Investigation of multiphase liquid systems in microchannels

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**NANYANG
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**INVESTIGATION OF MULTIPHASE LIQUID SYTEMS
IN MICROCHANNELS**

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SCHOOL OF MECHANICAL & AEROSPACE ENGINEERING

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Investigation of Multiphase Liquid Systems in Microchannels

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ABSTRACT

Droplet-based microfluidics plays an important role in biological and chemical sciences. Different operations such as transport, reagent reaction, particles sorting, and merging can be achieved within a confined droplet. Droplet manipulation can be achieved in a passive or active way. The passive way focuses on the ingenious device design and arrangement, while the active way incorporates external forces including but not limiting to thermocapillary force, electrowetting effect, and magnetic force.

Numerical procedures are implemented to study multiphase systems in microchannels. The combined governing equations are employed to calculate the physical fields. They are solved using a finite volume method on the uniform Cartesian grid. The interfacial tension force and the magnetic force are coupled in the Navier-Stokes equation. The interface between two phases is captured using a particle level-set method. The present numerical codes are validated to be accurate to use in the droplet-based microfluidics.

In this thesis, a series of diffuser/nozzle structures is employed to investigate the behavior of microdroplets flowing in microchannels. Depending on the imposed flow direction, the serial structures can act either as a series of diffusers or nozzles. On the experimental front, the fabrication of the test devices used the soft lithography techniques in polydimethylsiloxane (PDMS). T-junctions for droplet formation, diffuser/nozzle structures and pressure ports were integrated in a single device.

Mineral oil with surfactant and de-ionized water worked as the carrier phase and the dispersed phase, respectively. Each phase was driven by an individual syringe pump to give different flow rates. A high-speed camera mounted on the microscope recorded the deformation of water droplet in both diffuser and nozzle configurations at the fixed flow rate ratio between oil and water of 30. Two-dimensional and three-dimensional numerical models were employed to study the dynamics of the microdroplet during its passage through the diffuser/nozzle structures. The interface of the two phases was captured using a level-set method.

At first, the pressure drop between two ends of the diffuser/nozzle microchannel was measured with pressure sensor in both directions. The experimental and numerical results show that the pressure drop is linearly proportional to the flow rate. Furthermore, the rectification effect was observed in all tested devices. The pressure drop in the diffuser configuration is higher than that of the nozzle configuration. Finally, the measured results of the pressure with droplet and without droplet were analyzed and compared. The rectification characteristics can be used for the development of micropumps for multiphase systems.

Secondly, at the same flow rates of the continuous and the dispersed phases, the velocity of the droplet is determined by the viscosity of the continuous phase and the interfacial tension between the two phases. Both numerical and experimental results show that the velocity of the droplet increases with increasing capillary number. The droplet velocity is higher than the mean velocity of the fluid system and increases

with increasing viscosity of the continuous phase or decreasing interfacial tension. In all experiments, the droplet moves faster in the diffuser direction than in the nozzle direction. Our findings allow the development of a new measurement approach for interfacial tension.

As a second study case, the effect of magnetic force on the formation of ferrofluid droplets in a flow focusing channel was investigated numerically and experimentally. A three-dimensional model was built. Two phases of ferrofluid and silicone oil were employed in the simulation. The magnetic force as a body force was coupled into the Navier-Stoke equations. The interaction between hydrodynamics and capillarity force acting on the ferrofluid tip was analyzed numerically in the conditions of without and with magnetic field. The evolution of droplet formation and the time dependent velocity field are discussed. Increasing magnetic susceptibility or increasing magnetic field lead to the formation of larger droplets.

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NOMENCLATURE

b	radii of sessile droplet basement [m]
$\vec{B}$	magnetic flux density [T]
Bo	Bond number [-]
B_m	magnetic Bond number [-]
C	constant [-]
Ca	capillary number [-]
d	diameter of initial droplet [m]
D^*	dimensionless number of droplet diameters [m]
$\vec{F}_\sigma$	interfacial tension force per unit volume [Nm^{-3}]
$\vec{F}_g$	body force per unit volume [Nm^{-3}]
$\vec{F}_m$	magnetic force per unit volume [Nm^{-3}]
F_μ	viscous force [N]
F_P	pressure difference [Nm^{-2}]
F_σ	the interfacial tension force [N]
F_m	magnetic force [N]
g	gravitational acceleration [ms^{-2}]
h_0	droplet height [m]
H	smoothed Heaviside function
H_1	Hamiltonian
$\vec{H}$	magnetic field strength [Am^{-1}]

H_0	magnitude of magnetic field [Am^{-1}]
H_c	channel height [m]
l	capillary length [-]
L_D	droplet length [m]
L	character length [m]
L_t	triple contact line of the 3D model
$\vec{M}$	the magnetization of the ferrofluid [Am^{-1}]
M_0	mass [kg]
n	component of the normal to the contact line
$\hat{N}_F$	normal to the interface
P	pressure [Nm^{-2}]
P_{Ca}	capillary pressure [Nm^{-2}]
q	volumetric flow rate [$\mu\text{l/h}$]
Q	charge per unit area [C]
r_p	radii of the particles[m]
R	droplet radii [m]
R_{cri}	critical radii of droplet movement [m]
Re	Reynolds number [-]
S	source term in the general transport equation
t	time [s]
t'	pseudo-time [-]
T	temperature [$^{\circ}\text{C}$]

$\vec{u}$	velocity vector [ms^{-1}]
u	velocity component in the x -direction [kg s^{-1}]
$U_{\max}^*$	dimensionless maximum droplet velocity [-]
$\bar{u}_c$	mean velocity of the carrier fluid [ms^{-1}]
v	velocity component in the y -direction [kg s^{-1}]
V	droplet velocity [ms^{-1}]
V_0	voltage [V]
V_d	droplet volume [m^3]
V_{ori}	original droplet volume [m^3]
V_{cur}	current volume [m^3]
V^*	dimensionless number of droplet volumes [-]
w	velocity component in the z -direction [ms^{-1}]
W_c	channel width [m]
W_i	interfacial potential
x	coordinate axis
x_1, x_2	contact points of the 2D model [m]
$\vec{x}_p$	position vector of the center of the particles
y	coordinate axis

Greek symbols

σ	surface tension coefficient [Nm^{-1}]
μ	viscosity [$\text{Pa}\cdot\text{s}$]
ρ	density [kg m^{-3}]

θ	contact angle [°]
θ^*	the equilibrium static contact angle [°]
ζ	volume fraction in the computational domain
κ	curvature
ε	1.5 of control volume size [m]
η	gap between droplet interface and the channel wall [m]
ϕ	level-set function
φ	phase function
γ	width of the band around the interface [m]
Ω	fluid region
μ_0	permeability of the free space [NA ⁻²]
μ_1	permeability of the ferrofluid [NA ⁻²]
μ_b	permeability of the bubble [NA ⁻²]
χ_m	ferrofluid susceptibility
ψ	magnetic scalar potential
Γ_i	location of interface or the zero level-set value
λ	the slip length [m]
ω	effective friction coefficient [-]
Φ	the dependent valuable
ψ	the magnetic scalar potential
α	opening angle of the device [°]
Γ	diffusion coefficient [-]

Δx	width of a control volume [m]
ΔP	Pressure drop [Nm^{-2}]

Subscripts

C	channel
c	continuous flow
d	dispersed flow
p	particle
x	component in the x -direction
y	component in the y -direction
z	component in the z -direction
+	dispersed phase
-	continuous phase

Acronyms

2D	two-dimensional
3D	three-dimensional
CEW	continuous electrowetting
CFD	computational fluid dynamics
CLSVOF	coupling the level-set function with the VOF methods
CMC	critical micelle concentration
CSF	surface tension force
CVs	control volumes
DI	deionized

EDL	electric double layer
EW	electrowetting
EWOD	electrowetting-on-dielectric
FCT	flux-corrected transport
FVM	finite volume method
GMC	global mass correction
HJ	Hamilton-Jacobi
LBM	lattice Boltzmann method
LLF	local Lax-Friedrichs schemes
LSM	level-set method
L-PLIC	Lagrangian piecewise-linear interface reconstruction
NDV	Normalized Variable Diagram
PC	personal computer
PCB	printed circuit board
PDMS	polydimethylsiloxane
PLS	particle level-set method
PMMA	Polymethyl methacrylate
PROST	Parabolic Reconstruction of Surface Tension
TCP	thermocapillary pump
TVD	total variation diminishing
VOF	volume of fluid

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CHAPTER 1

INTRODUCTION

1.1 Background

Over the last decade, multiphase flows in microscale attract much attention due to the development of microfluidics in the applications of life sciences and analytical chemistry [1-3]. For example, microdroplets can represent a signal by decoding and encoding with their sequences in microfluidic networks [4]. Recently water-in-oil-in-water (W/O/W) droplets were used to avoid contamination from surfactant to form the spherical calcium alginate microbeads successfully [5]. The smart character display was realized with droplets [6].

A microfluidic system is characterized by a low-Reynolds-number flow indicating the laminar flow regime. Microfluidic systems can handle tiny amounts of fluid with remarkable small dimension ranging from millimeters to nanometers. Consequently, the reaction and residence time in these systems reduces significantly to a few seconds or milliseconds. The effects of volume-based inertia and gravity do not play an important role as compared to the macroscale case.

Droplet-based or digital microfluidics is one of the most important branches of microfluidics. The sample liquid is confined in a droplet that is surrounded by another immiscible phase such as air or liquid. In an integrated microchip, many tasks such as mixing [7], reaction [8], transport [9-10], and merging can be carried out in highly

precise and flexible manners. The materials suitable for microchip fabrication are developed include but not limit to PDMS, silicone, PMMA (Polymethyl methacrylate), and glass. Among them, PDMS is most popular, because of its easy and precise fabrication, optical transparency and low cost. However, the drawbacks are deformation under high pressure and swelling in the presence of strong organic solvents, which limit its application [11].

Droplets are generated in microfluidic devices by injecting immiscible fluids into microchannel configurations such as T-junction [12-13], flow focusing [5, 14], or co-flowing [15]. The droplets are the dispersed phase, while another fluid acts as the continuous phase. Generally, the stability of the droplet requires a low interface tension, the continuous phase wets the channel wall, while the dispersed phase does not wet the channel well. [16]. The droplet size and the formation frequency can be controlled directly by adjusting the flow rates, the flow rate ratio, the viscosities and the interfacial tension of the two phases [12, 16].

In the field of droplet-based microfluidics, because inertial effects are not significant in the microscale, much attention has been drawn to an inherently dominant force - surface tension at the interface boundary (liquid-liquid, or liquid-air) - to manipulate microfluidic droplets [17-18]. A number of techniques have been developed based on this attractive strategy to manipulate droplets easily and accurately. The most common methods are based on structural design of the channel, thermocapillarity, electrowetting, magnetism, acoustic, and others. They are flexible, efficient, accurate, rapid and capable of performing complicated and highly parallel

microfluidic tasks. A review presented by Teh et al. summarizes the various manipulation methods for droplet-based microfluidics [19]. Five approaches of droplet manipulation, different functions of droplet performed, and the application in chemical and biological fields was reviewed by Yang et al. [20]. They pointed out that there is still plenty of room for improvement in the field of droplet-based microfluidics.

Numerical methods provide powerful means to explore the microfluidic behaviors where the viscous force and interfacial tension dominate. Numerical simulation helps to understand complex multiphysics phenomena and the dynamics of a microfluidic system. For the purpose of numerical modeling, the above mentioned multiphase flow can be categorized as interface related problems. However, the challenge in solving multiphase flow problems is handling the interface separating the fluids involved and its evolution over time. The irregular interface shape deforms continuously as two phases significantly affect each other as well as the boundary conditions on it. Nevertheless, not only special treatments are needed to deal with the property jump (or gradient) across the interface, but the location of the interface also needs to be tracked at all time.

Numerical simulation has wide applications, and can be used to investigate the droplet formation in a T-shaped junction [21], the impact of viscosity on droplet size and the formation frequency [22], as well as the dynamics of droplet formation [23]. Phenomena such as transport, merging, rupture, separation and collision of droplets have also been modeled numerically [24-30]. Zheng and Zhang used level-set method

to simulate droplet spreading problems on a solid surface with solidification, which also involves a phase change process [31]. Recently, the phenomenon of a droplet falling on the liquid film with the phase field method was numerically simulated [32]. The motion of three coupled phases i.e. particle encapsulated droplets was readily simulated with the level-set method [33]. The problems of droplet evolution on a solid surface involving a triple phase line (gas, liquid and solid contact line) can also be solved numerically. Under a shear flow, Spelt numerically investigated a droplet rolling on a solid surface by adding the contact line velocity in the reinitialization function of the level-set approach [34-35].

Numerical methods also play an import role in solving problems of droplet actuated by external forces. In the presence of a magnetic field, Korlie used a VOF model to simulate the bubbles rising in a ferrofluid and magnetic droplet falling down in a non-magnetic fluid [36]. Among thermally mediated actuation problems, many available models were numerically studied such as the rising process of multiple bubbles [37] and thermally induced droplet breakup behavior [38].

1.2 Objectives and Scopes

This thesis aims to investigate multiphase flows in microchannels numerically and experimentally. First, a numerical scheme for solving droplet-related problems was established. Next, two case studies with representative problems of multiphase systems in microfluidics were carried out. A channel with a train of diffuser/nozzle

structures were employed for the first case study. The rectification behavior of the channel due to the droplet transport was tested. Given the interfacial tension and the viscous force dominating the multiphase flows in micro-scale, the droplet velocity was investigated with different liquid properties. The second case study represents active control of the formation process of droplets. Droplet formation process immersed in a magnetic field was investigated.

Following are the scopes of the thesis:

- (1) Use a level-set method to solve multiphase flow systems in both two-dimensional (2D) and three-dimensional (3D) models.
- (2) Design, fabricate and test diffuser/nozzle arrays for manipulating water droplets in oil. Using a camera setup and pressure sensor to measure the droplet and the corresponding pressure drop across a diffuser/nozzle array with oil flow rate ranging from 30 $\mu\text{l/h}$ to 300 $\mu\text{l/h}$ and water flow rate ranging from 1 $\mu\text{l/h}$ to 10 $\mu\text{l/h}$.
- (3) Use both 2D and 3D numerical models to calculate the droplet passage through the diffuser/nozzle array. Discuss the rectification effect and comparing numerical results with experimental results.
- (4) Fabricate the device with an opening angle of 35° . Mix surfactant of different concentration with mineral oil to change the interfacial tension between oil and water. Use a high speed camera to capture the droplet location in the diffuser/nozzle channels to test and investigate how the interfacial tension affects the droplet velocity. Calculate the influence of viscosity on the droplet velocity.

Measuring the channel dimension with confocal microscopy technique.

- (5) Develop a three-dimensional model to investigate how the magnetic field strength and magnetic susceptibility affect the droplet formation process in a flow focusing configuration. Discuss the hydrodynamics of droplet formation process and comparing with experimental results.

1.3 Outline of the Thesis

Chapter 1 gives a general introduction to the research topic and the significance of multiphase problems for droplet-based microfluidics.

Chapter 2 reviews the different manipulation schemes for droplet-based microfluidics including channel geometry, electrowetting, thermocapillarity, and magnetic actuation. Furthermore, numerical methods solving problems with moving interface are reviewed.

Chapter 3 establishes the numerical scheme using particle level-set method to solve the multiphase problem. The fluid velocity field and the magnetic field were solved with finite volume method. A uniform staggered grid was used. The triple contact line problems were evolved in the validation case by modeling sessile droplets on a solid surface.

Chapter 4 reports the fabrication of the diffuser/nozzle structures for the later experiments. Device configurations and experimental setup are described. Discussion on the droplet deformation and the rectification behavior will be carried out using the

images captured by a camera and data collected from the pressure sensor. Furthermore, comparison between experimental data and numerical data was carried out on the pressure drop and the rectification effect of the diffuser/nozzle structures. In addition, the traveling distance of the droplet advancing front was measured. The effects of the viscosities, droplet size, and the interfacial tension between two immiscible liquids on the droplet velocity are investigated in both experimental and numerical ways.

Chapter 5 discusses the effect of a magnetic field on the droplet shape, droplet size and the velocity field. The interaction among the interfacial tension force, viscous force, pressure difference, and the magnetic force acting at the droplet tip was investigated.

Chapter 6 summarizes the main results and suggests future works of this topic.

CHAPTER 2

LITERATURE REVIEW

2.1 Manipulation of Multiphase Flow and Droplets

Droplet-based manipulation in a channel can be accomplished using passive or active methods. Passive methods are realized by design of the microchannel geometry. Active methods rely on an external physical field such as temperature, electric or magnetic fields. An interfacial tension gradient can also be used for actuation. The gradient can be created by a temperature gradient, non-uniform surfactant concentration, or electric potential.

2.1.1 Channel Geometry

Channel geometry plays the most important role in the droplet formation process. It is reliable to produce highly stable, uniform, and monodisperse droplets in different geometries such as T-junction, flow focusing, and co-flowing configurations. Generally, two immiscible fluids such as oil and water are introduced into the channel and act as the dispersed fluid and the continuous fluid, respectively. The droplets exist in the form of emulsion.

The droplet formation regimes, the physical mechanisms, and the parameters affecting the droplet size were discussed by Christopher and Anna recently in a review paper [16]. The interfacial tension between the two fluids, the pressure difference, and

the viscous force determine the droplet formation process. The droplet size mainly depends on the geometry of the device, the capillary number (Ca), and the flow rate ratio. The droplet formation frequency at a T-junction was discussed by Nisisako et al. [39]. Garstecki et al. investigated the influence of the interfacial tension force, the shear stress and the resistance of the flow on the dispersed fluid tip [12]. The relationship between flow rate and droplet size, the viscosity of the continuous fluid, interfacial tension, as well as the geometry of the junction were experimentally studied.

Manipulation of microdroplets in a microfluidic network can be achieved through appropriate device designs. For example, rapid mixing in a curved winding channel was realized, Fig. 2-1. Mixing in a straight channel is inefficient because of the steady symmetric recirculating flow inside the plugs (black arrows in the rightmost plug in Fig. 2-1). Two counter-rotating vortices with different sizes can be formed in a curved winding channel (the white arrows in Fig. 2-1). This unsteady chaotic flow caused by the time-periodic alternating vortices can result in rapid mixing within millisecond inside droplets [40-41]. Since chaotic advection is based on the channel geometry, the droplet should be large enough to form a plug in the channel. As another typical case, a loop was used to realize coding/decoding of a droplet train (Fig. 2-2) [4]. The occupation of a single droplet in a branch results in an increasing resistance, which ensures that the next droplet enters another branch. A signal can be coded and encoded accurately with the time intervals between two droplets. With a similar concept, droplet traffic at a T-junction was regulated using a bypass which can also build

different resistance between two branches [42]. The breakup of a mother droplet forms two daughter droplets with different volumes at a T-junction because of the asymmetric resistance caused by the different lengths of the branches [43]. Through appropriate microfluidic network designs, logical operations of AND, OR and NOT can be performed with a droplet train [44].

Fig. 2-1 Rapid mixing inside plugs in a curved winding channel [40].

Fig. 2-2 A loop with two branches.

Serial diffuser/nozzle structures have been used as a flow rectification element for pumping applications in continuous-flow microfluidics. Stemme et al. reported for the first time the use of this structure in valveless micropump (Fig. 2-3) [45]. In serial diffuser/nozzle structures with a small opening angle, the pressure loss in the diffuser configuration is lower than that of the nozzle configuration for an identical flow rate. However, for serial diffuser/nozzle structures with a large opening angle such as the

one reported by Gerlach and Wurmus show the opposite behavior, the pressure drop in nozzle configuration is lower [46]. The cut-off angle is around 10° which were proposed by Gerlach [47]. He investigated the relationship between the rectification efficiency and the opening angles. Olsson studies this effect numerically and experimentally [48]. If the opening angle is small, selective rectification is caused by the pressure recovery in the expansion direction. If the opening angle is large, the rectification effect is caused by the “vena-contracta” phenomenon and the gross flow separation. All these works are based on high Reynolds number, and only a single diffuser/nozzle element was investigated. The flow fields at the inlet and outlet are different than those of the case of an array of diffuser/nozzle structures. The difference is caused by the different channel configuration at the inlet and the outlet. It should be noted that there were no droplets or bubbles flowing in the diffuser/nozzle. The influence of geometry on the rectification behavior of a serial diffuser/nozzle structure was investigated experimentally and numerically by Heschel et al. [49] and Olsson et al. [48], respectively. Nguyen and Huang used this concept to design and fabricate a micropump based on printed circuit board (PCB) technology [50]. Groisman and Quake as well as Nguyen et al. reported that the rectification effect is even stronger if a viscoelastic fluid is employed instead of a Newtonian fluid [51-52].

Fig. 2-3 A valveless micropump with a couple of diffuser/nozzle elements [45].

2.1.2 Thermocapillarity

The interfacial tension force is the summation of capillary force and the Marangoni force. The capillary force is resulted from the curvature changes, while the Marangoni force is related to the interfacial tension gradient ($\vec{\nabla}\sigma$). A nonuniform temperature field can induce an interfacial tension gradient on a droplet. The temperature field can be generated either by resistive heaters [9] or focused laser [53-55]. The Marangoni force can drive a droplet to move on a solid surface, inside a cylindrical capillary, or an enclosed channel.

The Marangoni force guides the droplet to move toward a place where the interfacial tension is lower. On an open hydrophilic solid surface, the relation between the advancing velocity of the droplet and temperature gradient are [56]

$$V = C \frac{h_0}{\eta} \left[\frac{\partial}{\partial x} (\sigma_{SA} - \sigma_{SL}) + \frac{\partial}{\partial T} (\sigma_{SA} - \sigma_{SL}) \frac{dT}{dx} + \frac{1}{2} \frac{\partial \sigma}{\partial T} \frac{dT}{dx} \right] \quad (2.1)$$

where C is a constant, h_0 is the thickness of the droplet at the center, σ_{SA} and

σ_{SL} are the surface tension of the solid/air and solid/liquid interfaces, respectively. The parameters of stationary and moving droplets are described in Fig. 2-4. However, the analysis did not fully consider the stress singularity happened at the contact line. Thus, the study was carried out by employing the lubrication theory based on an arbitrary droplet height [57]. For droplet moving on a hydrophobic surface, the effect of the horizontal thermal gradient on the movement was studied experimentally in another paper [58]. The droplets only move when a critical radius R_{cri} is reached. The droplet velocity V increases linearly with R and ∇T . In addition, V reaches a saturation value for large radii R . Here, R_{cri} depends on the contact angle hysteresis and is inversely proportional to ∇T . A very large contact angle hysteresis requires a higher temperature, and thus can cause a droplet to evaporate before it is able to move. Thus, Yakhshi-Tafti et al. used a chemically inert and thermally stable liquid coated on the platform to avoid the direct contact between the droplet and the solid substrate [59]. This method not only can avoid evaporative loss of the droplet and surface contamination due to contact angle hysteresis, but also can increase the droplet movement speed. The method was also adopted to actuate a sessile droplet with magnetism [60]. Darhuber et al. demonstrated thermal actuation to manipulate a continuous stream of discrete droplets on a solid substrate by electronically addressable microheater arrays. The method can electronically control the direction, timing, and speed of the streams and droplets [61].

Fig. 2-4 Thermocapillarity: (a) stationary droplet on homogenous surface; (b) droplet was moved by thermal gradient [56].

Fig. 2-5 Schematic of principle of thermocapillarity induced drop motion in a closed channel [9]. Γ_i is the interface between the air and liquid, P_{L1} is receding-edge pressure on the interface, P_{L2} is advancing-edge pressure on the interface and P_A is the air pressure.

The droplet can be actuated by thermocapillary force to move inside an enclosed microchannel. Burns et al. used local heating method to realize a thermocapillary pump (TCP) which can move discrete nanoliter droplets through square-cross-section microchannels. When the temperature of one end of the droplet increases, the capillary pressure, ΔP_{Ca} , across the liquid-air interface is a function of the surface tension

$$\Delta P_{Ca} = P_A - P_L = \frac{G\sigma \cos \theta}{H_C} \quad (2.2)$$

The parameters are depicted in Fig. 2-5. $G \approx 2(1 + H_C / W_C)$ for square or rectangular channels, H_C is the channel height, W_C is channel width. The interfacial tension σ is a function of temperature

$$\sigma = \sigma_0(1 - CT) \quad (2.3)$$

In Eq. (2.3), C is a positive empirical constant. Heating one end of the droplet in a uniform channel causes a pressure difference that pushes the droplet towards the lower pressure direction with a velocity of

$$V = (h / 6\mu L_D)[\sigma_a \cos \theta_a - \sigma_r \cos \theta_r] \quad (2.4)$$

where μ is the viscosity, L_D is droplet length, and subscripts a and r are the advancing and receding interfaces, respectively. Here, the contact angle hysteresis ($\theta_a \neq \theta_r$) should be reduced for the positive motion. Sammarco and Burns suggested several techniques for assisting TCP to reduce the contact angle hysteresis including the use of different channels, surface treatment, or reducing the external pressure [62]. DeBar and Liepmann fabricated three heaters to test TCP based on the Marangoni effect in a square channel (see Fig. 2-6) [63]. One heater generates a gas bubble, while other heaters control the temperature gradient along the fluid-vapor interface. Fluid flows from the hot region to the cold region until the temperature gradient does not exist. Glockner and Naterer developed a circular TCP with an embedded cyclic heat source inside an adjoining silicon substrate [64]. Unlike the past studies, the heat source is moved instead of being fixed. Very recently, a resistor pattern was employed

to drive the bubble or droplet toward a high surface tension region, to achieve displacing, switching and trapping of droplets [65].

Fig. 2-6 Thermocapillary pump with three heaters [63].

Nguyen and Huang presented the thermocapillary effect of a liquid droplet in a cylindrical capillary which is exposed to a transient temperature field [66]. The formulated one-dimensional analytical model agrees well with experimental measurement. The plug moves out of the high-gradient region before decelerates because of the lower thermal diffusivity. Three heaters wound around a cylindrical capillary were employed by Jiao et al. to study the behavior of a liquid plug [67]. Temperature cycling of heaters allows driving the plug back and forth. The periodic motion could be realized by thermal cycling and mixing inside a liquid plug. Theoretical analysis was given to describe the movement of the liquid plug by considering the spatio-temporal heat transfer effects and temperature dependent surface tension.

The droplet behavior can be changed by localized heating from a single or line

patterns laser. A single laser spot can increase the formed droplet volume in a cross flowing channel, and switch the droplet transport path at a bifurcation due to laser heating [54-55]. The interfacial film dynamics during droplets adhesion because of laser heating was studied by Dixit et al. as shown in Fig. 2-7 [53]. Laser heating can excite the thermal Marangoni effect and the solutal Marangoni effect. The former one can drive droplet to move away from the laser [Fig. 2-7 (a)]. The latter effect leads to the surfactant molecules movement towards the high interfacial tension region [Fig. 2-7 (b)]. In addition, laser heating can lead to asymmetric recirculation flow for efficient mixing inside the droplet [68]. Line patterns laser can be used to guide the droplet to the different channel branches by switching the orientation of the patterns [69].

Fig. 2-7 (a) The droplet is pulled away from the laser under the thermal Marangoni effect. (b) The surfactant molecules move to the high interfacial tension region under the solutal Marangoni effect [53].

