$$D = Ca \frac{19\lambda + 16}{16\lambda + 16}$$  \hspace{1cm} (2-12)

where $D$ is a deformation parameter defined as

$$D = \frac{L - B}{L + B}$$  \hspace{1cm} (2-13)

$L$ and $B$ are the axes of the drop defined in Figure 2-3. The value of $(19\lambda + 16) / (16\lambda + 16)$ varies from 1.0 to 1.187. Therefore, viscosity ratio has a minor effect on drop deformation when the drop is nearly spherical.

Experimental results (Taylor, 1934; Rumscheidt & Mason 1961; Torza, et al., 1972; Grace, 1971; Guido and Villone, 1998) and numerical calculations (Rallison 1981) showed excellent agreement with the small deformation theory, up to surprisingly high Capillary number. Recently, Guido and Villone (1998) studied the three-dimensional deformation of drops in simple shear flow by using two linear parallel-plate shear cells, one for observation along the vorticity direction ($z$ direction) and the other along the velocity gradient direction ($y$ direction). Each shear cell consists of two glass plates, one of which is movable, and the other is stationary. Their results of the deformation parameter as a function of $Ca$ (see Figure 2-4) agree with Taylor's small deformation theory (Equation 2-12) up to $Ca$=0.3-0.4 for viscosity ratio range from 1.3-2.1.

However, a detailed examination of the drop shape revealed discrepancies with the theory even at lower $Ca$. Theoretically, the drop width along the vorticity direction ($z$ direction) $W$ does not change under shear, because simple shear flow is a two-dimensional flow. Nonetheless, Guido and Villone's 3-D results (1998) (see Figure 2-5) showed that $W$ (solid and open circles) decreases as $Ca$ increases. The deviation from Taylor's theory (the
Figure 2-4 The deformation parameter $D$ as a function of $Ca$ (Guido and Villone, 1998). The solid line corresponds to Taylor's small deformation theory (Equation 2-12) for $\lambda=1.4$.

Figure 2-5 The three axes of a deformed drop as a function of $Ca$ at $\lambda$ around 1.4 (open symbols) and 2.0 (full symbols) (Guido Villone, 1998). The axes are nondimensionalized with respect to the initial drop diameter. $L$, $B$, and $W$ are defined in Figure 2-3. As $Ca$ increases, $B$ and $W$ approach each other, that means the cross-section of the drop becomes circular.

The horizontal line in Figure 2-5 is already apparent at $Ca$ as low as 0.1. The axes $B$ (marked by diamonds) and $W$ (marked by circles) become almost identical as $Ca$ increases, or in another words, the cross-section of the drop (in the $B$-$W$ plane) becomes circular at high $Ca$. One
possible reason is that the interfacial tension becomes increasingly important as \( Ca \) increases, which tends to pull the cross section of the drop into a circular shape. Numerical simulations (Kennedy et al., 1994; Uijtewaal and Nijhof, 1995) of the three-dimensional shape agrees with Guido and Villone’s experimental results (1998) up to \( Ca \) around 0.3, but the simulations were not able to predict the cross section becoming circular at higher \( Ca \), because they do not work well for highly non-ellipsoidal shapes.

2.2.2 Orientation of the drop

Taylor’s small deformation theory (1934) predicts that in simple shear flow the principal axis of a slightly deformed drop lies in the direction of maximum elongation, \( i.e. \) \( \theta=45^\circ \) (\( \theta \) is defined in Figure 2-3). Experiments (Taylor, 1934) agree for very small \( Ca \), but the drop rotates toward the flow direction as \( Ca \) increases. Cerf (1951) improved Taylor’s linear theory by taking into account the second order terms of the stress tensor, and derived \( \theta \) as a function of \( \lambda \) and \( Ca \),

\[
\theta = \frac{\pi}{4} + (1 + \frac{2\lambda}{5})Ca
\]

Chaffey and Brenner (1967) derived a similar formula

\[
\theta = \frac{\pi}{4} + \frac{(19\lambda + 16)(2\lambda + 3)}{80(1 + \lambda)} Ca
\]

The two equations are essentially the same when corrected for a small error (Roscoe, 1967).

Figure 2-6 compares Guido’s experimental results (1998) with Equation 2-15. The agreement is good up to \( Ca=0.4 \) for \( \lambda=1.4 \) and \( Ca\sim0.2 \) for \( \lambda=2 \). This indicates that the principal axis of the drop orients along the direction of maximum elongation (\( \theta=45^\circ \)) at \( Ca\rightarrow0 \), and it rotates toward the flow direction (\( \theta=90^\circ \)) as it elongates under increasing \( Ca \).
Both Equation 2-14 and 2-15 are invalid at high \( Ca \), because in both cases \( \theta \) increases monotonically as \( Ca \) increases, but they become unrealistic when \( \theta > 90^\circ \).

\[
\theta = \frac{\pi}{4} + \frac{1}{2} \tan^{-1} \left( \frac{19Ca\lambda}{20} \right) \quad (2-16)
\]

which captures the fact that in simple shear flow, as \( Ca \) increases, \( \theta \) approaches \( 90^\circ \). This agrees with their experimental results at high \( \lambda \), e.g. \( \lambda = 3.6 \), but significantly underestimates \( \theta \) for \( \lambda < 1 \) at low \( Ca \).

2.2.3 Critical capillary number, \( Ca_c \)

Torza et al. (1972) and Grace (1971) measured the critical capillary number, at a variety of viscosity ratios, by gradually increasing the shear rate up to a critical value where
the drop can no longer hold a steady shape, and thus breaks. Grace's results for simple shear flow are shown as the dashed line in Figure 2-7. Here, $Ca_c$ exhibits a minimum at $\lambda$ between 0.1 and 1. It is more difficult to break a drop with either low $\lambda$ or high $\lambda$. Note that $Ca_c$ goes to infinity at $\lambda=4$. It indicates the impossibility to break a drop with $\lambda>4$ in simple shear flow, which was first found by Taylor (1934). The above results of $Ca_c$ were obtained under quasi-equilibrium condition. Drop breakup under this condition is sometimes referred to as quasi-equilibrium breakup (a detailed discussion is included in Section 6.1). A drop may break up at a sub-critical capillary number if the shear rate undergoes a sudden increase (Torza et al., 1972; Hinch and Acrivos, 1980).

![Graph](image)

Figure 2-7 Variation of $Ca_c$ with $\lambda$ for simple shear flow (Adapted from Rallison, 1984). --- Experimental results from Grace (1971) (horizontal burst due to tip streaming); — Hinch and Acrivos' slender body theory for $\lambda\to0$ (1980); O second order theory of Barthes-Biesel & Acrivos (1973).

Taylor's small deformation theory (1932), being linear, was unable to predict the critical conditions for breakup. Barthes-Biesel and Acrivos' (1973) extended the theory by describing the drop shape in terms of second order harmonics, and made reasonable predictions of the critical shear rate (see the circles in Figure 2-7).
Taylor (1964) proposed another asymptotic theory, known as slender body theory, to account for large deformation at $\lambda \to 0$ in axisymmetric extensional flow. The theory was refined by Buckmaster (1972, 1973) and Acrivos and Lo (1978) for the same flow, and further extended by Hinch and Acrivos to plane hyperbolic flow (1979) and simple shear flow (1980). The idea is to solve the Stokes equations for noninertial flows by representing the influence of the drop as a distribution of singularities along the centerline of the drop. The theory predicts $Ca_c = 0.0541 \lambda^{2/3}$, which is in fair agreement with experiments, as shown by the solid line in Figure 2-7.

### 2.3 Transient (Pseudo-affine) Deformation ($Ca \gg Ca_c$)

The drop deformation considered in the above section is at equilibrium or quasi-equilibrium conditions, when $Ca$ is below or just above $Ca_c$. In this section, we will discuss the transient deformation of drops as a function of time (strain) for $Ca \gg Ca_c$. When $Ca \gg Ca_c$, the capillary pressure is much smaller than the shear stress. Thus, the viscous stress of the matrix acts on the drop as if the interface were not present. This is similar to affine deformation, where the drop deforms just like a sphere of the matrix phase deforming in the matrix itself.

Janssen (1997) calculated the drop shape under affine deformation as a function of the applied strain, for simple shear flow

$$\frac{L}{2a} = \frac{1}{2} \gamma + \frac{1}{2} \sqrt{4 + \gamma^2}$$  \hspace{1cm} (2-17)

$$\frac{B}{2a} = \frac{1}{2} \gamma + \frac{1}{2} \sqrt{4 + \gamma^2}$$  \hspace{1cm} (2-18)
\[
\tan \theta = \left( \frac{1}{2} \gamma + \frac{1}{2} \sqrt{4 + \gamma^2} \right)^{-1}
\]

(2-19)

where \( \gamma \) is the applied strain, \( i.e., \) the product of shear rate and time. Initially, \( \theta=45^\circ \), so that the drop orients toward the direction of maximum extension. As it elongates, it rotates toward the flow direction (\( \theta=90^\circ \)), and thus feels less stretching force. If the drop fully aligns with the flow direction, it will neither elongate nor contract.

Equations 2-17 and 2-18 imply that the drop radius along the vorticity direction does not change, and the cross section is noncircular, which is a 2-D feature of simple shear flow. However, as we previously mentioned, Guido and Villone's 3-D quantitative results (1998) showed that drop radius along the vorticity direction also decreases with \( Ca \). Furthermore, as the drop further elongates and the width continues to thin, the local capillary pressure becomes increasingly dominant and promotes a circular cross-section (Guido and Villone, 1998). Thus, the thinning of the drop width deviates from purely affine deformation at high deformations.

Allowing for both a circular cross section and volume conservation, Janssen (1997) found that the form of drop width thinning is given by

\[
\frac{B}{2a} = \left( \frac{1}{2} \gamma + \frac{1}{2} \sqrt{4 + \gamma^2} \right)^{-1/2}
\]

(2-20)

The equations above imply that the drop width thins monotonically with time (\( \gamma \)), and thus gives no prediction of drop breakup. When \( \gamma > 5 \), Equation 2-17, 2-19 and 2-20 can be simplified as

\[
\frac{L}{2a} \approx \gamma
\]

(2-21)

\[
\frac{B}{2a} \approx \gamma^{-1/2}
\]

(2-22)
\[ \theta = \tan^{-1} \gamma \quad \text{(2-23)} \]

Elemans et al. (1993) studied the time-dependent deformation of Newtonian drops in a simple shear flow generated by a Couette flow cell consisting of two counter-rotating concentric cylinders. By plotting the deformation parameter against the strain, he found that drop deforms affinely when \( Ca > 2Ca_c \) for a Newtonian system of \( \lambda = 0.135 \) (see Figure 2-8).

![Figure 2-8](image)

Figure 2-8 Deformation parameter \( D \) as a function of the strain shows that drop deforms affinely when \( Ca > 2Ca_c \) for a Newtonian system of \( \lambda = 0.135 \). The solid line corresponds to the pseudo-affine deformation (Elemans, 1989).

2.4 Quiescent Drop Breakup

If a drop is first stretched into a long liquid thread by an external flow, which is then stopped, the thread will break into smaller droplets in a quiescent matrix. Two different mechanisms, capillary instability and end pinching, may cause the drop to break. End pinching is the dominant mechanism on modestly stretched threads, while the capillary instability is the dominant breakup mode on highly stretched threads.

2.4.1 Capillary instability

This phenomenon was first observed by Taylor (1934) after flow cessation. An example is shown in Figure 2-9 (Rumscheidt and Mason, 1962). After the flow is stopped,
disturbances grow on the surface of the elongated drop like waves and finally cause the thread to break into smaller drops. The wavelength is uniform along the length of the thread, leading to uniformly distributed monodisperse daughter drops. There are even smaller satellite and sub-satellite drops between the daughter drops, which are not visible in the picture due to the poor reproduction. The thread breaks almost simultaneously at all points along the thread.

![Image](image_url)

Figure 2-9 The growth of capillary instability leading to the breakup of a stationary liquid thread at $\lambda=1.1$ (Rumscheidt and Mason, 1962). Note the uniform wavelengths and uniformly distributed monodisperse daughter drops.

Tomotika (1935) theoretically studied the instability on an infinitely long stationary viscous liquid thread surrounded by another viscous fluid. He assumed that the amplitude of disturbances ($\alpha$) grows exponentially with time,

$$\alpha = \alpha_0 \exp(\alpha t)$$  \hspace{1cm} (2-24)

where $\alpha_0$ is the initial disturbance amplitude. The growth rate of disturbances, $q$, is

$$q = \frac{\sigma \Omega(x,\lambda)}{2 \mu R_o}$$  \hspace{1cm} (2-25)
where $R_o$ is the initial thread radius, $\Omega$ is the dimensionless growth rate, which is a function of the viscosity ratio $\lambda$ and the dimensionless wavenumber,

$$x = \frac{2\pi R_o}{\omega}$$  \hspace{1cm} (2-26)

where $\omega$ is the wavelength of the disturbance.

![Graph showing $\Omega_m$ and $x_m$ vs $\lambda$]

Figure 2.10 The wavenumber $x_m$ and growth rate $\Omega_m$ of the fastest growing disturbances on an infinitely long stationary liquid thread (Tomotika, 1935; Janssen and Meijer, 1995)

Tomotika found that there is a non-zero fastest growing wavelength at any given $\lambda$, and assumed that the thread breaks up by this fastest growing disturbance. The dominant wavenumber $x_m$ and the corresponding dimensionless growth rate $\Omega_m$ are shown in Figure 2.10. The growth rate decreases as the viscosity ratio increases, and the wavenumber has a maximum at $\lambda$ around 0.3. Breakup is assumed when the amplitude of the disturbance grows to the size as of thread radius, thus the time required for thread breakup is

$$t_b = \frac{1}{q} \ln \left( \frac{R_o}{\alpha_o} \right)$$  \hspace{1cm} (2-27)

The initial amplitudes of disturbances caused by thermal fluctuation was estimated by Kuhn (1953)
\[ \alpha_0 = \left( \frac{21kT}{8\pi \gamma \sigma} \right)^{\frac{1}{2}} \]  

(2-28)

where \( k \) is the Boltzmann’s constant and \( T \) is absolute temperature. Typically, the initial amplitude is in the order of \( 10^{-9} \)m at room temperature. Assuming one wavelength forming one daughter drop and neglecting the volume of satellite drops, the drop radius can be calculated from volume conservation (Janssen 1997),

\[ a_d = R_0 \left( \frac{3\pi}{2\alpha_m} \right)^{\frac{1}{3}} \]  

(2-29)

Although Tomotika’s linear theory applies only when the amplitudes of the disturbances are small, experimental measurements of the dominant wavelength and \( \Omega_m \) agree with the theory (Rumscheidt and Mason, 1962; Janssen, 1997). Janssen also found that the growth time of the dominant wavelength \( t_b \) is in reasonable agreements with the theoretical prediction using Kuhn's estimation of \( \alpha_0 = 10^{-9} \)m.

The linear stability theory of Tomotika cannot predict the existence of the satellite and sub-satellite drops between the principal drops. Tjahjadi et al. (1992) have studied the formation of satellite drops of quiescent drop breakup by both experiments and boundary integral calculations. They found that satellite drops are formed due to the multiple breakup sequences around the neck regions of the capillary waves. The number of satellite drops and their relative sizes are governed by the viscosity ratio. For the range of viscosity ratios between 0.01 and 2.8, as the viscosity ratio increases, the number of satellite drops decreases. The size of largest satellite drop relative to the daughter drop increases due to the damping of the internal flow at high \( \lambda \). The time to the first breakup increases as \( \lambda \) increases.
2.4.2 End pinching

Stone et al. (1986, 1989a and 1989b) investigated the end pinching mechanism experimentally by using a computerized four-roll mill, and numerically via a boundary-integral method. Figure 2-11 shows typical end pinching at a variety of viscosity ratios (Stone et al., 1986). Once the flow is stopped after a drop has been significantly extended, the length of the elongated drop shrinks, the ends of the drop become rounded, and a neck forms between the bulbous end and the central thread. The neck continues to thin, and eventually causes a drop to be pinched off from each end of the thread. There are satellite and sub-satellite droplets between the pinched-off drop and the remaining thread. Similar processes may repeat on the remaining part of the thread.

Stone et al. (1986) described end pinching as interfacial tension-driven flow dominated by end effects. When the flow is stopped, the external viscous force exerted on the thread disappears, and the interfacial tension tends to make the threads return to a spherical

![Figure 2-11 End-pinching on modest long stationary liquid threads at a variety of viscosity ratios (Stone et al., 1986)]
shape. The capillary pressure varies with the curvature of the thread and has a maximum at the sharp ends of the threads, which generates a flow toward the center. Then the pressure gradient builds up in the transition region connecting the central cylindrical region with the bulbous end, and generates flow away from the center. The combined effects of these two types of flow cause a neck to form in the transition region. A pressure maximum develops at the neck, drives the fluid flow away from the neck, and makes it even thinner, which in turn increases the pressure gradient and finally causes the pinch off of the end. Once both ends pinch off, the same end-pinching process may repeat again and again, until the whole central part breaks up or relaxes into spherical drops.

![Diagram](image)

*Figure 2-12 Critical elongation ratio necessary to ensure breakup after the cessation of the flow vs. the viscosity ratio (Stone et al., 1986). The triangles denote the smallest \( L/2a \) for breakup (End-pinching) to occur, the squares denote the largest \( L/2a \) that a drop relaxed back to a sphere.*

End pinching only occurs when the elongation ratio \( (L/2a) \) prior to the cessation of the flow is above a critical value, which is a function of \( \lambda \) (see Figure 2-12). Otherwise, the elongated drop will relax back to a sphere. The capillary instability does not play a significant role at modest extensions of the drop. However, in cases (see Figure 2-13) when the drop is sufficiently stretched \( (L/2a \gtrsim O(20)) \), end pinching occurs at the ends of the thread first, and then capillary waves grow on the central part of the thread, and lead to its breakup.
into a line of small droplets. Stone and Leal (1989a) proposed that is because end pinching occurs on a faster time scale than that for the capillary instability. For the capillary instability to occur on the central part of the thread, the drop has to be sufficiently extended, initially, that the timescale for the complete breakup via multiple sequences of end pinching exceeds the timescale of capillary instability. Note that the daughter drops from the capillary instability are similar in size to the pinched-off drops except that the first principal drop pinched off at each end is much bigger (see Figure 2-13).

![Figure 2-13: End pinching and the capillary instability on sufficiently stretched liquid thread after the flow is stopped. Note that the daughter drops from the capillary instability are similar in size as the pinched-off drops except the first principal drop pinched off at each end is much bigger (Stone et al., 1986).]

The end pinching mechanism strongly depends on the viscosity ratio. Figure 2-12 indicates that it is difficult to break up drops with either large or small viscosity ratios via the end-pinching mechanism. Drops with a small viscosity ratio, e.g. \( \lambda < 0.01 \), can maintain highly elongated steady shapes, thus high capillary numbers are necessary to cause a transient elongation. For drops with \( \lambda > O(10) \), even highly stretched drops are able to relax back to a spherical shape owing to the significant resistance of the internal flow.
2.5 Drop Breakup in Flow: Theory and Simulation

2.5.1 Slender body theory

As mentioned in 2.2.2, Hinch and Acrivos (1980) have developed slender body theory for Newtonian drops suspended in Newtonian matrices at $\lambda \ll 1$ in simple shear flow. One consequence of this assumption of $\lambda \ll 1$ is that the drop is slender and nearly aligns with the flow. In addition, the shear stress exerted on the outer phase by the inner phase is negligible due to the low viscosity of the inner phase. The theoretical analysis of simple shear flow is much more complicated than for extensional flows, because the vorticity causes the drop to rotate in the flow. The orientation of the drop is then an additional unknown, which must be solved as part of the solution. Hinch and Acrivos (1980) made a simplifying assumption that the drop has a circular cross section. The assumption was made based on previous theoretical analysis for plane hyperbolic flow (Hinch and Acrivos, 1979) where they found that the cross section does not deviate greatly from circular shape, and the drop length at steady state and the critical capillary number were almost identical to those in axisymmetric flow (3-D extensional flow) where the drop has a circular cross section. They argued that noncircular cross section has little effect on the dynamics that determine the length of the drop. As mentioned previously, Guido (1998) experimentally observed that the cross section of the drop is non-circular at low shear rates, but it approaches circular shape as the shear rate increases. Therefore, the assumption of a circular cross section is justified at high shear rates (capillary numbers).

Based on the above assumptions, the evolution of the center line and the drop shape were calculated, and the following predictions were made for simple shear flow,
(1) At steady state, the angle of orientation between the long axis of the drop and the direction of flow is the same order of magnitude as the aspect ratio of the drop. This means that a highly deformed slender drop is almost aligned with the flow direction at steady state.

(2) The dimensionless length of the equilibrium drop \( \frac{L}{2\alpha \lambda^{1/3}} \) versus the dimensionless shear rate \( \tilde{G} = Ca \lambda^{2/3} \) is shown in Figure 2-14. There is a well-defined length corresponding to every shear rate, which means that a steady shape exists for all shear rates. However, a stability analysis shows that the equilibrium is unstable to small disturbances when \( \tilde{G} \) exceeds a critical value of 0.0541(point A in Figure 2-14). Thus, the critical capillary number is determined as \( Ca_c = 0.0541 \lambda^{2/3} \), as shown is Figure 2-7.

\[
\frac{L}{2\alpha \lambda^{1/3}} = 261G^2
\]  

(2-30)

(3) As \( G \to \infty \), the drop becomes a long thread with uniform radius except at the ends where it tapers. The length and the radius of the central uniform part are
\[
\frac{R}{a \lambda^{1/6}} = \frac{0.0505}{G}
\]

(2-31)

The central part aligns with the flow direction, while the center line of the ends slightly tilts. The end of the thread has a length of about 0.0231a\lambda^{1/3}G^{-1}.

(4) A stability analysis showed that the least stable mode of disturbance is on the scale of the length of the drop. It differs from the instability on infinitely long threads, where the least stable mode has a wavelength on the scale of the thread thickness (Tomotika, 1935).

(5) If the shear rate is increased suddenly instead of very slowly, the critical shear rate causing the drop to break decreases. This agrees with Torza et al.'s experiments (1972).

The steady shape of the drop and the wavelength of the least stable mode of disturbances have not been tested in experiments. We will provide the first experimental evidence in Chapters 4 and 5.

2.5.2 Capillary stability analysis for extending threads

Extensional flows

In section 2.4, we discussed Tomotika's capillary stability analysis for an indefinitely long liquid thread in a quiescent matrix. Tomotika (1936) further analyzed the stability of an infinitely long thread in an axisymmetric extensional flow. He found that the disturbances first initiated on the thread could only grow to limited amplitudes before being damped out by the flow. As the thread thins in the flow, the disturbances continue to be initiated until eventually they grow to the size of the thread radius, causing the thread to break. Tomotika (1936) assumed that the initial amplitude of disturbances (\(\alpha_0\)) is proportional to the thread radius, which varies with time. Mikami et al. (1975) improved the analysis for axisymmetric extensional flow by assuming that \(\alpha_0\) is independent of the thread radius, and of the
wavelength of the disturbances. They found that disturbances generally decay to a minimum amplitude, and then grow, and finally damp again. Following Tomotika (1936), they also assumed the disturbances are continually generated, and that thread breakup occurs when the amplitude of a disturbance grows to equal the radius of the thread. The model is capable of calculating the radius at breakup $R_b$, the time to breakup $t_b$, the final drop size $a_f$, and the wavenumber of dominant disturbances $x_{opt}$, given the initial thread radius $R_o$ and the initial amplitude of disturbances $\alpha_o$.

Khakhar and Ottino (1987) extended Mikami's analysis to a general linear flow (as described by Equation 2-10), by analyzing the problem with respect to a moving frame that rotates with the thread. Following Hinch and Acrivos' approach (1980), they assumed that the cross-section of the thread is circular. The Stokes equations for creeping flow inside and outside of the thread were solved together with the boundary conditions. It was found that the flow around the thread is an axisymmetric flow superimposed by a non-axisymmetric shear flow. Figure 2-15 shows a typical thread breakup process in a plane hyperbolic flow. Similar to what Mikami (1975) found in axisymmetric extensional flow, the disturbances first decay to a minimum amplitude, then grow to certain amplitude, before they are damped out by the flow (see Figure 2-15(a). With disturbances being continuously initiated on the thread surfaces, those that start later grow to larger amplitudes (see the series of solid curves in Figure 2-15(b)). At the same time, the thread radius (dashed line in the Figure 2-15 (b)) continues to thin as the thread is elongated by the flow. Eventually, the disturbances grow to amplitude equal in size to the thread radius, as manifested by the first intersection of the curve of amplitude and the dashed line of the thread radius. At this point, the thread breaks
up. The whole process is similar to what Tomotika (1936) and Mikami (1975) found in axisymmetric extensional flow.

\[ R_b \propto e_f^{-0.89} \lambda^{-0.44} \left( \sigma / \mu G \right)^{0.84-0.92} \quad (2-32) \]

where \( G \) is the velocity gradient of the flow. \( e_f \) is a stretching efficiency defined by Khakhar and Ottino (1987) and Tjahjadi and Ottino (1991) for linear flow. It is a constant for plane hyperbolic flow, while it changes with the orientation of the thread for simple shear flow. The above equation implies that the critical thread radius does not depend on the initial thread radius.

