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Study of Two-fluid Flows in Microchannels

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Abstract

In the first part of this project, the stratified two-fluid flow in microchannel under pressure gradient and electroosmosis was studied experimentally and analytically. Experimentally, the interface location control between two fluids under electroosmosis effect was investigated using fluorescein imaging techniques. The micro particle image velocimetry (micro-PIV) was utilized to study the velocity profile of the two fluids quantitatively under different flow conditions, such as flow rate and applied electric field. Theoretically, the electrical potential distribution in the microchannel was solved. The fully developed velocity profile of the two fluids have also been obtained by solving the modified Navier-Stokes equation. Comparison between the experimental and theoretical analysis shows good agreement.

In the second part of this project, the characteristics of droplet flow in microfluidics was investigated. Two immiscible liquids, water and oil, are introduced to a T-shaped microchannel to form water droplet flow. The oil acts as carrier fluid. The velocity field inside the water droplet was studied using the micro-PIV technique. By subtracting the average velocity of the droplet, recirculation was seen in the droplet. However, the result shows the central part of the droplet moves slower than the outer part. The recirculation flow field can improve mixing inside the droplet. The mixing of two droplets during the coalescence was studied using fluorescence imaging technique.

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Contents

Abstract	ii
Acknowledgement	iii
Contents	iv
List of Figures	vii
List of Symbols	xi
1 Introduction	1
1.1 Background	1
1.2 Objectives and Scope	3
1.3 Organization	4
2 Literature Review	5
2.1 Fabrication of microfluidic devices	5
2.2 Two-fluid and multi-fluid flow in microfluidics	11
2.2.1 Mixing in microfluidics	11
2.2.2 Diffusion-based microfluidics	12
2.2.3 Droplets in microfluidics	15
2.3 Electrokinetics	18
2.3.1 Electric double layer	18

2.3.2	Electrokinetic phenomena	21
2.4	Experimental techniques for microfluidics	24
2.5	Remarks on previous works	26
3	Two-fluid stratified flow under pressure and electroosmosis: modelling	28
3.1	Electrical potential distribution in microchannel	29
3.2	The flow field of two-fluid flow	34
3.3	The effect of the electroosmosis	35
3.3.1	The non-conducting fluid flow	37
3.3.2	The conducting fluid flow	37
3.3.3	The formulation of $\Phi_j(Y)$	40
3.3.4	Volumetric flow rate due to electroosmosis	41
3.4	The effect of pressure gradient	42
3.4.1	Evaluation of $\Phi_{3j}(Y)$ and $\Phi_{4j}(Y)$	45
3.4.2	Volumetric flow rate due to pressure gradient	46
3.5	Results and discussion	47
3.5.1	Electric potential distribution	47
3.5.2	Effect of viscosity ratio, β , on the velocity profile	48
4	Two-fluid stratified flow under pressure and electroosmosis: experiment	52
4.1	Fabrication of the microchannel	52
4.2	Experimental setup	53
4.3	Results and discussion	56
4.3.1	Fluorescence imaging measurement	56
4.3.2	Micro-PIV measurement	62
4.4	Comparison of PIV measurement and theoretical model	67
5	Flow field inside water droplet: experiment	68
5.1	Experimental setup	68

5.1.1	Microfluidic device	68
5.1.2	Materials	69
5.2	Results and discussion	70
5.2.1	Size and shape of the droplets	70
5.2.2	Velocity field inside the droplets	73
5.2.3	Velocity field during droplet formation	77
5.3	Summary	79
6	Mixing of droplets	80
6.1	Concept	80
6.2	Experiment and results	80
7	Conclusion and Future Works	84
7.1	Conclusion	84
7.2	Future works	85
	Bibliography	86

List of Figures

2.1	Scheme for PDMS by soft-lithography fabrication.	7
2.2	A T-sensor.	13
2.3	An H-filter.	14
2.4	Basic concept for preparing double emulsions (W/ O/W) using T-shaped microchannels [52].	17
2.5	Microcapillary geometry for generating double emulsions from coaxial jets. (A) Schematic of the coaxial microcapillary fluidic device. (B-H)Various form of double emulsions by varying the diameters of both the outer and inner drops and the number of inner droplets [58].	17
2.6	Schematic representation of ion distribution near contact surface [2]. . .	19
2.7	Structure of EDL for contact between glass and aqueous solution. . . .	20
2.8	Schematic of electroosmotic flow in a cylindrical capillary.	20
3.1	Schematic of two fluids flow in microchannel and the coordinates used for mathematical forumulation.	28
3.2	Dimensionless electrical potential distribution at the symmetrical line for different values of K , ($K = 103$ for $10^{-6}M$, and $K = 1030$ for $10^{-4}M$), $\zeta_1=\zeta_2=-25$ mV, $\zeta_3=0$	48
3.3	Dimensionless velocity distribution for different viscosity ratio ($\zeta_1=\zeta_2=-25$ mV, $\zeta_3=0$), β : (a) $\beta = 1$, (b) $\beta = 10$, (c) $\beta = 100$, (d) $\beta = 1000$	49

