Dynamics of a highly viscous core flow in a microfluidic flow focusing system

A thesis submitted in fulfilment of the requirements for the degree of Master of Engineering

Michael E. Kurdzinski
Bachelor of Mechanical Engineering, RMIT University

School of Engineering
College of Science, Engineering and Health
RMIT University

December 2016
Declaration

I certify that except where due acknowledgement has been made, the work is that of the author alone; the work has not been submitted previously, in whole or in part, to qualify for any other academic award; the content of this thesis/project is the result of work which has been carried out since the official commencement date of the approved research program; any editorial work, paid or unpaid, carried out by a third party is acknowledged; and, ethics procedures and guidelines have been followed.

Michael E. Kurdzinski

6 December 2016
To my beloved wife Berrak
Acknowledgements

First, I would like to express my profound gratitude to my senior supervisors, Dr. Khashayar Khoshmanesh and Professor Arnan Mitchell for their support throughout my Master’s candidature. Their knowledge and guidance through this time was paramount on completing my thesis. Their ability to motivate and inspire me combined with their continuous support gave me the ability to present this thesis.

I would like to thank researchers and students within the School of Engineering: Dr. Francisco Tovar-Lopez, Dr. Mahyar Nasabi, Dr. Kiplemo Yego, Dr. Shi-Yang Tang, Dr. Sivacarendran Balendhran, Dr. Kyle Berean, Mr. Peter Thurgood, Mr. Steffen Schoenhardt and Dr. Guanghui Ren for their support and for providing an exceptional research environment.

I would particularly like to thank Mr. Phred Petersen from School of Media and Communication for his time and for providing access to high-speed imaging hardware.

This research would not have been possible without access to state of the art equipment and facilities from Micro Nano Teaching Facility (MNTF), RMIT University and I would like to thank technical staff, Mr. Yuxun Cao and Mr. Paul Jones who work hard to keep these facilities operational.

Most importantly, I appreciate my parents, my parents in law and friends for their continuous support and encouragement. Finally, I am very thankful to my wife for her calm nature, support and encouragement, without which, I would have never reached so far.
Abstract

Multiphase microfluidic devices offer precise control of the flows in a wide range of applications such as chemical reactions, biological samples, mixing and droplet formation by applying different flow rates and channel geometries. However, in the majority of these cases, fluids used in these devices can have similar physical properties such as viscosity, density and surface tension.

Interaction of fluids with different physical properties can generate multifaceted fluid patterns in confined systems. In particular, applying fluids with a large viscosity contrast into a microfluidic system can generate quite complex instabilities arising from localised velocity and shear stress gradients. High viscosity contrast interfaces have been investigated within microfluidic flow focusing systems. However, in these works, the ratio of core/sheath flows as well as the diffusion coefficient between the core/sheath flows is such that the width of the core reduces very quickly at the entrance of the flow focusing channel. This motivated the author to change the set the operating conditions of the system to study the stability of the core flow such that the core flow can maintain its width at the entrance of the flow focusing channel.

**As the first research contribution**, the author investigated the dynamics of a highly viscous glycerol core confined between water sheath flows. Experiments indicate that the structure of core flow is determined by the ratio of core/sheath flows. At high flow rate ratios, parallel streams of core and sheath flows move alongside each other throughout the channel. At lower flow rate ratios, a tapered core flow is formed at the upstream of the channel before converting into a thin ‘viscous thread’. In this case, the stability of tapered core flow is determined by the magnitude of the sheath flow rate. At low sheath flow rates, the tapered core
remains stable. In contrast, at high sheath flow rates, ‘surface waves’ are induced at the interface of core/sheath flows. Such surface waves can buckle the core or even break it into two parts, which is referred to as ’core pinch off’ phenomenon, which has not been reported before.

**As the second research contribution**, the author characterised the dynamics of pinch off phenomenon under various combinations of core and sheath flow rates. In particular, using high speed imaging the author investigated the variations of core length and pinch off frequency. Experiments are conducted under a constant core flow rate but varying sheath flow rates as well as under varying core flow rates but a constant sheath flow rate.

**As the third research contribution**, the author explored the role of viscosity contrast between the core and sheath flows on the stability of the core. This has enabled the author to obtain the flow map of the core, representing the extent of the core not only under various combinations of core and sheath flows but also under two different viscosity contrasts.
Table of Contents

Chapter 1: Introduction

1.1 Multiphase flows

1.2 A summary of multiphase flows in microfluidics

1.3 A summary of high viscosity contrast immiscible flow instabilities

1.4 A summary of high viscosity contrast instabilities in miscible flow with
   a less viscous core

1.5 A summary of high viscosity contrast instabilities in miscible flow with
   a viscous core

1.6 Motivation of research

1.7 Layout of thesis

1.8 References

Chapter 2: Dynamics of a highly viscous core flow in a microfluidic flow focusing system

2.1 Abstract

2.2 Introduction
2.3 Materials and Method 32

2.4 Results 34

2.4.1 Low sheath flow rates: Stable core/sheath flows 34

2.4.2 Medium sheath flow rates: Onset of interfacial instabilities 37

2.4.3 High sheath flow rates: Dominance of interfacial instabilities 40

2.4.4 Characterisation of core pinch off 43

2.4.5 Mapping the core configuration under various core/sheath flow rates 50

2.5 Conclusions 53

2.6 References 54

Chapter 3: Conclusions and recommendations for future work ............57

3.1 Concluding remarks 57

3.2 Recommendations for future work 59

Chapter 4: Appendix .................................................................60

Appendix 1: Core/sheath viscosity ratio = 210:1, core flow rate = 61
1 µl/min

Appendix 2: Core/sheath viscosity ratio = 210:1, core flow rate = 62
5 µl/min

Appendix 3: Core/sheath viscosity ratio = 210:1, core flow rate = 63
2.5 µl/min

Appendix 4a: Core/sheath viscosity ratio = 210:1, core flow rate = 64
12.5 µl/min, sheath flow rates from 12.5 to 375 µl/min
Appendix 4b: Core/sheath viscosity ratio = 210:1, core flow rate = 12.5 µl/min, sheath flow rates are varying from 400 to 800 µl/min

Appendix 4c: Core/sheath viscosity ratio = 210:1, core flow rate = 12.5 µl/min, sheath flow rates are varying from 850 to 1500 µl/min

Appendix 5: Core/sheath viscosity ratio = 60:1, core flow rate = 1 µl/min

Appendix 6: Core/sheath viscosity ratio = 60:1, core flow rate = 5 µl/min

Appendix 7: Core/sheath viscosity ratio = 60:1, core flow rate = 12.5 µl/min

Appendix 8a: Core/sheath viscosity ratio = 60:1, core flow rate = 25 µl/min, sheath flow rates from 25 to 400 µl/min

Appendix 8b: Core/sheath viscosity ratio = 60:1, core flow rate = 25 µl/min, sheath flow rate from 450 to 800 µl/min

Appendix 8c: Core/sheath viscosity ratio = 60:1, core flow rate = 25 µl/min, sheath flow rates ranging from 850 to 1250 µl/min
**List of Abbreviations**

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>PDMS</td>
<td>Polydimethylsiloxane</td>
</tr>
<tr>
<td>Ca</td>
<td>Capillary number</td>
</tr>
<tr>
<td>Pe</td>
<td>Péclet number</td>
</tr>
<tr>
<td>Re</td>
<td>Reynolds number</td>
</tr>
</tbody>
</table>
List of Figures

CHAPTER 1

Figure 1.1  Folding and coiling instabilities within a diverging microfluidic channel using immiscible silicone oil/ethanol in a core/sheath arrangement

Figure 1.2  Surface bamboo waves along a vertical cylindrical channel using immiscible oil/water in a core/sheath arrangement

Figure 1.3  Pearl and mushroom instabilities along a vertical cylindrical channel using miscible water/Natrosol in a sheath/core arrangement at different Reynolds values

Figure 1.4  (a) Inertial instabilities along a flow focusing square channel using miscible silicone oil pairs in a sheath/core arrangement at various flow rates, (b) Flow map of diffusive, stable and unstable inertial regimes

Figure 1.5  Folding instabilities within a diverging microfluidic channel using amiscible PDMS oil pair at different sheath/core flow rates and viscosity contrasts

Figure 1.6  Swirling instabilities along a flow focusing square channel using a miscible PDMS oil pair created by applying mismatched sheath flows

Figure 1.7  (a) Diffusive instabilities along a flow focusing square channel using miscible silicone oil pairs in core/sheath
arrangement at various Péclet numbers, (b) Flow map of flow rate and Péclet number classifying stable, diffusive and ultra-diffusive flow regimes

**Figure 1.8** (a) Inertial instabilities along a flow focusing square channel using miscible silicone oil pairs in core/sheath arrangement, (b) Flow map of flow rate against Reynolds number

---

**CHAPTER 2**

**Figure 2.1** Schematics of the microfluidic flow focusing system. Inset shows the formation of the glycerol core flow between water sheath flows