The thermocapillary phenomenon can be solved numerically by coupling the energy equation with two-phase flow modeling using the finite volume method [38].

Glockner and Naterer solved a droplet pumping phenomenon inside a channel by considering the Navier-Stokes and energy equations [64]. The thermocapillary micropump for droplets was investigated by finite volume method (FVM) and analytical models. The pressure and velocity boundary conditions were used at the droplet/air interface using a sliding grid. Both numerical and analytical data agreed with each other closely.

2.1.3 Electrowetting

The implementation and control of microheaters are relatively simple. However, this technique has the disadvantage of the large energy consumption due to heat losses. The further drawbacks are the possible evaporation of the liquid droplet and the relatively slow velocity. In contrast, electrowetting allows manipulating droplets with a fast response and low energy consumption. Using electrostatic forces to control the surface tension between a solid electrode and a conducting liquid phase is known as electrowetting. Droplet manipulation is based on the change of the contact angle under an applied electric field [70]. This principle was used to yield a varying focal length of the liquid lens in response to the applied voltage [71]. Active manipulation tasks such as droplet splitting, merging, transporting and formation were realized [10]. The drawback is the complicated fabrication of the electrode structures as well as of the hydrophobic insulator film.

Lippman explored the changes in the interfacial tension as a function of the

applied voltage V_0 by employing a capillary electrometer filled with a mercury-electrolyte (Hg-el) meniscus during his PhD candidature [72]. The change in surface tension $\Delta\sigma_{SL}$ at the electronic conductor-electrolyte interface has the relation $\Delta\sigma_{SL} = -Q\Delta W_i$, where Q is the charge per unit area, and ΔW_i is the interfacial potential difference. The effect of applied voltage on the interfacial tension was explained with an integral version of Lippman's equation. In addition to show the electrowetting effect, Beni and Hackwood also derived the operation parameters in details [73]. Furthermore, they reported the experimental results to demonstrate the concept of electrowetting, and to prove that large contact angle changes can be induced quickly and reversibly. A comparison between electrowetting and electrocapillarity is shown in Fig. 2-8.

Fig. 2-8 Comparison between electrocapillarity and electrowetting [73].

A year later, Beni et al. introduced a new electrowetting effect, continuous electrowetting (CEW) [74]. The CEW liquids can be moved quickly and reversibly with a velocity of 10 cm/s using a relatively low driving voltage of 1 V in one or two dimensions. This reversible phenomenon was also demonstrated by Verheijen and

Prins [75]. The feasibility of a micropump based on electrowetting in a miniaturized closed channel system was first proposed in [76-77]. A prototype device was presented for the study of electrowetting. The experimental results indicated that the designed micropump could generate a pressure as high as 0.01 MPa. Yun et al. successfully demonstrated a micropump actuated by CEW with a low voltage ($< 2.3 V_{pp}$) and low power ($< 30 \mu W$) [78]. The movement of a mercury droplet was operated by an electrolyte as actuator. Besides the actuator, the pump consists of a silicone rubber membrane and copper check valves (Fig. 2-9). At an applied voltage of $2.3 V_{pp}$ and an operation frequency of 10 Hz, the output maximum value of the pump pressure is about 600 Pa.

Fig. 2-9 Schematic view of the micropump based on electrowetting actuation [78].

Pollack and Fair presented electrowetting as a concept for rapid manipulation of discrete microdroplets on a planar surface [18]. The concept directly uses electrodes to control the surface tension throughout the two parallel opposing planar electrodes fabricated on glass (Fig. 2-10). Using the configuration depicted in Fig. 2-10, the group has increased the maximum average droplet velocity to over 10 cm/s [79]. Colgate and Lee investigated and demonstrated the principles and fabrication process

of electrowetting (EW) and electrowetting-on-dielectric (EWOD) and their feasibility using MEMS technology [80]. EW employs the varying electric energy across the electric double layer (EDL) to modify the wettability of a certain electrolyte on a metal electrode, while EWOD employs the varying electric energy across the thin dielectric film between the liquid and conducting substrate to virtually any aqueous liquid. Fig. 2-11 shows the comparison between CEW, EW, and EWOD. An insulation film above the electrode can avoid the electrolysis in EWOD [20]. Therefore, EWOD is one of the most promising methods to actuate droplets. PDMS worked as a dielectric layer was studied experimentally [81]. Moon et al. used EWOD to drive liquids droplets and focused on how to lower the operating voltage [82]. He obtained a contact angle change from 120° to 180° with only 15 V. Cho et al. reported four fundamental fluidic operations: creation, transporting, cutting, and merging of liquid droplets, all with EW. He carried out a series of experiments to verify the criterion of the motion [10]. Mahmud et al. proved that the change of the applied potential across the electrodes of a micro-droplet can reverse the contact angle change in contrast to the conventional electrowetting results [83]. The polarity of the applied potential does not affect the contact angle change. Cooney et al. presented the method to enable more robust droplets translations using the strategy of grounding-from-below, unlike the structure showed in Fig. 2-10 with the ground electrode on the top [84].

Fig. 2-10 Schematic cross-section of the electrowetting microactuator [18].

Fig. 2-11 Electrocapillary : (a) continuous electrowetting (CEW); (b) electrowetting (EW); (c) electrowetting-on-dielectric (EWOD) [80].

A concept of the separation of two different types of particles was presented by Cho et al. [85]. The droplet actuation coupled both electrophoresis and EWOD. Electrophoresis isolated each type of particles at two ends of a mother droplet. The

EWOD actuation enforced splitting of the mother droplet into two daughter droplets, and consequently each type of particles was concentrated.

Recently, an analytical model was formulated for EWOD actuator by considering a droplet travelling between electrodes [86]. The model considers the effect of contact angle hysteresis, and force balance from the filler fluid, solid walls and the actuation force. Electrical actuation using AC voltages were explored by Kumari et al. [87]. Both experimental and analytical results showed the influence of the AC frequency and electrical properties on the droplet velocity.

2.1.4 Magnetism

A magnetic field can be used for manipulating droplets. This method reduces the costs further since no electrodes and heaters need to be integrated. The developed device is simple to prepare. Generally, a permanent magnet or planar coils are needed to provide the driving magnetic field. Magnetic particles inside a droplet are also required. The diameter of the particles ranges from one to few micrometers, which is relatively large. This kind of particles is not homogenously distributed in the droplet, and can be easily clustered or extracted from the suspending liquids under a strong magnetic force. Ferrofluids can avoid these problems. A ferrofluid consists of ferromagnetic particles such as magnetite (Fe_3O_4) with a few nanometers in size and coated by a layer of surfactants in a carrier liquid. Ferrofluid behaves as a liquid in a magnetic field. The development and application of magnetic microfluidics were

recently reviewed by Pamme [88].

Magnetic actuators have a wide use for manipulation of droplets and biological particles. Zborowski et al. built a novel magnetic quadrupole flow sorter [89]. The derived magnetic force was tested to successfully separate flowing cells continuously. Feldmann et al. combined the fluidic system with the electromagnetic actuators to develop a micropump [90].

Traditionally, magnetic field was employed as a separator in microfluidics. Fig. 2-12 shows a conventional design of a magnetic separator. Ahn et al. used a pair of permanent magnets holding removable ferromagnetic wires to separate magnetic particles suspended in a fluid flow [91]. Choi et al. also designed a bio-magnetic particle separator on a glass chip [92]. The relationship between the inductance variation and the number of the separated particles was investigated. The review by Safarik and Safarikova summarized the various strategies employed for separation of droplets in a magnetic field [93]. The position of the magnetic particles was derived as a function of the flow velocity and the frequency as well as the magnitude of the magnetic field strength [94]. The theory triggered further experimental study and application in bead-based assays.

Fig. 2-12 Traditional separator with the permanent magnets [91].

Rida et al. proposed an approach of combining the stationary permanent magnets and an array of simple planar coils for microbeads transport (Fig. 2-13) [95]. The stationary field provides a uniform static magnetic field, while the coils impose a small magnetic gradient field on the microbeads. Thus, the actuated force is supplied by specially arranged coils on a planar surface. When the coils are overlapped, there is no local energy minimum between the two coils. Synchronizing magnetic fields of the coils creates a magnetic field maximum propelling the microbeads to move effectively.

Fig. 2-13 Micro device for the transport of magnetic microbeads: (a) schematic view including (1) bead, (2) planar coils, (3) permanent magnets, and (4) metallic sheet; (b) arrangement of coils on a long planar of (2) [95].

Droplet manipulation was carried out by Lehmann et al. employing simple coils [96]. The same idea was used for actuating droplet with magnetic microparticles suspended in it. Magnetic particles of different sizes and droplet performance were discussed. Fig. 2-14 depicts the droplet manipulation process. Droplet manipulation tasks such as mixing, fusion, extraction, and condensation can be performed by handling the magnetic beads contained in aqueous droplets using a permanent magnet [97-98]. The basic idea is that the movement of permanent magnet will

correspondingly cause the migration of beads. The transport and separation of bead-containing droplets depend on the magnet speed and magnetic particles loading [60]. Coating an oil layer at the surface can make the droplet splitting easier and increases the speed of the droplet.

Fig. 2-14 Droplet merged, mixed, and split in a sequence cycle [96].

Fig. 2-15 Device used for driving the droplet [99].

A magnetic field can deform a ferrofluid drop [100]. The resulting magnetic force is comparable to the interfacial forces in magnitude. Ferrofluid droplet can be moved by employing two pairs of planar coils [101]. As shown in Fig. 2-15, two coils in y direction with the same current direction to limit the droplet moving inside a virtual channel. Two others coils drive the droplet back and forth. The four coils were fabricated on a double-sided printed circuit board (PCB). Significant deformations were observed for large droplets, low viscosity of the surrounding fluid, and high

driving currents. The magnitude of the current in the actuation coils determines the driving force. The direction of the droplet can be changed by switching the sign of the coil current. Recently, our group reported the effect of magnetowetting on a sessile ferrofluid droplet [102]. A permanent magnet was also used to control the formation process of ferrofluid droplets at a T-junction microchannel [103].

The mathematical formulation for hydrodynamics of ferrofluid was discussed by Rosensweig [100]. The problem of deformation of a ferrofluid droplet in a uniform magnetic field was numerically studied by coupling the magnetic field, the free surface, and the fluid flow in 2D and 3D models [104-105]. The magnetic force can be defined as a body force acting at the interface. A ferrofluid droplet falling down in a non-magnetic fluid was modeled by Korlie et al. [36]. In all these studies, the linearly magnetizable fluids were considered. The ferrofluid droplet shape for non-linear magnetic material was investigated by Afkhami et al. [106].

2.2 Numerical Methods for Interface Tracking in Multiphase Flow

The key challenge in modeling multiphase systems is describing the interface. Recently, different techniques were proposed for interface prediction. Traditionally, the interface separating two phases can be solved using volume-tracking method, front-tracking method, fractional volume of fluid (VOF), phase-field method, lattice Boltzmann method, the level-set method (LSM), and others. Slavov and Dimova categorized these methods into two types: explicit tracking methods and implicit

tracking methods [107]. The interface is explicitly tracked either with a moving grid or massless markers seeded at the interface. Either a moving grid or a moving grid coupled with the fixed grid is employed. The interface resolution depends on the number of points. It includes immersed interface method, front tracking method and boundary integral method,. In the implicit method, a simple function is defined on a fixed grid to capture the interface. The interface location is approximated by the indicator function. The method includes phase-field method, level-set method, and VOF. Many researchers classified the methods into interface tracking methods and interface capturing methods based on whether the computational mesh or massless markers is attached on the interface [108-109]. Garrioch and Baligo proposed to categorize these techniques as the moving font mesh to tack the interface and the fixed mesh to capture the interface location [110]. Each categorization approach summarizes one of characters of the numerical methods for the interface prediction. For example, the level-set method is an implicit, interface capturing and employing the fixed grid method.

2.2.1 Boundary-Integral Method

In the boundary integral method, the evolution of a deformed droplet is calculated by time integrating the fluid velocity of a set of marker points at the interface [25]. The evolution of the marker velocities are governed by a boundary-integral equation instead of solving the velocity field. Thus, the interface is explicitly tracked. The flow

solution is deduced from the information of the discrete points along the interface. The details of boundary integral method were reviewed by Pozrikidis [111]. Its application and development in multiphase fluid flow have been summarized in the literature [112]. The numerical instability and the limited accuracy due to the involvement of the interfacial tension were discussed. However, the presented theory only focused on 2D models. Actually, the previous 3D boundary integral method relying on stationary grid with uniform distributed marker points and cannot resolve the serious changes of the interface such as the droplet breakup. To address this problem, an alternative method employing remeshing algorithms was proposed. One algorithm involves local mesh refinement and reconnection. Another method is utilizing the adaptive discretization algorithm to resolve the interface to fit the deformed drop shape [25]. The method was later refined by minimizing the mesh configurational energy as the surface evolves [113]. This method is accurate in solving the drop breakup and coalescence. The limit of numerical instability was overcome by the method proposed by Zinchenko et al. [114]. A special mesh stabilization method was utilized to solve the 3D extremely deformed interface. Further developments showed that this method is very successful in solving single droplet breakup [115], droplet squeezing among spheres [116], and optical stretching and squeezing of a sessile droplet [117].

2.2.2 Front Tracking Methods

The front tracking method was developed by Glimm's group and Tryggvason's group. Glimm presented a 3D front tracking algorithm to solve Rayleigh-Taylor instability problems [118]. Unverdi and Tryggvason investigated the rising of one and two bubbles [119]. This method needs connected markers to reconstruct the interface grid dynamically in the calculation process. The front grid moves through the fixed grid giving the precise location and the geometry of the interface. The computation of the interfacial tension is carried out on the front grid and transferred to the global grid. As discussed in [119], a separate unstructured triangular grid was introduced to mark the interface position while the computational domain is discretized by a regular fixed rectangular grid, Fig. 2-16. Naturally, the Navier-Stokes equations are solved on the fixed grid to control the motion of the multiphase system. It assumes that the fluid properties are constant in each phase. The interfacial jump condition is added to the momentum equations via a discrete delta function along the smooth interface. This multi-grid method shows that the front-tracking method is complex. Furthermore, this method can not solve thin film resulting from the droplet rupture. A topological change algorithm near the interface needs to be considered. Problems with topological changes involving breakup of drops and jet [120]. A pressure driven cell was modeled as a Newtonian microdroplet with constant surface tension. The topological change of the interface was added by Torres and Brackbill to solve the problem of coalescence of two spherical drops [29]. Esmaeeli and Tryggvason extended this approach to cases with phase change [121-122].

Recently, Mao showed that the 2D discontinuity curves can be tracked in a 1D fashion with a 2D conservative front-tracking method [123]. The front tracking algorithm can also be applied uniformly to N dimensions [124]. A developed front-tracking method advects the unconnected points set [125]. The point set was used to mark the interface. The three-dimensional deformation of a spherical drop was modeled as the test case with topological changes. The improvement of the front tracking method was studied in the volume conservation and multiphase problems [126].

Fig. 2-16 The computational mesh. The 3D rectangular domain uses the uniform structure grid, and the interface uses a separate unstructured 2D grid.

2.2.3 Volume of Fluid Method

A review of volume of fluid method was given by Scardovelli and Zaleski [127]. Hirt and Nichols discussed the VOF techniques for interface capturing in non-uniform mesh [128]. The volume fraction function is introduced and defined in a volume fraction field throughout the whole computational domain. The volume fraction

function is

$$\frac{\partial \zeta}{\partial t} + \vec{u} \cdot \nabla \zeta = 0, \quad (2.5)$$

and can be solved on a fixed-grid. ζ represents the volume fraction and has a value between 0 and 1. Fig. 2-17(a) shows a typical distribution of the volume fraction values. The values are reconstructed to find the interface location. The interface lies within the computational cell with a volume fraction function of $0 < \zeta < 1$, and the location of the interface is 0.5, Fig. 2-17(b). For a volume fraction function of unity, the computational element is fully occupied by the primary phase. In the case of volume fraction function of zero, the computational element is fully occupied by the secondary phase. To solve the function, the initial approximate interface position needs to be found. In addition, both the determination of the weighted viscosity and density for the computational elements and the calculation of the volume flux of the convective terms in the governing equations are obtained by interface reconstruction. The crude reconstruction can generate a large error even with a simple velocity field such as translation or solid body rotation. The reconstruction methods include but not limit to Simple Line or Piecewise Linear Interface Construction (SLIC) [129-130], least-square fit with split Eulerian-Lagrangian advection [131] and Parabolic Reconstruction of Surface Tension (PROST) employing piecewise parabolic curves [132]. Instead of using interface reconstruction, Rudman introduced flux-corrected transport (FCT) method for volume capturing [133].

Fig. 2-17 Volume fraction values (a) and contour of volume fraction $\zeta = 0.01$, $\zeta = 0.21$, $\zeta = 0.5$ and $\zeta = 0.88$ (b).

After the interface reconstruction, the interface movement caused from the underlying flow field need a right advection algorithm to calculate. The discretization of advection term is the main difficulty to guarantee the physical volume fraction distribution and the sharpness of the interface. The lower order scheme can smear the interface due to the numerical diffusion. A higher order scheme can result in numerical oscillations. Gopala et al. reviewed and discussed the advantages and limitations of several volume advection techniques [134]. They are flux-corrected transport algorithm [135], compressive interface capturing scheme [136], gamma differencing scheme [137] and Lagrangian piecewise-linear interface reconstruction (L-PLIC) method [138]. The comparison was based on two practical examples of the Rayleigh–Taylor instability problem and sloshing of a liquid wave. Recently, a high resolution differencing schemes, Normalized Variable Diagram (NDV), was reported to preserve the sharpness of the interface and the boundedness of the volume fraction [139].

The advantage of the VOF method is its superior volume or mass conservation of

each fluid over other methods, for example, the level-set method. The interface could be captured implicitly, thus topological changes can be handled automatically. Problems with interfacial geometrical quantities such as the curvature and the unit normal can be encountered. These quantities are calculated from the volume fraction function which is discontinuously distributed, and are important in the calculation of surface tension. It is not straight forward to calculate these quantities accurately. Fortunately, the level-set method can fix this problem. As results, a number of researchers recently proposed a hybrid algorithm of coupling the level-set function with the VOF methods (CLSVOF) [140-142]. The VOF was used to reconstruct the interface, while the level-set method was used to compute the interface as well as its curvature. However, these methods are not easy to extend to three dimensions. Therefore, NDV was only used for the advection of the volume fraction function. Extending CLSVOF method to multi-dimensional unstructured grid is relatively complex and possible [139]. The three-dimensional shape of a rising bubble in a liquid was studied.

The VOF method can exhibit the high accuracy in the calculation of three dimensional flows. A pinching pendant drop was modeled and obtained a good agreement comparing to the experiment data [130]. The problems of bubble rising in the liquid [142-143], droplets adhering to a vertical wall [139], and a water drop impact onto a deep water pool with wave breaking and plunging [144] were modeled successfully. One of the most popular commercial CFD (computational fluid dynamics) softwares, Fluent, also employed VOF method to calculate multi-phase

flow.

2.2.4 Phase-Field Method

The phase-field method is also called the diffuse-interface method. In phase field method, a phase function $\varphi(\vec{x}, t)$ is introduced to represent the interface $\varphi=0$. Away from the interface, the function values of the two phases are $\varphi=+1$ and $\varphi=-1$, respectively. The evolution of the phase-field function is governed by a fourth order nonlinear parabolic diffusion equation, the Cahn-Hilliard equation [145]:

$$\varphi_t + (\vec{u} \bullet) \varphi = -\sigma \Delta [\Delta \varphi - f(\varphi)]. \quad (2.6)$$

The function $f(\varphi)$ is a polynomial of φ [146]. The method was used to model coalescent of two bubbles. Another alternative governing equation of the phase function is the Allen-Cahn type [147-148]. Because the phase-field method track the sharp interface with a first order approximation, a fine grid is required around the interface.

The advantages and disadvantages of phase-field method are discussed by Slavov and Dimova [107]. This method can be easily extended to a three-dimensional problem, and has a straight-forward extension to arbitrary multi-phase systems [149]. Moreover, the method can handle the topological changes of the interface automatically. Adaptive moving mesh algorithm can be used to increase the efficiency and accuracy of the method [148]. In this method, a mapping between the interface governing equations and the phase field equations is established by performing an asymptotic analysis. Thus, the phase field model only reproduces the motions of the

interface governing equations with the limitation that the density difference is sufficiently small. Also a refined grid is needed to resolve the interface. In contrast, the level-set approach is used to accurately track the interface in a simple fashion.

The recent applications of phase-field method are multiple pinch-offs of a long cylindrical thread at small Reynolds numbers [150], droplet pinch-off in liquid/liquid jet configuration [151], and two drops coalescence evolving four phases [152].

2.2.5 Lattice Boltzmann Method

The lattice Boltzmann method (LBM) is a relatively new simulation technique. The difference between phase field method and LBM only lies in the discretization to the Navier-Stokes equations [32]. The method is successful in dealing with the interface tension and the complex boundaries with a large density ratio between the two phases. The lattice Boltzmann method constructs the kinetic equation of the discrete-velocity distribution function utilizing discrete lattice and discrete time. Thus, it is viewed as a special finite difference scheme for the kinetic equation. But LBM distinguishes itself from the conventional numerical schemes which are based on the discretization of the continuum equations. The LBM is a derivative of the lattice gas theory. A lattice gas theory consists of a regular lattice with particles residing on the nodes. As a result, the N-S equation can be derived from the lattice Boltzmann equation. The macroscopic dynamics of fluid is the behavior of the particles in the system.

An overview of the LBM was presented and discussed by Chen and Doolen [153].

This review discussed the different methods for modeling interfacial tension in a multiphase fluid flow: the free energy model and the interparticle interaction potential model. Lamura employed the first approach to simulate oil-water-surfactant mixtures [154]. The latter method was first proposed by Shan and Chen [155]. Except for the above methods, another one is the He-Shan-Doolen approach [156]. Its limits and advantages were discussed by comparing with front tracking method based on the 2D bubble rising model [157]. Recently, LBM method becomes a popular modeling platform for droplet-based microfluidics. The droplet formation process at a microfluidic T-junction [158-159] and a microfluidic cross-junction [23] was investigated with LBM.

2.2.6 Level-Set Method

Osher and Sethian demonstrated a algorithm of level-set method to track the moving interface on the fixed-grid system [160]. The level-set function $\phi(\vec{x}, t)$ was introduced over the whole domain or near the interface and was defined as a signed shortest normal distance from the interface. Fluid properties can be calculated from the level-set function through a smoothed Heaviside function.

Currently, LSM has been used successfully for handling moving interface and free boundary problems in microfluidics [161]. Topological changes can be captured in a straightforward fashion, and thus the methods are suitable to solve the motion of the interface in both two and three spatial dimensions. We can conclude that LSM is a

powerful tool to solve the complicated deformation and to construct implicit surfaces on a fixed rectangular grid. The challenging problems such as coalescence/rupture of droplet and droplet falling down under difficult conditions can be simulated easily with the level-set method.

Numerically, it is desirable to ensure the level-set function approach a signed distance function of the interface. However, the fact is that the level-set function will generally deviate from a signed distance function. Moreover, the discretization of the level-set equation can cause numerical dissipation and inaccurate identification of interfaces. Flat or steep regions often occur near the interface. Generally, a reinitialization equation is required to ensure the level-set function to be a signed distance function to the interface with some degree of accuracy [27].

The evolution of the level-set and the reinitialization equation can be solved numerically with finite volume method [33, 161]. This method is susceptible to numerical fluctuations in some situations. Thus, many researchers prefer to add artificial viscosity as a source term to the level-set equation [109]. Its weakness is pointed out by Osher and Fedkiw [162]. The higher order advection schemes are presented through solving the general Hamilton-Jacobi (HJ) equation. The details are well documented in their book [162]. The gain in accuracy of the higher order schemes is compromised by the long time integration, which is not desirable. Therefore, new techniques need to be developed to reduce the computational time. An approach was introduced to only solve the level-set function within a band $|\phi(\vec{x})| < \gamma$ near the interface, where γ is the width of the interface [163].

Fig. 2-18 Mass error in the level-set method: (a) mass surplus; (b) mass loss.

Even using the higher-order advection schemes to evolve the level-set equation, the mass error still cannot be avoided. Fig. 2-18 depicts the typical examples of mass error. The error is caused because the sharpness of the interface is destroyed by under-resolved regions during the discretisation process. This mass conservation problem can be solved by many methods. One method employs the re-initialization equation as mentioned above. Another different reinitialization procedure is to solve the following equation for the steady state [164],

$$\phi_t + \Delta M_0 (-C + \kappa) |\nabla \phi| = 0 \quad (2.7)$$

where ΔM_0 is the difference of the mass between initial mass and the mass in the droplet deforming process, and κ is the curvature. Unfortunately, this improvement in the mass conservation cannot guarantee the mass conservation of the level-set method in some cases [164]. Therefore, a global mass correction (GMC) technique is introduced [33, 165]. A correction is added to the level-set equation in order to preserve the initial mass. Mass is conserved in a global sense.

Most researchers agree that employing the particle level-set method can improve

mass conservation. A number of particles are seeded into the grid cell to track the characteristic information, and thus can accurately reconstruct the interface where the level-set method failed to preserve mass. Traditionally, the particles are placed throughout the fluid domain and move with the local velocity [166-167]. This method is successful in solving the free surface problems i.e. falling free surface, splashing of water, and water sloshing in a tank. Later, an approach of seeding Lagrangian marker particles within a narrow band around the interface was proposed [168].

Recently, level-set methods have been used quite successfully for moving interface as well as multiphase flow in two- or three-dimensional problems: particle-encapsulated droplet transporting in the fluid flow [161], droplet spreading and solidification [31], bubble rising and growth in a stationary fluid [169], droplet falling down [170], and bubble adhering to the solid surface in the fluid [171], topological changes of the interface such as droplet pinching and connection [172], droplet collision and membrane break [173], breakup into smaller droplets [174]. In the field of the droplet-based microfluidics, numerical simulation with level-set method has the potential to play an important role.

2.3 Remarks

Application of diffuser/nozzle structures on the valveless micropump has its advantages over others which employ valves to pump liquid. The concept is passive, robust, and easy to be implemented. Thus, it is particularly important to study the physics of flow inside diffuser/nozzle structures. In the previous work, studies was

mainly focused on the effect of opening angle, the Re numbers, and the property of the liquid in a single phase flow. The objective was to improve the rectification, namely, the pumping efficiency. To our best knowledge, none of the published works used diffuser/nozzle structures for transporting droplets and investigating the droplet behavior as well as the rectification effect for a multiphase system. Due to its novelty and potential significance, rectification effect of diffuser/nozzle structure in a multiphase system is the preferred topic of this thesis.

In some applications, manipulating a multiphase flow by means of passive control has its limitations. Many practical applications use active methods to manipulate droplets. Due to its wireless nature, magnetism is the most simple active concept. For example, controlling of the droplet formation process can be realized by applying a magnetic field rather than changing channel geometry or the flow rate ratio between two immiscible liquids. On the one hand, the response of active manipulation is faster as compared to conventional methods. On the other hand, Magnetic manipulation of a microscale multiphase system is rich in physics, that can be extended to many practical applications. Therefore, coupling magnetism with microfluidics is our another topic of choice.