3.4 Dimensionless velocity distribution at the symmetrical line for different viscosity ratio ($\zeta_1=\zeta_2=-25$ mV, $\zeta_3=0$), β : (a) $\beta = 1$, (b) $\beta = 10$, (c) $\beta = 100$, (d) $\beta = 1000$ 50

3.5 Dimensionless velocity distribution for $\beta = 10000$: (a) three-dimensional view, (b) at the symmetrical line. 51

4.1 PMMA plates and adhesive tape drawings for the fabrication: (a) top layer PMMA plate , (b) bottom layer PMMA plate, (c) the middle layer adhesive tape. 53

4.2 H-shaped microchannel: (a) the fabricated device used in the experiment, (b) schematic drawing of the channel configuration (the main channel is between E and F with a cross section of $910\mu\text{m} \times 100\mu\text{m}$), in which two fluids flow side by side (electric field from A to B: +ve, from B to A: -ve). 54

4.3 Schematic of the experimental setup for fluorescence imaging and micro-PIV measurements. 55

4.4 Measurement results at a flow rate of 0.6 mL/h: (a) no electric field, original image; (b) under -0.8 kV, original image; (c) the normalized light intensity image for 0 kV; (d) the normalized light intensity image for -0.8 kV; (e) normalized concentration distribution of the fluorescent dye across channel width under different applied voltages, from which NaCl solution holdup is about 35% for 0 kV applied voltage, 47% for -0.8 kV applied voltage. 57

4.5 Relationship between NaCl solution holdup and (a) different applied voltage for the same volumetric flow rates of the two fluids or (b) volumetric flow rate under the same applied voltage. (The viscosity ratio between the two fluids is $\beta = 1.5$) 59

4.6 The holdup of NaCl, e_1 , for different β value, (channel size = $910\mu\text{m} \times 50\mu\text{m}$) 60

4.7 The holdup of NaCl, e_1 , for different β value, (channel size = $910\mu\text{m} \times 100\mu\text{m}$) 61

4.8 Micro-PIV measurements for 0.1 ml/h flow rate under different electric fields, (a) in the absence of external electric field: (a1) PIV image, (a2) velocity vector; (b) -1.0 kV voltage electric field: (b1) PIV image, (b2) velocity vector; (c) the streamwise velocity profile over the channel width. 63

4.9 The velocity profile under different applied voltage, (flow rate= 0.1mL/h, the left side is NaCl). 65

4.10 Comparison of the dimensionless velocity for two different flow rate 0.08 mL/h and 0.15 mL/h, (a) no applied electric voltage, (b) -0.2 kV applied electric voltage, (c) -0.6 kV applied electric voltage, (d) -1.0 kV applied electric voltage. 66

4.11 Comparison between PIV measurements and theoretical analysis at flow rate 0.1 mL/h, under various applied electric voltage. (a) under no applied electric voltage, (b) under -0.2 kV applied electric voltage, (c) under 0.2 kV applied electric voltage, (d) under 0.4 kV applied electric voltage. 67

5.1 The silicon/glass chip and the adapter used in our experiment. The inserted figure shows the concept of droplet formation investigated in our experiments. 69

5.2 Original PIV images of the droplets at different flow rates (the droplets flow from right to left). (a) $55\mu\text{L/h}$, (b) $165\mu\text{L/h}$, (c) $220\mu\text{L/h}$, (d) $275\mu\text{L/h}$, (e) $330\mu\text{L/h}$, (f) $440\mu\text{L/h}$ 71

5.3 The droplet length at different flow rates. 72

5.4 Velocity field inside the droplets at different flow rates (the droplets flow from right to left). (a) 55 $\mu\text{L/h}$, (b) 165 $\mu\text{L/h}$, (c) 220 $\mu\text{L/h}$, (d) 275 $\mu\text{L/h}$, (e) 330 $\mu\text{L/h}$, (f) 440 $\mu\text{L/h}$ 74