Under the same condition of constant stretching rate, Janssen and Meijer (1993) took Tjahjadi and Ottino’s (1991) approach and found that the critical thread width \( (R_b) \), the final
Figure 2.16 (a) Dimensionless critical thread radius, (b) Dimensionless daughter drop radius resulting from thread breakup, and (c) Growth time of the disturbance as a function of the dimensionless stretching rate. (Janssen and Meijer, 1993).
drop size \((a_f)\) and the growth time of disturbances from zero amplitude to breakup \((t_g^*)\) do not depend on the initial thread radius \(R_o\). Their dependence on the viscosity ratio and the dimensionless stretching rate \(\mu \dot{\varepsilon} \alpha / \sigma\) are shown in Figure 2-16. In general, the critical thread width and the final drop size decrease with the dimensionless stretching rate and the viscosity ratio. The product of the stretching rate and the growth time of the disturbance \((e_f t_g^*)\) is a weak function of the dimensionless stretching rate at a fixed viscosity ratio. They also predicted that the wavelength of the disturbance leading to the breakup is about the circumference of the thread \((2\pi R_b)\), and is independent of the viscosity ratio and the extension rate.

**Simple shear flow**

Simple shear flow is more complicated than the above extensional flows. In simple shear flow, the thread rotates toward the flow direction, thus the stretching rate decays with time. Therefore, the initial orientation of the thread must be known as well. For these reasons, Janssen and Meijer (1993) were not able to use Khakhar and Ottino's results (1987) for simple shear flow.

Khakhar and Ottino (1987) showed that in simple shear flow the time to breakup \((t_b)\) and the final drop radius \((a_f)\) are

\[
(1 + C t_b) \propto (CE)^{0.65}
\]

\[
\frac{a_f}{R_o} \propto (CE)^{-0.32}
\]

where \(C = 2\tan^{-1} \theta_o\) and \(\theta_o\) is the initial angle between the thread and the velocity gradient direction. \(E = \dot{\gamma} \mu R_o / 2 \sigma\) and \(R_o\) is the initial thread radius.
All of the stability analyses discussed above share the same inherent limitations:

(1) The analysis is for an infinitely long thread, thus no prediction is given to correlate with the initial spherical drop radius.

(2) The initial conditions such as the initial thread radius, the initial amplitude of disturbances as well as the initial thread orientation angle in flows with vorticity cannot be predicted by the theory itself. In practice, they are thus arbitrarily chosen.

By comparing with indirect experimental results, Mikami (1975) suggested that $\alpha_0$ is in a range of $10^{-8} - 10^{-7}$ m. Janssen and Meijer (1993) claimed that Kuhn's estimation of $10^{-9}$ m (Equation 2-28) was better. However, a physically unrealistic value of $10^{-10}$ m gave the best fit to experimental results. As suggested by Mikami (1975), the initial thread radius was usually arbitrarily chosen at a point where the drop is deformed into a long thread but without significant growth of disturbances on the thread surface. Janssen and Meijer (1993) arbitrarily chose $\theta_0 = 27^\circ$ corresponding to $C=1$ for simple shear flow.

Attempts have been made to compare theoretical results with experimental results for extensional flow (Mikami et al., 1975, Janssen and Meijer, 1993) and simple shear flow (Khakhar and Ottino, 1987). Overall, the comparisons are fair, however, as mentioned previously, the initial conditions are arbitrarily chosen.

2.6 Drop Breakup in Simple Shear Flow: Experiments

2.6.1 Drop breakup mechanism

While the steady shape of a slightly deformed drop and the critical conditions ($Ca_c$) for breakup (see Section 2.1) have been well studied, little is known about the actual fragmentation of the droplet, except in some special cases. One such example is thread
breakup in quiescent matrices as discussed in Section 2.4. Another example is quasi-equilibrium breakup. When the shear rate is slowly increased to just exceed the critical value, a neck forms at the center of the elongated drop and progressively thins to finally cause the drop to break into two daughter drops separated by satellite and sub-satellite drops (Bartok and Mason, 1959). Figure 2-17 shows the quasi-equilibrium breakup mode at a variety of viscosity ratios between $3 \times 10^{-3}$ and 3.0 (Torza et al., 1972).

![Figure 2-17 Quasi-equilibrium breakup in simple shear flow at a variety of viscosity ratio between 0.003 and 3.0 (Torza et al., 1972).](image)

A careful examination of the literature gives a vague picture about the transient drop breakup in supercritical shear ($Ca > Ca_c$) in simple shear flow. Taylor (1934) mentioned that a drop with $\lambda = 0.9$ is elongated into a thread, which then breaks up into smaller drops. Rumscheidt and Mason (1961) observed similar phenomena in both simple shear flow ($\lambda = 0.7 \sim 2.2$) and plane hyperbolic flow ($\lambda > 0.2$). However, in both papers, only the beginning state (a long thread) and the end state (a number of smaller drops) were mentioned, while the dynamic transitions between them were unknown. In addition, no quantative result about
the thread width and wavelength at breakup and the final drop size was available, nor were the effects of \( Ca \) and \( \lambda \) discussed.

Grace (1971) has done extensive studies of drop deformation and breakup in simple shear flow as well as plane hyperbolic flow. One of the most well known results is the dependence of \( Ca_c \) on \( \lambda \) shown is Figure 2-7. This is Grace's only published paper, and was edited long after his retirement according to the editorial note to the paper. Important results in the form of slides and movies were no longer available by the time it was published. There are no details about the actual drop breakup mechanism in shear flow in the paper.

Detailed observations of drop breakup were made in flows other than simple shear flow. For plane hyperbolic flow at \( \lambda = 0.148 \) and \( \lambda = 1.46 \), we quote from the paper of Mikami et al. (1975) “It was observed that breakup under extension of the liquid thread is characterized by the formation of large drops connected by thin liquid filaments, which on further extension develop a secondary varicosity and break up, leaving a myriad of satellite droplets between the larger principal drops.” The description implies breakup by the capillary instability. He further stated “Furthermore, in contrast to the breakup of a stationary liquid thread (Rumscheidt and Mason, 1962), breakup under extension does not occur simultaneously over the entire length of the thread, the spacing between principal drops is not constant and the filaments do not give rise to the same number of droplets. Thus the breakup products in this case lack uniformity in size.”

Tjahjadi and Ottino (1992) studied drop breakup in a chaotic flow produced by two periodically rotating eccentric cylinders. The drop undergoes stretching, folding and compression in the flow. They observed several drop breakup mechanisms including necking, end-pinching, capillary instability, and drop breakup due to folding of thread. The
capillary instability is the dominant mechanism for breakup of long threads, and the thread width at breakup differs from location to location due to the chaotic nature of the flow.

Tsakalos et al. (1998) recently studied the breakup of a viscoelastic drop in a Newtonian matrix (5x10^5 mPa.s polydimethylsiloxane (PDMS) or silicone oil) in simple shear flow with constant shear rate (0.08~1.6 s^{-1}), and observed drop breakup due to both end pinching and the capillary instability at $Ca >> Ca_c$ (see Figure 2-18). End pinching occurs earlier than the capillary instability, and the capillary instability is dominant on long threads. The capillary instability starts to develop at a constant thread diameter, which does not depend on the initial drop diameter. Furthermore, the critical diameter is inversely

Figure 2-18 Drop breakup by end pinching and the capillary instability in simple shear flow. The drop is viscoelastic, and the matrix is Newtonian with $\lambda$=0.70-0.85 (Tsakalos et al. 1998).
proportional to the shear rate. They found that the drop follows pseudo-affine deformation when $C_a > 2.5C_{ac}$ and for a strain greater than 20. Assuming pseudo-affine deformation, the time to breakup was estimated as

$$t_b = \frac{1}{\dot{\gamma}} \left( \frac{a}{R_b} \right)^2$$  \hspace{1cm} (2-35)

They estimated that the final drop size is half of the critical drop size corresponding to $Ca_c$, so that daughter drops will not break again in their emulsion system of $\lambda = 0.70-0.85$.

2.6.2 Final drop size and distribution

Grace (1971) found that the polydispersity of final drops widens as $Ca/Ca_c$ increases at $\lambda = 3.99 \times 10^3$. However, it is not specified what the drop breakup mechanism is, nor is the reason for the widening. He also found that the average final drop size to initial drop size first decreases and then tends to level out. (see Figure 2-19). However, the data raise questions. Taking the highlighted point in Figure 2-19 as an example, the data shows that the ratio of

![Figure 2-19 Grace's experimental data for effect of $Ca/Ca_c$ on the ratio of the final drop size to the original drop size (Grace 1971). $E = Ca(19\lambda+16)/(16\lambda+16)$, and $E_a$ is $E$ at breakup. Thus, for a certain emulsion system with a fixed $\lambda$, $E/E_a = Ca/Ca_c$.](image-url)
the final drop diameter to that of the original drop diameter is about 0.2, \( i.e., a_f \approx 0.2a \), at \( Ca/a_c = 10 \), \( i.e., a = 10a_c \). Thus, \( a_f = 2a_c \), which is unrealistic, because any drop larger than the critical drop size will be unstable and thus break again.

Bigio et al. (1998) studied the sizes of daughter drops from the breakup of single Newtonian drop in simple shear flow under a constant shear rate. Their emulsions cover a range of \( \lambda \) from 0.03 to 3. They found that the maximum daughter drop size is approximately the same as the maximum stable drop size at the corresponding shear rate, and the mean daughter drop size is about 70% of the maximum stable drop size.

### 2.7 Summary of Drop Deformation and Breakup in Simple Shear Flow

Since Taylor's pioneering work in the 1930s, fundamental research on drop deformation and breakup in flow has continued over the past 70 years. Many important issues such as the steady drop shape, the critical conditions for breakup, breakup of threads in quiescent matrix, and quasi-equilibrium breakup, have been well studied. However, relatively little is known about the dynamics of drop breakup. There is some theoretical analysis, such as slender body theory (Hinch and Acrivos, 1980) and stability analysis (Khakhar and Ottino, 1987), but few experimental results are available to verify them. The current state of fundamental researches about drop deformation and breakup was well stated by Janssen in a recent review (1997), “In contrast to thread breakup in a quiescent matrix and drop breakup under quasi-equilibrium conditions, the breakup of an extending thread has rarely been studied experimentally.”
2.8 Drop Collision and Coalescence in Flow

So far we have discussed the deformation and breakup of a single drop. In liquid-liquid systems consisting of multiple drops, drops may collide with each other and thus coalesce under the forces of the external flow. Drop coalescence is a complex problem, involving the interaction of two drops with each other and the external flow. For simplification, only the collision and coalescence of two equal sized drops are considered here. As in the analysis of drop deformation and breakup, inertia is neglected due to small drop sizes involved and therefore small Reynolds number. Buoyancy is also neglected because the density difference in most liquid-liquid systems is insignificant.

2.8.1 Sequential steps of flow-driven coalescence

Figure 2-20 taken from Ottino et al. (2000) shows schematically the collision and

![Figure 2-20 Schematic of the three basic steps of coalescence (Ottino, 2000)](image)
coalescence of two deformable drops driven by simple shear flow. The process generally consists of three sequential steps: (1) Two drops approach each other under the forces of the external shear flow and then collide; (2) The contact areas of the drops flatten, forming a liquid film between the drops, taken to be a flat circular film with radius \( r \) and thickness \( h \). The film continues to drain, while the two drops rotate together in the manner of a dumbbell. (3) The liquid film separating the drops ruptures. If no coalescence occurs during the contact and rotation process of the drops, the drops separate from each other and move apart.

Collision or close contact of drops is a necessary but not sufficient condition for coalescence. Coalescence only occurs when the liquid film is able to drain to a critical thickness \( h_{crit} \), when van der Waals forces become dominant and cause the rupture of the film. Otherwise, the two drops will move away from each other. The critical film thickness is estimated as (Chesters, 1991)

\[
h_{crit} \sim \left( \frac{Ha}{8\pi\sigma} \right)^{1/3}
\]  

Where \( H \) is the Hamaker constant (typically in the order of \( 10^{20} \) J), \( a \) is the initial drop radius, \( \sigma \) is the interfacial tension. For a drop of \( a=10\mu m \), and \( \sigma=5\times10^{-3} \) N/m, \( h_{crit} \) is about \( 10^{-8} \) m or 0.01 \( \mu \)m. The time for film drainage to \( h_{crit} \) is considered to be the rate-limiting step; rupture beyond this is dominated by van der Waals forces and is thought to occur very fast.

2.8.2 Film drainage models

As two drops are within a distance \( h_o \) of each other, their rate of approach, \( dh/dt \), is governed by the rate of film drainage. The time for a film thickness to decrease from \( h_o \) to \( h_{crit} \) is defined as the drainage time \( (t_{drain}) \). Table 2-1 summarizes the rate of film drainage and the drainage time for three different boundary conditions, the immobile interface (IMI),
partially mobile interface (PMI), and fully mobile interface (FMI) (Chester, 1991; Ottino et al., 2000). These equations describe the decrease in interfacial velocity (or mobility), which occurs as viscosity of the drops increases.

The driving force for film drainage, $F$, is assumed to be a constant. For solid particles, $F$ is the Stokes drag acting on the drops, to a first approximation (Chester, 1991),

$$F \sim 6\pi \mu \gamma a^2$$  \hspace{1cm} (2-40)

For liquid drops, $F$ is expected to be a weak function of the viscosity ratio, but no information on the dependence is available (Chesters, 1991). Since $h_o \gg h_{\text{crit}}$, $h_o^{-2}$ in Equation 2-37 (b) and $h_o^{-1}$ in Equation 2-38(b) are neglected comparing to $h_{\text{crit}}^{-2}$ and $h_{\text{crit}}^{-1}$ respectively. Equation 2-39(b) is insensitive to $h_o$. Following Janssen (1997), we approximate $h_o$ with the initial drop radius $a$.

Table 2-1 The rate of drainage and the drainage time for different interfacial models (Chesters, 1991; Ottino et al., 2000)

<table>
<thead>
<tr>
<th>Interfacial Model</th>
<th>Drainage Rate</th>
<th>Drainage Time</th>
<th>Criteria</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Immobile Interface (IMI)</strong></td>
<td>$-\frac{dh}{dt} = \frac{8\pi \sigma^2 h^3}{3\mu a^2 F}$ (2-37a)</td>
<td>$t_{\text{drain}} = \frac{3\mu a^2 F}{16\pi \sigma^2} \left( \frac{1}{h_{\text{crit}}^2} - \frac{1}{h_o^2} \right)$ (2-37b)</td>
<td>$\lambda &gt; 3r/h$ (2-37c)</td>
</tr>
<tr>
<td><strong>Partially Mobile Interface (PMI)</strong></td>
<td>$-\frac{dh}{dt} = \frac{2(2\pi \sigma / a)^{3/2}}{\pi \mu_i F^{1/2}} h^2$ (2-38a)</td>
<td>$t_{\text{drain}} = \frac{\pi \mu_i F^{1/2}}{2(2\pi \sigma / a)^{3/2}} \left( \frac{1}{h_{\text{crit}}} - \frac{1}{h_o} \right)$ (2-38b)</td>
<td>$6h/r &lt; \lambda &lt; 3r/h$ (2-38c)</td>
</tr>
<tr>
<td><strong>Fully Mobile Interface (FMI)</strong></td>
<td>$-\frac{dh}{dt} = \frac{2\sigma h}{3\mu a}$ (2-39a)</td>
<td>$t_{\text{drain}} = \frac{3\mu a}{2\sigma} \ln \left( \frac{h_o}{h_{\text{crit}}} \right)$ (2-39b)</td>
<td>$\lambda &lt; 6h/r$ (2-39c)</td>
</tr>
</tbody>
</table>

Where $r$ and $h$ are the radius and thickness of the film. $\mu_i$ and $\mu$ are the viscosity of the dispersed phase and the continuous phase, respectively. $a$ is the initial radius of the drop, $\sigma$ is the interfacial tension, and $F$ is the driving force for film drainage.

Several important conclusions can be drawn from the equations for the drainage time 2-37b, 38b and 39b:
(1) For all three cases, the drainage time increases with the drop radius, so that small drops are more likely to coalesce.

(2) For IMI and PMI, the drainage time increases as the drag force $F$ increases. Thus, low shear rates promote coalescence. However, for FMI, $t_{\text{drain}}$ is independent of the drag force and the shear rate.

(3) The drainage time increases as the mobility of the interface decreases. For example, for a system with $\mu_i=\mu=1\text{Pa.s}$, $\sigma=5\times10^{-3}\text{N/m}$, $a=10\mu\text{m}=10^{-5}\text{m}$, $h_{\text{crit}}=10^{-8}\text{m}$, $\dot{\gamma}=100\text{s}^{-1}$, the drainage times for IMI, PMI and FMI are calculated to be 449s, 0.387s, and 0.0207s respectively. The mobility of the interface increases as the viscosity ratio decreases. Therefore, drops with low viscosity ratio are more likely to coalesce. Surfactants on the interface reduce its mobility, and thus inhibit coalescence.

Bartok and Mason (1959) experimentally studied drop collision and coalescence by moving pairs of quasi-spherical drops ($Ca<<1$) toward each other in simple shear flow. They observed coalescence in two systems of $\lambda=2.2$ and $1.3\times10^{-4}$, and found that coalescence is more likely in freshly prepared systems and at low shear rates. Coalescence is inhibited by surfactants on the interface. The tendency of aged drops to coalesce decreases, indicating impurities may have accumulated on the interface. The experiments generally agree with the above theoretical predictions.

**Criteria for the models**

In general, IMI, PMI, and FMI are applicable for high, medium, and low viscosity ratios respectively. However, PMI does not asymptote toward IMI at high $\lambda$ or toward FMI at low $\lambda$. The applicability of the models can be obtained by equating the drainage rate of PMI
with that of the other two models. For example, equating Equation 2-37a and 2-38a gives the boundary between PMI and IMI,

$$\lambda = 3\sqrt{3}Ca^{1/2}a/h$$

(2-41)

Similarly, equating Equation 2-38a and 2-39a gives the boundary between PMI and FMI,

$$\lambda = 2\sqrt{3}Ca^{-1/2}h/a$$

(2-42)

In summary, the applicability of the three models is

**IMI:** \(\lambda > 3\sqrt{3}Ca^{1/2}a/h\)  

(2-43)

**PMI:** \(3\sqrt{3}Ca^{1/2}a/h > \lambda > 2\sqrt{3}Ca^{-1/2}h/a\)  

(2-44)

**FMI:** \(\lambda < 2\sqrt{3}Ca^{-1/2}h/a\)  

(2-45)

Chesters (1991) derived a formula of the film radius \(r\) as a function of the capillary number

$$r/a \sim (3Ca)^{1/2}$$

(2-46)

Ottino et al. (2000) further applied Equation 2-46 to Equation 2-43, 2-44, and 2-45, and obtained the criteria for the applicability of the models, listed in the right column of Table 2-1. The film thickness \(h\) in the criteria can be approximated by \(h_{cris}\), as the final stage of drainage is typically rate limiting (Chester, 1991; Janssen, 1997).

Based on a scaling analysis, Loewenberg and Hinch (1996) found a dependence of \(r/a\) on \(Ca\) and \(\lambda\)

For \(Ca<<h/a\), \(r\sim(ha)^{1/2}\)

(2-47)

For \(h/a\leq Ca\leq(l+\lambda)^{-2/3}\), \(r\sim Ca^{1/2}\)

(2-48)

For \(Ca=O(l+\lambda)^{-2/3}\), \(r\sim a(l+\lambda)^{-1/3}\)

(2-49)
For moderate deformations, Loewenberg and Hinch's estimate of $r \sim a Ca^{\frac{1}{6}}$ is on the same scale as Chesters' (Equation 2-46). Equation 2-49 indicates that $r$ could be as large as $a$ for highly deformed drops ($Ca=O(1)$) at very low viscosity ratios ($\lambda << 1$).

Chesters (1991) argued that $r<<a$ should be satisfied for coalescence to occur. Thus coalescence is only possible at very small values of $Ca$, as indicated by Equation 2-46. Guido and Simeone (1998) visualized drop collision and coalescence driven by the simple shear flow. Figure 2-21 shows a collision without coalescence at $Ca=0.13$ and $\lambda=1.4$. The observed value of $r/a$ can be as high as 0.7. Equation 2-46 gives a value of 0.62 for $Ca=0.13$, which is in reasonable agreement. Therefore, Chesters' assumption of $r<<a$, which is derived from solid or rigid particles that are almost point contacted, is invalid for highly deformable drops.

![Figure 2-21 Drop collision without coalescence at $Ca=0.13$, $\lambda=1.4$, and $a=20\mu m$ (Guido and Simeone (1998)).](image)

2.8.3 Critical drop radius for coalescence

A collision can lead to coalescence only when the contact time of the drops is longer than the required drainage time for the film to thin to $h_{crit}$. For solid particles, the contact time can be scaled by the reciprocal of the shear rate (Chesters, 1991),
\[ t_i \sim (\dot{\gamma})^{-1} \]  \hspace{1cm} (2-50)

Equation 2-50 indicates that the contact time decreases with the shear rate. Therefore, low shear rate promotes drop coalescence.

Equating \( t_i \) of Equation 2-50 and \( t_{\text{drain}} \) (Equation 2-37b, 38b, and 39b) gives a critical drop size, above which coalescence is unlikely (Janssen and Meijer, 1995),

**IMI:** \[ a_{cc} = \left( \frac{8}{9} \right)^{1/4} \frac{Y}{h_{\text{crit}}} (\mu \dot{\gamma}/\sigma)^{1/2} \]  \hspace{1cm} (2-51)

**PMI:** \[ a_{cc} = \left( \frac{4}{\sqrt{3}} \right)^{1/2} \frac{Y^{1/2}}{h_{\text{crit}}} \lambda^{1/2} (\mu \dot{\gamma}/\sigma)^{1/2} \]  \hspace{1cm} (2-52)

**FMI:** \[ a_{cc} \ln \left( \frac{a_{cc}}{h_{\text{crit}}} \right) = \frac{2}{3} (\mu \dot{\gamma}/\sigma)^{-1} \]  \hspace{1cm} (2-53)

Note that \( t_i \sim (\dot{\gamma})^{-1} \) is derived for solid particles. For liquid drops, \( t_i \) is expected as a weak function of the viscosity ratio. However, no such dependence is currently available (Chesters, 1991). As we will see in Chapter 4, \( t_i \sim (\dot{\gamma})^{-1} \) is likely to underestimate the contact time for deformable drops, particularly for drops with \( \lambda \ll 1 \). As a result, the above \( a_{cc} \)s may have been underestimated.

Hu et al. (2000) experimentally studied the collision and coalescence of quasi-spherical drops (\( Ca \ll 1 \)) in a linear flow that approximates plane hyperbolic flow (\( \alpha = 0.9 \)). They found that coalescence is possible only when \( Ca \) is below a critical value. This critical capillary number for coalescence \( Ca_{cc} \) is scaled with the drop radius as \( Ca_{cc} \sim \alpha^{-0.82} \), and with \( \lambda \) as \( \alpha^{-0.82} Ca_{cc} \sim \lambda^{-0.41} \). Experimental measurement of \( Ca_{cc} \) for simple shear flow is currently unavailable.
2.8.4 Coalescence and breakup

As discussed in Section 2.2.2, drops with radius larger than a critical value $a_c$, i.e., $Ca > Ca_c$, will break under shear. A drop breaks into 2 equal sized drops at $Ca = Ca_c$ under quasi-equilibrium conditions. The daughter drop size is thus,

$$a_f = 2^{-1/3} Ca_c \left( \mu \dot{\gamma} / \sigma \right)^{-1}$$  \hspace{1cm} (2-54)

Figure 2-22 compares $a_f$ as a function of $(\mu \dot{\gamma} / \sigma)^{-1}$ at $\lambda=1$ to the maximum drop size resulting from coalescence, i.e., $2^{1/3} a_{c,c}$, for the three interfacial models. Drop breakup dominates the area above the breakup line of $a_f$, while coalescence dominates the area below the coalescence line of $2^{1/3} a_{c,c}$. The breakup region and the coalescence region overlap when the shear rate is larger than a critical value $\dot{\gamma}_{c,c}$ (see the highlighted area). Therefore, drop breakup and coalescence may coexist at high shear rates. This is different than Chesters' argument (1991), "Coalescence is not in general to be expected in zones where drop breakup occurs (for which $Ca \geq 1$)." However, his argument was based on assumptions, e.g. $r \ll a$, which are derived from solid particles.

