Experimental investigations of a semi-batch Taylor vortex reactor using hexane and water

by

Charlton F. Campbell

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Major: Mechanical Engineering

Program of Study Committee:
Michael G. Olsen, Co-major Professor
R. Dennis Vigil, Co-major Professor
Alberto Passalacqua
Rodney Fox
Travis Sippel

The student author, whose presentation of the scholarship herein was approved by the program of study committee, is solely responsible for the content of this dissertation. The Graduate College will ensure this dissertation is globally accessible and will not permit alterations after a degree is conferred.

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Ames, Iowa
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DEDICATION

I dedicate my thesis primarily to my nieces and nephews. I thoroughly hope you show
determination to succeed in your endeavors despite obstacles you will have to face. Let my
experience be a guide to you all. I love all seven of you from the bottom of my heart.
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Finally, I want to thank my family. Most of you don’t know what I do, but know I have been struggling. That’s on me though for not properly explaining to you. Just know that I love you all, especially you mom who somehow seem to annoy me and make me feel happy at the same time, whenever you call. I thank you all for being my support system!
ABSTRACT

Two phase Taylor-Vortex reactors have been receiving increased attention due to its capacity to generate emulsions within the system. This dissertation focuses on understanding a Taylor-Vortex (TV) system when operating in a semi-batch manner. Optical experiments were aimed at identifying patterns and distributions that would aid in system operation as well as to better understand the canonical Taylor-Couette flow when multiple phases are involved.

Using two immiscible liquids, hexane and water, in the semi-batch TV system, droplet patterns were identified in the system in order to create a flow regime map. The regime map identified four unique stages of banding and non-banding, indicating scenarios where Taylor vortices present in the system are either strong enough to trap droplets or ineffective in compensating for the force of rising buoyant droplets.

Further optical investigation of the system identified three unique droplet size distributions, a unimodal distribution, a bi-modal distribution, and a right-skewed distribution. Additionally, the droplet sizes were investigated in relation to the system operation parameters. It was concluded that the droplet sizes and distribution had a strong dependence on the entry conditions of the dispersed phase and did not correlate to the flow patterns identified in the primary investigation of the system. The investigation of the entry conditions shed further insight into the drop sizes and distributions through the identification of jet breakup modes consistent with classical immiscible liquid jet systems, albeit in a narrow channel and a rotating fluid.
CHAPTER 1  GENERAL INTRODUCTION

There are many unique flows in fluid dynamics. One such flow is the flow developed as a result of rotating a liquid between two concentric cylinders. This flow, the Taylor-Couette flow, offers unique multi-directional mixing of the liquid under rotation.

As part of a major contribution to the field of rheology - the science of fluid flow and deformation – Maurice Marie Alfred Couette developed a method to measure the viscosity of air and water [1]. The fluid would rotate between two concentric cylinders, and under stable conditions, Couette was able to accurately measure the viscosity of air and water. Couette’s 1890 thesis describes his concentric cylindrical apparatus and experiments in greater detail [2]. A few decades later, Sir Geoffrey Ingram Taylor in his pioneering paper "Stability of a viscous liquid contained between two rotating cylinders" [3] details his advancement of Couette’s work by investigating the hydrodynamic instabilities that occur within the fluid rotating between two cylinders. This additional work serves as the cornerstone of one of several turbulent flows aptly named Taylor-Couette flow (TCF).

1.1 History of TCF

Taylor-Couette flow is a unique and canonical flow that offers mixing capabilities for various applications. As shown in Figure 1-1, TCF describes the flow between two concentric cylinders when one or both cylinders rotate. When either cylinder move at lower rotation rates, the flow is laminar, identified as Couette flow, which aids in the determination of fluid viscosities of Newtonian and shear thinning fluids [4]. Under normal conditions in low rotations
the flow continues in a steady pattern within the annulus between the two cylinders. When the cylinders rotate beyond a certain point, turbulence increases within the system leading to distinctive flow features that are of interest to many fluid dynamicists.

As with all fluidic systems, it is essential to identify a few key characteristics of the system, to aid in analyses and comparisons. Notable non-dimensional parameters of a Taylor-Couette (TC) system [5] are as follows. The first parameter is the radius ratio, $\eta$:

$$\eta = \frac{r_i}{r_o}$$  \hspace{1cm} (1.1)
$r_i$ and $r_o$ are the inner and outer radii of the cylinders, respectively. The next parameter is the aspect ratio, $\Gamma$:

$$\Gamma = \frac{L}{d} \quad (1.2)$$

$L$ is the length of the fluid column, and $d = r_o - r_i$ is the gap width of the reactor. The final parameter that characterizes this flow is the Reynolds number, $Re$:

$$Re_i = \frac{r_i \omega_i d}{\nu} \quad (1.3)$$

$Re_i$ is the Reynolds number of the inner cylinder, $\omega_i$ is the angular velocity of the inner cylinder, and $\nu$ is the kinematic viscosity of the fluid in the annular gap of the cylinder. Substituting the subscript 'o' in place of the subscript 'i' provides the Reynolds number of the outer cylinder, $Re_o$.

Increasing the rotation of one or both cylinders result in different flow features allowing the development of a phase space for the TC system. Figure 1-2 shows Andereck et al.'s work in 1986 [6], reproduced by fluid physicists at the Twente Turbulent Taylor-Couette group [7], identifies the various flow patterns observed beyond this critical value. The Reynolds number on the vertical axis represents the rotation of the inner cylinder, which only rotates in one direction. On the horizontal axis however, the Reynolds number represents the rotation of the outer cylinder which changes from negative to positive, indicating a change from counter-rotating, to co-rotation, with the inner cylinder. When either cylinder move at lower rotation rates, the flow is laminar, identified as Couette flow. Beyond a certain threshold, the first
instability occurs, and the system develops Taylor vortices. A critical Reynolds number describes this first instability point, which varies for different systems, depending on the radius ratio $\eta$ [8]. Most investigations using a TC reactor operate by solely rotating the inner cylinder, identified by the dotted line perpendicular to the horizontal axis in Figure 1-2. Some of the flow characteristics observed beyond the point of the first instability are the Wavy Vortex Flow, Modulated Wavy Flow, and Turbulent Taylor Vortices [6].

These flow features are what make TCF reactors an interesting apparatus as vortices formed within the annular gap offer controlled non-intrusive mixing between the two cylinders.


Figure 1-2. TC flow regime map identifying flow patterns based on inner and outer cylinder rotation [7].
It then begs the question, what would happen with the addition of a secondary fluid phase to the TC system. Researchers considered this question since the 1960s when Davis and Weber investigated a solvent extraction process that suited the needs for processing radioactive solutions where the degradation of the solvent was an issue [9]. Davis and Weber’s article shows the relation with dispersed phase residence time increased with smaller annular gaps. This led to further studies using the TC system in extraction processes due to the control that the system offers in terms of the simplistic concentric tubular design, rotation rates, and feed rates for secondary phases.

1.2 Applications for TCF

There are many fundamental studies associated with TC systems. A few notable applications encompass desalination tanks [10], blood and plasma filtration [11], [12], magnetohydrodynamic turbulence [13], mixing nutrients and oxygen to promote algal growth in bioreactors with suspended cultures [14], [15], photocatalysis [16], polymer processing [17], emulsion generation for liquid extraction [18], flocculation in water treatment [19], [20], food processing [21] separation of radio-nuclides from high level radiation liquid waste [17]. These investigations stem from the inability to inject fluid into the systems or systems where injection was only available at one port. As TC systems offer tunable multi-directional mixing via the developed hydrodynamic instabilities, liquid-liquid TCF studies progressed, which is the primary focus of this dissertation.
1.3 Liquid-Liquid Studies

Research expanded studies of the canonical flow surpassing the low-Reynolds regimes from $\text{Re} = 10^3$, to $\text{Re} = 10^6$. In their study, Grossman et al. (2016) provided a survey of the extended flow studies into the high-Reynolds regimes, characterizing boundary layer transition from low to high Re, and suggests the impact the TC system has on multiphase flow [7]. Experiments show that there is effectiveness in controlling a TC system due to it being a closed system, with a wide-ranging flow space, and thus appropriate for studying particles, bubbles, and droplets, in a multiphase system.

Due to the applicability of TC flows to enhance multiphase flows, research focus expanded to include gas-liquid and liquid-liquid systems. Studies covering both multiphase fields increased in the late 1990s, and early 2000s. Early aspects of gas-liquid TC systems initiated with experimental investigations identifying different flow patterns and regimes [23]–[25], and mass transfer characteristics [18], [19] before studies extended to computational simulation of similar systems [20], [21]. The same trend governed the study of liquid-liquid TC systems, with experimental studies arising as early as 1960, where Davis and Weber studied solvent extraction in a TC extractor [9]. Researchers continued exploration of liquid-liquid TC flows in the late 1990s, investigating flow features, patterns, and mass transfer within horizontally aligned systems [22]–[24]. Campero and Vigil in their 1997 work observed flow patterns in a horizontally aligned system, where they observed banded and oscillatory flow features when kerosene and water were introduced in the annular gap of the TC reactor [23]. The flow rate of the two immiscible liquids were separately controlled, allowing the reactor to
operate in a continuous flow, akin to a liquid-liquid extractor. In a separate study, Dluska and Markowska generated emulsions in a horizontally aligned TC contactor [25]. The system generated emulsions by introducing multiple liquid streams in the annular gap of the TC contactor. The water and silicone oil entered the contactor at different flow rates and volumetric fractions to generate the required droplet sizes. Furthermore, they presented a flow regime map that identified different droplet sizes based on the volume fraction of oil in the system, and the rotation rate of the inner cylinder. Sathe et al. in their computational and experimental investigation of a two-phase liquid-liquid flow in a vertical TC contactor introduced the dispersed phase after the inner cylinder began rotating [26]. Their contactor operated in a batch manner where they identified four distinct flow regimes and their respective criteria. Other researchers in the past two decades conducted experiments on liquid-liquid TC reactors either operating vertically or horizontally, operating as a centrifugal contactor, or in a batch manner [27]–[31]. The purpose of this dissertation is to showcase the semi-batch operation of a vertically oriented, liquid-liquid Taylor-Couette reactor.