As mentioned above, the investigation of multi-physics microfluidics involves the small scale, and complex and fast interactions. In practice, it is expensive or impossible to investigate the detailed dynamics of such processes by experiments only. The numerical solution plays a critical role to compensate this deficiency.

Solving mutiphase flow numerically is always a challenge because of the

difficulties in tracking the motion of the interface. It is not surprising that many interface tracking methods have been developed. Among them, the level set has some qualities which can not be substituted by other methods as mentioned in the literature review. Therefore, this method will be used in the study of multiphase liquid systems.

CHAPTER 3

NUMERICAL METHODS FOR MULTIPHASE SYSTEM IN MICROCHANNELS

The properties of multiphase system in microscale are still continuous except the jump at the interface. Thus, the governing equations describing the physical field inside each domain are still working here. This chapter presents the modeling of velocity field of the fluid flow, magnetic field, interface capturing, and even the triple contact line problems. The numerical procedure for solving the interface is described. The numerical methods are validated with both 2D and 3D cases including two phases fluid flow, the coupling of magnetic field and the fluid velocity field, and three-phase involving sessile droplets.

3.1 Mathematical Formulation

3.1.1 Velocity Field

The motions of two immiscible fluids are calculated with two sets of conservation equations within each region. The properties across the interface are discontinued, and can be defined using the smoothed Heaviside function $H(\phi)$ by either an arithmetic mean or a harmonic mean.

The continuity and momentum equations in Cartesian tensor notation for unsteady, viscous, incompressible, immiscible two-phase systems are defined as:

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

$$\frac{\partial}{\partial t}(\rho \vec{u}) + \nabla \cdot (\rho \vec{u} \vec{u}) = -\nabla P + \nabla \cdot [\mu (\nabla \vec{u} + \nabla \vec{u}^T)] + \vec{F} \quad (3.2)$$

where t is the time, $\vec{u}$ is the velocity vector with three components $\vec{u} = (u, v, w)$ in the 3D case, and P is the pressure. The discontinuous properties of the phases in the whole domain can be defined as an arithmetic mean

$$\alpha(\phi) = H\alpha_+ + (1-H)\alpha_-, \quad (3.3)$$

or a harmonic mean

$$\frac{1}{\alpha(\phi)} = \frac{H}{\alpha_+} + \frac{(1-H)}{\alpha_-}. \quad (3.4)$$

The arithmetic mean is used to calculate the density ρ , and the harmonic mean is used to calculate the viscosity μ . Since the viscosity is the momentum diffusion coefficient, just as the thermal conductivity for heat diffusion, harmonic mean should be more accurate for the viscosity. The smoothed Heaviside function $H(\phi)$ is expressed as [33]:

$$H(\phi) = \begin{cases} 0, & \text{if } \phi < -\varepsilon \\ (\phi + \varepsilon) / (2\varepsilon) + \sin(\pi\phi / \varepsilon) / (2\pi), & \text{if } |\phi| < \varepsilon \\ 1, & \text{if } \phi > \varepsilon \end{cases} \quad (3.5)$$

The parameter ε is set to 1.5 of the control volume thickness in a uniform grid.

In Eq.(3.2), the force per unit volume $\vec{F}$ can be the interfacial force $\vec{F}_\sigma$, gravitational force $\vec{F}_g$, and magnetic force $\vec{F}_m$. The interfacial force is determined using the surface tension force (CSF) model [175]:

$$\vec{F}_\sigma = -\sigma \kappa \hat{N}_F D(\vec{x} - \vec{x}_f) \quad (3.6)$$

where, $\hat{N}_F$ is a normal to the interface, and $D(\vec{x} - \vec{x}_f)$ is the delta function that is zero everywhere except at the interface:

$$\hat{N}_F = \frac{\nabla \phi}{|\nabla \phi|} \quad (3.7)$$

$$\kappa = \nabla \bullet \frac{\nabla \phi}{|\nabla \phi|} \quad (3.8)$$

$$D(\vec{x} - \vec{x}_f) = \begin{cases} (1 + \cos(\pi\phi/\varepsilon)) / (2\varepsilon), & \text{if } |\phi| < \varepsilon \\ 0, & \text{otherwise} \end{cases} \quad (3.9)$$

The gravity $\vec{F}_g$ is described as

$$\vec{F}_g = \rho \vec{g} \quad (3.10)$$

3.1.2 Magnetic Field

For the case of a magnetic fluid such as ferrofluid, the magnetic field also needs to be calculated and incorporated into the model. The Maxwell equations for nonconducting fluids are

$$\vec{\nabla} \times \vec{H} = 0 \quad (3.11)$$

$$\vec{\nabla} \bullet \vec{B} = 0 \quad (3.12)$$

where, $\vec{H}$ is the magnetic field strength, and $\vec{B}$ is the magnetic flux density. We consider a ferrofluid domain Ω_+ surrounded by a nonmagnetizable medium Ω_- .

The fields $\vec{B}$ and $\vec{H}$ satisfy

$$\vec{B} = \begin{cases} \mu_0 (\vec{H} + \vec{M}), & \text{if } \Omega_+ \\ \mu_0 (\vec{H}), & \text{if } \Omega_- \end{cases} \quad (3.13)$$

μ_0 is the permeability constant of the free space, and its value is $4\pi \times 10^{-7} \text{ N A}^{-2}$. $\vec{M}$

denotes the magnetization of the ferrofluid. It is assumed to be a linear function of the magnetic field strength

$$\vec{M} = \chi_m \vec{H} \quad (3.14)$$

where χ_m is the ferrofluid susceptibility. The magnetic permeability of the ferrofluid is defined as $\mu_1 = \mu_0 (1 + \chi_m)$. Thus, we have flux density of $\vec{B} = \mu_1 \vec{H}$ inside the ferrofluid. Introducing the magnetic scalar potential ψ in the form of $\vec{H} = -\nabla \psi$ yields

$$\nabla \cdot [(1 + \chi_m) \nabla \psi] = 0 \quad (3.15)$$

The permeability jumps across the interface between two immiscible phases. Thus, the scalar potential changes as the interface is moving. The magnetic susceptibility can be solved within the whole computational domain based on the harmonic mean

$$\frac{1}{1 + \chi_m} = \frac{1 - H}{1 + \chi_{m-}} + \frac{H}{1 + \chi_{m+}}. \quad (3.16)$$

The magnetic force in Eq. (3.2) is calculated by

$$\vec{F}_m = -\frac{1}{2} \mu_0 |\vec{H}|^2 \nabla \chi_m \quad (3.17)$$

Where $\nabla \chi_m = \frac{d\chi_m}{d\phi} \nabla \phi$

$$\frac{d\chi_m}{d\phi} = \frac{(1 + \chi_{m+})(1 + \chi_{m-})(\chi_{m+} - \chi_{m-})D(\phi)}{[(1 + \chi_{m+}) + H(\chi_{m-} - \chi_{m+})]^2} \quad (3.18)$$

The magnetic force only acts on the interface where the discontinuity in magnetic permeability takes place and will vanish when the permeability is constant. The magnetic field boundary condition satisfies

$$\vec{H} \cdot \hat{n} = -\frac{\partial \psi}{\partial n}, \quad \forall \vec{x} \in \partial\Omega \quad (3.19)$$

3.1.3 Interface Modeling

The moving interface Γ_i is traced with the level-set method [160]. The variable $\phi(\vec{x}, t)$ is used to define the interface that separates two regions Ω_+ and Ω_- (Fig. 3-1). Its values are positive inside the interface, and negative outside the interface. The different signs distinguish the two different fluids. Its value is a signed normal distance function $\phi(\vec{x}, t)$ from the interface:

$$\phi(\vec{x}, t) = \begin{cases} < 0, & \text{if } \vec{x} \in \Omega_- \\ 0, & \text{if } \vec{x} \in \Gamma_i \\ > 0, & \text{if } \vec{x} \in \Omega_+ \end{cases} \quad (3.20)$$

where the subscripts (+) and (-) refer to two immiscible phases. The interface Γ_i is represented as the zero level-set of function $\phi(\vec{x}, t)$. Fig. 3-2 shows the contours of the initial constant level-set function at $t = 0$ s. By taking the time derivative of $\phi(\vec{x}, t) = 0$, we get the motion of the interface Γ_i :

$$\frac{\partial \phi}{\partial t} + \vec{u} \cdot \nabla \phi = 0 \quad (3.21)$$

This function can be solved locally around a narrow band across the interface. The signed normal distance function $\phi(\vec{x}, t)$ satisfies $|\nabla \phi| = 1$ all the time.

Fig. 3-1 Schematic of the computational domain.

Fig. 3-2 Contours of constant level-set function at $t = 0$ s.

Numerically, to keep the level-set function to be a signed distance function to the interface, a reinitialization procedure is needed to reset the level-set function to be a signed distance function of the interface. In this thesis, we use the following reinitialization equation for the correction of ϕ at time t^* [27]:

$$\frac{\partial \phi^*}{\partial t^*} = \text{sign}(\phi_0)(1 - |\nabla \phi^*|) \quad (3.22)$$

$$\phi_0(\vec{x}) = \phi(\vec{x}, 0) \quad (3.23)$$

For numerical purposes, it is useful to smooth the signed function as

$$\text{sign}(\phi_0) = \phi / \sqrt{\phi^2 + |\nabla \phi|^2 \Delta x} \quad (3.24)$$

The steady-state solution of the equation will converge to the actual distance, and satisfy $|\nabla \phi| = 1$.

3.1.4 Contact Line Modeling

A liquid drop deposited on a solid surface forms an air-liquid-solid contact line along the three-phase intersection. The angle between the liquid surface and the solid is called the contact angle. Fig. 3-3 shows the investigated numerical models, a liquid

drop immersed in the gas and sitting on the solid surface. The free surface Γ_i separates the domain into two regions, i.e. Ω_+ and Ω_- , each filled with liquid and gas respectively. The droplet sits on the homogeneous surface, and its shape is axisymmetric. Thus, the droplet base is a circle [Fig. 3-3(b)].

Fig. 3-3 Schematic of a sessile droplet on the solid surface in the gas with (a) 2D model and (b) 3D model (x_1 and x_2 : contact points of the 2D model, L_t : contact line of the 3D model, θ : the contact angel of the three phases, h : droplet height, $2b$: droplet base).

The classical static equilibrium shape of a sessile drop is described by the Young's equation. However, numerical investigation of dynamic phenomena of a sessile droplet will encounter problems arising from the dynamic contact angle and the moving contact line. Dussan and Davis showed that if the traditional no-slip condition is enforced at the moving contact line by coupling of Navier-Stokes

equations, the numerical model will result into stress singularity [176]. Furthermore, the dynamic contact angles exhibit a hysteresis. Thus, the contact line speed needs to be prescribed.

The velocity of the contact line was derived in the previous literature using the force balance [177]. The results were utilized to numerically study the droplet spreading and recoiling by Li et al. [178]. The triple contact line problem was solved based on a single phase, and neglect the influence of surrounding gas on the droplet shape. This method does not consider the contact angle hysteresis. Here, we combine the same continuum model describing the moving contact line of a multiphase system to investigate the drop shape. The effect of surrounding gas is considered. The details are described in three-dimensional forms as follow.

To remove the force singularity, the slip law is employed by replacing the no-slip condition. Along the entire solid surface $y=0$, the Navier boundary condition is employed except points whose distances to the contact line are less than one grid size [179]

$$\begin{cases} u = \lambda \frac{\partial u}{\partial y} \\ v = 0 \\ w = \lambda \frac{\partial w}{\partial y} \end{cases} \quad (3.25)$$

where, λ is the slip length. $\lambda=0$ represents the no-slip condition, and $\lambda=\infty$ is the free-slip condition. Here, we assume a partial slip in the vicinity of the solid surface because of the flow friction [180]. In the work of Ren and E [177], λ is the ratio of viscosity and friction coefficient. In the equation (3.25), the velocities parallel

to the solid surface u and w are linear function of the velocity gradient. The velocity normal to the solid surface $v=0$ represents the no penetration boundary condition.

As long as the slip law is used to avoid force singularity, the speed of the contact line must be given to determine the Young's stress and the normal stress of free surface [177]. A simple dynamic velocity was imposed on the points within one grid from the contact line,

$$\begin{cases} u = \frac{\sigma}{\omega} (\cos \theta^* - \cos \theta) n_x e^{-d_c^2 / \Delta x^2} \\ v = 0 \\ w = \frac{\sigma}{\omega} (\cos \theta^* - \cos \theta) n_z e^{-d_c^2 / \Delta z^2} \end{cases} \quad (3.26)$$

where σ is the surface tension, and ω is the effective friction coefficient, d_c is the distance of the points to the contact line at the surface of $y=0$, n_x is the component of the normal to the contact line in x direction, and n_z is the component of the normal to the contact line in the z direction, θ^* is the equilibrium static contact angle, θ is the dynamic contact angle (Fig. 3-3), which is determined by the relationship:

$$\cos \theta = -\hat{n}_y \cdot \nabla \phi \quad (3.27)$$

where $\hat{n}_y$ is the unit normal vector of the solid surface. This method prescribing the dynamic contact line is relatively simple. This method is effective and easy to implement numerically. Since contact angle hysteresis is neglected, the advancing and receding contact angles do not need to be prescribed. The condition $|\nabla \phi|=1$ is imposed on the solid boundary. The typical contour of the level-set values near the

interface is shown in Fig. 3-4.

Fig. 3-4 The level-set contour around the free surface in 2D model.

3.2 Numerical Procedures

3.2.1 Physical Fields

The governing equations the fluid flow and the magnetic field are special cases of the general transport equation

$$\underbrace{\frac{\partial(\rho\Phi)}{\partial t}}_{\text{unsteady term}} + \underbrace{\nabla \cdot (\rho \vec{u} \Phi)}_{\text{convection term}} = \underbrace{\nabla \cdot (\Gamma \nabla \Phi)}_{\text{diffusion term}} + \underbrace{S}_{\text{source term}} \quad (3.28)$$

where Φ is the dependent variable and can stand for a velocity component, magnetic potential, temperature, or a displacement component of a linear-elastic isotropic material. Γ represents the diffusion coefficient. The source term S is the left terms except unsteady, convection, and diffusion terms. The equations have the same form as the general transport Eq. (3.28) and can be solved numerically. The 3D expression of Eq. (3.28) in Cartesian coordinate system is

$$\begin{aligned} \frac{\partial(\rho\Phi)}{\partial t} + \frac{\partial}{\partial x}(\rho u \Phi) + \frac{\partial}{\partial y}(\rho v \Phi) + \frac{\partial}{\partial z}(\rho w \Phi) \\ = \frac{\partial}{\partial x} \left(\Gamma \frac{\partial \Phi}{\partial x} \right) + \frac{\partial}{\partial y} \left(\Gamma \frac{\partial \Phi}{\partial y} \right) + \frac{\partial}{\partial z} \left(\Gamma \frac{\partial \Phi}{\partial z} \right) + S \end{aligned} \quad (3.29)$$

The values of density ρ , dependent variable Φ , diffusion coefficient Γ , source term S of continuity equation, momentum equation, and Maxwell equation are

tabulated in Table 3-1.

Table 3-1 Density, dependent variable, diffusion coefficient, and source term.

	ρ	Φ	Γ	S
Continuity Eq. (3.1)	ρ	1	0	0
Momentum Eq. (3.2)	ρ	u	μ	$\frac{\partial \mu}{\partial x} \frac{\partial u}{\partial x} + \frac{\partial \mu}{\partial y} \frac{\partial v}{\partial x} + \frac{\partial \mu}{\partial z} \frac{\partial w}{\partial x} + F_x$
	ρ	v	μ	$\frac{\partial \mu}{\partial x} \frac{\partial u}{\partial y} + \frac{\partial \mu}{\partial y} \frac{\partial v}{\partial y} + \frac{\partial \mu}{\partial z} \frac{\partial w}{\partial y} + F_y$
	ρ	w	μ	$\frac{\partial \mu}{\partial x} \frac{\partial u}{\partial z} + \frac{\partial \mu}{\partial y} \frac{\partial v}{\partial z} + \frac{\partial \mu}{\partial z} \frac{\partial w}{\partial z} + F_z$
Maxwell's Eq. (3.15)	0	ψ	$1 + \chi_m$	0

The Eq. (3.28) is solved on the Cartesian staggered grid by a finite volume method [181]. The computational domain is divided into a number of control volumes as shown in Fig. 3-5. The solid lines are the grid lines, and each grid point is the center of the control volume. Δx and Δy are grid sizes, and δx and δy are widths of the control volume. The control volume P has four neighbors labeled by W, E, N, and S, and four surfaces labeled by w, e, n, and s. The grid points store the scale variables, and control volume surfaces store the velocities.

Given the uniform grid, the channel structure is defined by the block-off approach of Patankar [181]. The velocity-pressure coupling utilizes the SIMPLER algorithm. The combined convection-diffusion effect is predicted by second-order accurate TVD (total variation diminishing) discretisation schemes which are given in the Appendix. The time integration used a fully implicit scheme.

Fig. 3-5 Control volumes for the two dimensional situation.

3.2.2 Interface Solver

The level-set equation (3.21) and the reinitialization equation (3.22) are special cases of the general Hamilton-Jacobi equation

$$\phi_t + H_1(\nabla \phi) = 0 \quad (3.30)$$

where $H_1(\nabla \phi)$ is the Hamiltonian, and it can be modeled as the convection term of level-set equation (3.21)

$$H_1(\nabla \phi) = \vec{u} \cdot \nabla \phi \quad (3.31)$$

This function is approximated with the local Lax-Friedrichs schemes (LLF) given in [182]. In 3D, the Hamiltonian function has the form of

$$\hat{H}^{LLF}(\phi_x^-, \phi_x^+, \phi_y^-, \phi_y^+, \phi_z^-, \phi_z^+) = H\left(\frac{\phi_x^+ + \phi_x^-}{2}, \frac{\phi_y^+ + \phi_y^-}{2}, \frac{\phi_z^+ + \phi_z^-}{2}\right) - \alpha^x \left(\frac{\phi_x^+ - \phi_x^-}{2}\right) - \alpha^y \left(\frac{\phi_y^+ - \phi_y^-}{2}\right) - \alpha^z \left(\frac{\phi_z^+ - \phi_z^-}{2}\right) \quad (3.32)$$

$$\begin{aligned} \alpha^x &= \max |H_{\phi_x}(\phi_x, \phi_y, \phi_z)| \\ \alpha^y &= \max |H_{\phi_y}(\phi_x, \phi_y, \phi_z)| \\ \alpha^z &= \max |H_{\phi_z}(\phi_x, \phi_y, \phi_z)| \end{aligned} \quad (3.33)$$

$\hat{H}^{LLF}$ is a numerical approximation of $H_1(\nabla\phi)$. Here, α^x , α^y and α^z are set to the maximum values of $|u|$, $|v|$, and $|w|$ in the neighbor nodes since $H_{\phi_x} = u$, $H_{\phi_y} = v$ and $H_{\phi_z} = w$.

Eq. (3.22) is another example of Eq. (3.30) with

$$H_1(\nabla\phi) = \text{sign}(\phi_0)(|\nabla\phi| - 1), \quad (3.34)$$

which is approximated by the Godunov's scheme [182]

$$\begin{aligned} \hat{H}^G(\phi_x^-, \phi_x^+, \phi_y^-, \phi_y^+, \phi_z^-, \phi_z^+) \\ = \begin{cases} s \left\{ \sqrt{\left[\max(\langle \phi_x^+ \rangle^-, \langle \phi_x^- \rangle^+) \right]^2 + \left[\max(\langle \phi_y^+ \rangle^-, \langle \phi_y^- \rangle^+) \right]^2 + \left[\max(\langle \phi_z^+ \rangle^-, \langle \phi_z^- \rangle^+) \right]^2} - 1 \right\}, & \text{if } \phi_{ijk}^o \geq 0 \\ s \left\{ \sqrt{\left[\max(\langle \phi_x^+ \rangle^+, \langle \phi_x^- \rangle^-) \right]^2 + \left[\max(\langle \phi_y^+ \rangle^+, \langle \phi_y^- \rangle^-) \right]^2 + \left[\max(\langle \phi_z^+ \rangle^+, \langle \phi_z^- \rangle^-) \right]^2} - 1 \right\}, & \text{otherwise} \end{cases} \end{aligned} \quad (3.35)$$

$$\begin{aligned} \langle a \rangle^+ &= \max(a, 0) \\ \langle a \rangle^- &= -\min(a, 0) \\ s &= \text{sign}(\phi) \end{aligned} \quad (3.36)$$

ϕ_x^- , ϕ_x^+ , ϕ_y^- , ϕ_y^+ , ϕ_z^- and ϕ_z^+ are reconstructed with a WENO procedure [182-183]. The total variation nonincreasing (TVN) Runge-Kutta methods were used for time discretization [184]. The Eq. (3.30) can be written as the form of

$$\frac{d\phi}{dt} = L(\phi) \quad (3.37)$$

The second order TVD Runge-Kutta scheme is

$$\phi^1 = \phi^n + \Delta t L(\phi^n) \quad (3.38)$$

$$\phi^{n+1} = \frac{1}{2}\phi^n + \frac{1}{2}(\phi^1 + \Delta t L(\phi^1)) \quad (3.39)$$

The third order TVD Runge-Kutta scheme is

$$\phi^1 = \phi^n + \Delta t L(\phi^n) \quad (3.40)$$

$$\phi^2 = \frac{3}{4}\phi^n + \frac{1}{4}(\phi^1 + \Delta t L(\phi^1)) \quad (3.41)$$

$$\phi^{n+1} = \frac{1}{3}\phi^n + \frac{2}{3}(\phi^2 + \Delta t L(\phi^2)) \quad (3.42)$$

They are solved within a band of certain width around the interface instead of over the entire computational domain to save computational time. This narrow-band approach is introduced by Peng et al. [163]. This approach will not affect the accuracy since the level-set value is important only around the interface. This method can reduce one order of magnitude of the computational time.

3.2.3 Particle Level-Set Method

The particle level-set method (PLS) is employed to alleviate the mass loss problem in the under-resolved region [168, 185]. Spherical particles are employed to track the characteristic information and to reconstruct the interface properly in three dimensions. The radii of the particle is within a range of $r_{\min} < r_p < r_{\max}$ ($r_{\min} = 0.1\Delta x$, $r_{\max} = 0.5\Delta x$). The center locations are $\vec{x}_p$. The particles are randomly seeded into the cells within a band of $b_{\max}$ from the interface ($b_{\max} = 2\Delta x$ in 3D space, and $b_{\max} = 3\Delta x$ in 2D space). Fig. 3-6(a) shows the result of a random distribution of the particles in

the 2D case.

The seeding particles are then attracted to its corresponding region. The negative particles will be attracted to the $\Omega_- (\phi \leq 0)$ region, the positive particles are attracted to the $\Omega_+ (\phi > 0)$ region as shown in Fig. 3-6(b). The boundary separating the two signed particles is the interface. In order to move the particle in the normal direction to the interface, ϕ_g is introduced:

$$\phi_g = r_{\min} + \text{ran}(\text{iseed}) \bullet (b_{\max} - b_{\min}) \quad (3.43)$$

where $\text{ran}(\text{iseed})$ is a random value between 0 and 1. Thus, $\phi_g \in (\pm b_{\min}, \pm b_{\max})$. The particles are attracted to the isocontour ϕ_g by,

$$\vec{x}_{\text{new}} = \vec{x}_p + \tau [\phi_g - \phi(\vec{x}_p)] \vec{N}(\vec{x}_p) \quad (3.44)$$

The level-set value $\phi(\vec{x}_p)$ and the unit normal $\vec{N}(\vec{x}_p)$ are obtained by the trilinear interpolation method from the values of $\phi(\vec{x}, t)$ and $\hat{N}_F$ at the eight neighboring corner nodes on the underlying grid. Initially, $\tau = 1$. The new particle has to satisfy two conditions: (a) $\vec{x}_{\text{new}}$ does not escape the calculation domain; and (b) $\phi(\vec{x}_{\text{new}}) \in (\pm b_{\min}, \pm b_{\max})$. Otherwise τ is to be reduced by half and Eq. (3.44) is repeated. If the run fails to meet both conditions after a maximum iteration number of 20, the particle is deleted. The next particle is randomly seeded until the particle number reaches 64 in 3D case and 16 in 2D case. The particle radius is defined as

$$r_p = \begin{cases} r_{\max}, & \text{if } S_{\text{new}} > r_{\max} \\ S_{\text{new}}, & \text{if } r_{\min} \leq S_{\text{new}} \leq r_{\max} \\ r_{\min}, & \text{if } S_{\text{new}} < r_{\min} \end{cases} \quad (3.45)$$

where $S_{\text{new}} = \text{sign}[\phi(\vec{x}_{\text{new}})] \phi(\vec{x}_{\text{new}})$. Particle overlapping is permitted. The mean distance between particles is generally less than Δx . These particles offer sub-cell

resolution, i.e. are more accurate in representing the interface than the level-set function. Therefore, the particles can be used to correct the level-set function. A successful particle attachment is shown in Fig. 3-6(b).

Fig. 3-6 Particle seeding (a) initial random seeding and (b) after attachment procedure (red indicate positive particles and blue indicate negative particles).

The particles are advected by the velocity field via

$$\frac{d\vec{x}_p}{dt} = \vec{u}(\vec{x}_p) \quad (3.46)$$

where $\vec{u}(\vec{x}_p)$ is the particle velocity. The velocities are obtained by trilinear interpolation method from the fluid velocities. In each time iteration step, $\phi(\vec{x}, t)$ is calculated from Eq. (3.21) and (3.22) first. Subsequently, the positions of particles are advected to their new spatial locations by applying the second-order TVD Runge-Kutta scheme to Eq. (3.46). In the well-resolved regions, the particles will follow the motion of the interface. However, if the level-set solution is not adequately accurate, the particles may escape across the interface to the opposite side during the advection process. It is therefore necessary to perform particle correction strategies to rebuild the positive region and the negative region. One correction technique was presented by Enright et al. [168]. The weak points of this method were pointed out by

Tran and Udaykumar [186] and Wang et al. [187], who proposed better methods. The negative and positive particles that happen to be in the opposite side of the interface by a distance more than its radius are identified as E_- and E_+ , respectively. The level-set functions with each associated with different sign particles, ϕ_- and ϕ_+ , are updated as

$$\phi_+ = \begin{cases} \max_{\forall p \in E_+} (|\phi_+|, |\phi_p|), & \text{if } \phi_+ > 0 \\ -\min_{\forall p \in E_+} (|\phi_+|, |\phi_p|), & \text{if } \phi_+ < 0 \end{cases} \quad (3.47)$$

$$\phi_- = \begin{cases} \min_{\forall p \in E_-} (|\phi_-|, |\phi_p|), & \text{if } \phi_- > 0 \\ -\max_{\forall p \in E_-} (|\phi_-|, |\phi_p|), & \text{if } \phi_- < 0 \end{cases} \quad (3.48)$$

where $\phi_p = \text{sign}[\phi(\vec{x}_p)](r_p - |\vec{x} - \vec{x}_p|)$. $\phi(\vec{x}_p)$ is the trilinear interpolation result from the values underlying the grid nodes. Finally, particle correction on the level-set function ϕ is

$$\phi = \begin{cases} \phi_-, & \text{if } |\phi_+| > |\phi_-| \\ \phi_+, & \text{if } |\phi_+| \leq |\phi_-| \end{cases} \quad (3.49)$$

After few time iteration steps, because of the interface movement and particle correction, the particles are either removed or added into the cell. Thus, some regions around the interface have fewer particles to track the interfaces as shown in Fig. 3-7(a). Reseeding algorithm is necessary to control the particle distribution at fixed time iteration steps. The particles should be deleted if they are beyond the computational domain or locate in the cell without a corner in a narrow bond b_{max} . If the number of particle is less than 64 in the 3D case, additional particles are seeded into the cell randomly. If the number is more than 64, the values of

$\text{sign}[\phi(\vec{x}_{\text{new}})]\phi(\vec{x}_{\text{new}}) - r_p$ are sorted in ascending order, and larger ones are deleted by heap sorting. Fig. 3-7(b) shows the completed reseeding results from the poor particle distribution depicted in Fig. 3-7(a). The reseeding algorithm is not a perfect method to smooth the interface. It is necessary to randomly seed and attract the particles repeatedly when the reseeding algorithm failed to maintain the smoothness of the interface.