![Figure 2-22 Comparison of drop size from breakup and coalescence (Janssen and Meijer, 1995).](image)

Minale et al. (1997) experimentally confirmed the coexistence of breakup and coalescence at high shear rates. They determined the average drop size of a polymer blend
(PIB-PDMS) under steady shear from rheological measurements of the linear dynamic moduli. They found a hysteresis of the steady drop size as a function of the shear rate. Multiple steady states exist under a critical shear rate, but only single steady drop size exists above it. They proposed that the single steady drop state at high shear rates is due to the equilibrium between breakup and coalescence, corresponding to a point in the overlapping area of Figure 2-22. Their polymer blend is fairly concentrated ($\phi=10\%$), thus they used $h_{\text{crit}}$ as an adjustable parameter rather than calculate it from Equation 2-36. We will present the first direct observation of concurring drop breakup and coalescence in Chapter 4.

### 2.8.5 Drop separation

Bartok and Mason (1959) observed that when two colliding quasi-spherical drops separate without coalescence, their distance in the velocity gradient direction increases, generating self-diffusion of drops. Recently, Loewenberg and Hinch (1997) numerically simulated the collision of two deformable drops in shear flow using the boundary integral method. They showed that drop collision results in self-diffusion of the drops across the flow direction, in agreement with Bartok and Mason's observation. Furthermore, they found that the self-diffusivity is asymmetrical, i.e. it is larger in the velocity gradient direction than in the vorticity direction.

Guido and Simeone (1998) also observed that drops separate further in the velocity gradient direction when they collide without coalescence. For example, Figure 2-21 shows that the distance between the two drops along the velocity gradient $\Delta y$ increases during the collision. This is consistent with Loewenberg and Hinch's results and with Bartok and Mason's observation (1959) for quasi-spherical drops ($Ca<<1$). In addition, they found that
drops accelerate as they separate from each other (no coalescence). Figure 2-23 shows the relative velocity of the same drops shown in Figure 2-21 as a function of their distance. The relative velocity after separation was almost three times the initial approach velocity. The acceleration is due to the increase of the distance of the two drops along the velocity gradient. For a steady simple shear, \( \dot{\gamma} = \frac{dv_x}{dy} = \text{constant} \), thus \( \Delta v_x \) increases as \( \Delta y \) increases. The solid line in Figure 2-23 is calculated from \( \dot{\gamma} \Delta y \), where \( \Delta y \) is measured from the same experiment. That calculated curve agrees with the direct measured \( \Delta v_x \).

![Figure 2-23](image)

Figure 2-23 The relative velocity between the drops, \( \Delta v_x \), increases during drop collision as a result of their separation in the velocity gradient direction. \( \Delta x/a \) is the nondimensionalized distance between the drops. Same experiment as in Figure 2-21. The continuous line was calculated as \( \dot{\gamma} \Delta y \) (Guido and Simeone (1998)).

Guido and Simeone (1998) observed a rare coalescence event at \( \lambda = 0.36 \). In two other systems with higher \( \lambda, \lambda = 1.4 \) and 2.0, they did not see any coalescence. The coalescence took place when the two drops were about to separate from each other in the extensional quadrants of the shear flow (see Figure 2-24), which agrees with Loewenberg and Hinch's numerical results (1996). Loewenberg and Hinch predicted that for nearly spherical drops \( (Ca << 1) \) or \( \lambda > O(1) \), coalescence is most likely to occur when drops are pressed together in
the compression quadrant of the simple shear flow. For $Ca=O(1)$ and $\lambda=O(1)$, coalescence is most likely to occur when closely contacting drops are pulled apart in the extensional quadrant of simple shear flow.

Figure 2.24 Sequence of images showing a collision with coalescence. $Ca=0.13$, $\lambda=0.36$ (Guido and Simeone, 1998).
Chapter 3 Experimental Section

3.1 Experimental Setup

Our principal experimental approach is to visually follow the deformation and breakup of drops in simple shear flow using high-speed photography. Figure 3-1 shows an experimental apparatus consisting of three units: a shear cell, a driving unit, and an image recording and processing unit.

(1) The shear cell (initial design)

We have built two linear shear cells on the stage of an inverted microscope (Nikon, Eclipse TE-300) to generate the simple shear flow field. The shear cell shown in Figure 3-1 is the initial design that was used. It consists of two glass plates (2mm x 30mm x 100mm), between which the emulsion sample is placed (see the front view). The upper plate is glued onto a moving aluminum frame, which moves smoothly along linear rollers (Schneeberger, RN-3-300). The lower plate is glued onto the microscope stage, so that it is stationary. The gap between the plates is maintained by three micrometers with electronic touch sensors, which are connected to three LEDs (light emission diodes). The typical gap thickness is between 100 and 200 microns.

(2) The driving unit

The upper plate is driven by an air cylinder (Bimba, MRS-092-DXPZ). A hydraulic damping cylinder (Deschner, model Kinechek 1003-31-3) reduces fluctuations in the speed of the plate. The inlet compressed air (~100 Psi) is first filtered and then adjusted with a
regulator to a lower pressure, usually around 60 Psi, before entering the air cylinder. The upper plate can move back and forth and is controlled by a direction control valve that determines the direction of flow of the air in the cylinder. The plate speed can be varied between 1 and 50 cm/s by adjusting either the air pressure or a valve of the damping cylinder.
(3) Image capturing and processing unit

The deformation and breakup of drops is observed through the microscope with a long working distance objective (20x or 40x). Images are obtained along the velocity gradient direction. The images are filmed using a high-speed digital camera (Kodak, Ektapro 1000HRC) that can record up to 1000 frames of digital images in one second. The exposure time is extremely short, typically 50μs. Thus a mercury lamp with strong light intensity is used as the light source to provide sufficient exposure for such a short time. The experiment usually lasts from 1 to 10 seconds. The captured images are then recorded on video types or downloaded to a computer for further processing.

(a) Initial design of shear cell   (b) Shear cell with improved design
Figure 3-2 Comparison of the former shear cell and the current shear cell
(a) The upper plate of the former shear cell moves, while the lower plate is stationary. Drops move in and out of the field of view (denote by the area between the two dotted lines) (b) The two plates of the current shear cell move in opposite direction, so the drop stays in the field of view as it deforms.

3.1.1 Current shear cell (improved design)

The lower plate of the previous shear cell is stationary, so that the drops move in and out of the field of view. Thus we are not able to follow the whole breakup history of a single drop (see Figure 3-2 (a)). Thus, we built the current shear cell in which the two plates move
in opposite direction at equal speed (see Figure 3-2 (b)). The drops then stay in the field of view while they deform.

The details of the improved design are shown in Figure 3-3. The upper glass plate (2mm x 25mm x 100mm) and the lower glass plate (6mm x 25mm x 100mm) are epoxied onto two aluminum moving frames. The two moving frames move smoothly along linear rollers, mounted on two lower fixed frames. The upper plate is driven by the same driving mechanism as for the previous cell, which in turn drives the lower plate through a gear system (see below). The moving distances for the two plates are both 37.5mm, and the relative moving speed of the two plates can be adjusted between 7.5 and 75 mm/s, corresponding to a shear rate range of 75-750s\(^{-1}\) for a 100\(\mu\)m gap between the two glass plates. Another improvement in the design of the cell is that a translating stage is built between the lower fixed frame and the microscope stage. By adjusting the two knobs of the translating stage, the whole cell can be moved horizontally in four directions to target a specific drop and move it into the field of view.

The gear system

The gear system is shown in the top view and the side view of the improved shear cell (Figure 3-3). A driving rack is mounted onto the upper moving frame. A slave rack is mounted onto the lower moving frame through an "L" shaped part (see side view). The two racks are parallel to each other, and each catches onto one side of a spur gear. Pushed by the driving rack, the gear rotates to move the slave rack at equal speed but in opposite direction with the driving rack. Gears with conjugate teeth are known for transmitting motions with uniform velocity (Ewert, 1997). Particularly, the spur gear with a 20° pressure angle has the
Figure 3-3 Schematic of the improved shear cell. The two plates move in opposite directions at equal speeds, facilitated by a gear system consisting of a driving rack, a spur gear and a slave rack (see the top view and the side view). The plates and frames are to scale in the horizontal plane (see the ruler bar in the top view), but not in the vertical direction.
advantage of maintaining a smooth transmission while withstanding a high load (Drago, 1991; Ewert, 1997). The two racks must be parallel for a smooth transmission. The parallelism is checked by adjusting the positioning screws of the slave rack and measuring the distance between the racks at several locations along the racks using a micrometer. In addition, the gear and the racks are tightly pressed against each other to attain a smooth transmission. Otherwise, the gear may jerk whenever it turns a tooth.

3.1.2 Parallelism of the plates

To ensure a uniform shear, it is essential that the two glass plates are parallel to each other. First, the surfaces of the glass plates are ground to the precision of 2μm. This is checked by putting an optical flat on top of the plate and counting the number of interference fringes (see Appendix A). Second, we carefully epoxy the glass plates on the moving frames to avoid any bumping and twisting of the plates. The frame is laid onto a solid horizontal surface (a level confirms that the surface is horizontal). A uniform layer of ultraviolet (UV) light cured epoxy (Norland Products, Inc., NOA-60) is applied between the surface of the glass plate and the moving frame. An optical flat is placed on top of the glass plate, both to check the parallelism as the epoxy cures and as a weight for better binding between the plate and the frame. UV light is used to cure the epoxy, and the whole setup is left overnight to guarantee a full aging of the epoxy.

The gap thickness of the two glass plates is set by three high precision micrometers (Thorlabs, Model 146-200) with electronic touch sensors (modified from BM-50 of Stanley Sheppard Co.). The micrometers are mounted on the top aluminum frame, while the electronic touch sensors are imbedded underneath the surface of the bottom frame (see
Figure 3-3 (c)). When the tip of a micrometer is in touch with the sensor, the circuit is closed and the corresponding LED turns on. The accuracy of the micrometers and the sensitivity of the sensors are both within 1μm according to the manufacturers' technical data. The micrometers are first loosened to bring the plates in contact with each other, and the parallelism of the plates is ascertained by counting the interference fringes visible on the upper plate. In general, the number of fringes is less than 10 along the length of the plate, corresponding to a gap of about 3μm (See Appendix A). The three micrometers are then adjusted so that the three LEDs connected with the touch sensors just turn on, zeroing the gap. Then the micrometers are raised to the height corresponding to the desired gap width between the two plates, which is typically 100μm. Finally, to ensure the parallelism is maintained as the gap is raised to 100μm, we check the parallelism of the plates again by a focusing technique. We put marks on the surface of one of the plates, for example, the lower plate. Then we adjust the focusing knob of the microscope to focus on the marks, and record the reading of the focusing knob. Moving the lower plate along the roller, we repeat the measurement every 5mm. The same measurement is repeated for the upper plate. The difference of the reading along the length of the plate is ±2μm.

3.1.3 Measuring the plate velocity

The shear rate is the relative speed of the two plates divided by the gap thickness. The speed of the plates is measured by two methods. In the first method, marks are made on the surface of one of the plates (without an emulsion sample). The distance that a mark moves from one frame to the other is then measured and multiplied by the exposure rate (frames per second). The results show that the plates accelerate to a steady speed in around 80ms for a
shear rate between 100 and 750s\(^{-1}\). The average speed is measured by dividing the total distance each plate moves (37.5mm) by the duration of the shear, which is counted by the number of frames from the start of the shear to the end of the shear (as observed by the motion of drops in the field of view). The average shear rate measured this way is within ±1% of the steady shear rate measured by the first method, and it can be obtained each time we shear a new emulsion sample. The shear rate given in the following chapters is therefore measured from the second method.

3.2 Newtonian Emulsions and Their Physical Properties

The dispersed phase and the continuous phase of the emulsions systems and their physical properties are listed in Table 3-1. The emulsions are made by adding drops of silicone oil with a capillary tube to the continuous phase and gently stirring with a spatula. The typical volume fraction of the inner phase is 0.1~0.2 wt%. No drop coalescence was observed in hours in the absence of shear at this low concentration.

3.2.1 Rheological measurements

The rheological properties of the fluids are measured in a strain-controlled rheometer (Rheometric, ARES) with a cone-and-plate geometry at 25°C. A general procedure of the measurements is included in Appendix B. The rheometer is calibrated with a standard silicone oil whose viscosity is known (Brookfield, 1000cps). The error is generally within 2%. All the fluids are Newtonian except for the aqueous solution of 12%PVP (polyvinyl pyrrolidone) and 2%SDS (Sodium dodecyl sulfate, an ionic surfactant), and their viscosities are listed in Table 3-1. The aqueous solution of 12%PVP+2%SDS is shear thinning (see
Figure 3-4). The emulsion for which this solution forms the continuous phase is sheared at 750s\(^{-1}\). At this shear rate, the continuous phase has a viscosity of 701mPa.s.

<table>
<thead>
<tr>
<th>Viscosity ratio</th>
<th>Inner phase</th>
<th>Density g/ml</th>
<th>Viscosity mPa.s</th>
<th>Outer phase</th>
<th>Density g/ml</th>
<th>Viscosity mPa.s</th>
<th>Interfacial Tension, mN/m</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.00166</td>
<td>SO5</td>
<td>0.9130</td>
<td>4.61</td>
<td>OCO, Z-1</td>
<td>0.9868</td>
<td>2780</td>
<td>3.57±0.1</td>
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<td>0.0182</td>
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<td>50.5</td>
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<td>0.9868</td>
<td>2780</td>
<td>4.85±0.1</td>
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<td>SO50</td>
<td>0.9588</td>
<td>50.5</td>
<td>OCO, XY</td>
<td>0.9792</td>
<td>1675</td>
<td>5.0±0.1</td>
</tr>
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<td>0.0748</td>
<td>SO50</td>
<td>0.9588</td>
<td>50.5</td>
<td>CO</td>
<td>0.9558</td>
<td>675</td>
<td>4.2±0.1</td>
</tr>
<tr>
<td>0.121</td>
<td>SO350</td>
<td>0.9671</td>
<td>337</td>
<td>OCO, Z-1</td>
<td>0.9868</td>
<td>2780</td>
<td>4.85±0.1</td>
</tr>
<tr>
<td>0.300</td>
<td>SO500</td>
<td>0.9676</td>
<td>502</td>
<td>OCO, XY</td>
<td>0.9792</td>
<td>1675</td>
<td>5.0±0.1</td>
</tr>
<tr>
<td>0.440</td>
<td>MSO-1</td>
<td>0.9692</td>
<td>1222</td>
<td>OCO, Z-1</td>
<td>0.9868</td>
<td>2780</td>
<td>4.85±0.1</td>
</tr>
<tr>
<td>0.496</td>
<td>MSO-2</td>
<td>0.9687</td>
<td>830</td>
<td>OCO, XY</td>
<td>0.9792</td>
<td>1673</td>
<td>5.0±0.1</td>
</tr>
<tr>
<td>0.500</td>
<td>SO350</td>
<td>0.9671</td>
<td>337</td>
<td>CO</td>
<td>0.9558</td>
<td>675</td>
<td>4.2±0.1</td>
</tr>
<tr>
<td>0.590</td>
<td>SO500</td>
<td>0.9676</td>
<td>502</td>
<td>Glycerol</td>
<td>1.2571</td>
<td>855</td>
<td>24.7±0.3</td>
</tr>
<tr>
<td>0.695</td>
<td>SO5k</td>
<td>0.9702</td>
<td>5842</td>
<td>94% Maltose + 6% water</td>
<td>1.3976</td>
<td>8411</td>
<td>39.8±0.3</td>
</tr>
<tr>
<td>1.01</td>
<td>MSO-3</td>
<td>0.9692</td>
<td>1684</td>
<td>OCO, XY</td>
<td>0.9792</td>
<td>1675</td>
<td>5.0±0.1</td>
</tr>
<tr>
<td>2.10</td>
<td>SO5k</td>
<td>0.9702</td>
<td>5842</td>
<td>OCO, Z-1</td>
<td>0.9868</td>
<td>2780</td>
<td>4.85±0.1</td>
</tr>
<tr>
<td>3.47</td>
<td>SO10k</td>
<td>0.9704</td>
<td>9655</td>
<td>OCO, Z-1</td>
<td>0.9868</td>
<td>2780</td>
<td>4.85±0.1</td>
</tr>
<tr>
<td>0.45</td>
<td>SO350</td>
<td>0.9674</td>
<td>338</td>
<td>2% Tween80 in Glycerol</td>
<td>1.2538</td>
<td>747</td>
<td>6.0±0.1</td>
</tr>
<tr>
<td>0.48</td>
<td>SO350</td>
<td>0.9674</td>
<td>338</td>
<td>12%PVP+2%SDS water solution</td>
<td>1.0257</td>
<td>701</td>
<td>4.8±0.1</td>
</tr>
</tbody>
</table>

Note:
1. SO--- Silicone oil (Rhone Poulenc), the number following "SO" denotes the nominal viscosity in mPa.s. MSO-1 is 80%SO1k+20%SO5k; MSO-2 is 70%SO1k+30%SO500, and MSO-3 is 74%SO1k+26%SO5k (all the percentage are in weight percent).
2. CO---Castor oil (Sigma-Aldrich).
   OCO--- oxidized silicone oil. OCO Z-1 (Pfau Industrial Animal Oils, Z-1), OCO XY (Degen Oil and Chemical Co., PC-30)
3. Maltose---50% Maltose corn syrup (Cargill)
4. Tween80---Polyoxyethylene Sorbitan Monooleate (Sigma-Aldrich), a nonionic surfactant.
   Glycerol from Sigma-Aldrich.
   PVP---polyvinyl pyrolidine (MW=1.3M, Sigma-Aldrich).
   SDS---Sodium dodecyl sulfate (Sigma-Aldrich), an ionic surfactant.
5. All emulsions were sheared in the improved shear cell, except for the last two, which were sheared in the previous shear cell.
6. The uncertainty in the viscosity value is 2%, and the uncertainty in the density value is 0.0001.

3.2.2 Interfacial tension

The interfacial tension of the emulsions is measured by a pendant drop technique (see Figure 3-5). A drop of liquid A is suspended from a tube in liquid B. Gravity tends to elongate the drop, while interfacial tension resists the elongation in order to minimize the
Figure 3-4 The aqueous solution of 12%PVP+2%SDS is shear thinning, and has a viscosity of 701 mPa.s at a shear rate of 750s⁻¹.

Figure 3-5 Schematic of the pendant drop technique for measuring interfacial tension

interfacial area. The equilibrium shape of the drop is governed by the balance of the two forces, according to the Laplace equation (Miller and Neogi, 1985):

\[ \Delta P = P_A - P_B = \left( \frac{1}{r_1} + \frac{1}{r_2} \right) \sigma \]  

(3-1)

where \( P_A \) and \( P_B \) are the pressure inside and outside of the drop, respectively. \( r_1 \) and \( r_2 \) are the radii of curvature at any point of the interface, and \( \sigma \) is the interfacial tension. Taking the tip of the drop, point O, as the reference point:
\[ P_A - P_B = (P_{Ao} - \rho_A gz) - (P_{Bo} - \rho_B gz) \]
\[ = (P_{Ao} - P_{Bo}) - (\rho_A gz - \rho_B gz) \]
\[ = \frac{2\sigma}{b} - (\rho_A - \rho_B)gz \]

where \( P_{Ao} \) and \( P_{Bo} \) are the pressures at point O, and \( b \) is the radius of curvature at point O.

For axisymmetric drops, \( b \) is the same for all orientations. Substituting Equation (3-2) into (3-1) gives

\[ \frac{2\sigma}{b} - (\rho_A - \rho_B)gz = \left( \frac{1}{r_1} + \frac{1}{r_2} \right) \sigma \]

(3-3)

For an axisymmetric drop, \( r_1 \) and \( r_2 \) can be related to the drop curvature by

\[ \frac{1}{r_1} = \frac{d^2z/dr^2}{\left[ 1 + (dz/dr)^2 \right]^{3/2}} \]

(3-4)

\[ \frac{1}{r_2} = \frac{dz/dr}{r\left[ 1 + (dz/dr)^2 \right]^{1/2}} \]

(3-5)

where \( r \) and \( z \) are the radius and vertical position of any given point on the drop surface (see Figure 3-5). Substituting Equation 3-4 and 3-5 into 3-3 gives a second order differential equation, whose numerical results of Equation 3-6 are available in the form of tables (Padday, 1969). The interfacial tension can be determined by measuring the maximum radius of the drop \( r_m \) and its position \( z_m \), and then referring to the tables.

In the experiment, the images of the equilibrium drop are taken by a CCD camera. The image processing and data analysis are done automatically by computer programs to obtain the interfacial tension (Kho Irani, 1995). The densities of the liquids are necessary for the calculation. They are determined using a pycnometer (25ml) and a digital scale (±0.1mg), and are listed in Table 3-1. To minimize the experimental error, the proper tube diameter is chosen to be close to the capillary constant,
\[ C = \sqrt{\frac{2\sigma}{(\rho_s - \rho_o)g}} \]  

The surface tension of water/air and silicone oil/air are measured and compared to literature values. The errors are within 2%. The interfacial tensions of the emulsions are listed in Table 3-1.

3.3 Loading Samples and Choosing Targeted Drops

We first position the glass plates at the center of the rollers. After zeroing the gap, we set the gap at 100\(\mu\)m with the three micrometers. The emulsion sample is loaded along the surface of the lower glass plate with a spatula. The upper plate is then very carefully placed on top of the lower one. The teeth of the gear and the slave rack are first matched before the upper plate is further lowered. The upper plate is lowered slowly until it is fully supported by the micrometers. A sudden drop of the upper plate may incorporate air bubbles into the sample, and may squeeze the fluids out of the plates, causing the drops to break. After loading the sample, we lock the plates in place, and search for drops with radii of 5~35 microns in the middle of the emulsion sample. Drops in the middle of the sample are identified by focusing on the surfaces of the upper plate and lower plate. We take the average of the two readings to be the middle of the sample, and search for suitable drops around this reading. Great care has been taken to avoid drops adjacent to air bubble or other drops. Usually, the targeted drop is the only drop in the field of view. For each sample, we record the deformation and breakup of a minimum of 20 drops (in some cases, the dynamics of 40 drops have been recorded). Between each load, the upper plate is disassembled. Both plates are thoroughly cleaned with Simple Green (Sunshine Makers), a detergent that is very
effective in cleaning up silicone oil. The plates are then rinsed three times with deionized water and air-dried.

3.4 Image Recording and Processing

After locating a target drop, we start the recording of the camera and then initiate the shear by pushing the direction control button on the control unit of the air cylinder (see Figure 3-1). The recording is manually stopped after the shear terminates, or it automatically terminates when the memory runs out. The images are replayed on the monitor connected to the camera processor. Valuable images are downloaded as tag image file format (tif) on the magneto-optical disk of the processor, and are transferred to a computer through a SCSI cable.

The images are processed using Photoshop (Adobe, Version 5.5) to measure the thread width, the wavelength, and the drop size. The typical procedure to process the images is:

1. Open the picture of interest in Photoshop;
2. Duplicate the image, and work on the duplication to avoid any unwanted irreversible changes.
3. Reduce the noise by a command of "Gaussian blur" with a radial value of 0.3 pixels.
4. Improve the contrast by a command of "auto contrast".
5. Highlight the area of interest, and apply a command of "threshold" to cutoff the pixels with gray values lower than a certain value. Usually, the automatically detected threshold value is used.
(6) Magnify the image to its maximum (16 times of the original size).

(7) Use the ruler bar to measure the size of the object of interest. The ruler has been previously calibrated by measuring the length of a micro-ruler of known length.

(8) Save the processed images for future reference.
Chapter 4 Drop Breakup Mechanism

As we mentioned in the Review section, the dynamics of drop breakup in flow has not been studied in depth experimentally (Stone, 1994). In this section, we will discuss the first systematic experiments on the mechanisms of drop breakup in steady simple shear flow. We focus on the following questions: “What are the drop breakup mechanisms in simple shear flow? How do the dynamics of drop breakup depend on the capillary number and the viscosity ratio?” First, in Section 4.1, we will study the effects of $Ca$ at a fixed $\lambda$, taking $\lambda=0.5$ as an example. As we shall see, at $Ca\sim Ca_c$, the drop breaks by necking; for $Ca<2Ca_c$, drop breakup is caused by end pinching; for $Ca>2Ca_c$, the capillary instability is the dominant mechanism for drop breakup. In section 4.2, 4.3, and 4.4, we will elaborate the above three cases, respectively. We will discuss and summarize our results in Section 4.5. In particular, the influence of $Ca$ and $\lambda$ on drop breakup is condensed into a single "map" (see Figure 4-22).