**Figure 2.2** Effect of water sheath flows on the structure of the glycerol core flow: (a) Results are obtained by applying glycerol to core inlet at 1 µl/min while applying water to sheath inlets at varying flow rates of 1 to 120 µl/min, (b) Core swelling and formation of thin streams of sheath flow along the sidewalls, (c) Gradual tapering of the core, (d) Formation of a tapered core at the upstream of the flow focusing channel followed by formation of a ‘viscous thread’ throughout the flow focusing channel, and (e) Shortened tapered core due to increased sheath flow rate
**Figure 2.3** Effect of water sheath flows on the stability of glycerol core flow: (a) Results are obtained by applying glycerol to core inlet at 5 µl/min while applying water to sheath inlets at varying flow rates of 5 to 600 µl/min, (b) ‘Core swelling’ and formation of thin streams of sheath flows alongside the sidewalls, (c) Formation of ‘viscous waves’ at the upstream of the flow focusing channel, (d) Strong ‘viscous waves’ lead to buckling of the core, and (e) Formation of a short and highly stable core at high sheath flow rates

**Figure 2.4** Dynamics of ‘core pinch off’: (a) Results are obtained by setting flow rates of glycerol core and water sheath flows to 12.5 and 400 µl/min, respectively, (b) The core flow is disturbed by asymmetric ‘viscous waves’ induced at the interface of core/sheath flows. The inset shows the filaments of the core shedding across the crest of a ‘viscous wave’, (c) ‘Viscous waves’ can create a large trough within the core, (d) The core is broken under the influence of a large trough, and (e) The separated part of the core is squeezed before merging with the ‘viscous thread’. The shedding filaments of the core can be clearly seen in the inset
**Figure 2.5** Characterisation of glycerol core flow dynamics under varying flow rates of the sheath flows: (a) Results are obtained by applying glycerol core at 12.5 µl/min while varying the sheath flow rate from 350 to 1000 µl/min, (b-e) Glycerol core flow reaches its longest/shortest extents before/after being pinched off by water sheath flows, respectively (f) Variations of core length against sheath flow rate, and (g) Variations of core pinch off frequency against sheath flow rate. The results are obtained over an 8-second interval.

**Figure 2.6** Characterisation of glycerol core flow dynamics under varying flow rates of sheath flows: (a) Results are obtained by applying glycerol core at 12.5 µl/min while varying the sheath flow rate from 350 to 1000 µl/min, (b) Variations of core average length against sheath flow rate, (c) Variations of core pinch off frequency against sheath flow rate, and (d) Variations of core pinch off frequency against core average length.

**Figure 2.7** Characterisation of glycerol core flow dynamics under varying flow rates of the core flow: (a) Results are obtained by applying water sheath flows at 850 µl/min while varying the core flow rate from 12.5 to 75 µl/min, (b-e) Glycerol core flow reaches its longest/shortest extents before/after being pinched off by water sheath
flows, respectively (f) Variations of core length against core flow rate, and (g) Variations of core pinch off frequency against core flow rate. The results are obtained over an 8-second interval.

**Figure 2.8** Characterisation of glycerol core flow dynamics under varying flow rates of the core flow: (a) Results are obtained by applying water sheath flows at 850 µl/min while varying the core flow rate from 12.5 to 75 µl/min, (b) Variations of core average length against core flow rate, (c) Variations of core pinch off frequency against core flow rate, and (d) Variations of core pinch off frequency against core average length.

**Figure 2.9** Extent of glycerol core flow at various combinations of core and sheath flow rates and two different viscosity ratios. (a) At a core/sheath viscosity ratio of 210:1, the core flow exhibits stable regime at a core flow rate of 1 µl/min, semi-stable regime at core flow rates of 2.5 and 5 µl/min, and unstable regime at core flow rates of 12.5 and 25 µl/min. (b) At a reduced core/sheath viscosity ratio of 60:1, the core flow exhibits stable regime at a core flow rate of 1 µl/min, semi-stable regime at core flow rates of and 12.5 µl/min, and unstable regime at a core flow rate of 25 µl/min.
List of Tables

Table 1.1  Examples of multiphase flows in microfluidics

Table 1.2  Examples of high viscosity contrast immiscible flow instabilities in core/sheath arrangement

Table 1.3  Examples of high viscosity contrast miscible flow instabilities in sheath/core arrangement

Table 1.4  Examples of high viscosity contrast miscible flow instabilities in core/sheath arrangement
Appendix Figures

**Figure A1** Expanded version of Figure 2.2, demonstrating the formation of stable core/sheath flows. Results are obtained under the following conditions: (i) Core flow rate is set to 1 µl/min, and (ii) Sheath flow rates are varying from 1 to 120 µl/min.

**Figure A2** Expanded version of Figure 2.3, demonstrating the inducing of interfacial instabilities and formation of ‘viscous waves’ at the interface of core/sheath flows. Results are obtained under the following conditions: (i) Core flow rate is set to 5 µl/min, and (ii) Sheath flow rates are varying from 5 to 600 µl/min.

**Figure A3** Lowering the core flow rate to 2.5 µl/min leads to similar instability patterns presented in Figure 2.3 and Figure A2, although the instabilities are weaker. Results are obtained by varying sheath flow rates from 2.5 to 300 µl/min.

**Figure A4** Increase of core flow rate to 12.5 µl/min leads to similar instability patterns presented in Figure 2.3 and Figure A3, although the instabilities are stronger. Results are obtained by varying the sheath flow rates from 12.5 to 375 µl/min. It should be noted that increasing the sheath flow rate beyond 375 µl/min leads to pinching off the core, as comprehensively presented in Figure A5.
Figure A5  Expanded version of Figure 2.5b-e, demonstrating the core pinch off at various sheath flow rates. Results are obtained under the following conditions: (i) Core flow rate is set to 12.5 µl/min, and (ii) Sheath flow rates are varying from 400 to 800 µl/min.

Figure A6  Expanded version of Figure 2.5b-e, demonstrating the harmonic oscillations of the core under asymmetric 'viscous waves'. Results are obtained under the following conditions: (i) Core flow rate is set to 12.5 µl/min, and (ii) Sheath flow rates varied from 850 to 1500 µl/min.

Figure A7  Formation of stable core/sheath flows. Results are obtained under the following conditions: (i) Core flow rate is set to 1 µl/min, and (ii) Sheath flow rates are varying from 1 to 75 µl/min.

Figure A8  Formation of ‘viscous waves’ at the interface of core/sheath flows. Results are obtained under the following conditions: (i) Core flow rate set to 5 µl/min, and (iii) Sheath flow rates varied from 5 to 250 µl/min.

Figure A9  Increase of core flow rate to 12.5 µl/min leads to similar instability patterns presented in Figure A6, although the instabilities are stronger. Results are obtained under the following conditions: (i) Viscosity ratio of core/sheath flows is set to 60, (ii) Sheath flow rates are varying from
12.5 to 900 µl/min.

**Figure A10**  Increase of core flow rate to 25 µl/min leads to similar instability patterns presented in Figure A10, although the instabilities are stronger. Results are obtained by varying the sheath flow rates from 25 to 400 µl/min. It should be noted that increasing the sheath flow rate beyond 400 µl/min leads to pinching off the core, as presented in Figure A11.

**Figure A11**  Referring to Figure A10, increase of sheath flow rate beyond 400 µl/min leads to pinching off the core. Results are obtained by varying the sheath flow rate from 450 to 800 µl/min. The left column presents the core before pinch-off, whereas the right column presents the core after pinch-off.

**Figure A12**  Referring to Figure A11, increase of sheath flow rate beyond 800 µl/min leads to shortening of the core. The shortened core harmonically oscillates under the influence of ‘viscous waves’ but does not pinch off for the sheath flow rates ranging from 850 to 1250 µl/min, similar to conditions shown in Figure 2.6. Further increase of sheath flow rate leads to immediate tapering of the core after leaving the central channel. The shortened core becomes very stable and does not oscillate, similar to conditions shown in Figure 2.5e.
Research Contributions


CHAPTER 1

Introduction

1.1 Multiphase flows

In microfluidics, multiphase flows have been well studied experimentally and theoretically whilst offering wide range applications such as chemical reactions, biological samples, mixing and droplet/bubble formation, as it has been well characterised and summarised in review papers [1-4]. In order to create multiphase flows, two or more immiscible/miscible fluids are required to contact each other within the confines of the system. These fluids generally can have similar physical properties including density, viscosity and surface tension or vastly different physical properties.
1.2 A summary of multiphase flows in microfluidics

Due to laminar characteristics of microfluidics, diffusion based microfluidic devices have been introduced to achieve mixing by molecular diffusion, which is a great example of two phase flows. The most common diffusion based microfluidic devices are Y-shape and T-shape, in which two fluids are introduced from separate inlets to enable the control of the flow rates, independently [5-7]. Diffusion occurs by increasing contact area of fluids and decreasing diffusion length. In order to enhance mixing, hydrodynamic focusing devices can be used to decrease diffusion length by decreasing the width of the flows where flow rates are increased, therefore mixing time can be reduced [8]. A fundamental hydrodynamic focusing device includes three inlets where central (core) flow and sheath flows are introduced through middle inlet and side inlets, respectively. These devices offer precise control of the flows by applying different flow rates and channel geometries as summarised in Table 1.1, where fluids have similar physical properties such as viscosity and density.
<table>
<thead>
<tr>
<th>Application</th>
<th>Working Principle</th>
</tr>
</thead>
</table>
| Diffusion based micromixers [5-7]               | Increasing contact surface of neighbouring flows  
Decreasing diffusion length of neighbouring flows  
Flow control by tuning the flow ratio of neighbouring flows |
| Flow cytometry [9-11]                           | Focusing cells along the central flow  
Optical detection/counting/sorting of cells  
Flow control by tuning the flow ratio of central and sheath flows |
| Lateral migration of cells/micro-particles [12]  | Control of parallel streams by tuning the flow ratio of neighbouring flows  
Migration of cells/micro-particles by lateral lift force |
| Flow switch [13, 14]                            | Sample flows focused by neighbouring flows  
Sample flows switched to specific channel by tuning the flow rate of neighbouring flows |
| Optical waveguides [15]                         | Optical fiber coupled into central flow |
| Optical light source [16]                       | Tuning the optical characteristics of the system by applying fluids with desired refractive indices |
| Droplet/bubble formation [17, 18]               | Breaking the central flow into droplets/bubbles by sheath flows  
Viscous forces dominate over surface tension forces |
| Cell encapsulation [19]                         | Droplet/bubble size dependent on Capillary number |
1.3 A summary of high viscosity contrast immiscible flow instabilities

