Thermally induced effects for droplet-based microfluidics

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**THERMALLY INDUCED EFFECTS
FOR
DROPLET-BASED MICROFLUIDICS**

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SCHOOL OF MECHANICAL AND AEROSPACE ENGINEERING

2013

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ABSTRACT

In recent years, the emergence of droplet-based microfluidics has brought new life to microfluidics technologies. Researchers have demonstrated the potential of droplet-based microfluidics in addressing issues encountered by the current continuous flow microfluidics especially in the area of analytical chemistry for lab-on-chip applications. This dissertation will, first, review the various droplet forming techniques and their underlying physics. Control and manipulation of droplets in microchannel will also be discussed. A class of fluids, which is referred to as functional fluid, will be introduced. These are fluids which have been reported to enhance the performance of microfluidic devices with their unique properties. To accelerate the understanding of droplet formation under various hydrodynamic flow conditions and operating environment, numerical modeling will be an economical tool to supplement conventional experimental approach. In this dissertation, a numerical model is developed to assist the study of droplet formation in a T-junction geometry. The model is successfully validated with experimental data and correlates well to the response of a temperature mediated T-junction geometry where the viscosities of the fluids are induced to vary accordingly. In the final part of the dissertation, the model is further extended to investigate the thermocoalescence of droplet in a microchannel geometry which consists of a non-temperature controlled droplet formation section of the similar T-junction geometry cascaded to a temperature-induced merging chamber. The validated model will pave the path for our future work on droplet-based microfluidic research such as the investigation of droplet formation and coalescent with a

non-Newtonian or functional fluid in which their fluid properties can induce to change according to the external perturbation they are subjected to.

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NOMENCLATURE

Re	Reynolds number [-]
Ca	capillary number [-]
F_p	pressure force [N]
F_v	viscous force [N]
F_{it}	interfacial force [N]
H	smoothed Heaviside function
L	baselength [m]
M_d	desired mass [kg]
M_c	current mass [kg]
$\hat{N}_F$	interface unit normal
p	pressure [Nm^{-2}]
c_p	specific heat capacity [$Jkg^{-1}m^{-3}$]
k	thermal conductivity
Q	volumetric flow rate [$\mu l/h$]
r_p	radii of the particle [m]
D	diameter of droplet [m]
a	combined advective flux and diffusive conductance of the discretized momentum equation
b	source term of the discretized momentum equation
t	time [s]
$\bar{t}$	pseudo-time [-]
T	temperature [$^{\circ}C$]
$\vec{u}$	velocity vector [ms^{-1}]

V_d	volume of droplet [m^3]
V^*	dimensionless number of droplet volume [-]
$\vec{x}_p$	position vector of the center of the particles
s_p	sphere of radius of particle
E^+	positive escaped particle
E^-	negative escape particle

Greek symbols

σ	surface tension coefficient [Nm^{-1}]
μ	viscosity [$\text{Pa}\cdot\text{s}$]
ρ	density [kgm^{-3}]
κ	curvature
ε	interface thickness
ϕ	level-set function
Ω	domain of interest in the fluid region
Γ_i	location of interface or the zero level-set value
Δx	grid spacing

Subscripts

p	particle
+	dispersed or +ve phase
-	continuous or -ve phase

Acronyms

CFD	computational fluid dynamics
CV	control volume
IPA	Isopropanol
PDMS	polydimethylsiloxane
PMMA	polymethyl-methacrylate
EWOD	electrowetting on dielectric
LBM	Lattice Boltzmann Method
VOF	volume of fluid
LSM	Level-set method
PLS	particle level-set method
SIMPLER	Semi-Implicit-Method for Pressure Linked Equation Revised
WENO5	Weighted Essentially Non-oscillatory 5th order
TVD-RK2	Total Variation Diminishing Runge-Kutta 2nd order

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Chapter 1 Introduction

1.1 Background

The miniaturization trend in microelectronics has not only brought about the realization of Moore's law on microprocessor development, but also the development of non-electronic devices. The know-how from the silicon processes have been extended beyond the making of transistors into the building and integration of electrically driven micromechanical structures, commonly known as Micro-Electrical-Mechanical Systems or MEMS. Because of the ability to build and realize some intended functionality on micro-mechanical structure, there has been great interest, over the past two decades, to explore and extend MEMS application to fluid flow at microscale regime, thus, the emergence of microfluidics technologies.

Since then, microfluidics applications have evolved from microflow sensors, micropumps, and microvalves to form the highly visible and dominant technology building block in life science, chemical and biochemical systems. The diverse applications include chemical reaction [1], high throughput screening [2], cell sorting [3], proteomics [4] and even the transport of nanolitres of material [5] etc.

Cost, ease of fabrication, reliability and operating environment are key consideration factors for the design of a microfluidic device. Because of that, the initial micromachining processes for MEMS on silicon based substrate has also been extended to a wider material of choice such as polymer and plastic.

As compared to macro system, microfluidic system requires significantly smaller volume of sample/reagent and also shorter time of analysis. These advantages offer lower cost and higher throughput which makes microfluidic systems extremely attractive as a replacement for their macroscale counterparts. Although the advantages are obvious, it is hard to ignore the associated challenges of handling the fluids at such length scale. Especially for microfluidic biochemical analytical application, effective mixing of fluids is the key performance index to determine the success of the device design.

In the early development phase of microfluidic devices for use in a biochemical analytical system, fluids are handled in continuous flows. The low Reynolds number at such length scale renders the fluid to behave in the laminar flow regime. Unless externally assisted or geometrically induced flow perturbation, the dominant mixing mechanism between the fluid streams is diffusive instead of convective. Diffusive is a slower and less effective process. To improve the effectiveness of mixing at low such Reynolds number, innovative designs (both active and passive) have actively been explored by researchers. Nguyen and Wu [6] have reviewed on the various micromixer types and designs reported and classify micromixers into passive or active designs.

Passive micromixer designs, though less effective than the active design, are generally a preferred choice for complex microfluidic system due to simpler fabrication technology and ease of implementation. However, for a continuous flow design, the device will be constrained by the number of fluids that can be mixed and by the number of reactions. Size of the device will grow according to the number of chemical reaction/analysis needed. Therefore, scaling up will be a challenge.

Figure 1.1 – Different mixing concepts [6]

To address such challenge, microfluidic design has, in the recent years, evolved into droplet-based approaches [7, 8]. While continuous flow is analogous to analog microfluidics, droplet-based device can be regarded as digital microfluidics. In droplet-based microfluidics, discrete volumes of fluid or droplets are generated with immiscible phases. Droplets can be transported and manipulated in the immiscible carrier fluid [9, 10]. Droplet-based microfluidic systems offer an important feature of compartmentalizing reaction within a droplet. Each droplet can be regarded as a self-contained microreactor. The number of droplets formed within a given space or volume will imply the number of microreactor available for chemical reaction/analysis required by any complex microfluidic system. This feature will facilitate expandability and scalability of the microfluidic device without added complexity to the microfluidic system.

Apart from the scalability feature, the volume of reagent and sample within the droplet can be reduced into nano- or even femtoliter which is hardly achievable at this current state of technology for a continuous flow type of microfluidic device. As mentioned earlier, diffusive mixing is the main mechanism for a microreactor at such length scale. Each droplet serves as a small container within which the reaction between minute volumes of fluids takes place. This will drastically help to reduce the reaction time needed for chemical analysis which otherwise, in a continuous flow type of device, either passive or active designs are being pursued to improve the mixing efficiency [11, 12]. As droplets function as multiple microreactor units, this feature enable parallel processing and large data set to be generated for analysis which can dramatically increase the throughput of the device.

Even though droplet-based microfluidics is still at the infant stage of development, several applications in biotechnology have already been reported. For instance, DNA analysis [4], cell encapsulation [13], polymerase chain reaction [14] and reaction platform for protein crystallization [15].

The key performance indices of a droplet-based microfluidic system are the ability to generate highly repeatable droplets of controllable size and at regular yet adjustable frequency. In order to achieve that, the understanding of the physics and mechanism underlying the formation of a droplet in immiscible phases will have to be established. More often than not, this exploratory phase is best achieved through experimentation. With that being said, numerous experiment setups with different conditions are inevitable. This requires a holistic commitment in terms of time and financial resources which are burden to some research institution and project. But it is a critical step that cannot be ignored. With the fundament understanding established through experiment, a less economically taxing

alternative for the next stage of the research process will be to embark on numerical approach. The advancement of computer technology in the last decade has drastically increased the computing power and made numerical solution approach a viable option to be considered to relieve the burden and limitation of experimental research processes. Numerical solution for the variety of conditions can be generated, analysed and optimised before narrowing down to some focused condition of choice to be validated with an experimental setup to confirm the finding and conclusion. This approach applies well to the research of droplet-based microfluidic since this field is still very much at the infant stage and there are many potential applications waiting to be discovered. With the fundamental understanding of the physics and mechanics underlying microdroplet formation, numerical model can be developed to expand the study to cover a broader range and larger permutation of various dependency parameters in different hydrodynamic settings and geometrical design. With the help of a numerical model, the study of droplet-based microfluidics can also be extended, with lesser burden than experimental approach, to other immiscible functional fluids such as viscoelastic fluids, ferrofluids, nanofluids etc.

Microfluidic research with functional fluid is growing but scattered. Functional fluid has some unique feature that can be offered to microfluidic devices to address certain shortfalls. Functional fluid is a class of fluid with certain unique fluid properties which can help to introduce new functionality into microfluidic devices. Nguyen has used ferrofluid to improve performance of CFPCR [16, 17] and viscoelastic fluid to enhance mixing performance [18]. In specific to droplet-based microfluidic, these functional fluids may

render different characteristics in droplet formation and can be further explored for its potential applications.

To date, there have been very few studies reported in the literature [19, 20] on numerical modeling of digital microfluidics. The main focus of this project is to establish a reliable numerical model on droplet formation in digital microfluidics and later, extend the modeling capability to a thermal forcing environment.

1.2 Objectives and scope

The objectives and scope of this dissertation are:

- To establish a numerical model of droplet formation in microfluidic channel.
 - Formulating a numerical model of droplet formation of an incompressible immiscible two-phase flow in a T-junction cross-flow geometry.
 - Validating the numerical results of the model using experimental data
- To numerically investigate the droplet formation process under a thermal forcing condition.
 - Perform numerical simulation of droplet formation in a thermally mediated T-junction microchannel
- To numerically investigate the behavior of droplet movement in a microfluid channel leading to droplet coalescence in a thermally mediated environment.
 - Perform numerical simulation of droplet coalescence in a temperature-controlled merging chamber and comparing the results for different heater configurations.

1.3 Organisation

This dissertation is organized as follows:

- Chapter 1 introduces the background, objectives and scope of the research.
- Chapter 2 describes the physics of droplet formation in immiscible phases, various method of droplet generation in different microfluidic channel geometry, transport and manipulation of droplets in digital microfluidics.
- Chapter 3 describes in detail the mathematical formulation and solution procedure of the numerical model.
- Chapter 4 describes the experimental setup, fabrication of the device under experiment and the numerical results of a thermally mediated droplet formation in a T-junction microfluidic device.
- Chapter 5 reports the numerical investigation of thermocoalescence of droplet in a microchannel designed with a merging chamber and compares with the experimental data.

Chapter 2 Literature Review

2.1 Droplet formation techniques

Unlike continuous flow microfluidics, digital or droplet-based microfluidic involves the discretisation of one or more fluid in another immiscible fluid acting as a carrier to create multiple individual microfluidic units or droplets which can be transported and manipulated to achieve an intended purpose. As each droplet is a self-contained microreactor, digital microfluidic has opened up a novel micro-environment which can be exploited to address the limitation of its continuous flow counterpart and even breaks into new applications.

Bubble formation refers to the emulsification of gaseous phase in liquid creating a gas-liquid interface. Droplet formation, in our context, is the emulsification of a liquid in another liquid forming a liquid-liquid interface. This dissertation will focus on liquid-liquid immiscible phases i.e. droplet. In most of the reported droplet-based devices, the dispersed phase is water or some aqueous solution and the continuous phase is usually oil. However, the choice of fluid is dependent on the fabrication material of the microfluidic device. As the droplet comes close to the wall surface of the microchannel, it will interact with the material of the microchannel. To achieve droplet formation, it is desirable for the wall to be wetted by the continuous phase. As such, the wettability of the continuous phase to wall material of the microchannel is a key consideration for the choice of fluid.

Prior to understanding the mechanism on droplet formation process, a few key parameters that characterize and have significant impact on the break-up process will be

discussed. In microfluidics, Reynolds number, Re , is commonly used to characterize the flow regime of the fluid flow in the microchannel. It indicates a transition from laminar to turbulence flow and also the significance of viscous stresses over inertia forces. Re is defined as

$$Re = \frac{\rho \bar{V} d_0}{\eta} \quad (2.1)$$

where $\bar{V}$, ρ , η and d_0 are the mean velocity, fluid density, viscosity and the characteristic length. For a given channel cross section, the flow is laminar below a critical Reynolds number and becomes turbulent when above the critical Reynolds number. However, external disturbance can impact the value of this critical Reynolds number. At $Re \ll 1$, which is typical in microscale flow, the viscous stresses and pressure gradients dominate inertia effects. Such flow characteristic allows a precise control on the trajectories of the fluid particles [21].

Another key and most important parameter that determines the droplet dynamic is the dimensionless capillary number Ca . Capillary number is used to characterize the relative importance of viscous stresses over capillary pressure. In other words, it indicates the dominating effects of interfacial forces over shear stresses. Capillary number is described by the flow field of the continuous phase which is acting to deform the fluid interface and has the expression below.

$$Ca = \frac{\eta \dot{\gamma} a}{\sigma} \quad (2.2)$$

where η is the dynamic viscosity of the carrier fluid, σ is the interfacial tension between the carrier fluid and aqueous fluid, a is the half-width of the aqueous fluid channel and $\dot{\gamma}$ is the effective shear rate.

There is a critical Ca value above which the droplet breakup will occur. The critical Ca value is specific to the geometrical design of a microfluidic system. Several values have been reported in various research papers [22-24].

A number of methods have been reported on the generation of droplets which can be classified as either passive or active. Passive methods rely mainly on the natural hydrodynamic settings to generate droplets and are more popular than active method. Unlike passive method, active methods require some external forcing mechanisms (can be mechanical, electrical, magnetic etc.) to create or induce the necessary actuation needed to assist the generation of droplet. In this review, passive methods will be emphasized although several active methods will also be briefly discussed. There are three groups of passive methods each has a specific nature of flow field that acts to deform the liquid interface near the break off region of the droplet. They are co-flow, cross-flow and flow-focusing configurations. Each method is facilitated by its unique geometrical channel design and will be described in the following sections.

2.1.1 Co-flow configuration

Co-flow droplet generation is achieved by driving the dispersed phase through a capillary or set of concentric channels into a co-flowing continuous phase. Umbanhowar et al [25] have reported to achieve the droplet generation through the co-flow method by using a macroscale experimental setup (Figure 2.1). They have shown to generate droplets of a

variety of fluid by driving the dispersed phase (silicone oil, hexadecane and liquid crystal) through a commercially available micropipette into a cup of continuous phase of water with added surfactant rotated by a speed-controlled motor to achieve a steady flow.

Figure 2.1 – Conceptual co-flow method: a) Experimental setup b) & c) Closed up view of tip/fluid contact geometry [25].

It can be seen that the interacting pathways of the dispersed and continuous phase are brought together not through a monolithic setup of microchannels but the integration of two separate flow apparatus. Another co-flow experimental setup by Cramer et al [26] has somewhat a similar concept (Figure 2.2).

Figure 2.2 – Cramer's Co-flow experimental setup [26].

The dispersed phase is driven through a capillary into a flowing stream of continuous phase in a rectangular channel. The only difference is the flow pattern of the continuous phase with respect to the dispersed phase. In Cramer's setup, the flow is parallel while Umbanhowar's [25] setup has a rotating flow.

There are 2 regimes of droplet formation – dripping and jetting. The regime is characterized by the distance from which the breakup of droplet occurs from capillary tip. Dripping takes place if the break up is near the tip. Whereas, jetting occurs at a distance further away from the tip. The transition between regimes is characterized by dynamics of the droplet formation process and the breakup mechanism.

Umbanhowar et al [25] reported that there is a critical flow rate of the dispersed phase where a transition between the regimes occurs. Below the critical flow rate dripping takes place and the droplet will grow spherically as the dispersed phase exits from the tip of the capillary. During this stage, the interfacial tension basically holds the forming droplet and acts in opposition to the flow field of the continuous phase. This will continue until the net force acting on the droplet become non-zero and the instability in the force equilibrium surrounding the droplet becomes overwhelming enough that a breakup eventually occurs. Above the critical dispersed flow rate, the droplet formation transition into jetting regime. At this regime, flow field of the continuous phase is more dominating than that of the dispersed phase such that it exerts a drag force on the forming droplet and stretching it downstream and away from the capillary tip forming a filament thread. The thread thins as it stretches further away from the tip and eventual breaks off. The two regimes are qualitatively described by the distance from capillary tip at which the breakup occurs.