Fig. 3-7 Particle reseeding procedure (a) before reseeding, (b) after reseeding.

3.3 Validations of Numerical Procedures

The mathematical formulation and associated numerical procedures were already discussed. In this section, the present numerical models are tested and validated against the existing numerical results of both 2D and 3D cases. They are proved to be accuracy and available in solving the variety of multiphase systems. The studied cases are two-phase fluid flow, the coupling of fluid flow and the magnetic field, sessile droplets involving three-phase.

3.3.1 2D Bubble Rising in the Liquid

A bubble is suspended in a container of $3d$ in width and $6d$ in height, and filled with

liquid. The bubble has a diameter of $d = 3 \text{ mm}$, and centered at $(x_c, y_c) = (1.5d, 1.8d)$ as shown in Fig. 3-8(a). Because of the buoyancy and the interfacial effect, the bubble rises and deforms from the initial stationary state. To maintain the approximately spherical shape of the bubble, Bond number $Bo (= g \Delta \rho d^2 / \sigma)$ is kept near unity. The properties of both fluids adopt the following values: $\rho_- = 1000 \text{ kg m}^{-3}$, $\rho_+ = 10 \text{ kg m}^{-3}$, $\mu_- = 0.01 \text{ Pa s}$, $\mu_+ = 0.001 \text{ Pa s}$, $\sigma = 0.04 \text{ N m}^{-1}$. These corresponding dimensionless numbers are $\rho_- / \rho_+ = 100$, $\mu_- / \mu_+ = 10$, $Bo = 1$. Only half of the domain is calculated due to the symmetry of the problem. . A mesh of 32×128 control volumes (CVs) with a time step size of $\Delta t = 5 \times 10^{-5} \text{ s}$ is used. The evolution of the bubble is given in Fig. 3-8. The present numerical results agree well with the VOF results given by Korlie et al. [36]. The large density and viscosity ratio involved in this case. The present numerical methods work well in 2D multiphase flow problems.

Fig. 3-8 A bubble rising in the liquid. The dashed line is the present numerical results, and the solid line are the VOF results.

3.3.2 3D Droplet Transport

As another 3D validation case, the results of the present numerical scheme are compared with the numerical results from Fluent. This software employs VOF method to solve the interface movement. Here, a droplet with a diameter of $d = 0.9L$ moving in a straight microchannel was simulated. The channel width $L = 200 \mu\text{m}$ is the characteristic length. The dimension of the channel is $5L \times L$. The droplet center is located at $(x_c, y_c) = (L, 0.5L)$ initially (Fig. 3-9 $t = 0 \text{ ms}$). Another immiscible continuous liquid occupies the entire channel. Initially, both liquids are at rest. A stream of the continuous fluid then enters the channel and its mean velocity is $u_c = 0.1 \text{ ms}^{-1}$, and carries the droplet forward. The parameters of both fluids are set to $\rho_+ = \rho_- = 2.74 \text{ kgm}^{-3}$, $\mu_+ = 0.9 \times 10^{-3} \text{ Pa s}$, $\mu_- = 1.8 \times 10^{-3} \text{ Pa s}$, $\sigma = 3.65 \text{ mN m}^{-1}$. The corresponding dimensionless numbers are $\rho_- / \rho_+ = 1$, $\mu_- / \mu_+ = 2$, $\text{Re} = 0.03$, $\text{Ca} = 0.05$, where $\text{Re} = \rho_- L u_c / \mu_-$, $\text{Ca} = u_c \mu_- / \sigma$. Given the symmetry of the problem, only half of the computational domain is simulated. The symmetry boundary conditions are used. No-slip condition is enforced at the walls. Outflow boundary condition is used at the exit.

A mesh with $100 \times 10 \times 10$ CVs with a time step size of $\Delta t = 4 \times 10^{-6} \text{ s}$ is used in our scheme and Fluent. After carried out grid independent study, we found that the mesh is sufficient to achieve grid and time independent solution as shown in

Fig. 3-9 and Fig. 3-10. Fig. 3-11 shows a good agreement of both results.

Fig. 3-9 Grid independent study of present numerical scheme.

Fig. 3-10 Grid independent study of VOF method with Fluent.

Fig. 3-11 The droplet passes through a straight channel on x-y symmetry plane.

3.3.3 Droplet in a Uniform Magnetic Field

The magnetic potential was obtained by numerically solving Eq. (3.15). We consider a permeable sphere exposed to a uniform magnetic field of strength H_0 (Fig. 3-12). The permeability ratio of two media is assumed to be $\mu_0 / \mu_1 = 1/6$. The magnetic dimensionless Bond number is $B_m = \mu_0 V_d^{1/3} \chi_{m+} H_0^2 / (2\sigma)$, where V_d is the volume of the sphere. The exact potential solution is [188]

$$\begin{cases} \psi_+ = -\frac{3(\mu_0 / \mu_1) H_0}{1 + 2(\mu_0 / \mu_1)} r \cos \theta, & \text{for } \Omega_+ \\ \psi_- = -H_0 r \cos \theta + \frac{1 - \mu_0 / \mu_1}{1 + 2(\mu_0 / \mu_1)} R^3 H_0 \frac{\cos \theta}{r^2}, & \text{for } \Omega_- \end{cases} \quad (3.50)$$

The contour of magnetic potential in and around a permeable sphere was calculated and compared with the exact solution in Fig. 3-13. Our numerical results match the exact solution. The magnetic potential varies across the interface where the permeability jumps in value, and is identical where the permeability is constant.

Fig. 3-12 A permeable sphere placed in a uniform magnetic field of strength H_0 . μ_1 is the permeability of the sphere, and μ_0 is the permeability of the surrounding medium.

Fig. 3-13 The magnetic potential for a permeable sphere placed in a uniform magnetic field.

To consider the coupling of the magnetic field, the velocity field and the evolution of the interface, a rising bubble in the presence of a uniform magnetic field was simulated in 2D (Fig. 3-14). The bubble of a diameter of $d = 2$ cm suspended in a ferrofluid is initially at rest. The computational domain is the whole container of dimensions $3d \times 3d$. The half domain was calculated due to the symmetry. A mesh of 100×200 with $\Delta t = 1.5 \times 10^{-4}$ s was used. The uniform magnetic field is parallel with the gravity and orients from bottom to top. The magnetic flux density is 0.002 T. The magnetic permeability of the ferrofluid is twice that of the bubble. Comparison was made against the solution in [189], both results show good agreement.

Fig. 3-14 A rising bubble in a ferrofluid. The bubble and ferrofluid permeability are $\mu_b = \mu_0$, $\mu_1 = 2\mu_0$, respectively.

For the 3D case, the steady-state shape of the ferrofluid drop was analyzed [190]. We consider a ferrofluid drop suspended in a miscible non-magnetic fluid. Initially, the drop has a spherical shape due to the minimized interfacial tension. An external uniform magnetic field was then applied. The drop is stretched in the direction of the uniform magnetic field until it reaches the stable equilibrium shape. The velocities of the computational domain relax to zero in the steady state. Therefore, the drop deformation depends on the balance of the magnetic force and the interfacial force. Their ratio is described by the dimensionless magnetic Bond number B_m . The numerical results are identical with results of Lavrova [190] as shown in Fig. 3-15.

Fig. 3-15 The equilibrium shape of a ferrofluid drop in a magnetic field (a) comparison with Lavrova's results [190], (b) 3D numerical results.

3.3.4 2D Contact Line Problem

The numerical model with moving contact line was first validated using a 2D model, Fig. 3-3(a). A numerical study was carried out based on the example presented by Li et al. [178]. The initial drop shape is a half circle with a radius of $r = 0.4$ m above the x -axis. Thus, the initial contact angle is 90° . The static contact angle $\theta^* = \pi/4$. The density, the viscosity, the surface tension, the ratio and the slip length are set as $\rho_+ = 1 \text{ kg m}^{-3}$, $\mu_+ = 2 \text{ Pa s}$, $\sigma = 0.5 \text{ N m}^{-1}$, $\sigma/\omega = 1 \text{ m s}^{-1}$, and $\lambda = 1 \text{ m}$, respectively. The gravity was taken as $g = 9.8 \text{ m s}^{-2}$. The above parameters are the same as used by Li et al. [178]. The density of gas is set to $\rho_- = 0.001 \text{ kg m}^{-3}$ to maintain the real density ratio between liquid and gas. The viscosity of gas is assumed to be $\mu_- = 2 \text{ Pa s}$, which is the same as the liquid viscosity. This assumption can dramatically decrease the computation time. The velocity field will relax to zero when the droplet reaches the equilibrium steady state. Thus, the droplet equilibrium shape is mainly determined by the surface tension, the density of the liquid, and the static contact angle. So the assumed values of gas viscosity have no significant influence on the steady shape of the droplet. This conclusion will be proved in the next section.

Results of the grid independence study are shown in Fig. 3-16. The dashed line is the result with a mesh of 256×102 CVs and a time step size of $\Delta t = 1.25 \times 10^{-3} \text{ s}$ throughout the entire computational domain. The solid line used a mesh of 128×51 CVs and $\Delta t = 2.5 \times 10^{-3} \text{ s}$. Initially, there are 102 cells across the droplet diameter for fine mesh, and 51 cells for coarse mesh. The two numerical results are nearly identical. The area losses of both meshes are shown in Fig. 3-17. The coarse

mesh results in a loss of less than 1.5% of its original area at the end of the simulation.

Although the fine mesh can reduce the area loss, adequate numerical accuracy is

reached with a mesh of 128×51 CVs, $\Delta t = 2.5 \times 10^{-3}$ s.

Fig. 3-16 Grid independence study.

Fig. 3-17 The normalized area versus time step.

Next, the problem of droplet spreading on the solid surface is simulated. The comparison of the drop shape is given in Fig. 3-18 resulting from our analysis with that of Li et al. [178]. The results match well in the case without gravity. Discrepancy

occurs if gravity is considered. Because the results proposed by Li et al. seem not to preserve the area. The area loss is calculated and presented in Table 3-2. The area loss is as much as 16.76%, and the loss of our present method is less than 2%. The area loss versus the time step of the present method is shown in Fig. 3-19. The droplet is easy to reach steady state with gravity. Therefore, more iteration steps are needed in the absence of the gravity with the same time step $\Delta t = 2.5 \times 10^{-3}$ s. The evolution of droplet profiles from initial shape to the equilibrium shape is shown in Fig. 3-20.

Table 3-2 2D drop area and area loss of simulation results.

	Area (No g)	Area loss(no g)	Area(With g)	Area loss(with g)
Li et al. [178]	0.251	0.039%	0.209	16.76%
Present method	0.248	1.3%	0.246	1.9%

Fig. 3-18 Droplet spreading with gravity and without gravity. The present results were compared with that of Li et al. [178].

Fig. 3-19 The comparison of area loss between considering and neglecting g .

Fig. 3-20 The changes of the droplet profiles with different iteration step with gravity (a) and without gravity (b).

Another validation case is droplet recoiling with the gravity effect. Recoiling happens on a non-wetting solid surface, and the droplet has a similar spherical cap.

Only the static contact angle is changed to $\theta^* = 3\pi/4$. The other parameters remain the same as in the spreading case. The initial droplet shape is still a half sphere. The comparison results are shown in Fig. 3-21. They matched well except the area of the present result is slightly larger than the results reported by Li et al. [178]. The difference is due to the mass loss as discussed in the last droplet spreading situation (Fig. 3-18).

Fig. 3-21 Droplet recoiling.

These two cases demonstrate that the wetting/nonwetting characteristics of the surface determine the spreading or recoiling behavior of the droplet for the same droplet size.

3.3.5 Sessile Droplet

In the experiment of Extrand and Moon [191], three deionized (DI) water drops were deposited on the solid surface. The volumes were $V_d = 1 \mu\text{l}$, $50 \mu\text{l}$, $2000 \mu\text{l}$ respectively. If we assume the initial drops are half sphere, the radii are summarized in Table 3-3. The solid surface is hydrophobic, and the static contact angle is $\theta^* = 108^\circ$. The properties of the liquids used in the simulation are $\rho_+ = 998 \text{ kg m}^{-3}$, $\rho_- = 1.25 \text{ kg m}^{-3}$, $\mu_+ = \mu_- = 2 \text{ Pa s}$, $\sigma = 0.072 \text{ N m}^{-1}$. The slip length is one grid distance $\lambda = \Delta x$ [35, 192]. As an input parameter, Spelt discussed the influence of the slip length λ value on the contact line region [34].

Table 3-3 The Volumes, Radii, and Bond number Bo of the investigated drops.

	Volume (μl)	Radii(mm)	Bo
Drop 1	1	0.62	0.052
Drop 2	50	2.88	1.127
Drop 3	2000	9.85	13.18

The droplet was first studied based on a 2D model. The initial drop is a semi circle sitting on the solid surface. Only half of the domain is simulated due to symmetry of the problem,. Before further investigation, the viscosity effect on the computational time and the final droplet shape was first studied. As shown in Fig. 3-22, the viscosity effect on the equilibrium steady-state shape is negligible. However, in the transient process, the viscosity magnitude plays an important role before the fluid velocity relaxes to zero, i.e. before the droplet reaches its steady state.

Employing real viscosities of water and air, $\mu_+ = 1 \text{ mPa s}$, and $\mu_- = 0.01 \text{ mPa s}$, results into the drop height oscillation before reaching the steady shape [Fig. 3-23(a)]. This unsteady oscillation is the results of exchanges between potential energy and the kinetic energy until the droplet reaches steady state. In this process, the energy dissipates because of the fluid friction. In the simulation, the time step is $\Delta t = 1 \times 10^{-6} \text{ s}$. The computational time is dramatically increased up to 5 times compared to the higher viscosity $\mu_+ = \mu_- = 2 \text{ Pa s}$. Fig. 3-23(b) shows the changes of drop height under higher viscosity conditions. No oscillation happens, and the high viscosity can dissipate the total energy quickly because of the stronger friction. The time step increases to $\Delta t = 1 \times 10^{-4} \text{ s}$, and was determined by fluids parameters and the grid size. The mass loss is much less than 0.5%. Therefore, the higher viscosity is adopted in the later simulation cases instead of the real experimental parameters. The main reason is that the higher viscosity can save the computational time in the simulation since the final equilibrium shape is our main objective.

Fig. 3-22 The drop equilibrium shape under different viscosities.

Fig. 3-23 Evolution of the droplet height in the transient process: (a) oscillation at low viscosity; (b) damping at high viscosity.

In order to get the stable state of the droplet of $\mu_+ = 1 \text{ mPa s}$, and $\mu_- = 0.01 \text{ mPa s}$, and to avoid serious mass loss, the global volume (or mass) correction procedure is used. The particle level set method is not used here. A dimensionless volume correction factor V_{cor} is used to ensure mass conservation, and is given as [33]

$$V_{cor} = \text{sign}(\phi_1) \frac{V_{ori} - V_{cur}}{V_{ori}} \quad (3.51)$$

where V_{ori} is the original droplet volume, V_{cur} is the current volume and $\text{sign}(\phi_1)$ is the sign of distance function of phase 1. The “steady-state” level-set function ϕ' is obtained using,

$$\frac{\partial \phi'}{\partial t'} = V_{cor} \quad (3.52)$$

The volume of phase 1 can be calculated using

$$V_{cur} = H \Delta V \quad (3.53)$$

where V_{cor} is the volume correction factor, t' is a pseudo-time. The initial value of this equation is the solution of Eq. (3.22).

For the small drop $V_d = 1 \mu\text{l}$, recoiling happens as shown in Fig. 3-24(a).

Increasing the drop size to $V_d = 50 \mu\text{l}$, the shape begins to spread to a puddle form

instead of remaining as a spherical cap, Fig. 3-24(b). If the drop volume is sufficiently large, the shape assumes a puddle form under the influence of gravity, Fig. 3-24(c). This phenomenon can be explained with a dimensionless number, the Bond number $Bo = \rho g R^2 / \sigma$, where R is of the order of the drop radius. If $Bo < 1$, the drop remains approximately spherical, and displays recoiling. Otherwise, the gravity flattens the drop on the solid surface, and it spreads to a puddle form. The values of Bo of three drops are shown in Table 3-3. The size of the second drop is the transient point for drop beginning to spreading. The capillary length of the system $l = \sqrt{\frac{\sigma}{\rho g}}$ is 2.7 mm. If the drop radius is less than this value, the drop assumes a spherical shape. If the radius is larger than the capillary length, the drop assumes a puddle shape. These drop behavior is consistent with the experiment results reported by Extrand and Moon which indicate that the volume transition point was $V_d = 39 \mu\text{l}$ [191], and the spreading phenomena occurred with a base diameter of $2b = 4.3 \text{ mm}$ and a height of $h = 2.7 \text{ mm}$.

Fig. 3-24 Equilibrium shape of droplets with initial radii of (a) $r = 0.62$ mm; (b) $r = 2.88$ mm; and (c) $r = 9.85$ mm.

If the problem is solved in a 2D Cartesian coordinate, the numerical results can not be validated, because the 2D drops have a cylindrical shape instead of the actual 3D spherical shape. The 2D model is not axial symmetric. To describe the problems more accurately, three dimensional models were built. The initial drop shape is a half sphere. The radii and the liquid properties are the same as those of the 2D models.

One quarter of drop was simulated according its symmetry. The numerical results are shown in Fig. 3-25. The simulated results show a good agreement with the experiment observation [191]. Table 3-4 compares the drop height and the base diameter resulting from numerical simulation and experiments.

Fig. 3-25 Drop shapes of 3D numerical results (a) $V_d = 1 \mu\text{l}$; (b) $V_d = 50 \mu\text{l}$; (c) $V_d = 2000 \mu\text{l}$.

Table 3-4 Drop height (h) and base diameter ($2b$) for a sessile drop sitting on a solid surface.

	Present (2D)		Present (3D)		Extrand and Moon [191]	
	$2b$ (mm)	h (mm)	$2b$ (mm)	h (mm)	$2b$ (mm)	h (mm)
Drop 1	1.02	0.69	1.02	0.72	1.052 ± 0.002	0.82 ± 0.02
Drop 2	5.54	2.82	5.26	2.9	5.46 ± 0.1	2.91 ± 0.04
Drop 3	33.6	4.85	23.6	5.1	26.52 ± 1.35	4.56 ± 0.05

In conclusion, the equilibrium steady-state drop shape is mainly determined by its size, surface tension, gravity, and wettability of the solid surface. For a fixed droplet size, the droplet can display spreading or recoiling depending on the wetting/nonwetting characteristics of the solid surface (see 3.3.4). On the non-wetting surface, the drop shape results from the equilibrium between the surface tension and the gravity. A drop with sufficiently small volume assumes a spherical shape because the surface tension dominates, while the shape of a larger drop is controlled by both gravitational force and surface tension.

CHAPTER 4

DROPLET MOVING IN A DIFFUSER/NOZZLE ARRAY

The first objective of this chapter is to investigate the effect of channel structure on the droplet behavior, i.e. its deformation and the corresponding pressure drop. The rectification effect was investigated in both situations of with and without the droplets travelling in the channel. They are quantified in both experimental and numerical results. Three different opening angles 15° , 30° , and 45° are selected. The pressure sensor is used to measure the pressure drop of a series of diffuser/nozzle structures with different flow rate of both continuous phase and dispersed phase.

The second objective is to study the influence of capillary number on the droplet velocity. Mineral oil mixed with different surfactant concentration is used to vary the interfacial tension. The diffuser/nozzle structures with different opening angles for continuous single-phase flow have been tested [52]. According to the study, an opening angle of 35° exhibits the largest rectification effect. Thus, this opening angle is selected in this study.

4.1 Problem Formulation and Numerical Model

4.1.1 Device Description

Fig. 4-1(a) shows the layout of the device used in the investigations. Each test device measures $10\text{ mm}\times 10\text{ mm}$. The test section is a series of ten identical diffuser/nozzle

structures. All structures have the same depth of 50 μm . Two pairs of oil and water ports form two T junctions to generate droplets in both directions. Fig. 4-1(b) shows the dimensions of the T-junction used in our later experiments. Water as the dispersed phase is injected through a 40 μm wide microchannel into the carrier channel with a width of 150 μm .

Two pressure ports are located at the two ends of the test section to allow access to a differential pressure sensor which is used to measure the pressure drop. Another purpose of pressure ports is their use for the injection of the continuous phase to accelerate the droplets. Consequently, the distance between droplets will be increased. This allows only one droplet to be in the test section even under the condition of a lower flow rate ratio between mineral oil and DI water.

Fig. 4-1 Device schematic for investigation of microdroplets in diffuser/nozzle structures: (a) microchannel layout; (b) T-junction used to form microdroplet.

4.1.2 Two-Dimensional Model

A two-dimensional model for the relatively low aspect ratio of our case gives sufficient qualitative insights into the physics of the problem at a reasonable computing time. The 2D model is suitable for the investigation of the influence of the geometry on the deformation of the droplet. The dimensions of the microchannels follow those of the fabricated devices as depicted in Fig. 4-2. All three designs with different opening angles $\alpha=15^\circ$, 30° and 45° were considered. The whole computational domain consists of a 2D uniform mesh. In all devices, the straight microchannels outside of the test section have the same width of $150\text{ }\mu\text{m}$. In order to reduce computational time, instead of ten only three diffuser/nozzle structures were considered in the simulation.

Fig. 4-2 Dimensions of diffuser/nozzle structures microchannels (a) $\alpha = 15^\circ$; (b) $\alpha = 30^\circ$; (c) $\alpha = 45^\circ$.

Two phases, the carrier fluid (mineral oil) and the dispersed fluid (water), are involved in the simulation. In the experiments, the carrier fluid (oil) always wet the channel wall. An oil film exists between the droplet (water) and the channel wall. Thus the contact angle at the wall/oil/water contact line is neglected. The droplet is pressure driven via the immiscible carrier fluid and squeezed through the diffuser/nozzle structures. The hydrodynamic force of the carrier fluid flow exerts on the droplet in the form of surface stress. Driven by the surface force, the droplet moves and simultaneously deforms.

Eqs. (3.1) and (3.2) are employed to solve the velocity field. The particle level-set method is used to capture the interface evolution. The boundary conditions are defined as described in the section 3.3.2. At the inlet, instead of using the uniform velocity, the carrier flow is assumed to be fully developed with an average velocity given by

$$\bar{u}_c = \frac{q_c}{W_c H_c} \quad (4.1)$$

where q_c is the volumetric flow rate controlled by the syringe pump, W_c is the channel width, and H_c is the channel height. Since the channel is oblique and the uniform grid is used, the boundary conditions are carried out with the block-off approach of Patankar [181].

The Reynolds number is defined here as

$$\text{Re} = \frac{2\rho q_c}{\mu_c (H_c + W_c)} \quad (4.2)$$

In the conducted experiments, the maximum Reynolds number is approximately

0.032 corresponding to the highest oil flow rate of 300 $\mu\text{l/h}$. The Reynolds number Re is low enough to warrant an assumption of laminar flow. The droplets size was extracted from the images captured using a (coupled charge device) CCD camera during the experiments. A program was written in MATLAB for this purpose. The initial droplet can be a circle [Fig. 4-3(a)], or two half circles sandwiching a rectangle [Fig. 4-3(c)]. After few time steps, the droplet shape was transformed from the initial defined shape to the original shape as observed in the experiment [Fig. 4-3(b) (d)]. Simulations were performed for flow rate ratios between water and oil of 3 $\mu\text{l/h}$: 90 $\mu\text{l/h}$, 6 $\mu\text{l/h}$: 180 $\mu\text{l/h}$, and 9 $\mu\text{l/h}$: 270 $\mu\text{l/h}$ for both configurations with different droplet sizes. It is worth to note that the flow rate ratio is fixed at 1 : 30. This large flow rate ratio is selected to ensure that there is only one droplet in the diffuser/nozzle microchannel (i.e. the test section) at any given time. With this, complex dynamic interactions between microdroplets are avoided. The high flow rate ratio only allows one droplet inside the channel in order to compare with the simulation data.

Fig. 4-3 (a) Initial droplet shape and (b) deformed droplet shape.

2D numerical simulations were performed on a uniform grid. Fig. 4-4 shows grid independent study for a nozzle configuration with an opening angle of $\alpha = 15^\circ$. Two different meshes of 470×40 CVs with a time step size of $\Delta t = 4 \times 10^{-5}$ s and 940×80 CVs with $\Delta t = 2 \times 10^{-5}$ s were used. The difference between the solutions of these meshes is small. Therefore, a mesh of 470×40 CVs with $\Delta t = 4 \times 10^{-5}$ s is sufficient to capture all the essential flow features. The same grid size in space and time step are used for other serial diffuser/nozzle microchannels.

Fig. 4-4 2D Grid independent study with $\alpha = 15^\circ$ in nozzle direction (solid line: 470×40 CVs with $\Delta t = 4 \times 10^{-5}$ s, dotted line: 940×80 CVs with $\Delta t = 2 \times 10^{-5}$ s).

4.1.3 Three-Dimensional Model

The 3D model of the first two structures of the array based on the measured geometry of the test device was built (Fig. 4-5). Because the real heights of the droplets were unknown, different lengths with the same height were assumed initially during the simulation. After a few time steps, the height and length of droplets can shrink to their real values. Then the droplet which has same size as displayed in the experiment was selected to continue the simulation. The cross section of the channel is assumed to be rectangular. Due to the symmetry, the one-quarter model as highlighted in Fig. 4-5 was considered. Symmetry boundary conditions are applied at $y = 0$ and $z = 0$. At the inlet, the oil flow is fully developed with an average velocity of $\bar{u}_c$.

Fig. 4-5 Numerical model (nozzle direction). The shaded region is the simulation domain, and it is one quarter of the whole model.

The grid independent study was also carried out in the nozzle direction. The flow rate ratio is 300 $\mu\text{l/h}$: 10 $\mu\text{l/h}$ between oil and water, and the surfactant concentration is 1.0% (w/w), the interfacial tension is 4.45 mN/m. Two different meshes of 227 $\times$ 34 $\times$ 18 CVs and a time step size of $\Delta t = 4 \times 10^{-5}$ s as well as 354 $\times$ 53 $\times$ 27 CVs with $\Delta t = 2 \times 10^{-5}$ s were used. Fig. 4-6 shows that the solution difference is very small.

Thus the mesh of $227 \times 34 \times 18$ CVs and $\Delta t = 4 \times 10^{-5}$ s is sufficient. Similar mesh density will be used in the other cases with different concentrations in both flow directions.