4.1 Drop Breakup at $\lambda=0.5$

We take $\lambda=0.5$ as an example to illustrate the effect of $Ca$ on drop breakup mechanism. The emulsion is 0.2wt% 337mPa.s silicone oil drops in a 675mPa.s castor oil matrix. Using the linear shear cell described in Chapter 3, we have sheared the emulsion in simple shear flow at a variety of shear rates from $130s^{-1}$ to $500s^{-1}$. At each shear rate, we have followed the deformation and breakup of at least 40 drops of various sizes, each run corresponding to a single drop. We will introduce the typical features of drop breakup at a fixed shear rate of $323s^{-1}$, and then describe other shear rates.
4.1.1 $\dot{\gamma}=323 s^{-1}$

Figure 4-1 shows the transient drop breakup for a variety of Ca$\alpha$s at $\dot{\gamma}=323 s^{-1}$. Taking the onset of the flow as time zero, the time (milliseconds) when each frame is taken is marked on the right side of each picture. At a given shear rate, drops that are smaller than a critical radius, $a_c$, reach steady ellipsoidal shapes under shear. Drops larger than $a_c$ go through a transient elongation and break into smaller droplets. The critical drop radius is experimentally determined as the radius of the smallest drop that breaks in the flows. The capillary number corresponding to this critical radius is defined as the critical capillary number, Ca$\alpha$. Figure 4-1 (I) shows such a drop breaking up at Ca $\sim$ Ca$\alpha$ for $\dot{\gamma}=323 s^{-1}$. Under shear, the initially spherical drop is deformed into an ellipsoid. The drop keeps stretching, and a neck forms in the middle of the drop. The neck thins gradually, and eventually causes the drop to break into two equal sized daughter drops separated by smaller satellite and sub-satellite drops. This phenomenon is similar to the quasi-equilibrium breakup (see Figure 2-17, Torza et al., 1972) or the necking mechanism as it sometimes is referred to (Ottino, et al. 2000). Our experiments, however, are different in that we have applied a constant shear flow, rather than a slowly increasing shear rate. The initial drop size in Figure 4-1 (I) is 7.4$\mu m$, which is taken as $a_c$, so that Ca$\alpha$=0.38. The distance between the two daughter drops keeps increasing after the breakup, indicating the two daughter drops move in opposite directions.

A slightly bigger drop corresponding to Ca=1.1Ca$\alpha$ (see Figure 4-1 (II)) forms a longer neck between the two daughter drops, which results in larger satellite and sub-satellite drops. The distance between the two daughter drops also increases after drop breakup. As the initial drop size further increases to Ca=1.3Ca$\alpha$, as shown in Figure 4-1 (III), the drop deforms into a long thread. As the drop stretches, its end bulbs up, and a neck forms between
Figure 4-1 Drop breakup at \( \lambda = 0.5 \) and \( \dot{\gamma} = 323 \text{s}^{-1} \).

(I) At \( Ca > Ca_c \) (\( a = 7.4 \mu m \)), the drop breaks up by necking into 2 equal sized daughter drops separated by smaller satellite and sub-satellite drops.
(II) $Ca = 1.1Ca_c$ and $a = 8.2\mu m$. The neck between the two daughter drops become longer, which results in relatively larger satellite drops compared to those in (I).

(III) $Ca = 1.3Ca_c$ and $a = 9.4\mu m$. The drop deforms into a thread. Drops are pinched off from the end. Similar processes repeat on the remaining part of the thread.

(IV) $Ca = 1.9Ca_c$ and $a = 14\mu m$. The drop deforms into a long thread. End pinching occurs at the end of the thread, while the capillary instability grows on the central part of the thread. The thread width at breakup as shown in (e) is $2.8\mu m$. The wavelength of the capillary instability is uniform along the length of the thread resulting in uniform daughter drops. Daughter drops from the capillary instability are similar in size to those from end pinched drops, except that the first end pinched drop is about twice as big as the other daughter drops. Frames (a)-(j) correspond to 0ms, 40ms, 120ms, 160ms, 200ms, 240ms, 260ms, 300ms, and 372ms respectively.

(V) $Ca = 3.4Ca_c$ and $a = 25\mu m$. The capillary instability is the dominant drop breakup mechanism. The thread width at breakup as shown in (e) is $2.8\mu m$, which is the same as that in (IV). The wavelength of capillary instability is similar to (IV), which results in a similar final drop size. Fewer drops can be observed in the field of view after the thread breaks (compare frame (h) and (i)), which indicates that the distance between the daughter drops increases after drop breakup. Frames (a)-(j) correspond to 0ms, 40ms, 120ms, 320ms, 400ms, 440ms, 480ms, 512ms, and 544ms respectively. The drop in Frame (b) at $t=40ms$ is slightly curved. It is probably due to the turning of the gear as the shear is initiated. The drop becomes straight in $80ms$, once the shear becomes steady. Breakup occurs much later, so its influence on drop breakup is probably minor.

the bulbous end and the uniform central thread. The neck continues to thin which leads to the pinch off of a daughter drop from the end. Similar processes repeat on the remaining part of the thread. Satellite and sub-satellite drops form between the pinched off daughter drops. This phenomenon is similar to the end pinching on an elongated thread in a quiescent matrix (Stone et al., 1986; Stone and Leal, 1989a, b).

Figure 4-1 (IV) shows the breakup of a drop of $Ca = 1.9Ca_c$. Similar end pinching occurs at the ends of the thread (frame (d) and (e)). From frame (f), we start to see the capillary instability growing on the central part of the thread. Taking the onset of the flow as time zero, we define the moment just before the capillary instability becomes visible as the time to breakup ($t_b$). In this case, $t_b = 200ms$ (corresponding to Frame (e)). We denote the radius at breakup $R_b$, as shown in frame (e) $R_b = 2.8\mu m$. When the amplitude of the instability has grown equal to in size to the thread radius, the thread breaks into a line of uniform sized
daughter drops, as a result of the uniformity of the wavelength along the length of the thread. Note that the daughter drops resulting from end pinching are similar in size to those resulting from the capillary instability, except for the first end pinched drops which are about twice as big as the other drops. There are also relatively few end-pinched drops. End pinching usually repeats up to 4-5 times before the capillary instability sets in. Smaller satellite and sub-satellite drops form between the daughter drops, but they are negligibly small comparing to them.

As the initial drop size increases from $Ca=1.9Ca_c$ to $Ca=3.4Ca_c$ (Figure 4-1 (V)), no change in the drop breakup modes is observed. Even though the drop size is 1.8 times bigger than the drop shown in Figure 4-1 (IV), the thread radius at breakup is still 2.8$\mu$m (Frame (f)). Since drops with different initial sizes deform into threads with the same radii at breakup, the larger drops must deform into longer threads. Comparing Figures 4-1 (V) and (IV), we see that the wavelengths appear comparable, so that the final daughter drops must also be similar in size. This can therefore lead to monodisperse emulsions, which we will show quantitatively in Chapter 5. Notice that the time to breakup increases with the initial drop radius, e.g. $t_b$ is 200ms for $Ca=1.9Ca_c$ and 440ms for $Ca=3.4Ca_c$.

Once formed, the daughter drops retract to an ellipsoidal shape, creating space between them. No further elongation and re-breaking of daughter drops have been observed. The daughter drops are carried away from each other by the flow. Figure 4-1 (IV) shows 14 daughter drops immediately after the disintegration of the thread (see frame (h), $t=300$ms), but only 11 drops 72ms later, (see frame (i), $t=372$ms). Similarly, in Figure 4-1 (V), two daughter drops move out of the field of view from $t=512$ms (frame (h)) to $t=544$ms (frame (i)). That fewer drops can be seen in the field of view as time goes on means that the distance
between the daughter drops increases after drop breakup. Consequently, no collision of daughter drops has been observed.

Figure 4-2 shows $R_b$ for a number of drops with a variety of initial drop sizes. The thread radius at breakup does not appear to depend on the initial drop radius. We define the polydispersity ($p$) as the standard deviation divided by the mean thread radius. Conventionally, a distribution is considered to be monodisperse when $p \leq 10\%$. In this case, the polydispersity is 6.6\%, i.e., the thread radius at breakup is monodisperse.

\begin{figure}
\centering
\includegraphics[width=0.5\textwidth]{thread_radius_breakup.png}
\caption{Thread radius at breakup is independent of the initial drop radius for $\lambda = 0.5$ and $\dot{\gamma} = 323s^{-1}$. The emulsion sample is 0.2\% 337mPa.s Silicone oil in a 675mPa.s Caster oil. The polydispersity or $R_b$ is 6.6\%.}
\end{figure}

4.1.2 $\dot{\gamma} = 132s^{-1}$, $197s^{-1}$, and $495s^{-1}$

The same emulsion has also been sheared at different shear rates. At each shear rate, we have followed the breakup of 40 drops with a variety of initial sizes. The breakup modes are no different from those at $323s^{-1}$. At $Ca \sim Ca_c$, the drop breaks via necking (see Figure 4-3 (I)). For $Ca < 2.0Ca_c$, end pinching is the dominant drop breakup mechanism. Figure 4-3 (II) shows an example at $132s^{-1}$. For $Ca > 2.0Ca_c$, the capillary instability is the dominant drop
Figure 4.3 Drop breakup at $\lambda=0.5$ and $\dot{\gamma}=132, 197,$ and $495 \text{s}^{-1}$ shows similar breakup modes as a function of $Ca$ as for $\lambda=0.5$ and $\dot{\gamma}=323 \text{s}^{-1}$: Necking mechanism at $Ca=Ca_c$ (series I), End pinching at $Ca<2.0Ca_c$ (series II), and the capillary instability at $Ca>2.0Ca_c$ (series III and IV). The thread width at breakup and the final drop size decrease with the shear rate.
breakup mechanism (see Figure 4-3 (III) for $197\,s^{-1}$ and (IV) for $495\,s^{-1}$). This is to be expected since Newtonian drop breakup only depends on $Ca$ and $\lambda$. The thread radius at breakup decreases as the shear rate increases: $R_b$ is $4.3\,\mu m$, $2.8\,\mu m$, and $2.0\,\mu m$ at $197\,s^{-1}$, $323\,s^{-1}$, and $495\,s^{-1}$ respectively. Consequently, the final drop size also decreases as the shear rate increases.

**SUMMARY**

Results at $\lambda=0.5$ show that the drop breaks via necking at $Ca \sim Ca_c$. For $Ca<2Ca_c$, drop breakup is caused by end pinching. For $Ca>2Ca_c$, the capillary instability is the dominant drop breakup mechanism. The thread width at breakup does not depend on the initial drop size. The wavelength of the capillary instability is uniform along the length of the thread. The daughter drops retract to an ellipsoidal shape after breakup. No further elongation and re-breaking of the daughter drops is observed. The daughter drops are carried away from each other by the flow. No collision of drops is observed.

**4.2 Drop Breakup at $Ca \sim Ca_c$**

Figure 4-4 shows drop breakup at $Ca \sim Ca_c$ for a range of $\lambda$s. The phases and physical properties of the emulsion corresponding to each $\lambda$ are listed in Table 3-1. In each case, the drop breaks by necking into 2 equal sized daughter drops moving in opposite directions. The critical capillary number is plotted against the viscosity ratio in Figure 4-5. The squares are our experimental results for a steady simple shear flow. The dashed line shows Grace's results (1971) for quasi-equilibrium breakup, where the shear rate was slowly increased until the drop breaks up. Our experimental results are consistently lower than Grace's. Torza, et
Figure 4.4 Drop breakup by necking at $Ca \sim Ca_c$ for $0.0017 < \lambda < 3.5$
Figure 4-5 Dependence of $Ca_c$ on $\lambda$ in simple shear flow shows our results for steady simple shear (squares) are consistently lower than Grace's results from pseudo-equilibrium breakup. The dashed line is an empirical fit to Grace's data (1971) by de Bruijn (1989).

... al.'s experiments (1972) and Hinch and Acrivos' theory (1980) have shown that the critical shear rate for a step shear flow is lower than that for pseudo-equilibrium breakup.

Form Figure 4-4, we can also see that the daughter drops become more slender at low $\lambda$. Quantitatively, we measure the length ($L'$) and width ($B'$) of the daughter drops from the images, and calculate the deformation parameter

$$D' = (L'-B')/(L'+B')$$

(4-1)

The prime denotes that the parameters are different from their original definition in Chapter 2. Since the drop is observed in direction of velocity gradient, the length $L'$ is the projection of $L$ onto the $x$-$z$ plane, i.e., $L' = L \sin \theta$, and $B'$ is the drop width along the vorticity direction instead of along the velocity gradient direction. According to Guido and Villone (1998), the differences between $L'$ and $L$ or $B'$ and $B$ are within 10% at $Ca=0.3$, and decrease with increasing $Ca$. Therefore, we will approximate $D'$ by $D$, where a larger $D'$ indicates a more slender drop. Figure 4-6 shows that $D'$ has a minimum at around $\lambda=0.5$. The drop becomes
more slender as $\lambda$ decreases. The deformation curve is similar to Grace's curve (1971) for critical drop deformation required for breakup, which shows a minimum in the range of $0.1<\lambda<1.0$.

![Deformation Parameter $D'$ vs Viscosity Ratio $\lambda'$](image_url)

Figure 4-6 Deformation parameter $D'$ of daughter drops resulting from necking mechanism (Figure 4-4) as a function of the viscosity ratio shows a minimum at $\lambda=0.5$

**SUMMARY**

At $Ca-Ca_c$, drops break by necking into two equally sized daughter drops for $0.0017<\lambda<3.5$. The $Ca$ in steady simple shear flow are slightly lower than results for pseudo-equilibrium breakup. The deformation parameters of the daughter drops have a minimum at around $\lambda=0.5$. The daughter drops become more slender as $\lambda$ decreases.

**4.3 Drop Breakup for $Ca<2.0Ca_c$**

**4.3.1 $0.1<\lambda<1.0$**

Drop breakup for $\lambda=0.12$ and $Ca=1.4Ca_c$ is shown in Figure 4-7 (I). Like the $\lambda=0.5$ case (Figure 4-1 (III) and 4-3(II)), end pinching is the dominant drop breakup mechanism. It
(I) $\lambda=0.12$ and $Ca=1.4Ca_c$ ($\dot{\gamma}=98.7\,s^{-1}, \, a=8.2\,\mu m$).

(a) 0ms  
(b) 60ms  
(c) 120ms  
(d) 200ms  
(e) 240ms  
(f) 260ms  
(g) 280ms  
(h) 300ms  
(i) 320ms  
(j) 340ms  
(k) 360ms

(II) $\lambda=1.0$ and $Ca=1.9Ca_c$ ($\dot{\gamma}=150\,s^{-1}, \, a=10.6\,\mu m$)

(a) 0ms  
(b) 40ms  
(c) 80ms  
(d) 120ms  
(e) 160ms  
(f) 200ms  
(g) 280ms  
(h) 320ms  
(i) 360ms  
(j) 400ms

(III) $\lambda=0.30$ and $Ca=1.6Ca_c$ ($\dot{\gamma}=145\,s^{-1}, \, a=10.6\,\mu m$)

(a) 0ms  
(b) 40ms  
(c) 80ms  
(d) 160ms  
(e) 240ms  
(f) 280ms  
(g) 300ms  
(h) 3320ms  
(i) 352ms  
(j) 328ms  
(k) 380ms  
(l) 420ms

50\mu m

Figure 4-7 End pinching for $0.1<\lambda<1.0$ and $Ca<2Ca_c$. 
repeats up to 4 times at each end of the thread until the whole thread is ruptured. The end pinched drops are uniform in size, except that the first pinched-off drops are about twice as large as the others. The distance between the pairs of pinched off drops at the two ends increases with time, which indicates that drops pinched off from opposite ends move in opposite directions. Thus, no drop collision is observed. The thread continues to stretch as end pinching occurs. This is different from end pinching in quiescent matrices, where the drop retracts during end pinching. Similar end pinching occurs at $\lambda = 0.30$ (Figure 4-7 (II)) and 1.0 (Figure 4-7 (III)). We start to see capillary wave on the middle part of the thread at $Ca = 1.6Ca_c$ for $\lambda = 0.30$ (Figure 4-7 (III), frame (k)).

4.3.2 $\lambda < 0.1$

Figure 4-8 shows end-pinning occurs up to around 4-5 times at each end of the thread at $\lambda = 0.075$, 0.030, 0.018, and 0.0017. As $\lambda$ decreases, the ends of the deformed drop become increasingly pointed, and the pinched off drops become more slender. No drop collision has been observed at $Ca < 2.0Ca_c$, because the end pinched drops move away from each other.

4.3.3 $1.0 < \lambda < 3.5$

End pinching at $1.0 < \lambda < 3.5$ (see Figure 4-9) is different from that for systems at lower $\lambda$. End pinching still occurs at the ends of the thread. However, it occurs only once to form a big drop at each end. The central part between the two end pinched drops is stretched into a long thin filament, which is broken by the capillary instability. The capillary instability occurs on the central thread even at fairly low $Cas$, e.g., Figure 4-9 (I) and (II) show drops with $1.3Ca_c$.
(I) $\lambda=0.075$ and $Ca=1.15Ca_c$ ($\dot{\gamma}=370\text{s}^{-1}$, $a=10.2\mu\text{m}$).

50$\mu\text{m}$

(II) $\lambda=0.018$ and $Ca=1.4Ca_c$ ($\dot{\gamma}=196\text{s}^{-1}$, $a=11.4\mu\text{m}$).

(III) $\lambda=0.030$ and $Ca=1.25Ca_c$ ($\dot{\gamma}=369\text{s}^{-1}$, $a=7.8\mu\text{m}$).

(IV) $\lambda=0.0017$ and $Ca=1.6Ca_c$ ($\dot{\gamma}=487\text{s}^{-1}$, $a=16\mu\text{m}$).

Figure 4-8 End pinching for $\lambda<0.1$ and $Ca<2Ca_c$. 
Figure 4-9 End pinching for $\lambda>1.0$ and $Ca<2Ca_c$. End pinching occurs only once. The central segment between the two end pinched drops is stretched into a long thin filament, which is broken by the capillary instability. The size of the pinched off drops is about 3 times larger than the daughter drops from the capillary instability.

and $1.6Ca_c$ respectively. The size of the pinched off drops is about 3 times larger than the daughter drops from the capillary instability.

**SUMMARY**

End pinching is the dominant drop breakup mechanism at $Ca<2.0Ca_c$. For $\lambda<1.0$, end pinching may occur up to 4-5 times depending on the length of the thread. The first end-pinched drop is about twice as large as the subsequent end-pinched drops. For $\lambda>1.0$, end pinching occurs only once. The capillary instability becomes dominant at capillary numbers as low as $1.3Ca_c$. 
4.4 Drop Breakup at $Ca > 2.0 Ca_c$

4.4.1 $0.1 < \lambda < 1.0$

Drop breakup processes for $\lambda = 0.12$, 0.30, and 1.0 (see Figure 4-10) are similar to those for $\lambda = 0.5$ and $Ca > 2.0 Ca_c$ (e.g. Figure 4-1 (IV) and (V)). The capillary instability is dominant on the central part of the thread, while end pinching occurs at the ends of the thread. As we will show quantitatively in Chapter 5, the thread radii are also independent of the initial drop size in each case. The wavelength of the capillary instability is uniform along the length of the thread and produce fairly monodisperse daughter drops uniformly distributed along the flow direction.

Figure 4-10 Drop breakup due to the capillary instability at $0.1 < \lambda < 1.0$ and $Ca > 2Ca_c$. 

$I$ $\lambda = 0.12$ and $Ca = 2.8 Ca_c$, $\dot{\gamma} = 98.7 s^{-1}$, $a = 16.2 \mu m$. 

(a) 0ms
(b) 40ms
(c) 280ms
(d) 320ms
(e) 360ms
(f) 400ms
(g) 440ms
(h) 480ms
(i) 600ms
(II) $\lambda = 0.3$ and $Ca = 2.6Ca_c$, $\dot{\gamma} = 145 \text{ s}^{-1}$, and $a = 17.2 \mu m$. Frames (a)-(g) correspond to 0, 40, 80, 160, 260, 420, 540 ms.

(III) $\lambda = 1.0$ and $Ca = 4.3Ca_c$, $\dot{\gamma} = 150 \text{ s}^{-1}$, and $a = 23.5 \mu m$. Frames (a)-(g) correspond to 0, 40, 80, 160, 920, 1080, and 1240 ms.

Figure 4-10 Drop breakup due to the capillary instability at $0.1 < \lambda < 1.0$ and $Ca > 2Ca_c$ (continued)

4.4.2 $1.0 < \lambda < 3.5$

Drop deformation and breakup at $\lambda = 2.1$ and 3.5 are shown in Figure 4-11. Similar to drop breakup for $0.1 < \lambda < 1.0$, the drop is stretched into a long thread, which is broken by the capillary instability. However, the wavelength is relatively longer than in the case of $0.1 < \lambda < 1.0$. The long filaments between the daughter drops form large satellite drops. The relative size of the satellite drops versus the daughter drops is much bigger than that for $0.1 < \lambda < 1.0$. The size of the satellite drops is no longer negligible, so the final drop size distribution is polydisperse, which we discuss in more detail in Chapter 5. No drop collision and re-breaking have been observed.
4.4.3 \( \lambda < 0.1 \)

For \( \lambda > 0.1 \) and \( Ca > 2Ca_c \), the capillary instability is the dominant mechanism of drop breakup. Once formed, the daughter drops retract to ellipsoidal shapes, and do not break again. In addition, the daughter drops are carried away from each other by the flow. Thus, no drop collision and coalescence have been observed under shear. For \( \lambda < 0.1 \), we observe two new features in the drop breakup mechanism. First, the wavelength of the capillary instability becomes very long, and the daughter drops from the capillary instability may break again. Second, the drop re-breaking induces drop collisions, which in turn leads to further drop re-
breaking or sometimes drop coalescence. This new re-breaking-collision-coalescence mechanism generates polydisperse drops.

**Re-breaking Mechanism**

Figure 4-12 illustrates this mechanism for $\lambda=0.018$ and $Ca=3.9Ca_c$. The drop is elongated into a long thread, which is then broken by the capillary instability with a very

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**Figure 4-12** Drop re-breaking and collision at $\lambda=0.018$ and $Ca=3.9Ca_c$ ($\dot{\gamma}=196s^{-1}$, $a=32.1\mu m$). Long wavelength capillary instability results in very slender daughter drops (e.g. Drop I in Frame (g)), which further breaks into smaller drops (II-1, II-2, and II-3 in Frame (i)). Drops II-1 and II-2 go through a similar drop re-breaking to form III-1, 2, 3, 4, and 5. Daughter drops form the capillary instability may collide with each other, for example III-2 and III-3 in frame (n). The collision induces irregular drop re-breaking generating nonuniform drops. The times corresponding to the frame (a) to (n) are (a) 0ms, (b) 40ms, (c) 120ms, (d) 160ms, (e) 640ms, (f) 680ms, (g) 720ms, (h) 740ms, (i) 760ms, (j) 800ms, (k) 820ms, (l) 840ms, (m) 860ms, (n) 888ms, (o) 960ms, (p) 1024ms, (q) 1040ms, (r) 1080ms.
long wavelength (see frame (g)). Instead of retracting to an ellipsoidal shape as for 
\(0.1 < \lambda < 1.0\), the daughter drop (labeled (l) in frame (g)) resulting from this long wavelength 
remains slender. It then further elongates and breaks again by a shorter wavelength instability 
into smaller drops labeled II-1, II-2, and II-3 (see frame (i)). Similarly, drop II-1 is broken 
again via even shorter wavelengths into drops III-1 and III-2, and drop II-2 is broken again 
into III-3, III-4, and III-5. The arrow in frame (f) marks the first observable breakup in the 
field of view, indicating that drop I may be part of an even longer daughter drop from an 
earlier generation that extends beyond the field of view.

The drops remain slender and close to each other after drop breakup. Some of the 
neighboring drops may approach each other, and then collide with each other, such as III-2 
and III-3 in the highlighted area of frame (n). The drop collision may force one or both of 
them to break irregularly into even smaller drops. The transient collision of drop III-2 and 
III-3 is enlarged in Figure 4-13. The arrows mark the front of drop III-3 from the overlapping 
images of drop III-2 and III-3. As a result of the collision, drop III-2 is irregularly broken 
into four smaller drops, IV-1, IV-2, IV-3, and IV-4. To differentiate the above breakup mode 
from re-breaking caused by the capillary instability, we call this type of drop re-breaking 
collision-induced-re-breaking. Note that the sizes of the four drops differ significantly from 
each other. Thus, we can expect that the final drop size distribution will be rather 
polydisperse, as demonstrated by frame (r) of Figure 4-12.

Why do the daughter drops re-break at \(\lambda < 0.1\)? Let us approximate a daughter drop by 
a liquid cylinder with radius \(R\) and length \(L\). The volume of the drop is then

\[
V = \pi R^2 L \tag{4-2}
\]
Figure 4.13 A detail of the collision of drops III-2 and III-3 from the highlighted area of Figure 4-12 n, o, and p. Collision of drop (III-2) and drop (III-3) results in the break of drop (III-2) into 4 smaller drops, IV-1, IV-2, IV-3, and IV-4, whose sizes are very different from each other. Frame (a), (d), and (h) correspond to frame (n), (o), and (p) of Figure 4-12.