Umbanhowar et al [25] have reported the production of droplets of diameter 2-200 μm with the co-flow technique using their experimental setup. It was also reported that the droplet diameter decreased with increasing velocity of the continuous phase. Effects associated with the velocity of continuous phase are also dealt with by Cramer et al. As described above, droplet break up is the result of loss of equilibrium between the surface tension and hydrodynamic force surrounding the forming droplet. The rate at which loss of equilibrium occurs is dependent on the velocity of the continuous phase. The higher the velocity the quicker will be the breakup rate. For a given dispersed phase flow rate, increase in break up rate will translate to shorter time for the droplet to grow and thus, lower volume and smaller droplet size will be formed. Experimentation on the effects of

disperse phase, viscosity, interfacial tension and channel aspect ratio on droplet formation process was carried out by Cramer. It was reported that, at low flow velocity of the continuous phase, the break up time will become approximately constant at a critical disperse flow rate. The disperse flow rate will complement the flow velocity in order for a constant break up to be reached. However, this behavior is not reported to be seen at higher flow velocity. Although the author did not pursue any further, it is likely that the behavior can be observed if the disperse flow rate can be adjusted beyond the experimental limit.

With regard to the effect of viscosity, it was reported to have minimal effect on the droplet size for a given velocity flow. Longer threads are, however, observed at higher disperse viscosities due to increase in viscous stresses that are acting in opposition to the capillary pressure. As a result of longer threads, more satellite droplets are formed after the breakup of the primary droplet.

The effect of interfacial tension on droplet formation is shown to have similar trend except that smaller droplet is obtained for the case of lower interfacial tension. This is because interfacial tension is the force holding the droplet at the tip during the forming stage and lower interfacial tension will lead to shorter time for the droplet to form thus results in smaller droplet size.

In Cramer's experiment, a change in the depth-width aspect ratio of the channel cross section from 8 to 2 is reported to have no significant impact to the general principles on the droplet formation. Size of the droplet is smaller for the low aspect ratio allowing the droplet to come close to the channel wall and thus wall effect prevails.

2.1.2 Cross-flow

Cross-flow droplet generation method is, by far, the most popular among the other methods. The droplet generation is supported by a T-junction geometry as shown in Figure 2.3 which Thorsen et al [27] are the first to implement this technique for the formation of droplet.

Figure 2.3 – T-junction Geometry [27].

The method is also commonly referred to as T-junction flow. The dispersed fluid phase enters at the inlet channel and the carrier phase in the main channel. This geometry allows the dispersed phase at the inlet channel to intersect perpendicularly with the carrier phase in the mainstream. The geometry of T-junction is characterized by the width of the main channel, the width of inlet channel and the height of the channels. The dynamic of the droplet formation process depends on the aspect ratio between the main and inlet channel width and also between the channel width and the height.

At small capillary number, the shear stresses at the break off region of the forming droplet are insignificant to distort the forming droplet and the droplet is held from detachment by interfacial tension. As a result, the droplet grows until it almost covers the

entire cross-section of the channel leaving a thin wetting film for the carrier fluid to flow through in the main channel. In this situation, pressure from the carrier fluid builds up at the upstream side of the forming droplet thus pushing or squeezing the droplet towards downstream and forming an immiscible thread. Thinning or necking of the thread will occur at a rate proportional to the flow rate of the carrier fluid. At the same time, the volume of the droplet will also grow at a rate proportional to the flow rate of the dispersed phase. The neck will eventually break as the fluid interface at the upstream side of the droplet touches the downstream edge of the inlet channel. The neck will eventually break as the fluid interface at the upstream side of the droplet touches the downstream edge of the inlet channel.

Figure 2.4 – Stages of droplet formation [28].

It can be seen that, at low capillary number, the breakup process is a balance of tangential shear stress due to the buildup of the continuous phase flow pressure across the droplet and the dominating interfacial forces at the neck of the forming droplet that act to resist the deformation. The forming process is described to be in the squeezing regime [29]. When the capillary number increases above certain critical value, droplet formation process transits into dripping regime which is similar to that seen in the case of co-flow droplet generation method. In this dripping regime, shear stresses become significant and are the main driving forces for the thinning and necking of the immiscible dispersed phase thread during the droplet formation process [30].

Based on the mechanics of droplet formation as described earlier, droplet formation is the results of interaction of the various forces around the forming droplet as the dispersed fluid enters into the flowing stream of immiscible carrier phase. As shown in Figure 2.5, the forces are made up of, F_p , the pressure force due to the stream-wise pressure difference across the forming droplet, F_v , the viscous force due to viscous stress around the forming droplet and, F_{st} , surface tension force between the phases.

Figure 2.5: Various forces acting on a forming droplet in a T-junction.

As the dispersed phase enters from the inlet channel into the main channel which carries a flowing stream of continuous phase, an interfacial boundary between two phases begins to form and it penetrates into the main channel with a spherical front. This growing interfacial boundary of spherical shape is pushed downstream by F_p and F_v . As the forming droplet grows, the interfacial area becomes larger and thus F_v , which exerts on the forming droplet as a form of surface stress, increases accordingly. F_v is proportional to viscosity and velocity gradient of the continuous phase. The droplet is withheld from detachment by F_{st} which is proportional surface tension, σ , and the circumference of the dispersed thread,

l . As the forming droplet is pushed downstream by F_p and F_v , the dispersed thread is stretched and the circumference of the thread, l , becomes smaller as a result. It will be good to note that, in the case of low capillary number, F_p , will be the dominating force in the droplet formation. As the thread thins, a region of high pressure is also created at the thinnest part of the thread due to the surface tension effects. This causes F_{st} to weaken further until a point where F_{st} is no longer strong enough to withhold the forming droplet and the thread breaks. Thus, a droplet is finally formed.

Depending on the value of capillary number which determines either the squeezing or dripping regime of the formation process, the shape of the droplet will follow accordingly. For example, the squeezing regime will produce droplets with an elongated body and spherical ends. Whereas, in the dripping regime, the droplet formed is almost spherical in shape. Regardless of the shape of the droplet, the potential applications of a droplet-based microfluidic device will increase drastically if it is able to precisely control the size of droplet generated. As explained earlier, the droplet size is dependent on

- Flow rates and flow rate ratio of dispersed and continuous phase.
- Viscosities of the phases
- Interfacial tension between the phases
- Geometrical dimension of the device

In most cases, the geometrical dimension, viscosities and interfacial tension are pre-determined by the design of the device and the choice of fluids used. Very little control to these parameters can be achieved during the device operation. Therefore, the only possible way to control or vary the size of the droplet is adjusting the flow rates of the

phases and let the fluid hydrodynamic settings take its natural course. Thus, this control method of the droplet size is classified as a passive method. If, however, the fluid properties can be altered during the device operation by inducing a stress or altering the environment thermally, electrically or magnetically this will open up a new and exciting perspective into the control of droplet size which is referred to as active control methods and will be discussed in section 2.2.

2.1.3 Flow focusing

Unlike the other two droplet formation methods, flow focusing leverage on the elongational flow. This is achieved by elongating the dispersed phase with a surrounding stream of continuous fluid flow and directing the fluid streams through a constriction to achieve the final breakup. Both the dispersed phase and continuous phase flow coaxially unlike the parallel flow pattern in the co-flow technique. Anna et al [31] is among the earliest research groups to have a microfluidic geometrical design to facilitate the flow focusing technique for two-phase liquid-liquid dispersion. It has a planar design with three microchannels built to direct fluid flow co-axially through an orifice into a collection channel.

Figure 2.6 – Flow focusing geometry [31].

The middle channel carries the dispersed phase while the two outer channels carry the continuous phase. At the exit of the channels, the two surrounding streams of continuous phase exert pressure and viscous stresses on the middle stream of dispersed phase thinning it into a narrow thread before flowing into the orifice. An expansion of the flow is experienced as the fluid stream exits the orifice. This process is accompanied by a slowdown in the flow velocity and the interfacial tension allows the formation of a spherical bulb at the end of the thread. Through this thin thread, the dispersed phase continues to be delivered into the bulb and gradually grows into a droplet. Once the fluid interfacial instability builds up to a critical point, the thread will break up at the base of the forming droplet or rather, the neck and the forming droplet are finally separated. Depending on the flow condition, the breakup can occur near or downstream of the orifice thus give a rise to the different regime of droplet formation – dripping or jetting.

Anna et al reported a wide range of droplet formation pattern observed with different flow rate ratio and also within different continuous flow rate. The observations range from droplet size changes (from approximately the size of orifice to much smaller), uniformity of droplet size (mono-disperse/bi-disperse/poly-disperse) and regime of droplet formation process (dripping/jetting). These observations are subsequently supported by theoretical and numerical models reported by various research groups. Besides the planar design, axisymmetric three-dimensional designs are also reported [32-34]. Except for being more robust, they are qualitatively similar in the mechanism of droplet formation process to that of the planar design. Generally, for flow focusing technique, the dispersed phase is co-axially manipulated and controlled by side stream of continuous phase prior to

flowing through an orifice. Other two-phase but liquid-gas dispersion is reported by Ganan but will not be focused on in this report [35, 36].

Similar to co-flow technique, dripping and jetting regimes can be observed in the droplet formation process in flow-focusing configuration. In the dripping regime, droplet breakup occurs near the exit of the orifice while for jetting, the breakup occurs at a few order of orifice diameter away from the orifice exit. Jetting can be easily observed as a long jet of thread trailing behind the forming droplet as it travels into the collection channel. But the physics underlying the droplet formation process and dripping-jetting transition differs from the co-flow and flow-focusing methods. Transition between the regimes is reported to be dependent on the flow rate ratio and capillary number.

In Utara's experiment [33], it was deduced that the breakup mechanism is solely attributed to the capillary instability. However, in Zhou's investigation [37] on the breakup mechanism, it was reported that both capillary instability and viscous drag force from the flow field are the underlying components for the breakup. Capillary waves, which are the telltale sign of capillary instability, can be witnessed as early as inside the orifice. The waves show up as a fluctuation of the radius of the dispersed phase thread as seen in Figure 2.7.

Figure 2.7 – Capillary wave in a long nozzle. [37] a) Channel geometry b) Radius of thread.

As Zhou [37] has proven in his simulation, capillary instability is not alone responsible for the breakup, the flow field after the exit of the orifice has a significant stake in the breakup.

The differing conclusions by Utara and Zhou could be attributed to the difference in the geometrical setup. The length of the orifice in Utara's setup is much shorter than Zhou's setup and this gives rise to the differences in the velocity profile of the fluid flow. The flow velocity starts to develop a parabolic profile as early as within the orifice in Zhou's setup. Whereas, in Utara's setup, the parabolic profile is only seen at a distance downstream in the collection channel during jetting. Thus, the breakup mechanism and

dripping-jetting transition in both setups are qualitatively different. As such, flow focusing technique is the most complex among the three droplet formation techniques and thus, requires a more comprehensive and robust model to accurately describe the flow.

The factors affecting the size of droplet are different for each regime. In the dripping regime, the droplets formed are highly monodisperse. The droplet diameter is dependent on the flow ratio and capillary number. For a given collection channel width, the droplet diameter is observed to decrease with decreasing flow ratio and increasing capillary number.

The deliveries of both the continuous and dispersed phase into the flow focusing device are commonly achieved through the use of syringe pumps. This method of fluid delivery provides a volume flow rate control. Ward has reported an alternative method of delivering the fluids by pressure pumping approach. This is to achieve a pressure controlled delivery. In the experiment, Ward [38] reported that pressure control has a more significant influence on the droplet formation than flow rate control. Droplet size varies more prominently to the changes in pressure than changes in flow rate. This is alluding to non linearities in the surface tension although it is yet to be fully understood.

2.2 Droplet control techniques

In the recent years, research on droplet-based microfluidics has gained a great interest in the area of analytical chemistry. The key motivation is that each droplet can act as a microscale chemical container which provides physical and chemical isolation for a pico- or micro-liters of content. Within each droplet, a desired chemical reaction can be contained. Multiple droplets containing a minute volume of reactants allow parallel reactions to happen at a reduced reaction time, and thus, result in an overall increase in the throughput of a chemical analysis system. To realize these benefits, it is crucial for the digital microfluidic device to manipulate the droplets. The operations include droplet sizing, control of pinch-off frequency, fission, fusion and mixing of the droplet contents. Several techniques are reported by researchers and will be discussed briefly in this section.

2.2.1 Droplet sizing and pinch-off frequency control

Passive technique relies mainly on geometrical design of microchannel to achieve the desirable droplet formation. This has already been discussed in co-flow, cross-flow and flow focusing method whereby droplet control depends purely on the dimension of the inlet or orifice as well as the hydrodynamic setting of the liquid phases such as flow rates. There is no induced change to the fluid properties or external physical perturbation in the droplet formation process.

Droplet sizing and pinch off frequency can be controlled through external forcing. External forcing includes but is not limited to thermal [28, 39, 40], mechanical actuation [41-43] and electric field [44, 45].

In thermal forcing, a localized heat source is introduced into a passive droplet generation device to modify the hydrodynamic settings and in turn alters the underlying conditions of the droplet formation process with an ultimate aim to achieve control over the droplet size or frequency of formation. Because of the thermal dependency of fluid properties, thermal forcing will change the viscosity of the fluids and the surface tension of fluid interface. Nguyen et al [40] have reported that the droplet size can be increased by as much as 2 times in a thermally mediated droplet formation environment. Knowing the temperature dependency of fluid properties, they have integrated a microheater and a temperature sensor at the droplet breakup location of a flow focusing device to induce a controlled heating effect targeting at modifying these fluid properties. The temperature range of the heater control unit is 25 to 70°C, a range deems to have minimal adverse chemical effect on droplet containing biological sample. With a thermally controlled change to the fluid properties, they reported that the droplet size of deionised water (DIW) increases with temperature. The change is less significant for channels with a larger depth. This is attributed to a larger thermal gradient around the location breakup as compared to channels with smaller depth which has a smaller cross-sectional area. As a result, the thermally induced effect is much smaller and the change in fluid properties with temperature has a lesser impact

Physical forcing is an alternative method to control droplet size and its production rate. Willaime et al [41] has integrated a deflecting membrane into a T-junction microfluidic device. The deflecting membrane is located between the actuation channel and the dispersed fluid channel. The membrane is deflected at controlled amplitude and frequencies by regulating the pressure of water flowing in the actuation channel.

Deflection of the membrane will have a bearing on the instantaneous flow velocity of the dispersed phase fluid. It has been shown that, when the membrane is actuated in the multiples of natural frequency of the droplet formation, the droplet size will reduce linearly and the rate of droplet production is higher than the natural frequency. Another form of physical forcing is by mechanically inducing a breakup during droplet formation process. Chen and Hsiung [42, 43] achieved this by pneumatically actuating the pressure chambers which, in turn, collapse the channel walls surrounding the thread of forming droplet causing the droplet to pinch off. The droplet size is thus highly dependent on the actuation frequency as well as the flow rate ratio of the working fluids.

Figure 2.8 – Physical forcing using pressure chambers[43].

The use of electric field to induce a breakup of liquid thread is another droplet forming technique [45, 46]. As shown in Figure 2.9a, two immiscible fluids with different conductivities and permittivities are driven into the main channel through each of the inlets. Electric field is generated across the main channel by two electrodes on opposite walls driven with a voltage potential difference. In the absence of the electric field (i.e. at zero voltage potential difference), the fluids remain separated in the main channel and is

depicted by a flat line of the interface shown in Figure 2.9b. When a voltage difference is applied, an electric field is generated and the line of interface deflects creating an interfacial instability (Figure 2.9c). If the electric field is sufficiently large, the interface will be deflected until it collides with the opposing wall causing the liquid thread to rupture and finally forming the droplet. By controlling the voltage, the magnitude of electric field will adjust accordingly which in turns, impact the rate of change of the interfacial deflection to reach the wall and the time to rupture the flowing liquid thread. This will have a bearing on the size of droplet formed thus, making it highly dependent on the magnitude of the electric field.

a) Device setup

b) Electric field OFF

c) Electric field ON

Figure 2.9 – External forcing technique using electric field [45, 46].

2.2.2 Droplet fission

Because each droplet can act as a vessel for chemical reaction, digital microfluidic has an edge over continuous microfluidic in applications for chemical analysis because of its high throughput and scalability feature. To augment this feature, one critical operation in digital microfluidics is droplet fission or splitting.

Passive fission relies on geometrical design of the microchannel network to split the droplets [47, 48]. Adamson et al [47] have designed a T-junction microchannel network to split a liquid plug. Liquid plug is a large droplet which shaped like a capsule and touches both sides of the channel wall. Symmetrical and asymmetrical splitting can be achieved depending on the pressure difference across the splitting arms. As shown in Figure 2.10 below, the daughter droplets are equal in size for symmetrical splitting as compared to the differing droplet volume in the asymmetrical split.