Fig. 4-6 3D Grid independent study in nozzle direction (solid line: $227 \times 34 \times 18$ CVs with $\Delta t = 4 \times 10^{-5}$ s, and dashed line: $354 \times 53 \times 27$ CVs with $\Delta t = 2 \times 10^{-5}$ s).

4.2 Materials and Experimental Setup

4.2.1 Device Fabrication

The test chip device was fabricated with PDMS (Sylgard 184, Dow Corning) using soft lithography techniques as described in Fig. 4-7. The photo mask was designed with AutoCAD and printed on a 5×5 in² plastic transparency film with a resolution of 8000 dpi. SU-8 with 2050 resists was spin coated on a cleaned 4" silicon wafer. After a soft bake to harden the SU-8 layer [Fig. 4-7(a)], UV exposure was carried out with transparency mask for 35 s [Fig. 4-7 (b)]. Developing in isopropanol results in the SU-8 mold for the later PDMS device [Fig. 4-7(c)]. It is called master mould [Fig. 4-7(g)]. The mixture of PDMS and curing agent with a weight ratio of 10 : 1 was then poured over the mould [Fig. 4-7(d)]. The thickness of PDMS was kept around 0.5cm to avoid the leakage at the fluidic interconnects in the later experiment. After curing,

the PDMS part was peeled off from the mould. The six ports depicted in Fig. 4-1(a) with 0.75 mm diameter were punched into the PDMS device [Fig. 4-7(e)]. The patterned PDMS surface and another blank PDMS piece were treated in oxygen plasma for 30 seconds at a power 100 W. Then two PDMS pieces were subsequently brought to contact [Fig. 4-7(f)]. An additional heat treatment returns the channel surface to a hydrophobic state.

Fig. 4-7 Fabrication of microchannels with soft lithography: (a) spin coating a silicon wafer with SU-8; (b) UV exposure with a clear mask; (c) development of SU-8 mold; (d) pouring PDMS on the mold; (e) peeling off the PDMS; (f) surface treatment of two pieces of PDMS with oxygen plasma, and contact immediately; (g) the fabricated microchannel.

Fig. 4-8 Investigated diffuser/nozzle structures: (a) $\alpha = 15^\circ$; (b) $\alpha = 30^\circ$; (c) $\alpha = 45^\circ$.

To investigate the effect of the device geometry on the droplet behavior, three different opening angles $\alpha=15^\circ$, 30° and 45° were used in this experiment. Fig. 4-8 depicts the detailed geometries of the different serial diffuser/nozzle microchannels used. For a fixed width of the diffuse/nozzle structures, channels with larger opening angle are shorter. The diffuser and nozzle flow direction are defined in Fig. 4-8.

4.2.2 Experimental Setup

In the experiment, de-ionized (DI) water and mineral oil work as the dispersed phase and the continuous phase, respectively. DI water was mixed with 0.01% w/w fluorescence dye (fluorescein disodium salt $C_{20}H_{10}Na_2O_5$, Acid Yellow 73 or C.I. 45350) to achieve better visualization with fluorescent microscopy. To reduce the interfacial tension between water and oil, 2% w/w surfactant span 80 was added to the oil. At a lower interfacial tension, it's easier to form small water droplets even at relatively low oil flow rates. The dynamic viscosity of mineral oil and the interfacial tension between these two immiscible liquids were measured at 25°C as 23.8 mPa s and 3.65 mN/m, respectively [14].

Fig. 4-9 (a) shows the schematic of the experimental setup. The working liquids were kept in 5 ml Hamilton glass syringes. Each syringe was driven by an individual syringe pump (KDS230, KD Scientific Inc, USA) to allow any flow rate ratio needed for the experiments. The pumps are connected in daisy-chain mode and can be controlled by a single serial port from the personal computer (PC). Hard Teflon tubes

connected the syringes with the inlets of the microfluidic chip. The inlets of the microfluidic chip were made of stainless steel needles. The needle has an inner diameter of 0.33 mm and an outer diameter 0.64 mm, which is press fit into the access hole previously punched into the PDMS substrate. The flow rate ratio was controlled by changing the volumetric flow rate of the pumps. The deformation of the droplet in the diffuser/nozzle microchannel was recorded with imaging software on the PC. As shown in Fig. 4-9(b), the test device was placed on the inverted microscope (Nikon EclipseTE 2000-S, Japan). Both bright-light and fluorescent modes were used to capture the image of the droplets. A sensitive CCD camera (HiSense MKII) attached to the microscope was used to capture the droplet image. In addition to the CCD camera, droplet images were also recorded using a commercial digital camcorder (DCR-DVD803E, Sony, Japan) attached to the eyepiece of the microscope.

The pressure ports on the chip were connected to a pressure sensor (HCX001, Sensortech, Germany) Fig. 4-9(a). The pressure sensor has a linear range from 0 to 1 bar corresponding to an output voltage ranging from 0.5 to 4.5 V. The sensor requires a calibrated supply voltage between 4.8 V and 15 V. A digital oscilloscope (TD220, Tektronix, USA) was used for collecting the electrical signal and converts it to a waveform, which was subsequently transferred to the PC via the serial interface. Fig. 4-1(b) already shows how a water droplet was formed at the T-junction. In the experiment, the large flow rate ratio of 30 was used to ensure that only one water droplet passing through the whole diffuser/nozzle microchannel at a given time, and the droplet size is on the order of the channel size. Since the flow rate ratio is fixed,

the droplet size is adjusted by the flow rates of both oil and water. A settling time of at least ten minutes was allocated for the flow to stabilize after changing the flow rates.

Fig. 4-9 Experimental setup (a) and the test device with 8 chips (b).

The pressure drop was measured in both diffuser and nozzle flow directions at different flow rates of oil. At a fixed flow rate ratio, the oil flow rate was varied from $30 \mu\text{l/h}$ to $300 \mu\text{l/h}$ corresponding to a water flow rate ranging from $1 \mu\text{l/h}$ to

10 $\mu\text{l/h}$. Hence, ten set of data were collected. The pressure data is considered as stable, if the standard deviation between the two sets of data averaged over an interval of 10 minutes is $< 1\%$. The pressure drop without droplets was also measured as a reference. Five data sets are considered. The flow rate between water and oil are 2 $\mu\text{l/h}$: 60 $\mu\text{l/h}$, 4 $\mu\text{l/h}$: 120 $\mu\text{l/h}$, and 6 $\mu\text{l/h}$: 180 $\mu\text{l/h}$, 8 $\mu\text{l/h}$: 240 $\mu\text{l/h}$, 10 $\mu\text{l/h}$: 300 $\mu\text{l/h}$.

To further investigate the effect of capillary number on the droplet behavior, the oil was mixed with surfactant Span 80 at different concentrations 0.1%(w/w), 0.5%(w/w), and 1.0%(w/w) to vary the interfacial tensions. The corresponding interfacial tensions are measured using a commercial tensiometer (FTA 200, First Ten Angstrom, USA) as 6.17 mN/m, 4.82 mN/m and 4.45 mN/m, respectively. The error of the measurement is less than 0.5 mN m^{-1} . The viscosities of the oil were measured with a Contraves Low Shear 40 rheometer at room temperature as 23.8 mPas and do not change significantly with the different surfactant concentrations. A high-speed camera (FASTCAM-SA1.1 675K, Photron) attached to an upright microscope (Olympus 171, Japan) captures the images of the droplet at a rate 500 fps and a size of 1024×1024 pixels. Unused pressure access ports were closed with solid needles (Fig. 4-10). For the majority of our experiments, the flow rates of oil and water were fixed at 300 $\mu\text{l/h}$ and 10 $\mu\text{l/h}$, respectively. The channels were flushed and cleaned carefully before a new set of liquid was used. The size and the velocity of the droplets were evaluated from the recorded images using a customized MATLAB program. The droplet diameter is obtained by assuming that the droplet has a discoidal shape.

Fig. 4-10 Experimental devices.

The channel dimension after bonding was measured with a confocal microscopy system [193]. The confocal microscopy is an optical imaging technique that can measure embedded micro-features bonded to a cover layer. It employs the laser fluorescent confocal microscopy and intensity differentiation algorithm to obtain the general profile of micro-features and further to ensure the device dimensions such as width, length, and depth with sub-micron measurement accuracy. To obtain the 3D profile of the microchannel device, DI water was mixed with fluorescence dye was filled into the channel by injecting the liquid from the inlet with a syringe. Then the sample was placed on a laser fluorescent confocal microscope (Zeiss LSM510) and scanned layer by layer. The cross section of the measured result is shown in Fig. 4-11. The cross section of the test device is trapezoid instead of rectangular considered in the numerical model.

Fig. 4-11 Cross sectional profile of the embedded channel.

4.3 Results and Discussions

4.3.1 Droplet Behavior Mediated by Geometry

The experimental droplets size show the same trend for all opening angles that the droplet size is smaller at a higher flow rate ratio (shown in Fig. 4-12). This behavior is consistent with the previous observation of droplet formation at a T-junction [12]. The larger shear rate caused by a higher flow rate makes the droplet breakup earlier leading to a smaller droplet. Fig. 4-13 depicts the droplet size as function of oil flow rate at the same flow rate ratio of 30. The higher the flow rate, the smaller is the droplet size.

 Fig. 4-12 Effect of flow rate ratio on droplet size ($q_c = 300 \mu\text{l/h}$).

 Fig. 4-13 Effect of oil flow rate on droplet size ($q_c : q_d = 30 : 1$).

Table 4-1 to Table 4-3 compare the droplet shapes obtained numerically and experimentally in structures with opening angles of $\alpha = 15^\circ$, 30° and 45° . Oil flow rates of $90 \mu\text{l/h}$, $180 \mu\text{l/h}$, and $270 \mu\text{l/h}$ were considered. The droplet shapes at the same position are compared. Because the different opening angle has the different length of the diffuser/nozzle structure, the droplet can only occupy one structure in Table 4-1, two structures in Table 4-2, and three structures in Table 4-3.

The agreement between simulation and measurement shows that the numerical method can describe well the deformation of a droplet in a diffuser/nozzle structure. From the experimental data, satellite droplets were formed at their tails. This breakup

was caused by the sudden pressure changes at the throats, as well as the lower interfacial tension. This phenomenon seems not to happen in the numerical case. The mesh size in space is not small enough to resolve the beads at the tails of droplets. The presence of underresolved regions lead to the mass loss phenomenon of level-set method [162]. Therefore, a lot of techniques were presented to improve mass conservation, as discussed in section 2.2.6. Here, it is the big deformed droplets mainly affect the characters of fluid flow. As a result, the present numerical results can be used to analyze hydrodynamics of droplets.

Table 4-1 Comparison of droplet shape between numerical data and experimental data for $\alpha = 15^\circ$.

	9 $\mu\text{l/h}$ water, 270 $\mu\text{l/h}$ oil		6 $\mu\text{l/h}$ water, 180 $\mu\text{l/h}$ oil		3 $\mu\text{l/h}$ water, 90 $\mu\text{l/h}$ oil	
	numerical data	experimental data	numerical data	experimental data	numerical data	experimental data
Nozzle
Diffuser

Table 4-2 Comparison of droplet shape between numerical data and experimental data for $\alpha = 30^\circ$.

	9 $\mu\text{l/h}$ water, 270 $\mu\text{l/h}$ oil		6 $\mu\text{l/h}$ water, 180 $\mu\text{l/h}$ oil		3 $\mu\text{l/h}$ water, 90 $\mu\text{l/h}$ oil	
	numerical data	experimental data	numerical data	experimental data	numerical data	experimental data
Nozzle
Diffuser

Table 4-3 Comparison of droplet shape between numerical data and experimental data for $\alpha = 45^\circ$.

	9 $\mu\text{l/h}$ water, 270 $\mu\text{l/h}$ oil		6 $\mu\text{l/h}$ water, 180 $\mu\text{l/h}$ oil		3 $\mu\text{l/h}$ water, 90 $\mu\text{l/h}$ oil	
	numerical data	experimental data	numerical data	experimental data	numerical data	experimental data
Nozzle
Diffuser

Fig. 4-14 shows the evolution of the velocity field as well as of the droplet motion in the case of $\alpha = 15^\circ$. The initial droplet diameter for this simulation is $d = 112 \mu\text{m}$. Fig. 4-14(a) shows an initially fully developed velocity field with the water droplet suspended in the microchannel which is filled with carrying fluid oil. Initially, two phases are at rest. The oil flow rate is $270 \mu\text{l/h}$, and the average velocity at the inlet is $\bar{u}_c = 0.01 \text{ m/s}$. A large deformation is expected as the droplet negotiates through the diffuser/nozzle structure.

The droplet shape mainly depends on the local channel structure. Fig. 4-14(b)-(c) describe its deformation process in time sequences. Along the center line of the channel, the carrying fluid flows faster than next to the wall. In one nozzle region, the velocity of main stream increases gradually in the flow direction. The narrowest section has the maximum velocity, while the larger end has a lower velocity. The droplet was squeezed as it went through the nozzle region due to the applied pressure. The inserts in Fig. 4-14(e) show that two vortexes appear in the two corners of the structure. For clarity, the velocity vector of the vortex was displayed with the same magnitude. The flow expands when the cross-sectional area increases suddenly at the tip of the nozzle region. The droplet has an obvious concave surface as shown in Fig. 4-14(d) because the velocity of the main fluid in the rear is much higher than that in the front part. At last, the interface becomes smoother as shown in Fig. 4-14(f) since the velocity does not change suddenly. The streamlines during the droplet evolution in the nozzle direction is depicted in Fig. 4-15.

Fig. 4-14 Droplet shape and velocity field with an opening angle of $\alpha = 15^\circ$ in the nozzle direction, and the flow rate ratio between water and oil is 9 : 270 (only few velocity vectors are shown for clarity).

Fig. 4-15 Streamlines with an opening angle of $\alpha = 15^\circ$ in the nozzle direction, and the flow rate ratio between water and oil is 9 : 270.

The velocity field in the diffuser direction is shown in Fig. 4-16. The droplet advancing tip is squeezed into a thinner finger [Fig. 4-16(c)], while it is spherical cap in nozzle flow direction [Fig. 4-14(e)]. From the maximum value at the narrowest portion shown in Fig. 4-16(f) and highlighted with the dashed circle, the magnitude decreases in the diffuser region gradually. There are still a concave in the rear as the droplet moves from one nozzle region to another. Fig. 4-16(g) shows the small vortex occurring at the corner as indicated by the insert with dotted line. Similar behaviors of the velocity field can be observed in structures with opening angles of $\alpha = 30^\circ$, 45° . The streamlines in the diffuser direction is depicted in Fig. 4-17.

Fig. 4-16 Droplet shape and velocity field with an opening angle of $\alpha = 15^\circ$ in the diffuser direction, and the flow rate ratio between water and oil is 9 : 270 (only few velocity vectors are shown for clarity).

Fig. 4-17 Streamlines with an opening angle of $\alpha = 15^\circ$ in the diffuser direction, and the flow rate ratio between water and oil is 9 : 270

Fig. 4-18 and Fig. 4-19 show the pressure distributions of the diffuser/nozzle portion along the centre line with different droplet location. They individually depict six pressure curves at different time instances corresponding to the time instances in Fig. 4-14 and Fig. 4-16, respectively. Due to the initial zero-flow condition, the initial pressure value is zero. The pressure shows a decreasing trend in flow direction for all cases.

Taking the results depicted in Fig. 4-18 as an example, the pressure jumps down at the narrowest section highlighted by the dashed circles in the graph. However, the pressure changes are slight at $t = 0.0448$ s and $t = 0.0656$ s located in the second and third throat, respectively. Fig. 4-14 shows that the droplet is going through the

narrowest throat at both time instances. In these situations, the droplet blocks the microchannel and is significantly deformed. Interfacial tension can keep the droplet from deforming. Furthermore, the viscosity of water is 0.9 mPa s, which is much less than the viscosity of the oil. So the velocity gradient inside the droplet is small. In another word, the droplet blocking the flow results in the stagnation that equalizes the pressure before and after the droplet. That is, the pressure difference of the whole channel will be decreased with the presence of the droplet. Thus, the droplet can significantly affect the pressure distribution along the array of the diffuser/nozzle structures. If the droplet is long enough to occupy several diffuser/nozzle structures at the same time, the pressure loss can be reduced even further.

The pressure distribution in the diffuser direction has similar characteristics (see Fig. 4-19). The only difference is that the pressure changes gradually in the nozzle region and then decreases steeply, while in the diffuser region the pressure jumps down first and then changes gradually. The pressure change is symmetric for the two opposite fluid flow directions.

Fig. 4-18 Pressure distribution along the centre line in the nozzle direction with $\alpha = 15^\circ$. The flow rate ratio between oil and water is 9 : 270. Six moments correspond to the time instances depicted in Fig. 4-14.

Fig. 4-19 Pressure distribution along the centre line in the diffuser direction with $\alpha = 15^\circ$. The flow rate ratio between oil and water is 9 : 270. Six time instances correspond to the time depicted in Fig. 4-16.

Fig. 4-20 shows the time history of the simulated pressure drop across the diffuser/nozzle structures. The fluctuation is caused by the change in pressure drop when the droplet passages the sudden expansion or sudden constriction. According to our design, the larger the opening angle the shorter is the diffuser/nozzle structure. Consequently, the pressure drop at a larger opening angle is lower. The lower pressure drop level shown in Fig. 4-20 corresponds to the situation when the droplet blocks the throat between two neighboring diffuser/nozzle structures. With an opening angle of $\alpha = 15^\circ$, the droplets is small compared to the length of the structure and the three low-pressure periods can be clearly observed, Fig. 4-20(a). In structures with larger opening angles of $\alpha = 30^\circ$ and $\alpha = 45^\circ$, the droplet is larger compared to the length of the structure and can occupy two throats at the same time. This situation leads to two low-pressure periods shown in Fig. 4-20(b) and Fig. 4-20(c).

Fig. 4-20 Pressure drop versus time of three diffuser/nozzle elements. The flow rates of oil and water are 270 $\mu\text{l/h}$ and 9 $\mu\text{l/h}$, respectively: (a) $\alpha = 15^\circ$; (b) $\alpha = 30^\circ$; (c) $\alpha = 45^\circ$.

In our experiments, the dynamics of the pressure measurement system can not follow the changes in pressure drop as shown in Fig. 4-20. Fig. 4-21 shows on set data of voltage as function of time. Fluctuation due to white noise can be observed. Thus, the recorded data is interpreted as the average pressure across the test section.

Fig. 4-21 Voltage waveform of the output signal.

The relationship between oil flow rate and the pressure drop across the diffuser/nozzle structures is shown in Fig. 4-22. The pressure drop is linearly proportional to the flow rate in both directions with the opening angles of $\alpha = 15^\circ$, 30° and 45° . Both fluids with droplet and without droplet are presented. Since the Reynolds number is much smaller than 1 in the experiment, the nonlinear inertial term in the Navier-Stokes equation is not significant. Thus, the measured results reflect this linear relationship. For the case without droplet, the magnitude of oil flow rate corresponds to the total flow rate in the case with droplet. The relative error increases with the increasing opening angles because of the shorter test structures and the corresponding lower pressure drop.

Fig. 4-22 Comparison of pressure drop between fluid flow with droplet and without droplet: (a) $\alpha = 15^\circ$; (b) $\alpha = 30^\circ$; (c) $\alpha = 45^\circ$.

The concept of diodicity was used to quantify the rectification effect of the tested structures. For the Stokes flow at a low Reynolds number, the pressure drop and the flow rate have the relationship:

$$\Delta P = Cq \quad (4.3)$$

where C is the proportional factor representing the pressure loss across the structure. Here, we define the ratio between factors of diffuser direction and nozzle direction as the diodicity

$$\zeta = C_{diffuser} / C_{nozzle} \quad (4.4)$$

The diodicity Eq. (4.4) is independent of the flow rate. Fig. 4-23 shows the diodicities for fluids with droplet and without droplet as a function of the opening angle α . The values are slightly bigger than 1 and the maximum at $\alpha = 15^\circ$ can be observed for both cases. Therefore, the rectification effect is not significant for our test structures. However, the existence of a droplet in the serial structures can slightly improve the rectification effect. If many droplets exist in the structure simultaneously, the diodicity can be improved further. If the structures are used in micropump, it indicates that the pumping direction is the nozzle direction.

The rectification effect in the Stokes regime at low Reynolds number observed may have different reasons. Compared to previous works on stand-alone diffuser/nozzle structures, the inlet and outlet conditions of the diffuser/nozzle structures connected in series are different. Furthermore, our designs have fixed widths at the two ends of the diffuser/nozzle structure. The structure length changes with the opening angles. The longer structure may allow flow separation even at very

low Reynolds number. A further possible reason for the rectification behavior at low Reynolds number is the interaction with the device material. The deformation of the microchannel due to the soft device material (PDMS) may also affect the pressure drop behavior.

Fig. 4-23 Diodicity as a function of the opening angle α .

To further investigate the diodicity of the structure, an opening contact angle of 45° was selected for the 3D simulation. In order to reduce the computation time, instead of ten structures only one diffuser/nozzle structure was solved (Fig. 4-24). Simulations were performed for flow rate ratios between water and oil of 6 : 180 $\mu\text{l/h}$, 8 : 240 $\mu\text{l/h}$, and 10 : 300 $\mu\text{l/h}$ for both flow directions with different droplet sizes. The droplet sizes were taken from the measurement in the experiments. Therefore, with a 3D model the flow behavior can be simulated and compared with the corresponding experimental results. In the case of a single-phase flow, the problem is steady state and only one phase of mineral oil needs to be considered.

Fig. 4-24 3D numerical model of diffuser/nozzle structure.

The pressure drops versus time of the 2D and 3D models are compared in Fig. 4-25. The flow rates of oil and water are 300 $\mu\text{l/h}$ and 10 $\mu\text{l/h}$, respectively. Fig. 4-25 shows that the droplet experiences a much longer passage time in the 2D simulation than in the 3D simulation. In the 2D simulation, the droplet thickness is assumed to be infinity. The volume of the droplet is larger than the actual amount. In the 3D simulation, the droplet thickness is confined by the microchannel height of 50 μm . Obviously the larger mass will take the longer time to pass through the model in the 2D case than the 3D case. Another reason for the discrepancy is the assumed droplet thickness. In the 3D simulation, the initial droplet thickness was assumed to be 30 μm which is less than the real value. The film between the droplet surface and the channel wall becomes thinner as the droplet moves through the model. The increase of droplet thickness decreases the passage time. Another issue shown in Fig. 4-25 is that the pressure drop of the 2D model is lower than that of 3D model. The reason for this discrepancy is that the friction effect at the bottom and top walls can not be considered in the 2D simulation. However, both simulation results show the same trend in pressure drop.

Fig. 4-25 Comparison of pressure drops from 2D and 3D simulations in nozzle and diffuser directions.

Fig. 4-26 Pressure drop versus oil flow rate of the two-phase flow (with droplet).

Fig. 4-27 Pressure drop versus oil flow rate without droplet.

The average pressure drop of simulation results are used to compare with the measured values. It is assumed that the measured pressure values are divided by ten to approximate the pressure drop of one structure. Fig. 4-26 and Fig. 4-27 show the pressure drop of one structure with droplet and without droplet, respectively. The flow rate between water and oil: 6 $\mu\text{l/h}$: 180 $\mu\text{l/h}$, 8 $\mu\text{l/h}$: 240 $\mu\text{l/h}$ and 10 $\mu\text{l/h}$: 300 $\mu\text{l/h}$, are simulated. The simulation results have the same order of magnitude as the measured results. A linear relationship was obtained from both simulation and measurement.

The diodicity values from simulation and experiments are shown in Fig. 4-28. The results indicate that the rectification effect can be observed even at low Re. The diodicity value with droplet is larger than that without droplet. Thus, we can conclude that the rectification effect is improved with droplet flowing in the microchannel.

Fig. 4-28 Diodicity comparison between the experimental, 2D numerical, 3D numerical results.

In Fig. 4-28, the values of simulation are higher than that from the experimental results. There are many reasons for this discrepancy. Firstly, we only take one structure for the simulation. The model is equivalent to ten droplets flowing in ten structures simultaneously. In the experiment, only one droplet exists in the entire test section at all instances. The sharp corners were used in the simulation model, while the corners of the actual device are rounded (see Fig. 4-8). The dimensions employed in the numerical models maybe different than the experiment device because of the fabrication error. Another possible reason is the deformability of the soft device material. And finally, the change in pressure drop involved in the experiments is relatively small and may not be captured precisely due to the accuracy limit of the sensor.

4.3.2 Droplet Behavior Mediated by Capillary Number

Beside the device geometry, the viscosity of continuous phase and interfacial tension are the key forces for droplet transporting in a diffuser/nozzle array. The capillary number Ca characterizes the ratio of viscous force to the interfacial tension,

$$Ca = \frac{\mu_c q_c}{W_c H_c \sigma} \quad (4.5)$$

Eq. (4.5) indicates that, at a fixed flow rate, the droplet velocity is dependent on the viscosity of the continuous phase, interfacial tension, and independent of the droplet size and its viscosity. For the convenience of generalization of the results, Ca will be used to represent their effect on droplet behaviors. In both simulation and experiment, the travelling distance of advancing front of droplet was measured. The velocity derived from this distance thereafter is referred to as the droplet velocity. As the numerical model does, the first two structures are also investigated in the experimental measurement.

The shapes of the deformed droplets from numerical and experimental results are compared in Table 4-4. The droplet surface is smooth, and no satellite droplets generated as listed in Table 4-1 to Table 4-3 because of the higher interfacial tension used in this experiment. The agreement shows that the numerical method can describe well the droplets deformation in a diffuser/nozzle array.

Table 4-4 Experimental and numerical deformation of droplets at different surfactant concentration.

The droplet diameter D^* is normalized by the channel width W_C . The capillary number was varied by adjusting the interfacial tensions with different surfactant concentrations. The higher capillary number leads to a smaller droplet formed at the T-junction of the test devices, Fig. 4-29. The droplet size in diffuser direction is slightly larger than that in nozzle direction. This phenomenon attributes to the rectification effect discussed in the previous section.

Fig. 4-29 The influence of capillary number on the droplet size ($q_c = 300 \mu\text{l/h}$, $q_d = 10 \mu\text{l/h}$).

The mechanism of droplet breakup in a T-junction was investigated in details by Garstecki et al. [12]. The forces involved in breakup are interfacial tension force F_σ , viscous force F_μ , the pressure difference F_P . Their values are estimated as

$$F_\sigma \approx -\sigma H_C \quad (4.6)$$

$$F_\mu \approx \frac{\mu q_c W_C}{\eta^2} \quad (4.7)$$

$$F_P \approx \frac{\mu q_c W_C^2}{\eta^3} \quad (4.8)$$

where η is the gap between droplet interface and the solid wall. F_σ orients upstream,

and keeps the water tip to move forward. F_μ and F_P orient downstream, and increase as η decreases. The higher interfacial tension can yield smaller gap ($\eta_3 > \eta_2 > \eta_1$), Fig. 4-30. Consequently, F_μ and F_P sharply increase to overcome the higher F_σ until breakup happens.

Fig. 4-30 Evolution of droplet generation in the forward direction: (a) $\sigma = 6.17$ mN/m; (b) $\sigma = 4.82$ mN/m; (c) $\sigma = 4.45$ mN/m.