1.4 Dissertation Organization

The organization of this dissertation mimics the outline proposed by the Graduate College of Iowa State University, meeting the Journal paper format. As such, there is a separate bibliography at the end of each chapter. Chapter 1 introduces the reader to the concept of Taylor-Couette flow, leading to a discussion on the present status of liquid-liquid TCF systems, thus highlighting the significance of the research. Chapter 2 presents an article published in the ASME Journal of Fluids Engineering, which discusses identified flow regimes in the semi-batch
TC reactor. Characterizing the flow regimes allows the identification of controlled space parameters for users of this particular system. Furthermore, there is presentation of the secondary phase hold-up as it relates to the feed and rotation rates of the system. Chapter 3 focuses on the droplet size distribution spanning the mapped phase space identified in Chapter 1. The chapter also provides a discussion on the dependence of droplet diameters on both rotation and feed rates of the system. Chapter 4 delves further in the entry conditions associated with the secondary phase in the system. Focusing on the breakup process of the dispersed phase sheds insight into the dependence on the drop formation process and the drop size distributions identified in Chapter 3. Chapter 5 revisits key conclusions identified in the previous chapters and discusses potential future work associated with the operation of the semi-batch TC system.

1.5 References


CHAPTER 2 FLOW REGIMES IN TWO-PHASE HEXANE/WATER SEMI-BATCH VERTICAL TAYLOR VORTEX FLOW

Charlton Campbell
Department of Mechanical Engineering,
2025 Black Engineering
Iowa State University, Ames, IA 50011-2030
campbel8@iastate.edu

Michael G. Olsen
Department of Mechanical Engineering,
2025 Black Engineering
Iowa State University, Ames, IA 50011-2030
mgolsen@iastate.edu

R. Dennis Vigil
Department of Chemical & Biological Engineering,
2114 Sweeney Hall
Iowa State University, Ames, IA 50011-2230
vigil@iastate.edu


2.1 Abstract

Optical-based experiments were carried out using the immiscible pair of liquids hexane and water in a vertically oriented Taylor–Couette reactor operated in a semi-batch mode. The dispersed droplet phase (hexane) was continually fed and removed from the reactor in a closed loop setup. The continuous water phase did not enter or exit the annular gap. Four distinct flow
patterns were observed including (1) a pseudo-homogenous dispersion, (2) a weakly banded regime, (3) a horizontally banded dispersion, and (4) a helical flow regime. These flow patterns can be organized into a two-dimensional regime map using the azimuthal and axial Reynolds numbers as axes. In addition, the dispersed phase holdup was found to increase monotonically with both the azimuthal and axial Reynolds numbers. The experimental observations can be explained in the context of a competition between the buoyancy-driven axial flow of hexane droplets and the wall-driven vortex flow of the continuous water phase. [DOI: 10.1115/1.4043493].

2.2 Introduction

The flow of two immiscible liquids in the annular region between a rotating inner cylinder and a concentric stationary outer cylinder (i.e., Liquid-Liquid Taylor-Couette flow) has practical applications (e.g. liquid extraction [1–4]; mixing and centrifugation [5–7]) and it can also serve as a canonical system for better understanding two-phase fluid flow. Whereas single phase Taylor-Couette flow has been extensively studied [8,9], and more recently gas-liquid Taylor-Couette flow has received attention [10–19], liquid-liquid Taylor-Couette flow is relatively poorly understood, particularly for the case of a vertically-oriented main axis with continuous feed of the dispersed phase, which is the subject of this work.

Most previous studies of liquid-liquid Taylor-Couette flow have focused on the behavior of systems with a horizontal main axis (i.e., axis of rotation), and in these systems several fluid flow pattern regimes have been identified, including banded droplet structures that typically arise at high inner cylinder rotation rates [20–29]. Under continuous co-current feed of both
immiscible phases, other interesting phenomena have been observed including phase inversions and periodic switching between banded and phase inverted flow patterns [22]. In addition to the aforementioned studies, flow structures in the annular region between co-rotating horizontal cylinders have also been explored [30,31]. Co-rotation of cylinders can cause centrifugation of an immiscible pair of liquids into a layered flow wherein one fluid wets the outer cylinder and the other wets the inner cylinder. For sufficiently high inner cylinder rotation speeds, Taylor vortices arising in both fluid layers can potentially be exploited for mass transfer applications [32].

Far less is known about liquid-liquid flow in vertically-oriented Couette devices, but a few experimental and computational investigations have been reported for reactors closed to mass exchange. Sathe et al. [33] surveyed fluid flow regimes in a closed vertical reactor and carried out corresponding computational fluid dynamics simulations. They identified four distinct flow patterns including a gravitationally segregated flow (no dispersion), a segregated dispersion which features droplets only in part of the reactor, a banded dispersion, and finally a homogeneous dispersion. These flow patterns can be organized using Taylor and Eötvös numbers to generate a phase regime map [33]. More recently, the motion of a single droplet within a vortex, where the droplet was injected in the quiescent fluid prior to data collection, has been studied in detail [34], and a population balance model for droplets undergoing coalescence and breakage in a vertical liquid-liquid Couette flow device has been proposed [28].

Here, we extend the study of flows in a vertically-oriented Taylor-Couette reactor operating in a semi-batch process. Using fluid pairs with different densities (where $\rho_i$ is
the continuous phase density, and \( \rho_d \) is the dispersed phase density), it is possible to operate the reactor in a semi-batch manner by providing for a constant feed and removal of the dispersed phase, which will either rise to the top (\( \rho_d < \rho_c \)) or settle at the bottom (\( \rho_d > \rho_c \)) of the reactor. For example, hexane droplets (\( \rho_d = 659 \text{ kg/m}^3 \)) continually introduced at the bottom of a reactor initially filled with water will eventually rise to the top where they will gather to form a continuous hexane layer at the free surface. By removing fluid from this hexane layer at the same rate that hexane is fed to the reactor at the bottom, a steady state can be achieved. Here we describe the results of optical based experiments carried out in a vertically-oriented Taylor vortex reactor operated using a hexane-water fluid pair in the semi-batch mode just described. Several distinct flow regimes were identified, and a flow regime map was constructed in terms of two key dimensionless parameters, namely the axial Reynolds number (which is a function of the volumetric flowrate of the disperse phase), \( \text{Re}_a \), and the azimuthal Reynolds number (which is a function of the rotational speed of the inner cylinder), \( \text{Re}_\theta \) [19]. The dependence of the dispersed phase holdup on these dimensionless quantities was also explored. The experimental observations can be interpreted in the context of a competition between the wall-driven azimuthal flow and the buoyancy-driven axial flow of the dispersed phase.

### 2.3 Experimental Apparatus and Procedures

The flow apparatus used in this investigation is depicted in Figure 2-1. The device consists of two concentric transparent acrylic cylinders confined between a top and bottom plate. The hollow rotating inner cylinder has an outer radius \( r_o = 15.2 \text{ cm} \) and a wall thickness of
0.64 cm. The stationary outer cylinder has an inner radius \( r_i = 19.4 \text{ cm} \) and a wall thickness of 0.95 cm. The resulting gap width between the two cylinders is 4.2 cm. The two cylinders are 152.4 cm long, and the annular gap was filled to a height \( h = 141 \text{ cm} \), resulting in a working fluid volume of 80 L. The corresponding aspect ratio and radius ratio of the system are given by \( \Gamma = \frac{h}{(r_o - r_i)} = 34 \) and \( \eta = \frac{r_i}{r_o} = 0.78 \), respectively.

Figure 2-1. Cross-sectional view of Taylor Couette apparatus in the experimental study. The inner cylinder has an outer radius of 15.2 cm, and the outer cylinder has an inner radius of 19.4 cm. The height of the working fluid in the annular gap is 141 cm. Note that the height of the apparatus has been truncated in the figure.
The inner cylinder was rotated along its major axis using a Dayton 3-phase, 1 hp general purpose motor coupled to the cylinder using an MTD-20 magnetic disc coupling from Magnetic Technologies Ltd. The motor was controlled remotely using a Schneider Electric ATV 12 variable frequency drive. Acrylic flanges, affixed on the outer cylinder, were used to attach the reactor to aluminum top and bottom plates, and leakage was prevented using an oil-resistant compressible Buna-N gasket from McMaster-Carr. The dispersed phase fluid was introduced into the apparatus through four blunt tip 16-gauge stainless steel needles mounted in the baseplate midway between the inner and outer cylinders and equally spaced azimuthally at 90-degree angles.

Figure 2-2. Schematic of the experimental arrangement. The annulus was filled with deionized water prior to introducing hexane into the system.
The continuous phase working fluid consisted of deionized water, and its volume was conserved because it was neither fed nor removed from the device after being initially introduced into the annulus. As the system was closed, it was important to obtain the fluid properties prior to performing the experiments. The measured viscosities of hexane and water were 0.451 mm$^2$/s and 1.00 mm$^2$/s, respectively, and the interfacial surface tension was 52.4 mN/m. The dispersed phase (hexane dyed with Oil Red O) was circulated through the apparatus using a (Cole-Parmer Model No. 75211-30) gear pump, with a full-scale accuracy of ±0.25% in a closed-loop arrangement, as depicted in Figure 2-2. Hexane injected through the needles in the baseplate formed droplets that rose through the flow device, eventually
coalescing into a phase-separated layer at the top of the reactor. This layer of hexane served as the feed to the pump for recirculation back into the flow cell.