Figure 2.10 – Passive droplet fission[47]

Other parameters to control the droplet splitting in passive fission technique are carrier fluid flow rate and resistance of the splitting arms [48]. These control parameters are not externally driven unlike the active fission technique. One such externally driven technique is EWOD – electrowetting on dielectric. As depicted in Figure 2.11, the technique is based on the principle that electric charge can modify the surface tension of a droplet by altering the free energy on the dielectric surface with a resultant change in the contact angle of the

droplet and thus, inducing wettability. When the electric potential is deactivated, the droplet returns to its original non-wetting shape indicating that the process is reversible.

Figure 2.11 – Illustration of the effect of EWOD[49].

Cho et al [49] implemented EWOD droplet fission using a system of surface electrodes that surrounds the droplet. Surface electrodes near the opposite ends of the droplet are activated which pull the droplet at each end until it pinches off and divided into two daughter droplets (Figure 2.12).

Figure 2.12 – EWOD droplet fission [49].

2.2.3 Droplet fusion

As important as how droplet fission operation can increase the throughput of the system, droplet fusion operation has to ensure that droplets containing different reagents are kept separated until required condition is reached when coalescence of droplets will take place.

A number of researchers have attempted to study the physics behind droplet coalescence. Jeffreys [50] has a good review on the coalescence. The process of droplet coalescence happens in three stages:

- Stage I - Distance between droplets decreases and brought closer to each other
- Stage II - When two droplets are brought close to each other, a thin film is formed between the interfaces. A disjoining pressure occurs within the thin film. The film will be in an equilibrium state and stable in the presence of positive disjoining pressure. However, if thinning of the film continues to happen to down under a thickness of $1000\text{\AA}$, a negative disjoining pressure will start to develop and destabilize the film.
- Stage III - When the negative disjoining pressure has reached a critical value, the thin film will become unstable and eventually rupture. At this point, droplet coalescence occurs.

The thin film has a non-uniform thickness. Prokhorov [51, 52] and Lindblad [52] have observed a dimpled film formed between the droplets (Figure 2.13).

Figure 2.13 – Dimpled film between spherical droplets [51]

The disjoining pressure of the thin film is the result of Van der Waals, electrical double-layer and structural forces [53, 54]. Chen [55] has attempted to model the coalescence of droplet with consideration to the forces from Van der Waals and electrical double layer. The Van der Waals force will develop a negative disjoining pressure that acts to attract the fluid-fluid interfaces of the adjacent droplets to each other. This negative disjoining pressure increases with decreasing thickness of the thin film. On the other hand, the electric double layer will develop a positive disjoining pressure that repel the fluid-fluid interfaces away from each other and its magnitude decreases with decreasing thickness of the thin film. Droplet coalescence is, thus, pivoted on the resultant of these oppositely acting forces which is highly dependent on the thickness of the thin film.

In the presence of surfactant, Bibette [56] attributed the occurrence of droplet coalescence to two microscopic parameters: surfactant chemical potential and the pressure exerted between the droplets in close contact. The surfactant chemical potential determines the density of surfactant molecules at the fluid interfaces. A higher surfactant chemical potential will imply a surfactant-rich situation whereby the interfaces are covered sufficiently with surfactant molecules to prevent coalescence. This will lead to greater

stability of the droplet. On the contrary, a low chemical potential will result in an increase susceptibility to coalescence. In the situation of high surfactant chemical potential, the condition for coalescence will rest heavily on the pressure exerted between the droplets. If sufficient pressure is presented, it will act to press the droplets together and squeeze away the water thus creating a favorable condition for coalescence. Nicolas [57] has, however, reported a counterintuitive phenomenon observed in their experiment which attributed droplet coalescence to separation of phases rather than upon impact. It was explained that separation induces the formation of two facing nipples at the contact area between the droplets. This has led to the depletion of surfactant molecules at the interface which destabilizes the droplet pair and eventually results in coalescence.

Both passive and active techniques have been demonstrated by researchers to induce droplet coalescence.

a) Straight expansion

b) Tapered expansion

c) Flow rectifying

Figure 2.14 – Various channel designs for passive droplet fusion [48].

The working principle of passive technique is to bring the droplets closer by reducing the flow rate thereby draining off the carrier fluid that separates the droplet until only a thin film is sandwiched between the droplet interfaces. Upon further increase of pressure on the film and imbalance in the surface tension, the film ruptures and droplet fusion is achieved. Tan et al [48] have demonstrated several geometrical designs to reduce the flow rate needed to bring the droplets closer. As shown in Figure 2.14, they use straight channel expansion, tapered expansion and flow rectifying design. In flow rectifying design, the droplets are directed and fused at the middle of the junction for fusion. The focusing of droplets is achieved by balancing the flow of the separating carrier fluid.

Figure 2.15 – Use of pillars structure to induce droplet coalescence [58]

Figure 2.16 – A trifurcating junction to rectify flow [59]

Niu [58] has explored the use of pillar structure in microchannel to induce droplet coalescence (Figure 2.15). Tan [59] employed a trifurcating junction to redistribute equally spaced droplet and bring the droplets closer to achieve coalescence (Figure 2.16).

a) Configuration setup

b) Electrocoalescence of droplet

Figure 2.17 – Active droplet fusion [60].

Similarly, active droplet fusion technique has been demonstrated by Priest et al [60] electrically using a short electrical pulse of 100 ms at 1 V (Figure 2.17). The electrical pulse breaks down the thin film of carrier fluid between the droplets and induces interfacial instability which leads to the coalescence of the droplets. Luong [61] and Xu [62] have demonstrated the thermocoalescence of droplets in a merging chamber. The experiment shows a dependency on flow rate for the critical temperature at which droplet coalescence occur.

2.2.4 Mixing in droplet

As each drop acts a vessel for chemical reaction, the efficiency of the reaction will be enhanced if the reagents within the droplet are properly mixed. In continuous microfluidics, it has been known that the laminar flow condition of the fluids has posed as a challenge to mixing. Mixing is still a challenge in digital microfluidics as the laminar flow condition of the fluids continues to exist within the droplet. In laminar flow, which

occurs at low Reynolds number, the fluids exist in distinct streams. As such, mixing between the fluids is due mainly to diffusion. However, the time required to completely mix the fluids is still long even at micrometer length scale. Several mixing techniques have been demonstrated by researchers. Basic concept is to induce flow instability to the fluids contained inside the droplet so as to create chaotic advection to facilitate mixing. This can be achieved either by subjecting the droplets to flow through undulating channel geometries [63] or maneuvering the droplet using externally induced thermocapillary forces [64, 65], electrostatic forces [66, 67] or magnetic forces [16, 17] to promote the necessary motions to the droplet for the purpose of creating chaotic advection to its content.

The former technique is classified as passive mixing where it relies only on the channel geometry and there is no externally assisted mechanism or forces. The channel design consists of a series of bends and turns through which the droplets will flow and chaotic advection within the content is achieved. As shown in the experiment conducted by Bringer et al [63] (Figure 2.18), the orientation of droplet will change uncontrollably as it travels through bends stretching and folding the droplet as well as its contents.

Figure 2.18 – Mixing pattern with the droplet for different geometrical design [63].

Pollack et al [66] have demonstrated the use of electrowetting to manipulate the movement of KCL (potassium chloride) droplet in their microactuator device shown in Figure 2.19.

Figure 2.19 – Electrostatic microactuator [66]

When a voltage difference of 40-80V is applied to adjacent control electrodes, the droplet is transferred between the electrodes. This is due to a reduction of interface energy as the charges build up at the interface between the droplet and the energized electrode resulting in the motion of the droplet. The electrodes can be controlled to create a reciprocating movement of the droplet so that flow turbulence can be induced within the droplet contents to achieve an efficient mixing.

Nguyen et al [16] have demonstrated another externally assisted mixing technique by actuating ferrofluid droplet using magnetic forces. The ferrofluid consists of magnetic particles of diameter in the order of few nanometers. Movement of droplet is induced by the activation of the planar micro-coils which generate the necessary magnetic field. Likewise, proper control on the movement of droplet will create the required chaotic advection needed to enhance the mixing efficiency.

The use of thermocapillary forces [64, 65, 68, 69] is another alternative to manipulate the droplet. Thermocapillary force is a result of Marangoni effect. Scriven and Sternling [70] explained the Marangoni effect as two associated effects. The first effect is a movement at the fluid interface due to a gradient of interface tension induced by temperature and concentration changes. And the second effect is a non-equilibrium tension at the interface as a result of contraction and expansion of interface. Ostrach [71] pointed out two mode of flows induced by the surface tension gradient. If the gradient is along the interface, it is termed as Marangoni convection. Since the gradient is due to a variation in temperature, composition, and electrical potential, the resulting flow can be termed as thermocapillary, diffusocapillary, or electrocapillary, respectively. These flows can affect transport processes. However, if the gradient is perpendicular to the interface, it will lead to an instability flow termed as Marangoni instability.

Young et al [72] has first demonstrated the thermocapillary motion of bubble in pure liquid. The bubble can be held stationary or driven downward by controlling a negative temperature gradient in the vertical direction. The effect is due to the stresses from the thermally-induced variation of surface tension on the bubble surface which is the outcome of Marangoni effect. Hardy [73] explained that the tangential shear stress at the interface has caused the fluid motion both inside and outside the bubble. Surface tension usually decreases at elevated temperature and fluid flows from hot to cold along the interface. Thus, the Marangoni force experienced by the bubble is opposite the fluid flow.

Jiao et al [68] used thermocapillary forces to generate a reciprocating liquid plug motion in an externally heated capillary. As shown in Figure 2.20, the liquid plug is positioned between two external heaters which are alternately activated by an electronic

control circuitry. This will alternate the direction of temperature gradient and consequently induce a reciprocating motion to the liquid plug under the thermocapillary effects.

Figure 2.20 – Thermocapillary microactuator [68]

Jiao et al [74] have also experiment the use of 4 integrated heaters surrounding a droplet to manipulate the droplet within the planar microchannel. By controlling the heating sequence of the heater array as shown in Figure 2.21, the droplet can be positioned anywhere within the microchannel.

Figure 2.21 – 4 heater array [68]

2.3 Functional fluids

Our definition of functional fluid refers to any special class of fluids with a unique set of properties that can be utilized to achieve the desirable outcome, effect, improved performance or control required of a microfluidic device under specific operating conditions. External forcing mechanisms as described in previous sections can constitute the operating condition. Functional fluids include but not limited to nanofluid, viscoelastic fluid (non-Newtonian), ferrofluid, optically or thermally sensitive fluid etc.

Murshed et al [28] have explored the use of nanofluid as a dispersed phase in cross-flow geometry (T-junction). Nanofluid is engineered by dispersing nanoparticles into conventional fluid such as deionised water (DIW). In their case of nanofluid, titanium oxide nanoparticles are dispersed into DIW. Addition of nanoparticles will have the effect of modifying the interfacial tension and introducing interfacial slip at the fluid interface. They have experimentally investigated the droplet formation process and droplet size control at different temperature. Their investigation showed that nanofluids not only behave differently from deionised water (DIW), the shape of nanoparticles also has significant bearings on the behavior. Nanofluid prepared with spherical shaped nanoparticles has a smaller droplet diameter and changes very little over the temperature range as compared with DIW. The result is totally different for the case of cylindrical shaped nanoparticles. At temperature below 32°C, the droplet size is bigger than the base fluid, DIW. But beyond this temperature, the droplet size decreases with temperature and become smaller than DIW. It is, thus, believed that the droplet formation process occurs at different flow regime during the temperature change. This phenomenon is yet to be thoroughly investigated. However, one key observation behind these observations is that

there exists a class of fluid that can be utilized as a control knob for a droplet-based microfluidic device to attain the desirable droplet size needed to support a specific application. The behavioral complexity associated with the property of the functional fluid in achieving different droplet size can pose a challenge in maintaining a consistency in the performance of the microchannel device. A more controlled technique can be achieved by using thermocapillary effect. This will be discussed later in this dissertation.

In the continuous flow microfluidic device, Gan et al [18] have reported the use of elastic property of viscoelastic fluid to enhance the mixing efficiency of fluid at low Reynolds number. Because of the fluid's elasticity property, the elastic stress experienced by the fluid will not immediately return to zero when fluid motion or externally induced force terminates. Instead, there will be a characteristic decay time. The experiment shows that this unique elastic property has induced flow instability to the flow streams which helps to facilitate mixing. The mixing efficiency is further enhanced by the expansion-flow created by the abrupt convergent/divergent geometry of the microchannel design. In digital droplet-based microfluidics, Zhou et al [37] have numerically simulated the effect of using viscoelastic fluid as the disperse phase in a flow focusing geometry. Comparing to a Newtonian fluid, the viscoelastic phase will give a rise to bigger droplet when working in the dripping regime. On the contrary, smaller droplet is obtained in the jetting regime. The tensile stress associated with the viscoelastic property of the dispersed phase has the effect of delaying the breakup time in the dripping regime and forming a shorter jet in the jetting regime. As compared to Newtonian fluid, the droplet forming process for viscoelastic fluid has different response at different regime and thus, the droplet size is affected correspondingly. It is, however, desirable to have a detail quantitative description of the

viscoelastic effect and supported by experimental measurement. Viscoelastic effects in other passive droplet formation techniques such as cross-flow and co-flow configurations are yet to be investigated too. These investigations can be economically achieved through numerical modeling. To the best of our knowledge, there is limited numerical model developed to study droplet formation let alone the response of viscoelastic effect. Beyond microfluidic applications, it has also been reported that viscoelastic fluid can behave like a transistor switch [41].

Optical properties can make liquids become functional. In the recent years, micro-optofluidic devices have been realized by some researchers [75-77] to handle light optics which are able to function as waveguides, splitters, switches, lens and mirrors. Unlike their solid-phase counterparts which make use of glass, semiconductor materials and metals, optofluidic device uses a liquid system. The optical waveguide designed by Wolfe et al [76] uses liquid-core/liquid-cladding. The refractive index of the liquid is a key selection criterion for the core and cladding liquid. For their device, deionized water with a refractive index of 1.335 and an aqueous solution of 5M CaCl_2 with a refractive index of 1.445 were used as cladding and core liquids respectively. Whereas, in Nguyen et al [75] optofluidic splitter/switching device, glycerol solution (water with 56 w% glycerol) with a refractive index of 1.445 was used as the core liquid. Regardless of the type of liquid used, the optical property of the fluids is a critical parameter to meet the intended application. And thus, these fluids can be classified as optically sensitive functional fluids.

Ferrofluid is another class of functional fluid which is composed of magnetic particles suspended in a liquid. Nguyen et al have reported [16] the use of ferrofluid to enhance the functionality and efficiency of microfluidic systems. The purpose of magnetic

particles is to facilitate the induction of magnetic forces thereby allowing the transportation and manipulation of the fluid for some intended application. The effectiveness of control on the ferrofluid depends on the uniformity of the distribution of magnetic particles within the liquid stream or droplet when subjected to the applied condition. In this aspect, the size of magnetic particles, which can range from micrometers to a few nanometers, has a significant impact. Smaller particles tend to move more randomly within the fluid and thus result in a more uniform distribution under any flow condition.

2.4 Numerical Methods in Multiphase Flow

In the modeling of multiphase flow system, the critical task lies in the tracking of fluid interface. Several methods have been employed by researchers in describing the interface for two phases. They are, but not limited to, volume tracking method, front tracking method, phase field method, fractional volume of fluid (VOF), lattice Boltzmann method (LBM) and Level-set method (LSM). These methods can be categorized in several perspectives:

1. Explicit or implicit tracking method [78]
2. Interface tracking or capturing method [79]
3. Moving or fixed mesh capturing technique [80]

No one single categorization rule can completely describe the method. Each of these describes the characteristic of the method. Level-set method, for example, is described as an implicit, interface capturing method using fixed mesh.

2.4.1 Front Tracking Method

The front tracking method is developed by Glimm [81] and Tryggvason [82]. The method uses connected markers to reconstruct the interface grid which moves through a fixed grid. As [82] illustrated, the computational domain is discretized by a fixed rectangular grid while the interface is described by a separate unstructured triangular grid. Computation of the interface is done on the interface grid and then transferred to the global grid. Incidentally, this multi-grid approach adopted by the front tracking method is complex.

2.4.2 Volume of Fluid Method

For this method, the interface is captured implicitly. A volume fraction field is defined for the entire computational domain where a volume fraction function will be solved on a fixed domain. The volume fraction function is described below:

$$\zeta_t + \vec{u} \bullet \nabla \zeta = 0 \quad (2.3)$$

Where ζ represent the volume fraction and has a value between 0 and 1. A sample distribution of the volume fraction is shown in Figure 2.22

Figure 2.22 Volume fraction value at the interface

If the volume fraction is 1, the computation cell is fully occupied with the primary phase. Whereas, a volume fraction value of 0 represents that the cell is fully occupied by secondary phase. Cell with volume fraction value of $0 < \zeta < 1$ will contain the interface. The values are reconstructed to obtain the interface location. In the reconstruction process, the weighted viscosity and density of the computational cells as well as the volume flux of the convective terms in the governing equations are determined. Some of reported reconstruction methods are Parabolic Reconstruction of Surface Tension (PROST) [83], Simple Line or Piecewise Linear Interface Construction (SLIC) [84] and least-square fit with split Eulerian-Lagrangian advection [85].