4.3.2.1 Effect of Viscosity on Droplet Velocity

To investigate the effect of viscosity on the droplet motion, we used a numerical model with fixed flow rates ($q_c = 300 \mu\text{l/h}$, $q_d = 10 \mu\text{l/h}$) and fixed interfacial tension ($\sigma = 4.82 \text{ mNm}^{-1}$). At the same flow rates and the same interfacial tension, the viscosity of the oil affects the droplet velocity significantly (Fig. 4-31). The results also show that the acceleration/deceleration pattern depends on the flow direction. The acceleration and deceleration are approximately symmetric about the maximum velocity in the nozzle direction, but asymmetric in the diffuser direction. A droplet of

the same size, the same interfacial tension and the same viscosity moves faster in a more viscous continuous phase. The higher viscosity of the continuous phase squeezes and elongates the droplet leading to a large difference in radius of curvature between the advancing and receding surfaces, Fig. 4-32(a) (c). Thus, the capillary force acting on the droplet is higher with a higher viscosity of the continuous phase. The different droplet lengths result in the different sizes of the cross section at the narrowest throat position, Fig. 4-32(b) (d).

Fig. 4-31 Simulated droplet velocities at different viscosities of the continuous phase in (a) nozzle and (b) diffuser directions ($q_c = 300 \mu\text{l/h}$, $q_d = 10 \mu\text{l/h}$, $\sigma = 4.82 \text{ mN/m}$).

Fig. 4-32 The higher viscosity of the continuous phase squeezes and elongates the droplet leading to a large difference in radius of curvature between the advancing receding surfaces in both (a, b) the nozzle direction and (c, d) the diffuser direction. The droplets also have different cross section at the narrowest throat in (b) the nozzle direction and (d) the diffuser direction.

Fig. 4-33 The influence of droplet viscosities on the droplet velocities: (a) nozzle direction; (b) diffuser direction ($q_c = 300 \text{ } \mu\text{l/h}$, $q_d = 10 \text{ } \mu\text{l/h}$, and $\sigma = 4.82 \text{ mN/m}$).

Fig. 4-34 The viscosities of the disperse phase have negligible effect on the droplet shape in both (a) the nozzle direction and (c) the diffuser direction.

At the same physical conditions ($q_c = 300 \text{ }\mu\text{l/h}$, $q_d = 10 \text{ }\mu\text{l/h}$, $\sigma = 4.82 \text{ mN/m}$), the viscosities effect of dispersed phase on the droplet velocities is investigated numerically at fixed droplet size. The viscosities 0.5 mPa s , 0.9 mPa s , and 9 mPa s are studied. From Fig. 4-33, we can conclude that the droplet viscosities have negligible effect on droplet velocities. The droplet shape is quite similar as shown in Fig. 4-34.

4.3.3.2 Droplet Size Effect on Droplet Velocity

We tested the effect of droplet size on the droplet velocity both numerically and experimentally. As the droplet size is affected by the interfacial tension, we design a control experiment to discriminate the effect of droplet size on the motion. The experiment only considers oil with $0.5 \text{ \% (w/w)}$ surfactant. The total flow rate was kept at $310 \text{ }\mu\text{l/h}$ so that the average mean velocity remains constant. The droplet size was changed by varying the flow rate ratio between oil and water. Both numerical and

experimental results indicate that the droplet velocity is not significantly affected by the droplet size, Fig. 4-35(a-d). The tested range of droplet size is approximately the range caused by the different interfacial tensions in our experiment.

Fig. 4-35 The effect of droplet size on the droplet velocity: (a) experimental data for nozzle direction; (b) numerical data for nozzle direction; (c) experimental data for diffuser direction; (d) numerical data for diffuser direction; (e) formed droplets at different flow rate ratios in both directions.

4.3.2.3 Interfacial Tension Effect on Droplet Velocity

In this part, we varied the interfacial tension with different surfactant concentrations.

As measured before, the viscosity of oil remains unchanged. The key observation in this experiment is that droplets with higher surfactant concentration or lower

interfacial tension always move faster in both flow directions, Fig. 4-36 (a) (c). Although the experimental results and numerical results show the same trends, their magnitudes are slightly different, Fig. 4-36(b), (d). The discrepancy of about 20% could be caused by the trapezoid cross section of the fabricated microchannel. The numerical model only considers a rectangular cross section and the first two in the array of the ten ratchets. Fig. 4-37 describes travelling distance of the droplet versus the time of both numerical and experimental data. The droplet moves faster in the diffuser direction than in the nozzle direction.

Fig. 4-36 The effect of different surfactant concentrations on the droplet velocities: (a) experimental data for nozzle direction; (b) numerical data for nozzle direction; (c) experimental data for diffuser direction; (d) numerical data for diffuser direction.

Fig. 4-37 Comparison of droplet travelling distance between nozzle direction and diffuser direction: experimental data (a), (c), and (e); numerical data (b), (d), and (f).

Fig. 4-38 shows the simulated droplet velocity $U_{\max}^* = U_{\max} / \bar{u}_c$ as a function of the capillary number Ca . Although the results were obtained by varying the dynamic viscosity of the continuous phase or by varying the interfacial tension, the same trend of the maximum droplet velocity is observed. At a given mean velocity (fixed flow rates), the maximum droplet velocity increases with increasing capillary number. The

velocity in diffuser direction is higher than that in nozzle direction.

Fig. 4-38 Dimensionless maximum velocity of the droplet as a function of capillary number ($q_c = 300 \mu\text{l/h}$, $q_d = 10 \mu\text{l/h}$).

Fig. 4-39 shows the experimental results of the maximum droplet velocities function of capillary number. The results indicate that a lower interfacial tension or a higher capillary number makes the droplet move faster in both directions of the microchannel. This trend agrees well with the results predicted by numerical simulation, Fig. 4-38. However, for the same range of capillary number the measured velocity is slightly lower than the simulated one. The discrepancy could be explained by the simplified numerical model. For the range of droplet size considered in the experiment, the velocity is independent of the droplet size (Fig. 4-35). Thus the magnitude of interfacial tension is the single parameter that determines the droplet velocity. Thus, measuring the droplet velocity can determine the interfacial tension. The difference in magnitudes of droplet velocity in both direction indicate that the channel configuration exhibit rectification characteristics which can be used for

designing micropumps for delivering multi-phase systems such as a droplet train.

Fig. 4-39 Dimensionless maximum velocity of the droplet as a function of capillary number ($q_c = 300 \mu\text{l/h}$, $q_d = 10 \mu\text{l/h}$).

4.4 Conclusions

In conclusion, we have presented the numerical and experimental results of the behavior of microdroplets in an array of diffuser/nozzle structures. The fabrication of the test devices used soft lithography techniques in PDMS. The deformation of a water droplet in diffuser/nozzle structures with an opening angles ranging from 15° to 45° were investigated. At a constant flow rate ratio between oil and water, both experimental and numerical results show that the pressure drop is linear proportional to the flow rates. A rectification effect was observed in all tested devices. Despite the low Reynolds number of less than 0.032, the pressure drop in diffuser direction is higher than in the nozzle direction. This effect is consistent with the behavior of a

continuous flow of a Newtonian fluid through a diffuser/nozzle structure and can be used for designing micropumps for two-phase flow.

In addition, the relationship between the droplet velocity and the capillary number was investigated. Mineral oils with different concentrations of surfactant Span 80 was tested at constant flow rates of oil and water. Densities of the liquid are insignificant because gravitational forces are negligible in microscale. The droplet velocity in an array of diffuser/nozzle structures is a function of interfacial tension, independent of the droplets size and the viscosity of the droplet liquid. A lower interfacial tension results in a smaller droplet size and an increase in the droplet velocity. The same phenomena were observed with a three-dimensional numerical model. Although only two structures are modeled in the simulation, numerical results agree relatively well with experimental data. The results presented here show new possibilities of measuring interfacial tension for multiphase systems.

CHAPTER 5

FORMATION OF FERROFLUID DROPLETS

In this chapter, the influence of a magnetic field on the droplet formation process in a flow focusing microchannel is studied. A linear magnetic fluid was assumed, and the magnetic field is uniform. The effects of magnetic Bond number and the susceptibility on the velocity field and the droplet size are analyzed. The droplet is bigger as the increase of magnetic strength and the susceptibility. The numerical results are compared with the experimental data at last.

5.1 Problem Formulation and Numerical Model

The flow focusing configuration is employed to form the uniform droplets. The test device has a size of $10\text{ mm} \times 10\text{ mm}$. Fig. 5-1 (a) shows the investigated layout of the device. Port one has two equal branches to guide the continuous flow with the same flow rate. Two branches acts as two lateral channels of the flow focusing structure. Port two is used to introduce dispersed fluid to flow in the main channel. The left port is an outlet.

Fig. 5-1(b) shows the geometry of a flow focusing configuration in 3D. Two lateral inlets and a single main inlet have a square cross section of $L \times L$. The characteristic length is L ($= 100\text{ }\mu\text{m}$). The droplets of dispersed flow were formed because of the squeezing of continuous flow. Inlet velocities were assumed to be fully developed with u_c and v_d . To save computational time, only one quarter of the

domain was calculated due to the symmetry of the channel geometry and the flow field.

Fig. 5-1 Device schematic for microdroplet formation: (a) test device layout; (b) geometry of the flow-focusing.

Before investigating the real case of this flow focusing structure, the validation and grid refinement were carried out. As a case study, the mean velocities of the main channel and the lateral channel are $u_c = 0.0025 \text{ m/s}$ and $v_d = 0.01 \text{ m/s}$. Both fluids

have the same densities and viscosities of $\rho_c = \rho_d = 1000 \text{ kg m}^{-3}$ and $\mu_c = \mu_d = 1 \text{ mPa s}$. The interfacial tension is set as $\sigma = 1 \text{ mN/m}$. The present results are compared with VOF method of the commercial code Fluent. Good agreement was achieved. The normalized dimensionless number of droplet volumes $V^* = V_d / L^3$ are shown in Table 5-1. Our numerical method uses the stagger grid to approach the flow focusing channel. The channel dimension will slightly change before attach to the exact position with grid refinement. The VOF method is utilized on the real channel structure shown in Fig. 5-1. The same grid size of $184 \times 62 \times 26$ will be used in the following simulation.

Table 5-1 Drop volume comparison between VOF results and LSM results.

VOF			LSM			
Cell number	Δt	V^*	Cell number	Δt	V^*	Difference
14280	2E-5	0.3127	93×32×14	2E-5	0.28038	10.3%
68663	1E-5	0.29986	116×40×17	2E-5	0.28656	4.4%
117560	5E-6	0.29633	184×62×26	1E-5	0.28828	2.7%

5.2 Materials and Experimental Setup

The test device was fabricated in PDMS, and was bonded to a spin coated PDMS membrane of a thickness of $200 \text{ }\mu\text{m}$ after oxygen plasma treating to the surface (790 Series, Plasma-Therm, Inc., FL, USA). The device was aligned on a glass slide to guarantee that the magnetic field is parallel to the main channel [Fig. 5-1(b)]. To generate a uniform magnetic field, a magnetic coil with 350 turns was spiraled round a “C” shape steel core with a small separation gap of 26 mm [194]. A commercial gaussmeter (Hirst, GM05, UK) was used to measure the magnetic flux density at

different current values. Fig. 5-2 shows the experiment setup. A DC power source (Instek, GPS-3030DD) was used to vary the magnitude of the magnetic field strength. Both continuous phase and dispersed phase were delivered to the device inlets by two separate precision syringe pumps. In the experiment, water-based ferrofluid (Ferrotech, EMG 807) works as the dispersed phase. The spherically shaped nanoparticles (Fe_3O_4) have a mean diameter of 10 nm. The particles volume concentration is 1.8% and the beginning susceptibility is $\chi = 0.39$. The magnetization of this ferrofluid has been described by Tan et al. [103]. The density and viscosity of ferrofluid are $\rho_c = 1100 \text{ kg/m}^3$, $\mu_c = 2 \times 10^{-3} \text{ mPas}$. Silicone oil (Sigma-Aldrich, 378364) works as the continuous phase. The properties are $\rho_d = 960 \text{ kg/m}^3$, $\mu_d = 96 \times 10^{-3} \text{ mPas}$.

Fig. 5-2 Experiment setup.

In the experiment, the flow rate of silicone oil varied from $Q_c = 20 \mu\text{l/h}$ to $24 \mu\text{l/h}$. The flow rates ratio of the continuous and dispersed phases were fixed at two. The current of the electromagnet was then varied at intervals of 0.5 A ranging from 0 to 3 A. The corresponding magnetic flux density varies from 0 to 40 mT (Fig. 5-3). The

ferrofluid behaves as a non-linear magnetizable material because of the stronger magnetic field (Fig. 5-4). The formed ferrofluid droplets were imaged with a high speed camera (Photron, APX RaS) using a 10X objective lens on a Nikon (TE2000) inverted fluorescence microscope. Images were acquired at a rate of 1000 frames per second. The droplet diameters were measured with a customized MATLAB program, and the droplets were assumed to be a discoid shape.

Fig. 5-3 The measured magnetic flux density in the air versus the different current.

Fig. 5-4 The relationship between the magnetization of the ferrofluid and magnetic field strength.

5.3 Results and Discussions

5.3.1 Droplet Formation with Flow-Focusing Configuration

The dynamics of droplet formation was first investigated without the magnetic field. To get the same droplet size with the experimental results, the interfacial tension is a varying input parameter and is set as $\sigma = 12$ mN/m. The flow rate of each oil inlet is $q_c = 10$ $\mu\text{l/h}$. The water flow rate is $q_d = 5$ $\mu\text{l/h}$. Thus, the mean velocities are $u_c = 2.78 \times 10^{-4}$ m/s and $v_d = u_c / 2$. The droplet formation process in the flow focusing configuration was calculated with the following dimensionless numbers,

$$Ca = \frac{\mu_c u_c}{\sigma}, \text{Re} = \frac{\rho_c u_c L}{\mu_2}, \mu^* = \frac{\mu_c}{\mu_d}, \rho^* = \frac{\rho_c}{\rho_d}, u^* = \frac{u_c}{u_d}, t^* = \frac{tu_c}{L}, \quad (5.1)$$

The values of the capillary number and the Reynolds number are $Ca = 2.22 \times 10^{-3}$, $\text{Re} = 2.78 \times 10^{-4}$.

The simulated evolution of the formed droplet is shown in Fig. 5-5. The numerical results are compared with the experimental results. The discrepancy occurs after the droplet breakup at $t = 187$ ms marked with a circle (Fig. 5-5(a), $t = 187$ ms, 207 ms). The tip of ferrofluid is sharper in the experimental results because of the rough surface of the channel. It caused from the swelling of the device material (PDMS) when exposed to silicone oil [11, 195]. The rough channel wall may result in stronger friction. The 3D numerical results with one planar slice located at the narrowest throat of the junction are depicted in

. The throat is blocked at $t = 47$ ms and then decreases gradually.

Fig. 5-5 Sequence of droplet formation without magnetic field: (a) experimental results, and (b) numerical results on x-y plane ($Ca = 2.22 \times 10^{-3}$, $Re = 2.78 \times 10^{-4}$, $q_c = 10 \mu\text{l/h}$, $q_d = 5 \mu\text{l/h}$).

Fig. 5-6 The behavior of ferrofluid flow without the magnetic field with section for the plane $y = 2.075 L$ ($Ca = 2.22 \times 10^{-3}$, $Re = 2.78 \times 10^{-4}$, $q_c = 10 \mu\text{l/h}$, $q_d = 5 \mu\text{l/h}$).

Fig. 5-7 The various forces acting on the ferrofluid tip during the droplet formation process in the flow focusing channel.

The forces acting on the ferrofluid tip are the viscous force F_μ , the pressure difference F_P , and the interfacial tension force F_σ as given in Fig. 5-7. F_μ is caused by the viscous stress acting on the interface, and is proportional to the tip area and the velocity gradient. Together with F_P , the viscous force F_μ provides a squeezing action on the tip to move it downstream. On the contrary, the interfacial tension force F_σ keeps the ferrofluid tip from moving forward. As shown in

, at the beginning of the formation process ($t = 0, 47$ ms), the curvature of the tip is bigger and results in a larger capillary force. The capillary force is proportional to the curvature and the interfacial tension. A higher pressure is needed to move the tip through the throat. As a result, the throat is blocked as the dispersed phase flows through. The velocities inside the throat orientate upstream instead of downstream (see Fig. 5-9 $t = 47$ ms). Because the ferrofluid blocks almost the entire throat, F_P increases dramatically at the region outside ferrofluid thread. The tip is pushed to move further forward. The curvature of the tip becomes smaller as the tip grows and weakens the capillary force. Thus, the direction of the fluid velocity located inside the rectangular window at $t = 116$ ms reverses to the opposite direction resulting in competition between F_P , F_μ and F_σ inside the ferrofluid thread (Fig. 5-9). This process happens instantaneous accompanying a couple of opposite velocities increase dramatically from $t = 166$ ms to $t = 173$ ms as marked with the rectangular window in Fig. 5-9. The changing thinner thread can weaken the interfacial tension effect, and increase its corresponding curvature. At this moment, the pressure difference is very high inside the thread. Under the stretching action, the ferrofluid inside the thread is

pushed to separate in both opposite directions until the weaker F_σ is no longer strong enough to hold the tip. The thread becomes thinner and breakups to form a droplet at $t = 187$ ms. Since droplet formation is a process of F_p and F_μ overcoming F_σ , the high interfacial tension can result into the larger droplet size Fig. 5-8 . This numerical result is consistent with our previous experimental results in a T-junction as shown in Fig. 4-29.

Fig. 5-8 Drop formation with different interfacial tension.

Fig. 5-9 Velocity field during droplet formation process without magnetic field effect ($Ca = 2.22 \times 10^{-3}$, $Re = 2.78 \times 10^{-4}$, $q_c = 10 \mu\text{l/h}$, $q_d = 5 \mu\text{l/h}$, x - y plane).

Fig. 5-10 shows the simulated instantaneous streamlines during the droplet formation process from the early stage to the breakup without the magnetic effect. The streamlines describe the direction of fluid flow over time. As shown in Fig. 5-10, from $t = 47 \text{ ms}$ to $t = 116 \text{ ms}$, the flow directions are changed and two counter rotating vortices near the wall appear inside the throat. Several pairs of vortices distribute along the interface of the thread at $t = 166 \text{ ms}$. It is interesting to see that two pairs of the vortices grow bigger when the thread is ready to breakup at $t = 173 \text{ ms}$.

Fig. 5-10 Instantaneous streamlines during droplet formation process without magnetic field effect ($Ca = 2.22 \times 10^{-3}$, $Re = 2.78 \times 10^{-4}$, $q_c = 10 \mu\text{l/h}$, $q_d = 5 \mu\text{l/h}$, x-y plane).

5.3.2 The Influence of Magnetic Field on the Formation Process

The magnetic force has a significant effect on the droplet formation process. When the microfluidic device is exposed to a uniform magnetic field and the direction is parallel to the ferrofluid flow direction, the ferrofluid is exposed to the combined action of interfacial tension, pressure difference, viscous force, and magnetic stretching force.

The values of dimensionless numbers in Eq. (5.1) are used here. A magnetic Bond

number B_m and the susceptibility χ_{m+} are used to represent the effect of magnetic field on droplet formation process. The susceptibility χ_{m+} represents the respond of ferrofluid to an applied magnetic field. The different χ_{m+} values denote the different ferrofluid types. The magnetic Bond number B_m ($=\mu_0 L H_0^2 / \sigma$) describes the ratio between the magnetic force and the interfacial tension force. The magnetic bond number increases by increasing the field strength H .

Fig. 5-11 shows the simulated velocity field during the droplet formation process under a magnetic field with $B_m = 0.1$, $\chi_{m+} = 8$. It takes much longer time for the ferrofluid tip to complete the formation process as compared to the process depicted in Fig. 5-6. The magnetic force F_m acts as a drag force on the tip. It enforces on the entire interface where the susceptibility changes and is zero inside each fluid. This force is related to the magnetic field strength and the magnetic susceptibility. The direction change of flow field inside the throat happens between $t = 43$ ms and $t = 108$ ms. As the channel throat is blocked as show in Fig. 5-11 ($t = 108$ ms), the pressure drop F_P increases near the region upstream of the throat. However, because of the forward drag force F_m acting on the tip along downstream direction, the whole tip is elongated. The elongation is more serious at increasing χ_{m+} , Fig. 5-14. As a result, the tip does not exhibit the spherical shape as in the case without the magnetic field in Fig. 5-6. The thread is relatively longer too (Fig. 5-11, $t = 223$ ms). Thus, from $t = 108$ ms to 288 ms in Fig. 5-11, the velocity direction inside the thread keeps orientating downstream. The magnitude of the velocities increases gradually until the breakup moment. These phenomena are different from the case without the magnetic

field. The pressure outside the thread increases slowly. The curvature of the thread is small and changes slowly because of the high pressure inside the thread. These behaviors increase the forming time and enlarge the ferrofluid tip. Finally, a bigger droplet is formed when the interfacial tension force can not withhold the ferrofluid. The formed droplet has a shape of an ellipsoid due to the stretching effect of the magnetic force ($t = 295 \text{ ms}$ to 309.4 ms in Fig. 5-13). The streamlines of droplet forming under a magnetic field is described in Fig. 5-12.

Fig. 5-11 Velocity field during droplet formation process with magnetic field ($B_m = 0.1$, $\chi_{m+} = 8$, $q_c = 10 \mu\text{l/h}$, $q_d = 5 \mu\text{l/h}$, x - y plane).

Fig. 5-12 Instantaneous streamlines during droplet formation process with magnetic field ($B_m = 0.1$, $\chi_{m+} = 8$, $q_c = 10 \text{ } \mu\text{l/h}$, $q_d = 5 \text{ } \mu\text{l/h}$, x-y plane).

Fig. 5-13 The behavior of ferrofluid flow in the magnetic field with section for the plane $y = 2.075 L$ ($B_m = 0.1$, $\chi_{m+} = 8$, $q_c = 10 \mu\text{l/h}$, $q_d = 5 \mu\text{l/h}$).

The droplet size depends on both the magnetic field strength and the magnetic susceptibility. Fig. 5-14 shows the relationship between the formed droplet volume V^* and the susceptibility. The nonlinear relationship shows a higher sensitivity at the higher susceptibility.

Fig. 5-14 Non-dimensional volume of droplet against susceptibility χ_{m+} on the condition of $B_m = 0.1$ ($q_c = 10 \mu\text{l/h}$, $q_d = 5 \mu\text{l/h}$).

Fig. 5-15 shows the evolution of the droplet formation process both experimentally and numerically. The ferrofluid tips were elongated, and the formed droplet size is big comparing with the case without the magnetic field as given in Fig. 5-5. In the experiment, the time to form a droplet is about 1.016 s when the magnetic flux density is $B = 42.3 \text{ mT}$. This time is almost doubles the time of the case without the magnetic field of 0.592 s.

Fig. 5-15 The evolution of droplet formation. Delay time of each frame is 58 ms ($q_c = 10 \mu\text{l/h}$, $q_d = 5 \mu\text{l/h}$): (a) experimental results of nonlinear magnetizable fluid at magnetic flux density of 42.3 mT, and current is 3A; (b) numerical results of linear magnetizable fluid at $\chi_{m+} = 5$, and $B_m = 0.3$.

The formed droplet becomes much bigger as the magnetic field strength increases. The relationship between the normalized diameter and the magnetic Bond number is shown in Fig. 5-16. The different trends between the numerical and the experimental results are caused from the different responses of the magnetization to the applied magnetic field strength.

The ferrofluid is assumed to be a linear magnetic material in the numerical models while it behaves as a non-linear magnetic material in the experiment. A non-linear model would be more accurate to obtain a quantitative comparison with the experiment results. However, the aim of the present numerical study is the qualitative understanding of the behavior of the ferrofluid under an applied magnetic field and of the magneto-hydrodynamic interactions. The present results provide the theoretical basis for further development of more comprehensive and accurate models describing the actual physics of the problem. Nevertheless, both results show the similar trends, and they obviously indicate the two regions affected by the flow rates.

There are the same two regions because of the influence of flow rate. The higher flow rate can form relatively bigger droplet in region I of Fig. 5-16, and smaller droplet in region II of Fig. 5-16. The experimental data shows the states where the ferrofluid tips are ready to breakup as depicted in Fig. 5-17. In region I, the tip diameter is smaller than the channel width. In region II, the blockage happens in case of $q_c = 10 \mu\text{l/h}$, $q_d = 5 \mu\text{l/h}$, and $\eta \ll W_C$. The pressure difference and the shear stress can increase dramatically. It reflects that lower flow rate can form bigger droplet in the stronger magnetic force. The same phenomenon happened at a T-junction [103].

Fig. 5-16 The magnetic effect on the droplet size.

Fig. 5-17 The shapes of the ferrofluid tips before breakup at the different magnetic Bond number B_m in the experiment.

5.4 Conclusions

This chapter focuses on the formation process of a ferrofluid droplet in a flow-focusing configuration with and without the magnetic effect. The formation process was investigated in details. In the absence of the magnetic field, a couple of opposite flow velocity appears in the flow focusing channel as results of the interaction between the pressure drop, viscous drag force and interfacial tension. The pressure drop and the viscous drag force push the ferrofluid forward while the interfacial tension keeps the tip from moving forward. In the presence of a magnetic field, the ferrofluid tip was pulled forward due the additional magnetic force. The thread and the tip become longer resulting in a longer formation time. The flow velocity inside the thread also increases. With a magnetic field, the forming droplet behaves in a different way as compared to the case without the magnetic field. Further simulations were carried out to investigate the influence of the magnetic Bond number and the susceptibility on the droplet size. The higher the magnetic Bond number and susceptibility are, the larger is the volume of the formed droplet. In addition, the flow rate effect on the droplet diameter was investigated using both experimental and numerical methods.

CHAPTER 6

CONCLUSIONS AND FUTURE WORKS

6.1 Summary and Conclusions

Investigation of the multiphase flow in a microchannel attracts much attention because of its wide application in microfluidics. Droplet-based microfluidics is an emerging research field. In this thesis, droplet behavior mediated by geometry, capillary number and an external magnetic field was investigated. The behavior of microdroplets in an array of diffuser/nozzle structures was investigated experimentally and numerically. In addition, the dynamics of droplet formation in a flow focusing channel was discussed.

To understand the complex multiphysics phenomena and the dynamics of a multiphase system in microchannels, a numerical scheme was first developed and employed. The governing equations of the physical fields were solved with finite volume method using a staggered grid. The interface between two immiscible fluids was tracked with the particle level-set method. The problem involving triple contact line was also solved successfully.

As the first case study, droplet behavior in a diffuser/nozzle array was studied. The deformation of a water droplet in a diffuser/nozzle array with opening angles ranging from 15° to 45° were investigated. A 2D numerical model was formulated and solved to track the deformation of a microdroplet during the passage through the

diffuser/nozzle array. In the experiment, the soft lithography techniques were used to fabricate the test chip in PDMS. The pressure drop of the test section of the channel was measured with a pressure sensor at the same flow rate ratio. The numerical and experimental results show that the numerical model can capture well the deformation of the droplet inside the diffuser/nozzle structure. Since the deformation is determined by the two key factors, the interfacial tension and the viscosity of the carrier fluid, the device and the experiment can be potentially used for rheometry. At a constant flow rate ratio between oil and water, the experimental and numerical results show that the pressure drop is linear proportional to the flow rates. A rectification effect was observed in all tested devices. Despite the low Reynolds number of less than 0.032, the pressure drop in diffuser direction is higher than in the nozzle direction. This effect is consistent with the behavior of a continuous flow of a Newtonian fluid through a diffuser/nozzle structure and can be used for designing micropumps for multiphase systems.

