Study of Immiscible Liquid-Liquid Microfluidic Flow using SPH-based Explicit Numerical Simulation

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A thesis submitted for the degree of Doctor of Philosophy

School of Chemical Engineering
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Australia

November 2015
Declaration

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Hamideh Elekaei Behjati

22 November 2015
Abstract

Microfluidic devices are utilized in a wide range of applications, including micro-electromechanical devices, drug delivery, biological diagnostics and micro-fuel cell systems. Of particular interest here are liquid-liquid microfluidic systems; which are used in drug discovery, food and oil industry amongst others. Increased understanding of the fundamentals of flows in such devices and an improved capacity to design them can come from modelling. In the case of liquid-liquid flows in microfluidic systems, it is necessary to explicitly model the behaviour of the individual liquid phases. Such explicit numerical simulation (ENS) as it is termed requires advanced numerical methods that are able to evaluate flow involving multiple deforming fluid domains within often complex boundaries. Smoothed Particle Hydrodynamics (SPH), a Lagrangian meshless method, is particularly suitable for such problems. This use of a CFD allows determination of parameters that are difficult to determine experimentally because of the challenges faced in microfabrication. The study reported in this thesis addresses these concerns through development of a new SPH-based model to correctly capture the immiscible liquid-liquid interfaces in general and for a microfluidic hydrodynamic focusing system in particular. The model includes surface tension to enforce immiscibility between different liquids based on a new immiscibility model, enforces strict incompressibility, and allows for arbitrary fluid constitutive models. This work presents a detailed study on the effects of various flow parameters including flowrate ratio, viscosity ratio and capillary number of each liquid phase, and geometry characteristics such as channel size, width ratio, and the angle between the inlet main and side channels on the flow dynamics and topological changes of the multiphase microfluidic system. According to our findings, both flowrate quantity and flowrate ratio affect the droplet length in the dripping regime and a large viscosity ratio imposes an increase in the flowrate of the continuous phase with the same capillary number of the dispersed phase to attain dripping regime in the outlet channel. Also, increasing the side channel width causes longer droplets, and the right-angled design makes the most efficient focusing behaviour. This study will provide great insights in designing microfluidic devices involving immiscible liquid-liquid flows.
Achievements

Three following papers were achieved from this work:


Acknowledgements

Over the past four years, I was inspired and supported by many people without whom I would never have been able to finalize this dissertation. This section is therefore dedicated to all those people who have shared their time, enthusiasm and expertise with me, admitting that I will never be able to thank enough those whom I am indebted the most. I would like to express my deepest gratitude and appreciation to all of them.

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<tr>
<td>Ca</td>
<td>Capillary Number</td>
</tr>
<tr>
<td>CAD</td>
<td>Computer Aided Design</td>
</tr>
<tr>
<td>CFD</td>
<td>Computational Fluid Dynamics</td>
</tr>
<tr>
<td>DNS</td>
<td>Direct Numerical Simulation</td>
</tr>
<tr>
<td>DPD</td>
<td>Dissipative Particle Dynamics</td>
</tr>
<tr>
<td>ENS</td>
<td>Explicit Numerical Simulation</td>
</tr>
<tr>
<td>Eo</td>
<td>Eotvos Number</td>
</tr>
<tr>
<td>FDM</td>
<td>Finite Difference Method</td>
</tr>
<tr>
<td>FEM</td>
<td>Finite Element Method</td>
</tr>
<tr>
<td>FVM</td>
<td>Finite Volume Method</td>
</tr>
<tr>
<td>HPC</td>
<td>High-Performance Computing</td>
</tr>
<tr>
<td>LBM</td>
<td>Lattice Boltzmann Method</td>
</tr>
<tr>
<td>LGA</td>
<td>Lattice Gas Automata</td>
</tr>
<tr>
<td>LOC</td>
<td>Lab-On-a-Chip</td>
</tr>
<tr>
<td>LoG</td>
<td>Laplacian of Gaussian</td>
</tr>
<tr>
<td>LSM</td>
<td>Level Set Method</td>
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<td>MD</td>
<td>Molecular Dynamics</td>
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<td>Mo</td>
<td>Morton Number</td>
</tr>
<tr>
<td>NNPS</td>
<td>Nearest Neighbouring Particle Searching</td>
</tr>
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<td>Oh</td>
<td>Ohnesorge Number</td>
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<tr>
<td>PPE</td>
<td>Pressure Poisson Equation</td>
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<tr>
<td>Re</td>
<td>Reynolds Number</td>
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<td>SD</td>
<td>Standard deviation</td>
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<td>SPH</td>
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<td>VOF</td>
<td>Volume of Fluid</td>
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<td>µTAS</td>
<td>Micro Total Analysis System</td>
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<th>Symbol</th>
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<th>Description</th>
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<tr>
<td>a</td>
<td>[m]</td>
<td>Length of the major axis of the ellipsoid</td>
</tr>
<tr>
<td>A</td>
<td>[m²]</td>
<td>Area</td>
</tr>
<tr>
<td>b</td>
<td>[m]</td>
<td>Length of the minor axis of the ellipsoid</td>
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<tr>
<td>C_D</td>
<td></td>
<td>Drag coefficient</td>
</tr>
<tr>
<td>g</td>
<td>[m/s²]</td>
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<tr>
<td>h</td>
<td>[m]</td>
<td>Cut off distance</td>
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<td>Unit tensor</td>
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<tr>
<td>L₀</td>
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<td>N_p</td>
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<tr>
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<td>τ</td>
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<td>∇</td>
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**Subscript**

- **c**: Value for continuous phase
- **i**: Value for particle of interest
- **j**: Value for neighbouring particles
- **j**: Value for jetting
- **d**: Value for droplet
- **p**: Value for particle
- **w**: Related to wall
- **α**: $\alpha$-coordinate direction
- **β**: $\beta$-coordinate direction
- *****: Intermediate state

**Superscript**

- **α**: Number of dimensions
- **β**: Number of dimensions
- *****: Dimensionless value
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Chapter 1: **Introduction**

Microfluidics, the study of fluid motion at a micro-scale, is currently a growing research area. It has practical applications in the design of systems in which a small amount of fluid in channels with dimensions in the order of tens to hundreds of micrometres is manipulated and processed [1-3]. Increased understandings and developments in microfluidics will lead to drastic progress in biotechnology, drug discovery, microelectronics, energy generation, pharmaceutical, food and many other industries. For instance, microfluidics allows the breakup of a droplet of blood into thousands of micro-droplets to perform biological tests in parallel [3], and this method is far better than those current ones, which require much larger samples and have slower reaction times.

At micro-scale, a high surface area to volume ratio results in that the interfacial forces, such as viscous force and surface tension, become dominant and intensely influences the fluid flow behaviour [4]. The advancement in micro-engineering and the scale-dependence behaviour of fluids allow us to be able to accurately control multiple fluid interfaces, which have been utilised in different areas ranging from chemical processing and energy to medical science and biology [4]. Practically, microfluidics offers a wealth of advantages including accurate manufacturing of devices, and precise control of interfaces. Microfluidic technology allows us to manipulate the deformable and moving interfaces in multiphase systems to achieve the benefits which cannot be obtained from the macro-scale flow devices.

Of particular interest are such multiphase microfluidic systems, such as micro-extraction used in drug discovery [5, 6], and micro-emulsification in food, cosmetic, pharmaceutical and oil industries [7-12]. The multiphase flow and moving/deformed interfaces unfortunately are far less well developed compared to the single-phase counterpart. Although a variety of experimental studies have been done so far on the droplet-based microfluidics [13-21], they are still sporadic, because of only limited material used for fabrication, a small range of flow parameters, and few configurations and geometries tested during measurement. Importantly, these reports are more focused on the observed phenomena, and the physics for such phenomena is not well clarified. Therefore, a full
understanding of the fundamental physics of multiphase liquid flow dynamics and the effects of geometry in the microfluidics and process parameters on the moving interfaces are essentially needed. Experimental studies for microchannel multiphase flows are quite challenging and expensive, while analytical solutions at such small scales are limited to very few simple cases. Consequently, modelling and numerical studies are often required to address complex flows, which are poorly understood, and to facilitate developments in microfluidics. To this end, this project aims to develop a numerical model for the immiscible liquid-liquid phase in microfluidics, which can be used as foundation for building a design capability for multichannel microfluidics devices.

Due to complex features of multiphase flow in microchannel devices including moving interfaces, a large ratio of the surface to volume and micro-mechanics such as capillary forces and potential intricate boundaries (e.g. fluid-fluid interface dynamics), numerical simulation of multiphase flows in microfluidics is not straightforward. Computational Fluid Dynamics (CFD) has been extensively used to simulate fluid flows in micro-fluidic devices as indicated by literature [22, 23]. These have been, however, largely attributed to a single phase systems. In the case of a multiphase system, the dearth of studies is due to challenges in explicitly tracking the individual phases. It is not possible to simply adopt a multi-fluid modelling approach for the immiscible-liquid multiphase microfluidic system, which treats the immiscible liquids as a homogeneous mixture without any interfaces and is typically applied for macro-scale counterparts [24].

Explicit numerical simulation (ENS) is a general concept where one attempts to model all the significant phenomena explicitly rather than in a mean field or average way as is done in many engineering models; in this sense, direct numerical simulation (DNS) of turbulence is a class of ENS [25]. Explicit simulation of the multiphase system allows us to track individual phases, deal with complex phase geometries and topologies, and consider multiple time and/or length scales in one simulation [26]. In most of the cases in microchannels, it is become important to explicitly model the behaviour of the individual phases and the way they have interaction in multiphase flow. In addition to that, based on the application of microfluidic devices, we are interested in the shape of dispersed phase rather than the other things. It means that, in addition to the continuous fluid phase, disperse phase must be explicitly modelled.
In the explicit simulation, modelling and tracking the interface with large deformation and topological changes is the main challenge of traditional CFD methods which need high resolution meshes, mesh refinement, proper mesh aspect ratios and intricate discretization techniques. Among those challenging issues, mesh generation and regeneration is a problematic task. It is a time-consuming and computationally-expensive process for problems with a complex geometry. Constructing a regular mesh for an irregular or intricate geometry may carry more computational load than solving the problem itself. Equally challenging tasks are to determining accurate locations of the free surfaces, deformable boundaries and moving interfaces within the frame of the fixed grid. This mesh-based numerical simulation is an expensive operation, which also bears another source for modelling errors and numerical instability [27-30]. Therefore, simulation of multiphase microfluidic systems using mesh-based ENS methods is computationally expensive and inefficient. Therefore, algorithms based on the Lagrangian meshless method should be considered in these problems.

Meshless methods have been developed over the past few decades with more efficient computational schemes designed for more complicated problems. Regarding dynamic simulations of continuum materials experiencing large deformations, the mesh creation processes in conventional mesh-based methods are involved by inevitability re-meshing the domain with the intention to keep away from weaken accuracy of the results owing to reduced aspect ratios of the mesh and termination of the simulation because of mesh complication and inversion. In this context, meshless particle methods have no inherent restrictions on the extent of deformation of the considered domain, since the connectivity between particles is updated for each computation step. Therefore, particle method holds advantages for large deformation simulations as mesh-based methods require constant re-meshing to obtain well-behaved elements. Moreover, the method is capable to be connected with a CAD database much simpler than mesh-based approaches due to the fact that this method does not need to generate an element grid. Meshless particle methods also produce more accurate results. Discretization techniques in meshless particle methods allow prices depiction of geometric object in compared with traditional mesh-based methods [27, 31, 32].
One preferable meshless particle method is Smoothed Particle Hydrodynamics (SPH) [33] which is a fully Lagrangian meshless method. For computational fluid dynamics problems governed by the Navier-Stokes equations, SPH is a powerful solution technique. In the SPH method, a set of particles that have material properties and interact with each other under the area delimited by a weight function or smoothing function, demonstrate the state of a system [34]. With the aid of these discrete particles, the governing equations are discretised and a diversity of particle-based formulations is used to calculate the acceleration, velocity and local density of the fluid. Afterwards through an appropriate equation of state, fluid pressure is calculated and hence the particle acceleration will be achieved using the pressure gradient and density. Since the movement of the fluid is illustrated by the movement of the particles, there is no extra procedure for interface tracking in multiphase flows [35].

Compared to the long-established mesh-based numerical methods, SPH has some particular benefits. Firstly, SPH is a meshless particle method with Lagrangian nature, which can record temporal profile of the material particles and so flow of the system can be accordingly obtained. Secondly, by positioning appropriate particles at particular locations at the early stage in the process of the simulation, free surfaces, interfaces and moving boundaries can be traced. As a consequence, interface tracking is explicit through capturing the locations of the particles, much simpler than Eulerian methods. Therefore, SPH is an ideal candidate for modelling free surface and interfacial flow problems. Thirdly, SPH is also appropriate for situations where the object of interest is not a continuum, such as micro-scale and nano-scale simulation in bio-engineering and nano-engineering. In addition, SPH is reasonably less complicated in numerical implementation, especially in comparison with mesh-based methods. Therefore, the SPH will be chosen to apply to the immiscible liquid-liquid flow in microchannel system in this project.

There are only a limited number of numerical studies with regard to immiscible-liquid multiphase microfluidics in literature [23, 35-37]. Among them, lattice Boltzmann method (LBM) has been mostly employed for simulation of droplet-based microfluidic systems [32, 38, 39], however, LBM-based simulation suffer from stability problems [40].

This project will be one of the first to apply SPH, a powerful meshless particle method, to explicitly modelling immiscible liquid-liquid flows in microchannels. This novel
modelling will enable us to better understand the effect of flow parameters, microfluidic geometry, and interfacial phenomena in such microfluidic devices. Moreover, this work will provide great insights in designing microfluidic devices involving immiscible liquid-liquid flows.

The aim of the work reported in this thesis was directed towards developing the application of meshless method to the explicit numerical simulation of immiscible-liquid flows in microfluidic systems so as to achieve an increased understanding of fluid dynamics and phase morphologies within the phases in such systems. The study reported in this thesis addresses these concerns through two related developments. ENS of immiscible liquid-liquid flows based on incompressible SPH method with particular regard to correctly capturing the liquid-liquid interface represents the first development. This goal was achieved through modelling of the system, code development, and validation of the model by applying it to a range of interfacial problems (Objective I). The second development of the work has been divided into two parts: the first involved the application of the proposed model for the study of hydrodynamic focusing of a liquid flowing in a deep microchannel by a second immiscible liquid introduced through identical but symmetrically opposed microchannels (Objective II). The second involved building a basis for optimal design of the microfluidic system through determining the effects of various flow parameters and geometry characteristics on the flow dynamics and topological changes of the multiphase flow system (Objective III).

The thesis is laid out as follows. Chapter 2 provides a literature review that identifies the strength and weak points of the different CFD methods, some research gaps that support the aim of this thesis, and the technical background of the method used. Chapters 3, 4 and 5 report the work undertaken to achieve Objectives I, II and III, respectively; these chapters are papers, all of which will be submitted to the Physics of Fluids. The final chapter draws together the work reported here to make overall conclusions and suggest future work.
Chapter 2: Literature Review

2.1 Microfluidics

2.1.1 A brief history of microfluidics

Microfluidics is defined as the study of the behaviour of fluid flow geometrically confined to a micro scale with dimensions varying roughly between 100 nm and 100 µm [1, 2]. The potential for microfluidics to revolutionize the energy, chemical processing and medical fields has attracted significant interest and research effort over the past few decades. In the 1980s, research on microfluidic devices appeared due to the availability of the micromechanics technology [41]. With the development of the concept of “Micro Total Analysis System” (µTAS) [42-46], and subsequently Lab-on-a-Chip (LOC) [47-50], micro-fabrication techniques are capable of making microchannels with complex structures from resistant and inactive substrate materials which are appropriate for dealing with chemicals and biological samples [43-45, 51, 52].