A right advection scheme to calculate the movement of the interface under the influence of the flow field is required after the interface reconstruction step. This is the key challenge for VOF method in order to achieve accurate representation of the physical volume fraction distribution and to ensure sharpness of the interface. A higher order scheme can lead to numerical oscillation whereas a lower order scheme will give a smear interface. Some of

the volume advection scheme are flux-corrected transport algorithm [86], Lagrangian piecewise-linear interface reconstruction (L-PLIC) method [87], gamma differencing scheme [88] and compressive interface capturing scheme [89]. Normalized Variable Diagram (NDV) [90] is a high order scheme which is reported recently to have preserved the interface sharpness and have boundedness of the volume fraction.

The key advantage of VOF method over other methods is its superior capability on volume or mass conservation of the fluid. This leads to high accuracy in the calculation of three dimensional flows. It is the engine for Fluent which is a popular commercial CFD (Computational Fluid Dynamic) software.

2.4.3 Phase Field Method

The interface is captured implicitly by the phase field method. For this method, a phase field function $\varphi(\vec{x}, t)$ is implemented to represent the interface. The function has a value of 0 at the interface and a value of +1 or -1 on either side of the phases away from the interface. The advection of the phase field function is governed by Cahn-Hilliard equation [145] which is a 4th order non-linear parabolic diffusion equation described as,

$$\varphi_t + \vec{u} \bullet \nabla \varphi = -\sigma \Delta [\Delta \varphi - f(\varphi)] \quad (2.4)$$

Where $f(\varphi)$ is a polynomial of φ [91]. An alternative governing equation of the phase field is Allen-Cahn [92, 93].

This method requires a fine grid at the interface and can be used to solve three dimensional problems. The phase field method only reproduces the advection of the interface by

mapping between the interface governing equation and phase field equation through asymptotic analysis.

2.4.4 Lattice Boltzmann Method (LBM)

This method is relatively new as compare to the other methods and has gained popularity in the modeling of droplet-based microfluidics [19, 20, 94]. The only difference between this method and the phase field method lies in the discretization of the Navier-Stokes equation [95]. The LBM is a derivation of the lattice gas theory which consists of a regular lattice with particles residing on the nodes. The macroscopic dynamic of the fluid is represented by the behavior of the particles.

2.4.5 Level Set Method

Level-set method (LSM) is an implicit, interface capturing method on a fixed grid domain. LSM is introduced by Osher and Sethian [96]. They employed an algorithm to track the moving interface on a fixed grid. A level-set function $\phi(\vec{x}, t)$ which defined as a signed shortest distance from the interface is implemented either over the computational domain or near the interface. The weighted fluid properties of a computational cell are calculated from the level-set function using a smoothed Heaviside function. Re-initialization equation is implemented to ensure that the level set function adhere to a signed distance function at the interface [97]. The accuracy on the advection of interface can be achieved with higher order scheme through solving the Hamilton-Jacobi equation. This is at the expense of higher computation which is undesirable. To reduce the computational time, a technique to

only solve the level set function within a band near the interface [98]. The main challenge of LSM is mass conservation. Several techniques have been reported [99-101] to address the problem. But Particle Level set method (PLS) is accepted by most researchers to be able to improve mass conservation. Particles are seeded near the interface to follow the advection and then draw from them the characteristic information to reconstruct the interface where level-set method failed to preserve mass [102].

LSM has shown success in the handling of moving interface in multiphase flow in two- or three-dimensional problem [103-105][161, 31, 169, 170, 171, 172, 173, 174]. It is a powerful and relatively easy to implement simulation scheme to study challenging problem such as coalescence or rupturing of droplets. Therefore, LSM has good potential as an important numerical tool in the field of droplet-based microfluidic.

2.5 Potential areas of work

This dissertation will expand further some of the existing works reported in the above literature review. The key areas of work are as follows:

- In the area of droplet sizing, Nguyen et al [40] has reported the use of thermal forcing in their experiment. The thermally induced effect will be further explored and analysed in this dissertation with different heater configurations through numerical simulation.
- Luong [61] has demonstrated the thermocoalescence of droplets in a merging chamber. As compared with the other passive technique, the merging chamber is simpler in structure and thus, easier to implement. Coupled with the use of thermal

forcing, a more consistent performance can be achieved. However, the combined effect of hydrodynamic and thermal forcing remains to be understood. Through the use of numerical simulation, the dissertation will analyse the impact of this effect on the movement of droplet across the merging chamber.

- Methods like VOF, LBM have been adopted by researchers to model droplet formation and control in microchannel. The work presented in this dissertation makes use of LSM to model droplet formation and the movement of the droplet in a microchannel device. The studies are also expanded to a thermal forcing environment and thermocoalescence of the droplet in a merging chamber where currently there exists limited research in this area. Another unique feature in the present numerical model is the use of high order WENO for interfacial advection to optimize and improve the accuracy of the numerical results. Commercial package is definitely limited in this aspect. The use of simulation will help to gain insights into the area within the microchannel which is otherwise difficult to reach out in an experimental setup. This will serve to provide more understanding on the effect of thermal forcing on the formation and coalescence of droplets.

Chapter 3 Numerical Modeling

This dissertation shall focus on the numerical modeling of droplet formation in a cross-flow (T-junction) channel design as shown in Figure 3.1. L is a variable used in the simulation program to represent the baselength for a specific microchannel geometry. In this dissertation, the typical value for L is $50\mu\text{m}$.

Figure 3.1 – Geometry of T-junction design with thermal forcing.

The carrier or continuous phase flows along the main channel and the dispersed phase is fed through an inlet perpendicular to the main channel. At the location of the intersection between the two channels where the droplet formation process begins, a heater is attached to the channel wall to provide a heat flux with the purpose to modify the temperature dependent properties of the phases. As explained in the previous session, the

temperature dependent properties will have bearings on the droplet size if they are able to be controlled via some form of thermal forcing unit. In this case, the thermal forcing unit comprises of the heater and a sensor to provide the feedback control.

The droplet formation process is treated as a flow interaction between the two incompressible immiscible phases. The general solution approach is to discretise the domain of interest by finite volume method and to solve the conservation equations at each individual control volume. As the motion of the two phases is closely coupled via a related interfacial condition, the interfacial boundary between the two phases will have to be determined in order for the respective phases to be spatially identified. Under the flow conditions of the two phases, this interfacial boundary is changing with time and space. To mitigate the abrupt transition between the phases right at the boundary, the domain is considered to be filled with a special fluid whose properties are a combinational effect of the two phases and is spatially dependent in reference to the changing interfacial boundary. Conservation equations are solved accordingly using the special fluid properties. Details will be discussed in the subsequent sessions.

3.1 Mathematical Formulation

The droplet formation process can be treated as a single phase flow problem of a special fluid even though there are two incompressible immiscible phases. The conservation of mass, momentum and energy of this special fluid are used to describe the two-phase microfluidic system and are formulated mathematically as:

$$\frac{\partial \rho}{\partial t} + \nabla \bullet (\rho \vec{u}) = 0 \quad (3.1)$$

$$\frac{\partial(\rho\vec{u})}{\partial t} + \nabla \bullet (\rho\vec{u}\vec{u}) = -\nabla p + \nabla \bullet \left[\mu (\nabla\vec{u} + \nabla\vec{u}^T) \right] + \vec{f}_F \quad (3.2)$$

$$\frac{\partial(\rho c_p T)}{\partial t} + \nabla \bullet (\rho c_p \vec{u} T) = \nabla \bullet (k \nabla T) \quad (3.3)$$

where ρ , μ , c and k are the density, the dynamic viscosity, the specific heat and the thermal conductivity, respectively.

Note that an external forcing term is included into the conservation of momentum equation. This forcing function is due to the interfacial force present on the fluid interface [106]. The interfacial force is due to surface tension force and it consists of two components acting on the surface. They are the tangential and normal forces as depicted by the first and second term on the right hand side of the following expression,

$$\vec{f}_F = -\kappa \hat{N}_F D(\vec{x} - \vec{x}_F) + (\hat{N}_F \times \nabla \sigma) \times \hat{N}_F D(\vec{x} - \vec{x}_F) \quad (3.4a)$$

where $D(\vec{x} - \vec{x}_F)$, $\hat{N}_F$ and κ are the dirac-delta function, interface unit normal and curvature respectively and are defined as

$$D(\vec{x} - \vec{x}_F) = D(\phi) = \begin{cases} \frac{1 + \cos(\pi(\phi)/\varepsilon)}{2\varepsilon} & |(\phi)| < \varepsilon \\ 0 & \text{otherwise} \end{cases} \quad (3.4b)$$

$$\hat{N}_F = \frac{\nabla(\phi)}{|\nabla(\phi)|} \quad (3.4c)$$

$$\kappa = \nabla \bullet \hat{N}_F \quad (3.4d)$$

Eqn. (3.4a) account for the effect of interfacial force induced in a temperature mediated environment. .

As mentioned earlier, the T-junction has a heater located at the intersection of the main and inlet channel. The heating effect of the heater is modeled by the equation of conservation of energy as shown in Eqn 3.3. The error criterion for temperature is such that, the summation of temperature difference between adjacent iteration for all the control volume shall be less than 1×10^{-3} . The energy equation will provide the temperature field distribution in the vicinity of the channel junction. The resulting temperature gradient will have the effect of modifying the viscosity of the fluid which is sensitive to temperature. The change in viscosity will have a bearing on the solution of the equations of conservation of mass and momentum and thus the flow field will change accordingly. This change will have underlying effects on the droplet formation process. The droplet size and rate of droplet formation are expected to vary accordingly and is the focal point of this numerical analysis.

At any particular location within the domain of interest, the properties of special fluid can assume that of -ve phase or +ve phase or a weighted average of both. Determining the interfacial boundary between the phases will be the first step in deciding the appropriate set of fluid properties for the special fluid to be used when solving the conservation equations. The relative normal distance of point of interest from the interfacial boundary will then be computed. This normal distance is signed to indicate on which side of the interface the point is located. If the point is exactly on the interface, the distance will be zero.

As shown in Figure 3.2, the domain of interest, Ω , consists of the -ve and the +ve fluid separated by the interface, Γ_i .

Figure 3.2 – Domain of interest separated by interface, Γ_i

Osher and Sethian [96] have devised a numerical procedure known as level-set method to represent the interface, Γ , and can be represented mathematically as,

$$\phi = \begin{cases} -d, & \vec{x} \in \Omega_- \\ 0, & \vec{x} \in \Gamma_i \\ +d, & \vec{x} \in \Omega_+ \end{cases} \quad (3.5)$$

where d is the normal distance from the interface, Γ_i . When the distance is negative, that particular point is located in the $-ve$ domain which contains $-ve$ fluid. The special fluid will thus assume the fluid properties that of the $-ve$ fluid. Likewise if the distance is $+ve$, the fluid properties of the special fluid will be that of the $+ve$ fluid. To alleviate the abrupt transition from one phase to another across the interface, a smoothing Heaviside function H , shown in Eqn 3.6, is applied for a given thickness of the interface. This finite thickness, ϵ , is related to the spatial mesh grid size and usually taken as a factor of the grid spacing and is set to be 1.5 times of the grid size.

$$H(\phi) = \begin{cases} 0, & \phi < -\varepsilon \\ \frac{\phi + \varepsilon}{2\varepsilon} + \frac{1}{2\pi} \sin\left(\frac{\pi\phi}{\varepsilon}\right), & |\phi| \leq \varepsilon \\ 1, & \phi > +\varepsilon \end{cases} \quad (3.6)$$

With the understanding of the distance function from the interface, Γ_i , expressed using the level-set method and coupled with a smoothing function, H , the properties of the special fluid can be calculated using,

$$\alpha(\phi) = (1 - H)\alpha_- + H\alpha_+ \quad (3.7)$$

At this point, it is important to note that the interface, Γ_i , evolves with time and velocity component of the flow. This relationship is investigated by Sussman [107] and it is governed by

$$\phi_t + \vec{u} \bullet \nabla \phi = 0 \quad (3.8)$$

As Eqn. 3.8 shows, the interfacial boundary between the two phases – moves with the flow velocity. Depending on the magnitude of differences in the flow condition of both phases which are present on the opposite sides of the interface, the interface function, ϕ , will lose its distance property and become irregular after some time making it no longer able to satisfy the condition as $|\nabla \phi| = 1$. To maintain ϕ as a distance function, it required to re-initialize ϕ and may take some iterations at each time step in order for the solution to be accurate. But such re-initialization will run into risk of amplifying any distortion seen due to computational error. A method has been devised by Sussman et al [97, 107] to re-initialise ϕ without the need to find the actual interface and thus help to minimize the risk of distortion. This is achieved by constructing a distance function, ϕ , which has a zero level set at ϕ_0 and still maintained as a signed normal distance to the interface. The distance

function can be obtained by solving the following partial differential equation and obtain the steady state solution,

$$\frac{\partial \phi'}{\partial \bar{t}} = \overline{\text{sign}}(\phi)(1 - |\nabla \phi'|) \quad (3.9a)$$

where $\bar{t}$ is a pseudo time for ϕ' and $\overline{\text{sign}}(\phi)$ is given by

$$\overline{\text{sign}}(\phi) = \frac{\phi}{\sqrt{\phi^2 + |\nabla \phi|^2 (\Delta x^2)}} \quad (3.9b)$$

and is subjected to the following initial condition.

$$\phi'(\vec{x}, 0) = \phi(\vec{x}) \quad (3.9c)$$

Sussman has proven that the method can converge to $|\nabla \phi'| = 1$ outside the interface and can provide the actual distance. This approach is numerically efficient as it takes only one iteration per time step for the computation to reach convergence criteria.

The level set method described above is simple to implement even for complex interfaces. However, one of its shortfalls is the loss of mass due to cumulative numerical error from the repeated/cyclic computational procedure. A number of mass loss correction methods have been explored by researchers to resolve this issue. Chang et al [100] included a mass conservation term into the level-set method. Yap et al [99, 108, 109] developed a local mass conservation scheme and later extended to a global mass correction scheme to ensure mass conservation in level set method. Son [110] has presented a coupled LSET-VOF approach to the preservation of mass. In this dissertation, a particle level set method [102] is used to handle the mass eroding issue. This method supplements the level set method with a marker particle scheme. Particles are placed randomly near the zero level

set of interface at a density of 4 particles per control volume per spatial variable. Each control volume near the interface will have 64 particles as shown in Fig 3.3 (b). These particles will follow according to the characteristic of the flow pattern and, under a normal condition, they are not supposed to cross the interface. In the event a particle is detected on the opposite side of the interface when under a flow condition, an error is said to be present in the level set function. By referring to the distance information of the escaped particle, this error is corrected and the level set function at the particular point of deviation will be rebuilt locally. Where necessary, the particle scheme will play a secondary role in providing sub-grid accuracy to the main interface defining level set function. This will reduce the tendency of the mass loss in under-resolved areas.

Massless particles of size in the range of $0.1\Delta x \leq r_p \leq 0.5\Delta x$, where r_p and Δx are particle radius and grid spacing respectively, are seeded randomly near the zero level set and on either sides of the interface but not necessarily the entire domain. The band of interest is ± 3 times the maximum of the grid spacing ($\Delta x, \Delta y, \Delta z$) from the zero level set. Positive particles are those in $\phi > 0$ phase whereas negative particles are in $\phi < 0$ phase. As shown in Figure 3.3(a), the size of the particles needs to be adjusted so that its boundary does not cross the interface and will be, at the maximum, tangential to the interface. As such, the radius of each particle will be set to,

$$r_p = \begin{cases} r_{\max} & \text{if } s_p\phi(\vec{x}_p) > r_{\max} \\ s_p\phi(\vec{x}_p) & \text{if } r_{\min} \leq s_p\phi(\vec{x}_p) \leq r_{\max} \\ r_{\min} & \text{if } s_p\phi(\vec{x}_p) < r_{\min} \end{cases} \quad (3.10)$$

where $s_p\phi(\vec{x}_p)$ is the sphere of radius centered at particle location, $\vec{x}_p$, $r_{\min}$ is the lower limit of the particle radius and $r_{\max}$ is the upper limit of the radius.

The particles motion will evolve with the flow according to the below expression,

$$\frac{d\vec{x}_p}{dt} = \vec{u}(\vec{x}_p) \quad (3.11)$$

$\phi(\vec{x}_p)$ and $\vec{u}(\vec{x}_p)$ are evaluated by trilinear interpolation of their respective values on the neighbouring nodes as depicted in Figure 3.3(b).