A 3D simulation was carried out to further investigate the rectification effect mediated by the channel geometry. The numerical results showed the same trend with the experimental results. The relationship between the pressure drop and the flow rate was given.

In a microfluidic system, the surface-based interfacial tension and the viscous shear dominate the behavior of the multiphase flow. Diffuse/nozzle structures with an opening angle of 35° , the droplet velocity was observed as a function of interfacial tension, and independent of the droplet size and the viscosity of the droplet liquid.

Mineral oils with different concentrations of surfactant Span 80 was tested at constant flow rates of oil and water. A lower interfacial tension results in a smaller droplet size and an increase in the droplet velocity. The same phenomena were observed with a three-dimensional numerical model. Although only two structures are modeled in the simulation, numerical results agree relatively well with experimental data. The results presented here show new possibilities of measuring interfacial tension.

As the second case study, the effect of the magnetic field on the formation of ferrofluid droplets in a flow-focusing configuration was investigated. In the absence of the magnetic field, a flow field direction upstream forms in the droplet as a result of interaction between the pressure drop, viscous drag force and interfacial tension. In the presence of a magnetic field, the ferrofluid tip was pulled forward under the additional magnetic force. The thread and the tip become longer resulting in a longer formation time. Further simulations were carried out to investigate the influence of the magnetic Bond number and the susceptibility on the droplet size. Furthermore, the experiment was carried out. The larger droplet formed in the stronger magnetic field strength. Both numerical and experimental data show that the flow rates of the two immiscible fluids also affect the droplet size in the magnetic field.

6.2 Suggestions for Future Works

A pump with an oscillating pressure port between two diffuser/nozzle arrays will be able to deliver a droplet train. In many applications such as protein crystallization in

microdroplets, an existing droplet train, immobile for the period of crystallization, will need a pump for on-chip transport and further processing.

Given the relationship between interfacial tension and the droplet velocity, the diffuser/nozzle array can be used to measure interfacial tension. If the concentration of surfactant is lower than the CMC (critical micelle concentration), the values of interfacial tension is a function of time [196]. The small time scale involved in the microfluidic device would allow the measurement of dynamic surface tension with high accuracy. Thus, a new measurement technique is needed with a microscale handling.

The present work established a method to calculate the magnetic force on the conditions of the linear magnetic material and the ferrofluid behaving as a uniform liquid. Although, the present numerical method gives a consistent trend with the experimental results, the exact quantitative comparison has not been carried out. The non-linear magnetizable fluids can be considered in the future [197]. Moreover, the present numerical method can be employed to study the droplet formation process in other channel geometries.

LIST OF PUBLICATION

Journal Papers:

[1] Liu, J.; Tan, S.H.; Yap, Y.F.; Ng, M.Y.; Nguyen, N.T. Numerical and experimental investigation of the formation process of ferrofluid droplets. Submitted to *Microfluidics and Nanofluidics*.

[2] Liu, J.; Nguyen, N.T.; Yap, Y.F. Numerical studies of sessile droplets shape with moving contact lines. Submitted to *Micro and Nanosystems*.

[3] Liu, J.; Yap, Y.F.; Nguyen, N.T. Numerical study of ferrofluid droplets formation. Submitted to *Physics of Fluids*.

[4] Liu, J.; Yap, Y.F.; Nguyen, N.T. Behavior of microdroplets in diffuser/nozzle structures. *Microfluidics and Nanofluidics*, 2009, 6(6), 835-846.

[5] Liu, J.; Yap, Y.F.; Nguyen, N.T. Motion of a droplet through microfluidic ratchets. *Physical Review E*, 2009, 80(4), 046319.

[6] Liu, J.; Nguyen, N.T. Numerical Simulation of Droplet-Based Microfluidics - a Review, *Micro and Nanosystems*, 2010, 2 (3), 193-201.

[7] Li, S.; Liu, J.; Nguyen, N.T.; Fang, Z.P.; Yoon, S.F. Measurement of buried undercut structures in microfluidic devices by laser fluorescent confocal microscopy. *Applied Optics*, 2009, 48(33), 6432-6441.

Conference Papers:

[1] Liu, J.; Nguyen, N.T.; Yap, Y.F. Experimental and numerical investigation of droplet transport in a diffuser/nozzle structure. In: *Proceedings of the 7th International Conference on Nanochannels, Microchannels, and Minichannels* South Korea, 2009.

[2] Liu, J.; Nguyen, N.T.; Yap, Y.F. Numerical study of ferrofluid droplets formation in the magnetic field. In: *The Second Conference on Advances in Microfluidics & Nanofluidics Asian-Pacific International Symposium on Lab on Chip*, Singapore, 2011.

REFERENCES

- [1] Burns, M.A.; Johnson, B.N.; Brahmasandra, S.N.; Handique, K.; Webster, J.R.; Krishnan, M.; Sammarco, T.S.; Man, P.M.; Jones, D.; Heldsinger, D.; Mastrangelo, C.H.; Burke, D.T. An integrated nanoliter DNA analysis device. *Science*, 1998, 282(5388), 484-487.
- [2] Gerdt, C.J.; Sharoyan, D.E.; Ismagilov, R.F. A synthetic reaction network: Chemical amplification using nonequilibrium autocatalytic reactions coupled in time. *J. Am. Chem. Soc.*, 2004, 126(20), 6327-6331.
- [3] Griffiths, A.D.; Tawfik, D.S. Miniaturising the laboratory in emulsion droplets. *Trends Biotechnol.*, 2006, 24(9), 395-402.
- [4] Fuerstman, M.J.; Garstecki, P.; Whitesides, G.M. Coding/decoding and reversibility of droplet trains in microfluidic networks. *Science*, 2007, 315(5813), 828-832.
- [5] Saeki, D.; Sugiura, S.; Kanamori, T.; Sato, S.; Ichikawa, S. Formation of monodisperse calcium alginate microbeads by rupture of water-in-oil-in-water droplets with an ultra-thin oil phase layer. *Lab Chip*, 2010, 10(17), 2292-2295.
- [6] Niu, X.; Zhang, M.; Wu, J.; Wen, W.; Sheng, P. Generation and manipulation of "smart" droplets. *Soft Matter*, 2009, 5(3), 576-581.
- [7] Srisa-Art, M.; Demello, A.J.; Edel, J.B. Fluorescence lifetime imaging of mixing dynamics in continuous-flow microdroplet reactors. *Phys. Rev. Lett.*, 2008, 101(1).
- [8] Song, H.; Chen, D.L.; Ismagilov, R.F. Reactions in droplets in microfluidic channels. *Angew. Chem. Int. Ed.*, 2006, 45(44), 7336-7356.
- [9] Burns, M.A.; Mastrangelo, C.H.; Sammarco, T.S.; Man, F.P.; Webster, J.R.; Johnson, B.N.; Foerster, B.; Jones, D.; Fields, Y.; Kaiser, A.R.; Burke, D.T. Microfabricated structures for integrated DNA analysis. *Proc. Nat. Acad. Sci. U.S.A.*, 1996, 93(11), 5556-5561.
- [10] Cho, S.K.; Moon, H.; Kim, C.J. Creating, transporting, cutting, and merging liquid droplets by electrowetting-based actuation for digital microfluidic circuits. *J. Microelectromech. Syst.*, 2003, 12(1), 70-80.

- [11] Lee, J.N.; Park, C.; Whitesides, G.M. Solvent Compatibility of Poly(dimethylsiloxane)-Based Microfluidic Devices. *Anal. Chem.*, 2003, 75(23), 6544-6554.
- [12] Garstecki, P.; Fuerstman, M.J.; Stone, H.A.; Whitesides, G.M. Formation of droplets and bubbles in a microfluidic T-junction - Scaling and mechanism of break-up. *Lab Chip*, 2006, 6(3), 437-446.
- [13] Xu, J.H.; Li, S.W.; Tân, J.; Wang, Y.J.; Luo, G.S. Preparation of highly monodisperse droplet in a T-junction microfluidic device. *AIChE J.*, 2006, 52(9), 3005-3010.
- [14] Nguyen, N.T.; Ting, T.H.; Yap, Y.F.; Wong, T.N.; Chai, J.C.K.; Ong, W.L.; Zhou, J.; Tan, S.H.; Yobas, L. Thermally mediated droplet formation in microchannels. *Appl. Phys. Lett.*, 2007, 91(8).
- [15] Xu, J.H.; Li, S.W.; Lan, W.J.; Luo, G.S. Microfluidic approach for rapid interfacial tension measurement. *Langmuir*, 2008, 24(19), 11287-11292.
- [16] Christopher, G.F.; Anna, S.L. Microfluidic methods for generating continuous droplet streams. *J. Phys. D: Appl. Phys.*, 2007, 40(19), R319-R336.
- [17] Mazouchi, A.; Homsy, G.M. Thermocapillary migration of long bubbles in cylindrical capillary tubes. *Phys. Fluids*, 2000, 12(3), 542-549.
- [18] Pollack, M.G.; Fair, R.B.; Shenderov, A.D. Electrowetting-based actuation of liquid droplets for microfluidic applications. *Appl. Phys. Lett.*, 2000, 77(11), 1725-1726.
- [19] Teh, S.Y.; Lin, R.; Hung, L.H.; Lee, A.P. Droplet microfluidics. *Lab Chip*, 2008, 8(2), 198-220.
- [20] Yang, C.G.; Xu, Z.R.; Wang, J.H. Manipulation of droplets in microfluidic systems. *TrAC, Trends Anal. Chem.*, 2010, 29(2), 141-157.
- [21] De Menech, M. Modeling of droplet breakup in a microfluidic T-shaped junction with a phase-field model. *Phys. Rev. E: Stat. Nonlinear Soft Matter Phys.*, 2006, 73(3), 1-9.
- [22] Sang, L.; Hong, Y.; Wang, F. Investigation of viscosity effect on droplet formation in T-shaped microchannels by numerical and analytical methods. *Microfluid. Nanofluid.*, 2009, 6(5), 621-635.

- [23] Wu, L.; Tsutahara, M.; Kim, L.S.; Ha, M. Three-dimensional lattice Boltzmann simulations of droplet formation in a cross-junction microchannel. *Int. J. Multiphase Flow*, 2008, 34(9), 852-864.
- [24] Coulliette, C.; Pozrikidis, C. Motion of an array of drops through a cylindrical tube. *J. Fluid Mech.*, 1998, 358, 1-28.
- [25] Cristini, V.; Bławdziewicz, J.; Loewenberg, M. Drop breakup in three-dimensional viscous flows. *Phys. Fluids*, 1998, 10(8), 1781-1783.
- [26] Quan, S.; Lou, J.; Schmidt, D.P. Modeling merging and breakup in the moving mesh interface tracking method for multiphase flow simulations. *J. Comput. Phys.*, 2009, 228(7), 2660-2675.
- [27] Sussman, M. A level set approach for computing solutions to incompressible two-phase flow. *J. Comput. Phys.*, 1994, 114(1), 146-159.
- [28] Sussman, M.; Fatemi, E. Efficient, interface-preserving level set redistancing algorithm and its application to interfacial incompressible fluid flow. *SIAM J. Sci. Comput.*, 1999, 20(4), 1165-1191.
- [29] Torres, D.J.; Brackbill, J.U. The Point-Set Method: Front-Tracking without Connectivity. *J. Comput. Phys.*, 2000, 165(2), 620-644.
- [30] Zhang, H.; Zheng, L.L.; Prasad, V.; Hou, T.Y. A curvilinear level set formulation for highly deformable free surface problems with application to solidification. *Numer. Heat Transfer, Part B*, 1998, 34(1), 1-20.
- [31] Zheng, L.L.; Zhang, H. An adaptive level set method for moving-boundary problems: Application to droplet spreading and solidification. *Numer. Heat Transfer, Part B*, 2000, 37(1-4), 437-454.
- [32] Takada, N.; Misawa, M.; Tomiyama, A. A phase-field method for interface-tracking simulation of two-phase flows. *Math. Comput. Simul.*, 2006, 72(2-6), 220-226.
- [33] Yap, Y.F.; Chai, J.C.; Wong, T.N.; Toh, K.C.; Zhang, H.Y. A global mass correction scheme for the level-set method. *Numer. Heat Transfer, Part B*, 2006, 50(5), 455-472.
- [34] Spelt, P.D.M. A level-set approach for simulations of flows with multiple moving contact lines with hysteresis. *J. Comput. Phys.*, 2005, 207(2), 389-404.

- [35] Spelt, P.D.M. Shear flow past two-dimensional droplets pinned or moving on an adhering channel wall at moderate Reynolds numbers: A numerical study. *J. Fluid Mech.*, 2006, 561, 439-463.
- [36] Korlie, M.S.; Mukherjee, A.; Nita, B.G.; Stevens, J.G.; Trubatch, A.D.; Yecko, P. Modeling bubbles and droplets in magnetic fluids. *J. Phys. Condens. Matter*, 2008, 20(20).
- [37] Tryggvason, G.; Bunner, B.; Esmaeeli, A.; Juric, D.; Al-Rawahi, N.; Tauber, W.; Han, J.; Nas, S.; Jan, Y.J. A Front-Tracking Method for the Computations of Multiphase Flow. *J. Comput. Phys.*, 2001, 169(2), 708-759.
- [38] Yap, Y.F.; Tan, S.H.; Nguyen, N.T.; Murshed, S.M.S.; Wong, T.N.; Yobas, L. Thermally mediated control of liquid microdroplets at a bifurcation. *J. Phys. D: Appl. Phys.*, 2009, 42(6).
- [39] Nisisako, T.; Torii, T.; Higuchi, T. Droplet formation in a microchannel network. *Lab Chip*, 2002, 2(1), 24-26.
- [40] Song, H.; Tice, J.D.; Ismagilov, R.F. A Microfluidic System for Controlling Reaction Networks in Time. *Angew. Chem. Int. Ed.*, 2003, 42(7), 768-772.
- [41] Bringer, M.R.; Gerdts, C.J.; Song, H.; Tice, J.D.; Ismagilov, R.F. Microfluidic systems for chemical kinetics that rely on chaotic mixing in droplets. *Philos. Trans. R. Soc. London, Ser. A*, 2004, 362(1818), 1087-1104.
- [42] Cristobal, G.; Benoit, J.P.; Joanicot, M.; Ajdari, A. Microfluidic bypass for efficient passive regulation of droplet traffic at a junction. *Appl. Phys. Lett.*, 2006, 89(3).
- [43] Link, D.R.; Anna, S.L.; Weitz, D.A.; Stone, H.A. Geometrically Mediated Breakup of Drops in Microfluidic Devices. *Phys. Rev. Lett.*, 2004, 92(5), 054503.
- [44] Prakash, M.; Gershenfeld, N. Microfluidic bubble logic. *Science*, 2007, 315(5813), 832-835.
- [45] Stemme, E.; Stemme, G. A valveless diffuser/nozzle-based fluid pump. *Sens. Actuators, A*, 1993, 39(2), 159-167.
- [46] Gerlach, T.; Wurmus, H. Working principle and performance of the dynamic micropump. In: *Proceedings of the 1995 IEEE Micro Electro Mechanical Systems Conference*, Amsterdam, Neth, Jan 29 - Feb 2 1995.

- [47] Gerlach, T. Microdiffusers as dynamic passive valves for micropump applications. *Sens. Actuators, A*, 1998, 69(2), 181-191.
- [48] Olsson, A.; Stemme, G.; Stemme, E. Numerical and experimental studies of flat-walled diffuser elements for valve-less micropumps. *Sens. Actuators, A*, 2000, 84(1), 165-175.
- [49] Heschel, M.; Miillenborn, M.; Bouwstra, S. Fabrication and characterization of truly 3-D diffuser/nozzle microstructures in silicon. *J. Microelectromech. Syst.*, 1997, 6(1), 41-46.
- [50] Nguyen, N.T.; Huang, X. Miniature valveless pumps based on printed circuit board technique. *Sens. Actuators, A*, 2001, 88(2), 104-111.
- [51] Groisman, A.; Quake, S.R. A microfluidic rectifier: Anisotropic flow resistance at low Reynolds numbers. *Phys. Rev. Lett.*, 2004, 92(9), 094501-1.
- [52] Nguyen, N.T.; Lam, Y.C.; Ho, S.S. Improvement of rectification effects in diffuser/nozzle structures with viscoelastic fluids. *Biomicrofluidics*, 2008, 2(3).
- [53] Dixit, S.S.; Kim, H.; Vasilyev, A.; Eid, A.; Faris, G.W. Light-driven formation and rupture of droplet bilayers. *Langmuir*, 2010, 26(9), 6193-6200.
- [54] Baroud, C.N.; Delville, J.P.; Gallaire, F.; Wunenburger, R. Thermocapillary valve for droplet production and sorting. *Phys. Rev. E: Stat. Nonlinear Soft Matter Phys.*, 2007, 75(4).
- [55] Baroud, C.N.; Robert De Saint Vincent, M.; Delville, J.P. An optical toolbox for total control of droplet microfluidics. *Lab Chip*, 2007, 7(8), 1029-1033.
- [56] Brochard, F. Motions of droplets on solid surfaces induced by chemical or thermal gradients. *Langmuir*, 1989, 5(2), 432-438.
- [57] Ford, M.L.; Nadim, A. Thermocapillary migration of an attached drop on a solid surface. *Phys. Fluids*, 1994, 6(9), 3183-3185.
- [58] Brzoska, J.B.; Brochard-Wyart, F.; Rondelez, F. Motions of droplets on hydrophobic model surfaces induced by thermal gradients. *Langmuir*, 1993, 9(8), 2220-2224.
- [59] Yakhshi-Tafti, E.; Cho, H.J.; Kumar, R. Discrete Droplet Manipulation on Liquid Platforms using Thermal Gradients. *Procedia Chemistry*, 2009, 1(1),

1519-1522.

- [60] Long, Z.; Shetty, A.M.; Solomon, M.J.; Larson, R.G. Fundamentals of magnet-actuated droplet manipulation on an open hydrophobic surface. *Lab Chip*, 2009, 9(11), 1567-1575.
- [61] Darhuber, A.A.; Valentino, J.P.; Davis, J.M.; Troian, S.M.; Wagner, S. Microfluidic actuation by modulation of surface stresses. *Appl. Phys. Lett.*, 2003, 82(4), 657-659.
- [62] Sammarco, T.S.; Burns, M.A. Thermocapillary pumping of discrete drops in microfabricated analysis devices. *AIChE J.*, 1999, 45(2), 350-366.
- [63] DeBar, M.J.; Liepmann, D. *Fabrication and performance testing of a steady thermocapillary pump with no moving parts*. In: *Proceedings of the IEEE Micro Electro Mechanical Systems (MEMS)*, Las Vegas, NV, USA, Jan 20-24, 2002.
- [64] Glockner, P.S.; Naterer, G.F. Thermocapillary control of microfluidic transport with a stationary cyclic heat source. *J. Micromech. Microeng.*, 2005, 15(12), 2216-2229.
- [65] Selva, B.; Miralles, V.; Cantat, I.; Jullien, M.C. Thermocapillary actuation by optimized resistor pattern: Bubbles and droplets displacing, switching and trapping. *Lab Chip*, 2010, 10(14), 1835-1840.
- [66] Nguyen, N.T.; Huang, X. Thermocapillary effect of a liquid plug in transient temperature fields. *Jpn. J. Appl. Phys., Part 1*, 2005, 44(2), 1139-1142.
- [67] Jiao, Z.; Nguyen, N.T.; Huang, X. Thermocapillary actuation of liquid plugs using a heater array. *Sens. Actuators, A*, 2007, 140(2), 145-155.
- [68] Cordero, M.L.; Rolfsnes, H.O.; Burnham, D.R.; Campbell, P.A.; McGloin, D.; Baroud, C.N. Mixing via thermocapillary generation of flow patterns inside a microfluidic drop. *New J. Phys.*, 2009, 11.
- [69] Cordero, M.L.; Burnham, D.R.; Baroud, C.N.; McGloin, D. Thermocapillary manipulation of droplets using holographic beam shaping: Microfluidic pin ball. *Appl. Phys. Lett.*, 2008, 93(3).
- [70] Sung, T.I.; Choi, H.S.; Kim, Y.S.; Park, I.H. *Modeling and analyzing of electrowetting using electromagnetic body force density and surface tension*. In: *Proceedings of 14th Biennial IEEE Conference on Electromagnetic Field*

- Computation (CEFC)*, Chicago, IL, May 9-12, 2010.
- [71] Liu, C.X.; Park, J.; Choi, J.W. A planar lens based on the electrowetting of two immiscible liquids. *J. Micromech. Microeng.*, 2008, *18*(3).
 - [72] Lippmann, G. Relations entre les phénomènes électriques et capillaires. *Ann. Chim. Phys.*, 1875, *5*, 494-549.
 - [73] Beni, G.; Hackwood, S. Electro-wetting displays. *Appl. Phys. Lett.*, 1981, *38*(4), 207-209.
 - [74] Beni, G.; Hackwood, S.; Jackel, J.L. Continuous electrowetting effect. *Appl. Phys. Lett.*, 1982, *40*(10), 912-914.
 - [75] Verheijen, H.J.J.; Prins, M.W.J. Reversible electrowetting and trapping of charge: Model and experiments. *Langmuir*, 1999, *15*(20), 6616-6620.
 - [76] Colgate, E.; Matsumoto, H. An investigation of electrowetting - based microactuation. *J. Vac. Sci. Technol. A*, 1990, *8*(4), 3625-3833.
 - [77] Matsumoto, H.; Colgate, J.E. *Preliminary investigation of micropumping based on electrical control of interfacial tension*. In: *Proceedings of IEEE Micro Electro Mechanical Systems-An Investigation of Micro Structures, Sensors, Actuators, Machines*, , Napa Valley, CA , USA, 11-14 Feb, 1990.
 - [78] Yun, K.S.; Cho, I.J.; Bu, J.U.; Kim, G.H.; Jeon, Y.S.; Kim, C.J.; Yoon, E. A micropump driven by continuous electrowetting actuation for low voltage and low power operations. In: *The 14th IEEE International Conference on Micro Electro Mechanical Systems*, Interlaken, August 07, 2002.
 - [79] Pollack, M.G.; Shenderov, A.D.; Fair, R.B. Electrowetting-based actuation of droplets for integrated microfluidics. *Lab Chip*, 2002, *2*(2), 96-101.
 - [80] Lee, J.; Moon, H.; Fowler, J.; Schoellhammer, T.; Kim, C.J. Electrowetting and electrowetting-on-dielectric for microscale liquid handling. *Sens. Actuators, A*, 2002, *95*(2-3), 259-268.
 - [81] Feng, J.T.; Zhao, Y.P. Experimental observation of electrical instability of droplets on dielectric layer. *J. Phys. D: Appl. Phys.*, 2008, *41*(5).
 - [82] Moon, H.; Cho, S.K.; Garrell, R.L.; Kim, C.J. Low voltage electrowetting-on-dielectric. *J. Appl. Phys.*, 2002, *92*(7), 4080.

- [83] Mahmud, T.; Rosengarten, G.; Holland, A. *Droplet manipulation for lab-on-a-chip using electrowetting*. In: *Progress in Biomedical Optics and Imaging - Proceedings of SPIE* 6036, Brisbane, Australia, December 12 2005.
- [84] Cooney, C.G.; Chen, C.Y.; Emerling, M.R.; Nadim, A.; Sterling, J.D. Electrowetting droplet microfluidics on a single planar surface. *Microfluid. Nanofluid.*, 2006, 2(5), 435-446.
- [85] Cho, S.K.; Zhao, Y.; Kim, C.J. Concentration and binary separation of micro particles for droplet-based digital microfluidics. *Lab Chip*, 2007, 7(4), 490-498.
- [86] Song, J.H.; Evans, R.; Lin, Y.Y.; Hsu, B.N.; Fair, R.B. A scaling model for electrowetting-on-dielectric microfluidic actuators. *Microfluid. Nanofluid.*, 2009, 7(1), 75-89.
- [87] Kumari, N.; Bahadur, V.; Garimella, S.V. Electrical actuation of electrically conducting and insulating droplets using ac and dc voltages. *J. Micromech. Microeng.*, 2008, 18(10).
- [88] Pamme, N. Magnetism and microfluidics. *Lab Chip*, 2006, 6(1), 24-38.
- [89] Zborowski, M.; Sun, L.; Moore, L.R.; Stephen Williams, P.; Chalmers, J.J. Continuous cell separation using novel magnetic quadrupole flow sorter. *J. Magn. Magn. Mater.*, 1999, 194(1), 224-230.
- [90] Feldmann, M.; Demming, S.; Büttgenbach, S. *Novel electro magnetic micro pump*. In: *NSTI Nanotechnology Conference and Trade Show - NSTI Nanotech 2007, Technical Proceedings* 3, Santa Clara, CA ,May 20-24 2007.
- [91] Ahn, C.H.; Allen, M.G.; Trimmer, W.; Jun, Y.N.; Erramilli, S. A fully integrated micromachined magnetic particle separator. *J. Microelectromech. Syst.*, 1996, 5(3), 151-158.
- [92] Choi, J.W.; Liakopoulos, T.M.; Ahn, C.H. An on-chip magnetic bead separator using spiral electromagnets with semi-encapsulated permalloy. *Biosens. Bioelectron.*, 2001, 16(6), 409-416.
- [93] Šafařík, I.; Šafaříková, M. Use of magnetic techniques for the isolation of cells. *J. Chromatogr. B*, 1999, 722(1-2), 33-53.
- [94] Rida, A.; Gijs, M.A.M. Dynamics of magnetically retained supraparticle structures in a liquid flow. *Appl. Phys. Lett.*, 2004, 85(21), 4986-4988.

- [95] Rida, A.; Fernandez, V.; Gijs, M.A.M. Long-range transport of magnetic microbeads using simple planar coils placed in a uniform magnetostatic field. *Appl. Phys. Lett.*, 2003, 83(12), 2396-2398.
- [96] Lehmann, U.; Hadjidj, S.; Parashar, V.K.; Vandevyver, C.; Rida, A.; Gijs, M.A.M. Two-dimensional magnetic manipulation of microdroplets on a chip as a platform for bioanalytical applications. *Sens. Actuators, B*, 2006, 117(2), 457-463.
- [97] Tsuchiya, H.; Okochi, M.; Nagao, N.; Shikida, M.; Honda, H. On-chip polymerase chain reaction microdevice employing a magnetic droplet-manipulation system. *Sens. Actuators, B*, 2008, 130(2), 583-588.
- [98] Shikida, M.; Takayanagi, K.; Inouchi, K.; Honda, H.; Sato, K. Using wettability and interfacial tension to handle droplets of magnetic beads in a micro-chemical-analysis system. *Sens. Actuators, B*, 2006, 113(1), 563-569.
- [99] Nguyen, N.T.; Ng, K.M.; Huang, X. *Magnetic actuation for microfluidics based on ferrofluid droplets*. In, 2007.
- [100] Rosensweig, R.E. *Ferrohydrodynamics*. Cambridge University Press, 1985.
- [101] Nguyen, N.T.; Ng, K.M.; Huang, X. Manipulation of ferrofluid droplets using planar coils. *Appl. Phys. Lett.*, 2006, 89(5).
- [102] Nguyen, N.T.; Zhu, G.; Chua, Y.C.; Phan, V.N.; Tan, S.H. Magnetowetting and sliding motion of a sessile ferrofluid droplet in the presence of a permanent magnet. *Langmuir*, 2010, 26(15), 12553-12559.
- [103] Tan, S.H.; Nguyen, N.T.; Yobas, L.; Kang, T.G. Formation and manipulation of ferrofluid droplets at a microfluidic T-junction. *J. Micromech. Microeng.*, 2010, 20(4).
- [104] Lavrova, O.; Matthies, G.; Mitkova, T.; Polevikov, V.; Tobiska, L. Numerical treatment of free surface problems in ferrohydrodynamics. *J. Phys. Condens. Matter*, 2006, 18(38), S2657-S2669.
- [105] Sero-Guillaume, O.E.; Zouaoui, D.; Bernardin, D.; Brancher, J.P. The shape of a magnetic liquid drop. *J. Fluid Mech.*, 1992, 241, 215-232.
- [106] Afkhami, S.; Renardy, Y.; Renardy, M.; Riffle, J.S.; St Perre, T. Field-induced motion of ferrofluid droplets through immiscible viscous media. *J. Fluid Mech.*, 2008, 610, 363-380.