5.5 The velocity profile at the middle line of the droplet. 75

5.6 Recirculation flow inside the droplets at different flow rates (the droplets flow from right to left). (a) 55 $\mu\text{L/h}$, (b) 165 $\mu\text{L/h}$, (c) 220 $\mu\text{L/h}$, (d) 275 $\mu\text{L/h}$, (e) 330 $\mu\text{L/h}$, (f) 440 $\mu\text{L/h}$ 76

5.7 Velocities of the droplet. The circles $\circ$ are the mean velocity of the droplet evaluated from the micro-PIV measurement. The dashed line $--$ depicts the averaged flow velocity in the microchannel calculated based on the total flow rate. The crosses $\times$ are measured Lagrangian velocity at the interface of the droplet. 77

5.8 The velocity fields inside the water droplet during the droplet formation stage, (a) at $t=0$ second, (b) at $t=0.1$ second, (c) at $t=0.2$ second, (d) at $t=0.3$ second, (e) at $t=0.4$ second, (f) at $t=0.5$ second. 78

6.1 Schematic of the microfluidic device for studying the coalesce and mixing of two droplets. 81

6.2 The fabricated device used in our experiment. 81

6.3 Concentration field of the dye during the coalescence of two droplets (channel size = $100\mu\text{m} \times 600\mu\text{m}$), total flow rate = 330 $\mu\text{L/h}$, flow rate ratio = 20:1:1. 83

List of Symbols

Greek symbols

- β dynamic viscosity ratio
- $1/\kappa$ characteristic thickness of the electrical double layer
- μ_1 viscosity of fluid 1
- μ_2 viscosity of fluid 2
- μ_{ref} reference viscosity
- $\Phi_j(y)$ a parametric function,, see equation (3.32), page 33
- Ψ non-dimensional electric potential
- ψ zeta potential
- ρ_1 density of fluid 1
- ρ_2 density of fluid 2
- ρ_e volume charge density
- ρ_{ref} reference density
- τ tangential direction along the fluid-fluid interface
- ϵ_0 permittivity of the vacuum

ϵ_r	dimensionless dielectric constant
ζ_1	zeta potential at the side wall
$\bar{\zeta}_1$	non-dimensional zeta potential at the side wall
ζ_2	zeta potential at the top wall
$\bar{\zeta}_2$	non-dimensional zeta potential at the top wall
ζ_3	zeta potential at the fluid-fluid interface
$\bar{\zeta}_3$	non-dimensional zeta potential at the fluids interface

Roman letters

u_1^*	velocity of fluid 1
u_2^*	velocity of fluid 2
$b1_j, b2_j$	coefficients function , page 34
$b3_j, b4_j$	coefficients,, page 40
D_h	hydraulic diameter
e	elementary charge
E_x	external electric field along x direction
$F_j(y)$	a parametric function , see equation (3.33), page 33
h	half channel height
$I_j^\Phi(Y)$	a parametric function , page 37
K	electrokinetic diameter
k_b	elementary charge

n_{i0} bulk concentration

$(\frac{dP}{dX})_1, (\frac{dP}{dX})_2$ dimensionless pressure gradient for fluid 1 and fluid 2, respectively

$(\frac{dp}{dx})_1, (\frac{dp}{dx})_2$ pressure gradient for fluid 1 and fluid 2, respectively

Q_1, Q_2 dimensionless volumetric flow rate of fluid 1 and fluid 2 electroosmosis

q_1, q_2 volumetric flow rate of fluid 1 and fluid 2 due to electroosmosis

Q_3, Q_4 dimensionless volumetric flow rate of fluid 1 and fluid 2 due to pressure gradient

q_3, q_4 volumetric flow rate of fluid 1 and fluid 2 due to pressure gradient

Re_0 reference Reynolds number

T absolute temperature

U_1 non-dimensional velocity of fluid 1 due to electroosmosis

U_2 non-dimensional velocity of fluid 2 due to electroosmosis

U_3 dimensionless velocity of fluid 1 due to pressure effect

U_4 dimensionless velocity of fluid 2 due to pressure effect

U_{ref} reference velocity

w_1 width occupied by fluid 1

w_2 width occupied by fluid 2

X, Y, Z non-dimensional coordinate

x, y, z local coordinate

z_i valence of type-i ions

Chapter 1

Introduction

1.1 Background

Micro-electromechanical Systems or MEMS are miniaturized electromechanical devices and systems that are realized using integrated micro fabrication methods. MEMS devices offer the advantages of micro/nano-scale sizes and increased degree of electromechanical functional integration. As a breakthrough technology, it allows unparalleled synergy between previously unrelated fields such as biology and mechanics.