Characteristically, a microfluidic system is composed of microfluidic channels, micro-reservoirs and reaction chambers at micro-meter length scales. It is possible to accomplish the unit operations including mixing, separation, reaction, detection and analysis, through one integrated micro-device [53, 54]. There have been several attempts for integration and parallelization of microfluidic devices. Best examples include Quake et al [48] for biological systems, Wei et al [55] and Torii et al [56] on droplet generation, and Tetradis-Meris et al [57] for emulsion formation. Applications of microfluidics have been explored in colloid science [58], plant biology [59] and process intensifications [60-62]. In particular, process intensifications involve the micro-chemical engineering technology [63-67], which can lead to improved quality products, increasing yields, reduction in investment costs, lower energy consumption, reduced environmental and safety risks [68], and control of extreme reactions [66, 69-71].
2.1.2 **Advanced techniques to make microfluidics**

Since the beginning of the microfluidic era, the techniques of fabrication of microfluidic devices have been in constant improvement. Examples of the fabrication techniques include soft-lithography [72], laser ablation [73], and X-ray lithography [74]. The main challenges of the common microfabrication techniques are connected with the construction of open microchannels and assembly of machined substrates to create an integrated structure. Open microchannels can be made through etching process which is expensive, and may involve aggressive materials [75].

Among the materials used for fabrication of the microfluidic chips, polydimethylsiloxane (PDMS) is of particular interest, since it is transparent, flexible, replicable, and low-cost, however, it swells and deforms in the presence of organic oils. Therefore, more robust materials such as glass, metal, silicon, and thiolene are needed to be applied for droplet-based microfluidic systems.

Moreover, significant efforts have to be done to develop functional parts of the microfluidic system, including microvalves for isolation purposes, and digital pumps for precise control of flow rates [76]. These difficulties associated with the microfabrication techniques and materials restrict the experimental research studies. Numerical studies, therefore, should be called to facilitate building of understanding of flow behaviour in such systems.

2.1.3 **Brief overview of current microfluidic flows**

Single phase microfluidic systems are based on continuous liquid flows through microchannels and are efficiently used for trapping, focusing and/or separation of solid particles [77], cells [78], and single DNA molecules [79]. Protein analysis through immobilized microfluidic enzymatic reactors is also another application of the single phase microfluidics [80]. Single phase microfluidic systems, however, are less suitable for tasks requiring a high degree of flexibility.

In contrast, multiphase flows provide different mechanisms for improving the performance of single phase systems. For example, immiscible multiphase microfluidic systems enable manipulating of discrete volumes of fluids in immiscible phases that benefit large
interfacial areas, fast mixing and reduced mass transfer limitations [81]. Examples include emulsions and droplet-based microfluidics with a wide range of applications from chemical and material processing to biology and medicine [4].

2.2 Multiphase microfluidics

Fluids of interest in many engineering and industrial applications involve more than a single phase. Multiphase microfluidic systems play a significant role in drug discovery, biomedical purposes and many other industrial fluid dynamical problems involving fluid transport and interfacial phenomena [43]. For instance, biomedical examples in Lab on Chip devices include emulsions of bacterial cells (solid-liquid-liquid flow) and analysis of whole blood samples (solid-liquid flow) [49, 82]. Other applications of multiphase microfluid flow systems are synthesis of a small-amount of fine chemicals or active pharmaceutical ingredients in chemical and pharmaceutical industries, where solid catalysts are used for gas or liquid reactants [44, 83, 84]. In this case, the hydrodynamics of multiphase flow has the most important effect on the reactor performance and consequently in the design of a reactor [44]. Some investigation has been done on the design of microchannel mixers in order to enhance the advection phenomena in liquid-liquid mixing processes, such as the micro-mixer designed by Bringer et al [85] for dilute solutions in aqueous drops dispersed in an oily continuous phase. Liquid-Liquid phase interactions have also been observed in mono-dispersed emulsions such as water-oil two phase flows in microfluidic devices [11, 57].

2.3 Immiscible Liquid-Liquid Microfluidics

Of particular interest in this project is multiphase flow of immiscible liquids in microfluidics. Through interaction of immiscible-liquid micro-flows, the flow and capillary instabilities emerge and threads, jets or monodispersed droplets form. This is the physical basis for producing emulsions and dispersion of one liquid phase in the other one [86, 87]. Such microfluidic systems are of relevance to emulsifications used in food [8, 9], cosmetics [7], pharmaceutical [6, 12] and oil industries [10, 11, 88], plus liquid-liquid extraction used in drug discovery [5, 89-91]. Figure 2-1 displays a schematic illustration of
(a) integrated microfluidic liquid-liquid extraction [5, 92] and (b) water-in-oil-in-water (W/O/W) emulsion [93].

Figure 2-1 Examples of liquid-liquid microfluidics; (a) integrated microfluidic liquid-liquid extraction [92] and (b) water-in-oil-in-water (W/O/W) emulsion in microchannels [93]

Immiscible liquid interfaces are important throughout many areas, particularly chemistry and biology, because of their unique chemical and interfacial behaviour. There is a capillary pressure across a curved liquid interface at equilibrium, which is described by Young-Laplace equation as:

\[ \Delta P_{cap} = \gamma k \]  

(1)

Where, \( \gamma \) and \( k \) are interfacial tension and local mean curvature at the interface, respectively. According to the equation, the variation of the pressure drop across the interface is a function of the interface curvature. For instance, the capillary pressure across the interface of a spherical water droplet with a radius of 1 \( \mu \)m in air is approximately 1000 times, larger than that of the capillary pressure of the same droplet with a radius of 1 mm [81]. When two immiscible liquids are brought into contact with each other, various flow patterns and phase
distributions can occur due to the interplay between various physical forces such as interfacial, gravitational, viscous and inertial forces. Figure 2-2 shows the different flow patterns such as threading, jetting, dripping, tubing, and viscous displacement [14] in the immiscible-liquid microchannel flow. At micro-scale, because of the laminar nature of the flow, the flow pattern is dictated by the competition between viscous forces and capillary force at the interface. The former tends to drag the liquids along the microchannel, while the latter tends to minimize the interface of the two liquids.

The competition of the various forces is affected by flow parameters, microchannel geometry and surface chemistry. These effects can be captured by a number of dimensionless numbers expressing the relative importance of the competing forces [2, 81, 86]. Scaling these forces based on flow conditions and fluid characteristics helps predict multiphase microfluidic behaviour, and introduce design guidelines for microfluidic devices.

One of the most important dimensionless numbers is capillary number (Ca) which is defined as the relative effect of viscous force to interfacial tension acting across the interface between two media:

Figure 2-2 Possible flow patterns in immiscible-liquid microfluidics at (a) cross-junction [14], the flow regimes from top to bottom show threading, jetting, dripping, tubing, and viscous displacement regimes, (b) T-junction [94], the flow patterns from top to bottom show slugs, monodispersed droplet, droplet populations, parallel flows, parallel flows with wavy interface, chaotic thin striations flow.
\[ Ca = \frac{\mu U}{\gamma} \] (2)

Where \( \mu \), \( U \) and \( \gamma \) denote viscosity, velocity and interfacial tension. Capillary number is calculated for both dispersed phase \( Ca_d \) and continuous phase \( Ca_c \). According to literature in the microfluidic context, Capillary numbers are usually less than 1.0 which means that capillary forces (or interfacial forces) play a more important role in the flow behaviour than viscous forces. When Ca is very small around zero, the viscous shearing effect can be neglected and the interfacial tension is enough to keep the interface away from any deformation, however by increasing the Ca, deformation of the interface increases and may lead to formation of droplets and threads. Figure 2-3 shows a typical flow map for an immiscible-liquid pair [14] based on Ca values of the dispersed and continuous phases. The flow map reveals how changes in capillary numbers can influence the flow patterns and instabilities. Five typical flow patterns are indicated by the map including threading, jetting, dripping, tubing, and displacement. The threading flow pattern refers to a long core thread of the main channel liquid. The jetting flow pattern refers to a thin thread that emits droplets smaller than size of the channel at a distance from the junction. In dripping flow regime, the droplet forms near the junction. The tubing flow pattern is viscous stress controlled and refers to a viscous core. At low momentum of the continuous phase viscous displacement occurs [14].

![Figure 2-3 Typical Ca-based flow map for immiscible-liquid two-phase microfluidics [14]](image-url)
In addition, the influence of flow rates on the flow pattern and topological changes can be determined from Reynolds number (Re), which represents the ratio of inertia to viscous forces:

\[ Re = \frac{\rho Ud}{\mu} \]  

(3)

Where \( \rho \) and \( d \) are fluid density and hydraulic diameter of microchannel, respectively. Literature shows that the flow rate of about 1 \( \mu l/s \) is typically applied in microchannels with a dimension in the order of 100 \( \mu m \) [14, 95]. Using these data, Re can be estimated in the orders of 0.1 to 1. At a small Reynolds Number (Re < 1), the flow is governed by viscous stresses and pressure gradients, and inertial effects can be omitted, which reflects the typical microfluidic characteristics. The other important dimensionless numbers in multiphase microfluidic systems include Weber number, which implies the influence of inertia with respect to surface forces (i.e. interfacial tension), and Eotvos number which refers to the ratio of gravity force to surface forces.

2.3.1 Experimental study of immiscible liquid-liquid microfluidic system

Nakajima et al. [17, 96] and Knight et al. [97] were pioneers for creating monodisperse emulsions in a microfluidic device, the former through an array of cross-flow microchannel network and the latter using a hydrodynamic focusing microfluidic system. Similarly, Thorsen et al [15] employed a T-junction microfluidic configuration for generating dispersed water droplets in oil emulations, and they demonstrated how the interface instability evolves due to a competition between shear forces and surface tension. Flow instability usually causes the formation of monodispersed droplets in microfluidic devices, whereas, a capillary instability is anticipated to create segmented flows with identical droplets in liquid-liquid systems [98, 99]. Stone et al. [100] experimentally investigated formation of liquid droplets in an immiscible liquid through a flow-focusing microfluidic device. They concluded a phase diagram that illustrates the drop size as a function of flowrate and flowrate ratio of two liquids. Dummann et al. [101] developed a capillary-microreactor concept and experimentally studied nitration of a single ring aromatic in an exothermic liquid-liquid capillary-microreactor which is proposed for intensification of heat and mass transfer. Dreyfuss et al. [102] used a microfluidic cross-junction for their experiments to examine the
role of wetting properties in controlling flow patterns. Moreover, they demonstrated that phase distribution is influenced by superficial velocities of both continuous and dispersed phases in immiscible-liquid microchannel flows. Cramer et al. [103] investigated dripping and jetting flow regimes in co-flowing streams. Furthermore, Okushima et al. [104] reported a new technique for producing double emulsions (water-in-oil-in-water dispersions) using a two-step droplet formation in T-shaped microchannels. Over the past decade, different microchannel configurations including T-junctions [35, 105], Y-junctions [18], cross-junctions [20, 38], flow focusing [14, 16, 100, 106-109], concentric injection [110] and membrane emulsification [111-113] have been reported for droplet formation process.

Understanding of flow dynamics and droplet formation mechanisms in the proposed microfluidic configurations has also been well progressed to facilitate a smart design of liquid-liquid microfluidic devices. In this context, Garstecki et al. [21] studied the effects of flow rates and material parameters on the mechanism of droplet formation and breakup in liquid-liquid emulsions using a microfluidic T-junction. Constraining the liquid-liquid flow between microchannel walls influences the rupture of an annular liquid core flow into droplets. At a small Capillary number, less than $10^{-2}$, the shear stress forced on the interface of the appearing droplet becomes so inadequate that the force is not able to deform significantly the interface. Thus, the droplet blocks approximately the whole cross-section of the microchannel and confines the continuous flow to the walls. Garstecki et al. [21] concluded that increasing Ca in a liquid-liquid system may lead to three different patterns for the formation of droplets including squeezing, dripping and jetting. Droplet generation in the squeezing regime is stimulated with pressure instabilities during the breakup process.

A successful example of the microfluidic configuration for producing monodispersed droplets is microfluidic hydrodynamic focusing wherein a centrally located flow is narrowed or hydrodynamically focused by two neighbouring flows through cross-like microchannels. For example, Xu et al. [16] set up a three-stream laminar flow configuration to hydrodynamically focus a more viscous liquid surrounded by two inviscid liquids to facilitate droplet formation through control of the flow rate ratio between the continuous phase and the dispersed phase. The efficiency of droplet formation in microchannels is mainly affected by the nature of liquids (viscosity and interfacial tension), flow parameters (flowrate ratio, capillary number), and geometrical characteristics. In this respect, Cubaud et al. [14] performed a
comprehensive experimental study on a few Newtonian immiscible liquids in a microfluidic hydrodynamic focusing device. They investigated the competition between viscosity and interfacial tension effects on the formation of different flow regimes using hydrodynamic focusing over a range of viscosities and interfacial tensions at a fixed geometry. Although their experiments cover a useful range of flowrate ratios, viscosity ratios and Ca, their results provide an insight to the flow behaviour in the conditions limited to the examined cases. We meant to develop a reliable simulation to cover a wide range of parameters in order to increase our predictive capacity for a smart design of immiscible-liquid microfluidic devices.

Despite the various studies on droplet formation and breakup process through different microfluidic junctions have been reported so far, a detailed study on the effects of geometrical characteristics and junction/inlet design of microchannels is still missing. The flow and junction geometry, the cross-section of the channels, the dimensions and the angle made by the inlet channels all can be influential on flow pattern. There are only a few researches on the geometry effects reported by literature. For instance, Ménétrier-Deremble et al. [114] found out the controlling effect of flow geometry in breakup process through their experiments related to droplet breakup in different-angled microfluidic T-junction configuration. Also, Tan et al. [115] considered a microfluidic system through a trifurcating junction to illustrate the effect of junction geometry on droplet coalescence. They deduced that wider junction lessens the shear force and aligns the droplets to the core section of the channel and let droplets to create a zigzag pattern resulting in a lateral coalescence. Abate et al. [116] demonstrated that droplet formation through a flow-focusing device occurs at higher capillary numbers than to that of a T-shaped microfluidic junction. Furthermore, Priest et al. [117] designed a Y-shaped microchip for the recovery of copper from leach solutions in which two liquid phases (organic and aqueous) are contacted in parallel through the main channel and separated at a Y-junction. They also concluded that the proposed microchip has greater potential for recycling of volatile liquids. More recently, Li et al. [19] focused on the droplet formation mechanism in an immiscible liquid-liquid system with T-junction distributor and noticed that smart geometric design of micro-fluidic devices can affect controlling the droplet formation on demand. Also, Kunstmann-Olsen et al. [108] studied experimentally and theoretically a two-dimensional microfluidic cross-flow and showed that the local geometry of the microchannel-junction affects the hydrodynamic focusing.
behaviour. We aimed to figure out these effects through our simulation and present a robust model to predict behaviour of immiscible-liquid flows under the different conditions of process, material and geometry in a microfluidic hydrodynamic focusing system.

2.4 Numerical study of immiscible liquid-liquid microfluidic system

There is an increasing need to understand and exploit the link between liquid-liquid phase morphology and fluid dynamics within the phases on the one hand and the processing conditions that produce them on the other. For example, the droplet size distribution of an emulsion produced through agitation is a function of the balance between droplet breakup and coalescence, which can be controlled by the surfactant and stabilizer concentration and relative velocities of the phases [118-120]. Microfluidic production of encapsulates provides a further example: in this case, encapsulate morphology can be varied through the nature of the flow-focusing in the microchannels and, amongst other things, the continuous-to-dispersed phase flow rate ratio [121, 122]. The challenges faced in experimentally elucidating these types of relationships are significant, however. For example, the visualization of the morphologies of the phases and flow fields therein are still very much in their infancy [123-126]. Models that treat the phases and interfaces between them explicitly have, therefore, an important role to play in building understanding of liquid-liquid systems and exploiting this understanding in a systematic way.