Two phase immiscible fluid lubrication has been investigated thoroughly due to the importance in crude oil processing [20], oil recovery [21], pipelining oil transport [22], and bio diesel production [23]. Water due to its abundance, modifiable nature and chemical properties is the perfect lubrication fluid. The oil density could either be higher or lower than the sheath fluid dependent on the location of the drill/processing site. Oil in these cases would vary in viscosity, density, interfacial tension and therefore the sheath fluid (water) could be dosed/tuned with chemicals accordingly to modify the viscosity contrast and reduce such instabilities along the pipelines. Utilising water to lubricate the walls of pipes leads to reduced friction losses and increased energy efficiency along large stretches of piping, which not only reduces pumping load but also reduces fouling and pipe blockage [22].

As the viscosity decreases, fluid tends to arrange itself in the high region of shear, oil and water concurrent flows include a sheath made of water which lubricates the highly viscous core flow of oil from touching the channel walls. Table 1.2 shows high viscosity contrast immiscible flows within a fluid system, various flow instabilities are present. According to Table 1.2, the most common instabilities are dripping, jetting, folding, coiling and surface waves which are named by their shape.

Core break-up is associated with droplet formation where the fluid thread is ‘broken up’ by an interfacial tension instability that overcomes either inertial or viscosity forces resulting in separated discrete packets of fluid. Dependent on the flow rates and fluid properties, various regimes and hence droplet sizes are
created, jetting creates smaller droplets further down the channel with the utilisation of a thinner core thread. Folding and coiling regimes are created by a diverging channel where the core/sheath fluid velocity is quickly reduced due to the larger cross sectional area, as further discussed in Figure 1.1. Surface wave instabilities are created along the core sheath interface and are driven by the viscosity contrast and flow rate ratio between both fluids as further discussed in Figure 1.2.

The most common core fluids used include oil, glycerol, silicone oil and polydimethylsiloxane (PDMS). PDMS oil and glycerol are of great importance as their viscosity is easily tunable. Common lower viscosity sheath fluids include water, ethanol and isopropanol alcohol.

The sheath fluid is generally the lower viscosity fluid that entrains the high viscous core fluid. Common channel geometries utilised are square and rectangular cross sections with flow focusing and diverging channels and cylindrical channels. Interfacial tension between the core and sheath fluids range from 1.5 to 27 mN/m. Viscosity of core flows range from $1.93 \times 10^{-1}$ to 4.87 Pa.s whilst sheath viscosity ranges from $1.00 \times 10^{-3}$ to $4.59 \times 10^{-3}$ Pa.s.
<table>
<thead>
<tr>
<th>Instability</th>
<th>Geometry</th>
<th>Core viscosity (Pa.s)</th>
<th>Sheath viscosity (Pa.s)</th>
<th>Fluids (core/sheath)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Core break-up (dripping, jetting)</td>
<td>Flow focusing square channel</td>
<td>1.21</td>
<td>4.59×10^{-3}</td>
<td>Glycerol / PDMS oil</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>[24]</td>
</tr>
<tr>
<td>Core folding</td>
<td>Diverging rectangular channel</td>
<td>1.93×10^{-1} to 4.87</td>
<td>2.24×10^{-3}</td>
<td>PDMS oil / Isopropanol</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>[25]</td>
</tr>
<tr>
<td>Core folding, coiling</td>
<td>Diverging rectangular channel</td>
<td>1.94×10^{-1} to 4.87</td>
<td>1.14×10^{-3}</td>
<td>PDMS oil / Ethanol</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>[26]</td>
</tr>
<tr>
<td>Surface wave (saw tooth)</td>
<td>Cylindrical channel</td>
<td>1.15</td>
<td>1.00×10^{-3}</td>
<td>Oil / Water</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>[22]</td>
</tr>
<tr>
<td>Surface wave (bamboo, corkscrew)</td>
<td>Vertical cylindrical channel</td>
<td>6.01×10^{-1} to 1.33</td>
<td>1.00×10^{-3}</td>
<td>Oil / Water</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>[27-30]</td>
</tr>
</tbody>
</table>
Folding and coiling flow regimes using immiscible fluids (silicone oil core and ethanol for sheath) are shown in Figure 1.1. A large viscosity contrast of 4344 is used, with the flow rates of the core and sheath flows set to 1 and 600 µl/min, respectively. The folding pattern is induced by the diverging channel such that the velocity reduces by a factor of 6. The core flow then morphs from a two dimensional folding pattern to a coil like pattern with three dimensional helical threads as the core fluid tries to rapidly reduce its interfacial area through the central channel. Interestingly the thread diameter is nearly unchanged through the whole process [26].

![Figure 1.1: Folding and coiling instabilities within a diverging microfluidic channel using immiscible silicone oil/ethanol in a core/sheath arrangement [26].](image)
Bamboo surface waves are shown in Figure 1.2, by utilising an up flow capillary instability through a vertical cylindrical channel of diameter 10 mm. Shear stabilisation along the immiscible interface of an oil core entrained by a water sheath, with the viscosity of the core and sheath flows set to $6.01 \times 10^{-1}$ and $1.00 \times 10^{-3}$ Pa.s, respectively [30]. Wavy flow with sharp crests at the tip of each wave is called bamboo waves, these waves are stretched due to buoyancy and lubrication forces (Figure 1.2a). By increasing the core flow and keeping the sheath water flow rate constant, the core is thickened and the average length between waves decrease (Figure 1.2b).

![Figure 1.2](image)

**Figure 1.2:** Surface bamboo waves along a vertical cylindrical channel using immiscible oil/water in a core/sheath arrangement [30]: (a) slow core flow rate, versus (b) fast core flow rate.
1.4 A summary of high viscosity contrast instabilities in miscible flow with a less viscous core

In the case where large viscosity contrasts are used, fluid flows tend to be laminar with little mixing capability and rely on the diffusion characteristics of the paired miscible fluids. Miscible fluid instabilities at the flow boundaries show promise to better understand the mixing capabilities within a flow system.

**Table 1.3** shows high viscosity contrast miscible flows with a fluid arrangement in which core fluid is less viscous than the surrounding sheath fluid. Low viscosity core miscible fluids include water and PDMS oil while high viscosity fluids include Natrosol and PDMS oil. Due to the miscible nature of the study within flow systems, liquid fluid pairs such as PDMS oil / PDMS oil are possible. The fluid arrangement choice is critical for the instability pattern desired, hence **Table 1.3** shows less viscous core miscible flows. Instabilities present from miscible fluids differ from their immiscible counterparts due to the lack of any influential interfacial tension, therefore a stratified interface and diffusion mixing capability can be created. Core fluid viscosities range from \(4.90 \times 10^{-4}\) to \(2.69 \times 10^{-3}\) Pa\(\cdot\)s, whilst high viscosity sheath fluids range from \(1.00 \times 10^{-2}\) to \(4.87\) Pa\(\cdot\)s, as shown in **Table 1.3**.

Surface wave instabilities present along the two fluids interface include pearl, mushroom, roll-up, pinching and corkscrew. These instabilities are named by their respective authors and the name chosen identifies the shape of the wave. In this case, the two stratified interfaces are seen as two separate interfaces and they do not converge. Utilising square section, flow focusing and cylindrical channel geometries, these instabilities have been experimentally and numerically identified.
Table 1.3 Examples of high viscosity contrast miscible flow instabilities in sheath/core arrangement

<table>
<thead>
<tr>
<th>Instability (geometry)</th>
<th>Core viscosity (Pa.s)</th>
<th>Sheath viscosity (Pa.s)</th>
<th>Fluids (core/sheath)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Surface wave (pearl, mushroom) Cylindrical channel</td>
<td>1.00×10^{-3}</td>
<td>2.50×10^{-2}</td>
<td>Water / Natrosol mixtures [31, 32]</td>
</tr>
<tr>
<td>Surface wave (roll-up, convective mixing) Square channel</td>
<td>1.00×10^{-3}</td>
<td>1.00×10^{-2} to 4.00×10^{-2}</td>
<td>Artificial fluids for numerical analysis [33]</td>
</tr>
<tr>
<td>Surface wave (pinching, corkscrew) Flow focusing square channel</td>
<td>4.90×10^{-4} to 2.69×10^{-3}</td>
<td>4.80×10^{-2} to 4.87</td>
<td>PDMS oil / PDMS oil [34]</td>
</tr>
</tbody>
</table>
Pearl and mushroom patterns are shown in **Figure 1.3**. A vertical cylindrical tube of 20 mm diameter is used. A miscible central core of water with the viscosity of 1.00×10⁻³ Pa.s flows inside a Natrosol sheath with the viscosity 2.50×10⁻² Pa.s. The sheath/core flow ratio is set to 4. Instabilities are induced due the lower viscosity fluids tendency to arrange itself in high shear stress locations while the high viscosity sheath flow tends to move toward low shear stress locations. Under these conditions, increasing the core and sheath flow rate leads to progression from a pearl like pattern to a mushroom like surface wave pattern. The mushroom pattern also shows viscous thread formation along crest of the wave [31].