![Graph showing droplet measurements](image)

**Figure 2-4.** Droplet measurements obtained showing size dependence on the axial and the azimuthal Reynolds number.

Images of the dispersed droplets in the annulus were acquired using a digital camera (Canon Rebel T3i) fitted with a Canon 18-55 mm f/3.5-5.6 zoom lens and aided by two off-camera electronic flashes. The camera was placed 1 m away from the test section, at a height approximately midway between the top and bottom of the outer cylinder. A flat, matte white backdrop was placed behind the reactor, opposite to the camera, to increase contrast between
the two liquid phases. Additionally, two synchronized flashes were used to provide sufficient lighting to the test section. Droplet diameters were measured from postprocessed digital images as shown in Figure 2-3.

Figure 2-4 shows preliminary results from a subsequent experiment showing droplet size varying in the range 0.5-2.8 mm depending upon the axial Reynolds number. Observations indicate the droplet diameters to be monodisperse, with no growth or coalescence prior to reaching the hexane layer at the top of the test section.

The effects of two operating variables on the observed fluid flow patterns were investigated. These variables were the inner cylinder rotation speed and the hexane volumetric flow rate. The range of operating values of these parameters can be expressed in dimensionless form by computing the corresponding azimuthal and axial Reynolds numbers defined here as:

\[
Re_\theta = \frac{\omega r_i (r_o - r_i)}{\nu_c} \tag{2.1}
\]

and

\[
Re_a = \frac{u_d (r_o - r_i)}{\nu_d} \tag{2.2}
\]

respectively. In the above equations, \(\omega\) is the angular velocity of the inner cylinder, \(\nu_c\) and \(\nu_d\) are the kinematic viscosities of the continuous and dispersed phases, respectively, and \(u_d\) is the superficial velocity of the dispersed (hexane) obtained by diving the volumetric flow rate of the dispersed phase by the cross-sectional area of the reactor annulus. For the experiments
discussed here, the Reynolds numbers were varied over the following ranges: $6612 < \text{Re}_\theta < 67,448$ and $1.27 < \text{Re}_\theta < 11.55$. It should be noted that for a single-phase batch system with the same geometry (i.e. radius ratio $\eta = 0.78$) of the flow cell used here, the critical azimuthal Reynolds number for onset of laminar Taylor-Couette flow is approximately 90.6, based on a cubic spline fit to published data [35,36]. Since the values of $\text{Re}_\theta$ considered here are 2-3 orders of magnitude greater than the critical azimuthal Reynolds number for single phase flow, it is likely that turbulent vortex flow existed in all the experiments presented here.

In addition to identifying flow patterns in the Taylor-Couette reactor, estimates of hexane droplet holdup were obtained by photographic measurements of the difference in the height of the hexane layer at the top of the flow cell. To accomplish this measurement, the volume of hexane introduced in the system was fixed. The first height measurement was taken when all tubing was filled with hexane and with no droplets being dispersed into the system. With the use of the gear pump to remove and add the dispersed phase simultaneously at any given flow rate, images of the top hexane layer were obtained and compared with the initial height measurement during each test case. To acquire sharp images of the hexane layer, the camera was placed 12 cm from the test section, at the height of the hexane layer. A calibration target of known length and width was placed in the center of the annulus to provide accurate height measurements of the hexane layer.
2.4 Results and Discussion

Examination of images acquired at various operating conditions leads to the identification of at least four distinct flow regimes, examples of which are shown in Figure 2-5. These flow patterns are classified here as 1) a pseudo-homogeneous dispersion 2) a weakly banded dispersion, 3) a horizontally banded dispersion, and 4) a helical dispersion. A phase diagram delineating these flow regimes as functions of $Re_\theta$ and $Re_a$ is shown in Fig. 6.

The pseudo-homogeneous flow pattern, as observed in Figure 2-5(a), occurs at the lowest azimuthal Reynolds numbers investigated ($Re_\theta < 20,000$), irrespective of the value of the axial Reynolds number investigated. Because the minimum value of $Re_\theta$ is significantly
greater than the critical Reynolds number for onset of single phase Taylor vortex flow (and the
dispersed phase holdup is < 3%, as is discussed shortly), one can infer that Taylor vortices are
present but are simply not sufficiently strong to trap rising hexane droplets. However, as the
azimuthal Reynolds number is increased into the range \(2.5 \times 10^4 < \text{Re}_\theta < 4 \times 10^4\), the flow enters
the weakly banded regime, as shown in Figure 2-5(b). This flow regime is characterized by a
dispersion that is no longer spatially homogeneous and there is an obvious banded structure.
Although the location of hexane droplet bands within a Taylor vortex was not identified here
using flow visualization, it is worth noting that the measured distance between adjacent droplet

![Flow regime map based upon photographic observations. Corresponding example images of the identified flow regimes are shown in Figure 2-5.](image.png)
bands was typically found to be 2.1 times the cylinder gap width. A spatial periodicity of twice the gap width suggests that droplets accumulate at vortex inflow boundaries rather than inside the cores of the toroidal vortices, in which case the expected periodicity would be approximately equal to the annular gap width [37]. Lastly, we note that significant migration of droplets between adjacent Taylor vortices in the weakly banded flow regime can be observed in Figure 2-5(b), as evidently the buoyant force is sufficiently strong to cause some rising droplets to escape from one vortex to the next.

When $Re_o > 4.3 \times 10^3$, the vortices have a strong impact on droplet rise trajectories, leading to either a horizontally banded dispersion (as shown in Figure 2-5(c)), or helical dispersion flow pattern (as shown in Figure 2-5(d)). In cases for which $Re_a < 5$, the horizontally banded flow pattern is observed, suggesting that Taylor vortices trap droplets at inflow boundaries, and that droplet movement between adjacent vortices is diminished compared to the weakly banded flow regime. When the imposed dispersed phase axial flow is sufficiently strong (i.e. $Re_a > 6$), the spatial distribution of droplets transitions from the horizontal banded structure to a helical flow pattern wherein relatively few droplets traverse the boundary between adjacent helix windings. It should be noted that the helical pattern is stable in the sense that there is no spatio-temporal change in the helices. This observation is consistent with a hypothesis that the axial flow of the dispersed phase is sufficiently strong to disrupt Taylor vortices and to form helical vortices in their place, as occurs similarly in the case of single phase Taylor vortex flow with an imposed axial feed of working fluid [36].
Figure 2-7 shows hexane holdup as a function of azimuthal and axial Reynolds numbers. Because the top of the hexane layer at the top of the reactor is a free surface and the bottom of this hexane layer is continually being broken by rising hexane droplets, there are innate difficulties in measuring the hexane droplet volume. The resulting error bars in this measurement, shown in Figure 2-7, were computed from the magnitude of observed fluctuations in the thickness of the hexane layer at the top of the reactor. In all cases studied, droplet holdup increases monotonically with both \( \text{Re}_\theta \) and \( \text{Re}_a \). However, for the lowest non-
zero azimuthal Reynolds number considered ($\text{Re}_\theta = 10,000$), hexane holdup is nearly identical to the observed holdup with no cylinder rotation ($\text{Re}_\theta = 0$, annular bubble column) for all hexane flow rates considered. For azimuthal Reynolds numbers greater than 20,000, however, holdup increases approximately linearly with $\text{Re}_\theta$. Note that a value $\text{Re}_\theta = 20,000$ corresponds to the boundary between the pseudo-homogeneous and the weakly banded flow regimes, as shown in Figure 2-6. This observation is consistent with the hypothesis that the increase in hexane holdup with increasing azimuthal Reynolds number is caused by entrapment of rising droplets in Taylor vortices. Further increases in hexane holdup at the highest azimuthal Reynolds numbers studied suggest that the horizontal banded and helical flow structures permit even fewer droplets to escape from one vortex to the next or from one helical winding to the next (as can be verified visually for example in Figure 2-5(c) and 5(d)), thereby leading to increased droplet residence time in the annulus.

### 2.5 Conclusions

An experimental investigation was conducted on a vertically-oriented liquid-liquid Taylor-Couette system with hexane as the disperse phase and water as the continuous phase with continuous feed and removal of the dispersed droplet phase. Four distinct flow regimes were identified, each characterized by the spatial distribution of the droplet phase. At the lowest azimuthal Reynolds numbers considered, a pseudo-homogeneous dispersion was observed irrespective of the value of the axial Reynolds number (i.e., the dispersed phase flow rate). Centrifugal forces generated by rotation of vortices at these relatively low azimuthal Reynolds numbers are easily overcome by the buoyant force acting on rising droplets.
In contrast, when the azimuthal Reynolds number is greater than approximately 30,000, droplets are organized into either banded or spiral structures. This observation is consistent with the interpretation that for the larger azimuthal Reynolds numbers considered, the centrifugal force generated by vortex rotation becomes comparable to the buoyant force acting on droplets, thereby leading to enhancement of droplet concentration in either toroidal or helical vortices and a concomitant increase in droplet holdup. The droplet holdup in this experiment shows a monotonic increase with both azimuthal and axial Reynolds numbers. Although the overall droplet holdup is low, the increase is noted primarily in the identified banded regimes, more so with the axial Reynolds number increasing from \( \text{Re}_a = 6.80 \) to \( \text{Re}_a = 10.19 \). The axial Reynolds number, which in this work was modulated by changing the dispersed phase feed rate, appears to be important for determining the extent to which the axial flow of droplets disrupts toroidal vortices. In summary, the observed flow patterns can be explained in the framework of a competition between wall-driven azimuthal flow of the continuous fluid and buoyancy-driven axial flow of the droplet phase.