Models of liquid-liquid systems in which the individual phases and interfaces between them are treated explicitly are long-standing [127-130]. The earliest models, which focus on droplets in a continuous phase, include those of Taylor [131], Mason and co-workers [132, 133], Cox and co-workers [134, 135], and Acrivos and co-workers [136-138] amongst others. Whilst these models were important in building understanding, they are limited by a good number of the simplifying assumptions [139], including negligible inertial effects (i.e. small Reynolds numbers), small viscosity ratio ranges, and regular droplet shapes (e.g. spheres; ellipsoids). Indeed, it was shown by Mason et al. [135] that no wholly theoretical approach can encompass the entire range of process parameters adequately. Phenomenological approaches have been adopted to overcome some limitations associated with wholly analytical models. For example, Maffettone et al. [87]
used such an approach, determining its parameters by ensuring it matches analytical model results in appropriate limits. Such models are, however, also limited by, for example, underpinning assumptions about droplet shape and the absence of any connection between the model parameters and underlying fundamentals.

Adoption of wholly numerical approaches can, in principle, overcome limitations faced by analytical and related models. Some of the earliest numerical models include those of Acrivos and co-workers [139-141], who studied the deformation and breakup of a viscous droplet freely suspended in another liquid within extensional and shear flow fields. More recent examples include the works of Loewenberg, Hinch and Davis [142-148], which are based on a boundary-integral approach. Afterwards, interface tracking techniques such as Volume of Fluid (VOF) [130, 149, 150] were used for simulation of immiscible fluids. For instance, Richards et al. [151] simulated formation of immiscible liquid jets and their transient to breakup into drops numerically based on the VOF method. Although this model represent acceptable mass conservation properties, the geometrical characteristics of the interface, the maintenance of a sharp boundary between the different fluids, and the surface tension computation are cumbersome to be implemented by VOF, since the volume fractions are non-smooth functions [149]. As a remedy, a number of techniques such as Level Set method (LSM) [152, 153] were developed to improve the interface resolution and include the surface tension computations.

Level Set method set up a smooth distant function in which zero value displays the position of the interface. As LSM is based on a stream-function formulation and needs to use a vector potential and solve three elliptic equations, therefore, it becomes more expensive in three dimensions [154]. Unlike the Level Set method, improved VOF methods usually involve an efficient interface reconstruction step based on the local volume fraction and orientation of its gradient. However, it is worthwhile adding that three-dimensional reconstruction process and as discussed before tracking of curved interfaces by this method is truly problematic [155]. Alternatively, some techniques made use of markers to follow the interfaces. Marker methods set up a secondary moving grid aimed to present precisely the interface location and curvature. Particle-in-Cell methods[156] which pack the area close to the interface with markers moving with the flow are another example of this category. The downside of these methods, however, is
that they can become rather expensive as the number of markers increases. In addition, they may suffer from difficulties in controlling the addition or deletion of markers when the interface is extended or condensed by the flow.

As all these methods involve the use of computational meshes, mesh adjustment is required as the phases deform. Tracking the interface with substantial deformation and topological changes is the main challenge of the traditional methods which need high resolution meshes, mesh refinement, proper mesh aspect ratios and intricate discretization techniques. Generally, in traditional mesh-based methods, the finite element and finite volume methods solve the Navier-Stokes equations by integrating the equations over a mesh of finite elements or volumes. To make the set of nonlinear equations linear, unknowns are substituted with finite difference equations, and subsequently the linear equations are solved through an iterative method. To solve the governing equations, the flow domain has to be properly discretised into tiny volumes or elements resulting in the solution of a partial differential equation. This process is referred to as mesh or grid generation. If a proper grid or mesh is not generated, geometric reliability problems will occur regarding the representation of the computational domain. Furthermore, if a proper grid is not produced, instabilities will happen when trying to solve the governing equations. Figure 2-4 illustrates this difficulty with meshing and re-meshing process in different microfluidic systems. Once a solution has been formed, the flow parameters can be extracted for each individual cell or element for analysis. These various operations are complex, computationally expensive and prone to numerical instability [27]. Treatment of coalescence and rupture of interfaces between phases is extremely challenging if not impossible in many of these techniques [157]. In conclusion, simulation of immiscible liquid-liquid systems using mesh-based Computational Fluid Dynamics (CFD) methods will be computationally very expensive and a formidable task to undertake [27].

Above and beyond the mentioned strengths and weaknesses, modelling of multiphase micro-flows is particularly challenging because it is not possible to simply adopt a multi-fluid modelling approach— which treats the immiscible liquids as a homogeneous mixture without any interfaces—as is typically done for macro-scale counterparts [24, 158]. At micro-scale, the flow is predominantly laminar, the discrete phases are controllable and of the order of the size of the geometry. Existence of movable and complex boundaries at the
liquid-liquid interface, and liquid-solid contact line dynamics makes the simulation to be problematic. In such scales, the surface to volume ratio is extremely high and notable, constitutive relations of liquids are mainly under the influence of the boundary, and the no-slip boundary condition is not valid. To overcome these issues, we must explicitly model the behaviour of the individual phases and their interaction at the interface. Explicit Numerical Simulation (ENS) of multiphase systems takes advantages of investigating complex phase geometries and topologies and considering multiple time and length scales in one simulation [26]. As a result, traditional mesh-based CFD methods cannot be efficiently applied for simulation of multiphase microfluidics. Thus, for some cases such as immiscible-liquid flows in microchannels with complicated geometries, a more powerful approach should be called to remove restrictions on mesh entanglement, complex geometry, Reynolds number and so forth. This study aims to develop a meshless method for modelling of such systems.

Figure 2-4 Computational mesh close to the interface for a rising droplet [159] and (b) adaptive mesh refinement for a droplet impact on a Liquid-Liquid interface [160]
Particle-based approaches have been used as a remedy to the issues faced with traditional mesh-based approaches. Historically, particle methods present the most primitive efforts of developing meshless numerical schemes. Particle methods have been demonstrated to be powerful in numerically treatment of large deformation processes and discrete phenomena [27]. These methods take the advantage of having no inherent restrictions normally on the permissible deformation of the considered domain in opposition to mesh-based methods. As for the former, the connectivity between particles is created as a part of computation and can vary with time which is not the case for 3D meshes. This positive side of particle methods is very important for substantial deformation simulations as mesh-based methods may necessitate constant re-meshing to keep well-behaved elements up. Moreover, the meshless particle method is capable to be connected with a CAD database more effortlessly than mesh-based approaches, due to the fact that it is not essential to generate an element grid. The other considerable aspect of particle methods is that the accuracy and damage of the components can be simply controlled and meshless discretization supplies precise depiction of geometric object [27, 31].

Lattice Boltzmann method (LBM) [32, 89] is a mesh-based particle method which was developed for fluid flow applications. The idea behind this method is to construct simplified kinetic models integrating the essential physics of microscopic or mesoscopic systems to facilitate the macroscopic averaged parameters follow the desired macroscopic equations [32]. LBM has been widely employed in numerical simulation of multiphase microfluidic systems. For instance, Wu et al. [38] presented a lattice Boltzmann model for simulation of immiscible two-phase flows in a cross-junction microchannel and also studied the effect of Ca on droplet formation in such a cross-flow systems. Liu et al. [20] studied the effect of flowrate ratio and capillary number on droplet formation in a microfluidic cross-flow configuration using a multiphase lattice Boltzmann model. They used a limited range of capillary numbers (Ca<0.01) and viscosity ratio (λ≤1/50), but a variety of flowrate ratios in their simulation and recognized three different flow regimes for their different flowrate ratios at a fixed capillary number. Gupta et al. [161] used LBM to show how the length of the plugs in the squeezing regime are influenced by the width of the two channels and the depth of the assembly in a microfluidic T-junction. Additionally, they examined the channel width ratio effect on the droplet formation process in a T-
junction microchannel at high capillary numbers. The benefit of LBM, against the conventional interface tracking methods, lies in its capability that maintains sharp interfaces without weighty effort. However, this method as well as its less widely-used antecedent, lattice-gas automata (LGA) [162, 163], are also not without their problems and limitations. For example, Galilean invariance is lost in LGA, leading to a spurious density-dependent factor multiplying the inertial term in the momentum conservation equation whose effect cannot be removed [164]. On the other hand, LBM-based simulation of immiscible multiphase fluid flows tend to suffer from stability problems [40]. In the LBM, the spatial space is discretised in a way that it is consistent with the kinetic equation. Therefore, it requires not only grid, but also the grid has to be uniform. This actually causes problems at curved boundaries and so development of irregular grids is needed [27]. In addition, LBM needs the input thermophysical properties to be adjusted by the microscopic parameters as a result of microscopic dynamics of distribution functions, and also due to the kinetic nature of the LB method, hydrodynamic boundary conditions are difficult to satisfy on a grid point exactly [157]. Therefore, specific techniques are required to enforce boundary conditions. This work is to develop a robust numerical model for immiscible liquid microfluidic systems that eliminates the aforementioned needs exist for LBM simulations of microfluidic systems.

An alternative approach is Dissipative Particle Dynamics (DPD) [164-166], which has been used to study the rheology of complex fluids [167]. DPD is an off-lattice mesoscopic simulation technique which involves a set of particles moving in continuous space and each particle represents a cluster of atoms or molecules, and, as a result of the internal degrees of freedom associated with each particle, the particle-particle interactions include random and dissipative contributions [168]. The drawback of DPD is determination of liquid properties and transport coefficients of the system. Also, because of limitation of small time steps, DPD is not computationally efficient [169, 170].

One preferable particle method that has a much longer history is Smoothed Particle Hydrodynamics (SPH) [33, 171-173]. In addition to avoiding all the issues associated with mesh-based methods, SPH has the advantages that it is derived directly from the Navier-Stokes equations and, hence, can be easily extended to any non-Newtonian matter. It also has an innate capacity to adapt through time to better capture steep gradients as they form.
Explicit numerical simulation of liquid-liquid micro-flows using SPH can be the most appropriate approach for capturing liquid-liquid interface deformations [26] thanks to its advantages over the traditional methods. For example, moving and deformable interfaces can be easily traced using SPH apart from the complexity of the motion of the particles, and so numerical investigation of multiphase flows through microfluidic devices with similar behaviour can be reliably and efficiently relied on the Smoothed Particle Hydrodynamics Method.

Our findings on the subject of multiphase microfluidic systems are still shallow and need more attempts, however, experimental studies of such micro-scale flows are quite challenging and expensive, and the analytical solutions are limited to a few simple cases. Therefore, development of a powerful numerical technique is highly needed for comprehensive and effective study of multiphase microfluidic flows with the intention of achieving a better understanding of dynamics of such flows. The significance of this thesis lies in the development of a new incompressible SPH-based model to the ENS of immiscible-liquid microchannel flows with particular regard to correctly capture the liquid-liquid interface. This work contributes in building of a basis for optimal design of the microfluidic systems through determining the effects of various flow parameters and geometry characteristics on the dynamics of the multiphase flow system.

### 2.5 Smoothed Particle Hydrodynamics (SPH) Method

Smoothed Particle Hydrodynamics, considered as the oldest up to date meshless particle method, was initially developed by Gingold et al. [174] to solve astrophysical problems in three dimensional open spaces. They introduced SPH as a technique for finding approximate numerical solutions of the momentum equations by replacing the fluid with a set of particles [173]. In this technique, an integral representation of a function, called kernel approximation, is used to solve the governing partial differential equations using a Lagrangian approach. A weakly compressible SPH algorithm [175] was developed for simulation of fluids for which the assumption of slightly compressible is valid such as coastal applications[176]. The weakly compressible SPH method was also used in some multiphase flows as dust gas flows [177], interfacial flows [178] and so forth.
Morris et al. [179] incorporated the standard SPH formulation with a quasi-incompressible equation of state to model incompressible flows of low Reynolds number. Later on, Shao et al. [180] presented the incompressible SPH model to study numerically both Newtonian and non-Newtonian flows based on the concept of projection SPH [175] and the moving particle semi-implicit method [181]. In this method, an invariant density equation was used to calculate the required fluid pressures. Moreover, Hu et al. [182, 183] extended the incompressible SPH model for simulation of multiphase flows under a wide range of density ratios. In this approach, the standard SPH formulations were modified to account for the discontinuity of densities across the interface boundary. Correspondingly, Hu et al. [29] examined the applicability of SPH for simulation of multiphase flows both at macro- and meso-scales.

In addition to being used to model multiphase fluid systems via the two-fluid model [177, 184], SPH has been used extensively to model such systems in which the interfaces are explicitly resolved. Many of these ignore surface tension (e.g. [185-190]) and are, hence, of limited use for liquid-liquid systems. Others have, however, sought to include surface tension effects. One broad approach is that first proposed by Morris [191], which embedded the continuum surface force (CSF) approach [192] within SPH. This approach was disadvantaged by the need to evaluate interface curvature, a numerically challenging task that is prone to numerical instability. Adams and co-workers [29] addressed this issue, although computational expense is still an issue, and the approach has since been extended to systems involving solid surfaces of desired wettability [29, 193] and, strictly incompressible systems [182, 183, 194, 195]. A second broad approach sees SPH combined with the Cahn-Hilliard model [196, 197]; this diffuse interface approach is yet to see wide use. The third and final broad approach to including surface tension within SPH is via incorporation of inter-particle forces that lead to phase separation. The earliest example of this approach was due to Nugent and Posch [198] whose inter-particle force was inspired by the intermolecular interaction term that arises in the van der Waals equation of state that was used to close their compressible flow SPH model; this approach has been widely used since (e.g. [199, 200]) as it is far simpler and cheaper than the CSF-based approach, although it requires calibration through modelling of the Young-Laplace problem. A variety of other inter-particle force models have also been proposed since,
some were inspired by DPD [170, 201], others are density-dependent [202], whilst various arbitrary forms have also been used, including trigonometric [203] and inverse square law [204] forms. In all these cases, however, strict incompressibility has not been imposed; this has only been done once before [205], using the trigonometric interaction model [203].

Although there is an increasing interest in meshless methods specifically SPH, only a limited number of their application in porous media and microfluidics have been reported so far [157]. In this context, Tartakovsky et al. [206] utilized a SPH-based simulation to investigate behaviour of immiscible and miscible fluid flows in porous media. They also explored the influence of pore scale heterogeneity and anisotropy on such flows. Additionally, they applied SPH for modelling of reactive transport and mineral precipitation in porous media [207]. Successful examples of SPH in other applications include free surface flows and simple unsteady flows in microfluidics [208, 209]. In the following, a brief description of general SPH formulation is given.

### 2.5.1 General SPH Formulation

In SPH formulation the various influences of the Navier-Stokes equation are simulated by a set of forces that act on an individual particle at a location \( \mathbf{r} \), where these forces are given by scalar quantities that are interpolated at a location \( \mathbf{r} \) by a weighted sum of contributions from all neighbouring particles inside a cut-off distance \( h \) in the space. This can be expressed in integral form as follows[33]:

\[
A_i = \int_{\Omega} A(\mathbf{\hat{r}}) W(r - \mathbf{\hat{r}}, h) d\mathbf{\hat{r}}
\]  

(4)

By approximating the integral interpolant by a summation interpolant, the numerical equivalent to Equation (4) is obtained:

\[
A_i = \sum_j A_j V_j W(r_{ij}, h)
\]  

(5)

where \( j \) is iterated over all particles, \( V_j \) the volume attributed implicitly to the particle \( j \), \( r_{ij} = r_i - r_j \), where \( r \) is the position of a particle, and finally \( A \) is the scalar quantity that is
being interpolated. The summation is over particles which lie within the radius of a circle centered at \( r_i \). The following relation between volume, mass and density applies\[33\].

\[
V = \frac{m}{\rho}
\]  

(6)

where \( m \) is the mass and \( \rho \) is the density. Combining this we get the basic formulation of the SPH interpolation function.

\[
A_i = \sum_j A_j \frac{m_j}{\rho_j} W(r_{ij}, h)
\]  

(7)

The function \( W(r_{ij}, h) \) is the smoothing kernel, which is a scalar weighted function. The function uses a position \( r \) and a smoothing length \( h \). This radius can be seen as a cut-off for how many particles will be considered in the interpolation. This cut-off radius sets \( W = 0 \) for \(|r_{ij}| > h\) \[33\].