**Figure 1.3**: Pearl and mushroom instabilities along a vertical cylindrical channel using miscible water/Natrosol in a sheath/core arrangement at different Reynolds values [31]: (a) Re=5, (b) Re=9, (c) Re=12, and (d) Re=18.
Inertial destabilisation of high viscous contrast miscible fluids is shown in Figure 1.4. A miscible central core of silicone oil of viscosity $4.90 \times 10^{-4}$ Pa.s is surrounded by a silicone oil sheath fluid of $1.94 \times 10^{-1}$ Pa.s. Diffusion coefficient between the two fluids is $7.7 \times 10^{-10}$ m$^2$/s. A microfluidic chip of silicon and glass with 250 µm square section is used. Core/sheath flow rates are set to (i) 1/1 µl/min, (ii) 50/20 µl/min, (iii) 200/100 µl/min and (iv) 400/100 µl/min. As the flow rate is increased, the flow regime varies from diffusive to inertial instabilities with waves forming along the two fluid interfaces. (iii, iv) shows the core flow increase with a fixed sheath flow, in which the position of the wave disturbance moves closer to the focusing inlet (Figure 1.4a). A flow map comparing core flow rate against sheath flow rate classifies diffusive, stable and unstable inertial regimes (Figure 1.4b) [34].
Figure 1.4: (a) Inertial instabilities along a flow focusing square channel using miscible silicone oil pairs in a sheath/core arrangement at various flow rates, (b) Flow map of diffusive, stable and unstable inertial regimes [34].
1.5 A summary of high viscosity contrast instabilities in miscible flow with a viscous core

Currently there is limited experimental research into flow instabilities with miscible fluid flows in microfluidics. Table 1.4 summarises instabilities created within micro environments. Fluid viscosities for the core fluid range from $4.59 \times 10^{-3}$ to 9.74 Pa.s whilst sheath viscosities range from $4.90 \times 10^{-4}$ to $9.60 \times 10^{-2}$ Pa.s, instabilities include folding, coiling, swirls, diffusive and inertial. Like their immiscible counterparts folding and coiling instabilities are created with either diverging or constricting geometric channel arrangements. Swirl instabilities have been shown as a unique instability within miscible fluid contact and can be created if the core fluid thread is off centre within the channel. This can be implemented if a two-step flow focusing device is used or if the incoming sheath flow rates are mismatched. Diffusive instabilities have been shown to be dependent on Péclet number, flow rate ratio and viscosity contrast, whilst inertial instabilities are heavily dependent on the flow rate ratio and high sheath flow rate.
Table 1.4 Examples of high viscosity contrast miscible flow instabilities in core/sheath arrangement

<table>
<thead>
<tr>
<th>Instability</th>
<th>Geometry</th>
<th>Core viscosity (Pa.s)</th>
<th>Sheath viscosity (Pa.s)</th>
<th>Fluids (core/sheath)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Core folding</td>
<td>Diverging rectangular channel</td>
<td>4.86×10^{-1} or 9.74</td>
<td>4.59×10^{-3} or 1.90×10^{-2} or 9.60×10^{-2}</td>
<td>PDMS oil / PDMS oil [26]</td>
</tr>
<tr>
<td>Core folding</td>
<td>Diverging rectangular channel</td>
<td>1.00×10^{-1} to 5.00×10^{-1}</td>
<td>5.00×10^{-4} to 6.00×10^{-3}</td>
<td>PDMS oil / PDMS oil [35]</td>
</tr>
<tr>
<td>Core folding, coiling</td>
<td>Converging / diverging rectangular channel</td>
<td>1.00×10^{-1}</td>
<td>8.2×10^{-4}</td>
<td>PDMS oil / PDMS oil [36]</td>
</tr>
<tr>
<td>Core swirling</td>
<td>Two-step square flow focusing channel</td>
<td>5.00×10^{-1}</td>
<td>6.00×10^{-3}</td>
<td>PDMS oil / PDMS oil [37, 38]</td>
</tr>
<tr>
<td>Core diffusive/Inertial disturbance</td>
<td>Flow focusing square channel</td>
<td>4.59×10^{-3} to 4.87</td>
<td>4.9×10^{-4} to 4.59×10^{-3}</td>
<td>Silicone oil / Silicone oil [39]</td>
</tr>
</tbody>
</table>
Folding flow regimes using miscible fluids (silicone oil pair) is shown in Figure 1.5. The incoming channel shown on the left is 250 µm × 250 µm, the folding pattern is induced by the diverging channel such that the velocity reduces by a factor of 6. Amplitude variation of viscous folding is controlled by the sheath/core ratio. As the viscous thread is swept through the channel by the sheath flow the lower viscosity contrast of 52 tends to ‘reflow’ within its neighbouring fold (Figure 1.5a) whilst as the viscosity contrast is increased, higher sheath flow ratios are needed to entrain and maintain the viscous core width. The higher viscosity contrast of 512 leads to more rigid and solid like behaviour (Figure 1.5b) [26].

Figure 1.5: Folding instabilities within a diverging microfluidic channel using a miscible PDMS oil pair at different sheath/core flow rates employing viscosity contrasts of (a) 52, and (b) 512 [26].
Viscous swirl instability is shown in Figure 1.6. A single off center miscible thread is created by mismatching the two incoming sheath fluid channel flow rates. A microfluidic flow focusing device of 100 µm square section is utilised. The viscous core flow rate is 1 µl/min, whilst the two mismatched sheath flows are 2 and 10 µl/min, accordingly. The viscosity contrast of 83 is obtained by setting the core viscosity to 5.00×10⁻¹ Pa.s and the sheath viscosity to 6.00×10⁻³ Pa.s (Figure 1.6a). The thread instability is caused by the viscous torque induced due to difference between drag forces on each side of the thread. The thread gain angular momentum and continue to rotate following they break up into discrete viscous swirls (Figure 1.6b) [38].

**Figure 1.6**: (a) Schematic representation of the square microchannel used for studying swirling instabilities using a miscible PDMS oil pair created by applying mismatched sheath flows, (b) Top view of the channel depicting the viscous swirls [38].
Diffusive flow regimes shown in Figure 1.7 are obtained within a flow focusing square channel. Strongly diffusive and high viscosity contrast fluid pairs have been used by setting the values of mass diffusivity and viscosity contrast to $5.5 \times 10^{-10} \text{ m}^2/\text{s}$ and 982 for cases (i) and (ii) while $1.2 \times 10^{-9} \text{ m}^2/\text{s}$ and 97 for case (iii). Diffusive instabilities are less sensitive to viscosity contrast and high flow rates. The viscous core instability is due to highly complex viscous stratifications downstream of the fluid junction. The diffusive mixing of neighbouring fluids can be seen by the shade gradient of the greyscale images and are arranged by Péclet number in which (i) high Péclet number shows thin stable thread with very little diffusion (ii) moderate Péclet number shows thread formation and oscillation downstream, increasing mixing capability (iii) low Péclet number shows large diffusion gradient increasing downstream of the channel (Figure 1.7a). These diffusive regimes depend on the sheath flow rate and Péclet number, and can be categorised into stable thread, diffusive instability and ultra-diffusive regimes. (Figure 1.7b) [39].
Figure 1.7: (a) Diffusive instabilities along a flow focusing square channel using miscible silicone oil pairs in core/sheath arrangement at various Péclet numbers, (b) Flow map of flow rate and Péclet number classifying stable, diffusive and ultra-diffusive flow regimes [39].
Inertial instability can significantly disturb the central core thread flow, as shown in Figure 1.8. A microfluidic chip of silicon and glass with 250 µm square section is used. By utilising viscosity contrasts ranging from 10 to 1000 whilst Reynolds number values kept at roughly 103, the slender viscous core flow is influenced by inertial flow fields along the core/sheath interface. The structural integrity of the core is analysed at moderate Reynolds values. Fixed flow rates for core and sheath are 1 and 1000 µl/min, respectively. As the viscosity contrast increases, the width of core thread increases and sheath flow deformations reduce. Interestingly at low viscosity contrasts the centralised core thread ceases to exist. (Figure 1.8a). Using a fixed viscosity contrast of $10^3$ and a low viscosity sheath entrainment fluid viscosity of $4.95 \times 10^{-4}$ Pa.s, stable and inertial regime flow map is shown by core/sheath flow rate ratio against Reynolds number. The sheath velocity profile plays an important role destabilising the core flow. The more viscous the core the more resilient the core flow is to inertial deformation due to its width along the channel, interesting to note the higher the sheath flow rate the closer the instability moves towards the flow focusing junction. Interestingly, inertial instabilities are dependent on the flow rate ratio and Reynolds numbers. Thicker threads are more resilient to similar Reynolds numbers, and as velocity increases the thread destabilisation moves toward the fluid junction. These inertial regimes are categorised based on the core/sheath flow rate ratio and Reynolds number. There are two main regimes; stable thread and inertial (Figure 1.8b) [39].
Figure 1.8: (a) Inertial instabilities along a flow focusing square channel using miscible silicone oil pairs in core/sheath arrangement, (b) Flow map of flow rate against Reynolds number [39].
1.6 Motivation of research

This thesis investigates the dynamics of highly viscous core flow confined by low viscous sheath flows within a microfluidic flow focusing channel. High viscosity contrast core/sheath flows have been investigated before [26, 39]. However, in such works, the ratio of core/sheath flows as well as the diffusion coefficient between the core/sheath flows is such that the core becomes thin very quickly at the entrance of the flow focusing channel. This motivated the author to change the set of operating conditions of the system to investigate the structure and stability of the highly viscous core flow under the conditions that the core flow can maintain its width at the entrance of the flow focusing channel.