Competition between buoyancy-driven axial and wall-driven azimuthal flow also plays a significant role in explaining flow patterns in semi-batch gas-liquid flow in vertical Taylor-Couette reactors [16,18,38]. A significant difference between gas-liquid and liquid-liquid systems is that dispersed phase liquid droplets have greater inertia than do gas bubbles, and as a result they are more capable of disrupting the continuous phase flow field and they also may be less susceptible to confinement near the inner cylinder wall, since there is a smaller density difference to drive the centrifugal separation observed in gas-liquid systems [16,18].
2.6 Acknowledgement

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2.7 References


CHAPTER 3  DROPLET SIZE DISTRIBUTION IN A HEXANE/WATER SEMI-BATCH TAYLOR VORTEX REACTOR

Charlton Campbell  
Department of Mechanical Engineering,  
2025 Black Engineering  
Iowa State University, Ames, IA 50011-2030  
campbel8@iastate.edu

Michael G. Olsen  
Department of Mechanical Engineering,  
2025 Black Engineering  
Iowa State University, Ames, IA 50011-2030  
mgolsen@iastate.edu

R. Dennis Vigil  
Department of Chemical & Biological Engineering,  
2114 Sweeney Hall  
Iowa State University, Ames, IA 50011-2230  
vigil@iastate.edu

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3.1 Abstract

Optical methods were used to measure droplet size distributions in a liquid-liquid Taylor vortex reactor oriented vertically along its main axis and operated in semi-batch fashion with continuous feed of the dispersed phase and no feed or removal of the continuous liquid. The effects of two operational parameters on droplet size distributions were considered, including the inner cylinder rotation rate (azimuthal Reynolds number), and the dispersed phase inlet flow rate (jet Reynolds number). Both the mean droplet diameter and the shape of the droplet size distribution were found to depend upon the jet Reynolds number and were independent of
cylinder rotation speed up to the largest azimuthal Reynolds number investigated (60,000). The shape of the droplet size distribution underwent transitions from a unimodal distribution at low cylinder rotation speeds to a bimodal distribution at intermediate speeds. At the largest rotation speeds considered, the bimodal distribution became right-skewed. These observations provide strong support for the hypothesis that the droplet size and distribution are determined by droplet breakage dynamics at the tips of inlet nozzles, and it can be shown that mean droplet size data collected from two geometrically distinct reactors can be collapsed onto a universal curve that relates the Weber and jet Reynolds numbers.

3.2 Introduction

Taylor-Couette (TC) flow offers a canonical system for studying hydrodynamic instabilities and as such, it has been explored extensively for a single fluid phase. However, many practical applications of TC flow devices involve multiple phases. Some examples include simultaneous extraction and phase separation in the processing of spent nuclear fuels [1], blood and plasma filtration [2], and the culturing of microalgae [3]. In all such multiphase processes, the size and spatial distributions of dispersed phase bubbles, drops, or particles play a crucial role in the performance of Taylor vortex devices.

Here we consider the specific case of two immiscible liquids that constitute a continuous and a dispersed phase. Often, the motivation for creating such droplet systems is to enhance the interfacial area for mass transport via emulsions. Hence, the performance of liquid-liquid Taylor vortex devices depends sensitively on the droplet size distribution, droplet interactions, and for continuous or semi-batch operations also on the fluid holdup. Despite extensive study
of single-phase TC flow, relatively few reports regarding liquid-liquid TC flow have been published, and of those, even fewer mention droplet size variation in the TC system as it relates to the emulsion generation method [4]–[6]. Joseph et al. [4] reported torque measurements and observed the stability of generated emulsions. While operating a horizontally oriented TC device in batch mode and with high dispersed phase hold-up (up to 50%), these investigators reported observing a decrease in droplet size with increasing inner cylinder rotation speed. Qiao et al. [5] took a different approach by observing temporal droplet size evolution in a vertically oriented batch TC system in which droplets were introduced immediately prior to commencing inner cylinder rotation. They observed an increase in the number of small droplets after initiating cylinder rotation followed by a rapid decrease in the number of small droplets due to coalescence. Farzad et al. [6] measured drop sizes in a vertically oriented batch reactor and found that droplet size decreases with increasing cylinder rotation speed.

To further illuminate the understanding of drop size distributions in TC devices, this report addresses the specific case of vertical semi-batch liquid-liquid TC flow. In this system, the dispersed phase is fed and removed continuously, whereas the continuous phase remains in the reactor, which is a configuration well-suited for both chemical and biological applications [7]–[10]. The operating parameters that impact droplet size distribution include the dispersed phase feed rate and inner cylinder rotation speed. In this study, droplet size distributions are measured using optical methods as the cylinder rotation speed and dispersed phase flow rates systematically vary. It is then shown that the mean droplet diameters obtained from these droplet size distributions can be collapsed onto a universal curve by plotting the Weber number
versus jet Reynolds number, even for data obtained in a TC reactor with different geometric characteristics (e.g. inner cylinder radius and gap width).

3.3 Experimental Apparatus and Procedure

3.3.1 Fluid Test Section Description and Operating Procedure

Figure 3-1 shows a schematic of the Taylor vortex flow apparatus used in this study. The device consists of two transparent concentric acrylic cylindrical tubes. The inner cylinder has an outer radius, \( r_i = 15.2 \text{ cm} \) whereas the outer cylinder has an inner radius, \( r_o = 19.4 \text{ cm} \), resulting in an annular gap of 4.2 cm. The thickness of the outer acrylic cylinder is 0.95 cm. The outer cylinder has flanges fitted at both the top and bottom ends that attach to an aluminum top cap and a base plate, respectively. A magnetic coupling provides for vibration-free rotation of the inner cylinder using a motor mounted beneath the bottom plate. A Dayton 3-phase, 1 hp general purpose motor remotely controlled by a Schneider Electric ATV 12 variable frequency drive is used to rotate the magnetic coupling, which was obtained from Magnetic Technologies Ltd. Oil-resistant compressible Buna-N gaskets, obtained from McMaster-Carr, act as a sealant for leak free operation. Fluids fill the 4.2 cm gap-width to a height \( h = 142 \text{ cm} \), resulting in a working fluid volume of 65 L. The resulting radius ratio and aspect ratio associated with the system are given by \( \Gamma = h/(r_o - r_i) = 34 \) and \( \eta = r_i/r_o = 0.78 \), respectively.
Four 16-gauge blunt tip stainless steel nozzles mounted in the baseplate midway between the outer and inner cylinders and evenly spaced azimuthally (90-degree angles) provide the means to introduce the dispersed phase into the system. The system operates in a semi-batch manner by first filling the device with the continuous phase (deionized water) and then continuously injecting a less dense droplet phase through the nozzles. This less-dense continuously fed dispersed droplet phase consists of reagent grade hexane dyed with catalytic amounts of Oil Red O. A gear pump (Cole-Parmer Model No. 75211-30) re-circulates the dispersed phase by siphoning hexane from a phase-separated layer accumulated at the top of the device and injecting it through the nozzles mounted in the base plate as shown in Figure 3-2. For the range of hexane flow rates employed, the dispersed phase hold-up does not exceed 4% by volume.
The operating temperature of all experiments was 20 °C, and no temporal temperature changes were observed, as viscous heating was not important at the low cylinder rotation speeds used and the magnetic coupling between the motor and inner cylinder prevented heating of the fluid by the motor. The DSD experiments spanned a range of two non-dimensional operating variables, including the azimuthal Reynolds and jet Reynolds numbers. The azimuthal Reynolds number was varied by changing the rotation speed of the inner cylinder, and it is defined as

\[
\text{Re}_\theta = \frac{\omega r_i (r_o - r_i)}{v_c}
\]  

(3.1)
In equation (3.1), $\omega$ is the angular velocity of the inner cylinder and $\nu_c$ is the kinematic viscosity of the continuous phase. The jet Reynolds number, defined by equation (3.2), was varied by changing the dispersed phase fluid inlet flow rate through the nozzles.

$$\text{Re}_j = \frac{\nu_j d_n}{\nu_d}$$  \hspace{1cm} (3.2)

Here, $\nu_j$ is the superficial jet velocity based on the hexane volumetric flowrate and the cross-sectional area of the injection nozzle. The terms $\nu_d$ and $d_n$ are the kinematic viscosity of the dispersed phase and the inner diameter of the injection nozzle (1.194 mm), respectively. Various combinations of cylinder rotation speeds in the interval $0 – 90$ RPM ($0 \leq \text{Re}_\theta \leq 5.98 \times 10^4$) and volumetric flow rates in the range $60 – 270$ mL/min ($2,366 \leq \text{Re}_j \leq 10,650$) were chosen to produce a total of 56 unique experimental conditions.

3.3.2 Data Acquisition and Image Processing

A digital camera (Canon Rebel T6i) fitted with a Canon 18-55 mm f/3.5-5.6 zoom lens placed 50 cm in front of the test section (and midway between the top and bottom plates) is used to acquire images of the test section, aided by two off-camera electronic flashes. A matte white backdrop provides contrast to the dyed droplets, as the entire test section is optically transparent at any given angle. Camera RAW image files were converted to TIFF format for use in subsequent analyses. ImageJ, an open-source platform for image analysis, was used for post-processing and thresholding these 16-bit TIFF image files.
Figure 3-3 shows a typical example of a raw image and the corresponding post-processed image used for identifying and measuring droplet diameters. Droplets were observed to be nearly spherical, and therefore droplets that appear to overlap in the image plane can appear to be a single elongated drop. To avoid these perspective artifacts, only droplets with aspect ratios (ratio of major/minor axis length) within the range 1.0 – 1.35 were counted. In addition, to avoid random noise, any identified droplet with size less than 0.3 mm was excluded. For each of the 56 experiments carried out using different combinations of axial and jet Reynolds numbers, 20-40 independent images were processed, resulting in measurement of 772 - 13,140 hexane droplet diameters for a single experiment.
3.4 Results and Discussion

3.4.1 Classification of Droplet Size Distributions

Representative droplet size distributions (DSDs) for several experimental conditions are shown in Figure 3-4, including examples of three characteristic DSD shapes that were observed. Figure 3-4, panels A and B, show unimodal DSDs, exhibiting a single major peak with little asymmetry – a shape observed for the smallest values of \( \text{Re}_j \) investigated. Examples of bimodal DSDs, observed at moderately high jet Reynolds numbers, are shown in Figure 3-4, panels C and D. A third DSD classification, observed at the largest jet Reynolds numbers studied and associated with small mean droplet sizes, can be characterized as right-skewed, as shown in Figure 3-4, panels E and F.