In SPH, the derivatives of a function can be obtained by using the derivatives of the smoothing kernel that results in the Basic Gradient Approximation Formula \[172\]:

\[
\nabla A_i = \sum_j A_j \frac{m_j}{\rho_j} \nabla W(r_{ij}, h)
\]  

(8)

\[
\nabla^2 A_i = \sum_j A_j \frac{m_j}{\rho_j} \nabla^2 W(r_{ij}, h)
\]  

(9)

These formulations can produce spurious results, and several corrected formulations have been developed. One of these is the Difference Gradient Approximation Formula, which has the advantage that the force vanishes exactly when the pressure is constant.

\[
\nabla A_i = \frac{1}{\rho_i} \sum_j m_j (A_j - A_i) \nabla W(r_{ij}, h)
\]  

(10)

The forces between two particles must observe Newton’s Third Law that for every action there is an equal and opposite reaction. Pair-wise forces must be equal in size with opposite sign \( (f_i = -f_j) \). This means that the differentials in momentum equations that
create these forces must be symmetrized. Monaghan developed a symmetrisation, referred to as the Symmetric Gradient Approximation Formula [33]. It is commonly used for the pressure gradient.

\[ \nabla A_i = \rho_i \sum_j m_j \frac{A_i - A_j}{\rho_i^2 - \rho_j^2} \nabla W(r_{ij}, h) \]  

(11)

An alternative symmetrical formulation is as Equation (12) [34]:

\[ \nabla A_i = \sum_j \frac{m_j}{\rho_i} (A_i + A_j) \nabla W(r_{ij}, h) \]  

(12)

The basic formulation of the Laplacian has been found to be somewhat unstable under certain conditions, and there exist a wide range of possible corrections. Shao and Lo established a correction well suited for the correction of the Laplacian in the viscous force [180].

\[ \nabla \left( \frac{1}{\rho} \nabla A \right) = \sum m_j \frac{8}{(\rho_i + \rho_j)^2} \frac{(A_i - A_j) \nabla W(r_{ij}, h)}{|r_{ij}|^2 + \eta^2} \]  

(13)

To remain the denominator non-zero, \( \eta \) variable is used, which is equal to 0.1h.

2.5.2 Neighbour searching algorithm

In the SPH method, only a limited number of particles are inside the support domain of dimension \( kh \) for the target particle, which are applied in the particle approximations. These particles in the support domain are usually mentioned to as nearest neighbouring particles (NNP) for that target particle. The procedure of detecting the nearest particles is known as nearest neighbouring particle searching (NNPS). Unlike a grid based numerical method, where the position of neighbour grid-cells are well defined once the grids are given, the nearest neighbouring particles in the SPH for a given particle can vary with time. The different NNPS approaches which can be used in SPH implementations include all-pair search, linked-list search algorithm, and tree search algorithm [33].
2.5.3 SPH Algorithm

The SPH algorithm approximates a function and its spatial derivatives through averaging or summation over neighbouring particles. The special features in the SPH coding are generally involved under the main loop of time integration process, including the smoothing function and derivative calculation, particle interaction calculation, smoothing length evolution, spatial derivative estimation, boundary treatment, etc. A typical procedure for SPH simulation includes three main steps and some sub-steps, as below.

I. Initialization step: includes the input of the initial configuration of the problem geometry (dimensions and boundary conditions), discretization information of the initial geometry of particles, material properties, time step and other simulation control parameters.

II. Main SPH step: contains the major parts in the SPH simulation, and is implemented in the time integration loop. The following steps are required to be considered into the time integration process:

- Nearest neighbouring particle searching (NNPS)
- Calculating the smoothing function (for the summation density approach) and its derivatives from the generated information of interaction particle pairs
- Calculating the density
- Calculating the stress term (viscous force and pressure gradient), internal forces arising from the particle-particle interactions, and the body forces
- Updating particle momentum, density, particle position, velocity, and checking the momentum conservation

III. Output step: the time step reaches to a prescribed one or at some interval, the resultant information is saved for later analyses or post-processing.

2.6 Conclusion

Even with the recent developments being made in the field of immiscible-liquid microfluidics, an in-depth understanding of flow behaviour in micro-geometry is needed to
improve the design and optimization of the microfluidic devices. The studies have been done so far in this context, are still sporadic due to variations in the material used for fabrication, flow rates, channel dimensions and geometries. Experimental studies at such small scales are quite challenging and expensive, therefore numerical studies have to be called to become complementary to the experiments. On the other hand, the numerical studies involving microchannel flow simulations using mesh-based methods face a variety of obstacles. As discussed, the most important problem is the difficulties along with mesh generation which would be a problematic task, time consuming and an expensive process for cases with complex geometry. Another challenge in mesh-based simulations is to track the movement of the interfacial boundaries in multiphase fluid flows using explicit numerical simulation (ENS) method, which causes another source for modelling error and numerical instability. However, meshless particle methods such as SPH have been the focus recently to develop the next generation of more efficient computational schemes designed for more complex problems. However, there isn’t any report in the literature on the dynamics of immiscible-liquid microfluidics which have been studied based on SPH technique.

The space of parameters which can influence the flow pattern of two liquids interacting in immiscible-liquid microfluidics is very extensive and involves the nature of the liquids, the details of flow injections, geometry of the channels, surface characteristics and so forth. For instance, droplet formation and breakup processes depend on capillary number, superficial velocities of the continuous and dispersed phase, flowrate ratio, viscosity ratio, interfacial tension and geometry of microchannel. Despite various experimental and numerical studies have been reported on droplet-based microfluidics so far, the detailed flow physics and process parameters is still missing and much effort is still required to accurately control interfaces and flow patterns, make use of their interfacial properties and also to capture all the key parameters for design development of microfluidic devices.
Chapter 3: Modelling of Immiscible Liquid-Liquid Systems by Smoothed Particle Hydrodynamics

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Abstract

Immiscible liquid systems are ubiquitous in industry, medicine and nature. Understanding the relationship between the phase morphologies and fluid dynamics within the phases for the conditions they experience is critical to understanding in these many occurrences. In this paper, we detail a Smoothed Particle Hydrodynamics (SPH) model that facilitates building of this understanding. The model includes surface tension, enforces strict incompressibility, and allows for arbitrary fluid constitutive models. The validity of the model is demonstrated by applying it to a range of model problems with known solutions, including a liquid droplet falling under gravity through another quiescent liquid, Young-Laplace equation, and droplet deformation and rupture under shear whilst confined.

Keywords: Incompressible Smoothed Particle Hydrodynamics; Newtonian liquids; Immiscible liquid-liquid microfluidics; Droplet deformation; Drag coefficient
3.1 Introduction

Processing of immiscible liquid-liquid dispersions occurs widely in the manufacture of foods [210], pharmaceuticals [211], cosmetics [212], paints [213], and oil [214]. Examples of processes include emulsification [213, 215], encapsulation [216, 217], multiphase reaction systems [218, 219], electrochemical processes [220], bioprocesses [221], separation processes [222] such as liquid-liquid extraction [223], polymer blending [224], and oil recovery [225]. Processes involving immiscible liquids are also found beyond industry including in environmental clean-up [226], artificial oxygen carriers [227], oil spills [228], and pyroclastic flows [229].

There is an increasing need to understand and exploit the link between liquid-liquid phase morphology and fluid dynamics within the phases on the one hand and the conditions that lead to them on the other. For example, the droplet size distribution of an emulsion produced through agitation is a function of the balance between droplet breakup and coalescence, which can be controlled by the surfactant and stabilizer concentration and relative velocities of the phases [118-120]. Microfluidic production of encapsulates provides a further example: in this case, encapsulate morphology can be varied through the nature of the flow-focusing in the microchannels and, amongst other things, the continuous-to-dispersed phase flow rate ratio [121, 122]. The challenges faced in experimentally elucidating these types of relationships are significant, however. For example, the visualization of the morphologies of the phases and flow fields therein are still very much in their infancy [123-126]. Models that treat the phases and interfaces between them explicitly have, therefore, an important role to play in building understanding of liquid-liquid systems and exploiting this understanding in a systematic way.

Models of liquid-liquid systems in which the individual phases and interfaces between them are treated explicitly are long-standing [127-130]. The earliest models, which focus on droplets in a continuous phase, include those of Taylor [131, 230], Mason and co-workers [132, 133], Cox and co-workers [134, 135], and Acivos and co-workers [136-138] amongst others. Whilst these models were important in building understanding, they are limited by a good number of simplifying assumptions [139], including negligible
inertial effects (i.e. small Reynolds numbers), small viscosity ratio ranges, and regular droplet shapes (e.g. spheres; ellipsoids). Phenomenological approaches have been adopted to overcome some limitations associated with wholly analytical models. For example, Maffettone and Minale [87] used such an approach, determining the model parameters by ensuring it matches analytical results in appropriate limits. Such models are, however, also limited by underpinning assumptions such as, for example, specific droplet shapes and the absence of any connection between the model parameters and underlying fundamentals.

Adoption of wholly numerical approaches can, in principle, overcome limitations faced by analytical and related models. Some of the earliest numerical models include those of Acrivos and co-workers [139-141], who studied the deformation and rupture of a viscous droplet suspended in another liquid within extensional and shear flow fields. More recent examples include the works of Loewenberg, Hinch and Davis [142-144, 146-148, 231], which are based on a boundary-integral approach. Interface tracking techniques such as Volume of Fluid methods (VOF) [150, 232] and level set methods [152, 153] have been used even more recently. Whilst these works represent major advances in the field, they have two significant disadvantages that lead to algorithmic complexity, computational expense and numerical stability challenges [130, 165]: (1) the need to adjust the mesh as the phases deform; and (2) the need to track the interfaces between the different phases. The former can be eliminated by selecting a single set of mesh [233] with very high resolution enough to correctly capture the interface without adjusting the mesh as the phases deform but this again would be computationally very expensive. Additionally, the treatment of coalescence and rupture of the interfaces between the phases is extremely challenging if not impossible in many of these techniques [234, 235].

Particle-based approaches have more recently been used as a remedy to the issues faced by mesh-based approaches. The most widely-adopted is the lattice Boltzmann method (LBM) [32, 236]. This method as well as its less widely-used antecedent, lattice-gas automata (LGA) [162, 163], are also not without their problems and limitations, however. For example, Galilean invariance is lost in LGA, leading to a spurious density-dependent factor multiplying the inertial term in the momentum conservation equation whose effect cannot be removed [164]. LBM-based simulation of immiscible multiphase fluid flows, on the other hand, tend to suffer from stability problems [40]. An alternative approach is
Dissipative Particle Dynamics (DPD) [164-166], which has been used to study the rheology of complex fluids [167]. DPD is an off-lattice mesoscopic simulation technique which involves a set of particles moving in continuous space and each particle represents a cluster of atoms or molecules, and, as a result of the internal degrees of freedom associated with each particle, the particle-particle interactions include random and dissipative contributions[168]. However, the disadvantage of DPD is determination of liquid properties (e.g. viscosity) and transport coefficients of the system such as self-diffusion coefficient. Since the validity of the equilibrium properties and analytical expressions is restricted to the limit of small time steps, DPD is not computationally as efficient as LBM [169, 170]. A related but computationally far cheaper approach [209] of much longer standing is Smoothed Particle Hydrodynamics (SPH) [171]. In addition to avoiding all the issues associated with mesh-based methods, SPH has the advantages that it is derived directly from the Navier-Stokes equations and, hence, can be easily extended to any non-Newtonian matter.

In addition to being used to model multiphase fluid systems via the two-fluid model [177, 184], SPH has been used extensively to model such systems in which the interfaces are explicitly resolved. Many of these ignore surface tension (e.g. [185-190]), making them of limited value when interest lies in liquid-liquid systems where it plays an important role in dictating behaviour. Some have, however, sought to include surface tension effects. The first broad approach is that initially proposed by Morris [191], which embeds the continuum surface force (CSF) approach [192] within SPH. This approach is disadvantaged by the need to evaluate the curvature of interfaces, a numerically challenging task that also leads to numerical instabilities. Adams and co-workers [29] addressed this issue, although computational complexity and expense is still an issue. The CSF-based SPH approach has since been extended to systems involving solid surfaces of desired wettability [29, 193] and strictly incompressible systems [182, 183, 194, 195, 237-240]. A second broad approach sees SPH combined with the Cahn-Hilliard model [196, 197]; this diffuse interface approach is yet to see wide use, however. The final broad approach to including surface tension within SPH is via incorporation of inter-particle forces that lead to phase separation. The earliest example of this approach was due to Nugent and Posch [198], whose inter-particle force was inspired by the intermolecular
interaction term that arises in the van der Waals equation of state used to close their compressible SPH model. This particular model, which is far simpler and cheaper than the two alternative approaches mentioned above, has seen some use since \( \text{(e.g. [199, 241])} \) along with similar models based on other inter-particle force models, including some inspired by DPD \([170, 201]\) along with various other more arbitrary forms \([202]\), including trigonometric \([203]\) and inverse square law \([204]\). In all these cases, however, strict incompressibility has not been imposed; In this paper, we report incorporation of surface tension within strictly incompressible SPH framework by including a Lennard-Jones (LJ) interaction between particles \([242]\) in immiscibility model. The adoption of the LJ interaction to bring about surface tension has the advantage that it mirrors the almost universally used approach to incorporating solid boundaries within SPH, thus opening the way to unifying the treatment of interfaces in SPH. The paper first provides details of the SPH model and how it is parametrized to yield the desired surface tension. The new method is then validated by comparing its predictions to analytical results and experiments for a number of model problems, including a liquid droplet in a linear shear field and the same falling freely in a quiescent continuous phase.

3.2 The Method

3.2.1 Governing Equations

Smoothed-particle hydrodynamics builds on the Navier–Stokes equations expressed in the Lagrangian frame, which for an incompressible, isothermal system are of the form

\[
\nabla \cdot \mathbf{v} = 0
\]

(1)

\[
\rho \frac{d\mathbf{v}}{dt} = \nabla \cdot \mathbf{\sigma} + \rho \mathbf{g} + \mathbf{F}_i
\]

(2)

where \(\rho\), \(\mathbf{v}\), and \(\mathbf{\sigma}\) are the fluid density, velocity and stress tensor, respectively, and \(\mathbf{g}\) is the acceleration due to gravity or other body forces. The term \(\mathbf{F}_i\) is introduced into the momentum equation here so as to enable the imposition of immiscibility between the different liquids as described in more detail below. The form of this force is selected in a
way that its sum across the entire multiphase system disappears, ensuring overall conservation of momentum.

The stress tensor is described here by

$$\sigma = -P I + \tau$$  \hspace{1cm} (3)

where $P$ is the pressure, $I$ the unit tensor, and $\tau$ the shear stress tensor. The latter for a Newtonian fluid may be expressed as

$$\tau = -\mu \left[ \nabla v + (\nabla v)^T \right]$$  \hspace{1cm} (4)

where $\mu$ is the dynamic viscosity.

3.2.2 SPH Discretization

In SPH, the fluid is represented by a discrete set of particles of mass, $m_i$, and viscosity, $\mu_i$, that move with the local fluid velocity, $v_i$. Here, each particle also carries a colour, $c_i$, to indicate which of the immiscible fluids it belongs to. The velocity and other quantities associated with any particle are interpolated at a position, $r$, through a weighted summation of contributions from all neighbouring particles within a compact support of size $O(h)$, as illustrated in Figure 3-1.