This is quite important, as previous studies by the same research group towards hydrodynamic migration of liquid metal droplets along flow focusing channel (in which the author of this thesis has been involved) has shown massive flow instabilities at the interface of core sheath flows [40]. However, the above mentioned study has been conducted in the presence of liquid metal droplets, which means that such flow instabilities could have been induced by migration of droplets. The existence of a long highly viscous core flow can lead to new types of flow instabilities, which are investigated in this thesis in a systematic manner, which can be summarised in the form of the following three research questions:

Research Question 1: What types of flow instabilities are induced along the highly viscous core flow, if the core flow can maintain its width along the entrance of the flow focusing channel?

Research Question 2: How does the sheath flow rate influence the flow instabilities observed in Research Question 1?
Research Question 3: How does the viscosity contrast between the core/sheath flows impact the flow instabilities observed in Research Question 1?

1.7 Layout of thesis

This research aims to experimentally explore the effect of sheath flow rate on a highly miscible core centralised core within a microfluidic system. The outcomes of this work are presented in the following chapters, as summarised below:

Chapter 2: Presents the dynamics of a glycerol core flow confined by water sheath flows within a microfluidic flow focusing system by utilising high speed imaging. A comprehensive set of experiments are conducted and the influence of a high flow rate sheath flow are investigated.

Chapter 3: Presents conclusion, concluding remarks and recommendations for future work
1.8 References


CHAPTER 2

Dynamics of a highly viscous core flow in a microfluidic flow focusing system

2.1 Abstract

This chapter investigates the dynamics of a glycerol core flow confined by water sheath flows within a microfluidic flow focusing system. The configuration of the core flow depends on the ratio of core/sheath flows. At low core/sheath flow ratios, the core flow becomes tapered and eventually narrows down to form a ‘viscous thread’ along the middle of the channel. The large viscosity contrast between the core and sheath liquids induces local disturbances, which propagate in the form of asymmetric ‘viscous waves’ at the interface of the two flows. Such ‘viscous waves’ can significantly impact the stability of the tapered core flow, and depending on the magnitude of the sheath flow rate can buckle or even break the core flow into two parts. Using high speed imaging, the dynamic characteristics of highly viscous core flow under various combinations of core/sheath flows have been studied.
2.2 Introduction

Microfluidic flow focusing systems enable multiple fluids to be introduced into the device. Applying a pair of immiscible fluids (e.g. water/oil or air/water) into a flow focusing system enables the continuous generation of microscale droplets or bubbles of the low viscosity fluid encapsulated within the high viscosity sheath flow [1-3].

Alternatively, utilising a pair of miscible fluids (e.g. two water based solutions) into a flow focusing system enables a core flow surrounded by sheath flows. The width of core and sheath flows can be precisely modulated according to the ratio of the core and sheath flows. Owing to the laminar characteristics of flow, the mixing between the neighbouring flows is governed by diffusion, which is a slow process, since it is limited to the interface between the fluids. Therefore, the width of the neighbouring flows remains almost constant along the focusing channel. Microfluidic flow focusing systems have been widely used for flow cytometry [4, 5], and lateral migration of suspended particles (e.g. colloidal particles and DNA molecules) [6].

However, in the majority of the above cases, the core and sheath flows have similar viscosities. Some unique instability patterns can be observed when there is a large viscosity contrast between the neighbouring flows [7-10], as comprehensively reviewed by Sahu et al. [11]. For example, both experimental [12, 13] and numerical [14] analyses have shown the existence of pearl and mushroom instability patterns when applying two miscible liquids into a circular pipe with the viscous liquid acting as the sheath (annular) flow. Likewise, it has been shown both experimentally [15] and numerically [16] that applying two immiscible liquids into a circular pipe with the viscous liquid acting as the core
flow is associated with bamboo instability patterns at the interface of the liquids. In particular, the stability of core-annular flows is important for transporting heavy and extra-heavy crude oil within a sheath of lubricating water [17], which is used as an effective means for reducing friction losses through long pipelines [18-20].

Over the last decade, Cubaud et al. [21, 22] have comprehensively studied the fascinating flow dynamics of highly viscous core flow confined between low viscosity sheath flows in microchannels in a wide range of core/sheath flow ratios, viscosity ratios and diffusion coefficient ratios. These studies have demonstrated the formation of a highly stable ‘viscous thread’ at high Péclet numbers with a smooth interface formed between the core/sheath flows [23]. In contrast, applying moderate Péclet numbers is associated with some periodical oscillations at specific distances from the entrance of the core flow channel, where thread experiences ‘diffusive instabilities’ in the form of diffusive coiling. Applying low Péclet numbers leads to ‘ultra-diffusive instabilities’ where the core and sheath flows are mixed very quickly [23]. Also, it is shown that the combination of high sheath flow rates and low core/sheath viscosity ratios is associated with rolling up thin filaments of ‘viscous thread’, leading to ‘inertial instabilities’ [23].

In addition, deceleration of ‘viscous threads’ by means of diverging of the focusing channel is shown to induce ‘viscous buckling instabilities’, leading to formation of periodic folding threads [21, 24, 25], similar to the case of a viscous liquid falling on a solid surface [26]. Furthermore, offsetting the viscous core flow from the middle of the focusing channel by means of applying asymmetric sheath flow rates is shown to induce sinuous perturbations in the direction transverse to
the flow, which eventually leads to disintegration of the ‘viscous thread’ into isolated ‘viscous swirls’ [22].

In this chapter, the exciting dynamics of a highly viscous core flow made of glycerol confined by sheath flows made of water within a microfluidic flow focusing system is studied. Experiments indicate that the structure of core flow is determined by the ratio of core/sheath flows. At high flow ratios, parallel streams of core/sheath flows move alongside each other throughout the channel. At lower flow ratio, a tapered core flow is formed at the upstream of the channel before narrowing down to a thin ‘viscous thread’. Studies using high speed imaging indicate that the stability of tapered core flow is determined by the magnitude of the sheath flow rate. At low sheath flow rates, the tapered core remains very stable. Increasing the sheath flow rate is associated with inducing local disturbances at the interface that propagate in the form of ‘viscous waves’. Such disturbances intensify by increasing the sheath flow rate, which can severely deform the core or even break it into two parts. The dynamic characteristics of ‘core pinch off’ are studied under various combinations of core/sheath flows.
2.3 Material and Methods

The experimental setup consists of a microfluidic flow focusing device for producing core/sheath flows of glycerol/water, two syringe pumps (Harvard 2000, Harvard PicoPlus) for introducing glycerol and water into the device, and a high-speed camera (Phantom Miro 310, 1000 fps) attached to an inverted microscope (Nikon Eclipse Ti) for visualisation of the core/sheath flow dynamics in real-time.

Figure 2.1 presents the schematic of the microfluidic flow focusing device. The device has three inlet channels with inlet-1 to infuse glycerol, and inlets-2 and -3 to infuse water ($W_1 = W_2 = W_3 = 300 \mu m$). Channel inlets-2 and -3 are connected to channel inlet-1 at an angle of 30°. All three inlet channels converge into a flow focusing channel ($W_4 = 700 \mu m$ and $L = 30 mm$). The height of the microfluidic device is 100 µm.

The microfluidic device is fabricated from polydimethylsiloxane (PDMS) using soft lithography techniques. PDMS moulded channel is sealed onto a 1 mm thick glass slide and plasma treated in a Harrick plasma cleaner for 5 minutes. Microfluidic device is then post baked at a 75°C oven for 2 hours to permanently bond the glass slide to the PDMS moulded channel.

Glycerol is diluted with water at a volume ratio of 9:1 to produce a viscosity ratio of 210:1 ($\mu_{\text{glycerol}} = 0.210 \text{ Pa.s}, \mu_{\text{water}} = 0.001 \text{ Pa.s}$). The fluidic input includes three plastic syringes, consisting of one 5 ml filled with diluted glycerol and two 20 ml syringes filled with water. The 5 ml syringe is inserted onto the Harvard PicoPlus syringe pump whilst the two 20 ml syringes are coupled together onto the Harvard 2000 syringe pump. The outlet of the microfluidic device is coupled to a collection beaker via a 0.06 inch Tygon® tube. Figure 2.1 inset shows the
formation of a single central glycerol core flow focused between water sheath flows entering from both sides.