If it retracts to a spherical drop, its equivalent radius will be,

\[ a_d = \left(\frac{3}{4} R^2 L \right)^{1/3} \]  \hspace{1cm} (4-3)

For drop (I) in Figure 4-13 (g), its equivalent drop radius is calculated to be 14.6\( \mu \)m, while the critical drop radius \( a_c \) corresponding to \( Ca_c \) is 8.3\( \mu \)m for \( \lambda = 0.018 \) at \( \dot{\gamma} = 196 \text{s}^{-1} \).

According to the definition of \( a_c \), drops larger than \( a_c \) are unstable and thus will break into drops smaller than \( a_c \). This is the underlying reason that daughter drops from long wavelengths, such as drop (I), will break again under steady shear. Similarly, the equivalent
drop radii of drops II-1, II-2, and II-3 are calculated to be 11.5µm, 10.9µm, and 8.1µm respectively. Accordingly, drop II-1 and II-2 will break again, and II-3 will not. This is exactly what happens to those three drops.

**Drop collision**

A second question we can ask is why drop collision occurs so frequently for λ<0.1? One factor is that the daughter drops are very close to each other due to their slenderness. However, two drops need to be moving toward each other for a collision to occur. Figure 4-14 shows how drop re-breaking induces a drop collision. Drop A and B are two neighboring daughter drops. They each break again into two daughter drops, A-1, A-2, and B-1, B-2, similar to necking mechanism shown in Figure 1-1 (I). Each pair of daughter drops is carried away in opposite directions by the flow, again similar to the necking mechanism (see frame (h) - (j) of Figure 1-1 (I)). The directions that the drops move in are marked by the arrows (frame (c)). Consequently, A-2 and B-1 approach each other and finally collide with each other. The two drops roll over each other and continue to move in their original direction (frame (e) and (f)). Therefore, drop collision appears to be a natural consequence of drop re-breaking.

Notice that both A-2 and B-1 accelerate as they separate from each other, *i.e.* drops B-1 and A-2 overtake drop A-1 and B-2, respectively, in 24ms after the separation of A-2 and B-1 (see frame (f) to frame (i)). Why do colliding drops accelerate as they separate from each other? Guido and Simeone (1998) experimentally studied drop collision by moving two drops toward each other under shear flow. They also observed that colliding drops accelerate as they separate from each other, and attributed it to the further separation of the drops in the
Figure 4-14 Drop re-breaking naturally leads to drop collision ($\lambda=0.075$ and $Ca=4.5Ca_c$, $\dot{\gamma}=505s^{-1}$, $a=29.7\mu m$). Drop A and B each break into 2 daughter drops, A-1/A-2 and B-1/B-2. Each pair of daughter drops moves in opposite directions as marked by the arrows (frame (c)). As a result, A-2 and B-1 collide with each other. The two drops both gain momentum and accelerate as they collide and then separate from each other, i.e. drops B-1 and A-2 overtake drops A-1 and B-2 in 24ms after the separation of A-2 and B-1 (see frame (f) to frame (i)). Drop A-1 is broken into 2 drops as drop B-1 collides with it and finally overtakes it (see frame (g) to (i)).

velocity gradient direction. For a steady simple shear, $\dot{\gamma}=dy/dt=constant$, thus $\Delta v_2$ increases as $\Delta y$ increases. Separation of drops in the velocity gradient direction was confirmed by Bartok and Mason (1959) and Guido and Simeone's experimental observations (1998) and Loewenberg and Hinch's numerical analysis (1996). An example is shown in Figure 2-21. Our observations are made along the velocity gradient direction, so we cannot measure how far apart the drops are in that direction. Nonetheless, we can get some idea from the focus of
the drops. When drop A-2 and B-1 first contact each other (Figure 4-14(c)), we see a bright circle around the edge of each drop, indicating they both are slightly out of focus. The thickness and brightness of the circle are similar, showing their positions in the velocity gradient direction are similar, i.e., \( y_{A,2} = y_{B,1} \). After the drops separate from each other (Figure 4-14(f)), drop B-1 becomes in focus (bright circle disappeared, drop edge became sharp and clear), and drop A-2 becomes more out of focus (bright circle become thicker and brighter). This means their distance must be enlarged in the velocity gradient direction during their collision.

Apparently, the accelerated drops tend to overtake other daughter drops along the same streamline and thus cause more drop collisions, which in turn will further accelerate even more drops. Figure 4-15 shows the collision frequency (number of collision in each 40ms interval) as a function of time. These data are measured from the transient breakup sequences shown in Figure 4-12. The x mark at \( t = 680 \)ms in Figure 4-15 corresponds to the thread breakup shown in frame (f) of Figure 4-12. From 680ms till around 950ms, no more

![Graph showing collision frequency over time](image)

**Figure 4-15** Collision frequency (number of collision in every 40ms) as a function of time for the breakup shown in Figure 4-12. The x mark in the plot corresponds to frame (f) of Figure 4-12 at \( t = 680 \)ms.
than one collision occurs every 40ms. Then the collision frequency suddenly jumps, and then
decreases before finally vanishing. The sudden increase of the collision frequency is due to
the self-accelerating nature of the collision process. However, the stretching effect of simple
shear flow eventually dominates and carry the drops further away from each other, thus the
collision frequency decreases.

Moreover, the high momentum of these accelerated drops is more likely to cause
collision induced re-breaking. For example, drop A-1 is broken into two drops as drop B-1
collides with it and finally overtakes it.

**Coalescence**

Sometimes, drop collision also leads to the coalescence of drops. Figure 4-16
illustrates some typical examples. The first frame is taken as time zero. Series (I) shows the
collision and coalescence of three drops C, D, and E at $\lambda=0.075$ and $\dot{\gamma}=505\text{s}^{-1}$. First, drop E
collides with drop D. The leading edge of drop E is shown by arrows in frames (b) and (c).
Drop D and E appear to meet head on. However, since our observations are made along the
velocity gradient direction, we do not know their actual locations in that direction. One drop
may lie slightly above the other drop. In frame (d), the interface between D and E
disappears, and drops D and E coalesce into one drop DE. Drop DE then immediately break
into two drops, DE-1 and DE-2. Next, drops C and DE-1 collide with each other. The arrows
in frame (f) to (i) show the leading edge of drop C advancing towards DE-1. Finally, the two
drops coalesce into one drop CDE (see Frame (j)). Drop CDE then breaks into two drops,
CDE-1 and CDE-2.
Figure 4-16 Drop Coalescence at λ=0.075 and 0.0017. (I) shows that drop coalenscence occurs even when drop D is undergoing breakup. (II) shows two drops coalesce into a drop larger than a*, which breaks up immediately after the coalescence.
Note that a neck forms in the middle of drop D even before the collision of D and E. The length and width of drop D are measured, and its equivalent radius is calculated to be 7.3 μm according to Equation 4-3. The corresponding capillary number is 0.59, which is higher than the critical capillary number \( Ca_c = 0.54 \) for \( \lambda = 0.075 \). Therefore, drop D is undergoing a transient breakup as it coalesces with drop E. This is to the best of our knowledge the first observation of concurrence of drop coalescence and breakup at a supercritical \( Ca \).

Figure 4-16 (II) shows another such example at \( \lambda = 0.0017 \) and \( \dot{\gamma} = 487 \text{s}^{-1} \). Two drops collide with each other as they disintegrate from their mother drops, and they coalesce into one drop (see frame (h)). The coalesced drop further elongates and eventually breaks into
two more or less equally sized drops, which are about the size of the mother drops. However, each drop consists of liquid from both of the two initial drops. Two small drops are shed during the collision and coalescence event (frame (g)). We should point out that there is no surfactant present in the system, so this is different from tip streaming, where surfactants or impurities accumulate at the pointed drop ends and small drops are ejected (De Bruijn, 1993). In fact, coalescence is normally inhibited when surfactants exist on the interface (Bartok and Mason, 1959). Also, the tiny drops are only shed once during the coalescence, whereas drops are continually ejected during tip streaming.

Figure 4-16 (III) shows another coalescence event at the same viscosity ratio and shear rate as Figure 4-16 (II). The two drops collide with each other sideways as seen from the velocity gradient direction. Frame (a) is the first frame after the drops enter the field of view, and we do not have data on their initial approach. Their contact area shrinks from frame (a) to (c), indicating they are moving away from instead of towards each other. The two drops coalesce into a flat "s" shape, which then realigns with the flow. As in Figure 4-16 (II), two small drops are shed during coalescence. We did not observe re-breaking of the coalesced drop, as the flow was stopped shortly after the coalescence event, and the drop retracted to a sphere. Figure 4-16 (IV) shows the coalescence of two drops with significantly different sizes. The small drop is sucked into the big one as it is moving away from the big drop. In general, the drops are more likely to coalesce as they separate from each other. In numerical simulations of drops with $0.25<\lambda<20$, Loewenberg and Hinch (1996) found that for drops with $Ca\sim O(1)$ and $\lambda\sim O(1)$ the tendency to coalesce is greatest when closely spaced drops are separated in the extensional quadrant of the flow field.
Qualitatively, we see more coalescence events at lower viscosity ratios. In particular, we see more coalescence at $\lambda=0.0017$ than at any other viscosity ratios. This is in agreement with Chesters' theoretical prediction (1991, see Chapter 2) that drops with low viscosity ratio are more likely to coalesce.

Effects of $Ca$ and $\lambda$

The re-breaking mechanism described above depends both on $Ca$ and $\lambda$. At $\lambda=0.12$, we have observed no drop re-breaking and collision at the highest attainable $Ca$ of the apparatus, $Ca=12Ca_c$. At $\lambda=0.075$ and $Ca=2.8Ca_c$ (see Figure 4-17 (I)), drop breakup is similar to that for $0.1<\lambda<1.0$. The capillary instability is the dominant drop breakup mechanism. The wavelength is uniform along the length of the thread, resulting in uniform daughter drops. The distance between daughter drops increases after drop breakup (compare (g) and (h)). No drop re-breaking and collision has been observed.

As $Ca$ increases to $4.5Ca_c$ (see Figure 4-17 (II)), the re-breaking mechanism becomes dominant. The drop is first broken by the capillary instability. The wavelengths become nonuniform along the length of the thread (see frame (g)), and some daughter drops formed from long wavelengths will break again by short wavelengths. For example, the detailed re-breaking and subsequent collision of drops (A) and (B) highlighted in Frame (g) have been shown in Figure 4-14. The re-breaking is followed by drop collision, which further induces irregular drop re-breaking and in some cases, drop coalescence. Figure 4-16 (I) has shown the coalescing of drop C, D, and E highlighted in frame (h). As a result of drop re-breaking, collision, and coalescence, the final drop sizes are less uniform than those for $Ca=2.8Ca_c$. 
Figure 4-17 Drop Breakup at $\lambda=0.075$ and $\dot{\gamma}=505s^{-1}$. (I) $Ca=2.8Ca_c$ ($a=18.4\mu m$). Drop breakup due to the capillary instability results in uniform daughter drops. The distance between daughter drops increases after drop breakup (compare (g) and (h)). No drop collision has been observed. Frames (a) through (h) correspond to 0ms, 20ms, 40ms, 80ms, 200ms, 240ms, 256ms, and 336ms. (II) $Ca=4.5Ca_c$ ($a=29.7\mu m$). The drop is first broken by the capillary instability. The wavelength is nonuniform along the length of the thread, and some daughter drops formed from long wavelengths will break again. The re-breaking of drops A and B highlighted in frame (g) is shown in Figure 4-14. The re-breaking is followed by drop collision, which further induces drop re-breaking and drop coalescence. The coalescence of drop C, D, and E in frame (h) is shown in Figure 4-16 (I). The final drop sizes for the initial drop size of $Ca=4.5Ca_c$ are less uniform than those of $Ca=2.8Ca_c$ (compare frame (j) of (II) with frame (g) of (I)). Frames (a) through (j) correspond to 0ms, 40ms, 120ms, 320ms, 400ms, 440ms, 460ms, 480ms, 528ms, and 640ms.

Figure 4-18 shows drop breakup at a lower viscosity ratio, $\lambda=0.030$. At $Ca=2.8Ca_c$ (see Figure 4-18(I)), daughter drops born of a long wavelength capillary instability break again. Few drop collisions occur afterwards. The final drops are more slender than those at $\lambda=0.075$. At $Ca=4.7Ca_c$ (see Figure 4-18(II)), daughter drops form the capillary instability
collide with each other right after the first breakup. Collisions occur much more frequently.

The final drop sizes are rather nonuniform.

(1) $Ca=2.8Ca_c$
(II) $Ca=4.7Ca_c$

Figure 4-18 Drop Breakup at $\lambda=0.030$ and $\gamma=369s^{-3}$. (I) $Ca=2.8Ca_c$ ($a=17.2\mu m$). Some daughter drops resulting from the long wavelength capillary instability break again. The final drops are somewhat nonuniform, and they are more slender than those for $\lambda=0.075$ (comparing with Figure 4-17 (I) (g)). Frames (a)-(g) correspond to 0, 40, 120, 160, 256, 264, and 300ms. (II) $Ca=4.7Ca_c$ ($a=29.3\mu m$). Daughter drops from the capillary instability collide with each other right after the first breakup. The number of collision event is much higher compared to that at $\lambda=0.075$ (Figure 4-17 (II)). Frames (a)-(j) correspond to 0, 20, 120, 360, 440, 480, 500, 520, 560, 600ms, respectively.

Figure 4-19 shows drop breakup at $\lambda=0.018$, $Ca=2.4Ca_c$. At $Ca=2.4Ca_c$, we see similar drop re-breaking as Figure 4-18 (I). Another case at $\lambda=0.018$ and $Ca=3.9Ca_c$ has been shown in Figure 4-12 as a typical example of the re-breaking mechanism. Figure 4-20 show similar phenomena at $\lambda=0.0017$, $Ca=1.7Ca_c$ and $2.4Ca_c$. 
Figure 4-19  Drop Breakup at $\lambda=0.018$ and $Ca=2.4Ca_c$.
($\phi=196s^{-1}$ and $a=19.9\mu m$).
Some daughter drops resulting from the long wavelength capillary instability break again.

Figure 4-20  Drop breakup for $\lambda=0.0017$ and $\phi=487s^{-1}$ (I) $Ca=1.7Ca_c$ ($a=17.2\mu m$). Some daughter drops resulting from long wavelength capillary instability break again. Frames (a)-(h) correspond to 0, 40, 120, 200, 320, 400, 440, and 480ms. (II) $Ca=2.4Ca_c$ ($a=24.2\mu m$). Daughter drops form the capillary instability collide with each other right after the first breakup. Collision-induced re-breaking results in nonuniform drops. Frames (a)-(m) correspond to 0, 120, 280, 440, 560, 600, 680, 720, 800, 880, 1000, and 1120ms.
The maximum collision frequencies \( f_{\text{max}} \) (number of collisions in 40 ms) for the above cases are measured and listed in Table 4-1. A clear trend is that at a fixed \( \lambda \) the maximum collision frequency increases as \( Ca \) increases. Comparing Figures 4-12, 4-17~4-20, we can see that the final drop size become more polydisperse at higher \( Ca \). This is due to more collisions occurs at higher \( Ca \), and collision induced drop re-breaking usually generates irregular sized drops.

<table>
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<th>( f_{\text{max}} )</th>
<th>( Ca )</th>
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<td>0</td>
<td>2.4</td>
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</table>

**SUMMARY**

We observe a new drop breakup mechanism at \( \lambda < 0.1 \) and \( Ca > 2.0 Ca_c \), which we call the re-breaking mechanism. Its features include:

(a) The drop is elongated into a long thread, which is broken by the capillary instability with very a long wavelength. The daughter drops are slender and they do not retract to ellipsoids. In addition, some of them may be larger than the critical drop size corresponding to the \( Ca_c \) and thus will further elongate and break again. The re-breaking may repeat multiple times.

(b) The drop re-breaking appears to initiate drop collision. Drop collision may cause drop re-breaking and less frequently, drop coalescence.

(c) Colliding drops accelerate as they separate from each other, causing more drop collisions and re-breaking. The collision frequency increases as \( Ca \) increases at fixed \( \lambda \).
(d) Qualitatively, more coalescence occurs at lower viscosity ratios. We presented the first observation of concurring drop breakup and coalescence for $Ca>Ca_c$.

(e) The final drops generated by this re-breaking mechanism are generally polydisperse, mainly because of the irregular sized drops formed during collision-induced-re-breaking.

4.5 Discussion

4.5.1 Scaling analysis of pseudo-steady state

Hinch and Acrivos (1980) argued that shear flow is different from extensional flow, because there is no internal pressure driven return flow. This is because the highly elongated drops align closely with the flow, thus the flux of fluid along the top surface is offset by an opposite flux from the bottom surface. Therefore, the internal pressure difference does not play an important role in simple shear flow; drop deformation and breakup at a fixed viscosity ratio are mainly governed by the viscous stress and the interfacial pressure. The viscous stress tends to deform the drop, while the interfacial pressure tends to resist the deformation. Initially, the viscous stress is far greater than the interfacial pressure, thus the drop is deformed into a liquid thread. Disturbances grow on the surface of the thread and are damped out by the flow. As the drop elongates, it rotates toward the flow direction. Finally, the thread almost aligns with the flow direction so that there is little driving force to further elongate the thread. Meanwhile, the interfacial pressure increases as the thread thins. Consequently, the two forces approach a balance between each other and the drop reaches a pseudo-steady state. Thus the amplitude of the capillary instability is able to grow as large as the radius of the thread resulting in breakup.
The viscous stress acting on a thread can be characterized by $\mu \dot{\gamma}$, and the interfacial pressure on the central part of the thread can be characterized by $\sigma/R_b$, where $R_b$ is the thread radius at breakup. Then at pseudo-steady state,

$$\mu \dot{\gamma} \sim \sigma/R_b$$

(4-4)

The thread radius at breakup is therefore determined by the balance of the two forces, which is independent of the initial drop size.

End pinching in flow is different from end pinching in a quiescent matrix (Stone et al. 1986). In the latter case, the viscous stress disappears after the flow stops, and end pinching is driven by interfacial tension. However, end pinching in flow depends on the balance of the viscous stress and the capillary pressure at the end of the thread. If we assume the tip of the thread has a hemispherical shape with a radius of $R_b$, the interfacial pressure on the tip is $2\sigma/R_b$, which is twice that on the central part of the thread. As the thread thins, it reaches a point where $\sigma/R_b < \mu \dot{\gamma} < 2\sigma/R_b$. This means on the central part of the thread, the viscous stress is higher than the interfacial pressure. This generates a flow from the center toward the end of the thread. On the other hand, at the ends, the interfacial pressure dominates the viscous stress, which generates a flow from the end toward the center. The combined effect of the two flows results in a neck forming between the end and the central part. A maximum pressure builds up at the neck, leads to further thinning of the neck, and eventually causes a drop to be pinched off from the end. End pinching starts when $\mu \dot{\gamma} \sim 2\sigma/R_b$, whereas the capillary instability begins when $\mu \dot{\gamma} \sim \sigma/R_b$ thus end pinching always occurs earlier than the capillary instability. For drop breakup in quiescent matrix, Stone et al. also found that end pinching occurs earlier than the capillary instability, and they attributed it to the shorter timescale of end pinching comparing to that of the capillary instability.
4.5.2 Re-breaking and slender body theory ($\lambda<<1$)

Our experimental results show that for $\lambda<<1$ the wavelength of capillary instability is very long, forming large daughter drops whose equivalent radii are larger than the critical drop radius $a_c$ corresponding to $Ca_c$. Thus they will break again until the drop radii are smaller than $a_c$. No existing theory explicitly explains such drop re-breaking. Hinch and Acrivos' slender body theory (1980) predicts that for $\lambda<<1$ in simple shear flow, the length scale of the least stable modes of disturbances is on the scale of the length of the thread. However, there is no quantitative prediction of the wavelength, nor any prediction of the re-breaking mechanism. However, let us do some further analysis on their theory.

The theory predicts that at high shear rates, the drop reaches a pseudo-equilibrium shape as a long cylinder with radius $R$ and length $L$ given by (note in their paper, Hinch and Acrivos used half-length)

$$\frac{R}{a\lambda^{1/6}} = \frac{0.0505}{G}$$

(4-5)

$$\frac{L}{2a\lambda^{1/5}} \approx 261G^2 + \frac{0.0231}{G}$$

(4-6)

where

$$G=Ca\lambda^{2/3}$$

(4-7)

and $a$ is the initial drop radius. The above equations are valid as $G \rightarrow \infty$. In practice, the length predicted by Equation 4-6 lies within 10% of the numerical solution for $G>0.05$. The theory further predicts that the pseudo-equilibrium shape is unstable to small disturbances when

$$G > G_c = 0.0541$$

(4-8)

Rearranging the above equation gives a prediction for

$$Ca_c = 0.0541 \lambda^{2/3}$$

(4-9)
which agrees with Grace's experimental data reasonably well as shown by Figure 2-7.

Next, we apply the slender body theory to calculate the daughter drop size $a_d$ and compare with the calculated critical drop size $a_c$. If $a_d > a_c$, the daughter drops are unstable and have to break again. Otherwise, the daughter drops are stable under shear. The theory predicted that the wavelength of the least stable mode is on the scale of the length of the thread. Here we first make a conservative assumption that the wavelength is an order of magnitude shorter than the length of the drop. Taking our system of $\lambda = 0.0017$ as an example, the known parameters are $\mu = 2.78 \text{Pa.s}$, $\sigma = 3.57 \times 10^3 \text{N/m}$, and $\gamma = 487 \text{s}^{-1}$. For a drop of radius $a = 25 \mu m$, the pseudo-equilibrium shape is calculated from Equations 4-5 and 4-6 to be $R = 3.23 \mu m$ and $L = 2066 \mu m$. Thus, the wavelength is $\omega = 1/10L = 206.6 \mu m$. If the resulting daughter drop retracts to a sphere, its equivalent radius is calculated by volume conservation (Equation 4-3) as $a_d = 11.7 \mu m$. The critical capillary number is calculated by Equation 4-9 to be $Ca_c = 3.8$, accordingly, $a_c = 10.0 \mu m$. Therefore, $a_d > a_c$, the daughter drop is unstable under this shear rate and doomed to break again. Therefore, the daughter drops have to re-break even when the wavelength is an order of magnitude shorter than the scale of the thread length. Apparently, daughter drops from longer wavelengths that are on the same scale of the thread length are doomed to break again.

The daughter drop is smaller than $a_c$ only when the wavelength is shorter than $1/16$ of the thread length. In our experiments, the wavelength observed in the field of view is about $1/2$ to $1/5$ of the length of the mother drop. If we assume the wavelength is half of the cylinder length, the breakup must repeat at least four times to satisfy $a_d < a_c$. That means multiple sequences of re-breaking are possible, depending on the length of the thread and the
scale of the wavelength. In conclusion, drop re-breaking can be inferred from the slender body theory.

4.5.3 Drop collision initiated by drop re-breaking ($\lambda<<1$)

Our experimental results show that drop re-breaking initiates the collision of drops (see Figure 4-14). Our observations were made along the velocity gradient direction, while a schematic diagram (Figure 4-21) in the vorticity direction offers more information, such as the orientation angle of the drop. Under steady shear, drop I reaches a pseudo-steady state that almost aligns with the flow direction. The orientation angle between the centerline of the drop and the flow direction is denoted as $\phi_o$, which is exaggerated in the diagrams. This pseudo-steady state is not stable for small disturbances (Hinch and Acrivos, 1980). For

Figure 4-21 Schematic diagram of drop re-breaking and collision at $\lambda<0.1$
simplicity, we assume that the dominant wavelength of the disturbances is half of the length of the thread. Thus, drop I breaks into two daughter drops, II-1 and II-2. We further assume that drop I initially lies right in the middle of the flow field. Drop II-2, which lies slightly above the central line of the flow field, is carried toward the right (marked by the arrow). On the contrary, drop II-1, which lies below the central line, moves in the opposite direction. This is consistent with our observation that the two daughter drops move away from each other after the drop is broken by necking (see Figure 4-1 (i)). When the mother drop is not in the middle of the flow field, it will move at a certain speed toward one direction. However, the two daughter drops still move in opposite directions, taking the initial speed of the mother drop as a reference.

After the first breakup, drops II-1 and II-2 realign with the flow direction, as shown by (b) and (c) of Figure 4-21. Hinch and Acrivos' slender body theory (1980) predicts that at pseudo-steady state the orientation angle is in the same scale as the aspect ratio of the drop (drop width divided by its length). Thus, the orientation angle of drop II-1 and II-2 is about $2\phi_0$, since their length is half that of their mother drop.