MEMS can range from simple mechanical structures to completely miniaturized fluidic multi-functional systems containing actuators, sensors and electronics. MEMS-based components are used in many applications, such as medical, automotive, telecommunication, IT and space. Typical mass-produced examples are: accelerometers for airbag systems, heads for ink jet printers and hard disk drivers, pressure and chemical sensors for medical applications and quartz watch crystals resonators for time-keeping.

MEMS technology is currently enjoying a moment of formidable expansion in synergy with the health sciences, giving rise to the notion of MEMS for biomedical applications, known as **BioMEMS** or **lab-on-a-chip**. One of the examples is the Micro Total Analysis Systems (μ -TAS). Ideally, a Total Analysis Systems (TAS) performs all the component stages of a complete chemical analysis in an integrated and automated fashion. These stage can include sampling, sample pre-treatment, chemical reactions,

Chapter 1 Introduction

analytical separations, analyte detection, product isolation and data analysis. There exist significant drawbacks of the conventional TAS, such as slow sample transport, high reagent consumption, poor separation efficiencies and the need of to fabricate interfaces between distinct components.

Improvements can be achieved by minimizing the scale on which the analysis is performed. The "downsized" TAS, μ -TAS, was suggested by Manz *et al* [1] in 1990. Essentially, μ -TAS is a device that improves the performance of an analysis by reduced size. Typically, the μ -TAS is a micro-chip, biochip or lab-on-a-chip. Besides the reduced reagents consumption, greater separation efficiencies, and reduced analysis time, the advantages of improved analytical performance, component integration are much more important.

To manipulate biological reagents, usually in the fluid sample form, on the micron-sized chip, microfluidics plays an important role. Microfluidics refers to devices and methods for controlling and manipulating fluid flows in flow length scales less than a millimeter. As the geometry shrinks, the volume to surface ratio increases dramatically, many other forces which are not significant on macroscale now play important parts in the microfluidics. These includes, capillary effects, surface tension, and electrokinetics. Other characteristic associated with microfluidics is the low Reynolds number, usually in the order of 0.01 to 100. The extreme low Reynolds number assures the laminar flow. Diffusion is thus the only mechanism through which transport occurs between phases. The laminar flows of several streams allow separation or extraction from one phase to another phase.

Most solid surfaces acquire a certain amount of electrostatic charges when they are in contact with polar liquids, and hence a difference in potential is developed across the interface between the two phases. The charged interface attracts ions of opposite charge (counter-ion) and repels ions of like charge (co-ions) in the liquid. The arrangement of charges that occurs near the interface leads to the development of electric double layer

Chapter 1 Introduction

(EDL) [2]. Electroosmosis refers to the fluid movement relative to a stationary solid wall under an externally applied electric field. Electrokinetics provides another driving force to control the fluid flow in microfluidics.

1.2 Objectives and Scope

The project aims to study the characteristics of two-fluid flows in microfluidics. In the first part, the stratified two-fluid flow in microchannel under pressure gradient and electroosmosis effects is studied. The objectives are:

- To study and characterize the interface location between two fluids under the pressure and electroosmosis
- To investigate the fully developed velocity profile of the two fluids inside the microchannel using micro-PIV technique
- To develop a mathematical model of two fluids flow inside microchannel

In the second part of the project, two immiscible fluids are brought together to form droplet flow. The flow field inside a water droplet is investigated. The objectives are:

- To study the velocity field inside a moving water droplet using micro-PIV technique
- To study the mixing of two water droplets during the coalescence

1.3 Organization

In Chapter 2, the literature of microfluidics fabrication, experimental techniques and characteristics, electrokinetics theory is reviewed. In Chapter 3, the mathematical formulation of the two-fluid flow in microchannel with electroosmosis and pressure gradient effect is presented. The experimental study utilizing fluorescein imaging and micro-PIV is given in Chapter 4. In Chapter 5, the flow field inside a moving droplet in microchannel is investigated using micro-PIV technique. The experimental measurement of mixing of droplet coalescence is presented in Chapter 6. In Chapter 7, the project is concluded and the future works are recommended.

Chapter 2

Literature Review

This chapter gives a literature survey, including fabrication, experimental techniques and various works of two-fluid and multi-fluid flows in microfluidics. Background about electrokinetics and electrokinetics in microfluidics are also reviewed in the later section of this chapter.