Figure 3.3 – a) Correction of particle radius b) Particle seeding within CV

During the advection process, particles that cross the interface by a distance more than its radius are referred to as escaped particles. Escaped positive and negative particles are denoted as E^+ and E^- respectively. These escaped particles will be used to reconstruct

the interface. To be specific, E^+ will rebuild the $\phi > 0$ region and E^- will rebuild the $\phi \leq 0$ region. The spherical boundary surrounding a particle can be regarded as locally defined level set function given as,

$$\phi_p(\vec{x}) = s_p \left(r_p - |\vec{x} - \vec{x}_p| \right) \quad (3.12)$$

There will be a ϕ_p value for each of the eight grid points of the control volume containing the particle as depicted in Figure 3.3(b). Each ϕ_p is compared to the corresponding local value of ϕ and the maximum of the two values will be taken as ϕ^+ or ϕ^- depending on the particular particle set. With this step, both ϕ^+ and ϕ^- will be initialized using ϕ and will then subsequently be updated via

$$\phi^+ = \begin{cases} \max_{\forall p \in E^+} (|\phi^+|, |\phi_p|), \phi^+ > 0 \\ -\min_{\forall p \in E^+} (|\phi^+|, |\phi_p|), \phi^+ < 0 \end{cases} \quad (3.13a)$$

$$\phi^- = \begin{cases} \min_{\forall p \in E^-} (|\phi^-|, |\phi_p|), \phi^- > 0 \\ -\max_{\forall p \in E^-} (|\phi^-|, |\phi_p|), \phi^- < 0 \end{cases} \quad (3.13b)$$

where

$$\phi_p = \text{sign}[\phi(\vec{x}_p)] (r_p - |\vec{x} - \vec{x}_p|) \quad (3.13c)$$

The particle correction for the level set function ϕ is finally performed via

$$\phi = \begin{cases} \phi^+, & \text{if } |\phi^+| \leq |\phi^-| \\ \phi^-, & \text{if } |\phi^+| > |\phi^-| \end{cases} \quad (3.14)$$

Once the level set function can be accurately implemented, the information will be used to determine the fluid properties of the special fluid at any particular grid point in the domain. The appropriate fluid properties will then be used to compute the solution to the equation of conservation of mass (Eqn.3.1), momentum (Eqn. 3.2) and energy (Eqn. 3.3).

3.2 Boundary Conditions

Fully developed flow as shown by the profile in Figure 3.1 is assumed at inlets. In a fully developed flow, the velocity profile and the interface become independent of the axial coordinate. The velocity profile in the x-direction of a fully developed flow in a rectangular channel of width, W and height, H , is given below [19]:

$$u_x(y, z) = v_o \left[1 - \left(\frac{z}{H/2} \right)^2 + 4 \sum_{k=1}^{\infty} \frac{(-1)^k}{\alpha_k^3} \frac{\cosh\left(\frac{\alpha_k y}{H/2}\right)}{\cosh\left(\frac{\alpha_k W/2}{H/2}\right)} \cos\left(\frac{\alpha_k z}{H/2}\right) \right] \quad (3.15)$$

where

$$v_o = \frac{(H)^2 \Delta P}{8\mu L} \quad \text{and} \quad \alpha_k = (2k - 1) \frac{\pi}{2}, \quad k = 1, 2, \dots$$

In the model, a velocity profile at the inlet is initialised using Eqn 3.15. The initialization program is run for a few time cycles to evolve the fluid interface prior to the actual simulation. Outflow boundary condition is assigned for the outlet. Non-slip flow condition is also assumed at the channel wall. A layer of oil always exists between the droplet and the channel wall. Therefore, the droplet will not come into direct contact with the wall. Thus, there is no moving three-phase contact line. Based on this assumption, the contact angle does not need be taken into account in our problem.

The fluid temperatures at both inlets are set to T_o . As shown in Figure 3.1, the temperature field is induced into the device by a heater attached to the wall. The boundary conditions are set such that the temperatures of the locations on the wall directly adjacent to the heater are set to T_{set} . Other locations of the wall which are outside the heater are maintained at T_o . At the outlet, a zero temperature gradient condition is employed. With these boundary conditions, a non-uniform temperature field is established in the channel and changes the properties of both fluids.

3.3 Solution Procedure

By employing the SIMPLER algorithm [111], the conservation equations (Eqn. 3.1 – 3.3) are solved to obtain the pressure and velocity components of the special fluid flow.

In the SIMPLER algorithm, the discretised momentum equation will have the form

$$a_{i,j} \vec{u}_{i,j}^* = \sum a_{nb} \vec{u}_{nb}^* + p^* A_{i,j} + b_{i,j} \quad (3.16)$$

where coefficients, $a_{i,j}$ and a_{nb} contain a combination of advective flux and diffusive conductance at the control volume cell face predicted by the Power Law and $b_{i,j}$ is the momentum source term which contains the interfacial force.

The equation is solved using pressure field, p^* , to yield the velocity components $\vec{u}^*$. It starts by using an initial guessed value of p^* and $\vec{u}^*$. Iteration will continue until the source, b , becomes sufficiently small everywhere in the discretised domain. The source term, b , is the corrective factor of the guessed pressure for the each subsequent iteration. For convergence, the value of mass source term will practically become zero for all the control volumes. The criteria for convergence are

1. The maximum value of the source term for all the control volume is less than 1×10^{-12} ,

and

2. The summation of source term for all the control volume is less than 1.

The level-set function and re-distancing procedure are implemented using the PDE-based fast local set method [98]. Both functions are spatially discretized with WENOS [112] and advected using TVD-RK2 [113]. It has been reported in [107] that WENO5 scheme has a gain in accuracy which has helped to reduce long iteration time. Internally in our simulation program, we use a mass loss index to monitor the health status of the numerical solution. The requirement is not more than 10% mass loss. The mass loss index is described as below

$$\Delta M = \frac{M_c}{M_d} \quad (3.17)$$

where M_d and M_c are the desired mass and the most current mass of the reference phase respectively. The values are computed purely based on the volume flow rate of fluid and the calculated mass of the fluid from the numerical results of each iterations cycle.

From the boundary and initial conditions, ϕ^n , $\bar{u}^n$, p^n and T^n are known. Their corresponding value, ϕ^{n+1} , $\bar{u}^{n+1}$, p^{n+1} and T^{n+1} , for the next time step is obtained with the following procedure:

- (1) Set $\bar{u}^{n+1} = \bar{u}^n$
- (2) Compute level set function, ϕ^{n+1} from Eqn 3.8 using a narrow band procedure[98].

- (3) Perform redistancing procedure on ϕ^{n+1} using function ϕ' from Eqns. 3.9
- (4) Perform particle correction on the level set solution in the previous step using Eqns. 3.10 – 3.14 to obtain the final ϕ^{n+1} solution.
- (5) Calculate the properties of the special fluid with the information from ϕ^{n+1}
- (6) Solve the conservation equations of mass and momentum Eqns 3.1-2 using the SIMPLER algorithm to obtain $\vec{u}^{n+1}$ and p^{n+1}
- (7) Solve the conservation equation of energy, Eqn 3.3, to obtain T^{n+1}
- (8) Repeat step (2) to (7) until solution converges

3.4 Solution Validation

The purpose of validation is to show,

- (i) The accuracy of present numerical approach for use as a simulation tool in the study of droplet formation, and
- (ii) The expected response and outcome of the model to changes in flow parameters or fluid properties induced by external means.

Validation is performed using a flow focusing configuration i.e. cross-junction geometry. The size of the domain is made as small as possible due the daunting requirement on computer resources – computational power as well as memory size – for the simulation of a three dimensional flow problem of such nature.

(a) Experimental[20] (b) Numerical[20] (c) Present

Figure 3.4 - Droplet formation in a cross-junction.

The numerical solution from the present approach is compared to that of experiments and Lattice Boltzmann simulation reported in [20]. As shown in Figure 3.4, except for a slightly smaller droplet diameter in the present solution, there is a reasonable agreement in the results.

3.4.1 Evaluating the Solution against Mass Loss

As described earlier, the solution is subjected to mass loss phenomenon due to accumulative buildup of small numerical errors over the numerous computational cycles. And, in the present solution, the mass correction scheme used is particle level-set (PLS). The mass loss phenomena is evaluated with and without PLS. Solution without PLS is obtained from pure level set method (LS). Figure 3.5 shows the results of mass loss ratio with LS and PLS over time at $T_{set}^* = 0$.

Figure 3.5 - Mass losses of the +ve fluid (LS: level-set method only, PLS: particle level-set method)

The mass loss ratio is defined as M_+/M_{+0} where M_{+0} is the correct mass and M_+ is the current mass at the specific time step in the computation. For a perfect mass conservation, the mass loss ratio shall be unity i.e. $M_+/M_{+0} = 1$. As shown in Figure 3.5, without mass correction scheme as in LS, the mass loss is ~17% at the end of the time period of interest. This mass loss is expected to trend down further if the time period is extended. However, with PLS, the mass loss is maintaining around 3% for the same period with no obvious trend of degradation for any extended time frame.

Though the result is not presented in this dissertation, similar trend can also be observed with thermal forcing. The mass loss phenomenon for the case of zero thermal forcing is, however, sufficient to illustrate the effectiveness of the mass loss correction scheme (PLS) used in the present solution.

3.4.2 Sensitivity of Solution to Mesh Density and Time Granularity

The settings of the particle related parameters are validated prior to the start of the actual numerical investigation. This is carried out by simulating a spherical water droplet flowing in a square channel of $50\mu\text{m} \times 50\mu\text{m} \times 250\mu\text{m}$ with the same hydrodynamic flow condition as the intended study - Figure 3.6. The hydrodynamic flow condition is governed by 7 dimensionless numbers: the Reynolds number $\text{Re}_0 = \rho u_{-0} L / \mu_{-0}$, the capillary number $\text{Ca}_0 = u_{-0} \mu_{-0} / \sigma_{-0}$, the Prandtl number $\text{Pr}_0 = \mu_{-0} / a_{-0}$, the dimensionless heater temperature $T_{\text{set}}^* = (T_{\text{set}} - T_0) / T_0$, the viscosity ratio $\mu^* = \mu_{+0} / \mu_{-0}$, the flow rate ratio $u^* = u_{+0} / u_{-0}$ and the ratio of thermal diffusivities $a^* = a_{+0} / a_{-0}$. L is the base length. The conditions are $\text{Re}_0 = 4.63 \times 10^{-4}$, $\text{Ca}_0 = 3.65 \times 10^{-2}$, $\text{Pr}_0 = 1.02 \times 10^3$, $\mu^* = 8.33 \times 10^{-2}$, $u^* = 1.0$ and $a^* = 1.21$.

Figure 3.6 - Droplet flow model

As mass conservation is a critical index to the success of a PLS numerical model, the validation is based on the mass loss performance. Two different mesh setups are evaluated – coarse and fine meshes. A coarse mesh will have 12k nodes for the modeled domain and a fine mesh will have 81k nodes. For each of the mesh setup, different settings are tested: 1) particle re-initialisation at post advection of the interface versus at both pre- and post-advection 2) particle density of 64 particles per CV versus 32 particles and 3) seeding interval of 200 time steps versus 100. The baseline settings are re-initialisation at post-advection, 64 particles per CV and 200 time step seeding interval. Figure 3.7 and 3.8 shows the mass loss of the droplet over time for coarse and fine meshes respectively at different settings.

Figure 3.7 - Impact of various PLS settings on a Coarse mesh setup

In the case of coarse mesh (Figure 3.7), the mass of droplet for the baseline setting is eroding over time and has reached 0.9 of the exact mass before 700 time steps. As

compared to the baseline setting, similar trend on the degradation of mass loss for the other PLS settings can also be observed (i.e. pre- and post-advection particle re-initialization scheme, 100 time steps seeding interval and 32 per CV particle density). The worst among them is the seeding density 32 particles per CV. If the flow condition is isolated, as in the case of the static flow, the mass loss phenomenon is less drastic. However, a significant mass loss still happens eventually after 800 time steps. To a certain extent, this illustrates the impact of flow condition and would mean that the optimal grid for any domain is dependent on the flow condition.

Figure 3.8 - Impact of various PLS settings on a Fine mesh setup

For the case of fine mesh (Figure. 3.8), tremendous improvement can be seen in the mass loss performance as compared to coarse grid. The fine mesh has also reduced the sensitivity of the various PLS settings. Similar to the case of coarse mesh, the mass loss performance is most sensitive to particle density although the impact is not considered to pose any major concern as it has shown to reach stability. Based on the results from fine

mesh, the baseline condition is, thus, validated to be acceptable for the present study. Under a no flow condition, the static case shows a superior performance in term of the mass loss phenomena. This has further confirmed the effect of flow condition.

Considering the computational burden which increase exponentially with mesh density, a moderate mesh of 36k nodes is selected for evaluation. Based on the mass loss performance index as shown in Figure 3.9, the moderate mesh is acceptable for the purpose of qualitative investigation that is required of the present study as compared to the fine and coarse mesh discussed earlier. At this point, it can be noted that there is no absolute one-size-fits-all mesh setup. It is specific to the hydrodynamic flow condition employed in the numerical simulation and with mass loss performance as the backdrop.

Figure 3.9 - Mass loss performance of different mesh

Chapter 4 Thermally mediated droplet formation

This chapter reports the experimental setup, fabrication of the device under experiment and the numerical result of thermally mediated droplet formation at a microfluidic T-junction. The temperature field generated by an integrated heater causes changes in properties of the fluids and affects the droplet formation process.

The first objective of this chapter is to understand the formation of water droplet in oil. Various forces acting on the droplet will be discussed.

The second objective is to investigate the effect of thermal forcing on the droplet formation. The effectiveness of the penetration of temperature field induced by different heater geometries that resulted in different incremental change in droplet size over a temperature range will be discussed.

4.1 Problem Description

The geometry of the T-junction is shown in Figure 3.1. The channel dimension is the same as that of the $30\mu\text{m}$ depth channel reported in the experiment [28]. The $-ve$ fluid and the $+ve$ fluid represent mineral oil and water respectively. The $-ve$ fluid flows in the main channel with a velocity of u_{-o} and the $+ve$ fluid enters through the inlet channel with a velocity of u_{+o} . The main channel is filled with continuously flowing mineral oil as the forming droplet develops. A layer of oil is always present between the droplet and the channel wall. Therefore, the droplet will not come into direct contact with the wall and thus, the absence of moving contact line. Based on this assumption, contact angle will not be taken into account in our problem. Both fluids are maintained at a temperature of T_o . The heater, located at the intersection of the main and inlet channel, can be controlled to

maintain at a temperature of T_{set} . The value of T_{set} is usually above T_o . As the heat source is localized, it will generate a non-uniform distribution of heat field in the channel with a corresponding influence on the fluid property. This will have a bearing on the droplet formation process and thus, acts as additional forces.

4.2 Material and Experimental Setup

4.2.1 Device Fabrication

The T-junction microchannel device was designed in accordance to the geometry outlined in Figure 3.1. The necessary aspect ratios of the width and depth of the main and inlet channel were maintained. In order to minimize the surface effects of the channel wall on the fluid flows, the device was fabricated using micromachining of glass and polydimethylsiloxane (PDMS). This was to take advantage of the better surface finish as compared to laser machining of polymethyl-methacrylate (PMMA) [114, 115].

4.2.1.1 Design and fabrication of microchannel device

To kick-start the fabrication process of microchannel network using soft lithography technique on PDMS, the master mold was, first, fabricated. A silicon substrate was coated with photoresist made of SU-8 (Figure 4.1a) to a thickness of 30 μ m which is depth of the microchannel network. This was followed by photo lithography of the thick-film SU-8 photoresist using a mask (Figure 4.1b). The mask carried the microchannel network design and was made of low cost transparency. After the SU-8 is exposed, the master mold was formed by developing and removing the unexposed portion

Figure 4.1- Fabrication process of PDMS microchannel device

of the material since SU-8 was a negative photoresist (Figure 4. 1c). The next step was to fabricate the microchannel network from the master mold. The material for the microchannel was prepared by mixing silicon elastomer base and curing agent in a 10:1 weight/weight (w/w) ratio. To prevent bubbles or voids from being formed within the base material as a result of out-gassing, the mixture was left to de-gas in a vacuum flask for 1 hour prior to transferring the mixture into a Petri dish containing the SU-8 master mold

(Figure 4.1d). The mixture was then allowed to oven cure for 2 hours at 80°C. After curing, the microchannel network was obtained by removing it from the SU-8 mold (Figure 4.1e).

Inlets for the fluids into the microchannel network were formed by creating microfluidic accesses using Harris uni-core hole puncher of 0.75mm diameter.

The microchannel was then thoroughly cleaned by soaking in Isopropanol (IPA) for 15 minutes followed by rinsing with distilled (DI) water to prevent the accumulation of dust and dirt on the surfaces. Any residual traces of Isopropanol had to be removed from the surfaces to ensure no bond failure in the subsequent bonding process because Isopropanol has a bond-deterrent effect. This was done by subjecting the microchannel under high pressure nitrogen gas (N₂). The microchannel was then placed in an oven at 150°C for 30 minutes to dry and to ensure the channel material is hydrophobic in preparation for the next process step - bonding.