**Figure 2.1:** Schematics of the microfluidic flow focusing system. Inset shows the formation of the glycerol core flow between water sheath flows.
2.4 RESULTS

2.4.1. Low sheath flow rates: Stable core/sheath flows

To explore the dynamics of the highly viscous core flow through the flow focusing channel, experiments are started by applying glycerol at a flow rate of 1 µl/min while applying water at flow rates varying from 1 to 120 µl/min (Figure 2.2a).

The variations of core structure at different sheath flow rates are presented in Appendix Figure A1, with highlights discussed here. By setting the sheath flow rate to 1 µl/min \((Q_{core}/Q_{sheath}=1:1)\), the highly viscous core occupies most of the flow focusing channel (Figure 2.2b), which is referred to as ‘tubing regime’ in the literature [23]. The core flow advances toward the two sheath channels, which is coined as ‘core swelling’ here. Passing the swelled region of the core, the sheath flows form thin streams against the sidewalls where they can flow alongside the core. The two sheath flows create interfaces on each side of the core, which remain stable through the flow focusing channel. A dark line can be seen between the core and sheath flows in Figure 2.2b. As the two fluids are both transparent, this line should be produced by reflection/refraction of the incident illumination from the refractive index change between the two fluids \((n_{glycerol} = 1.4716, \ n_{water} = 1.330)\). As the two fluids are miscible, the exact location of the interface cannot be identified. However, to simplify descriptions, this dark line is referred to as the core/sheath flow interface.

By increasing the sheath flow rate to 30 µl/min \((Q_{core}/Q_{sheath}=1:30)\) the width of the core flow and consequently the ‘core swelling’ reduce. Interestingly, the continuous mixing at the core/sheath flow interfaces leads to tapering of the core flow at an angle of 1.7° toward the outlet of the flow focusing channel (Figure...
As the sheath flow rate is increased to 60 µl/min ($Q_{core}/Q_{sheath} = 1:60$) the tapering angle increases to 4.4°. Under this condition, the length of the core reduces to ~2.3 mm, after which the core flow transforms into a thin stream along the middle of the flow focusing channel (Figure 2.2d). This thin stream of highly viscous core liquid is referred to as ‘viscous thread’ in the literature [21]. Likewise, by increasing the sheath flow rate to 120 µl/min ($Q_{core}/Q_{sheath} = 1:120$), the tapering angle increases to 27.5°, and the length of the core flow reduces to ~0.3 mm (Figure 2.2e).
Figure 2.2: Effect of water sheath flows on the structure of the glycerol core flow. Results are obtained by applying glycerol to core inlet at 1 µl/min while applying water to sheath inlets at varying flow rates of 1 to 120 µl/min: (a) Schematic representation, (b) Core swelling and formation of thin streams of sheath flow along the sidewalls, (c) Gradual tapering of the core, (d) Formation of a tapered core at the upstream of the flow focusing channel followed by formation of a ‘viscous thread’ throughout the flow focusing channel, and (e) Shortened tapered core due to increased sheath flow rate.
2.4.2 Medium sheath flow rates: Onset of interfacial instabilities

It was hypothesised that increasing the sheath flow rate can cause instabilities at the interface of the core/sheath flow. To prove this hypothesis, the flow rate of glycerol is set to 5 µl/min and accordingly the flow rate of the water is varied from 5 to 600 µl/min to produce the flow ratios presented in the previous example (Figure 2.3a).

The variations of core structure at different sheath flow rates are depicted in Appendix Figure A2, with highlights discussed here. Setting the sheath flow rate to 5 µl/min \( \left( \frac{Q_{\text{core}}}{Q_{\text{sheath}}} = 1:1 \right) \) leads to ‘core swelling’ at the upstream of the flow focusing channel, and formation of thin streams of sheath flow alongside the sidewalls (Figure 2.3b). The core/sheath interface remains stable throughout the flow focusing channel, very similar to the conditions observed in Figure 2.2b.

As the sheath flow is increased to 150 µl/min \( \left( \frac{Q_{\text{core}}}{Q_{\text{sheath}}} = 1:30 \right) \), the width of the core flow is reduced, as expected. However, unexpectedly, despite the formation of a stable interface at the upstream of the flow focusing channel, minor instabilities are induced at the interface of the core/sheath flows (Figure 2.3c). At close inspection, these instabilities induce ‘viscous waves’ along the interface with thin filaments of viscous liquid shedding across the crest of the ‘viscous waves’ (Figure 2.3c’), similar to what has been numerically predicted in ref [27]. These shedding filaments move alongside the core flow and eventually merge with the ‘viscous thread’ formed at the tip of the tapered core flow.

By increasing the sheath flow rate to 300 µl/min \( \left( \frac{Q_{\text{core}}}{Q_{\text{sheath}}} = 1:60 \right) \), the core flow is tapered and eventually transforms into a ‘viscous thread’ at ~ 4.2 mm downstream of the flow focusing channel (Figure 2.3d). This is similar to the
conditions when the core flow rate is set to 1 µl/min (**Figure 2.2d**). However, the presence of strong but asymmetric ‘viscous waves’ lead to the local buckling of the core (**Figure 2.3d**). Interestingly, the base and the tip of the tapered core are mechanically robust and do not deform. As a result, the core behaves as a ‘simply supported beam’ and experiences severe deformations along its middle regions.

Increasing the sheath flow rate to 550 µl/min shortens the core at the same time intensifies the ‘viscous waves’ induced at the interface of the core/sheath flows. However, by increasing the sheath flow rate to 600 µl/min ($Q_{\text{core}}/Q_{\text{sheath}} = 1:120$), the core becomes tapered just after leaving the core channel (**Figure 2.3e**). In this condition, the core is so short that the ‘viscous waves’ do not develop at its interface. In the absence of ‘viscous waves’ the interface of the core flow is very smooth and the core maintains its stability, very similar to the conditions observed in **Figure 2.2e**.

Extended experiments at the core flow rate of 2.5 µl/min indicate similar instability patterns, as presented in **Appendix Figure A3**. The ‘viscous waves’ start to emerge at the sheath flow rate of 120 µl/min. This is very close to the sheath flow rate of 150 µl/min, beyond which ‘viscous waves’ were observed at a core flow rate of 5 µl/min, as presented in **Figure 2.3c**.
Figure 2.3: Effect of water sheath flows on the stability of glycerol core flow. Results are obtained by applying glycerol to core inlet at 5 µl/min while applying water to sheath inlets at varying flow rates of 5 to 600 µl/min: (a) Schematic representation, (b) ‘Core swelling’ and formation of thin streams of sheath flows alongside the sidewalls, (c) Formation of ‘viscous waves’ at the upstream of the flow focusing channel, (d) Strong ‘viscous waves’ lead to buckling of the core, and (e) Formation of a short and highly stable core at high sheath flow rates.
2.4.3 High sheath flow rates: Dominance of interfacial instabilities

To further investigate the stability of the core flow at higher sheath flow rates, the flow rate of glycerol is set to 12.5 µl/min and accordingly the flow rate of water is varied from 12.5 to 750 µl/min. Experiments indicate the formation of ‘viscous waves’ when applying sheath flow rates ranging from 125 to 375 µl/min (Appendix Figure A4), which cause the buckling of the core, similar to the conditions shown in Figure 2.3d.

Surprisingly, by increasing the sheath flow rate to 400 µl/min, a completely new phenomenon is observed, in which the core cannot maintain its continuity and is broken into two parts. This phenomenon, which is referred to as ‘core pinch off’, which is discussed in more detail here.

Figure 2.4a shows the flow conditions with the core flow rate set to 12.5 µl/min and the sheath flow rate set to 400 µl/min, corresponding to a core/sheath flow ratio of 1:32. The core is constantly disturbed due to formation of ‘viscous waves’, which lead to formation of ‘troughs’ and ‘crests’ at the interface of the core/sheath flows (Figure 2.4b-b’). These ‘troughs’ expand very quickly and depending on their size can severely reduce the local width of the core flow. However, at the base the core, the core width is determined by the width of the core channel, whereas at the tip of the core, the core width is reduced to almost zero due to formation of ‘viscous thread’. Therefore, the middle regions of the core experience the highest amount of localised width reduction under the influence of ‘troughs’ (Figure 2.4c).

The ‘troughs’ are formed along the two sides of the core in an asymmetric manner, which can cause severe buckling along the middle regions of the core.
Once these localised deformations have sufficiently narrowed the core, an upcoming ‘trough’ formed at the opposite side of the core is able to break the core into two parts (Figure 2.4d-d’).