Until recently, the few existing studies on droplet size distributions in liquid-liquid Taylor vortex flow suggested that droplet emulsions in these systems only exhibit right-skewed lognormal droplet size distributions [6], [11]–[14]. However Grafschafter and Siebenhofer [15] recently reported observing bimodal DSDs in a TC contactor system. This discrepancy is attributable to the method of emulsion generation in Grafschafter and Siebenhofer’s system compared with previous reports. In particular, most previous studies were conducted in batch mode wherein the two phases were introduced into the annular space without any attempt at dispersing the droplet phase. After introduction of the two phases, the emulsion was generated
by spinning the inner cylinder at a high angular velocity to break up the dispersed phase. In contrast, the Grafshafter apparatus operated continuously and counter currently, with the
continuous phase fed at the top and the less dense dispersed phase injected at the bottom of the reactor, similar to the method used here.

The existence of bimodal droplet distributions has been associated with the breakup of inlet jets as the dispersed phase is introduced into a continuous phase, whether or not an additional cross flow is present. For example, Li et al. [16] observed a transition from a unimodal distribution to a bimodal distribution of droplets when varying the rotation rate, by using a narrow gap rotor-stator batch mixing device. Similarly, Leiva and Geffroy [17] reported observing a transition from a broad single-peak distribution to a narrower bimodal droplet size distribution in an immiscible liquid-liquid shear flow between two plates. Tang [14], using a quiescent pressurized cylindrical tank with a dispersed phase inlet jet at the bottom, found three types of DSDs including unimodal, bimodal, and right-skewed, depending upon the jet inlet flowrate. The findings from previous investigations of droplet size distributions discussed above, taken together with the observation of several distinct DSDs in this work, suggest that the nature of the DSD in semi-batch vertical liquid-liquid Taylor vortex flow is primarily determined by factors impacting jet breakup near nozzle tips.

3.4.2 Mean Droplet Diameter Dependence on $Re_\theta$ and $Re_j$

In previous experiments on vertical semi-batch liquid-liquid Taylor vortex flow using the same apparatus as in this study, four distinct flow regimes were identified depending on the azimuthal and jet Reynolds numbers [18]. For rotation rates such that $Re_\theta < 25000$, a pseudo-homogeneous dispersion of hexane droplets is observed, regardless of the hexane feed rate.
Similarly, when $2.5 \times 10^4 < \text{Re}_\theta < 4.2 \times 10^4$, hexane droplets appear as a weakly banded dispersion, irrespective of changes in hexane feed rate. However, if $\text{Re}_\theta > 4.2 \times 10^4$, the flow pattern attained does depend upon the hexane flowrate. Specifically, for flowrates with $\text{Re}_j < 6000$, droplets form distinct horizontal bands whilst a value $\text{Re}_j > 6000$ results in a helically banded droplet flow pattern. These flow patterns and their relation to the azimuthal and jet Reynolds numbers provide context for the analysis of droplet diameters.

Figure 3-5. Normalized droplet diameter plotted as a function of azimuthal Reynolds number. Shaded regions denote the indicated flow pattern regimes. Diamond symbols indicate unimodal drop size distribution, circles indicate a bimodal distribution, and square symbols indicate a right-skewed distribution.
Figure 3-5 shows the observed mean droplet diameter, $d_\text{d}$, normalized by the injection nozzle diameter ($d_n$) as a function of the azimuthal Reynolds number for several values of the jet Reynolds number. Corresponding droplet flow regimes are also shown in Figure 3-5, and it is evident from this plot that the mean drop size depends only weakly (and apparently non-systematically) on $\text{Re}_\theta$, a variable that primarily determines the fluid flow regime accessed. This weak dependence of droplet diameter on azimuthal Reynolds number suggests that droplet size is determined primarily at the point of injection, with little subsequent downstream change due to coagulation and breakage. Figure 3-5 also demonstrates that flow pattern selection is primarily governed by the cylinder rotation speed and is independent of the mean drop size when $\text{Re}_\theta < 4.2 \times 10^4$. However, when $\text{Re}_\theta > 4.2 \times 10^4$, the flow pattern selection clearly depends upon the jet Reynolds number and a helical spatial distribution of droplets correlates to small mean droplet sizes (large $\text{Re}_j$) whereas large droplets (small $\text{Re}_j$) are associated with strong horizontal banding of droplet swarms.

Figure 3-6 shows that the dependence of droplet diameter on jet Reynolds numbers is non-monotonic such that a maximum droplet size is reached when $\text{Re}_j$ is in the approximate range $5000 < \text{Re}_j < 6000$. At $\text{Re}_j = 2366$, the semi-turbulent [19] dispersed phase flow forms an axisymmetric jet [20] which then develops instabilities, leading to the sudden decrease in drop diameter at $\text{Re}_j = 3549$. As the instability increases with $\text{Re}_j$, smaller droplets begin appearing in the flow, leading to a decrease in mean droplet diameter as well as the bimodal droplet size distributions observed at $\text{Re}_j = 7098$. 
3.4.3 Similarity Relation for Droplet Diameter

The Weber number, which characterizes the ratio between the inertial force and surface tension, plays a significant role in determining droplet size in agitated liquid-liquid emulsions, and therefore we can anticipate that it will be an important part of a similarity criterion capable of collapsing droplet size data obtained using Taylor vortex reactors operated at different cylinder rotation speeds and dispersed phase injection flow rates. The data presented in
Section 3.2 shows that droplet size in a vertical liquid-liquid semi-batch Taylor vortex reactor primarily depends upon the jet Reynolds number, and this observation implies droplet diameter is determined by dynamics at the tip of the injection nozzle. It follows that, in the current context, the Weber number should be defined using the characteristic jet velocity, rather than the azimuthal velocity, as shown in equation (3.3).

\[ \text{We} = \frac{\rho_d v_d^2 d_d}{\sigma} \]  (3.3)

In order to generalize a relation between \( \text{We} \) and \( \text{Re}_j \) capable of predicting mean droplet diameter in reactors with geometries (e.g. radius ratio, \( \eta \), and needle diameter) different than the reactor used here, it is necessary to add a scaling factor to equation (3.3) that accounts for differences in the ratio of annular and injection nozzle cross-sectional areas, as shown in equation (3.4).

\[ \text{We}_g = \frac{\rho_d v_d^2 d_d}{\sigma} \left( \frac{r_i (r_o - r_i)}{d_n^2} \right) \]  (3.4)

The applicability of equation (3.4) for predicting droplet size in a vertical semi-batch Taylor vortex reactor that is not geometrically similar to the apparatus described in Section 3.3.1 can be tested by considering the results of droplet size measurements using a second Taylor vortex reactor with significantly different geometry (radii, radius ratio, and ratio of gap width to length). Here, we chose to carry out such experiments (using hexane and water) in a significantly smaller apparatus than the one described in Section 3.3.1. Table 3-1 shows a
comparison of the geometrical specifications of the two reactors, labeled (L) for the large
reactor described in Section 3.3.1 and (S) for the small reactor.

Table 3-1  Comparison of dimensional specifications for droplet size
distribution studies in a large (L) and small (S) Taylor vortex reactors.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>(L)</th>
<th>(S)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inner Cylinder Radius ( r_i )</td>
<td>15.2 cm</td>
<td>3.81 cm</td>
</tr>
<tr>
<td>Outer Cylinder Radius ( r_o )</td>
<td>19.4 cm</td>
<td>5.08 cm</td>
</tr>
<tr>
<td>Radius Ratio ( \eta )</td>
<td>0.79</td>
<td>0.75</td>
</tr>
<tr>
<td>Height of Fluid Region</td>
<td>142 cm</td>
<td>48 cm</td>
</tr>
<tr>
<td>Aspect Ratio ( \Gamma )</td>
<td>34</td>
<td>38</td>
</tr>
<tr>
<td>Nozzle Diameter</td>
<td>1.194 mm</td>
<td>0.413 mm</td>
</tr>
<tr>
<td>Re range</td>
<td>0-59,751</td>
<td>5,067-20,268</td>
</tr>
</tbody>
</table>

A plot of the adjusted Weber number defined by equation (3.4) as a function of the jet
Reynolds number is shown in Figure 7, and it is evident from this plot that these two
dimensionless quantities are sufficient to collapse the droplet size data onto a universal curve
that is largely insensitive to the azimuthal Reynolds number, except perhaps at high jet
Reynolds numbers. Furthermore, in contrast to the plot of normalized droplet diameter versus
jet Reynolds number (Figure 3-6), Figure 3-7 exhibits a monotonic and approximately linear
increase in \( \text{We}_a \) with increasing \( \text{Re}_j \), eventually leading to a plateau at the largest jet Reynolds
numbers considered.
The plateau in the value of $\text{We}_a$ at $\text{Re}_j = 7098$ is a consequence of the fact that the mean droplet diameter decreases with increasing $\text{Re}_j$ (see Figure 3-6) in a manner that offsets the concurrent increase in the nozzle jet velocity. It is worth noting that the jet Reynolds number at which the plateau in Weber number is observed occurs at the same value of $\text{Re}_j$ at which the droplet size distribution changes from unimodal to bimodal, suggesting that a change in the droplet breakup mechanism near the injection nozzle occurs at this critical $\text{Re}_j$. Specifically, the
onset of a bimodal distribution reflects the appearance of small droplets that are formed during shear-induced breakage of drops emerging from the nozzle tip. Figure 3-7 also shows that the appearance of the plateau is accompanied by an increase in scattering of the data.