Figure 3-1 An illustration of an SPH weighting function with compact support sited on particle-i (shown in red) that leads to the particle interacting with all other particles-j within the support of size $O(h)$.
For example, the density of a particle-\(i\) is given by [172]

\[
\rho_i = \sum_j m_j W(r_{ij}, h)
\]  

(5)

where \(r_{ij}\) is the distance between particles \(i\) and \(j\).

The pressure gradient associated with particle-\(i\) is given by [172, 207]

\[
\nabla P_i = \rho_i \sum_j m_j \left( \frac{P_j}{\rho_j^2} + \frac{P_i}{\rho_i^2} \right) \nabla_i W_{ij}
\]  

(6)

where \(P_i\) is the pressure associated with particle-\(i\).

Finally, the divergence of the shear stress tensor attached to a particle-\(i\) is given by [173]

\[
(\nabla \cdot \mathbf{\tau})_i = \rho_i \sum_j m_j \left( \frac{\tau_{ij}}{\rho_j^2} + \frac{\tau_{ji}}{\rho_i^2} \right) \cdot \nabla_i W_{ij}
\]  

(7)

The components of the shear stress tensor in this expression, which are derived from Equation (4), are given by

\[
\tau_{ij}^{\alpha\beta} = - \left( \sum_j m_j \mu_j \frac{v_{ij}^\beta}{\rho_j} \frac{\partial W_{ij}}{\partial x_i^\alpha} + \sum_j m_j \mu_j \frac{v_{ji}^\alpha}{\rho_j} \frac{\partial W_{ij}}{\partial x_i^\beta} \right)
\]  

(8)

where \(v_{ij} = v_i - v_j\)

We have employed the following quintic spline kernel to ensure the second derivatives of the smoothing kernel that arise in the viscous SPH model are smooth, thus avoiding instabilities that are known to arise with lower order kernels [33, 179]
\[ W(r^*, h) = \frac{7}{478\pi h^2} \times \begin{cases} (3 - r^*)^5 - 6(2 - r^*)^5 + 15(1 - r^*)^5 & 0 \leq r^* < 1 \\ (3 - r^*)^5 - 6(2 - r^*)^5 & 1 \leq r^* < 2 \\ (3 - r^*)^5 & 2 \leq r^* < 3 \\ 0 & r^* > 3 \end{cases} \]  

where \( r^* = r/h \) is a dimensionless radius.

### 3.2.3 Immiscibility Model

The immiscibility between the different fluids is enforced through the force, \( F_I \), in Equation (2), which is dependent on the colour of the particles. The force is based on the following functional form between particle pairs-\( ij \) that was arrived at through experimentation:

\[ \phi_{ij} = \begin{cases} \varepsilon_{ij} \left( \frac{L_0}{r} \right)^{12} & \text{if } c_i \neq c_j \\ -\varepsilon_{ij} \left( \frac{L_0}{r} \right)^6 & \text{if } c_i = c_j \end{cases} \]  

where, \( L_0 \) is a reference length, and \( \varepsilon_{ij} \) is a parameter that is linked to the relevant interfacial tension as outlined further below. The first part of this expression ultimately leads to a short range repulsive force of equal magnitude but opposite direction between pairs of different coloured SPH particles (i.e. different liquids), whilst the second part leads to a somewhat longer ranged attractive force of equal magnitude but opposite direction operating between pairs of particles of the same colour (i.e. same liquid). Although the first is clearly necessary to cause phase separation, the less intuitive second force was found to lead to improved behaviour from the model when included.

The force between particles is obtained from the gradient of the expression:

\[ (F_I)_{ij} = -\nabla \phi_{ij} \]  

Bringing together equations (10) and (11), the SPH formulation for the force acting on particle-\( i \) that brings about phase separation is
\[(F_i)_i = \sum_j \frac{m_j}{\rho_j} \phi_{ij} \nabla_i W_{ij} \quad (12)\]

### 3.2.4 Solution Algorithm

Combining equations (2), (6), (7), and (12) leads to the following SPH formulation for the momentum equation:

\[
d\frac{dv_i}{dt} = -\sum_j m_j \left( \frac{p_j}{\rho_j^2} + \frac{P_i}{\rho_i^2} \right) \nabla_i W_{ij} + \sum_j m_j \left( \frac{\tau_j}{\rho_j^2} + \frac{\tau_i}{\rho_i^2} \right) \cdot \nabla_i W_{ij} - \frac{1}{\rho_i} \sum_j \frac{m_j}{\rho_j} (F_i)_j \nabla_i W_{ij} \quad (13)\]

This is solved using a two-step predictor-corrector scheme based on that proposed by Cummins and Rudman [175] for single phase strictly incompressible flows and later extended to multiphase flows without surface tension by Shao and Lo [180]. Firstly, an initial estimate of the particle velocities, \(v_*\), is made using only the shear stress, gravitational and surface force terms in Equation (13) at time step \(t\) (indicated by the subscript \(-t\); particle indices have been dropped for convenience) [175, 180].

\[
v_* = v_t + \left( \frac{1}{\rho} \nabla \cdot \tau + g + \frac{1}{\rho} F_i \right) \Delta t \quad (14)\]

where \(\Delta t\) is the time step size. The corresponding initial estimates for the particle positions are then evaluated using [180]

\[
x_* = x_t + v_* \Delta t \quad (15)\]

Use of these initial particle position estimates in Equation (5) would reveal that the density \(\rho_*\), is not fixed as desired. Incompressibility is, thus, enforced by correcting the initial estimates of the particle velocities using [180]
\[ v_{t+1} = v_* + \Delta v_* \]  \hspace{1cm} (16)

where the velocity correction is evaluated using the pressure gradient term of the momentum equation only

\[ \Delta v_* = -\frac{1}{\rho_*} \nabla P_{t+1} \Delta t \]  \hspace{1cm} (17)

To obtain the pressure at the new time, \( P_{t+1} \), Equation (16) and (17) are combined and the divergence is taken to give [175]

\[ \nabla \cdot \left( \frac{v_{t+1} - v_*}{\Delta t} \right) = -\nabla \cdot \left( \frac{1}{\rho_*} \nabla P_{t+1} \right) \]  \hspace{1cm} (18)

Imposing the incompressibility condition at the new time step, \( \nabla \cdot v_{t+1} = 0 \), leads to the Pressure Poisson Equation (PPE) [175]

\[ \nabla \cdot \left( \frac{1}{\rho_*} \nabla P_{t+1} \right) = \frac{\nabla \cdot v_*}{\Delta t} \]  \hspace{1cm} (19)

The left hand side is discretized based on Shao’s approximation for the Laplacian in SPH [180], which is a hybrid of a standard SPH first derivative with a finite difference computation [175]

\[ \nabla \cdot \left( \frac{1}{\rho} \nabla P \right)_i = \sum_j m_j \frac{8}{(\rho_i + \rho_j)^2} \frac{(P_i - P_j \cdot \mathbf{r}_{ij} \cdot \nabla_i W_{ij}}{||\mathbf{r}_{ij}||^2 + \omega^2} \]  \hspace{1cm} (20)

where \( \omega \) is a small value (e.g. 0.1\( h \)) to ensure the denominator is always non-zero.

The right hand side of equation (19) is discretised in SPH using

\[ (\nabla \cdot v_*)_i = \rho_i \sum_j m_j \left( \frac{v_{*,j}}{\rho_j^2} + \frac{v_{*,i}}{\rho_i^2} \right) \cdot \nabla_i W_{ij} \]  \hspace{1cm} (21)
Discretization of the PPE equation leads to a system of linear equations, $Ay = b$, in which $y$ is the vector of unknown pressure gradients to be determined, and the matrix $A$ is not necessarily positive definite or symmetric. In the present work, the biconjugate gradient algorithm [243] was used to solve this set of equations.

Once the velocity at the next time step is determined via use of Equation (16), the new particle positions are finally obtained using

$$x_{t+1} = x_t + \frac{1}{2}(v_t + v_{t+1})\Delta t$$  \hspace{1cm} (22)

3.3 Results and Discussion

3.3.1 Young-Laplace and Parameterization of Immiscibility Model

The validity of the new method was first assessed by ensuring that a circular droplet of one liquid is recovered when sitting in a second, quiescent immiscible liquid of the same density ($\eta = \rho_d/\rho_c = 1$) and viscosity ($\lambda = \mu_d/\mu_c = 1$), in line with physics. This was done by observing the change in the shape of a stationary droplet over time using a time step size of $\Delta t^* = \Delta t\sqrt{\gamma/\rho h^3} = 0.1$ when the drop is initially a square of size $6h \times 6h$ ($h = 1.25L_0$, where $L_0 = 5 \times 10^{-5}m$) within a periodic domain of $24h \times 24h$; the total number of SPH particles was $N_p = 961$. As Figure 3-2 shows for a typical simulation, as expected, the perimeter of the droplet gradually decreases as it changes from its initial square shape to a circle. This figure also shows that the pressure difference across the interface between the two phases, shown in dimensionless form ($\Delta P^* = \Delta P h/\gamma$), increases as the droplet transforms in shape, once again in line with the physics where the pressure difference is counter-balanced by the interfacial tension (in this case, $\gamma = 0.045$ N/m, which corresponds to $\varepsilon = 56.25$ N/m$^2$).
Figure 3-2 Typical variation through time, $t^* = t\sqrt{\gamma/\rho h^3}$, of the circumference of an initially square neutrally buoyant droplet suspended in a second continuous phase and the associated change in pressure difference, $\Delta P^* = \Delta Ph/\gamma$, across the interface between the two liquids. Snapshots of the droplet along the transformation pathway are shown at various points; the continuous phase is not shown for simplicity.

Figure 3-3 shows the variation of the pressure drop across the interface between the drop and continuous phase as a function of the inverse of the drop size. The linear behaviour seen in this figure clearly conforms to the Young-Laplace equation

$$\Delta P = \frac{\gamma}{R}$$  \hspace{1cm} (23)

The relationship between the interaction model parameter in equation (10), $\varepsilon$, and the interfacial tension, $\gamma$, can be determined by repeating the simulations that lead to Figure 3-3 for various values of the former. Doing this leads to Figure 3-4, this shows that the interaction model parameter varies in a linear fashion with the interfacial tension.
3.3.2 Deformation of a Confined Droplet in a Linear Shear Field

The new method was further validated by considering through time the deformation under linear shear of an initially circular droplet in a second immiscible liquid of the same density ($\eta = \rho_d/\rho_c = 1$) and viscosity ($\lambda = \mu_d/\mu_c = 1$), Figure 3-5. The initial droplet of diameter $D = 17h$ ($h = 1.2L_0$, where $L_0 = 1 \times 10^{-5}$ m) was located centrally within a domain of height $H = 34h$ and length $L = 100h$; the total number of SPH particles was $N_p = 4961$. To compensate truncation of the support domain of the kernel at the wall
boundary, at initial state 3h layers of dummy particles are regularly arranged at the other side of the wall boundary [110, 111]. Equal but opposite velocities of magnitude $U$ were applied to the top and bottom of the domain with no slip boundary conditions (i.e. the top and bottom were treated as moving solid surfaces) to yield a linear shear field with shear rate $G = 2U/H$. The state of the system was evolved through time using timesteps of size $\Delta t^* = G \Delta t = 0.001$.

Under suitable conditions, the droplet can deform to take on an ellipsoidal shape as illustrated in Figure 3-5. The character of this droplet may be described in part by the deformation parameter [244]

$$D_f = \frac{a - b}{a + b}$$ (24)

where $a$ and $b$ are the lengths of the major and minor axes of the ellipsoid, respectively. The other key parameter characterizing the droplet behaviour is the angle the major axis of the ellipsoid subtends to the streamlines of the shear field, $\varphi$.

---

**Figure 3-5** Droplet shape and velocity field obtained from a typical SPH simulation with the key characteristics shown.

Figures 3-6 illustrates the deformation and breakup of a droplet under a confined shear flow through dimensionless times, $t^* = G t$, at $\text{Re} = \rho R^2 G / \mu = 0.1$, $\lambda = 1$, $D/H = 0.5$ and $\text{Ca} = \mu R G / \gamma = 0.5$, a capillary number greater than the critical value where droplet breakup is ensured. In comparison, Figure 3-7 indicates the velocity field and shape change of the same droplet for a Ca value smaller than the critical value.
Figure 3-6 Variation of velocity field and droplet morphology with time for $Ca = 0.5$, $Re = 0.1$, $\lambda = 1$, and $D/H = 0.5$. 

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Figure 3-7 Variation of velocity field and droplet morphology with time for $Ca = 0.2$, $Re = 0.1$, $\lambda = 1$, and $D/H = 0.5$. 
The velocity fields show changes in x-velocity component along the height of the domain. This induces a velocity field inside the droplet which reduces the shear stress on the droplet. As a result, the droplet starts elongation and a rotational motion so that a circulating flow is set up inside the droplet. At higher Ca, viscous forces are more important than interfacial forces and therefore, the droplet undergoes a larger deformation and experiences a breakup, as indicated by figure 3-6.

Figure 3-8 compares the deformation parameter as obtained from the SPH simulations at a confinement ratio of $D/H = 0.5$ with the experimental results of Sibillo et al. [245] and Taylor’s model for an unconfined droplet in a shear field [244]

$$D_f = \frac{19\lambda + 16}{16\lambda + 16} Ca$$  \hspace{1cm} (25)

This figure shows that the deformations predicted by SPH match very well the experimental results up to the $Ca = 0.3$ limit probed by Sibillo et al. [245]. The results also match well those predicted by Taylor’s model up to this Capillary Number. However, beyond this one can see a significant deviation from Taylor’s model, indicating that confinement has an effect at higher Capillary Numbers where shear is starting to dominate over surface tension.

Figure 3-8 Variation of the deformation parameter as defined in Equation (24) with Capillary Number as predicted by SPH with Np=7701 (solid circles with linear fit shown as a line; $R^2=0.99$), SPH with Np=4961 (open circles) Taylor theory as defined by Equation (25) (solid line) [244], and experimental data (open squares) [245].
Figure 3-9 compares the tilt angle of the ellipsoid (see Figure 3-5) as obtained from the SPH simulations at a confinement ratio of $D/H = 0.5$ with experimental data and the value predicted from the Taylor model [244]

$$\varphi = \frac{\pi}{4} - \frac{(19\lambda + 16)(2\lambda + 3)}{80(\lambda + 1)} Ca$$

Once again, comparison with experiment is very good. The deviation of the SPH values from Taylor’s theory is modest but grows with Ca, indicating that the tilt angle is more sensitive to confinement.

Figure 3-9 Variation of the droplet tilt angle with Capillary Number as predicted by SPH with $N_p=4961$ (open circles), SPH with $N_p=7701$ (solid circles), Taylor theory as defined by Equation (25) (solid line) [244], and the experimental values of Sibillo [246] (open squares).

3.3.3 Deformation of Free-Falling Droplet

The new method was finally validated by considering the descent of an initially stationary circular droplet under gravity through a second stationary liquid whose density, $\rho_d = 1150 \, kg/m^3$ differed from that of the continuous phase, $\rho_c$, by $\Delta \rho = \rho_d - \rho_c = 150 \, kg/m^3$. The droplet was initially of diameter $D = 14h$ ($h = 1.2L_0$, where $L_0 = 1 \times 10^{-4} m$), and was located within a periodic domain of size $34h \times 34h$; the total number of SPH particles was $N_p = 1681$. The droplet motion and deformation was followed through time ($\Delta t^* = \Delta t\sqrt{g/D} = 0.004$) until the terminal velocity was reached and the droplet
shape remained unchanged \((t^* = 45)\). A range of combinations of Morton Number, \( \text{Mo} = g\Delta \rho \mu_c^4 / \rho_c^2 \gamma^3 \), Ohnesorge Number, \( \text{Oh}_d = \mu_d / \sqrt{\rho_d \gamma D} \), and \( \text{Oh}_c = \mu_c / \sqrt{\rho_c \gamma D} \) and Eötvös Number, \( \text{Eo} = g \Delta \rho D^2 / \gamma \) were considered.