If the two drops are larger than $\alpha_c$, they will further elongate and then break again. Similarly, each pair of daughter drops II-1-1 and II-1-2, II-2-1 and II-2-2 move in opposite directions. Consequently, the two neighboring drops II-1-2 and II-2-1 move toward each other and collide. This is exactly what we observed in Figure 4-14, except that we were not able to see the orientation angle. The drops are tilted as they collide with each other, but it appears to be head on collision when observed from the velocity gradient direction. In addition, the realignment of the daughter drops and the drop elongation during each re-breaking increase the contact area of the two drops at collision. As Hinch and Acrivos' theory
(1980) pointed out, drop breakup in simple shear flow occurs due to the pseudo-steady state is not stable to small disturbances. It implies that a drop needs to reach the pseudo-steady state before disturbances cause it to break. Therefore, drop re-breaking always occurs after it has realigned with the flow.

If the four drops in (d) go through further re-breaking, it can be expected that more pairs of neighboring drops will move toward each other, leading to more collisions. Therefore, the collision frequency increases as the number of re-breaking sequences increases. If the drop breaks into more than two daughter drops in each breakup event, collision will still occur as long as re-breaking occurs, because neighboring second-generation daughter drops from different mother drops will move toward each other. In summary, drop collision is a natural result of drop re-breaking.

4.5.4 Drop coalescence ($\lambda<<1$)

**The drainage time**

Figure 4-16 (I) shows that it takes about 32ms (frame (a) to (d)) for drops D and E to coalesce into one drop, *i.e.*, the drainage time of the film between drop D and E is 32ms. Similarly, $t_{drain}$ of drop C and DE-1 is around 64ms (frame (a) to (j)). As mentioned in Section 2.8, several models have been developed for the film drainage between colliding drops, depending on different viscosity ratios (see Table 2-1). Next, the fully mobile interface (FMI) model is used to calculate the drainage time and compared with the experimental results. The FMI model is chosen for the following reasons:

1. The viscosity ratios of the emulsions in which coalescence occurs are very low, $\lambda<0.1$, while FMI model generally applies for low viscosity ratios.
(2) The immobile interface model and the partially mobile interface model require the interaction force between two colliding drops, $F$. However, $F$ for deformable drops is currently unavailable (Chesters, 1991).

We are not able to precisely measure the film thickness during the collision and coalescence. However, we can calculate the drainage time according to Equations 2-39(b) with some approximations. Equation 2-39(b) is insensitive to $h_o$. Following Janssen (1997), we approximate $h_o$ with the initial drop radius $a$. The critical film thickness can be estimated by Equation 2-36, using a Hamaker constant of $10^{-20}J$.

The physical properties of the systems and the shear rate are $\lambda=0.075$, $\mu=0.675$Pa.s, $\mu_r=0.0505$Pa.s, $\dot{\gamma}=505s^{-1}$, and $\sigma=4.2\times10^{-3}$N/m. The width and length of drop C, D, and E in Figure 4-16 are measured, and their equivalent radii are estimated as 6.4$\mu$m, 7.3$\mu$m, and 5.6$\mu$m, respectively. For simplification, we take the average as the radius for all the three drops, i.e., $a=6.4$ $\mu$m, accordingly, $Ca=0.519$. The critical film thickness is estimated by Equation 2-36, $h_{crit}=8.5\times10^{-9}$m. The drainage time calculated from the FMI model (Equation 2-39b) is 11ms, which is in the same order of magnitude as the experimental measurements, 32ms and 64ms.

In general, the FMI is applicable when $\lambda<6h/r$ (see Table 2-1), where $r$ and $h$ are the radius and thickness of the circular film between the colliding drops. For drops with $Ca=O(1)$, $r$ can be approximated by Equation 2-48

$$r=Ca^{1/2}=0.72a$$

$h$ can be approximated by $h_{crit}$, since the final stage of drainage is typically rate limiting (Chester, 1991). Hence,

$$6h/r=0.011$$
Therefore, $\lambda=0.075$ is higher than but fairly close to $6h/r=0.011$, which means the FMI model is approximately applicable.

Similarly, we can calculate the drainage time for the two drops in Figure 4-16 (II). The physical properties and the shear rates are $\lambda=0.0017$, $\mu=2.78\text{Pa.s}$, $\mu_e=4.6\times10^{-3}\text{Pa.s}$, $\dot{\gamma}=487\text{s}^{-1}$, $\sigma=3.57\times10^{-3}\text{N/m}$. The equivalent drop radii of the two drops are estimated to be $8\mu\text{m}$, and the corresponding $Ca=3.03$. The critical film thickness is estimated to be $h_{\text{crit}}=9.6\times10^{-9}\text{m}$. According to Equation (2-49), $r\sim a=8\mu\text{m}$.

Thus,

$\lambda=0.0017<6h/r=7.2\times10^{-3}$

indicating that the FMI model best describes the film drainage for this case. The drainage time is calculated to be $t_{\text{drain}}=62\text{ms}$, which is on the same order of magnitude as the experimental measurement of $128\text{ms}$ (from frame (a) to (h) of Figure 4-16 (II)). The drainage time for the case of Figure 4-16 (IV) is around $200\text{ms}$ (from frame (a) to (g)), which is also in the similar scale, even though one of the drops is much smaller than the other. The drainage time of case (III) of Figure 4-16 is unknown, because data of their initial approach are unavailable. In conclusion, the FMI model gives reasonable estimates of the drainage time for highly deformable drops ($Ca=O(1))$ for the two systems $\lambda=0.075$ and 0.0017.

The contact time

In general, drop coalescence occurs only when the interaction time, $t_i$ exceeds the required drainage time. Our experiment indicates that $t_i \geq t_{\text{drain}}=32\text{ms}$ for the case of $\lambda=0.075$ and $t_i \geq t_{\text{drain}}=128\text{ms}$ for the case of $\lambda=0.0017$. Chester (1991) proposed that $t_i$ can be scaled by $\dot{\gamma}^{-1}$. However, in the above two cases, $\dot{\gamma}=500\text{s}^{-1}$, thus $t_i \sim 0.002\text{s}=2\text{ms}$, which is far less
than the corresponding $t_{\text{drain}}$. As realized by Chester (1991), $t \sim \dot{\gamma}^{-1}$ is valid for solid particles, and it is expected to be a weak function of the viscosity ratio for deformable drops. Our experimental results suggest that $t \sim \dot{\gamma}^{-1}$ is likely to underestimate the interaction time of highly deformable drops at very low viscosity ratios. In contrast to Chesters' assumption of $r \ll a$ (1991), Loewenberg and Hinch's numerical simulations (1996) and Guido and Simeone's experimental results (1998) show that the radius of the contacting area of highly deformable drops is on the scale of the initial drop radius, i.e. $r \sim a$. It is very likely that the large contacting area of the highly deformable drops hinders them from rolling over each other, prolonging their contact time.

**Simultaneous coalescence and breakup**

Our experimental observation (Figure 4-16) shows that coalescence is possible even when drops are undergoing breakup. Chesters (1991) argued that coalescence occurs at much smaller $Ca$ than breakup. However, his argument was based on assumptions derived from solid particles, such as $r \ll a$. Janssen's theoretical analysis (1995) suggested that drop coalescence is possible in the breakup region. By indirectly determining the average drop size under steady shear from rheological measurements, Minale et al. (1997) verified Janssen's analysis. Our direct observation not only verifies that coalescence is possible in the breakup region, but also shows that the same drops undergo coalescence and breakup at the same time. This is the first direct observation of concurring coalescence and breakup.

**4.5.5 Final drop size distribution as a function of $Ca$ for $\lambda<0.1$**

We have shown that the final drops from a single drop become more polydisperse at higher $Ca$ for $\lambda<0.1$ (e.g. compare the final drop size in Figure 4-12 (r) and 4-19 (h)). Our
results also show that the collision frequency increases with $Ca$. Collision induced drop re-breaking usually forms irregularly sized drops. Therefore, the final drop sizes become more polydisperse at higher $Ca$. Grace (1971) found that final drop size distribution broadens as $Ca/Ca_c$ increases at $\lambda=3.99 \times 10^{-3}$, but he did not mention what the drop breakup mechanism was and why final drop size distribution broadened.

4.5.6 Final drop size distribution as a function of $\lambda$

We found that daughter drops from the capillary instability do not re-break for $\lambda>0.1$, but may break again for $\lambda<0.1$. Tjahjadi and Ottino (1991) studied drop breakup in chaotic flow, and found that drops for systems $\lambda>1$ extend $O(10^3 \sim 10^4)$ times their original length before they break, and the resulting daughter drops rarely break again. By comparison, drops for systems with $\lambda<1$ break at relatively low stretch rates ($O(10^1 \sim 10^2)$), resulting in large drops that may break again. However, the re-breaking in Tjahjadi’s system may be due to the variation of the shear strength at different locations in the chaotic flow. They also found that a succession of breakup events at low viscosity ratios results in more uniform drop distributions. In simple shear flow, however, we find that re-breaking generates much more nonuniform drops for systems with $\lambda<0.1$. We present more analysis on drop size distribution in Chapter 5.

4.5.7 Satellite and sub-satellite drops

For viscosity ratios $0.0017<\lambda<3.5$, our results (see Figure 4-10 to 4-25) show that the size of satellite drops generally decreases as $\lambda$ decreases. In particular, the relative size of the satellite drops to the daughter drops is the largest at $\lambda=3.5$, and no satellite drops are visible
at $\lambda=0.018$ and 0.0017. Tjahjadi et al. (1992) studied both experimentally and theoretically the formation of satellite and sub-satellite drops from the breakup of a long liquid thread in a quiescent matrix. They found that satellite drops are formed by multiple breakup sequences around the neck region of a thread. The relative size and the number of satellite drops are determined by the viscosity ratio. For $0.01<\lambda<2.8$, they found that the relative size of the two largest satellites (compared to their mother drops) are bigger for $\lambda>0.4$, and the largest satellites occur at $\lambda=1.0$.

In our experiments on drop breakup due to the capillary instability under shear, satellite and sub-satellite drops form from thin filaments between the daughter drops. The filament is generally about an order of magnitude thinner than the thread width at breakup (see for example Figure 4-10 (III) (e) and (f)). As mentioned previously, the thread reaches a pseudo-steady state before the capillary instability causes the breakup, whereas the viscous stress $\mu \dot{\gamma}$ and capillary pressure $\sigma R_b$ balance each other. Thus, for a filament that is an order of magnitude thinner than $R_b$, the viscous stress is dominated by the capillary pressure. The formation of satellite drops from these very thin filaments in flow approximates the formation of satellite drops in a quiescent matrix driven by interfacial tension. We find that in general the size of satellite drops increases as the viscosity increases. The trend is similar to what Tjahjadi et al. (1992) found in quiescent breakup, except we do not find a maximum at $\lambda\sim 1.0$. This may be due to the fact that the highest viscosity ratio Tjahjadi et al. studied ($\lambda=2.8$) is relatively low. Tjahjadi et al. (1992) also found that the number of all visible satellite and sub-satellite drops increases as $\lambda$ decreases. In our case, sub-satellites after the first two generations are too small to see.
4.5.8 Map of factors influencing drop breakup

We summarize the different drop breakup modes as a function of \( \lambda \) and \( Ca \) in Figure 4-22.

![Diagram showing the relationship between viscosity ratio \( \lambda \) and capillary number \( Ca \) for drop breakup modes]

Figure 4-22 Drop breakup mechanism as a function of \( \lambda \) and \( Ca \).

At \( Ca \sim Ca_c \), drops are broken by necking into two equally sized daughter drops. For \( Ca < 2.0Ca_c \), end pinching is the dominant drop breakup mechanism.

For \( Ca > 2.0Ca_c \), the capillary instability is the dominant drop breakup mechanism. At \( 0.1 < \lambda < 1.0 \), the thread width at breakup is uniform. The wavelength is also uniform along the length of the thread and from thread to thread. This mechanism may result in the production of monodisperse emulsions under shear. At \( 1.0 < \lambda < 3.5 \), large satellite drops lead to a polydisperse drop size distribution. At \( \lambda < 0.1 \), daughter drops from long wavelength capillary instability break again. The drop re-breaking induces drop collision, which further causes
irregular drop re-breaking or drop coalescence. This re-breaking mechanism will result in a polydisperse drop size distribution.
Chapter 5 Thread Radius at Breakup, Dominant Wavelength, and Final drop size

A principal goal of research on drop deformation and breakup is to control the final drop size and distribution (Rallison, 1984). As demonstrated in Chapter 4, the final drop sizes are determined by the drop breakup mechanism, which in turn depends on the capillary number and the viscosity ratio. Qualitatively, fairly uniform drops are obtained for $0.1 < \lambda < 1.0$ due to the size selection of the thread radius and the wavelength at breakup. Conversely, polydisperse drops are generated for $\lambda > 1.0$ owing to the relatively large size of satellite drops; and for $\lambda < 0.1$, because of the re-breaking, collision and coalescence of daughter drops. In this chapter, we will present quantitative results for the thread radius at breakup, the dominant wavelength, and the final drop size. We will also discuss the time for a thread to breakup and the pseudo-affine nature of drop deformation.

5.1 Thread radius at Breakup, $R_b$

We showed in Chapter 4 that the capillary instability is the dominant mechanism of drop breakup at $Ca > 2.0 Ca_c$, and that the thread radius at breakup does not depend on the initial drop size for one emulsion system of $\lambda = 0.5$ at $\dot{\gamma} = 323 \text{s}^{-1}$ (see Figure 4-2). At a fixed viscosity ratio, the thread radius at breakup is governed by the balance of viscous stress ($\mu \dot{\gamma}$) and the interfacial pressure ($\sigma / R_b$) of the elongated drop. Therefore, $R_b$ is independent of the initial drop size. To characterize the balance of the two forces, we define a dimensionless thread number as

$$Th = \frac{\mu \dot{\gamma} R_b}{\sigma}$$

(5-1)
In this section, we first fix the viscosity ratio at $\lambda=0.5$ and investigate the dependence of $R_b$ on the shear rate, the outer phase viscosity, and the interfacial tension by correlating these parameters with the thread number. Next, we present results for the thread number as a function of the viscosity ratio.

5.1.1 $\lambda=0.5$

(a) Effect of shear rate

We keep all the other parameters the same by shearing the same emulsion sample (0.2% 337mPa.s silicone oil in 675mPa.s castor oil) at a variety of shear rates, 132s$^{-1}$, 197s$^{-1}$, 323s$^{-1}$, and 495s$^{-1}$. At each shear rate, we collect $R_b$ of 40 threads. The polydispersity of each case is below or just above 10% (see Table 5-1). Figure 5-1 shows that the thread radius at breakup is linearly proportional to the reciprocal of the shear rate. As a result, the thread number is a constant of about 0.13 for the different shear rates, as listed in Table 5-1.

![Figure 5-1](image)

Figure 5-1 Thread radius at breakup versus inverse shear rate for $\lambda=0.5$ shows that thread radius at breakup is inversely proportional to the shear rate. The emulsion is 0.2% 337mPa.s silicone oil in 675mPa.s castor oil. The circles are average thread radius of 40 threads. The dashed line is the best linear fit.
Tsakalos et al. (1998) also found that the thread width at breakup is inversely proportional to the shear rate for a viscoelastic emulsion consisting of drops of isotropic hydroxypropylcellulose solution suspended in a polydimethylsiloxane (PDMS, or silicone oil) matrix at a viscosity ratio of 0.70-0.85.

<table>
<thead>
<tr>
<th>Inner Phase</th>
<th>Outer Phase</th>
<th>λ</th>
<th>σ</th>
<th>Shear rate</th>
<th>( R_b )</th>
<th>Polydispersity</th>
<th>( T_h )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silicone oil</td>
<td>Castor oil</td>
<td>337/675=0.50</td>
<td>4.2</td>
<td>495</td>
<td>1.93</td>
<td>0.146</td>
<td>0.153±0.016</td>
</tr>
<tr>
<td>Silicone oil</td>
<td>Castor oil</td>
<td>337/675=0.50</td>
<td>4.2</td>
<td>323</td>
<td>2.66</td>
<td>0.066</td>
<td>0.138±0.011</td>
</tr>
<tr>
<td>Silicone oil</td>
<td>Castor oil</td>
<td>337/675=0.50</td>
<td>4.2</td>
<td>197</td>
<td>3.8</td>
<td>0.092</td>
<td>0.120±0.008</td>
</tr>
<tr>
<td>Silicone oil</td>
<td>Castor oil</td>
<td>337/675=0.50</td>
<td>4.2</td>
<td>132</td>
<td>6.1</td>
<td>0.063</td>
<td>0.129±0.006</td>
</tr>
<tr>
<td>Silicone oil</td>
<td>OCO-XY</td>
<td>830/1675=0.50</td>
<td>5.0</td>
<td>114</td>
<td>3.11</td>
<td>0.062</td>
<td>0.119±0.008</td>
</tr>
<tr>
<td>Silicone oil</td>
<td>OCO-Z1</td>
<td>1222/2780=0.44</td>
<td>4.85</td>
<td>94.9</td>
<td>2.32</td>
<td>0.111</td>
<td>0.126±0.011</td>
</tr>
<tr>
<td>Silicone oil</td>
<td>Corn syrup</td>
<td>5842/8411=0.69</td>
<td>39.8</td>
<td>328</td>
<td>1.9</td>
<td>0.13</td>
<td>0.130±0.016</td>
</tr>
<tr>
<td>Silicone oil</td>
<td>Glycerol</td>
<td>502/855=0.59</td>
<td>24.7</td>
<td>718</td>
<td>3.0</td>
<td>0.073</td>
<td>0.150±0.006</td>
</tr>
</tbody>
</table>

**(b) Effect of the outer phase viscosity**

We change the outer phase viscosity while keeping all the other parameters constant. This is done by using oxidized castor oil with different degrees of oxidization. Generally, viscosity increases with the degree of oxidation. The viscosities of the two oxidized Caster oil samples are 1675mPa.s (OCO-XY) and 2780mPa.s (OCO-Z1), respectively. The inner phase viscosity is adjusted by mixing silicone oils of two different viscosities to keep the viscosity ratio at around 0.5. The interfacial tension of the two samples (OXY-XY and OCO-Z1) are 5.0 mN/m and 4.85mN/m, respectively, which are reasonably close to that of 337mPa.s silicone oil/675mPa.s castor oil, \( \sigma =4.2 \) mN/m. There is no surfactant in any of the emulsions. The mean thread radius, the polydispersity, and the corresponding thread number from 40 threads for each sample are listed in Table 5-1. The thread numbers in both cases are close to those of 337mPa.s silicone oil/675mPa.s castor oil.
(c) Effect of the interfacial tension

Newtonian systems with significantly different interfacial tension, such as silicone oil in maltose syrup ($\sigma=39.8\text{mN/m}$) and silicone oil in glycerine ($\sigma=24.7\text{mN/m}$) are used. The viscosity of the silicone oil is adjusted to keep the viscosity ratio at around 0.5. The thread numbers of the two systems (see Table 5-1) are similar to those of the previous cases.

SUMMARY

The thread radius at breakup does not depend on the initial drop radius. The thread number is approximately a constant of 0.13 at a fixed viscosity ratio $\lambda=0.5$, and is independent of the outer phase viscosity, the shear rate, and the interfacial tension.

5.1.2 Results from the previous shear cell (Zhao and Goveas, 2001)

All the above experiments are done by using the improved shear cell (see Figure 3-3), where the top plate and the bottom plate of the cell move in opposite direction. This setup enables us to follow the whole history of deformation and breakup of drops. Previously, we have studied two emulsions by using a previous shear cell (see Figure 3-1), where the top plate moved while the bottom plate was stationary. In that case, drops flowed in and out of the field of view. We could only view a partial history of a particular drop. Nonetheless, because the deforming threads are very long, we normally could follow an already substantially deformed drop up to its breakup. Thus, we could measure the thread width at breakup for drops that happened to be in the field of view while breaking.

The two emulsions that we studied using this old shear cell are: a purely Newtonian emulsion (NE) and a viscoelastic emulsion (VE). The dispersed phase in both emulsions was
338mPa.s silicone oil, which is Newtonian. The continuous phase of NE was 2wt% of the nonionic surfactant TWEEN-80 in glycerine, which is a Newtonian liquid with a viscosity of 747mPa.s. The continuous phase of VE consisted of an aqueous solution of 2wt% sodium dodecyl sulfate (SDS, an ionic surfactant) and 12wt% polyvinyl pyrrolidone (PVP). This is a viscoelastic shear-thinning solution (see Figure 3-4). The emulsion is sheared at 750s⁻¹, where the continuous phase has a viscosity of 701mPa.s.

The deformation process for the NE and VE were compared by shearing both emulsions at fixed viscosity ratio and external stresses. Both experiments were performed at a shear rate of 750s⁻¹ in a 100-micron gap. At this shear rate, the continuous phase of the VE emulsion had a viscosity of 701mPa.s. The viscosity ratios of the NE and VE emulsions were thus 0.45 and 0.48, respectively. In the VE case, we observed similar deformation and breakup mechanisms as the capillary instability at λ=0.5 shown in Chapter 4. The thread width at breakup was almost the same for all the drops (see Figure 5-2). The wavelength was more or less uniform along the thread length, resulting in uniform daughter drops. We did not know the exact initial sizes of the drops, because we were not able to follow the motion of a single drop. However, it was clear that the threads originated from different-sized mother drops, because they had similar radii but different lengths. Conversely, we observed a variety of thread widths at breakup in the NE case (see Figure 5-3). The wavelength was also nonuniform along the thread length, generating polydisperse drops.

To quantify the narrowing of the thread width distribution for the VE, we measured $R_b$ of 40 threads. For both emulsions, drops did not re-break in the flow. Figure 5-4 shows that the polydispersity for the NE was 3 times as large as that of the VE case.
Figure 5-2 (a) Pre-sheared viscoelastic emulsion containing 2wt% silicone oil in an aqueous solution of 12wt% PVP and 2wt% SDS, which corresponds to a viscosity ratio of 0.48. (b) Viscoelastic emulsion under shear, showing that threads have similar width at breakup. The wavelength is uniform along the thread length, resulting in uniform daughter drops.

Figure 5-3 (a) Pre-sheared Newtonian emulsion containing 2wt% silicone oil in glycerine with 2wt% TWEEN-80, which corresponds to a viscosity ratio of 0.45. (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 thread length.

We have sheared both the VE and NE using the improved shear cell, and observed similar results, i.e., uniform thread width at breakup in the VE, but polydisperse thread width at breakup in the NE. Initially, we attributed the difference to the viscoelasticity of the VE. However, as shown by the Newtonian emulsions listed in Table 5-1, we have obtained uniform thread widths at breakup in various Newtonian emulsions without any surfactant. In particular, the thread width at breakup is also uniform for silicone oil in pure glycerine with
no surfactant. This convinces us that the nonuniformity of thread width at breakup in the NE case was due to surfactant effect. However, we have not proved whether this is due to the surfactant itself or due to the interaction of the surfactant with glycerine. We also do not know whether these results are particular to the TWEEN-80 surfactant or whether all surfactants will behave the same manner.

5.1.3 Effects of $\lambda$ on $R_b$

The thread radius at breakup vs. the initial drop radius at a variety of viscosity ratios between $10^{-3}$ and 3.5 is shown in Figure 5-5. The emulsion and its physical properties corresponding to each $\lambda$ are listed in Table 3-1. The shear rate is adjusted so that the thread radius at breakup is in the range of 2-4$\mu$m. The error of $R_b$ is larger than 10% at an $R_b$ thinner than 2$\mu$m. An $R_b$ thicker than 4$\mu$m requires drops with $a > 35 \mu$m, for which the confinement effect of the shear cell may be significant. The shear rate is not a constant for all the cases. However, we have showed that the thread radius at breakup is inversely proportional to the shear rate, and that the thread number is independent of the shear rate at $\lambda = 0.5$ (see Figure
\[ \lambda = 0.0017, \ \dot{\gamma} = 487 \text{s}^{-1}, \ \bar{R}_e = 3.74 \mu\text{m}, \ \text{and} \ p = 10.4\% \]

\[ \lambda = 0.075, \ \dot{\gamma} = 370 \text{s}^{-1}, \ \bar{R}_e = 3.43 \mu\text{m}, \ \text{and} \ p = 8.4\% \]

\[ \lambda = 0.018, \ \dot{\gamma} = 196 \text{s}^{-1}, \ \bar{R}_e = 3.93 \mu\text{m}, \ \text{and} \ p = 13.6\% \]

\[ \lambda = 0.12, \ \dot{\gamma} = 99 \text{s}^{-1}, \ \bar{R}_e = 2.83 \mu\text{m}, \ \text{and} \ p = 9.6\% \]

\[ \lambda = 0.030, \ \dot{\gamma} = 369 \text{s}^{-1}, \ \bar{R}_e = 3.02 \mu\text{m}, \ \text{and} \ p = 14.8\% \]

\[ \lambda = 0.30, \ \dot{\gamma} = 145 \text{s}^{-1}, \ \bar{R}_e = 2.82 \mu\text{m}, \ \text{and} \ p = 9.8\% \]

Figure 5-5 The thread radius at breakup vs. the initial drop radius for viscosity ratio between 0.0017 and 3.5. The scale of x-axis is 10-35\( \mu\text{m} \) for all the figures, and the y-axis is scaled as twice of the mean thread radius at breakup (\( \bar{R}_e \)) for each case (to be continued on the next page).
Figure 5-5 The thread radius at breakup vs. initial drop radius for viscosity ratio between 0.0017 and 3.5. The scale of x-axis is 10-35µm for all the figures, and the y-axis is scaled as twice of the mean thread radius at breakup ($\overline{R}_b$) for each case.