### 3.5 Conclusions

Drop size distributions in a vertical, semi-batch liquid-liquid Taylor-Couette flow device were measured using photographic methods for a range of combinations of inner cylinder rotation speed and dispersed phase flow rate. Three distinct types of DSDs were observed, including unimodal, bimodal, and skewed distributions, depending only on the jet Reynolds number. Specifically, values of \( \text{Re}_j < 7098 \) resulted in unimodal DSDs, bimodal distributions were observed for \( \text{Re}_j = 7098 \), and right-skewed distributions were produced when \( \text{Re}_j > 7098 \). Similarly, the mean droplet diameter was found to depend only on \( \text{Re}_j \) and it is independent of the azimuthal Reynolds number. This fact suggests that droplet size distributions are primarily determined at inlet nozzle tips, with relatively little downstream coagulation and breakage. Indeed, at least for the range of azimuthal Reynolds numbers considered, the changes in DSD and mean droplet size are consistent with droplet generation mechanisms that have been elucidated for injection of droplets into quiescent droplet columns using nozzles. Lastly, it has been demonstrated that the mean droplet size in vertical semi-batch liquid-liquid Taylor vortex flow can be well-predicted from an empirical plot of Weber number versus jet Reynolds number, and data collected from two reactors that do not preserve geometric similarity and that have widely different sizes collapse onto this universal Weber number-jet Reynolds number curve.
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3.7 References


4.1 Abstract

The entry conditions in a semi-batch Taylor vortex reactor (TVR) was explored. Using two immiscible liquids, hexane and water, in a TVR such that the secondary hexane phase enters through nozzles in the system provided insight to the droplet sizes and distribution within the system. Optical experiments resulted in the identification of four jet breakup behaviors. A) Regime I – Varicose jetting, B) Regime II – Sinuous jetting without entrainment C) Regime III – Sinuous jetting with entrainment, and D) Regime IV – Atomized jetting. These
regimes confirmed that the droplet size distribution is strongly dependent on the jet Reynolds number.

4.2 Introduction

A Taylor vortex reactor (TVR) consists of one or more fluids confined to the annular space between a rotating inner cylinder and a concentric fixed outer cylinder. When two immiscible liquids are present, these devices can be used to generate emulsions without the use of impellers, which produce highly non-homogeneous turbulence. In many applications of liquid-liquid TVR flow, such as for extraction or carrying out chemical reactions, emulsion droplet size distributions play a key role in determining device performance because of the critical role that interfacial area can play in interphase mass transfer [1], [2].

In most previously studied liquid-liquid TVR systems, droplets form as a result of fluid shear produced by rotating the inner cylinder at sufficiently high speeds in co-flowing [3]–[7] or batch operation [8]–[11]. However, it is also possible to generate droplets in these systems by injecting the dispersed phase into the continuous phase via a nozzle. Here, we consider such a system operated in semi-batch mode (continuous feed and removal of the dispersed phase, no feed or removal of the continuous phase). Operating a TVR in this manner leads to droplet development governed not only by wall-driven turbulent shear, but also by hydrodynamic instabilities that occur at the tip of the injection nozzle. Indeed, in a previous investigation, the drop size distribution in a semi-batch vertically oriented TVR with continuous feed of the dispersed phase showed much stronger dependence on the volumetric flow rate of the dispersed phase than it did on the rotation speed of the inner cylinder [12]. This result is in
stark contrast to the analogous gas-liquid counterpart where gas bubble sizes not only grow downstream from the injection site, but the bubble size distribution shows a strong dependence on the rotation rate of the inner cylinder.

Several previous studies on drop formation due to breakup of submerged jets in liquid-liquid systems [13]–[18] have been published, although most of these reports are limited to the case when an immiscible liquid jet enters a quiescent liquid, in contrast to the current study wherein an immiscible liquid jet enters a second liquid undergoing a turbulent crossflow. For example, a few studies have focused on liquid plumes formed as a result of deep oil spills [19]–[21]. However, several experimental investigations of liquid jets entering a gaseous crossflow for applications such as fuel injection systems, and various mechanistic features of jet breakup in these systems have been identified and classified, such as jet trajectories and jet atomization [22]–[31]. These previous investigations of systems where either a liquid jet enters a gaseous crossflow or a liquid jet enters a quiescent immiscible liquid are useful in characterizing the entry conditions of the present study, as described in subsequent sections. Thus, the purpose of this report is to characterize the jet entry conditions in a semi-batch Taylor-Couette system, in accordance with the aforementioned studies and to add to the minimal literature available for liquid-liquid jet systems. Furthermore, given that the droplet diameter of the system is dependent on the jet inlet condition, this paper seeks to provide an explanation for the system’s droplet diameter variation.
4.3 Control Parameters

Two variables, including volumetric feed rate of the dispersed phase and inner cylinder rotation speed, were manipulated in order to elucidate the effects of these variables on jet breakup and drop generation in a semi-batch, vertical TVR. Because the dispersed phase was introduced into the TVR through nozzles, the appropriate dimensionless parameter characterizing the strength of the dispersed phase feed rate is given by the jet Reynolds number, $Re_j$, defined as

$$Re_j = \frac{V_d D_N}{\nu_d}$$

(4.1)

where $V_d$ is the velocity of the dispersed phase at the nozzle exit, $D_N$ is the nozzle diameter, and $\nu_d$ is the kinematic viscosity of the dispersed phase. This quantity has been used extensively in previous studies of jet breakup to characterize various breakup regimes.

In the TVR considered here, the angular velocity of the inner cylinder can be used to vary the strength of the cross flow at the nozzle tip, and therefore we define an azimuthal Reynolds number, $Re_\theta$, given by equation (4.2).

$$Re_\theta = \frac{V_c (r_p - r_i)}{\nu_c}$$

(4.2)

The characteristic velocity of the continuous phase, $V_c$, is chosen as the linear velocity on the surface of the rotating inner cylinder, i.e. $V_c = \omega r_i$, where $\omega$ is the angular velocity. The characteristic length scale in Eq. (4.2) is given by the annular gap width and the kinematic viscosity of the continuous phase is used because of the low holdup (< 5%) of the dispersed
phase observed for all conditions studied. For consistency with terminology used in previous studies of liquid jet breakup in gaseous crossflows, the azimuthal Reynolds will henceforth be referred to as the crossflow Reynolds number as it relates to the conditions at the nozzle tips.

Based upon our earlier study of droplet size distributions in semi-batch vertical liquid-liquid Taylor vortex flow [12] and on studies of liquid jet breakup in gaseous crossflows, it is useful to define a crossflow Weber number,

\[ \text{We}_c = \frac{\rho_c V_c^2 D_n}{\sigma} \]  

(4.3)

where \( \rho_c \) represents the density of the continuous phase, and \( \sigma \) describes the interfacial surface tension between the two immiscible phases. This crossflow Weber number is useful for classifying jet breakup regimes caused by shearing due to the perpendicular flow of the continuous phase. Finally, the momentum flux ratio, shown in equation (4.4), has been used extensively to characterize jet breakup regimes, such as by Leask et al. [29], where \( \rho_d \) represents the dispersed phase density.

\[ q = \frac{\rho_d V_d^2}{\rho_c V_c^2} \]  

(4.4)

### 4.4 Experimental Methods

#### 4.4.1 Working Fluids

All experiments were carried out at 20 °C. Deionized water with density \( \rho_c = 998 \text{ kg/m}^3 \) and viscosity \( \nu_c = 1.0 \times 10^{-6} \text{ m}^2/\text{s} \) was chosen as the continuous phase fluid. Reagent grade hexane purchased from Fisher Scientific, was used as the continuously-fed dispersed phase and
the measured density and kinematic viscosity are \( \rho_d = 659 \, \text{kg/m}^3 \) and \( \nu_d = 4.51 \times 10^{-7} \, \text{m}^2/\text{s} \). The interfacial surface tension between the two fluids was measured using a Du Nouy ring tensiometer (Fisher Model 21 tensiomat) and the value measure was \( \sigma = 52.4 \times 10^{-3} \, \text{N/m} \).

4.4.2 Taylor Vortex Reactor

The vertically oriented semi-batch TVR consists of two concentric acrylic cylinders. The gap width between the two cylinders is 4.2 cm, with annular inner and outer radii of \( r_i = 15.2 \, \text{cm} \) and \( r_o = 19.4 \, \text{cm} \), respectively. In Taylor vortex flow devices, the radius and aspect ratios of the system are parameters known to impact flow pattern selection, and the values for

Figure 4-1. Image rendering at the entry section of the semi-batch TC reactor. Shown at the very center of the system is one half of the magnetic coupling.
the apparatus described here are \( \eta = r_i / r_o = 0.78 \), and \( \Gamma = h / (r_o - r_i) = 34 \), respectively, where \( h = 142 \text{ cm} \) is the height of fluid in the annulus.

The flanged outer cylinder is affixed to an aluminum baseplate and an aluminum top cap. The inner cylinder, which contain stainless steel shafts at both ends, rotates via a magnetic coupling to minimize motor vibrations from being transmitted to the cylinder. Concentricity of the cylinders is maintained by stainless-steel shafts on the inner cylinder rotation axis and centered in holes in the baseplate and the top caps. Low friction sleeve bearings placed within these holes eliminate metal on metal contact. A lubricated turntable from McMaster-Carr eliminates rotational friction on the top aluminum. A 1-hp general purpose motor, coupled with an MTD-20 magnetic disc coupling from Magnetic Technologies Ltd., rotates the inner cylinder. A Schneider Electric ATV 12 variable frequency drive was used to remotely control the motor.