- [107] Slavov, V.; Dimova, S. *Phase-field versus level set method for 2D dendritic growth*. In, 2007.
- [108] Cristini, V.; Tan, Y.C. Theory and numerical simulation of droplet dynamics in complex flows - A review. *Lab Chip*, 2004, 4(4), 257-264.
- [109] Yap, Y.F. *Numerical modeling of multiphase flows in microchannels*. PhD Thesis, Nanyang Technological University: Singapore, 2007.
- [110] Garrioch, S.H.; Baliga, B.R. A PLIC volume tracking method for the simulation of two-fluid flows. *Int. J. Numer. Methods Fluids*, 2006, 52(10), 1093-1134.
- [111] Pozrikidis, C. Interfacial dynamics for Stokes flow. *J. Comput. Phys.*, 2001, 169(2), 250-301.
- [112] Hou, T.Y.; Lowengrub, J.S.; Shelley, M.J. Boundary Integral Methods for Multicomponent Fluids and Multiphase Materials. *J. Comput. Phys.*, 2001, 169(2), 302-362.
- [113] Cristini, V.; Blawdziewicz, J.; Loewenberg, M. An Adaptive Mesh Algorithm for Evolving Surfaces: Simulations of Drop Breakup and Coalescence. *J. Comput. Phys.*, 2001, 168(2), 445-463.
- [114] Zinchenko, A.Z.; Rother, M.A.; Davis, R.H. Cusping, capture, and breakup of interacting drops by a curvatureless boundary-integral algorithm. *J. Fluid Mech.*, 1999, 391, 249-292.
- [115] Janssen, P.A.; Anderson, P.D. Boundary-integral method for drop deformation between parallel plates. *Phys. Fluids*, 2007, 19(4).
- [116] Zinchenko, A.Z.; Davis, R.H. *Boundary-integral algorithm for drop squeezing through a granular material*. In: *9th International Conference on Multiscale and Functionally Graded Materials, FGM IX*, Oahu Island, HI, October 15-18, 2008.
- [117] Chraïbi, H.; Lasseux, D.; Arquis, E.; Wunenburger, R.; Delville, J.P. Stretching and squeezing of sessile dielectric drops by the optical radiation pressure. *Phys. Rev. E: Stat. Nonlinear Soft Matter Phys.*, 2008, 77(6).
- [118] Glimm, J.; Grove, J.W.; Li, X.L.; Shyue, K.M.; Zeng, Y.; Zhang, Q. Three-dimensional front tracking. *SIAM J. Sci. Comput.*, 1998, 19(3), 703-727.

- [119] Unverdi, S.O. A front-tracking method for viscous, incompressible, multifluid flows. *J. Comput. Phys.*, 1992, 100(1), 25-37.
- [120] Agresar, G.; Linderman, J.J.; Tryggvason, G.; Powell, K.G. An Adaptive, Cartesian, Front-Tracking Method for the Motion, Deformation and Adhesion of Circulating Cells. *J. Comput. Phys.*, 1998, 143(2), 346-380.
- [121] Esmaeeli, A.; Tryggvason, G. Computations of Explosive Boiling in Microgravity. *J. Sci. Comput.*, 2003, 19(1-3), 163-182.
- [122] Esmaeeli, A.; Tryggvason, G. Computations of film boiling. Part I: Numerical method. *Int. J. Heat Mass Transfer*, 2004, 47(25), 5451-5461.
- [123] Mao, D.k. Towards front-tracking based on conservation in two space dimensions II, tracking discontinuities in capturing fashion. *J. Comput. Phys.*, 2007, 226(2), 1550-1588.
- [124] Liu, J.; Lim, H.K.; Glimm, J.; Li, X. A conservative front tracking method in N-dimensions. *J. Sci. Comput.*, 2007, 31(1-2), 213-236.
- [125] Gois, J.P.; Nakano, A.; Nonato, L.G.; Buscaglia, G.C. Front tracking with moving-least-squares surfaces. *J. Comput. Phys.*, 2008, 227(22), 9643-9669.
- [126] Dijkhuizen, W.; Roghair, I.; Annaland, M.V.S.; Kuipers, J.A.M. DNS of gas bubbles behaviour using an improved 3D front tracking model-Model development. *Chem. Eng. Sci.*, 2010, 65(4), 1427-1437.
- [127] Scardovelli, R.; Zaleski, S. Direct numerical simulation of free-surface and interfacial flow. *Annu. Rev. Fluid Mech.*, 1999, 31, 567-603.
- [128] Hirt, C.W.; Nichols, B.D. Volume of fluid (VOF) method for the dynamics of free boundaries. *J. Comput. Phys.*, 1981, 39(1), 201-225.
- [129] Noh, W.; Woodward, P. SLIC-simple line interface calculation *International Conference on Fluid Dynamics*, 1976, 59, 330-340.
- [130] Gueyffier, D.; Li, J.; Nadim, A.; Scardovelli, R.; Zaleski, S. Volume-of-Fluid Interface Tracking with Smoothed Surface Stress Methods for Three-Dimensional Flows. *J. Comput. Phys.*, 1999, 152(2), 423-456.
- [131] Scardovelli, R.; Zaleski, S. Interface reconstruction with least-square fit and split Eulerian-Lagrangian advection. *Int. J. Numer. Methods Fluids*, 2003,

41(3), 251-274.

- [132] Renardy, Y.; Renardy, M. PROST: A parabolic reconstruction of surface tension for the volume-of-fluid method. *J. Comput. Phys.*, 2002, 183(2), 400-421.
- [133] Rudman, M. Volume-tracking methods for interfacial flow calculations. *Int. J. Numer. Methods Fluids*, 1997, 24(7), 671-691.
- [134] Gopala, V.R.; van Wachem, B.G.M. Volume of fluid methods for immiscible-fluid and free-surface flows. *Chem. Eng. J.*, 2008, 141(1-3), 204-221.
- [135] Boris, J.P.; Book, D.L. Flux-corrected transport. I. SHASTA, a fluid transport algorithm that works. *J. Comput. Phys.*, 1973, 11(1), 38-69.
- [136] Ubbink, O.; Issa, R.I. A Method for Capturing Sharp Fluid Interfaces on Arbitrary Meshes. *J. Comput. Phys.*, 1999, 153(1), 26-50.
- [137] Jasak, H.; Weller, H.G.; Gosman, A.D. High resolution NVD differencing scheme for arbitrarily unstructured meshes. *Int. J. Numer. Methods Fluids*, 1999, 31(2), 431-449.
- [138] Van Wachem, B.G.M.; Schouten, J.C. Experimental validation of 3-D Lagrangian VOF model: Bubble shape and rise velocity. *AIChE J.*, 2002, 48(12), 2744-2753.
- [139] Park, I.R.; Kim, K.S.; Kim, J.; Van, S.H. A volume-of-fluid method for incompressible free surface flows. *Int. J. Numer. Methods Fluids*, 2009, 61(12), 1331-1362.
- [140] Son, G. Efficient implementation of a coupled level-set and volume-of-fluid method for three-dimensional incompressible two-phase flows. *Numer. Heat Transfer, Part B*, 2003, 43(6), 549-565.
- [141] Sussman, M.; Puckett, E.G. A Coupled Level Set and Volume-of-Fluid Method for Computing 3D and Axisymmetric Incompressible Two-Phase Flows. *J. Comput. Phys.*, 2000, 162(2), 301-337.
- [142] Sussman, M.; Smith, K.M.; Hussaini, M.Y.; Ohta, M.; Zhi-Wei, R. A sharp interface method for incompressible two-phase flows. *J. Comput. Phys.*, 2007, 221(2), 469-505.

- [143] van der Pijl, S.P.; Segal, A.; Vuik, C.; Wesseling, P. A mass-conserving Level-Set method for modelling of multi-phase flows. *Int. J. Numer. Methods Fluids*, 2005, 47(4), 339-361.
- [144] Wang, Z.; Yang, J.; Koo, B.; Stern, F. A coupled level set and volume-of-fluid method for sharp interface simulation of plunging breaking waves. *Int. J. Multiphase Flow*, 2009, 35(3), 227-246.
- [145] Cahn, J.W.; Hilliard, J.E. Free energy of a nonuniform system. III. Nucleation in a two-component incompressible fluid. *J. Chem. Phys.*, 1959, 31(3), 688-699.
- [146] Liu, C.; Shen, J. A phase field model for the mixture of two incompressible fluids and its approximation by a Fourier-spectral method. *Physica D*, 2003, 179(3-4), 211-228.
- [147] Liu, C.; Walkington, N.J. An Eulerian description of fluids containing visco-elastic particles. *Arch. Ration. Mech. Anal.*, 2001, 159(3), 229-252.
- [148] Zhang, Z.; Tang, H. An adaptive phase field method for the mixture of two incompressible fluids. *Comput. Fluids*, 2007, 36(8), 1307-1318.
- [149] Kim, J.; Kang, K.; Lowengrub, J. Conservative multigrid methods for Cahn-Hilliard fluids. *J. Comput. Phys.*, 2004, 193(2), 511-543.
- [150] Kim, J. A diffuse-interface model for axisymmetric immiscible two-phase flow. *Appl. Math. Comput.*, 2005, 160(2), 589-606.
- [151] Kim, C.H.; Shin, S.H.; Lee, H.G.; Kim, J. Phase-field model for the pinchoff of liquid-liquid jets. *J. Korean Phys. Soc.*, 2009, 55(4), 1451-1460.
- [152] Kim, J. A generalized continuous surface tension force formulation for phase-field models for multi-component immiscible fluid flows. *Comput. Meth. Appl. Mech. Eng.*, 2009, 198(37-40), 3105-3112.
- [153] Chen, S.; Doolen, G.D. Lattice Boltzmann model for fluid flows. *Annu. Rev. Fluid. Mech.*, 1998, 30, 329-364.
- [154] Lamura, A.; Gonnella, G.; Yeomans, J.M. A lattice Boltzmann model of ternary fluid mixtures. *Europhys. Lett.*, 1999, 45(3), 314-320.
- [155] Shan, X.; Chen, H. Lattice Boltzmann model for simulating flows with

- multiple phases and components. *Phys. Rev. E*, 1993, 47(3), 1815-1819.
- [156] Nourgaliev, R.R.; Dinh, T.N.; Theofanous, T.G.; Joseph, D. The lattice Boltzmann equation method: Theoretical interpretation, numerics and implications. *Int. J. Multiphase Flow*, 2003, 29(1), 117-169.
- [157] Sankaranarayanan, K.; Kevrekidis, I.G.; Sundaresan, S.; Lu, J.; Tryggvason, G. A comparative study of lattice Boltzmann and front-tracking finite-difference methods for bubble simulations. *Int. J. Multiphase Flow*, 2003, 29(1), 109-116.
- [158] Van Der Graaf, S.; Nisisako, T.; Schroën, C.G.P.H.; Van Der Sman, R.G.M.; Boom, R.M. Lattice Boltzmann simulations of droplet formation in a T-shaped microchannel. *Langmuir*, 2006, 22(9), 4144-4152.
- [159] Gupta, A.; Kumar, R. Effect of geometry on droplet formation in the squeezing regime in a microfluidic T-junction. *Microfluid. Nanofluid.*, 2010, 8(6), 799-812.
- [160] Osher, S.; Sethian, J.A. Fronts propagating with curvature-dependent speed: Algorithms based on Hamilton-Jacobi formulations. *J. Comput. Phys.*, 1988, 79(1), 12-49.
- [161] Yap, Y.F.; Chai, J.C.; Wong, T.N.; Nguyen, N.T.; Toh, K.C.; Zhang, H.Y.; Yobas, L. A procedure for the motion of particle-encapsulated droplets in microchannels. *Numer. Heat Transfer, Part B*, 2008, 53(1), 59-74.
- [162] Osher, S.; Fedkiw, R.P. *Level set methods and dynamic implicit surfaces*. Springer-Verlag, New York, 2003.
- [163] Peng, D.; Merriman, B.; Osher, S.; Zhao, H.; Kang, M. A PDE-Based Fast Local Level Set Method. *J. Comput. Phys.*, 1999, 155(2), 410-438.
- [164] Chang, Y.C.; Hou, T.Y.; Merriman, B.; Osher, S. A level set formulation of Eulerian interface capturing methods for incompressible fluid flows. *J. Comput. Phys.*, 1996, 124(2), 449-464.
- [165] Lakehal, D.; Meier, M.; Fulgosi, M. Interface tracking towards the direct simulation of heat and mass transfer in multiphase flows. *Int. J. Heat Fluid Flow*, 2002, 23(3), 242-257.
- [166] Harlow, F.H.; Welch, J.E. Numerical calculation of time-dependent viscous incompressible flow of fluid with free surface. *Phys. Fluids*, 1965, 8(12),

2182-2189.

- [167] Chen, S.; Johnson, D.B.; Raad, P.E.; Fadda, D. The surface marker and micro cell method. *Int. J. Numer. Methods Fluids*, 1997, 25(7), 749-778.
- [168] Enright, D.; Fedkiw, R.; Ferziger, J.; Mitchell, I. A hybrid particle level set method for improved interface capturing. *J. Comput. Phys.*, 2002, 183(1), 83-116.
- [169] Son, G. A numerical method for incompressible two-phase flows with open or periodic boundaries. *Numer. Heat Transfer, Part B*, 2001, 39(1), 45-60.
- [170] Ni, M.J.; Abdou, M.; Komori, S. A variable-density projection method for interfacial flows. *Numer. Heat Transfer, Part B*, 2003, 44(6), 553-574.
- [171] Son, G. A level set method for incompressible two-fluid flows with immersed solid boundaries. *Numer. Heat Transfer, Part B*, 2005, 47(5), 473-489.
- [172] Nochetto, R.H.; Walker, S.W. A hybrid variational front tracking-level set mesh generator for problems exhibiting large deformations and topological changes. *J. Comput. Phys.*, 2010, 229(18), 6243-6269.
- [173] Tanguy, S.; Berlemont, A. Application of a level set method for simulation of droplet collisions. *Int. J. Multiphase Flow*, 2005, 31(9), 1015-1035.
- [174] Croce, R.; Griebel, M.; Schweitzer, M.A. Numerical simulation of bubble and droplet deformation by a level set approach with surface tension in three dimensions. *Int. J. Numer. Methods Fluids*, 2010, 62(9), 963-993.
- [175] Brackbill, J.U. A continuum method for modeling surface tension. *J. Comput. Phys.*, 1992, 100(2), 335-354.
- [176] Dussan V, E.B.; Davis, S.H. The motion of a fluid-fluid interface along a solid surface. *J. Fluid Mech.*, 1974, 65(Part 1), 71-95.
- [177] Ren, W.; E, W. Boundary conditions for the moving contact line problem. *Phys. Fluids*, 2007, 19(2).
- [178] Li, Z.; Lai, M.C.; He, G.; Zhao, H. An augmented method for free boundary problems with moving contact lines. *Comput. Fluids*, 2010, 39(6), 1033-1040.
- [179] Batchelor, G.K.; Moffatt, H.K.; Worster, M.G. *Perspectives in fluid dynamics*.

- cambridge University Press, Cambridge, 2000.
- [180] Ganesan, S.; Tobiska, L. Modelling and simulation of moving contact line problems with wetting effects. *Comput Visual Sci*, 2009, 12(7), 329-336.
 - [181] Patankar, S.V. *Numerical heat transfer and fluid flow*. Hemisphere New York, 1980.
 - [182] Jiang, G.S.; Peng, D. Weighted ENO schemes for Hamilton-Jacobi equations. *SIAM J. Sci. Comput.*, 2000, 21(6), 2126-2143.
 - [183] Balsara, D.S.; Shu, C.W. Monotonicity Preserving Weighted Essentially Non-oscillatory Schemes with Increasingly High Order of Accuracy. *J. Comput. Phys.*, 2000, 160(2), 405-452.
 - [184] Jiang, G.S.; Shu, C.W. Efficient implementation of weighted ENO schemes. *J. Comput. Phys.*, 1996, 126(1), 202-228.
 - [185] Ho, P.C.; Yap, Y.F.; Nguyen, N.T.; Chai, J.C.; Wong, T.N.; Yobas, L. A numerical investigation of thermally mediated droplet formation in a T-junction. In: *Proceedings of the 7th International Conference on Nanochannels, Microchannels, and Minichannels*, Pohang, South Korea, June 22–24, 2009.
 - [186] Tran, L.B.; Udaykumar, H.S. A particle-level set-based sharp interface cartesian grid method for impact, penetration, and void collapse. *J. Comput. Phys.*, 2004, 193(2), 469-510.
 - [187] Wang, Z.; Yang, J.; Stern, F. An improved particle correction procedure for the particle level set method. *J. Comput. Phys.*, 2009, 228(16), 5819-5837.
 - [188] Cochran, J.F.; Heinrich, B., *Application of Maxwell's equations*. 2004.
 - [189] Ki, H. Level set method for two-phase incompressible flows under magnetic fields. *Comput. Phys. Commun.*, 2010, 181(6), 999-1007.
 - [190] Lavrova, O. Numerical methods for axisymmetric equilibrium magnetic fluid shapes. Dissertation, Otto-von-Guericke-Universität Magdeburg, 2006.
 - [191] Extrand, C.W.; Moon, S.I. When Sessile Drops Are No Longer Small: Transitions from Spherical to Fully Flattened. *Langmuir*, 2010, 26 (14), 11815–11822.

- [192] Liu, H.; Krishnan, S.; Marella, S.; Udaykumar, H.S. Sharp interface Cartesian grid method II: A technique for simulating droplet interactions with surfaces of arbitrary shape. *J. Comput. Phys.*, 2005, *210*(1), 32-54.
- [193] Li, S.; Liu, J.; Nguyen, N.T.; Fang, Z.P.; Yoon, S.F. Measurement of buried undercut structures in microfluidic devices by laser fluorescent confocal microscopy. *Appl. Opt.*, 2009, *48*(33), 6432-6441.
- [194] Ogawa, C.; Masubuchi, Y.; Takimoto, J.I.; Koyama, K. Domain structure and MR effect of ferrofluid emulsion. *Int. J. Mod. Phys. B*, 2001, *15*(6-7), 859-863.
- [195] Venkatraman, S.; Nixon, A.; Highe, A. Deformation behavior of poly(dimethyl siloxane) networks. II. Equilibrium swelling. *J. Appl. Polym. Sci.*, 1994, *52*(11), 1619-1627.
- [196] Biresaw, G. Adsorption of amphiphiles at an oil-water vs. an oil-metal interface. *J. Am. Oil Chem. Soc.*, 2005, *82*(4), 285-292.
- [197] Rosensweig, R.E. Magnetic fluids. *Ann. Rev. Fluid Mech.* , 1987, *19*, 437-463.
- [198] Sweby, P.K. High resolution schemes using flux limiters for hyperbolic conservation laws. *SIAM J. Numer. Anal.*, 1984, *21*(5), 995-1011.
- [199] Versteeg, H.K.; Malalaseker, W. *An Introduction to Computational Fluid Dynamics: The Finite Volume Method*, 2nd ed. Prentice Education Limited, England, 2007.

APPENDIX: TVD SCHEMES FOR THE COMBINED CONVECTION AND DIFFUSION

The 3D convection-diffusion equation is

$$\frac{\partial}{\partial x}(\rho u \Phi) + \frac{\partial}{\partial y}(\rho v \Phi) + \frac{\partial}{\partial z}(\rho w \Phi) = \frac{\partial}{\partial x} \left(\Gamma \frac{\partial \Phi}{\partial x} \right) + \frac{\partial}{\partial y} \left(\Gamma \frac{\partial \Phi}{\partial y} \right) + \frac{\partial}{\partial z} \left(\Gamma \frac{\partial \Phi}{\partial z} \right) \quad (\text{A.2})$$

The central difference is used to discretize the diffusion terms. The discretisation of the equation is

$$\begin{aligned} F_e \Phi_e - F_w \Phi_w + F_n \Phi_n - F_s \Phi_s + F_t \Phi_t - F_b \Phi_b \\ = D_e (\Phi_e - \Phi_p) - D_w (\Phi_p - \Phi_w) \\ + D_n (\Phi_n - \Phi_p) - D_s (\Phi_p - \Phi_s) \\ + D_t (\Phi_t - \Phi_p) - D_b (\Phi_p - \Phi_b) \end{aligned} \quad (\text{A.3})$$

where $F = \rho u$ and $D = \frac{\Gamma}{\delta x}$. The subscripts are shown in Fig. 3-5. Φ_e , Φ_w , Φ_n , Φ_s , Φ_t , Φ_b use the TVD scheme. The 3D convection-diffusion equation can be written in a form of

$$a_p \Phi_p = a_w \Phi_w + a_e \Phi_e + a_n \Phi_n + a_s \Phi_s + a_t \Phi_t + a_b \Phi_b + S^{DC} \quad (\text{A.4})$$

where S^{DC} is the deferred correction source term. The coefficients and the source term are

$$a_w = D_w + \max(F_w, 0) \quad (\text{A.5})$$

$$a_e = D_e + \max(-F_e, 0) \quad (\text{A.6})$$

$$a_n = D_n + \max(-F_n, 0) \quad (\text{A.7})$$

$$a_s = D_s + \max(-F_s, 0) \quad (\text{A.8})$$

$$a_t = D_t + \max(-F_t, 0) \quad (\text{A.9})$$

$$a_b = D_b + \max(-F_b, 0) \quad (\text{A.10})$$

$$\begin{aligned}
S_{DC} = & \frac{1}{2} F_e \left[+ (1 - \alpha_e) \Psi(r_e^-) - \alpha_e \psi(r_e^+) \right] (\Phi_E - \Phi_P) \\
& + \frac{1}{2} F_w \left[- (1 - \alpha_w) \Psi(r_w^-) + \alpha_w \psi(r_w^+) \right] (\Phi_P - \Phi_W) \\
& + \frac{1}{2} F_n \left[+ (1 - \alpha_n) \Psi(r_n^-) - \alpha_n \psi(r_n^+) \right] (\Phi_N - \Phi_P) \\
& + \frac{1}{2} F_s \left[- (1 - \alpha_s) \Psi(r_s^-) + \alpha_s \psi(r_s^+) \right] (\Phi_P - \Phi_S) \\
& + \frac{1}{2} F_t \left[+ (1 - \alpha_t) \Psi(r_t^-) - \alpha_t \psi(r_t^+) \right] (\Phi_T - \Phi_P) \\
& + \frac{1}{2} F_b \left[- (1 - \alpha_b) \Psi(r_b^-) + \alpha_b \psi(r_b^+) \right] (\Phi_P - \Phi_B)
\end{aligned} \tag{A.11}$$

$$r_e^- = \frac{(\Phi_{EE} - \Phi_E)/(x_{EE} - x_E)}{(\Phi_E - \Phi_P)/(x_E - x_P)}, \quad r_e^+ = \frac{(\Phi_P - \Phi_W)/(x_P - x_W)}{(\Phi_E - \Phi_P)/(x_E - x_P)} \tag{A.12}$$

$$r_w^- = \frac{(\Phi_E - \Phi_P)/(x_E - x_P)}{(\Phi_P - \Phi_W)/(x_P - x_W)}, \quad r_w^+ = \frac{(\Phi_W - \Phi_{WW})/(x_W - x_{WW})}{(\Phi_P - \Phi_W)/(x_P - x_W)} \tag{A.13}$$

$$r_n^- = \frac{(\Phi_{NN} - \Phi_N)/(x_{NN} - x_N)}{(\Phi_N - \Phi_P)/(x_N - x_P)}, \quad r_n^+ = \frac{(\Phi_P - \Phi_S)/(x_P - x_S)}{(\Phi_N - \Phi_P)/(x_N - x_P)} \tag{A.14}$$

$$r_s^- = \frac{(\Phi_N - \Phi_P)/(x_N - x_P)}{(\Phi_P - \Phi_S)/(x_P - x_S)}, \quad r_s^+ = \frac{(\Phi_S - \Phi_{SS})/(x_S - x_{SS})}{(\Phi_P - \Phi_S)/(x_P - x_S)} \tag{A.15}$$

$$r_t^- = \frac{(\Phi_{TT} - \Phi_T)/(x_{TT} - x_T)}{(\Phi_T - \Phi_P)/(x_T - x_P)}, \quad r_t^+ = \frac{(\Phi_P - \Phi_B)/(x_P - x_B)}{(\Phi_T - \Phi_P)/(x_T - x_P)} \tag{A.16}$$

$$r_b^- = \frac{(\Phi_T - \Phi_P)/(x_T - x_P)}{(\Phi_P - \Phi_B)/(x_P - x_B)}, \quad r_b^+ = \frac{(\Phi_B - \Phi_{BB})/(x_B - x_{BB})}{(\Phi_P - \Phi_B)/(x_P - x_B)} \tag{A.17}$$

$$\alpha_w = \begin{cases} 1, & F_w > 0 \\ 0, & \text{else} \end{cases} \tag{A.18}$$

$$\alpha_e = \begin{cases} 1, & F_e > 0 \\ 0, & \text{else} \end{cases} \tag{A.19}$$

$$\alpha_n = \begin{cases} 1, & F_n > 0 \\ 0, & \text{else} \end{cases} \tag{A.20}$$

$$\alpha_s = \begin{cases} 1, & F_s > 0 \\ 0, & \text{else} \end{cases} \tag{A.21}$$

$$\alpha_t = \begin{cases} 1, & F_t > 0 \\ 0, & \text{else} \end{cases} \quad (\text{A.22})$$

$$\alpha_b = \begin{cases} 1, & F_b > 0 \\ 0, & \text{else} \end{cases} \quad (\text{A.23})$$

where $\Psi(r)$ is the flux limiter function. Various flux limiters can be employed. The Sweby limiter is [198]

$$\Psi(r) = \max[0, \min(\beta r, 1), \min(\beta, r)] \quad (\text{A.24})$$

The Sweby limiter becomes the Minmod limiter if $\beta = 1$ and the Superbee limiter if $\beta = 2$ [199]. Among these limiters, Minmod is more diffusive. Superbee is less diffusive and works better to preserve sharp jump in the solution. Similar correction is employed for the convective terms in the momentum equations.