5-1). It is further confirmed by the thread radius at breakup for $\lambda=0.075$ at $\dot{\gamma}=200$ s$^{-1}$, 370 s$^{-1}$, and 500 s$^{-1}$. The polydispersity of $R_b$ (see Table 5-2) is close to 10% for all viscosity ratios, which means that the thread radius at breakup is nearly monodisperse.

The thread numbers are plotted against the viscosity ratios in Figure 5-6. The circles are our experimental results corresponding to the average of the thread radius at breakup shown in Figure 5-5. The thread number is almost a constant around 0.13 for viscosity ratios between 0.1 and 1.0, and it increases as the viscosity ratio decreases from 0.1 or increases from 1.0.
Table 5-2 Summary of experimental results of critical drop size, thread radius at breakup, wavelength at breakup, and final drop size and distribution as a function of $\lambda$ for 0.0017$<\lambda<$3.5

<table>
<thead>
<tr>
<th></th>
<th>$\lambda$</th>
<th>0.0017</th>
<th>0.018</th>
<th>0.030</th>
<th>0.075</th>
<th>0.12</th>
<th>0.30</th>
<th>0.50</th>
<th>1.0</th>
<th>2.1</th>
<th>3.5</th>
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<tbody>
<tr>
<td>Physical properties &amp; shear rate</td>
<td>$\sigma$ (mN/m)</td>
<td>3.57</td>
<td>4.85</td>
<td>5.0</td>
<td>4.2</td>
<td>4.85</td>
<td>5.0</td>
<td>4.2</td>
<td>5.0</td>
<td>4.85</td>
<td>4.85</td>
</tr>
<tr>
<td></td>
<td>$\mu$ (mPa.s)</td>
<td>2780</td>
<td>2780</td>
<td>1675</td>
<td>675</td>
<td>2780</td>
<td>1675</td>
<td>675</td>
<td>1675</td>
<td>2780</td>
<td>2780</td>
</tr>
<tr>
<td></td>
<td>$\dot{\gamma}$ (s$^{-1}$)</td>
<td>487</td>
<td>196</td>
<td>369</td>
<td>370</td>
<td>99</td>
<td>145</td>
<td>323</td>
<td>151</td>
<td>149</td>
<td>197</td>
</tr>
<tr>
<td>Critical drop size</td>
<td>$a_c$ (µm)</td>
<td>10.2</td>
<td>8.25</td>
<td>6.25</td>
<td>9.10</td>
<td>5.85</td>
<td>6.70</td>
<td>7.40</td>
<td>5.50</td>
<td>5.72</td>
<td>6.72</td>
</tr>
<tr>
<td></td>
<td>$Ca_c$</td>
<td>3.87</td>
<td>0.93</td>
<td>0.77</td>
<td>0.54</td>
<td>0.33</td>
<td>0.32</td>
<td>0.38</td>
<td>0.28</td>
<td>0.49</td>
<td>0.76</td>
</tr>
<tr>
<td>Thread radius at breakup</td>
<td>$R_b$ (µm)</td>
<td>3.74</td>
<td>3.93</td>
<td>3.02</td>
<td>3.43</td>
<td>2.83</td>
<td>2.82</td>
<td>2.66</td>
<td>2.20</td>
<td>1.95</td>
<td>2.57</td>
</tr>
<tr>
<td></td>
<td>$p(R_b)$ /%*</td>
<td>10.4</td>
<td>13.6</td>
<td>14.8</td>
<td>8.4</td>
<td>9.6</td>
<td>9.8</td>
<td>6.6</td>
<td>12.4</td>
<td>14.6</td>
<td>11.9</td>
</tr>
<tr>
<td></td>
<td>$Th$</td>
<td>1.42</td>
<td>0.44</td>
<td>0.37</td>
<td>0.20</td>
<td>0.16</td>
<td>0.14</td>
<td>0.14</td>
<td>0.11</td>
<td>0.17</td>
<td>0.29</td>
</tr>
<tr>
<td></td>
<td>$a_d/R_b$ (=$Ca_d/Th$)</td>
<td>2.73</td>
<td>2.10</td>
<td>2.07</td>
<td>2.65</td>
<td>2.07</td>
<td>2.38</td>
<td>2.78</td>
<td>2.50</td>
<td>2.93</td>
<td>2.61</td>
</tr>
<tr>
<td>Wavelength at breakup</td>
<td>$k$</td>
<td>13.1</td>
<td>8.2</td>
<td>7.6</td>
<td>6.9</td>
<td>6.7</td>
<td>5.7</td>
<td>6.3</td>
<td>6.4</td>
<td>7.4</td>
<td>11.5</td>
</tr>
<tr>
<td></td>
<td>$p(k)$ /%</td>
<td>26.5</td>
<td>20.8</td>
<td>20.6</td>
<td>15.0</td>
<td>14.5</td>
<td>15.7</td>
<td>17.1</td>
<td>14.8</td>
<td>21.1</td>
<td>25.4</td>
</tr>
<tr>
<td>Final drop size and distribution</td>
<td>$a_t$ (µm)</td>
<td>4.69</td>
<td>5.7</td>
<td>4.05</td>
<td>7.75</td>
<td>5.68</td>
<td>5.24</td>
<td>5.35</td>
<td>4.47</td>
<td>3.76</td>
<td>4.33</td>
</tr>
<tr>
<td></td>
<td>$p(a_t)$ /%</td>
<td>32.1</td>
<td>27.0</td>
<td>26.0</td>
<td>16.5</td>
<td>10.9</td>
<td>13.6</td>
<td>12.6</td>
<td>12.9</td>
<td>16.6</td>
<td>34.8</td>
</tr>
<tr>
<td></td>
<td># of drops</td>
<td>132</td>
<td>30</td>
<td>327</td>
<td>59</td>
<td>83</td>
<td>96</td>
<td>134</td>
<td>67</td>
<td>163</td>
<td>164</td>
</tr>
<tr>
<td></td>
<td>$a_t/a_c$</td>
<td>0.46</td>
<td>0.69</td>
<td>0.65</td>
<td>0.85</td>
<td>0.97</td>
<td>0.78</td>
<td>0.72</td>
<td>0.81</td>
<td>0.66</td>
<td>0.64</td>
</tr>
<tr>
<td></td>
<td>$a_n$ cal (µm)**</td>
<td>10.1</td>
<td>9.08</td>
<td>6.80</td>
<td>7.47</td>
<td>6.11</td>
<td>5.78</td>
<td>5.63</td>
<td>4.68</td>
<td>4.34</td>
<td>6.64</td>
</tr>
<tr>
<td></td>
<td>$p(a_t)$ /%, cal</td>
<td>13.6</td>
<td>15.3</td>
<td>16.3</td>
<td>9.78</td>
<td>10.7</td>
<td>11.1</td>
<td>8.72</td>
<td>13.3</td>
<td>16.2</td>
<td>14.6</td>
</tr>
</tbody>
</table>

* $p$ --- polydispersity=standard deviation/mean. Generally, $p<$10% is considered as monodisperse.

** $cal$ --- calculated results.
Figure 5-6 The dependence of the thread number on the viscosity ratio. The circles are experimental data; each point is an average of around 20 threads. The solid line corresponds to the slender body theory, $\text{Th} = 0.0505\lambda^{-0.5}$.

For Newtonian systems with $\lambda<<1$ in simple shear flow, Hinch and Acrivos' slender body theory (1980) predicted that drops reach a pseudo-steady state before small disturbances cause it to break up. As $G=Ca^2\lambda^{2/3} \to \infty$, the thread radius, $R_b$ at this pseudo-steady state is given as

$$\frac{R_b}{a\lambda^{1/6}} = \frac{0.0505}{G}$$

(5-2)

We can rewrite this as:

$$R_b = \frac{0.0505\sigma}{\lambda^{0.5}\mu \dot{\gamma}}$$

(5-3)

Equation 5-3 shows that the thread radius is independent of the initial drop size. Using the definition of the thread number, Th, into the above equation, we obtain

$$\text{Th}=0.0505\lambda^{-0.5}$$

(5-4)
This implies that the thread number is only a function of the viscosity ratio. The prediction of Hinch and Acrivos’ theory is plotted as the solid line in Figure 5-6. The experiments are in good agreement with the theory results for $\lambda<0.1$.

Figure 5-7 shows both $Th$ and $Ca_c$ as a function of $\lambda$. We can see that $Th$ and $Ca_c$ follow a similar trend. For all viscosity ratios, $Ca_c$ is about $2.5\pm0.4$ times of $Th$. The possible underlying reason will be explored in the discussion section.

\[\text{Figure 5-7 } \text{Comparison of } Th \text{ (circles) and } Ca \text{ (squares) shows } Ca\sim2.5Th \text{ for } 0.0017<\lambda<3.5.\]

**SUMMARY**

For the Newtonian emulsions studied, the thread number is only a function of the viscosity ratio. The thread number is not sensitive to the viscosity ratio for $0.1<\lambda<1.0$, and it increases as $\lambda$ increases from 1.0 or decreases from 0.1. The experimental results of the thread number agree with Hinch and Acrivos’ slender body theory.
5.2 Wavelength at Breakup

We have measured the wavelength of the capillary instability from the same sets of images used to quantify the thread radius at breakup. The measurements are made when the undulation of the instability along the thread first becomes visible. For each set of experiments, we measure at least 40 wavelengths on threads from drops with different initial sizes. At a fixed viscosity ratio $\lambda=0.5$ and shear rate of $323\text{s}^{-1}$, the dimensionless wavelength, $k$, (the wavelength divided by the thread width at breakup) does not depend on the initial drop radius (see Figure 5-8). Each datum point is an average of the wavelengths along a single thread, and the error bar shows the standard deviation around the mean. The overall polydispersity of the wavelengths is 17.1%.

![Graph showing the relationship between dimensionless wavelength and initial drop radius.](image)

Figure 5-8 Dimensionless wavelength $k$ ($k=$wavelength/thread width at breakup) as a function of the initial drop radius at $\lambda=0.5$. The emulsion is 0.2wt% 337mPa.s silicone oil in 675mPa.s castor oil. The shear rate is $323\text{s}^{-1}$. Each datum is an average of the wavelengths along a single thread, and the error bar shows the standard deviation. The overall polydispersity, $\sigma=17.1\%$.

Results from different shear rates for the same sample with $\lambda=0.5$ (Figure 5-9) show that the dimensionless wavelength is independent of the shear rate. As the shear rate increases, the thread width at breakup decreases (see Figure 5-1), and the absolute value of
wavelength also decreases. However, the ratio of the wavelength to the thread width at breakup remains the same.

![Graph](image)

**Figure 5-9** Dimensionless wavelength as a function of the shear rate at $\lambda=0.5$. The emulsion is 0.2wt% 337mPa.s silicone oil in 675mPa.s castor oil. Each datum is an average of at least 40 wavelengths on threads from drops with different initial sizes. The error bar denotes the standard deviation.

Figure 5-10 shows the wavelength at breakup vs. the initial drop radius for 0.0017<$\lambda<$3.5, measured from the same set of images from which we measured the thread radius at breakup shown in Figure 5-5. As shown in Chapter 4, at $\lambda<$0.1, daughter drops resulting from the capillary instability can re-break by a capillary instability with a shorter wavelength. The re-breaking may continue multiple times. The wavelengths observed therefore are the result of different generations of the capillary instability. The wavelength measured is the wavelength of the last generation. For viscosity ratios between 0.1 and 1.0, the dimensionless wavelength is distributed relatively uniformly around the mean, which is between 6 and 7. As $\lambda$ increases from 1.0 or decreases from 0.1, the wavelength increases and the dispersion around the mean widens. The polydispersity of wavelength is around 15%
Figure 5-10 Dependence of the dimensionless wavelength $k$ on the initial drop radius at a variety of viscosity ratios between $10^3$ and 3.5. The scale of $x$-axis is 10-35$\mu$m for all the figures, and the $y$-axis is scaled as twice of the mean wavelength at breakup ($\bar{k}$) for each case. (to be continued on the next page)
for $0.1 < \lambda < 1.0$, while it is generally above 20% when $\lambda$ is out of this range. The dependence of the wavelength and its polydispersity on the viscosity ratio is summarized in Table 5-2.

**The dominant wavelength predicted by stability analysis**

Tomotika (1935) theoretically analyzed the capillary instability on an infinitely long liquid thread immersed in another quiescent liquid. He found that there is a fastest growing instability with a certain wavelength at any given viscosity ratio. Rumscheidt and Mason's experiment (1962) showed surprisingly good agreement with the predictions of final drop size from linear stability analysis, despite Tomotika's theory is a small deformation theory.
The theoretical results are compared with our experimental results in Figure 5-11. Our measured wavelength at breakup coincides with 1.2 times of the theoretical values (the dashed line) except for $\lambda=3.5$. This appears to be surprising at the first glance, because our experiment is for simple shear flow, while Tomotika's theory is for quiescent breakup. However, as we have discussed previously, in steady shear, the drop aligns with the flow such that it may be considered to be in a pseudo-quiescent state at breakup.

![Comparison of experimental results of dimensionless wavelength with Tomotika's theoretical results. The solid line is the prediction of Tomotika's theory for quiescent breakup. The experimental results coincide with the dashed line, which is 1.20 times of the theoretical values. Each datum is an average of at least 40 wavelengths on threads from drops with different initial sizes. The error bar denotes the standard deviation.](image)

During quiescent breakup, the whole length of the thread barely changes, thus the wavelength is a constant as its amplitude grows. While the pseudo-steady state under shear is not a true steady state, the thread is still extending, albeit slowly. Therefore, the wavelength along the length of the thread is also elongating slowly as it grows. For example, for $\lambda=0.5$ and $\dot{\gamma}=495s^{-1}$, the wavelength elongates an average of 7% in 20ms after it first becomes visible (see Table 5-3). We cannot directly measure the wavelength elongation as its amplitude grows from zero to when it first becomes visible. Our experiments show that it
takes about 40ms for the amplitude to grow from zero to first become visible in the above case. For pseudo-affine deformation, the drop length elongates linearly with time (see Equation 2-21). Therefore, we can estimate that the wavelength elongates about 14% as the amplitude grows from zero to when it first becomes visible. This accounts for the fact that our measured wavelength in simple shear flow is about 20% larger than the prediction of Tomotika's theory for quiescent breakup.

<table>
<thead>
<tr>
<th>Thread</th>
<th>Wavelength when first becomes visible</th>
<th>Wavelength after 20ms</th>
<th>Elongation percentage in 20ms</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>6.37</td>
<td>6.57</td>
<td>3.1%</td>
</tr>
<tr>
<td>2</td>
<td>5.9</td>
<td>6.52</td>
<td>10.5%</td>
</tr>
<tr>
<td>3</td>
<td>5.45</td>
<td>5.79</td>
<td>6.2%</td>
</tr>
<tr>
<td>4</td>
<td>6.02</td>
<td>6.57</td>
<td>9.1%</td>
</tr>
</tbody>
</table>

**SUMMARY**

The wavelength at breakup is only a function of the viscosity ratio. For the system studied, it is between 6 and 7 for 0.1<\(\lambda\)<1.0, and increases as \(\lambda\) increases from 1.0 or decreases from 0.1. The polydispersity of the wavelength is around 15% for 0.1<\(\lambda\)<1.0 and above 20% otherwise. The wavelengths are 20% larger than the predictions of Tomotika's stability analysis for quiescent breakup, due to wavelength elongation in shear flow.

**5.3 Final Drop Size**

In the experiments, after the shear flow is stopped, the drops in the field of view are recorded and their sizes are measured. Figure 5-12 shows the final drop size distribution for
Figure 5-12 Final drop size distribution for 0.0017<λ<3.5 shows nearly monodisperse drops are obtained for 0.1<λ<1.0, but not otherwise. The x-axis of the figures is scaled as twice of the corresponding average drop size, and the y-axis is nondimensionlized by the total number of drops (to be continued on the next page).
$\lambda=0.50$, $\dot{\gamma}=323s^{-1}$, $a_f=5.35\mu m$, and $p=12.6\%$

$\lambda=2.1$, $\dot{\gamma}=149s^{-1}$, $a_f=3.76\mu m$, and $p=16.6\%$

$\lambda=1.0$, $\dot{\gamma}=151s^{-1}$, $a_f=4.47\mu m$, and $p=12.9\%$

$\lambda=3.5$, $\dot{\gamma}=197s^{-1}$, $a_f=4.33\mu m$, and $p=34.8\%$

Figure 5-12 Final drop size distribution for $0.0017<\lambda<3.5$ shows nearly monodisperse drops are obtained for $0.1<\lambda<1.0$, but not otherwise. Note the bimodal distribution obtained at $\lambda=3.5$.

Figure 5-13 Final drop size distribution by volume for $\lambda=3.5$, corresponding to the drop size distribution by number for $\lambda=3.5$ in Figure 5-12.
0.0017<\lambda<3.5, corresponding to the same sets of experiments that we measured the thread radius at breakup (Figure 5-5) and the wavelength at breakup (Figure 5-10). For each case, the distribution corresponds to a measurement of at least 60 drops, except that we have only 30 drops for \lambda=0.018 and 57 drops for \lambda=0.075. In particular, we have measured 327 drops for \lambda=0.030. The results show that the average drop size measured from sub-samples consisting of more than 50 drops is within 5% that of the whole sample of 327 drops. The polydispersity of the sub-samples is within 3% that of the whole sample. To compare the size distribution between different viscosity ratios, the x-axis of each figure is scaled as twice of the corresponding average drop size, and the y-axis is nondimensionlized by the total number of drops. As we can see, the dispersion around the mean drop size is fairly narrow for 0.1<\lambda<1.0. Quantitatively, the polydispersity is between 10-14% for 0.1<\lambda<1.0, which means the emulsions become fairly monodisperse under shear. The polydispersity increases as \lambda decreases from 0.1 or increases from 1.0. Note that the drop size distribution is bimodal at \lambda=3.5. This is due to the relatively big satellite and sub-satellite drops formed from the filaments between the daughter drops (see Figure 4-11 (II)). The higher peak at the smaller drop size indicates that there are more satellites than daughter drops. However, the volume percentage of the daughter drops is still larger than that of the satellites, as shown by the drop size distribution by volume in Figure 5-13. The experimental results of the mean drop radius \alpha and the polydispersity are summarized in Table 5-2.

The experimental results of the dependence of mean drop size as a function of the inverse shear rate at \lambda=0.5 (see Figure 5-14) indicates that \alpha is in inverse proportion to the shear rate.
Figure 5-14 Final drop size is in direct proportion to the inverse shear rate at $\lambda=0.5$. The emulsion is 0.2wt% 337mPa.s silicone oil in 675mPa.s castor oil.

**SUMMARY**

Fairly monodisperse dilute Newtonian emulsions are obtained under shear for $0.1<\lambda<1.0$. Conversely, polydisperse drops are generated for $\lambda>1.0$ and $\lambda<0.1$. The average final drop size is inversely proportional to the shear rate at $\lambda=0.5$.

**5.4 Time to Breakup and Pseudo-Affine Deformation**

When a drop deforms affinely, the disperse phase deforms as if it were the matrix itself, i.e., as if no interface were present. The strain of the drop is then given by

$$\gamma = \frac{l}{a} \quad (5-5)$$

where $l$ is the half-length of the drop at time $t$. Volume conservation gives

$$a^3 \sim R^2 l \quad (5-6)$$

where $R$ is the radius of the thread at time $t$. Substituting Equation 5-6 into Equation 5-5, we have

$$\gamma = (a/R)^2 \quad (5-7)$$
Figure 5-15 Strain to breakup ($\gamma = \dot{\gamma}t_b$) as a function of $a/R_b$ for $0.0017<\lambda<3.5$. Drops deform affinely for $0.1<\lambda<1.0$, and it deviates from pseudo-affine deformation as the difference in the viscosities of the two phases increases. The dotted dashed line corresponds to pseudo-affine deformation of Equation 5-9.
Figure 5.16 Thread radius thinning as a function of strain ($\gamma = \dot{\gamma} t_s$) for $0.0017 < \lambda < 3.5$. Note the thread radius is normalized with the initial drop radius. The dashed line corresponds to pseudo-affine deformation (Equation 5.7).
The strain experienced by the drop in time $t$ can also be expressed as

$$\gamma = \dot{\gamma} t$$  \hspace{1cm} (5-8)

Therefore, at the time to breakup, $t_b$,

$$\dot{\gamma} t_b \sim (a/R_b)^2$$  \hspace{1cm} (5-9)

Figure 5-15 shows the strain at breakup ($\gamma = \dot{\gamma} t_b$) as a function of the ratio of initial drop radius and the thread radius at breakup for a variety of viscosity ratios. The circles are experimental data, and the dashed line corresponds to the pseudo-affine deformation of Equation 5-9. We can see that drop deforms affinely for $0.1 < \lambda < 1.0$, and it deviates from pseudo-affine deformation as the difference in the viscosities of the two phases increases. It is expected that when the viscosities of the two phases match each other, they tend to behave the same way under shear. When the viscosities are mismatched, the inner phase and the outer phase will deform differently and thus less affinely.

Figure 5-16 shows the thread radius thinning as a function of strain for drops at different viscosity ratios. The drop size in each case is about 50$\mu$m. For $0.1 < \lambda < 1.0$, the thread radius follows the pseudo-affine deformation curve. For $\lambda < 0.1$, the thread radius thinning deviates from affine deformation, particularly, at $\lambda = 0.0017$. At $\lambda = 2.1$ and 3.5, the thread radius thinning appears to follow pseudo-affine deformation. In general, the curve of thread radius thinning in Figure 5-16 is less sensitive than the curve of strain to breakup in Figure 5-15 in identifying pseudo-affine deformation.

**SUMMARY**

Drops deform pseudo-affinely for $0.1 < \lambda < 1.0$, but the deformation process deviates from pseudo-affine as the difference in the viscosity of the two phases increases.
5.5 Discussion

5.5.1 Critical capillary number and thread radius at breakup

Figure 5-7 shows that $Th$ and $Ca_c$ follow similar trends over $\lambda$, and $Ca_c$ is about twice (2.5±0.4 times to be precise) the value of $Th$ for all the viscosity ratios. This is not surprising because both the thread number and the critical capillary number characterize the balance of shear stress and interfacial pressure.

For a drop suspended in another quiescent liquid, the interfacial tension maintains the drop in a spherical shape. Under shear, the viscous stress ($\mu \dot{\gamma}$) overcomes the interfacial pressure ($\sigma/\alpha$), and deforms the drop into an ellipsoidal shape. As the viscous stress (shear rate) increases, the drop becomes more and more elongated. When the shear rate increases to a certain critical value, the drop can no longer keep a steady shape, and then undergoes a transient elongation, which finally leads to the disintegration of the drop. The capillary number corresponding to this critical shear rate is the critical capillary number ($Ca_c$).

In simple shear flow, as the drop elongates, it rotates toward the flow direction. Eventually, it almost aligns with the flow direction so that there is little driving force to further elongate the thread. Meanwhile, the interfacial pressure ($\sigma/R_b$, here $R_b$ is the thread radius and not the initial drop radius) increases as the thread thins. Consequently, the two forces almost balance each other and the drop reaches a pseudo-steady state. There is thus enough time for the amplitude of the capillary instability to grow to the radius of the thread and cause it to break.

For an ellipsoidal drop with circular cross-section, the interfacial pressure is

$$\Delta p = \frac{\sigma}{b_1} + \frac{\sigma}{b_2}$$

(5-10)
where \( b_1 \) and \( b_2 \) are the two principal radii of the drop. For a spherical drop with radius \( a \), \( b_1 = b_2 = a \), the interfacial pressure is then

\[
\Delta p = \frac{2\sigma}{a} \tag{5-11}
\]

For a long thread, the length is much greater than the width, \textit{i.e.}, \( b_1 \gg b_2 \). Thus, Equation 5-10 can be simplified as

\[
\Delta p \approx \frac{\sigma}{b_2} \tag{5-12}
\]

\( b_2 \) can be approximated by the thread radius. The circular cross-section was justified by Guido and Villone's experiments (1998) for highly deformed drops. At the pseudo-steady state,

\[
\Delta p \approx \frac{\sigma}{R_b} \tag{5-13}
\]

To balance the same shear stress,

\[
\frac{2\sigma}{a_c} \sim \frac{\sigma}{R_b} \tag{5-14}
\]

\textit{i.e.}, \( a_c \sim 2R_b \).