Once the core tip is separated from the core, it is squeezed and merges with the previously formed ‘viscous thread’ along the middle of the flow focusing channel. At the same time, a new tip is formed at the front of the broken core. The core then advances through the flow focusing channel before the ‘viscous waves’ can break it apart again (Figure 2.4e). Depending on the magnitude of the disturbance caused by the expansion of ‘troughs’, the core can be pinched off even before reaching its longest possible length. The dynamic characteristics of ‘core pinch off’ are further explored in Figure 2.5.
**Figure 2.4:** Dynamics of ‘core pinch off’. Results are obtained by setting flow rates of glycerol core and water sheath flows to 12.5 and 400 µl/min, respectively:  
(a) Schematic representation, (b) The core flow is disturbed by asymmetric ‘viscous waves’ induced at the interface of core/sheath flows. The inset shows the filaments of the core shedding across the crest of a ‘viscous wave’, (c) ‘Viscous waves’ can create a large trough within the core, (d) The core is broken under the influence of a large trough, and (e) The separated part of the core is squeezed before merging with the ‘viscous thread’. The shedding filaments of the core can be clearly seen in the inset.
2.4.4 Characterisation of core pinch off

Further experiments are conducted to characterise the dynamics of glycerol core flow under varying flow rates of the core and sheath flows. Firstly, the flow rate of the glycerol is set to 12.5 µl/min while the flow rate of the water is varied from 400 to 1500 µl/min (Figure 2.5a). It should be noted that our observations are limited to the first 5 mm of the flow focusing channel that can be monitored in one field of view using a 4× objective.

Applying sheath flows within the range of 400 to 800 µl/min is associated with the ‘core pinch off’ phenomenon, in which the core length varies between maximum/minimum limits corresponding to pre/post pinch off conditions (Appendix Figure A5). Figures 2.5b-d show the core structure while at its minimum length at four representative sheath flow rates falling within the range of 400 to 800 µl/min. Increasing the sheath flow rate to 850 µl/min is associated with the formation of a short core, which oscillates almost harmonically under the influence of ‘viscous waves’ but does not pinch off (Figure 2.5e). Increasing the sheath flow rate to 1500 µl/min leads to significant reduction of the core length, which is immediately tapered after leaving the core channel. Such a short core is very stable and does not oscillate. The dynamics of core structure at various sheath flow rates falling within the range of 850 to 1500 µl/min is presented in Appendix Figure A6.

Figure 2.5f shows the extent of the core under various sheath flow rates. It reveals that the average length of the tapered core reduces almost exponentially with respect to sheath flow rate. For the pinched off core, the average length of the core is defined as the average value of minimum and maximum core lengths. Furthermore, results indicate that the pinch off frequency increases linearly.
against the sheath flow rate (Figure 2.5g), while decreases exponentially against the average core length (Figure 2.5g-inset). The details of calculations are presented in Figure 2.6.
Figure 2.5: Characterisation of glycerol core flow dynamics under varying flow rates of the sheath flows. Results are obtained by applying glycerol core at 12.5 μl/min while varying the sheath flow rate from 350 to 1500 μl/min: (a) Schematic representation, (b-e) Glycerol core flow reaches its longest/shortest extents before/after being pinched off by water sheath flows, respectively (f) Variations of core length against sheath flow rate, and (g) Variations of core pinch off frequency against sheath flow rate. The results are obtained over an 8-second interval.
Figure 2.6: Characterisation of glycerol core flow dynamics under varying flow rates of sheath flows. Results are obtained by applying glycerol core at 12.5 µl/min while varying the sheath flow rate from 350 to 1500 µl/min: (a) Schematic representation, (b) Variations of core average length against sheath flow rate, (c) Variations of core pinch off frequency against sheath flow rate, and (d) Variations of core pinch off frequency against core average length.
Secondly, the flow rate of water sheath flows is set to 850 µl/min while the glycerol core flow rate is varied from 12.5 to 75 µl/min (Figure 2.7a). Applying core flow at 12.5 µl/min leads to formation of an oscillating core which does not pinch off (Figure 2.7b). Increase of core flow rate beyond 25 µl/min leads to increasing the length and width of the core. The ‘viscous waves’ induced at the core/sheath flow interface can cause huge disturbances along the core structure and pinch it off (Figure 2.7c-e).

The average length of the core increases linearly with respect to core flow rate (Figure 2.7f). In comparison, the pinch off frequency increases linearly against both the sheath flow rate (Figure 2.7g) and the average core length (Figure 2.7g-inset). The details of calculations are presented in Figure 2.8.
**Figure 2.7:** Characterisation of glycerol core flow dynamics under varying flow rates of the core flow. Results are obtained by applying water sheath flows at 850 µl/min while varying the core flow rate from 12.5 to 75 µl/min: (a) Schematic representation, (b-e) Glycerol core flow reaches its longest/shortest extents before/after being pinched off by water sheath flows, respectively (f) Variations of core length against core flow rate, and (g) Variations of core pinch off frequency against core flow rate. The results are obtained over an 8-second interval.
Figure 2.8: Characterisation of glycerol core flow dynamics under varying flow rates of the core flow. Results are obtained by applying water sheath flows at 850 $\mu$l/min while varying the core flow rate from 12.5 to 75 $\mu$l/min: (a) Schematic representation, (b) Variations of core average length against core flow rate, (c) Variations of core pinch off frequency against core flow rate, and (d) Variations of core pinch off frequency against core average length.
2.4.5 Mapping the core configuration under various core/sheath flow rates

Figure 2.9a illustrates the extent of glycerol core under varying flow rates of glycerol core and water sheath flows at the core/sheath viscosity ratio of 210:1. The flow rate of the core flow is varied from 1 to 25 µl/min while the flow rate of the sheath flow is varied from 1 to 2000 µl/min.

The core structure is governed by the ratio of core/sheath flows, and varies from ‘tube’ configuration [23] obtained at high core/sheath flow ratios to tapered configuration obtained at low core/sheath flow ratios. At a core flow rate of 1 µl/min, the core structure is highly stable and the core/sheath interface is very smooth (Figure 2.2). In order to produce similar core structures at a higher core flow rate of 2.5 µl/min, the sheath flow rate should be increased. However, the increase of sheath flow rate is associated with inducing of ‘viscous waves’ at the core/sheath interface and disturbing of the smoothly formed interface (Appendix Figure A3). A similar trend is observed at a core flow rate of 5 µl/min (Figure 2.3), although the ‘viscous waves’ have become stronger and larger due to increased sheath flow rate. By increasing the core flow rate to 12.5 µl/min, the ‘viscous waves’ become so intense that they are able to break the core into two parts. Under these conditions, the core will have a dynamic structure, and its length varies between maximum/minimum values corresponding to pre/post pinch off conditions (Figure 2.4). The core pinch off happened at the sheath flow rates ranging from 400 to 800 µl/min (Appendix Figure A5). Applying sheath flow rates ranging from 850 to 1250 µl/min led to harmonic oscillation of the core while applying higher sheath flow rates formed a highly stable core structure (Appendix Figure A6). Similar trends can be observed at a core flow rate of 25 µl/min with the core pinch off occurring at the sheath flow rates ranging from
450 to 2000 µl/min. The core is expected to oscillate harmonically at sheath flow rates higher than 2000 µl/min, which are not analysed here due to leakage issues.

According to Figure 2.9a, the dynamics of highly viscous core can be classified into stable, semi-stable and unstable regimes. For the core/sheath viscosity ratio of 210:1 studied here, ‘stable’ core (stable core and with smooth core/sheath interface) is formed at the core flow rate of 1 µl/min, ‘semi-stable’ core (disturbed core with ‘viscous waves’ induced at the core/sheath interface) is formed at the core flow rates of 2.5 and 5 µl/min, and finally ‘unstable’ core (pinched off or oscillating core) is formed at the core flow rates of 12.5 and 25 µl/min.

Extended experiments are conducted to explore the dynamics of highly viscous core flow at the core/sheath viscosity ratio of 60:1. The results of these experiments are presented in Appendix Figures A7 to A12 for various core flow rates of 1, 5, 12.5 and 25 µl/min. Figure 2.9b depicts the extent of glycerol core under varying flow rates of core and sheath flows obtained at the reduced core/sheath viscosity ratio of 60:1. Reducing the viscosity ratio leads is associated with reducing the sheath flow rates to produce core structures similar to Figure 2.9a. Experiments indicate the formation of ‘stable’ core structures at the core flow rate of 1 µl/min (Appendix Figure A7), ‘semi-stable’ core structures at the core flow rates of 5 and 12.5 µl/min (Appendix Figures A8 and A9), and ‘unstable’ core structures at the core flow rate of 25 µl/min (Appendix Figures A10 to A12).
Figure 2.9: Extent of glycerol core flow at various combinations of core and sheath flow rates and two different viscosity ratios. (a) At a core/sheath viscosity ratio of 210:1, the core flow exhibits stable regime at a core flow rate of 1 µl/min, semi-stable regime at core flow rates of 2.5 and 5 µl/min, and unstable regime at core flow rates of 12.5 and 25 µl/min. The unstable regime is characterised by the length of the core varying within a maximum/minimum range before/after pinch off. (b) At a reduced core/sheath viscosity ratio of 60:1, the core flow exhibits stable regime at a core flow rate of 1 µl/min, semi-stable regime at core flow rates of 5 and 12.5 µl/min, and unstable regime at a core flow rate of 25 µl/min.
2.5 Conclusions

In summary, the dynamics of a highly viscous core flow confined by low viscosity sheath flows in a microfluidic flow focusing channel has been investigated. The mixing between the core and sheath flows reduces the width of the core flow continuously. At high core/sheath flow ratios, the reduction of core width is minimal and does not affect the overall structure of the core flow. However, at low core/sheath flow ratios, the reduction of core width is so quick that the core flow becomes tapered before forming a ‘viscous thread’ along the middle of the channel.