Figure 3-10 compares the variation with time of the droplet descent speed, \( u_d^* = u_d / \sqrt{gD} \), obtained here from SPH with that predicted by Han and Tryggvason [247] using a finite difference front tracking method. This figure shows that the two predictions are qualitatively very similar. Whilst there are some quantitative differences between the two predictions, these differences are small about 1.36%. Snapshots of the deforming droplet at various times indicate how the deformation affects the droplet descent speed. After the initial rise the droplet descent slows down as it starts deforming, until reaches to a steady state condition.

![Figure 3-10 Variation of the droplet descent speed with time as predicted by SPH (dash line) and Han and Tryggvason [247] (solid line) for \( \eta = 1.15, \lambda = 1, \text{Eo} = 10, \text{Oh}_d = 0.24 \) and \( \text{Mo} = 0.04 \). Snapshots of the deforming droplet are shown at various times](image)

Figure 3-11 shows the evolution of the droplet descent speed with time for different Eötvös Numbers at \( \eta = 1.15, \text{Oh}_d = 0.23, \text{Oh}_o = 1.25 \). Although the falling velocity ends to a steady state, the simulation results reveal some dissimilarity in the droplet velocity trends for the different \( \text{Eo} \). According to the figure at the fixed Ohnesorge numbers, the smaller \( \text{Eo} \) reaches to the steady state faster, while for the larger \( \text{Eo} \), the droplet experiences an
overshoot in the descent speed that implies larger deformation for the falling droplet. Figures 3-11 (b) and (c) show the droplet shape changes at the different Eötvös Numbers.

Figure 3-11 (a) variation of droplet descent speed with time as predicted by SPH for $\eta = 1.15$, $Oh_d = 0.23$, $Oh_o = 1.25$, $Eo = 24$ (solid line), and $Eo = 144$ (dash line). Droplet deformation for (a) $Oh_d = 0.23$, $Oh_o = 1.25$, $Eo = 24$, and (c) $Oh_d = 0.23$, $Oh_o = 1.25$, $Eo = 144$.

Figure 3-12 illustrates the effect of $Oh_c$ and the corresponding Morton numbers on the droplet shape and descent speed at the fixed $Eo = 24$, and $Oh_d = 1.16$. According to the figure, the shape change occurs from spherical to oblate and indented oblate as $Oh_c$ and $Mo$ decrease, implying the less viscous droplet experiences the more deformation. These results are in agreement with the shape regime maps developed by Han and Tryggvason.

Figure 3-12 (a) variation of droplet descent speed with time as predicted by SPH for $\eta = 1.15$, $Oh_d = 1.16$, $Eo = 24$, $Oh_c = 0.05$ (solid line), and $Oh_c = 1.25$ (dash line). Droplet deformation for $Oh_d = 1.16$, $Eo = 24$, (b) $Oh_c = 0.05$ and $Mo = 0.00015$, and (c) $Oh_c = 1.25$ and $Mo = 58$. 

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3.4 Conclusions

In this work, a SPH-based model was developed to study immiscible liquid-liquid systems. Strict incompressibility was enforced through solving the Pressure Poisson Equation. A surface tension model was proposed to enforce immiscibility between the different fluids. The model was validated against different model problems including Young-Laplace equation, droplet deformation and breakup under confined shear field, and a liquid droplet falling under gravity through another quiescent liquid. The results suggest that the model is capable to predict phase morphologies and fluid dynamics of immiscible liquid-liquid systems.
Chapter 4: Smoothed Particle Hydrodynamics-based Simulations of Immiscible Liquid-Liquid Microfluidics in a Hydrodynamic Focusing Configuration

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Abstract

Smoothed Particle Hydrodynamics (SPH) was applied to the study of hydrodynamic focusing of a liquid flowing in a deep microchannel by a second, immiscible less-viscous liquid introduced at right angles through identical but symmetrically opposed microchannels. To investigate the flow dynamics and topological changes of the two phases, the phases and the interfaces between them were explicitly modelled. The fluids were truly incompressible, viscous effects were treated rigorously, and surface tension was included. The model is able to reproduce the experimental results of Cubaud and Mason (Phys. Fluids. 20, 053302, 2008), including the flow morphology mapping with the Capillary Numbers of the two liquids, and the characteristics dimensions of the focused liquid phase as a function of the liquid flow rate ratio. Going beyond the experimental results, the model was used to build understanding of the velocity fields.

Keywords: Microfluidics; Hydrodynamic focusing; Smoothed Particle Hydrodynamics; Immiscible liquids; Surface tension.
4.1 Introduction

The potential for microfluidics to revolutionize the energy, chemical processing and medical fields has attracted significant interest and research effort over the last few decades [1, 4, 81, 248-253]. Whilst many microfluidic systems involve single phase fluids only (e.g. lab-on-a-chip analysis of biological fluids [254, 255]), microfluidic processing of multiphase fluids also offers a wealth of opportunities. For example, adding a second immiscible fluid phase to single-phase microfluidics facilitates high specific interfacial area, mixing and mass transfer, leading to increased reaction yields for mass transfer-limited reactions [81, 248, 256, 257]. Multiphase microfluidics also provides possibilities to precisely control fluid-fluid interfaces and flow patterns for a wide range of applications ranging from production of novel materials [81, 258, 259] through to medicine [51, 260, 261]. The production of monodispersed droplets of controlled size in micro-emulsions for pharmaceutical applications [104, 262] is just one example.

In the absence of electric or magnetic fields, the morphology of the phases in isothermal two-fluid microfluidic flows is dependent on the balance between the two dominant forces at play – viscous and surface tension – which can be quantified by the Capillary Number, the relative flow rates of the two phases, the ratio of their viscosities, and the geometry of the microfluidic pathways. For example, when one liquid (L_1) is ‘focused’ by a second immiscible liquid (L_2) introduced through two side channels as illustrated in Figure 1(a), the phase morphology can take on one of five characters depending on the Capillary Numbers of the two streams, $Ca_i$, as indicated in Figure 1(b) [14]: (a) threading in which L_1 is focused into a thin thread by L_2; (b) jetting where the thread breaks up due to instabilities to produce droplets of L_1 in L_2 down-stream of the flow focusing junction; (c) dripping in which the droplets of L_1 are formed at constant frequency at the junction; (d) tubing in which L_1 occupies much of the outlet cross-section; and (e) viscous displacement where fingers of L_1 penetrate into the side channels. The thickness of the thread in the threading regime, $\varepsilon$, is related to the ratio of the flow rates [14]

$$\frac{\varepsilon}{H} \approx \left(\frac{Q_1}{2Q_2}\right)^{1/2}$$

(1)
In the dripping regime, the droplet length, $l_d$, is given by

$$\frac{l_d}{H} \approx 0.5 \left( \frac{Q_2}{Q_1 + Q_2} C a_2 \right)^{-0.17}$$

(2)

In the jetting regime, the diameter of the droplets, $d_j$, and the thread length, $l_j$ are given by

$$\frac{d_j}{H} \approx 3.1 \left( \frac{Q_1}{2 Q_2} \right)^{1/2}$$

(3)

$$\frac{l_j}{H} \approx 8 C_j \mu_1 \left( \frac{Q_1 Q_2}{\pi H^2 C a_c \gamma} \right)^{1/2}$$

(4)

where $C_j$ is a constant close to unity and $C a_c$ is the critical capillary number which was found to be approximately 0.1 [14]. A range of other configurations beyond that of Figure 4-1 have also been studied over the last two decades, including T-junctions [15, 35, 105], Y-junctions [18], and concentric injection [110] to reveal a rich phase morphology behaviour that is dependent on the nature of the flow focusing junction. For example, a cross-junction like that shown in Figure 4-1 allows formation of symmetrical droplets whilst only asymmetrical drops are possible with T-junctions [263].
Figure 4-1 (a) schematic of cross-junction flow focusing geometry considered in detail in [14] and here, where $Q_i$, $Ca_i$, $\mu_i$ and $U_i = Q_i/H^2$ are the flow rate, Capillary Number, viscosity and superficial velocity of liquid $Li$, respectively, and $\gamma_{12}$ is the interfacial tension for the liquid pair; (b) phase morphology map based on the Capillary Numbers of the liquid $L1$ ($Ca_1$) and the focusing liquid $L2$ ($Ca_2$): (a) threading; (b) jetting; (c) dripping; (d) tubing; and (e) viscous displacement. The numbers on the map refer to the conditions that were considered in the study reported on here.

Though there has been extensive study of the effect of fluid properties and flow geometries on phase morphology, there are still many gaps in understanding due to the sporadic sampling of the range of materials, fluids, flow rates, channel dimensions and geometries [264]. Additionally, understanding of the flow fields in microfluidic devices is even less well explored because of the challenges faced in measuring these at such small scales. Numerical studies can assist in filling this gap. For instance, De Menech et al [35] studied transition regimes in a microfluidic T-junction using a phase field model to show the importance of viscosity ratio for the droplet formation as well as the effect of confinement.
on the shape of droplet and slugs when the size of the side-channel is less than size of the continuous phase channel. Similarly, the different flow patterns obtained through droplet formation process in a T-junction microchannel have been numerically investigated by the same group [250]. Liu and Zhang [265] studied the effects of flowrates, viscosity and wettability on the process of droplet formation at a microfluidic junction using an improved phase field lattice Boltzmann method. Also, Gupta and Kumar [161] applied lattice Boltzmann method (LBM) to show how the dimensions of the channels in a microfluidic T-junction influence the droplet size. Recently, Ngo et al. [266] investigated numerically the droplet formation process in a double T-shaped microchannels using a Volume of Fluid (VOF) model. Liu et al. [267] employed Dissipative Particle Dynamics method to study multiphase flow behaviour in an inverted Y-shaped microchannel and a microchannel network. Wu et al. [268] developed a multiphase LBM for droplet formation in a microfluidic cross-junction flow geometry and demonstrated the dependence of dripping flow pattern on both Capillary and Weber Numbers. Likewise, Liu et al. [20] reported three different flow regimes for varied flowrate ratios at a fixed Capillary number using LBM. Their study was, however, limited to small Capillary Numbers (Ca<0.01) and viscosity ratios (\(\lambda\leq1/50\)). Shi et al. [269] performed a comparative study on the dynamics of the droplet formation in a microfluidic T-junction and a flow-focusing configuration using LBM. They indicated that a spherical droplet can be formed through a flow-focusing geometry, whereas T-junction is likely to produce an elliptical droplet.

Numerical modelling of immiscible liquid-liquid microfluidics requires the explicit modelling of the two phases and the interfaces between them. This is necessary for two reasons at least. Firstly, as the characteristic dimensions of the individual phases are typically comparable to that of the confining geometry, homogenisation approaches are inappropriate. The second reason is the fact that interest often lays in understanding the detailed morphology of the phases, something that is not accessible in more mean field models such as the two-fluid model. Although explicit modelling of the individual phases and the interfaces between them through space and time is necessary, it is particularly challenging. If mesh-based numerical methods are used, for example, the evolution of the mesh in space and time requires complex and expensive algorithms that can bring error propagation and numerical instabilities [130, 165]. These methods are also often unable to
handle the coalescence or break-up of phases (e.g. break-up of the thread in jetting to form the droplets) [241, 270]. Meshless methods such as Smoothed Particle Hydrodynamics (SPH) [27, 171] and the more expensive [209] DPD [271, 272] avoid these issues, on the other hand.

In the work reported in this paper, an incompressible SPH-based model [273] is shown to be able to reproduce the experimental results obtained by Cubaud and Mason [14] for the flow-focusing geometry shown in Figure 4-1(a). The emphasis of the study was on the effect of the flowrate ratio and Capillary Numbers. The paper first presents the details of the SPH model in brief along with the details of the study. Results are then presented for the range of conditions shown on the Capillary Number map in Figure 4-1(b) and compared with the experimental data of Cubaud and Mason [14], thereby demonstrating that the SPH method is suitable for building detailed understanding of the influence of fluid properties and process conditions on the phase morphology. The flow fields are also explored, adding understanding to the experimental results.

4.2 Model and Study Details

4.2.1 Model

A microfluidic right angled cross-junction consisting of three inlet microchannels and one outlet microchannel as shown in Figure 4-1(a) was considered; the depth of the microchannels was assumed to be much greater than their width, \( H \), and study was restricted to a plane away from the top and bottom of the channels (i.e. the simulations were limited to two spatial dimensions). A Newtonian liquid \( L_1 \) of viscosity \( \mu_1 \) entered the main inlet channel at a volumetric flow rate \( Q_1 = U_1 H^2 \). Likewise, a second Newtonian liquid \( L_2 \) of viscosity \( \mu_2 < \mu_1 \) was injected symmetrically into the two lateral inlet channels each at a volumetric flow rate \( (Q_2/2) = U_2 H^2 \). This configuration leads to the second liquid hydrodynamically focusing the first as outlined in the Introduction to the report here.

The SPH-based model proposed in our earlier work [273] for simulation of immiscible liquid-liquid flows was applied here to study the flow configuration outlined immediately above; full details of the SPH model can be found in Elekaei et al. [273]. A total of 6562
SPH particles were initially arranged uniformly within the microchannels with a spacing of $L_0 = 1 \times 10^{-5}$ m and the fluid particles were assigned an initial speed of $U_i = Q_i/H^2$. The particles located in the main microchannel were initially designated as liquid $L_1$, and those in the two transverse channels as liquid $L_2$. No-slip boundary conditions and confinement of the SPH particles within the microchannel walls was implemented as described by [273]. Dirichlet velocity boundary conditions were applied at the inlets and outlet of the microchannels using a similar approach without, of course, the particle confinement. Whenever a particle of a liquid leaves the outlet, it is fed back into the simulation through the appropriate inlet, being sure to maintain symmetry of the lateral flows, thus conserving the number of particles (and mass) in the simulations. The timestep size used was $1 \times 10^{-7}$ s, and the kernel size was $1.4 L_0$.

4.2.2 Study Details

The study reported here focused on the two immiscible Newtonian liquids summarized in Table 4-1. The channel cross sectional size was also set to $H = 100$ μm for all simulations. The 16 Capillary Number pairs $(Ca_1,Ca_2)$ shown in Figure 4-1(b) were considered by varying independently the superficial velocity of the two liquids, $U_i$. These conditions were selected to widely probe the capacity of the model to reproduce the map determined experimentally by Cubaud and Mason [14], including the boundaries between the different phase morphologies.

<table>
<thead>
<tr>
<th>Property/Fluid</th>
<th>Glycerol ($L_1$)</th>
<th>PDMS Oil ($L_2$)</th>
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<tr>
<td>Viscosity ($\mu_i$; Pa·s)</td>
<td>1.214</td>
<td>0.00459</td>
</tr>
<tr>
<td>Surface tension ($\gamma_{12}$; N/m)</td>
<td>0.027</td>
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4.3 Results and Discussion

Figure 4-2 shows the simulation result for the Capillary Number doublet of (0.5, 0.5), which corresponds to the point 1 in Figure 4-1(b). It is clear that the model is able to reproduce the threading flow expected at this point. As shown in Figure 4-10, the same can be said for the three other simulations undertaken in this part of the phase morphology
map. More quantitatively, as Figure 4-3 shows, the SPH model is able to predict very well the variation of the thread thickness, $\varepsilon$, obtained experimentally, which is defined by equation (1); the relative error between the correlation and the model is on average 8%, which is anticipated the deviations are smaller than the experimental uncertainties. The last point in this figure at $Q_1/Q_2 = 0.24$ shows the upper limit for $Q_1/Q_2$ in the aforementioned conditions for which a threading flow regime is valid.