Thus, \( Ca_c \sim 2Th \).

### 5.5.2 Estimate the average final drop size and the polydispersity

\textbf{(a) Calculating mean drop size}

We can estimate the final drop size by assuming all the daughter drops come from the capillary instability and one wavelength generates one daughter drop as shown in Figure 5-17.
Consider a deformed thread to be a cylinder with radius $R_b$. The volume of a section of this cylinder that is one wavelength long is

$$V_{\omega} = \pi R_b^2 \omega \tag{5-15}$$

where $R_b$ is thread radius at breakup, and $\omega$ is the wavelength, which can be expressed in terms of the dimensionless wavelength $k$ as

$$k = \omega / 2R_b \tag{5-16}$$

Thus,

$$V_{\omega} = 2k \pi R_b^3 \tag{5-17}$$

The volume of the final drop is

$$V_f = 4/3 \pi a_f^3 \tag{5-18}$$

From volume conservation we have

$$V_f = V_{\omega} \tag{5-19}$$
so that
\[ a_f = (1.5k)^{1/3} R_b \]  \hspace{1cm} (5-20)

Therefore, the final drop size is directly proportional to the thread radius at breakup, and to the 1/3 power of the wavenumber. Table 5-2 shows calculated values of the final drop size based on experimentally obtained values of the thread radius and the wavelength at breakup. The calculated values generally agree with the direct measurements (within 10%) for \(0.1 < \lambda < 1.0\), but overestimate for \(\lambda < 0.1\) due to drop re-breaking and for \(\lambda > 1.0\) due to large satellite drops.

For \(0.1 < \lambda < 1.0\), the calculated results are about 10% larger than the experimental results. Some of the reasons for the discrepancy are:

(1) When we calculate the volume of the cylinder of one wavelength long, we need the measurements of the thread radius and the wavelength simultaneously. However, we are not able to measure the wavelength as we measure the thread radius at breakup, because the amplitude of the wavelength at that moment is zero. Instead, we measure the wavelength a little later when it first becomes distinguishable. In Section 5.2, we have estimated that the wavelength elongates averagely about 14% as the amplitude grows from zero amplitude to it first becomes visible in one case at \(\lambda = 0.5\).

(2) We neglected the satellite and sub-satellite drops in the calculation. Incorporating the volume of satellite drops into the volume of daughter drops would also increase the calculated value of the final drop size. In one case at \(\lambda = 0.5\) and \(\dot{\gamma} = 130\, \text{s}^{-1}\), we were able to measure all the 21 daughter drops from one mother drop. The total volume of the 21 daughter drops is about 96% of the volume of the mother drop.
The above factors will result in an overestimate of the average drop size roughly by 6%.

\textbf{(b) Correlating daughter drop size with the critical drop size $a_c$}

As shown by Figure 5-7, \( Ca = (2.5 \pm 0.4)Th \), \textit{i.e.}, \( a_c = (2.5 \pm 0.4)R_b \). Substituting into Equation 5-20 gives \( a_f = 0.4(1.5k)^{1/3}a_c \). The average of experimentally determined \( ks \) for \( 0.1 < \lambda < 1.0 \) is 6.3 (Table 5-2), thus \( a_f \sim 0.85a_c \). Our direct measurement of \( a_f/a_c \) for \( 0.1 < \lambda < 1.0 \) gives an average of 0.81 (Table 5-2), which is consistent with the above estimate. The ratio of \( a_f/a_c \) is generally lower than the estimate for \( \lambda < 0.1 \) due to drop re-breaking and for \( \lambda > 1.0 \) due to large satellite drops. In particular, \( a_f/a_c \) is as low as 0.46 for \( \lambda = 0.0017 \) and 0.49 for \( \lambda = 3.6 \). Bigio \textit{et al.} 's experimental results (1998) for simple shear flow showed that the mean daughter drop size approximates 0.7 times the critical drop size for \( 0.03 < \lambda < 3.0 \), which is in fair agreement with our results. As argued by Bigio \textit{et al.}, since the critical conditions are well studied (\textit{e.g.} Grace (1971)), the above scaling gives a priori prediction of the average final drop size. The above analysis shows that the scaling between \( a_f \) and \( a_c \) is due to the size selection of the thread radius and the wavelength at breakup, and the scaling between the thread radius at breakup and the critical drop size. In addition, the scaling is applicable for \( 0.1 < \lambda < 1.0 \).

\textbf{(d) Calculating the polydispersity in the final drop size for 0.1<\lambda<1.0}

The polydispersity of final drop size can be calculated from the polydispersity of the thread radius and the wavelength through the propagation of error. For a function such as,

\[ z = x^ay^b \]  

(5-21)
the propagation of the error is (Bevington, 1969)

\[(\sigma_y/z)^2 = a^2(\sigma_x/x)^2 + b^2(\sigma_y/y)^2\]  \hspace{1cm} (5-22)

Therefore, the uncertainty of the final drop size could be derived from Equation 5-20 as,

\[(\sigma_i/a_i)^2 = (\sigma_{R_i}/R_i)^2 + 1/9(\sigma_i/a_i)^2\]  \hspace{1cm} (5-23)

where \(\sigma_{R_i}\) and \(\sigma_i\) are standard deviation of \(R_i\) and \(a_i\). The calculated polydispersity of final drop size is listed in Table 5-2 as well. We can see that for \(0.1 < \lambda < 1.0\), the calculated polydispersities are close to the experimental measurements. Thus the major source of polydispersity of final drop size can be attributed to the polydispersity of \(R_i\) and the wavelength. The effects of end pinched drops do not appear to be significant.

The actual polydispersity is much higher than the calculated value at \(\lambda = 3.6\) because of the large satellite drops. In particular, the distribution becomes bimodal at \(\lambda = 3.6\), as shown by Figures 5-12 and 5-13. The actual polydispersity is also much higher than that of the calculated results for \(\lambda < 0.1\), since the major source of polydispersity is no longer the polydispersity of thread radius and wavelength, but arise from the re-breaking and coalescence of the daughter drops.

\textbf{(d) Effects of the shear rate}

Our experimental results (see Figure 5-14) indicate that \(a_i\) is in inverse proportion to the shear rate at a fixed viscosity ratio of \(\lambda = 0.5\). From the definition of thread number, we have

\[R_i = Th\sigma\mu \dot{\gamma}\]  \hspace{1cm} (5-24)

Substituting Equation 5-24 into Equation 5-20, we obtain
\[
\frac{\mu \dot{\gamma} a_f}{\sigma} = (1.5k(\lambda))^{0.7} Th(\lambda)
\]  
(5-25)

Equation 5-25 indicates that the final drop size is in inverse proportion to the shear rate for a given emulsion system. For the same set of experiments, the averages of \(k\) and \(Th\) are determined as \(\bar{k} = 6.25\) (see Figure 5-9) and \(\bar{Th} = 0.135\) (see Table 5-1). Substituting into Equation 5-26, we calculate the slope of \(a_f\) vs. \(1/\dot{\gamma}\) to be \(1.77 \times 10^3 \mu m/s\). The actual slope in Figure 5-14 is \(1.59 \times 10^3 \mu m/s\), which agrees with the calculated value. With pre-measured \(k(\lambda)\) and \(Th(\lambda)\) such as Figure 5-11 and 5-6, the average drop size for a certain emulsion system at a given shear rate can be predicted by Equation 5-25. We expect Equation 5-25 to be applicable for \(0.1<\lambda<1.0\), when drop breakup is dominated by the capillary instability with uniform thread radius and wavelength at breakup.

By contrast, Khakhar and Ottino (1985) predicted that in a simple shear flow the daughter drop size scales as the inverse shear rate to the 0.32 power,

\[
\frac{a_f}{R_o} \propto \left(\frac{\tan^{-1} \theta_o \mu \sigma}{\dot{\gamma}}\right)^{-0.32}
\]  
(5-26)

where \(\theta_o\) and \(R_o\) are the initial orientation angle and the initial radius of the thread, respectively. However, \(\theta_o\) and \(R_o\) are initial conditions required by the theory, and it is not clear whether these should be shear rate dependent, since the theory does not relate \(\theta_o\) and \(R_o\) to the initial drop size.

For \(0.1<\lambda<1.0\) and \(Ca>2Ca_c\), we found that the thread radius and the wavelength at breakup are independent of the initial drop size, thus the final drop size is also independent of \(a\) according to Equation 5-25. Grace (1971) showed that as \(Ca/Ca_c\) increases (i.e., \(a\) increases when \(\dot{\gamma}\) fixed) the ratio of final drop diameter to original drop diameter \((=a_f/a)\) first
decreases and then tends to level out (see Figure 2-19). However, the result raises questions as we discussed in Section 2.6.2.

5.5.3 Making monodisperse dilute Newtonian emulsions

Fairly monodisperse dilute Newtonian emulsions are produced under shear for $0.1<\lambda<1.0$. The uniformity can be attributed to the following factors:

(1) The thread radius is uniform at breakup. The typical polydispersity of $R_b$ is around 10%.

(2) The wavelength of capillary instability is fairly uniform along the length of the thread and from thread to thread. The typical polydispersity of the wavelength is around 15%.

(3) The size of end-pinched drops is similar to that of the drops from the capillary instability. In addition, there are very few end-pinched drops.

(4) The satellite drops and sub-satellite drops are negligibly small in comparison to the daughter drops.

(5) Daughter drops do not break again.

The drops become more nonuniform for $\lambda>1.0$ due to the relatively large satellite and sub-satellite drops, and for $\lambda<0.1$ due to drop collision, re-breaking and coalescence, which generate drops of irregular sizes.

Therefore, to make monodisperse dilute Newtonian emulsions,

(1) We should choose emulsion systems with $\lambda$ between 0.1 and 1.0.

(2) We should first make a pre-sheared emulsion consisting of large drops and then apply a high shear rate. Drops smaller than $a_c$ will not break under the corresponding shear
rate. Therefore, fine emulsions with pre-existing small drops are not recommended. In addition, drop breakup is dominated by the capillary instability at high shear rate (high \( Ca \)), leading to monodisperse drops due to the size selection of the thread width and wavelength at breakup. Breakup dominated by end pinching at relatively low shear rate tends to produce more polydisperse drops.

(3) The shear needs to be homogeneous in space and steady with time. In addition, the shear needs to be sustained long enough to break all the drops. Otherwise, polydisperse drops will be obtained if the flow is stopped before all the extending threads have broken up. The size of drops resulting from quiescent thread breakup is proportional to the radius of the thread when the flow is stopped.
Chapter 6 Conclusions & Future Work

Conclusions

We have set up an experimental apparatus to visualize transient deformation and breakup of dilute Newtonian drops in Newtonian matrices under steady shear. The following conclusions can be drawn about the breakup mechanism, the thread radius and dominant wavelength at breakup, the final drop size and distribution, and the pseudo-affine nature of the breakup process:

1. Dependence of the breakup mechanism on $Ca$ and $\lambda$.

   a. At $Ca=C_{a*}$, drops are broken up via necking into two daughter drops separated by satellite drops. The daughter drops are fairly spherical around $\lambda=0.5$, and become increasingly slender as $\lambda$ decreases. The experimental measurements of $Ca_c$ under steady shear are smaller than results in the literature for pseudo-equilibrium breakup.

   b. At $Ca<2Ca_c$, end pinching is the dominant drop breakup mechanism. For $\lambda<1.0$, end pinching may repeat up to 4-5 times depending on the length of the thread. The first end-pinched drop is about 1.5~2.0 times larger than subsequent end-pinched drops. For $\lambda>1.0$, end pinching occurs only once on a given thread.

   c. At $Ca>2Ca_c$, the dominant mechanism for drop breakup is the capillary instability. For $0.1<\lambda<1$, the thread width at breakup does not depend on the initial drop size, and the wavelength of capillary instability is uniform along the length of a thread and from thread to thread. Once formed, the daughter drops retract to an ellipsoidal shape, and they are carried away from each other by the flow. No drop collision or drop re-breaking has been observed.
In addition, the satellite drops are negligibly small compared to the daughter drops. Size selection of the thread width and wavelength at breakup leads to monodisperse emulsions.

For $1.0 < \lambda < 3.5$, the breakup mechanism is similar to that for $0.1 < \lambda < 1.0$. However, the satellite drops are larger, which results in polydisperse emulsions.

For $\lambda < 0.1$, the wavelength at breakup is very long, generating daughter drops larger than the critical drop size, which then break again. The re-breaking causes drops to move toward each other and collide, which induces irregular drop re-breaking and coalescence. Colliding drops accelerate as they separate from each other, resulting in more collisions. Drop coalescence is more likely at low viscosity ratios, and occurs even for $Ca > Ca_c$ when drops are undergoing breakup. This drop re-breaking mechanism results in polydisperse emulsions.

(2) Drops reach a pseudo-steady state where they are almost aligned with the flow direction before the capillary instability starts to grow. At the pseudo-steady state, the viscous stress ($\mu \dot{\gamma}$) and the interfacial tension ($\sigma/R_b$) almost balance each other. The thread radius at breakup is determined by the balance of the two forces, which does not depend on the initial drop size. We define a thread number $Th = \frac{\mu \dot{\gamma}}{\sigma / R_b}$, and find that the thread number is a constant at a fixed viscosity ratio. The thread number exhibits a minimum as a function of $\lambda$. The measured thread number agrees with Hinch and Acrivos' slender body theory. The thread number is generally about half of the corresponding $Ca_c$, over a range of viscosity ratio between $10^{-3}$ and 3.5.
(3) The dimensionless wavelength at breakup (the wavelength divided by the thread diameter at breakup) is a function of the viscosity ratio. For systems studied, it is between 6 and 7 for $0.1<\lambda<1.0$, and increases as $\lambda$ increases from 1.0 or decreases from 0.1. Its polydispersity in the wavelength is around 15% for $0.1<\lambda<1.0$, and it is above 20% otherwise. The measured wavelengths agree with Tomotika's predictions for the dominant wavelength on quiescent threads. In steady shear, the drop is almost aligned with the flow such that it may be considered to be in a pseudo-quiescent state at breakup.

(4) The final drop size distribution is nearly monodisperse for $0.1<\lambda<1.0$, and the average drop size is inversely proportional to the shear rate. Volume conservation gives an estimate of the average drop radius about twice of the thread radius at breakup, which agrees with the experimental results. The final drops become polydisperse for $\lambda>1.0$ due to large satellite drops and for $\lambda<0.1$ because of drop re-breaking and coalescence.

(5) Drops deform pseudo-affinely for $0.1<\lambda<1.0$, but the deformation process deviates from pseudo-affine as the difference in the viscosity of the two phases increases.
Future Work

This thesis has focused on the breakup of single Newtonian drops in Newtonian matrices under steady shear, and it lays the groundwork for studies on more complicated systems involving surfactants, concentrated emulsions, and viscoelastic phases. The emulsions considered here are very dilute. The typical volume fraction of the drop is 0.1-0.2wt%, so that the interactions between drops can be neglected. Most industrial emulsions are more concentrated, and they usually contain some surfactant to stabilize them. One or both phases of an emulsion can be viscoelastic, and almost all polymer blends are viscoelastic. To investigate the above effects, we want to answer a series of questions (see Figure 6-1). The primary question is how each of these factors affects the drop breakup mechanism. In particular, we can ask: what the dominant drop breakup mechanism is, how it affects the size and uniformity of the thread width and the wavelength at breakup, and how it affects the satellite drop sizes and drop re-breaking mechanisms? Answering these questions will enable us to determine how these factors affect the final drop size and size distribution.

(1) Effect of surfactants

(a) Effect on breakup mechanism

Our experimental results indicate that certain surfactants may dramatically affect the drop breakup mechanism. For a system of silicone oil in a solution of 2%Tween-80 (a nonionic surfactant) in glycerine (λ=0.5), we observed nonuniform thread widths and wavelengths at breakup, which leads to polydisperse final drop sizes (see Section 5.1.2). One question to be answered is whether this is a phenomenon specific to this surfactant, or
general to all surfactants. To test this idea, a variety of combinations of different surfactants with different Newtonian matrices need to be studied.

(b) Effect on the thread radius size at breakup.

The thread radius at breakup is determined by the balance of the viscous stress \( (\mu \dot{\gamma}) \) and the interfacial pressure \( (\sigma/R) \). It is expected that under the same shear stress, a decrease of the interfacial tension will lead to smaller thread radii at breakup and smaller daughter drops.

(c) Prevention of drop re-breaking and coalescence upon drop collision.

We found that for \( \lambda < 0.1 \), daughter drops may re-break, which causes drop collision, which in turn induces drop re-breaking into irregular sized drops or drop coalescence. It is known that surfactants tend to inhibit drop coalescence. Hu et al. (2000) put a small amount
of polybutadiene (PB)-polydimethylsiloxane (PDMS) copolymer on the interface of PB drops in PDMS matrices. They found that the critical capillary number for coalescence was reduced by a factor of 6, even though the interfacial tension was reduced by only 3%. Surfactant also tends to immobilize the interface, which may prevent irregular drop re-breaking during collision.

(2) Effect of viscoelasticity

Current experimental evidence shows that drop breakup mechanism in emulsion systems consisting of viscoelastic polymer solutions is similar to that in purely Newtonian systems. In a viscoelastic system consisting of viscoelastic drops (aqueous solution of hydroxypropylcellulose) of in Newtonian matrix, Tsakalos et al. (1998) found that the capillary instability is the dominant drop breakup mechanism, and the thread width at breakup is independent of the initial drop size. We found similar results for a viscoelastic emulsion of Newtonian drops (338cP silicone oil) in a viscoelastic polymeric (12%PVP +2%SDS) water solution. However, several new phenomena, such as thread-folding, drop widening (Levitt et al., 1996), and vorticity alignment (Migler, 2000) have been observed in polymer blends. In addition, the viscoelasticity of polymer solutions may affect the thread width and the wavelength at breakup, and the satellite drop sizes.

(a) Effect on thread width at breakup

For the silicone oil/12%PVP+2%SDS water solution system we just mentioned, the thread number is calculated to be 0.41 at λ=0.48, which is higher than that of purely Newtonian systems at the same viscosity ratio, T=0.13. More experiments are needed to
quantify the effects of viscoelasticity on the thread width at breakup. Most polymer solutions are both viscoelastic and shear thinning. Using Boger fluids, i.e. viscoelastic fluids with constant viscosity can isolate the effects of viscoelasticity from those of shear thinning.

(b) Effect on dominant wavelength

Lee et al. (1981) extended Tomotika's stability analysis (1935) of quiescent thread breakup to viscoelastic systems, assuming that both the continuous phase and the dispersed phase are linearly viscoelastic. They found that the growth rate of the capillary instability is faster for viscoelastic systems than for Newtonian system at the same viscosity ratio. When the viscosity ratio (zero shear rate viscosity) is below 1.0, the critical wavelength increases as the elasticity ratio (disperse phase elasticity over the continuous phase elasticity) increases. When the viscosity ratio is above 1.0, the opposite is true but the effect is minor. The daughter drop size will change with the wavelength accordingly. Our results show that the dominant wavelength under steady shear is 20% higher than the predictions of Tomotika's linear stability theory for quiescent breakup. Experiments are needed to compare with the prediction of Lee et al. for the viscoelastic systems.

(c) Effect on satellite drop sizes

Varanasi et al. (1994) experimentally studied the deformation and breakup of viscoelastic drops suspended in Newtonian matrices. They found that drop breakup is due to the capillary instability at high shear rates, which is similar to that in Newtonian systems. However, the filament joining two neighbor daughter drops is very thin, which forms satellite drops much smaller than those in purely Newtonian systems. While Varanasi et al. studied
systems consisting of viscoelastic drops in Newtonian matrix; similar studies for Newtonian drops in viscoelastic matrices are still needed.

(3) Concentration Effects

(a) Production of monodisperse emulsions in concentrated systems

A few years ago, Mason and Bibette (1996) found that monodisperse emulsions can be produced by shearing certain highly viscoelastic and concentrated emulsions. Their emulsion system was quite complicated. It was highly viscoelastic, highly concentrated, and the continuous phase contained a high concentration of surfactant. A typical continuous phase is an aqueous solution of 40wt% of nonionic surfactant Tergitol NP-7, which serves both to stabilize the interface and to make the solution viscoelastic. The typical volume fraction of the silicone oil drop is 70%, which is above the close packing concentration. Thus, the viscoelasticity comes from both the high concentration surfactant and the deformed close packed drops. The mechanism producing the monodispersity remains unknown. Visualization of drop breakup in these emulsions under shear seems to be the most direct approach to explore the mechanism.

Marcu (2003) has successfully visualized drop breakup in concentrated emulsions. His emulsion system consists of castor oil drops suspended in corn syrup with surfactant AOS-1416. The refractive index of castor oil and corn syrup are almost identical. Dyeing a few drops of castor oil with Oil Red makes those drops stand out from the other undyed drops and the matrix. He was able to observe drop breakup at a volume fraction up to 70%, and found that the thread radius becomes nonuniform along the length of a thread with increasing concentration. No data about the final drop size distribution were reported. This
methodology proved to be feasible for concentrated emulsion systems. Searching for a concentrated emulsion system that can produce monodisperse emulsions under shear and can be visualized by the dyeing technique will be instrumental in finding out the mechanism producing the monodispersity in Mason and Bibette's emulsion. The continuous phase of Mason and Bibette is highly viscoelastic, while both the dispersed phase (castor oil) and the continuous phase (corn syrup) of Marcu's emulsion are Newtonian. Adding a small amount of high molecular weight polymer such as PAA into corn syrup will make it into a viscoelastic Boger fluid, and probably will not change the refractive index too much. Comparing the drop breakup mechanism and the final drop size distribution in this Boger fluid system with Marcu's purely Newtonian system may be helpful in figuring out the mechanism producing monodispersity in concentrated emulsions.

(b) What is the role of drop interaction when the concentration of drop is increased from very dilute to close packing?

Marcu (2003) has done some qualitative work on this subject. Quantitative analysis for a wider range of viscosity ratios is needed.

Each of the above problems is a rich project with practical significance. These effects may also couple together in an emulsion or polymer blend increasing the complexity of the problem. Good understanding of each individual effect is essential to solve the more difficult puzzles.
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Appendix A  Calculating parallelism of the glass plates by light interference

When the three micrometers are set to zero, the two glass plates are in close contact with each other. For simplicity, let us assume that the two plates are perfectly flat, and the lack of parallelism is caused by a prop of height $h$ at one end of the plates (see Figure A-1). The dimensions of the plates are 100mm x 30mm x 2mm for the bottom plate and 100mm x 30mm x 6mm for the top plate. The gap in the diagram is exaggerated.

![Diagram of interference of reflective lights from two glass plates.](image)

Figure A-1 Schematic diagram of interference of reflective lights from two glass plates.

The light beams reflected from two surfaces interfere only when the gap between the two surfaces is on the scale of the wavelength of light. White light consists of a spectrum of wavelengths, on the length scale of 600nm. Therefore, only light reflected from surface $b$ and $c$ will interfere. The interference is constructive (bright fringes) when the two beams are in phase, *i.e.*, they shift by an integer multiple of wavelengths relative to each other. On the contrary, the interference is destructive (dark fringes) when the beams are out of phase. We can see alternate bright and dark fringes, when we look through the plates from above. The number of visible fringes is determined by the gap thickness of the plates. The reflected beam experiences a shift of half a wavelength when it is reflected from a surface with higher refractive index, but experiences no shift when it is reflected from a surface of lower
refractive index. Since the refractive index of glass is higher than that of the air, the light reflected from surface \( b \) experiences no phase shift, while that reflected from surface \( c \) experiences a shift of half a wavelength.

The gap thickness \( h \) is related with the number of visible constructive fringes, \( n \), by (Halliday D. et al., 1997)

\[
2h + \lambda/2 = n\lambda
\]

hence,

\[
h = (2n-1)\lambda/4
\]

For example, when \( n=10 \), \( h=2.85\mu\text{m} \). A rule of thumb for checking the parallelism is that the number of visible bright fringes along the length of the glass plate is less than ten at zero gap.
Appendix B General procedures for measuring the rheological properties of liquids

The rheological properties of the fluids are measured in a strain-controlled rheometer (Rheometric, ARES) with a cone-and-plate geometry at 25°C. A general procedure of the measurements include:

(1) Turn on the rheometer and the connected computer, and start the software (RSIOrche600).

(2) Turn on the circulator, set the temperature, and wait until it stabilizes at the temperature.

(3) Clean the surfaces of the cone and the plate, and mount them onto the rheometer.

(4) Zero the gap between the cone and the plate.

(5) Raise the cone, and load the sample on the center of the plate.

(6) Lower the cone very slowly to avoid incorporating air bubbles into the sample, until the desired gap is reached.

(7) Clean up the liquid that squeezes out between the cone and plate.

(8) Set the experimental parameters and start the measurement.

(9) Save the data once the measurement is finished.