4.4.3 Reactor Operation and Data Acquisition

The continuous phase fluid (deionized water) is placed into the annular gap prior to rotating the inner cylinder and continuously injecting the less dense dispersed phase (hexane) through four equally spaced (azimuthally) threaded, blunt-tipped nozzles located on the baseplate at the midpoint of the annular gap, as shown in Figure 4-1. Each nozzle has an inner and outer diameter of 1.194 mm and 1.651 mm, respectively. The rising hexane droplets accumulate at the top of the annular column to form an approximately 5-cm thick phase-separated hexane layer between the water-hexane emulsion and a small air-filled head space. This phase-separated layer of hexane is then cycled back to the injection nozzles at the bottom
of the reactor via four gear pumps (Cole–Parmer model no. 75211-30, one for each nozzle). The operating RPM range for the inner cylinder is 15-90 RPM, which corresponds to $9.96 \times 10^3 \leq \text{Re}_\theta \leq 5.98 \times 10^4$. The hexane volumetric flowrate was varied from 40-270 mL/min, corresponding to a jet Reynolds number range $1.58 \times 10^3 \leq \text{Re}_j \leq 1.06 \times 10^4$.

Hexane jet breakup regimes were identified by analyzing photographic images using a high-speed camera (Photron FASTCAM APX RS), fitted with a Canon 18-55 mm f/3.5-5.6 zoom lens, situated 10 cm in front of the outer cylinder and centered on one of the inlet nozzles. A Lowel ViP Pro LED provided backlight illumination to increase contrast. The high-speed camera generates video files of jets at 3000 frames per second, within a 512x1024 pixel field of view. Images acquired from a total of 45 experiments at various combinations of azimuthal and jet Reynolds numbers contributed to the analysis provided in subsequent sections.

### 4.5 Results and Discussion

Based upon analysis of images acquired under various combinations of hexane flow rate and inner cylinder rotation speed, four distinct jetting regimes were identified in the semi-batch TVR described here and representative images of these regimes are shown in Figure 2. The jetting behaviors shown closely mirror those identified in a recent study by Saito et al. of a liquid jet entering through a nozzle into a quiescent pool comprised of a second immiscible liquid [32]. Because of strong similarities in the observed jetting behavior between the two studies, the jetting regimes shown in Figure 2 were organized according to the classifications described by Saito et al. Regime I, known as varicose jetting, occurs when droplets detach from the ends of smooth, tendril-like columns of the disperse phase fluid [14], [15], [33].
A transition from Regime I to Regimes II is observed upon increasing $Re_j$, which produces sinuous-like movements of the inlet jet due to a Kelvin-Helmholtz [32] instability. In Regime II, drops are primarily produced by breakage at the tip of the sinuous column of dispersed phase fluid, whereas in Regime III, in addition to droplets being generated by column breakage at the tip of the jet, also features the appearance of relatively smaller droplets on the leeward side of the primary sinuous column (due to transport by the cross flow) generated by shear-induced breakage from the column surface. Regime IV – also referred to as atomized jetting, exhibits an even broader distribution of droplet sizes due to the generation of more and smaller droplets (than Regime III) arising from column surface breakage.
In a previous study, four distinct reactor-scale flow patterns in the TVR considered here were identified for various combinations of azimuthal and jet Reynolds numbers [34], as shown in Figure 4-3. These patterns include a pseudo-homogeneous flow regime observed at relatively small values of $\theta Re$ (and independent of $Re_j$) in which dispersed phase droplets disperse uniformly throughout the annulus as they rise through the reactor. At intermediate azimuthal Reynolds numbers (again, independent of the jet Reynolds number), some periodic horizontal banding of droplet swarms is observed, but a considerable number of droplets continue to rise freely throughout the system. At high values of $\theta Re$ and relatively low jet
Reynolds numbers, the horizontal bands become very distinct due to centrifugally-driven concentration of almost all of the less-dense droplets at the inflow boundaries of Taylor vortices. Lastly, for large values of both $Re_\theta$ and $Re_j$, the concentrated droplet swarm forms a helical coil. In addition to the droplet flow pattern regime map shown in Figure 4-3, the observed jetting behavior is also plotted. In contrast to the flow pattern regime map, which depends almost exclusively on the value of the azimuthal Reynolds number (except for at the highest values of considered), the jetting regime depends strongly on the value of the jet Reynolds number and is relatively insensitive to the azimuthal Reynolds number.

Figure 4-4 shows a comparison of the jetting regimes described here to those observed in the experiments by Saito et al. [32]. Although both studies consider an immiscible liquid injected via a nozzle into a second liquid phase, there exists two important differences in the experiments performed. First, in the present study a crossflow is present, whereas Saito used a quiescent continuous liquid phase. Second, in the experiments presented here the dispersed phase is less dense than the continuous phase and it enters the TVR at the bottom of the reactor. In contrast, the liquid jet is comprised of a dense fluid that enters the top of the flow cell so that droplets fall through the less dense quiescent continuous phase in Saito et al.’s experiments.

Saito et al. classified jetting regimes using the Ohnesorge number [35],

$$Oh = \frac{\mu_d}{\sqrt{\rho_d \sigma D_N}}$$  \hspace{1cm} (4.5)

as it compares the Rayleigh timescale ($t_R \sim \sqrt{\rho_d D_N^3/\sigma}$), and the viscocapillary ($t_{visc} \sim \mu_d D_N/\sigma$) timescale. Both timescales are used to characterize breakup of an inviscid fluid jet, and
thinning dynamics of a viscous liquid thread [36], [37], respectively. Here, $\mu_d$ is the dynamic viscosity of the dispersed phase.

Figure 4-4 shows that a jet breakup regime map can be constructed by plotting the Ohnesorge number against the jet Reynolds number on a log-log scale. Saito et al. [32], who

Figure 4-4. Breakup regimes identified in the present study compared with a previous study by Saito et al. in 2017. Dashed lines corresponding to $Oh = 2.8Re^{-1}$ and $Oh = 22Re^{-1}$ indicate transition from regimes I-II and III-IV, respectively, in a liquid-liquid system with no crossflow, while the line $Oh = 9.5Re^{-1}$ indicates the transition from regimes III-IV in a liquid-liquid system with crossflow.

Figure 4-4 shows that a jet breakup regime map can be constructed by plotting the Ohnesorge number against the jet Reynolds number on a log-log scale. Saito et al. [32], who
carried out experiments with no crossflow, identified transitions from regimes I to regime II (\( \text{Oh} = 2.8\text{Re}^{-1} \)), and from regimes III to regime IV (\( \text{Oh} = 22\text{Re}^{-1} \)). Note that the line demarcating regimes I and II coincides with is consistent with the transition from regime I to regime II observed in the present study. However, the transition between regimes III and IV observed by Saito et al. occurs along a different boundary in Figure 1-4 than the data obtained in the present study. Specifically, the regime III-IV transition in the present experiments, which were carried out in the presence of a crossflow, occurs at lower jet Reynolds numbers consistent with a boundary line defined as \( \text{Oh} = 9.5\text{Re}^{-1} \).

In addition to comparing the observed jet breakup behavior in this study to the findings of Saito et al. for jet breakage in a quiescent liquid, it is also interesting to compare these results with the larger available body of literature concerning liquid jet breakup in gaseous crossflow. Most of these liquid-gas studies identify jet breakup regimes on phase maps using the crossflow Weber number, \( \text{We}_c \), as one of the organizing parameters. For example, a commonly used breakup regime map is the \( \text{We}_c - q \) map, which makes use of the relative strengths of the jet and crossflow, as quantified by the momentum flux ratio \( q \), defined in section 1.3. Wu et al. [38] proposed a \( \text{We}_c - q \) breakup regime map based upon experiments using aqueous liquid jets with various compositions injected transversely in a crossflow of air. Six breakup regimes were identified by Wu et al., and boundary lines for three of these regimes are plotted in Figure 4-5.
Wu et al. classify jet breakup under low crossflow Weber numbers, $\text{We}_c < 10$, as the enhanced capillary breakup regime, since inertial force associated with crossflow are relatively small compared with the surface tension force. A capillary breakup regime was not observed in the current study due to the limited range of inner cylinder rotation speeds investigated. A second breakup regime characterized by Wu et al. as “column breakup” occurs when $\text{We}_c > 10$.

Figure 4-5. Comparison of the liquid-liquid jet breakup regimes observed in this study with regimes observed by Wu et al. [35] for aqueous liquid jets breaking up in gaseous (air) crossflow. The dashed red lines separate the proposed breakup regions based on results of the present study and the dashed grey lines separate the same transition regions identified by Wu et al., although unmarked in the figure.
resulting in droplets formed from the breakup of ligaments and bag-like structures that break away from the main jet column, referred to as the secondary droplet formation process. For the current study, this “column breakup” regime occurs when $We_c > 0.8$, however, stable droplets are immediately formed after breaking away from the main liquid column, as opposed to the secondary droplet formation described for Wu et al.’s study. Coincidentally, the column breakup process spans Regimes I and II of the present study. Thirdly, Wu associate “surface breakup” with a combination of drop formation by pinch-off and by shearing of the jet column resulting in the appearance of small droplets on the leeward side of the jet. When the value of $q$ is low, the liquid jet experiences column breakup without surface breakage, while surface breakup occurs at higher values of $q$. The surface breakage criterion is a function of the $We_c$, identified by the sloped line in Figure 4-5. Although multiple modes of breakup occur in this regime, the primary modes are surface and column breakup but for simplicity and alignment with other classification schemes Wu et al. refer to this regime as simply surface breakup. Not surprisingly, the boundaries identified in the liquid-gas study by Wu et al. do not align with boundaries for the breakup regimes I-IV identified in the current study. Nevertheless, if one associates regimes I and II with “column breakage” and regimes III and IV with “surface breakage”, the resulting flow regime map would be reminiscent of the map produced by Wu et al. except that the linear column-surface boundary is shifted to lower values of $q$ and the vertical capillary breakage boundary occurs at a crossflow Weber number approximately one order of magnitude smaller. One explanation is based on surface tension forces associated with the drop breakup process. In the case of the present liquid-liquid study, droplets form immediately once separated the main liquid jet column due to stronger surface tension forces
that the crossflowing momentum of the fluid. In the gas-liquid case, the crossflowing aerodynamic forces tend to be stronger than the surface forces resulting in droplets being formed from the secondary breakup process described earlier.