Figure 4-2 also shows the velocity field at steady state, something that is difficult to measure experimentally. As a result of a flowrate ratio of $Q_1/Q_2 = 0.1$, the two branches of the continuous phase suppress the dispersed phase at the cross-junction and makes a thinning process which leads to the formation of a long thread of $L_1$.

![Figure 4-2](image)

*Figure 4-2 Representative outcome for threading regime at $Ca_1 = 0.5$ and $Ca_2 = 0.02$, which corresponds to point 1 on Figure 4-1(b).*
Figure 4-3 Dimensionless thickness of threads as a function of flowrate ratio as evaluated using the SPH model (open circles) and equation (1) (solid line).

Figure 4-4 shows the SPH prediction at a Capillary Number doublet of (0.1, 0.003), which corresponds to point 2 on Figure 4-1(b). Once again, it is clear that the model is able to reproduce the dripping flow expected at this point. As shown in Figure 4-11, the same can be said for the three other simulations undertaken in this part of the phase morphology map. Figure 4-5 shows, the SPH model is able to predict very well the variation of the droplet length, $l_d$, obtained experimentally, which is defined by equation (2); the relative error between the correlation and the model is on average less than 8%, which is anticipated that the error is smaller than the experimental uncertainties.

Figure 4-4 also indicates the evolution of the droplet formation process. At higher flowrate ratios that allows a larger momentum for the dispersed phase, $L_1$ is broken into droplets. According to the figure, initially the velocity of the dispersed phase reaches to its maximum where occupies the width of the main channel until collapses under the effect of the continuous phase momentum that acts against interfacial tension, and leads to a thinning pinch-off process. The built-up pressure in the side channels makes acceleration and drives $L_1$ to squeeze until reaches to a constant superficial velocity of $U_1+U_2$. At the end of pinching process, breakup occurs and interfaces retract as a result of surface tension.
\[ x (\mu m) \]

\[ y (\mu m) \]

(a)

(b)

(c)
Figure 4-4 Representative outcome of the SPH model for dynamics of the droplet formation in dripping regime at $C_{a1} = 0.1$ and $C_{a2} = 0.003$, which corresponds to point 2 on Figure 4-1(b). The snapshots belong to (a) $t^{*} = 0.07$, (b) $t^{*} = 0.2$, (c) $t^{*} = 0.38$, and (d) $t^{*} = 0.4$, where $t^{*} = U_1 t/H$.

Figure 4-5 Dimensionless length of droplet as estimated by the SPH model (open circles) and equation (2) (solid line).

Figure 4-6 shows the SPH prediction at a Capillary Number doublet of $(0.3, 0.005)$, which corresponds to point 3 on Figure 1(b). As with the two other cases considered thus far, it is clear that the SPH model is able to reproduce the jetting flow expected at this point. As shown in Figure S3, the same can be said for the three other simulations undertaken in this part of the phase morphology map. Figure 4-7(a) shows, the SPH model is able to predict very well the variation of the droplet length, $d_j$, obtained experimentally, which is defined by equation (3); the relative error between the correlation and the model is on average 6%.
which is anticipated that the deviation is smaller than the experimental uncertainties. Similar to the dripping regime, a droplet formation and pinching process occurs in the jetting regime but away from the junction after forming a very thin thread of the liquid in the main channel, $L_1$. Similarly, Figure 4-7(b) indicates the SPH results for variation of the thread length of the jet, $l_j$, as defined by equation (4); the relative error between the correlation and the model is on average 10%.

![Figure 4-6 Representative outcome of the SPH model for dynamics of the jetting regime at $Ca_1 = 0.3$ and $Ca_2 = 0.005$, which corresponds to point 3 on Figure 4-1(b)). The snapshots belong to (a) $t' = 0.42$, and (b) $t' = 0.44$, where $t' = U_1t/H$]
Figure 4-7 Dimensionless (a) droplet diameter (equation (3)), and (b) thread length (equation (4)) in dripping regime as evaluated by the SPH model (open circles) and the related correlations (solid line).

Figure 4-8 shows the SPH prediction at a Capillary Number doublet of \((1.0, 0.006)\), which corresponds to point 4 on Figure 4-1(b). As with the all the other cases considered thus far, it is clear that the SPH model is able to reproduce the expected flow pattern (i.e. tubing).

The controlling agent in tubing regime is the viscous stress by which a long viscous core occupies the most cross section of the main channel as can be observed by the figure.

Figure 4-8 Representative outcome of the SPH model for the tubing regime at \(Ca_1 = 1.0\) and \(Ca_2 = 0.006\), which corresponds to point 4 on Figure 4-1(b).
And, finally, Figure 4-9 shows the SPH prediction at a Capillary Number doublet of (0.19, 0.0002), which corresponds to point 16 on Figure 4-1(b). Viscous displacement regime, also known as fingering, happens at a large q, when the low momentum of L₂ is not enough to drive L₁, and so L₁ starts to occupy the side channels.

Figure 4-9 Representative outcome of the SPH model for the tubing regime at $Ca₁ = 0.19$ and $Ca₂ = 0.0002$, which corresponds to point 16 on Figure 4-1(b).

### 4.4 Conclusion

An incompressible Smoothed Particle Hydrodynamics-based model was applied to study immiscible-liquid flows through microfluidic hydrodynamic focusing configuration for a wide range of capillary numbers ($0.001 < Ca \leq 1.0$). For a Newtonian liquid pair with a large viscosity ratio, the different flow patterns known as threading, dripping, jetting, and tubing regime were achieved through our simulations under the effects of the capillary number of each liquid phase, as expected by the experimental results of Cubaud and Mason (Phys. Fluids. 20, 053302, 2008). The characteristics dimensions of the liquid phase in the main channel such as the thickness of the thread in the threading regime, the droplet length in the dripping regime, and the droplet diameter in the jetting regime were validated by the experimental results. Moreover, dynamics of the focused liquid flow and the droplet formation evolution were discussed for different flow patterns.
4.5 Supplementary Information

Figure 4-10 Representative outcome for threading regime at (a) $Ca_1 = 0.5$ and $Ca_2 = 0.008$, which corresponds to point 12 on Figure 4-1(b), (a) $Ca_1 = 0.4$ and $Ca_2 = 0.01$, which corresponds to point 11 on Figure 4-1(b), and (a) $Ca_1 = 0.3$ and $Ca_2 = 0.025$, which corresponds to point 10 on Figure 4-1(b).
Figure 4-11 Representative outcome for dripping regime at (a) $\text{Ca}_1 = 0.085$ and $\text{Ca}_2 = 0.002$ which corresponds to point 15 on Figure 4-1(b), (a) $\text{Ca}_1 = 0.1$ and $\text{Ca}_2 = 0.001$, which corresponds to point 8 on Figure 4-1(b), and (a) $\text{Ca}_1 = 0.05$ and $\text{Ca}_2 = 0.0006$ which corresponds to point 7 on Figure 4-1(b).
Figure 4-12 Representative outcome for jetting regime at (a) $Ca_1 = 0.19$ and $Ca_2 = 0.005$ which corresponds to point 15 on Figure 1(b), (a) $Ca_1 = 0.17$ and $Ca_2 = 0.008$, which corresponds to point 14 on Figure 1(b), and (a) $Ca_1 = 0.2$ and $Ca_2 = 0.008$ which corresponds to point 13 on Figure 1(b).
Chapter 5: **Effects of Flow Parameters and Geometrical Characteristics on the Dynamics of Two Immiscible Liquids in a Hydrodynamic Focusing Microgeometry**

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**Statement of Authorship**

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**Author Contributions**

By signing the Statement of Authorship, each author certifies that their stated contribution to the publication is accurate and that permission is granted for the publication to be included in the candidate’s thesis.

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<td>Contribution</td>
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Abstract

Recently we reported a numerical study on immiscible-liquid flows in hydrodynamic focusing microchannels using Smoothed Particle Hydrodynamics (SPH) technique. The optimal design of the system, however, needs a detailed understanding of the dynamics of flows and the capability to predict such behaviours. To respond to this need, we utilized our SPH-based simulation to predict the behaviour of the multiphase flow system for a wider range of flow parameters including the flowrate ratio, viscosity ratio and capillary number of each liquid phase. The flowrate ratio of the continuous phase to the dispersed phase affects the flow behaviour and causes different flow patterns depending on $Ca_1$ and $Ca_2$. The droplet length also appears to be influenced by the both flowrate quantity and flowrate ratio in dripping regimes. In addition, we discuss the viscosity ratio effects on the dynamics of the two-phase flow. Subsequently, we explore the effects of geometry characteristics such as channel size, width ratio, and the angle between the main and side inlet channels, on the flow dynamics in a hydrodynamic focusing configuration. According to our findings, increasing the side channel width causes longer droplets, and the right-angled design effects the most efficient focusing behaviour.

Keywords: Microfluidics; Hydrodynamic focusing; Smoothed Particle Hydrodynamics; Immiscible liquids; Surface tension.
5.1 Introduction

The flow structures formed by two-phase flows in microchannels have attracted significant scientific and industrial attention in recent years. The importance of the micro-systems lies in the dominance of interfacial forces such as viscosity and surface tension over bulk forces, which enable controlled flow patterns, such as droplet formation, with precise control over the size of the droplets. Such controlled behaviour of two immiscible liquids interacting in an emulsion inside a microchannel is highly needed for a variety of industrial applications such as pharmaceuticals, food processing, chemical processing, enhanced oil recovery, and biotechnology [274-277].

Nakajima et al [17, 96] and Knight et al [97] were pioneers in generating monodisperse micro-droplets, the former through an array of cross-flow microchannels and the latter using a hydrodynamic focusing microchannel. Over the past decade, a number of research studies have reported developments being made in the field of droplet formation and breakup using different microchannel configurations, including T-junctions [15, 35, 105, 278], Y-junctions [18], Cross-junctions [20, 38], Hydrodynamic focusing and flow focusing [14, 16, 100, 106-109], Concentric injection [110] and Membrane Emulsification [111-113]. As is addressed by these studies, a potential strategy to increase the degree of monodispersity during droplet production in the micro-geometries is to draw from the characteristic features of multi-stream laminar flow systems, using continuous flows. In laminar flows, the multi-liquid flows through microchannels without any turbulence; interfacial forces dominate inertia and make it possible to control the flow structure, size of the formed droplets and threads by altering the relative flowrates of the multi-stream.

Hydrodynamic focusing is a successful example of monodispersed droplet formation micro-configuration, wherein a centrally located flow is narrowed or hydrodynamically focused by two neighbouring flows through cross-like microchannels. Xu et al [16] set up a three-stream laminar flow configuration to hydrodynamically focus a more viscous liquid surrounded by two inviscid liquids in order to facilitate a final breakup through control of the flow rate ratio. Cubaud and Mason [14] performed a comprehensive experimental study on Newtonian immiscible liquids in a hydrodynamic focusing microfluidic device. They investigated the competition between viscosity and interfacial tension effects on the
creation of capillary threads and droplets using hydrodynamic focusing over a large range of viscosities and interfacial tensions at a fixed geometry, which provides a useful range of flowrate ratios, viscosity ratios and \( Ca \). In addition, they continued their experiments on miscible fluids in order to investigate miscible interfacial morphologies and flow patterns [109]. Recently, Kunstmann-Olsen et al[108] studied experimentally and theoretically a two-dimensional microfluidic cross-flow and showed that the local geometry of the microchannel-junction affects the hydrodynamic focusing behaviour.

As demonstrated by Baroud et al. [277], the parameters by which the flow pattern of liquids interacting in a microchannel can be influenced, are very extensive and variables include the nature of the liquids, the details of the flow injections, the geometry of the channels, the surface characteristics and so forth. Despite various studies on droplet formation through microfluidic devices reported so far, a full understanding of the flow physics and the effective process parameters is still missing. On the other side, experimental studies for microchannel flows are quite challenging and expensive, so analytical solutions at such a small scale are limited to a few simple cases. Therefore, numerical studies can be complementary to these experiments. To date, there are only a limited number of numerical investigations in the literature. For instance, Wu et al. [38] presented an improved immiscible lattice Boltzmann model for simulation of immiscible two-phase flows in a cross-junction microchannel and also studied the effect of \( Ca \) on droplet formation in such a cross-flow systems. Furthermore, Liu et al. [20] studied the effect of flowrate ratios and capillary numbers on droplet formation in a microfluidic cross-flow configuration, using a multiphase lattice Boltzmann model. They used a limited range of capillary numbers \((Ca<0.01)\) and viscosity ratios \((\lambda\leq1/50)\), but a variety of flowrate ratios in their simulation and recognized three different flow regimes for their different flowrate ratios at a fixed capillary number. In spite of these numerical simulations, much effort is still required to accurately control interfaces and capture all the key parameters for the design development of microfluidic devices.

Nevertheless, modelling of multiphase micro-flows is not easy because of some of their special properties like free surfaces, moving interfaces, largescale differences in each dimension, and microscale phenomena [279]. Numerical studies involving microchannel flow simulations using mesh-based methods face a variety of obstacles. The most
important problem lies in the difficulties associated with generating the meshes, which would be a problematic task, time consuming and an expensive process for problems with complex geometry. Another challenge in mesh-based simulations is to track the movement of the interfacial boundaries of multiphase fluid systems. This is an expensive operation which affords another source for modelling error and numerical instability [130, 165]. Meshless methods have been the focus of research over the past few decades, in order to develop the next generation of more efficient computational schemes designed for more complex problems. One preferred meshless method is Smoothed Particle Hydrodynamics (SPH) [171, 182].

Recently, we presented a novel attempt to model the flow behaviour of two immiscible liquids in a hydrodynamic focusing micro-geometry using an incompressible Smoothed Particle Hydrodynamics (SPH) method. The dynamics of the two-phase flows at various capillary numbers were studied through the SPH-based simulation. Moreover, the simulation results were compared and validated against the existing experimental data and the comparison showed good agreement between the model prediction and experimental outcomes [280]. The efficiency of droplet formation in microchannels is mainly affected by the nature of the liquids and flow parameters such as viscosity, interfacial tension, flowrate ratio, capillary number and geometrical characteristics. Therefore, in the present work, we develop our SPH-based simulation to numerically study the effects of flow parameters and geometry on immiscible-liquid flow behaviour in hydrodynamic focusing microfluidics. The results are discussed in three sections covering the effects of the flowrate and flowrate ratio, the effects of the viscosity ratio, and finally the effects of the inlet channels’ geometry.

5.2 Model Details

A microfluidic right angled cross-junction consisting of three inlet microchannels and one outlet microchannel, as shown in Figure 5-1, was considered; the depth of the microchannels was assumed to be much greater than their width, $H$, and the study was restricted to a plane away from the top and bottom of the channels (i.e. the simulations were limited to two spatial dimensions). A Newtonian liquid, $L_1$, of viscosity $\mu_1$ entered the main inlet channel at a volumetric flow rate of $Q_1 = U_1 H^2$. Likewise, a second
Newtonian liquid \( L_2 \) of viscosity \( \mu_2 < \mu_1 \) was injected symmetrically into the two lateral inlet channels each at a volumetric flow rate \( (Q_2/2) = U_2 H^2 \). This configuration leads to the second liquid hydrodynamically focusing the first as outlined in the Introduction to the report here.

\[
Ca_1 = \frac{\mu_1 U_1 \gamma}{Q_1} \quad , \quad Ca_2 = \frac{\mu_2 U_2 \gamma}{Q_1 + Q_2}
\]

Figure 5-1 Schematic of cross-junction flow focusing geometry

The SPH-based model proposed in our earlier work [273] for simulation of immiscible liquid-liquid flows was applied here to investigate the effects of various flow parameters and geometry characteristics; full details of the SPH model, simulation parameters and boundary conditions can be found in Elekai et al. [273, 280].