The massive viscosity contrast between the core and sheath flows induces local disturbances at the interface of the two liquids, which can greatly impact the stability of the tapered core. Experiments indicate that the stability of the tapered core depends on the magnitude of the sheath flow rate. At low sheath flow rates, the tapered core remains ‘stable’. In contrast, at high sheath flow rates, ‘viscous waves’ are induced at the interface of core/sheath flows, which can greatly impact the stability of the core flow. Depending on the magnitude of sheath flow rate, the induced ‘viscous waves’ can cause local deformation of the tapered core, making it ‘semi-stable’, and even can break it into two parts in an almost harmonic manner, making it ‘unstable’. The dynamics of broken core is studied under various combinations of core and sheath flows.
2.6 References


CHAPTER 3
Conclusions and recommendations for future work

3.1 Concluding remarks

As the first research contribution, the author investigated the stability of a highly viscous core flow made of glycerol confined between low viscosity sheath flows made of water within a microfluidic flow focusing channel. Experiments using high speed imaging indicated the existence of three core regimes, including stable, semi-stable and unstable ones. The ‘stable’ core regime is associated with the gradual tapering of the core flow until forming a thin ‘viscous thread’ along the middle of the channel. The core/sheath flow interface remains highly smooth in this regime. The ‘semi-stable’ core regime is associated with inducing of ‘viscous waves’ at the interface of the core/sheath flows. These ‘viscous waves’ disturb the interface and lead to local deformation of the tapered core. The ‘unstable’ core regime is associated with strong and asymmetric ‘viscous waves’ at the interface, such that the core cannot maintain its integrity and is broken into two parts. This phenomenon, which has not been reported before is referred as to ‘core pinch off’ phenomenon.

As the second research contribution, the author studied the effect of sheath flow rate on the stability of core flow. Experiments indicate the formation of ‘stable’ core structures at low core/sheath flow rates, formation of ‘semi-stable’ core structures at intermediate core/sheath flows, and finally formation of ‘unstable’
core structures at high core/sheath flows. For the case of ‘unstable’ core structures, the length of the core varies between maximum/minimum values corresponding to pre/post pinch off conditions. The average length of the pinched off core reduces almost exponentially with respect to sheath flow rate, while the frequency of pinch off increases almost linearly with respect to sheath flow rate. Reducing the average length of the core below ~1 mm, leads to harmonic oscillation of the core under the asymmetric ‘viscous waves’. Alternatively, reducing the average length of the core below ~0.5 mm, leads to formation of highly stable core with a smooth interface.

As the third research contribution, the author studied the dynamics of highly viscous core flow at two core/sheath viscosity ratios of 210:1 and 60:1, and demonstrated the existence of stable, semi-stable and unstable core regimes in both viscosity ratios. These studies indicate that lowering the viscosity ratio reduces the ‘viscous wave’ disturbances at the interface of core/sheath flows, which can be attributed to reducing of velocity gradients across the interface. This means that the sheath flow rate (and consequently the core flow rate) needs to be increased to produce the disturbances observed at a higher viscosity ratio. For example, for a core/sheath viscosity ratio of 210:1 the unstable core regime is obtained at core flow rates of 12.5 and 25 µl/min, whereas for a core/sheath viscosity ratio of 60:1 the unstable core regime is obtained at a core flow rate of 25 µl/min.
3.2 Recommendations for future work

In this work, the core/sheath flow has been produced by applying a pair of miscible core/sheath flows (glycerol/water) through a flow focusing channel. The work can be extended by applying a pair of immiscible core/sheath such as oil/water, in which interfacial forces might govern the stability of the core.

Here, extensive experimental analysis has been conducted to investigate the dynamics of core/sheath flows under various combinations of flow rate, flow ratios and viscosity contrasts. The work can be extended by performing numerical analysis using computational fluid dynamics techniques, which enables better understanding of the physics underlying the stability of the core, especially when undergoing the ‘pinch off’ phenomenon.

The unstable characteristics of the core flow during the ‘pinch off’ phenomenon might enhance the convective heat transfer at the entrance of the flow focusing channel, which require further investigation.
Appendix 1: Core/sheath viscosity ratio = 210:1, core flow rate = 1 µl/min

Figure A1. Expanded version of Figure 2.2, demonstrating the formation of stable core/sheath flows. Results are obtained under the following conditions: (i) Core flow rate is set to 1 µl/min, and (ii) Sheath flow rates are varying from 1 to 120 µl/min.
Appendix 2: Core/sheath viscosity ratio = 210:1, core flow rate = 5 µl/min

Figure A2. Expanded version of Figure 2.3, demonstrating the inducing of interfacial instabilities and formation of ‘viscous waves’ at the interface of core/sheath flows. Results are obtained under the following conditions: (i) Core flow rate is set to 5 µl/min, and (ii) Sheath flow rates are varying from 5 to 600 µl/min.
Appendix 3: Core/sheath viscosity ratio = 210:1, core flow rate = 2.5 µl/min

Figure A3. Lowering the core flow rate to 2.5 µl/min leads to similar instability patterns presented in Figure 2.3 and Figure A2, although the instabilities are weaker. Results are obtained by varying sheath flow rates from 2.5 to 300 µl/min.
Appendix 4a: Core/sheath viscosity ratio = 210:1, core flow rate = 12.5 μl/min, sheath flow rates from 12.5 to 375 μl/min

Figure A4. Increase of core flow rate to 12.5 μl/min leads to similar instability patterns presented in Figure 2.3 and Figure A3, although the instabilities are stronger. Results are obtained by varying the sheath flow rates from 12.5 to 375 μl/min. It should be noted that increasing the sheath flow rate beyond 375 μl/min leads to pinching off the core, as comprehensively presented in Figure A5.
Appendix 4b: Core/sheath viscosity ratio = 210:1, core flow rate = 12.5 µl/min, sheath flow rates are varying from 400 to 800 µl/min

Figure A5. Expanded version of Figure 2.5b-e, demonstrating the core pinch off at various sheath flow rates. Results are obtained under the following conditions: (i) Core flow rate is set to 12.5 µl/min, and (ii) Sheath flow rates are varying from 400 to 800 µl/min.
**Appendix 4c:** Core/sheath viscosity ratio = 210:1, core flow rate = 12.5 µl/min, sheath flow rates are varying from 850 to 1500 µl/min

![Diagram of flow rates and core/sheath viscosity ratio]

**Figure A6.** Expanded version of Figure 2.5b-e, demonstrating the harmonic oscillations of the core under asymmetric ‘viscous waves’. Results are obtained under the following conditions: (i) Core flow rate is set to 12.5 µl/min, and (ii) Sheath flow rates are varying from 850 to 1500 µl/min.
**Appendix 5:** Core/sheath viscosity ratio = 60:1, core flow rate = 1 µl/min

**Figure A7.** Formation of stable core/sheath flows. Results are obtained under the following conditions: (i) Core flow rate is set to 1 µl/min, and (ii) Sheath flow rates are varying from 1 to 75 µl/min.
Appendix 6: Core/sheath viscosity ratio = 60:1, core flow rate = 5 µl/min

Figure A8. Formation of ‘viscous waves’ at the interface of core/sheath flows. Results are obtained under the following conditions: (i) Core flow rate is set to 5 µl/min, and (iii) Sheath flow rates are varying from 5 to 250 µl/min.
Appendix 7: Core/sheath viscosity ratio = 60:1, core flow rate = 12.5 \mu\text{L}/\text{min}

**Figure A9.** Increase of core flow rate to 12.5 \mu\text{L}/\text{min} leads to similar instability patterns presented in **Figure A6**, although the instabilities are stronger. Results are obtained under the following conditions: (i) Viscosity ratio of core/sheath flows is set to 60, (ii) Sheath flow rates are varying from 12.5 to 900 \mu\text{L}/\text{min}. 
Appendix 8a: Core/sheath viscosity ratio = 60:1, core flow rate = 25 µl/min, sheath flow rates from 25 to 400 µl/min

Figure A10. Increase of core flow rate to 25 µl/min leads to similar instability patterns presented in Figure A9, although the instabilities are stronger. Results are obtained by varying the sheath flow rates from 25 to 400 µl/min. It should be noted that increasing the sheath flow rate beyond 400 µl/min leads to pinching off the core, as presented in Figure A11.
Appendix 8b: Core/sheath viscosity ratio = 60:1, core flow rate = 25 μl/min, sheath flow rate from 450 to 800 μl/min

Figure A11. Referring to Figure A10, increase of sheath flow rate beyond 400 μl/min leads to pinching off the core. Results are obtained by varying the sheath flow rate from 450 to 800 μl/min. The left column presents the core before pinch-off, whereas the right column presents the core after pinch-off.
Appendix 8c: Core/sheath viscosity ratio = 60:1, core flow rate = 25 \mu l/min, sheath flow rates ranging from 850 to 1250 \mu l/min

Figure A12. Referring to Figure A11, increase of sheath flow rate beyond 800 \mu l/min leads to shortening of the core. The shortened core harmonically oscillates under the influence of ‘viscous waves’ but does not pinch off for the sheath flow rates ranging from 850 to 1250 \mu l/min, similar to conditions shown in Figure 2.6. Further increase of sheath flow rate leads to immediate tapering of the core after leaving the central channel. The shortened core becomes very stable and does not oscillate, similar to conditions shown in Figure 2.5e.