The microchannel network was encapsulated by bonding it to a thin layer of pre-prepared PDMS (Figure 4.1f). Surface preparation on the bonding surfaces were carried out using oxygen plasma treatment at 70(W), 60(mTorr) and 50(s). The oxygen plasma treatment will have a side effect of temporarily changing the material properties to hydrophilic. After the surface treatment, the alignment between the microchannel network and the thin PDMS layer was done manually. The encapsulated microchannel network was finally left in the oven at 150°C for 2 hours to ensure good bonding. The baking process will also assist hydrophobic recovery of the material.

4.2.1.2 Design and fabrication of microheater and sensor

A low cost transparency mask containing the design of the microheater and sensor was, first, prepared. By using a lithography process, the mask design was transferred and patterned onto a glass wafer coated with a layer of photoresist (Figure 4.2a and 4.2b). The required pattern was obtained by developing away the unexposed photoresist (Figure 4.2c). The

Figure 4.2 - Fabrication process of microheater and sensor.

After the development process, a layer of titanium of 500 Å thickness was deposited onto the glass wafer followed by a platinum layer of thickness 1000Å (Figure 4.2d). The titanium acted as an adhesion layer between glass and platinum. The microheater and sensor was finally obtained by removing the excess metal using a lift-off process (Figure 4.2e).

The microheater and sensor will, next, be integrated to the microchannel device prepared in section 4.1. Prior to the integration process, the glass wafer will be diced into chip size of 1cm x 1cm in order to fit the microheater and sensor within the land estate of the T-junction microchannel device. After dicing, the condition of the microheater and sensor was tested by measuring their resistance using a handheld multimeter. If the thermal forcing chip is in good condition, there will be a finite resistance reading. And when the chip is subjected to temperature change e.g. touching it with finger, a resistance change can be observed. The purpose of the check tests is to pick proper functioning thermal forcing chip to be used for integration with the microchannel network.

The chips will then be thoroughly cleaned by soaking in Isopropanol (IPA) for 30 minutes followed by rinsing with deionized (DI) water and drying under high pressure nitrogen gas (N₂). This was to make sure any dirt and contaminant left over from the dicing process were thoroughly removed and the chips are free of isopropanol as IPA is a bond deterrent agent.

4.2.1.3 Integration of microchannel device with thermal forcing chip

To start off the bonding process, the microchannel device and thermal forcing chip were, first, soaked in isopropanol (IPA) for 30 minutes. Both were then dried with high pressure nitrogen gas (N_2) and oven baked at 150°C for 1 hour. The bonding surfaces of both devices were surface treated with oxygen plasma treatment at 70(W), 60(mTorr) and 50(s) (Figure 4.3a). The devices were manually aligned under a microscope in a clean room environment (Figure 4.3b). Care and precautions have to be taken during the manual handling of both devices to avoid contaminating the bonding surfaces by accidental contact with hand. Any contamination will result in inadequate bonding. Once aligned and integrated, the devices were placed in the oven at 150°C for 2 hours to ensure a good bonding result between the surfaces (Figure 4.3c).

After the bonding process, the final step was to make electrical connection to the microheater and sensor of the integrated device (Figure 4.3d) so that the microheater and sensor can be connected to a direct current (DC) power supply and digital multimeter respectively during the experiment. The electrical contacts were made by attaching wires to the contact pads of the microheater and sensor using conductive epoxy compound from RS component. A 24 hour curing time for the epoxy compound at room temperature is necessary to ensure good contact between the wire and contact pad.

After the curing process, the electrical connections were tested by taking measurement of the resistance. Any abnormality in the resistance reading, for example, zero, infinity or fluctuating value was a tell-tale sign of poor electrical contact and the electrical connection process had to be repeated. A stable resistance reading will indicate that the integrated device is ready to support the experiment.

Figure 4.3 - Integration process of Microchannel device and Thermal forcing chip.

4.2.2 Experimental Setup

The experimental setup is shown in Figure 4.4a. There were four components: precision syringe pumps, DC power supply, digital multimeter and inverted microscope.

The precision syringe pumps (KDS200 from KD Scientific Inc, USA) were used to deliver the carrier and dispersed phase into the T-junction droplet forming microchannel device. In the present study, the carrier phase and dispersed phase were oil and water

respectively. The flow rates of the fluid can be adjusted appropriately by the syringe pumps.

The DC power supply provided the microheater with an appropriate current by changing the voltage. This will allow the microheater to act as a localized heat source in providing the required temperature effect to the droplet formation process at the T-junction.

The heater temperature was monitored using the sensor which was connected to the digital multimeter to measure its resistance value. As the heater temperature increased, the resistance value recorded from the sensor will also increase correspondingly. With the help of an external thermometer, the sensor can be calibrated by determining its corresponding resistance value at various heater temperatures which can be achieved by changing the heater supply voltage. Once calibrated, the sensor resistance value will, thus, indicate the actual temperature of the thermal forcing device. The calibration curve is shown in Figure 4.4b.

The inverted microscope provided the functions of magnifying the droplet formation process and capturing pictures of the water droplet for post-processing works like measurement of droplet diameter, identifying different regimes of droplet formation.

Figure 4.4 – a) Experimental setup b) Calibration curve for heater sensor

4.2.2.1 Imaging technique

The imaging system (Figure 4.5) consisted of illumination system, CCD camera and control system.

Figure 4.5 - Block diagram of fluorescence microscope

The control system was made up of a desktop computer with Flow Manager System hub and corresponding software programs. All imaging related functions e.g. illumination, recording and post-analysis of images, are coordinated and managed by the control system.

The optical system used is Nikon, ECLIPSE TE2000-S fluorescence microscope with the components listed in Table 4.1.

Light source	Mercury lamp
Epi-fluorescent attachment	Nikon, B-2A
Excitation filter	450-490 nm
Dichroic mirror	505 nm
Emission filter	520 nm

Table 4.1 - Parameters of the optical setup

To perform image recording for the purpose of post-processing of image, an interline transfer CCD camera (NISENSE MKII) was used to capture the emitted light output from the fluorescence microscope. The CCD camera captures the image with a resolution of 1344 x 1024 pixels with 12 bit greyscale. In the present study, green fluorescent dye of fluorescent band 590/520nm was added to the dispersed fluid. Images of the fluid flows in the microchannel device were then captured by epi-flourescent attachments of the microscope with a green filter of 25nm bandwidth. The digitally recorded images were transferred to the desktop computer for analysis using a customized MATLAB program.

4.2.2.2 MATLAB program

Post-analysis on the image was carried using a customized MATLAB program. The program was used to calculate the size of the droplets formed. The image recorded was read by the program and the droplet diameter is approximated by the number of pixels within a sphere. The program applied a conversion ratio that relates the number of pixels to

length scale in micrometers. The droplet diameter, D , was calculated by $D = \alpha \sqrt{\frac{4A}{\pi}}$

where α is the conversion ratio and A is the measured area in pixel.

4.2.2.3 Fluid preparation and characterization

Two different fluids, carrier and dispersed, were prepared for the experiment. The dispersed fluid was DI water and was mixed with 0.05% w/w green fluorescence dye (Sigma F6377). Mineral oil (Sigma M5904) was used as the carrier fluid and was mixed with 2% w/w Span 80 surfactant (Sigma S6760). The surfactant had an effect of reducing the interfacial tension between oil and water. This will enhance the ease of water droplet formation in the oil for being the carrier fluid. By reducing the interfacial tension force between water and oil, the droplet has a greater freedom of movement within the carrier fluid especially so under a thermocapillary force, if present [116]. Incidentally, the reduced interfacial tension also resulted in larger deviation of the droplet shape from that of a spherical formation.

As described in the previous sections, the temperature dependency of the fluid properties, viscosities and interfacial tension, can affect the droplet formation process under different temperature environment. Prior to the experiment, these fluid properties for both the carrier and dispersed fluid were characterized over a temperature range of 25°C to 39°C. The fluid viscosity was measured using a low-shear rheometer (LS40, Mettler Toledo, Switzerland) and the interfacial tension was measured with a tensiometer system (FTA 200, First Ten Angstroms, USA). The measured values were normalized using their nominal values at 25°C i.e. $\eta^* = \eta(T)/\eta(T = 25^\circ\text{C})$ and $\sigma^* = \sigma(T)/\sigma(T = 25^\circ\text{C})$. The

trending of their values over the temperature range was used to support the experimental investigation on the effect of temperature on droplet formation.

For use in numerical analysis, the interfacial and viscosities were normalized with their corresponding values at $T_o = 25^{\circ}\text{C}$ and were curve fitted as polynomial functions from the experimental data reported in [40] by the following expressions:

$$\mu_{-}^{*} = \frac{\mu_{-}}{\mu_{-o}} = 4.56997 \times 10^{-2} T^{*2} - 3.46447 \times 10^{-1} T^{*} + 9.98367 \times 10^{-1} \quad (4.1)$$

$$\mu_{+}^{*} = \frac{\mu_{+}}{\mu_{+o}} = -1.40625 \times 10^{-1} T^{*3} + 3.75000 \times 10^{-1} T^{*2} - 6.37500 \times 10^{-1} T^{*} + 1.0 \quad (4.2)$$

$$\sigma^{*} = \frac{\sigma}{\sigma_o} = -4.70007 \times 10^{-2} T^{*3} + 3.07364 \times 10^{-1} T^{*2} - 8.37126 \times 10^{-1} T^{*} + 9.98779 \times 10^{-1} \quad (4.3)$$

where $T^{*} = (T - T_o) / T_o$ is the dimensionless temperature, $\sigma_o = 3.65 \times 10^{-3} \text{ N/m}$, $\mu_{-o} = 1.20 \times 10^{-2} \text{ Pa.s}$ and $\mu_{+o} = 1.00 \times 10^{-3} \text{ Pa.s}$ are the interfacial tension, viscosity of oil and viscosity of water at the reference temperature of $T = 25^{\circ}\text{C}$. Figure 4.6 shows the relationships between the above properties and the temperature. In general, these properties vary almost linearly over the small range of temperature change considered in our problem.

Because of the low Reynolds number, the inertia effects are insignificant as compared to viscous effects. At this microscale, gravitational (or buoyancy) effects is generally small and can be neglected. Thus, the densities of the fluid are deem to be not critical in this problem. Besides that, the densities of both fluids are considered as temperature independent in the problem. With all these considerations, the densities of both fluids are

conveniently set to 1000 kg/m^3 for both fluids just like the other workers [19, 20, 30].

However, the densities are not ignored and are part of the governing equations.

Figure 4.6 - Variation of fluids' viscosities and interfacial tension with temperature.

4.3 Results and Discussions

4.3.1 Formation of Water Droplet in Oil

To generalize the numerical results, instead of presenting them with dimensions, it is more convenient to display them in a dimensionless form. With geometrical similarity maintained, the present problem is governed by a total of 7 dimensionless numbers as described section 3.4.2. As the purpose of the present investigation is to study the effect of a thermally mediated droplet formation process, all the above dimensionless parameters are fixed except the dimensionless heater temperature. Different dimensionless heater temperatures, T_{set}^* , of values 0.0, 0.04, 0.2, 0.36, 0.44 and 0.56 were investigated. The time t and the volume V_d of the formed droplet size are non-dimensionalized as $t^* = t / (L/u_o)$ and $V^* = V_d / L^3$ respectively. As shown in the validation exercise, a moderate mesh is selected

for the case study. For the same hydrodynamic flow conditions, this corresponds to 15 control volumes (CVs) spanning across L which gives a total of 56k computational nodes for the domain employed in the present study. A time step size of $t = 5 \times 10^{-5}$ sec is used.

The various forces acting on a forming droplet will be discussed next. Figure 4.7 shows the pressure force F_p generated by the streamwise pressure difference across the forming droplet, the viscous force F_v due the viscous stress acting on the interface of the forming droplet and the interfacial tension force F_{it} . Pressure force F_p and viscous force F_v which act normal to the fluid interface push the forming droplet downstream. Interfacial tension force, F_{it} , which acts tangential to the interface increases with larger interfacial area of the forming droplet. Interfacial tension force F_{it} withholds the forming droplet from detachment. This force is proportional to the interfacial tension σ and the circumference of the water thread l .

Figure 4.7 - Various forces acting on a forming droplet in a T-junction.

Given the capillary number of 3.65×10^{-2} , the formation process in the present investigation should fall within the dripping regime [30]. In this particular regime, both the pressure force F_p and viscous force F_v play the critical role in determining the formation process. For the present study, the thermally mediated environment will, however, allow F_v to exhibit a more dominating effect because of its association with the change in fluid properties with temperature.

Figures 4.8 shows the droplet formation process for the case without the induced temperature field ($T_{\text{set}}^* = 0.0$). Two additional sections of the channel at $y/L = 2.0$ and $x/L = 2.5$ are included. The section at $x/L = 2.5$ indicates the profile of the forming droplet across the channel.

Figure 4.8. Droplet formation without induced temperature field ($T^* = 0.0$) with sections for the planes $y/L = 2.0$ and $x/L = 2.5$.

Figure 4.9. Droplet formation with induced temperature field of $T_{\text{set}}^* = 0$ for the plane $z/L = 0.3$.

Figure 4.9 shows another section of the channel at plane $z/L = 0.5$. These sectional views of the channel are presented for the ease of explanation.

As the water enters into the channel and intersects with the continuous flow of oil, an interface layer is formed between the two phases and the water droplet begins to form. This forming droplet is withheld from detaching by the interfacial tension force F_{it} . As the drop grows, it progressively blocks the cross section of the main channel and obstructs the flow of the continuous phase (oil). This will cause some pressure build-up to occur at the location labeled "X" upstream of the forming droplet (Figure 4.9d) and increase the pressure force F_p . At the same time, the flowing oil exerts a viscous force F_v in the form of surface stress on the forming droplet. Both the pressure force F_p and viscous force F_v push the forming droplet downstream causing the water thread to be thinner. This process

continues to stretch the water thread and making it thinner as shown in Figure 4.9d-e. A thin water thread has a smaller circumference l and therefore weakens F_{it} which withholds the forming droplet from detachment. Besides, interfacial tension effect creates a region of high pressure within the thread at the location where it is the thinnest and has the largest curvature. The water in the thread is then pushed both upstream and downstream away from this region of high pressure within the thread. This further makes the water thread even thinner until the interfacial tension force F_{it} is no longer sufficiently strong to withhold the forming droplet. The water thread breaks and results in the formation of a water droplet. As shown in Figure 4.10, the various stages of droplet formation process of the present procedure agree qualitatively well with the images extracted from the experiment [28].

Figure 4.10 - Droplet formation process: a) Experiment [28] b) Present numerical procedure

4.3.2 Effects of Thermal Forcing

The effect of thermally induced droplet formation will now be discussed. In the present study, it has found that the heater geometry has an important effect in the numerical model. Different heater geometries are modeled and evaluated to emulate the same effect of temperature field distribution within the channel as that of the experiment and ultimately to

achieve the same order of increase in droplet size at elevated temperature. Unfortunately, no experiment data on temperature distribution within the channel was reported [28] for a more precise correlation.

In the experiment, the heater is attached to one side of the channel and is located at the intersection of inlets. This is modeled in the present study. Figure 4.11 shows the droplet formation process with a heater temperature of $T_{\text{set}}^* = 1.0$. The temperature field at the plane $y/L = 2.0$ is also shown. The sectional views of the front and top of the channel are presented in Figure.4.13.

Figure 4.11 - Droplet formation with a thermal forcing of $T_{\text{set}}^* = 1$ at planes $y/L = 2.0$ and $x/L = 2.5$

Figure 4.12 - Sectional views of droplet formation with thermal forcing of $T_{\text{set}}^* = 1$ at planes $y/L = 2.0$ and $z/L = 0.3$

Because the heater is attached to one side of the channel, the temperature field is non-symmetrical. The temperature field generated is rather localized with a much higher temperature near the inner wall where the heater is located. From the plots, the temperature field does not appear to penetrate very far into the flowing fluids. However the layer of oil between the forming droplet and the hot wall is thin, and much of the heat generated can be transferred into the forming droplet. As water has a much larger thermal conductivity than oil, the temperature field penetrates much further into the forming droplet as evidently shown in Figure 4.12b-c. This is a desirable effect because the induced temperature field helps to cause a larger change in properties on a larger portion of the oil-water interface. Therefore, the small patch of localized heating is sufficient to cause visible change in

droplet size. Figure 4.13 shows the size of the droplet formed at different heater temperatures. Without the induced temperature field, a droplet of size $V^* = 1.15$ is formed. The size increases monotonically to $V^* = 1.38$ at a heater temperature of $T_{\text{set}}^* = 0.56$ which corresponds to a temperature of $T = 39^\circ\text{C}$. This means an increase of 20% in the droplet size.

Figure 4.13 - Dimensionless droplet size corresponding to different thermal forcing.