5.3 Results and Discussion

5.3.1 Effects of flowrate and flowrate ratio

To investigate the influence of flowrate ratio, we keep the size of the microchannels constant at \( H = 100 \mu m \), and consider a reference liquid pair and constant flowrate of the main channel phase \( Q_1 \), whilst changing the flow rate of the continuous phase \( Q_2 \) and thus \( Ca_2 \). Firstly, we study the effect of flowrate ratio \( q = \frac{Q_2}{Q_1} \) on the flow behaviour of each liquid phase in a pair, including an 80 volume percentage of Glycerol in water as \( L_1 \) with
\( \mu_1 = 0.077 \text{ Pa.s} \) and PDMS oil as \( L_2 \) with \( \mu_2 = 8.2 \times 10^{-4} \text{ Pa.s} \) plus the interfacial tension of \( \gamma_{12} = 0.0304 \text{ Nm}^{-1} \) [14] at constant \( Ca_1 \).

The simulation results show various flow regimes at different flowrate ratios. Figure 5-2 represents the influence of the flowrate ratio on the flow pattern of the aforementioned liquid pair with a fixed viscosity ratio of \( \frac{\mu_1}{\mu_2} = 94 \), for which \( Q_1 = 25 \mu l/min \) and \( Ca_1 = 0.1 \) are fixed, while \( Q_2 \) and \( Ca_2 \) are variable with regard to a range of flowrate ratios at 0.1, 0.5, 1, 2, 3, 5, 10, and 20.
Figure 5-2 Effect of flowrate ratio on the flow pattern at (a) $q=0.1$, (b) $q=0.5$, (c) $q=1$, (d) $q=2$, (e) $q=3$, (f) $q=5$, (g) $q=10$, and (h) $q=20$, with a fixed viscosity ratio of $\frac{\mu_1}{\mu_2} = 94$

The flow structure of the liquid pair changes from the displacement pattern in Figure 5-2(a) and the tubing regime in Figures 5-2(b) and (c), to dripping and jetting regimes as shown in Figures 5-2(d) to 5-2(f) respectively, and finally the threading regime in Figures 5-2(g) and (h), as the volume fraction of the continuous phase $\alpha = \frac{Q_2}{Q_1 + Q_2}$ increases.

Moreover, it can be understood from the figures that the flowrate ratio affects the size of the formed droplets and the thickness of the threads, as well as the thickness of the core plug flow in the tubing regime. Comparing Figure 5-1(d) and (e) indicates that the droplet length in the dipping regime decreases when the flowrate ratio $q = \frac{Q_2}{Q_1}$ or the continuous phase volume fraction $\alpha$ increases.

In addition to the flowrate ratio, the flow rate quantity itself can influence the flow regime and associated features at a constant flowrate ratio. To study this effect, we have
performed a number of simulations in several groups for which various flowrates of the dispersed phase at a fixed flowrate ratio were considered. Figure 5-3 indicates a flow pattern map for the above liquid pair, based on the capillary numbers of both liquids, $Ca_1$ and $Ca_2$, and the flowrate ratio $q$. Note that changes in $Ca_1$ and $Ca_2$, here, were caused simply by the changes in the flowrates, $Q_1$ and $Q_2$.

![Flow regime map in terms of $Ca_1$, $Ca_2$ and flowrate ratio. Dripping (solid square), displacement (solid circles), tubing (solid triangle), threading (solid diamond), and jetting flow regime (solid pentagon).](image)

According to the Figure 5-3, variations of the flowrate quantities even at the same flowrate ratio can change the flow regime. In addition, the figure shows that at smaller values of $Q_1$ for the liquid pair, the dripping regime is achievable for a wider range of $q$ and $Ca_2$.

Furthermore, Figure 5-4 shows how the flowrate quantity can affect the length of the droplets of the aforementioned liquid pair in the dripping flow regime at a constant flowrate ratio of 2. Increasing the flowrate of the dispersed phase results in a smaller droplet and more stabilized tail, as is obvious in the Figures 5-4(a) to (c) and measured by Figure 5-4(d).
Figure 5-4 Effect of the flowrate quantities on the droplet size for the constant flowrate ratio of q=2 and at (a) $Q_1 = 10 \ \mu\text{l/min}, \ Ca_1 = 0.04, \ Ca_2 = 0.00086$, (b) $Q_1 = 25 \ \mu\text{l/min}, \ Ca_1 = 0.1, \ Ca_2 = 0.0022$ and (c) $Q_1 = 40 \ \mu\text{l/min}, \ Ca_1 = 0.168, \ Ca_2 = 0.0036$, (d) dimensionless droplet diameter vs. flowrate of $Q_1$

5.3.2 The effect of the viscosity ratio

Structure of two-phase flows in microchannels is controlled by the domination of one of the competing forces such as inertia, viscous forces and interfacial tension. In addition, the relative importance of the viscous stress of one phase acting on the other is a factor in determining the flow pattern. Viscous forces tend to deform droplets and keep the interface of the two-liquid phase smooth by dissipating the energy of the perturbations at the interface. To find out the effects, we have implemented three groups of simulations for Glycerol as the dispersed phase and PDMS oil as the continuous phase. During each group of simulations $\mu_1 = 1.214 \ \text{Pa.s}$ and $\gamma_{12} = 0.027 \ \text{Nm}^{-1}$ and other parameters such as the flowrates and flowrate ratio have been kept constant whilst a range of viscosity ratios,
denoted by $\lambda = \frac{\mu_1}{\mu_2}$ have been considered. Figure 5-5 shows the simulation results for the fixed flowrate ratio of $q=10$ and the different viscosity ratios as shown in Figure 5-5(a) $\lambda=1484$, Figure 5-5(b) $\lambda=264$, and Figure 5-5(c) $\lambda=24$. By decreasing the viscosity ratio and increasing viscosity of the continuous phase, the viscous stress of $L_2$ on the interface increases, which results in a change in the flow pattern from a tubing flow regime to a threading regime.

![Figure 5-5 Effect of viscosity ratio of (a) $\lambda=1484$ and $Ca_2=0.0036$, (b) $\lambda=264$ and $Ca_2=0.02$, and (c) $\lambda=24$ and $Ca_2=0.2$, on flow pattern at $q=10$ and $Ca_1=0.5$](image)

Figure 5-6 indicates how the droplet formation can be influenced by the viscosity ratio at the flowrate ratio of $q=8$ and $Ca_1 = 0.1$. As can be seen from the figure, at high values of $\lambda$ (when $\mu_2$ is very small), there must be an increase in the flowrate of the continuous phase at the same capillary number of the dispersed phase in order to achieve a dripping flow.
regime in the outlet channel. A greater disparity in viscosities of the two phases allows for a larger deformation and deviation from droplet generation. Furthermore, the figure indicates that longer droplets are formed at higher viscosity ratios.

![Figure 5-6](image)

*Figure 5-6 Effect of viscosity ratio of (a) \(\lambda=1484\) and \(Ca_2=0.00054\), (b) \(\lambda=264\) and \(Ca_2=0.003\), and (c) \(\lambda=24\) and \(Ca_2=0.033\) on flow pattern at \(q=8\) and \(Ca_1=0.1\)*

Figure 5-7 displays the results of another group of simulations when \(q=4.4\) and \(Ca_1 = 0.3\). By increasing \(\mu_2\), the viscous shear of the continuous phase acting on the dispersed phase increases, resulting in a change in the two-phase flow regime from displacement as shown in figure 5-7(a) to a jetting regime in figure 5-7(b) and then to a threading flow regime in figure 5-7(c).
Figure 5-7 Effect of viscosity ratio of (a) $\lambda=1484$ and $Ca_2=0.0009$, (b) $\lambda=264$ and $Ca_2=0.005$ and (c) $\lambda=24$ and $Ca_2=0.054$, on flow pattern at $q=4.4$ and $Ca_1=0.3$

5.3.3 Effect of geometry

In this section, we investigate the effects of the width ratio and inlet angle on the two-phase flow behaviour in the crossed microchannels.

5.3.3.1 Width ratio effect

The width ratio of the main channel $W_2$ with the side channel $W_1$ at a constant channel height $H$ can affect the length of the droplets in a dripping flow regime. In an attempt to study this effect, we have implemented the simulations at a fixed width $W_1 = 100 \, \mu m$ and various width ratios, denoted as $\zeta = \frac{W_2}{W_1}$. Figures 5-8 represent the simulation results for effects of $\zeta$ on the droplet size when $q = 2$ and $Ca_1 = 0.04$. According to the figure, the wider the side channel width, the longer the droplet length.
Figure 5-8 Effect of width ratio of (a) $\zeta = 0.5$ and $Ca_2=0.0036$, (b) $\zeta = 1$ and $Ca_2=0.0009$, and (c) $\zeta = 2$ and $Ca_2=0.00026$ on droplet length at $q = 2$ and $Ca_1 = 0.04$.

Figure 5-9 summarises the simulation results related to the width ratio effect on the droplet length as a function of the flowrate ratio at $Ca_1 = 0.04$. The figure confirms that there is a good agreement between the predicted results and the values calculated from the following correlation that was obtained by Cubaud and Mason [14] for droplet length measurement in a dripping regime:

$$\frac{l_d}{H} \approx \frac{1}{2} \left( \frac{Q_2}{Q_1 + Q_2} \right)^{0.17} \frac{Ca_2}{x_2}$$  \hspace{1cm} (1)

By increasing the width ratio in a dripping flow regime the size of the droplets increases at every flowrate ratio in this range. In addition, the changing trends at each $\zeta$ are quite similar with approximately equal difference which shows the identical effects of width ratio on the droplet size against the different flowrate ratios.
5.3.3.2 Inlet angle effect

In order to study the effect of the inlet angle, which is made by the main channel and the side channel in the cross-like configuration, three different angles were investigated and compared at the fixed flowrate ratio $q=2$, viscosity ratio $\lambda=94$, and width ratio $\zeta = 1$ with a width of 100 $\mu m$. The results obtained by the simulations at $\theta = 45^\circ, 90^\circ, 135^\circ$ are shown in Figures 5-10 (a), (b) and (c) respectively. As is observed in the figures, increasing the inlet angle leads to an increase in the droplet length. In addition, the breakup process is affected by changing the inlet angle from $90^\circ$. According to the results, the sharp-angled design delays the droplet breakup, whereas the obtuse-angled design hastens the separation. In the right-angled design, the maximum flow focusing is obtained, however, for a $45^\circ$ or $135^\circ$-angled design, the inclined flow direction of the continuous phase influences the focusing behaviour unfavourably. This effect arises from the decomposition of the inertial force into a smaller vertical component plus a horizontal component which is co-current with the dispersed phase in the $45^\circ$ design and counter-current with that in the $135^\circ$ design. Nonetheless, according to Figures 5-8 and 5-10, at a constant flowrate ratio,
the influence of the inlet angle is less important as compared with the effect of the width ratio.

![Diagrams showing the effect of different inlet angles on droplet length](image)

**Figure 5-10** Effect of an inlet angle of (a) $\theta=45^\circ$, (b) $\theta=90^\circ$ and (c) $\theta=135^\circ$ on the droplet length at $q=2$, $Ca_1=0.1$ and $Ca_2=0.002$

### 5.4 Conclusion

In this study, we investigated numerically the behaviour of immiscible-liquid pairs interacting through hydrodynamic focusing microchannels, based on the Smoothed Particle Hydrodynamics (SPH) method. We discussed the various aspects of the system, including the effects of the capillary numbers of both dispersed and continuous phases, the influence of important flow parameters and geometrical characteristics. Different flow patterns including viscous displacement, tubing, jetting, dripping and threading regimes were
obtained through variations of the flowrate ratio. The results show that the dripping regime only happens for \( q \geq 1 \), meaning that \( L_2 \) must be at least equal to or greater than \( L_1 \) to achieve a dripping flow regime. In addition, we indicated that the flowrate quantity and flowrate ratio affect the droplet length in dripping regimes. We studied the effects of the viscosity ratio on the flow structures and the dynamics of the droplets. As a consequence, a large value of \( \lambda \) imposes an increase in the flowrate of the continuous phase with the same capillary number of the dispersed phase to attain a dripping regime in the outlet channel. Subsequently, we investigated how the geometry of the channel junction influences the flow pattern and the hydrodynamic focusing. More specifically, we discussed the effects of the different width ratios as well as the inlet angle of the main and side channels in cross-like microfluidic configurations. According to our findings, increasing the side channel width causes longer droplets, and the right-angled design gives the most efficient focusing behaviour.
Chapter 6: Conclusion

This thesis is focused on outlining work that was focused on developing the application of a meshless explicit numerical simulation method for the study of immiscible liquid-liquid flows in microfluidic systems, so as to improve understanding of the fundamentals of flows in such devices and develop a design capacity for immiscible-liquid microfluidics. This work was undertaken through two related developments. The first one involved developing a Smoothed Particle Hydrodynamics (SPH) model for explicit numerical simulation of immiscible liquid systems. The model includes the surface tension, enforces strict incompressibility, and allows for arbitrary fluid constitutive models. The validity of the model was demonstrated by applying it to a range of interfacial problems with known solutions, including a liquid droplet falling under gravity through another quiescent liquid, Young-Laplace equation, and the deformation and breakup of a droplet under a shear field, whilst confined.

The second development in this research has been divided into two parts: the first involved the application of the proposed model for the study of hydrodynamic focusing of a liquid flowing in a deep microchannel by a second immiscible liquid introduced at right angles through identical but symmetrically opposed microchannels. The model was shown to be able to reproduce the experimental results of Cubaud and Mason (Phys. Fluids. 20, 053302, 2008), including the flow morphology mapping with the Capillary Numbers of the two liquids, and the characteristic dimensions of the focused liquid phase as a function of the liquid flow rate ratio. Going beyond the experimental results, the model was used to build understanding of the velocity fields.

The second part involved building a basis for the optimal design of the microfluidic system by determining the effects of various flow parameters and geometry characteristics on the flow dynamics and topological changes of the multiphase flow system. The different flow patterns, including displacement, tubing, jetting, dripping and threading regimes were identified as a result of changes in the capillary numbers of the interacting liquids. The effects of flowrate quantity and flowrate ratio on the droplet length in dripping regimes were also shown. The viscosity ratio effect was studied and it was demonstrated that a high value of the viscosity ratio imposes an increase in the flowrate of the continuous phase,
with the same capillary number of the dispersed phase, to attain a dripping regime in the outlet channel. Moreover, it was indicated how the geometry of the channel junction influences the flow pattern and hydrodynamic focusing. In this regard, the effects of the different width ratios, as well as the inlet angles of the main and side channels, were investigated in the cross-like microfluidic configuration. We concluded that increasing the side channel width causes longer droplets, and the right-angled design gives the most efficient focusing behaviour. The two-dimensional model developed in this thesis is expected to predict the parameters and variables with less than 10% deviation from the experimental results: that is reasonably useful for the prediction of the hydrodynamics of a 3D system. These developments will provide great insights for designing microfluidic devices involving immiscible liquid-liquid flows.

In terms of future work, there is clearly an opportunity to expand on the work concerned with different microfluidic configurations such as T-shaped junctions. It would also be of interest to extend the method to study double-emulsion microfluidic systems. In the case of the numerical simulation work, this could also be extended to a three-dimensional model. In addition, the model can be developed for fluids beyond those considered here, including non-isothermal conditions and non-Newtonian fluids. The standard SPH algorithm explicitly solves the conservation equations that is not efficient for the prediction of the rheological properties of non-Newtonian fluids with high viscosity and therefore an implicit SPH method is also required [281]. Before this happens, however, parallelisation of the code and adoption of more efficient methods for solving the Pressure Poisson Equation will be necessary.
References


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