Figure 4-6. Defining a transition from laminar to turbulent regime for the liquid-jet in a liquid rotating crossflow as compared to a liquid-gas study [36].

As discussed above, the Ohnesorge number and momentum flux ratios plotted against jet Reynolds number have been used with some success to construct jet breakup regime maps in quiescent liquids and in gas crossflows, respectively. However, a simpler way of determining
breakup regime boundaries is to consider how these flow regimes are related to the transition of the jet from laminar to turbulent flow, as suggested by Madabhushi et al. [39]. For example, Figure 4-6 shows a breakup regime map from the data obtained in the current study plotted in \( \text{Re}_j-\text{We}_c \) space. The blue horizontal line is the critical \( \text{Re}_j \) for transition of the jet from laminar to turbulent flow, as reported by Madabhushi et al. in their study of liquid jets in gaseous crossflow [39]. Note that this laminar-turbulent transition occurs at a value of \( \text{Re}_j \) that is close to the value that defines the transition between breakup regimes II and III (dashed line). As was discussed previously, the transition from regimes II and III can also be associated with transition from column to surface breakup regimes.

The jet breakage regime map in Figure 4-6 demonstrates that the jet Reynolds number is the dominant factor in determining the primary breakage mechanism. Consequently, it is useful to consider how \( \text{Re}_j \) (and hence the breakage regime) impacts droplet size distributions in the TVR. Utilizing data for droplet sizes associated with \( \text{Re}_\theta = 2.98 \times 10^4 \) in Campbell et al.’s investigation [34], it is possible to infer droplet sizes, \( D_\theta \), from different regimes, and vice versa. Figure 4-7 shows a number of unique features. There is an initial decrease in the mean drop diameter, transitioning from Regime I to Regime II. This initial decrease is a result of increased hydrodynamic instabilities of the surface of the jet as seen in Figure 4-2B. The transition from Regime II – III leads to a rise in the mean drop diameter as droplets continue to form at the end of the jet column, with more fluid entering droplets prior to detachment [40]. The droplet size decreases again transitioning from Regimes III – IV due to the crossflow now having some impact on the jet, shearing droplets from the leeward edge of the jet. This surface breakup mechanism results in a broader droplet size distribution because
of the increase in small size droplet population, which in turn causes a significant decrease in the mean drop size.

![Figure 4-7. Correlation of the four identified regimes with droplet diameter, based on Re_j.](image)

### 4.6 Conclusions

In a semi-batch Taylor vortex reactor that utilizes nozzles to inject the dispersed phase liquid into a rotating immiscible liquid, it was shown that the jet Reynolds number is the dominant factor that determines the breakup mode of the jet. Because of our previous finding [12] that the strength of the crossflow, quantified by the azimuthal Reynolds number Re_ϕ, has little
impact on downstream evolution of droplets in semi-batch TVRs, it can be surmised that $Re_j$ is also the dominant factor in determining downstream droplet size distribution. This hypothesis also provides a rational explanation for the experimentally-observed non-monotonic dependence of mean droplet size on jet Reynolds number shown in Figure 1-7. In addition, four jet breakup regimes were identified and compared to analogous liquid jet breakage mechanisms for injection into quiescent liquids or gas phase crossflows. At the lowest jet Reynolds numbers considered, the breakup mode is primarily due to a competition amongst buoyant and interfacial forces, in addition to the liquid momentum. As the jet Reynolds number increases, other factors become important including destabilization of the cylindrical jet fluid column into a sinuous shape and formation of small droplets due to shearing from the surface of the fluid column.

4.7 Acknowledgements

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4.8 References


CHAPTER 5  GENERAL CONCLUSIONS

5.1 Review of Results

Operating a Taylor-Couette (TC) reactor in a semi-batch manner offers unique control in emulsion generation when using two immiscible liquids. Several experiments were performed on the TC system, the first of which was to observe bulk flow patterns while varying the rotation rate of the inner cylinder and the flow rate of the dispersed hexane phase. Additional experiments were carried out to observe the droplet size distribution within the system as well as to characterize different entry conditions of dispersed phase jet breakup in the presence of the rotating liquid crossflow.

Initial optical experiments revealed four distinct flow patterns characteristic of the spatial distribution of the dispersed phase. At the lowest rotation of the inner cylinder, the droplets rose through the system despite the changing flowrate. This pattern was characterized as the pseudo-homogenous regime as droplets showed no unique structure as they rose through the system. Increasing the rotation of the inner cylinder led to another observation where the droplets showed weak banding for all dispersed phase flowrate. Further increasing the inner cylinder rotation rate led to two banding patterns, primarily characterized by the dispersed phase flow rate. At the highest studied rotation rate, a lower dispersed phase flowrate led to horizontal banding of the droplets and at higher flowrates, the droplets rose in a helical band towards the top of the system. The banding is best explained as droplets trapped in developed Taylor vortices along the vertical axis of the system. The experimental holdup was
observed and showed a monotonic increase with both the rotation rate and dispersed feed flowrate. The secondary phase holdup was greatest at the highest rotation and feed rate, but never exceeded 4%.

A second set of optical experiments focused on the droplet size distributions associated with varying the rotation and feed rates. Results showed three droplet size distributions: a unimodal distribution, a bi-modal distribution, and a right-skewed distribution. Experiments revealed that the distributions were not dependent on the rotation rate of the inner cylinder but on the dispersed phase flowrate. The drop size distribution varied from a unimodal distribution, observed at lower dispersed flowrates, to a right-skewed distribution characterized at higher dispersed flowrates, with a single transitory flowrate characterizing the bi-modal distributions. Further investigations into the sizes of the droplets also revealed a strong dependence on dispersed phase flow rate. The droplet size was observed to increase to a maximum diameter then decrease to a minimum at the highest observed dispersed phase flowrate.

A third set of experiments focused on the entry conditions of the dispersed phase flowrate. Four breakup regimes were identified, a varicose jetting regime, a sinuous jetting regime, with and without entrainment droplets, and finally an atomized jetting regime. While the breakup modes primarily relied on the dispersed phase flowrate, there was some dependence on the rotation rate of the inner cylinder, primarily at the highest observed rotation and feed flowrates. The observed breakup modes provided insight into the variation of
the droplet size, where the drop size decrease is best explained by the atomized jetting breakup regime.

### 5.2 Future Work

The current system has much potential for further investigation without significant funding efforts. To best identify such investigative efforts, it would be necessary to investigate non-dimensional parameters that govern the system. Chapters 2 – 4 introduced dimensionless parameters such as the Reynolds number governing both the rotational flow and the axial flow of the dispersed hexane phase. Although discussed briefly in Chapter 3, the Weber number,

\[ We = \frac{\rho V^2 D_d}{\sigma} \]  

(5.1)

sheds insight into the breakup mode of the jets and subsequently the droplet sizes. Here, \( \rho \) and \( V \), are the density and velocity of the dispersed phase, respectively, \( \sigma = 52.4 \times 10^{-3} \) N/m is the interfacial surface tension between the hexane-water pair, and \( D_d \) is the diameter of the droplets. The droplet experiments covered a range of \( 20 \leq We \leq 250 \). Lower Weber numbers indicate strong interfacial forces leading to larger drop sizes and as that Weber number increases, inertial forces effect the dispersed liquid jet, leading to smaller drop sizes. Plotting the Weber number against the fixed viscosity ratio of 0.45, it is possible to see the vertical transition between the breakup modes of the droplets [1]. Unfortunately, a single liquid pair would not shed much insight into a generalization of the breakup regimes. The Ohnesorge number,
\[ Oh = \frac{\mu}{\sqrt[3]{\rho \sigma D_d}} \] 

where \( \mu \) is the density of the dispersed, was discussed in Chapter 4, also aids in identifying the breakup modes of droplets, and shows the impact that viscous forces have on surface tension forces. For the present system, the Ohnesorge values have a range between 0.0013 and 0.0022. As the Ohnesorge number incorporates the Weber number, it serves as a more useful identifier of the drop formation regimes, when plotted with the Reynolds number. There are two other unique dimensionless parameters which aids in classifying droplets in liquid-liquid systems. These are the Morton number, \( M = g \mu^4 \Delta \rho / \rho^2 \sigma^3 = 2.3 \), with \( \Delta \rho \) is the density difference between the hexane-water pair, and the Bond (Eötvös) number, \( \text{Bo} = g \Delta \rho D_d^2 / \sigma \), which has a range of \( 0.05 < \text{Bo} < 0.5 \). Together, these two numbers describe the shape of the droplets as they rise through the system. For the current system parameters, the Bond and Morton numbers indicate droplets maintaining their sphericity [2] as observed in the study. Furthermore, similar to the Weber and the Ohnesorge numbers, the Bond number is dependent of the surface tension forces, i.e. the Bond number relates gravitational to surface tension forces. An interesting quality of the Bond number is that incorporates the density difference of the liquid pairs. As such, reducing that density difference would serve to increase not just the holdup of hexane droplets but also the residence time of the dispersed phase.

Using silicone oil 350 cSt (CAS 63148-62-9), which has a density and viscosity of 970 kg / m\(^3\) and \( 42.5 \times 10^{-3} \text{ N} / \text{m} \) [3], respectively, and assuming similar droplet size ranges, the Bond numbers
would potentially be lowered to \(0.006 < Bo < 0.05\), which is an order of magnitude lower than the hexane-water pair used in the present study.

5.3 References