To understand such an increase in the formed droplet size, attention is paid to the change in fluid properties at elevated temperature as shown in Figure 4.6. An increase in the overall temperature of both oil and water results in smaller interfacial tension and viscosities. The sole effect of a smaller interfacial tension tends to suggest that a smaller interfacial force F_{it} is withholding the forming droplet and therefore the forming droplet can easily break off, leading to smaller droplet. This is however not the case as the decrease in viscosities is more significant (Figure 4.6) and therefore plays the dominant role.

A smaller viscosity effectively reduces the viscous force F_v . With a reduction in F_v , the detachment of the forming droplet is delayed. For the case without induced temperature field, the detachment of the forming droplet occurs at $t^* = 2.2$. With the induced temperature field, detachment of the forming droplet is delayed and occurs at around $t^* = 2.4$. During the additional time $\Delta t^* = 0.2$, more water can flow into the forming droplet through the thread leading to a larger droplet size.

Figure 4.14 - Droplet size of simulation results and experiment data [28] at thermal forcing

At 39°C, the increase in droplet size is 20%. This increase is much smaller and does not agree well with the data reported in the experiment [28] which shows a 50% increase for the same temperature difference (Figure 4.14). Even though, the single-sided heater patch has demonstrated its effect in changing the fluid properties, the limited penetration depth of heat flux from the heat source into the critical droplet forming area can only have influence on a limited volume of fluid mass which are exposed to the heat flux. In order to further

increase the heater effectiveness, it is necessary to model a second heater of the same geometry and mirrored onto the opposite side of the channel. As seen in Figure 4.15, mirroring a 2nd heater has improved the penetration of the heat flux from the heater to the fluid mass inside the channel. A more uniform heat distribution can be observed at the critical droplet formation area. If only the experiment data can be made available, it will be most ideal to compare and correlate the temperature field distribution profile of the heater model with that of the experiment.

Figure 4.15 - Temperature distribution of single- and double-sided heater geometry

Figure 4.16 and 4.17 show the isometric and sectional views of the droplet formation process of the double-sided heater model at $T_{\text{set}}^* = 1.0$. It should be noted that the exit channel is longer than the single-sided case. This is to accommodate the development of a larger forming droplet as expected from the improved heater model. The solution of our present numerical procedure shall be independent of the change as the critical aspect ratios of the channel are maintained as before.

Figure 4.16. Droplet formation of double-side heater model at $T_{\text{set}}^* = 1$ with sections for the planes $y/L = 2.0$ and $x/L = 1.5$

Figure 4.17 Sectional views of droplet formation in a double-side heater model at $T_{\text{set}}^* = 1$ at planes $y/L = 2.0$ and $z/L = 0.3$

With the improved heat distribution within the channel, the change in the fluid properties will be more prevailing. This will have a cascading effect on the mechanics of droplet formation and the simulation results are shown in Figure 4.18.

Figure 4.18. Droplet size of optimized simulation results and experiment [28]

As compared with single-sided heater model, the increase in droplet size of the double-sided heater geometry has very good agreement with that of the experiment. Although only one heater is used in the experiment, it is not clear for how long the heater is turned on prior to the start of the experiment. This is an important consideration as the longer the heater is turned on, the more time it will allow the heat to penetrate into the fluid mass inside the channel. This allows the fluid to be well soaked at the desired temperature. As such, the temperature effect on the droplet forming stage will completely take effect by the time the experiment has started.

Numerical model, on the other hand, will present a more ‘localised’ heating effect as compare to the actual experiment setup. In order to correlate their effect with experiment, the actual penetration and spreading of the heat flux will have to be better represented in the numerical model. This can be achieved by optimising the heater geometry which has been illustrated in this present study with the use of double-sided heat geometry.

At thermal forcing, the phenomenon of thermo-capillary effect is present. Figure 4.19 shows the cross sectional view of the droplet at the plane $x/L = 2.5$ (single-sided heater) and 1.5 (double-sided heater). In the case of the single-sided heater, the droplet is observed to be inclining towards the right side of the channel where the heater is sited. The non-symmetrical temperature field (Figure 4.11) of a single-sided heater has created the thermo-capillary effect which draws the forming droplet toward the region of higher temperature. This effect is even more apparent for a channel with square cross section [117] However, for the case of double-sided heater model, this effect cannot be clearly observed from the cross-sectional droplet profile even though the thermo-capillary effect is present if not stronger. Due to the symmetrical temperature field distribution, the forming droplet in the double-sided heater geometry develops centrically with respect to the channel cross section. This profile is similar to one without thermal forcing.

Figure 4.19 Cross-sectional view of the forming droplet at $x/L = 2.5$ (single-sided heater) and 1.5 (double-sided heater).

4.4 Conclusions

The chapter presented a three-dimensional simulation of the droplet formation process in a microfluidic T-junction under an induced temperature field. Formation of water droplet in oil in the dripping regime was performed as a case study and compare with experiment data. In the present model, the roles of lower viscosities and interfacial tension of the fluids were explored in the droplet formation process. This is achieved through thermal forcing provided by a heat source. Different heater geometries – single and double-side – are modeled in this present study to achieve a correlation to experiment data. It is found that the double-sided heater model has agreed well with the experiment results in regards to the increase in droplet size at elevated temperature. Thermo-capillary effect drives the forming droplet towards the region of higher temperature allowing the generated heat to penetrate further into the forming droplet. This phenomenon increases the effectiveness of the applied temperature field. The effect is more noticeable for the case of single-sided heater

geometry which has a non-symmetrical temperature field. However, this does not undermine the absence of thermo-capillary effect on double-sided heater geometry. In fact, double-sided heater geometry has shown to provide a more uniform heat distribution and thus better heat penetration within the channel. As a result, a larger droplet size can be seen at elevated temperature. In terms of the increase in droplet size over a temperature range from 25°C to 39°C, the present numerical solution with the optimized heater geometry has shown good agreement with the experiment data. As demonstrated, the present procedure can be employed to investigate the case of an active droplet formation which makes use of thermal forcing. The procedure can also be extended to other channel geometries.

Chapter 5 Thermal Coalescence of droplets

This chapter reports the numerical investigation of thermocoalescence of droplet in a microchannel geometry which consist of a non-temperature controlled droplet formation section cascading to a temperature-induced merging chamber. The numerical model is formulated as an incompressible immiscible two-phase flow problem with oil and water as the continuous and dispersed phase respectively.

The first objective of this chapter examines the droplet formation process and droplet size at various combination of oil and water flow rate.

The second objective analyses the motion of the droplet across the heat-induced merging chamber. The numerical method is able to provide a detail visual presentation of the droplet movement in a heated environment under the Marangoni effects which otherwise cannot be observed in experiment.

The third objective investigates the relationship between the merging temperature and the liquid flow rates.

5.1 Problem Description

The geometry of the microchannel for the present study is shown in Figure 5.1. The microchannel is similar to the experiment setup reported by Luong [61].

Figure 5.1 - Thermally controlled droplet coalescence microchannel

It consists of a T-junction droplet forming section and a merging chamber of 1mm x 1mm. The T-junction has a square cross section and two inlets - one for the continuous phase (oil) and the other for dispersed phase (water). The continuous phase and the dispersed phase are immiscible. For the convenient of the later implementation of level-set simulation, they are designated as the negative (-ve) fluid and the positive (+ve) fluid for the continuous and dispersed phase respectively. These phases are distinguished in the level set function by their signs. The continuous phase (-ve fluid) flows into the channel from the left inlet with an average inlet velocity of u_{-o} at an initial temperature of T_o . At the same time, the dispersed phase (+ve fluid) enters the other inlet with an average velocity of u_{+o} of T_o . Droplet formation occurs at the T-junction where the two channels join. The shear force and the pressure build-up at the junction causes a breakup of the disperse phase and as a result, droplets of the +ve fluid are formed. The droplets flow downstream into the merging chamber where droplet coalescence occurs under a thermally controlled condition. Fully

developed flows are assumed for both inlets. Non-slip condition is enforced at the walls of the channel. Out flow boundary condition is employed at the outlet.

Temperature field is induced into the merging chamber by a heater attached to the wall of the merging chamber at the location shown in Figure 5.1. When the heater is switched on, the temperature of the wall directly adjacent to the heater is raised to T_{set} . Other location of the wall is maintained at a temperature of T_o . At the outlet, zero-gradient is employed. With these boundary conditions, a non-uniform temperature field is established in the channel and changes the properties of both fluids. The temperature field induced additional forces acting on the droplet and together with the changes of fluid properties within the merging chamber, creates the right condition for droplet coalescence to happen.

5.2 Material and Experimental Setup

The microchannel device is designed in accordance to the geometry outlined in Figure 5.1 and fabricated using micromachining of glass and polydimethylsiloxane (PDMS) as described in section 4.2. The heater attached to the wall of the merging chamber is fabricated on a Pyrex glass wafer by a lift-off process. The microchannel and the heater were treated by oxygen plasma, aligned under a microscope and brought into contact. The fabricated microchannel device is shown in Figure 5.2a.

The experiment setup consisted of:

1. Precision syringe pump which is meant to feed the fluid into the microchannel.
2. DC power supply and digital multimeter which are meant to control the heater temperature

3. Imaging system which is meant to capture the image of the droplet formation and coalescence processes.

Details on these individual instrument have been described in section 4.2. The final setup is shown in Figure 5.2b.

Figure 5.2 – a) Microchannel device; b) Experiment setup

The dispersed phase is DI water and continuous phase is mineral oil (Sigma M5904) mixed with 2% w/w Span 80 surfactant (Sigma S6760). As in the experiments, the flow rate of water in this numerical study is kept at $Q_+=100 \mu\text{l/h}$ in combination with the various flow rate for oil at $Q_-=150 \mu\text{l/hr}$, $175 \mu\text{l/hr}$, $200 \mu\text{l/hr}$ and $225 \mu\text{l/hr}$. The densities

and viscosities and interfacial tension of the fluids at reference temperature of $T_o = 25^\circ\text{C}$ are $\rho_- = \rho_+ = 1000 \text{ kg/m}^3$, $\mu_{-o} = 2.64 \times 10^{-2} \text{ Pa.s}$, $\mu_{+o} = 1.00 \times 10^{-3} \text{ Pa.s}$ and $\sigma_o = 3.65 \times 10^{-3} \text{ N/m}$. As viscosities and interfacial tension are temperature dependent fluid properties, they are normalized with their corresponding values at $T_o = 25^\circ\text{C}$ and are curve fitted as polynomial functions from the experimental data reported in [40] by Eqn 4.1- 4.3 for use in numerical analysis.

The densities of the fluids are set to be the same since the densities of both fluids are considered as temperature independent in the problem and buoyancy is thus neglected [19, 20, 30].

Table 5.1 below shows the respective Reynolds number and Capillary numbers for the various combinations of flow rates.

Q_- flow rate, $\mu\text{l/hr}$	Q_+ flow rate, $\mu\text{l/hr}$	Re	Ca
150	100	0.395×10^{-2}	0.753×10^{-2}
175	100	0.460×10^{-2}	0.879×10^{-2}
200	100	0.526×10^{-2}	1.005×10^{-2}
225	100	0.592×10^{-2}	1.130×10^{-2}

Table 5.1 - Re and Ca for different combinations of flow rates

As mentioned in Section 4.2.2.3, the densities of the fluid are deemed to be not critical in this problem and are conveniently set to be the same for both fluids

5.3 Results and Discussions

In the present numerical study, the coalescence of the water droplet at various flow ratio and temperature of the merging chamber are considered in the numerical study. The numerical results are compared with the experiment data reported previously by our group

[61]. At the upstream of the merging chamber, the formation of water droplet is realised by T-junction geometry as described in Figure 5.1. The size of droplet and rate of droplet generation are determined by the flow ratio between the continuous phase (oil) and the dispersed phase (water). The main channel is filled with continuously flowing mineral oil as the forming droplet develops. A layer of oil is always present between the droplet and the channel wall. Therefore, the droplet will not come into direct contact with the wall and thus, the absence of moving contact line. Based on this assumption, the contact angle will not be taken into account in our problem.

5.3.1 Time function of the droplet formation stage

The formation of droplet at the T-junction of the microchannel device under the flow conditions employed in our previous experiment [61] is discussed in this section.

As explained in 4.3.1, the forces acting on the forming droplets are the pressure force F_p , the viscous force F_v and the interfacial tension force F_σ . And, droplet formation is the resultant of these forces under the hydrodynamic flow condition. In this present study, the capillary number ranges between 0.008 and 0.011 and squeezing regime can be observed from the numerical results. This agrees well with De Menech findings [30].

Figure 5.3 compares the formation of droplet from the simulation results and the experiment. The shape of the droplet tip, the shape of droplet at breakup and the size of the droplet is slightly small for the numerical results. This is attributed to the impact of surfactant. But the physics of droplet formation remains consistent.

It has to be noted that surfactant is used in the experiment to stabilize the formation of droplet and the effect of surfactant is not considered in the present numerical model. The surfactant affects the shape of the droplet at breakup and the tip.

Figure 5.3 - Droplet formation process a) Experiment [61] b) Present numerical procedure

Figure 5.4 shows the size of droplet decreases with increasing flow rate. Higher oil flow rate leads to higher pressure force and viscous force to overcome the interfacial force that is withholding the droplet from detachment. As a result, it takes shorter time to achieve droplet breakup at higher flow rate and the droplet size reduces almost linearly. Similar trend was also observed in Garstecki [29] experiment where the droplet length, or indirectly the droplet volume, reduces with increase flow rate ratio.

Figure 5.4 - Droplet size at different flow rates.

5.3.2 Movement of droplet in the merging chamber at elevated temperature

As shown in Figure 5.1, a heater is attached to the wall of the merging chamber. The temperature of the wall directly adjacent to the heater will be raised to T_{set} when the heater is switched on. Figure 5.5 shows the temperature profile on both horizontal and vertical planes in the merging chamber when the heater is set at 60°C . Because the heater is attached to one side of the merging chamber, the temperature distribution within the chamber can be observed to be asymmetrical.

Figure 5.5 - Temperature distribution in the merging chamber

It can be seen in Figure 4.6 that the surface tension and fluid viscosities reduce at elevated temperature. In fact, the –ve fluid (i.e. oil) shows a bigger reduction in viscosity than water. Under such thermally induced condition, the forces acting on the droplet change accordingly and thus, affecting its movement across the merging chamber. With a decrease in the viscosity, the continuous phase (i.e. oil) loses some extent of the hydrodynamic drag force to push the droplet downstream and instead, flows around the droplet at a faster velocity towards the outlet of the merging chamber. At the same time, the droplet is also subjected to the temperature distribution generated by the heater. As shown in Figure 5.5, it has an asymmetric profile and the droplet will be drawn toward higher temperature region similar to the thermocapillary phenomena. The overall effect of these forces results in a decrease in velocity of the droplet in the direction towards the outlet of the chamber. The phenomena can be observed by comparing the movement of the droplet in the merging chamber for the case of 25°C and 60°C as shown in Figure 5.6. By the time when the droplet at 25°C has already exit the chamber at 0.68sec, the droplet in a 60°C chamber

temperature is still in the chamber. These results show that the droplet velocity decreases at elevated temperature.

Figure 5.6 – Droplet position in the merging chamber at a) 25°C and b) 60°C at flow rate of 225ul (-ve fluid) and 100ul (+ve fluid).

Luong et al [61] reported in their experiment the droplet velocity decreases with increasing heating temperature, Figure 5.7(a). The droplet velocity is normalized at $T_o = 25^\circ\text{C}$ such that, $v^* = v / v_o$. It was explained that the normalized droplet velocity is exponentially related to heating temperature as,

$$v_+^* \propto \frac{\sigma^*}{\eta_+^*} \propto \exp\left(-\frac{\theta}{\theta_o}\right) \quad (5.1)$$

where $\theta = T - T_o$.

From the present numerical results of Q_-/Q_+ flow rate of 150uL/100uL as shown in Figure 5.7(b), it can also be seen that the droplet experienced an exponential decay on its velocity over increasing temperature. Thus, our numerical results agree well with the experiment.

Figure 5.7 - Relationship between droplet velocity and increase in temperature

It is expected that the same behaviour will be exhibited at other flow rate. Thus, in the present study, it does not necessitate the need to run the numerical simulation at other flow rate.

Next, we examine the time history of the velocity. Figure 5.8(a) shows the velocity profile of the 1st and 2nd droplet in the merging chamber for the case of 25°C, 50°C and 60°C. The velocity of the droplet is normalised to its velocity in the forming section of the microchannel device just before entry into the chamber. The three cases represent the following scenarios:

Case 1: 25⁰C. No heating

Case 2: 50⁰C. Heating but no droplet coalescence

Case 3: 60⁰C. Heating and droplet coalescence

Figure 5.8(b) shows the corresponding velocity profile of the droplet for Case 3 taken from actual experiment. The present numerical solution and experiment data show similar trend. For all the three cases, the present numerical solution shows that the velocity of the first droplet decreases as it enters the chamber. The expansion in the channel geometry induces a sudden decrease in flow velocity leading to a slowdown in the droplet velocity. This behaviour can also be observed in the experiment, Figure 5.8(b).

Immediately after the droplet enters the chamber, it accelerates for the case of elevated temperatures (i.e. 50⁰C and 60⁰C). Such behaviour is not seen in the case of no heating (i.e. 25⁰C). This phenomenon is attributed to the thermocapillary effect.

Figure 5.8 – Velocity profile of droplet velocity across the merging chamber
a) Present numerical procedure; b) Experiment, 60°C

Figure 5.9 shows the shape of droplet superimposed on the temperature distribution within the chamber. Different temperature is experienced by the leading and the trailing face of the droplet as it enters the heated chamber. Right at the entrance to heated merging chamber, the trailing face of the droplet is colder than the leading face. This results in the surface tension force on the trailing face being higher than that on the leading face. As shown in Figure 5.9, the radius of curvature of the trailing face is higher in the case of elevated temperature as compared to non-heating. This affects the shape of the droplet such that it appears less spherical at elevated temperature. The higher surface tension force on the trailing face has given an added thrust to the droplet. It is this difference in the surface tension force between the trailing and leading face that accelerates the droplet forward. However, once the droplet is completely inside the chamber, the droplet velocity reduces as it traverses further into the chamber.

Figure 5.9 – Droplet shape at position right after entry point of merging chamber

It can also be seen that the rate of acceleration increases with the chamber temperature. At the time instant of 0.6sec, the normalized velocity of the droplet reaches 1.19 for 60°C. Whereas, the value at 50°C is only 1.02. This result demonstrates the effect of thermocapillary force at elevated temperature. The effect is not so obvious in the experiment although a small ‘bump’ in the velocity profile can be seen at the beginning of the drop movement in the heated chamber. Further investigation is needed to understand if presence of surfactant is withholding this effect. However, this is beyond the present scope of work.

The thermocapillary effect begins to wear off quickly as the droplet travels further into the chamber. A progressive reduction in droplet velocity can be observed until the droplet reaches the chamber exit. In the heated chamber, the viscosity of the fluid reduces with increasing temperature as shown in Figure 4.6. This change in the fluid property allows the fluid to flow around the droplet more easily instead of dragging it forward. This effect leads to a reduction in droplet velocity and is more dominant than the reduction in the velocity of the fluid due to the expanded geometry of the chamber as explained earlier. In the case of no heating, the velocity profile of the droplet does not change significantly in the merging chamber after the initial decrease in velocity. The effect of expanded geometry does not cast a consistent impact on the droplet. Otherwise, a decreasing trend in the velocity profile of the droplet will be observed across the chamber. Therefore, it is evident that the decreased in fluid viscosity at elevated temperature is the dominant factor responsible for the progressive decrease in droplet velocity in the merging chamber. With heating, a decreasing trend in the velocity of the droplet is observed across the chamber. The trend is in agreement with the experiment data.

A further investigation was conducted to understand the magnitude of influence of the thermocapillary force as compared to the hydrodynamic drag force of the oil on the droplet movement. Two simulations were carried out by limiting the change in fluid viscosity and surface tension with temperature. The separation of temperature dependencies of viscosity and interfacial tension is not possible in the real situation and such investigation can only be carried out numerically instead of the actual experiment. As shown in Figure 5.10, the velocity of the droplet at elevated temperature is almost the same as at 25°C if the surface tension is kept constant, e.g. temperature independent. This result indicates the significance of thermocapillary force on the thermocoalescence process. On the other hand, the velocity profile of the droplet is close to the case of elevated temperature if the viscosity is kept constant. This result further ascertains the dominating effect of thermocapillary force on the droplet movement.

Figure 5.10 - Velocity of the droplet with isolated temperature dependent parameters

As mentioned in Eqn.3.6, the thickness of fluid interface for the present study is set at 1.5 times of grid size, Δx . A thickness of 3 times Δx is employed in a separate simulation to check the sensitivity of smearing factor on the numerical results. It has been observed in Figure 5.10 that the droplet movement is the same as the reference and coalescence also happened at 60°C. The results have shown that changing the interface thickness does not significantly affect the numerical results.

So much on the behavior of the 1st droplet, the movement of 2nd droplet will now be discussed. At the entrance of the merging chamber, the normalized velocity of the 2nd droplet is slightly higher than the 1st droplet. As mentioned earlier that the velocity is normalized to the velocity of travel in the forming section of the device before it enters the

chamber. Because of the presence of 1st droplet in the merging chamber by the time the 2nd droplet enters, the 2nd droplet experiences a smaller expanded volume as compared to what the 1st droplet sees whereby the merging chamber is empty. The fluid velocity entering into the merging chamber will thus experience a smaller reduction in its velocity. Because the flowing fluid is the transport medium of the droplet, a reduction in the fluid velocity will translate to a smaller decrease in the velocity of 2nd droplet as it enters the chamber. Therefore, its normalized velocity is slightly higher than the 1st droplet. Other than this observation, the 2nd droplet exhibits similar trend as the 1st droplet for all the 3 cases.

Another phenomenon that can be observed in this numerical study is the skewed movement of the droplet in the merging chamber as it enters the chamber and interacts with the temperature field. As described earlier, the temperature field is asymmetrically distributed and does not penetrate evenly across the horizontal plane (Figure 5.5). Figure 5.11 show that this asymmetrical temperature field has drawn the droplet horizontally towards the region of higher temperature close to the heated side of the chamber. As the droplet is drawn towards inner wall, it flattens out and appears to be slightly bigger if viewed from the z-direction, Figure 5.9.

Figure 5.11 – Position of droplet in the heated chamber

5.3.3 Droplet coalescence

This section discussed the mechanics behind the droplet coalescence in the present study and compare with experiment [61]. The relationship between the critical merging temperature and flow ratio will also be discussed.

The mechanism behind droplet coalescence is the rupture of the bridging oil film between two droplets due to their close proximity. The high curvature at the liquid bridge results in high force associated with surface tension effect around the bridge thus pulling the two droplets together and leading to droplet coalescence [48, 118]. The idea of having a merging chamber is to create an expansion in the channel geometry to slow down the droplet movement so as to close up the distance between two adjacent droplets. To enhance the condition of droplet coalescence, heat is induced into the merging chamber with the help of a heater to further slowdown the velocity of droplet to a point where it will contact with the next incoming droplet. As explained in the previous section, increasing the

temperature of the merging chamber will lower the viscosity of the fluid as well as creating an imbalance in the surface tension of the droplet leading to a reduced velocity of the droplet.

Figure 5.12 – Droplet coalescence at elevated temperature:

a) No Heating b) Heating at 60°C

As shown in Figure 5.12, droplet coalescence occurs at 60°C at flow rate ratio of $225(Q_-)/100\mu\text{L}(Q_+)$. With heating, the droplet slows down and arrives near the exit point only at 0.68 sec compared to 0.56 sec for the case with no heating. The droplet in a non-heated chamber exits the channel very much earlier at 0.64 sec, Figure 5.12(a). However, the droplet in the heated chamber can be observed to remain almost the same position near the exit of the merging chamber after 0.68 sec, Figure 5.12(b). This is attributed the surface tension gradient on the droplet as a result of the non-uniformity of the temperature distribution around the droplet. When the droplet approaches the exit of the channel, its leading face experiences a colder temperature than the trailing face. This is because the heat flux is concentrated mainly within the merging chamber due to the location of the heater. As shown in Figure 5.5, the intensity of the heat flux decays towards the outlet. The colder leading face of the droplet will experience a higher surface tension force than at the trailing face causing the leading face to exert a thermocapillary force that pushes the droplet backward acting against the forward hydrodynamic drag force of the fluid flow. When force equilibrium is achieved between these two forces, the droplet appears to be ‘trapped’ at the exit of the merging chamber waiting for the next droplet to arrive. The incoming droplet eventually comes into contact with the ‘trapped’ droplet leading to droplet coalescence.

After coalescence, the merged droplet is twice as big and blocks the exit of the merging chamber, increasing its pressure. The pressure builds up in the merging chamber until it has gained sufficient magnitude to be able to push the merged droplet out of the chamber eventually.

In the present work, the key factor for droplet coalescence to occur depends on the ability to trap the droplet at the exit of the merging chamber. Coalescence relies on the thermocapillary force to counter the hydrodynamic force of the fluid flow. And, the thermocapillary force is dependent on the temperature of the heater while hydrodynamic force is a function of fluid flow rate. For a particular flow rate, there is a threshold temperature beyond which droplet coalescence will take place. We describe the difference between this threshold temperature and room temperature of 25°C as the critical merging temperature, θ_{cr} , with the following relationship.

$$\theta_{cr} = T_{merging} - T_o$$

As depicted in Figure 5.13(a), the present numerical study shows that the critical temperature increases with increasing flow rate. This is because a higher thermocapillary force, which is achieved by higher temperature, is needed to overcome the higher hydrodynamic force from a higher flow rate. This trend is in agreement with our previous experiment [61]. However, the critical merging temperature for the range of oil flow rate spans from 27°C to 30°C for the present numerical study which is much smaller than the experiment. This difference is due to the effect of the surfactant which is not considered in the present numerical model. Lu et al [119] discussed the effect of surfactant in preventing droplet coalescence. The uneven distribution of surfactant on the surface of droplet will directly affect the coalescence of droplet. Their findings also revealed that the Marangoni effect which is caused by surface tension gradient will further hamper the even distribution of the surfactant.

Figure 5.13 – Critical merging temperature versus flow rate a) Present numerical results
b) Experiment

The numerical study is extended to a merging chamber with a top heater in addition to the original bottom heater. This additional heater allows a wider span of critical merging temperature as shown in Figure 5.13(a).

In the case with bottom heater only, the asymmetric temperature distribution in the merging chamber cause the droplet to be drawn towards the heated side resulting in a gap

between the droplet and the top side of the device, Figure 5.11. This allows the fluid to flow pass the droplet and the droplet experiences a lower hydrodynamic drag from the fluid flow. However, in the case of two heaters, the symmetrical temperature distribution within the chamber keeps the droplet in the middle of the chamber and occupies the cross section of the channel with no gap between the droplet and the channel wall, Figure 5.14. The droplet will thus experience a higher hydrodynamic force from the fluid flow. In order to counteract this hydrodynamic force so that the droplet can be ‘trapped’ to meet the incoming droplet, a higher thermocapillary force is necessary. This can only be achieved with higher temperature and thus leading to higher critical merging temperature especially obvious at higher oil flow rate.

Figure 5.14 – Temperature profile with heaters on both sides of the chamber.

5.4 Conclusions

The study reported a three-dimensional simulation of temperature-induced droplet coalescence in a microchamber with oil as the continuous phase and water as the disperse phase. The flow rate of oil ranges between 150 $\mu\text{l/h}$ to 225 $\mu\text{l/h}$ with a constant water flow rate of 100 $\mu\text{l/h}$. The process of droplet formation is validated with the experiment data and has shown to be in agreement.

Based on the numerical results, the size of droplet reduces with increasing oil flow rates. The higher viscous force from the increased oil flow rate causes the droplet breakup to occur earlier and thus, shortens the droplet growing cycle and the volume of droplet reduces accordingly.

The movement of droplet in the heated chamber is examined. The numerical results show that the velocity of the droplet reduced across a heated chamber. Upon more detail analysis, the results show that the droplet has initially accelerated when it first enters the chamber and thereafter slows down as it travels across the heated chamber. When subjected to the temperature profile of the heated chamber, the droplet will experience a temperature gradient across its surface. The temperature dependent surface tension difference between the leading and trailing face of the droplet has pushed the droplet towards the heated region. Analysis on the next adjacent droplet shows similar trend on the velocity profile as compare to the 1st droplet although the normalized velocity is slightly lower due to a reduction in the expanded volume of the merging chamber as a result of the pre-occupation of the 1st droplet.

At some critical temperature, it can be observed that the droplet remains at the same position near the exit of the merging chamber. This phenomenon favors the condition for

droplet coalescence by holding the droplet at the chamber exit while waiting for an incoming droplet to arrive and merge with it. Incidentally, the critical temperature for droplet coalescence is dependent on the flow rate. The numerical results show that at higher oil flow rate, a higher temperature is required for droplet coalescence to occur. The relationship between critical temperature and flow rate is almost linear. Similar trend can be seen in the experiment except that the temperature range is wider for the same range of oil flow rate. This is attributed to the effect of surfactant present in the experiment which induces an additional factor into the physics behind the droplet coalescence.

In the present study, the numerical simulation on droplet coalescence is extended to a configuration with top and bottom side heater. This is beyond the actual experiment setup in which the heater is only implemented on the bottom wall of the microchannel device. With only one heater, the asymmetrical heat distribution results in the droplet being drawn towards the heater side leaving a gap between the droplet and top wall of the microchannel. Fluid is able to flow through this gap and the hydrodynamic flow effect on the droplet is lessened as compared to the case of two heater configuration where there is no gap between the droplet and wall. As a result, a lower thermocapillary force is needed to overcome the hydrodynamic drag in order to hold the droplet at the exit. Therefore, bottom side heater sees a lower critical merging temperature as compared to the case of two heater configuration.

Overall, the present numerical simulation has demonstrated the advantage over the actual experiment in providing more insights on the physical behavior of the droplet under the thermocapillary effect.

Chapter 6 Conclusions and Future Works

6.1 Conclusion

The key objectives of the work are to establish a numerical model on droplet formation in a microchannel device and to use the model to study droplet formation and droplet coalescence in a thermal mediated environment.

A numerical solution procedure for three-dimensional simulation of droplet formation in T-junction geometry has been formulated. The simulation results have been validated using experimental data. The results are qualitatively close to that of the experiments. Thus, the competency of the solution procedure has successfully been demonstrated.

However, the model does not take into account the effect of surfactant which is added into the fluid used in the experiment. Surfactant will alter the surface tension. Besides that, the thermal property of surfactant will need to be considered. The thermal property may impede heat transfer into the droplet, and thus affecting the droplet size and coalescence temperature reported in this dissertation. The surfactant effect could have contributed to some of the differences seen between the simulation results and the experiment. This deficiency can be understood and addressed by including the surfactant effect into the model. And this will be considered for future enhancement work.

In a thermal forcing environment, the thermocapillary effect has shown up clearly in the simulation results. Under the thermocapillary effect, the forming droplet can be seen to be leaning toward region of higher temperature. Such observation cannot be easily realized in experimental setup due to limited angle of view by the vision system. This phenomenon is desirable as it helps to facilitate the penetration of heat into the forming droplet which thus,

increases the effectiveness of localized heating provided by the microheater. Thermal forcing has induced a reduction in the viscosities and surface tension of the fluids thereby causing an impact to the physics of droplet formation. The end result is that the droplet size increases. This is primarily due to the more significant reduction in the viscosities of the fluids as compared to the surface tension which allows more time for the droplet to grow prior to detachment. The delay in detachment has resulted in bigger droplet size.

A thermally mediated environment has also helped to enhance the controllability of a geometrically expanded chamber intended for the purpose of droplet coalescence. Without thermal control, the chamber size has to be carefully designed with due consideration to the fluid flow rate in order to create the right condition to narrow the distance between adjacent droplets up to a point where they are brought in contact for coalescence to occur. Thermal forcing has introduced an additional control knob into the expanded chamber design to supplement the necessary condition for droplet coalescence. The reduction in fluid viscosity at elevated temperature has further reduced the movement of droplet in the chamber. Thus, there is an improved opportunity for the droplet to slow down to be in contact with incoming droplet even at higher flow rate. The thermal feature has, therefore, allowed a given geometrical design - which is intended for droplet coalescence - to accommodate a higher range of fluid flow rate. Different heater configurations are also numerically experimented to study its effectiveness on a thermally mediated merging chamber. It has been understood that a single-side heater requires a lower temperature for droplet coalescence even for a higher fluid flow rate.

6.2 Future Works

The work in this dissertation has demonstrated the impact of viscosity change to the droplet size and droplet movement. The effect of surfactant will need to be considered as an enhancement to the existing model. This will help to bring the numerical result even closer to the experiment. In addition, the hydrodynamic part of droplet formation mechanism deserved a closer examination. Thus, the present work can be extended to study the effect of Reynolds number, capillary number, viscosity ratio, surface tension etc.

Instead of thermal forcing, change of viscosity can be effected using functional fluid such as non-Newtonian fluid which has a viscosity value that is dependent on the shear rate or stress. This unique characteristic deserves further investigation in our research in droplet-based microfluidic.

Since the model has already been validated, it will be more than appropriate to deploy numerical modeling to gain a head-start in the investigation process. Experiments can eventually be conducted to correlate with the simulation results. Numerical modeling provides a means to streamline and reach down to a set of desirable experimental conditions that is able to maximize the probability of capturing the expected outcome as a final validation step. This will help to reduce the amount of time, labour and financial resources needed to support the usual iteration cycles of experimentation. In this aspect, we believe the research process will be optimized and refined using by this combination of numerical and experimental approaches.

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