2.1 Fabrication of microfluidic devices

As the microfluidics develop rapidly in recent years, microfluidic devices can be fabricated from a variety of materials. Based on fabrication techniques developed in the microelectronics industry, silicon has been used extensively to create microfluidic devices. The microelectronics fabrication techniques based on silicon material are the most promising method to fabricate microfluidic devices. This method has been dominant since the introduction of "lab-on-a-chip" since 1990s, primarily driven by the fact that fabrication methods were well established by the semiconductor industry, and surface properties and derivatization methods were well characterized and developed by the chromatography industry among others. The general procedures of silicon microfabrication techniques can be found in various literature [3].

Wong *et al* [4] fabricated the micro T-mixers out of a silicon substrate and bonded to a Pyrex glass plate to study the mixing performances. Electrokinetic micromixers fabricated from the silicon were investigated by Lee *et al* [5], Biddiss *et al* [6] and Wu

Chapter 2 Literature Review

et al [7]. The mature microelectronics fabrication techniques allow the modification of the wall surface of the microchannel. The basic characteristic of the electrokinetic micromixer is the zeta potential variation of the channel wall surface. Externally electric field is usually applied to establish localized zeta potential variation near the electric double layer. Deval *et al* [8] demonstrated the reconfigurable hydrophobic-hydrophilic surfaces by reversibly adsorbing and removing avidin from a hydrophobic self-assembled monolayer in micro-machined silicon microchannel. The microstructures used by Dreyfus *et al* [9] and Thorsen *et al* [10] to in their studies of two-phase flow were all fabricated out from silicon.

Complex systems can be manufactured out of silicon. However, silicon is often not the ideal material for microfluidic applications due to optical opacity, cost, difficulty in component integration, and surface characteristics that are not well suited to biological applications. The needs of many microfluidic applications do not require the precision that silicon micromachining can offer. In addition, silicon micromachining techniques are costly, labor intensive, and require highly specialized skills, equipment, and facilities. Silicon and glass-based microfluidic devices are, however, well suited to some chemistry applications that require strong solvents, high temperatures, or chemically stable surfaces cite.

The cost of producing microfluidic devices in silicon, glass is high. Commercial manufacturers of microfluidic devices see many benefits in employing plastics that include reduced cost and simplified manufacturing procedures, particularly when compared to glass and silicon. Another advantage of the plastic materials is the wide range of plastics available, which allows the manufacturer to choose materials properties suitable for their specific application. In addition, it is much faster to build prototype based on the plastic material.

One of plastic fabrication methods, which is getting more attention, is the polydimethylsiloxane (PDMS) by soft-lithography. Fabrication of microfluidic devices in

Chapter 2 Literature Review

Figure 2.1: Scheme for PDMS by soft-lithography fabrication.

PDMS by soft-lithography provides faster, less expensive routes than these conventional methods. These soft-lithographic methods are based on rapid prototyping and replica molding and are more accessible to chemists and biologists working under benchtop conditions than the microelectronics-derived methods because, in soft lithography, devices do not need to be fabricated in a cleanroom [11, 12].

The method of fabrication of PDMS by soft-lithography can be described as the following, shown in Figure 2.1: A liquid pre-polymer is poured onto a master and after curing the PDMS layer can be peeled off the master. The low surface free energy and elasticity of PDMS allow it to release from masters without damaging the master or itself. The master can be made from a variety of method, from standard microelectronics

Chapter 2 Literature Review

fabrication techniques. A common method is through photolithography: After designing the microstructures, the CAD patterns are printed on transparencies. Lateral resolutions of $25\text{ }\mu\text{m}$ can be routinely achieved with image setters operating at 5080 dpi. They can be expanded to $8\text{ }\mu\text{m}$ using photoplotters operating at 20000 dpi. The transparencies are then used as photomasks in UV lithography and structures are transferred into SU-8 resist which serves as a master for fabricating PDMS moulds [13].

The use of PDMS as a material for microfluidic applications originates mostly from the work of George Whitesides' research group of Harvard university [14, 15]. It was rapidly used by other researchers. Ichikawa *et al* [16] studied the interface motion driven by capillary action in a rectangular microchannel. Rectangular microchannels with several sizes (about 50 to $100\text{ }\mu\text{m}$ square) of glass capillaries and $85 \times 68\text{ }\mu\text{m}$ and $75 \times 45\text{ }\mu\text{m}$ of PDMS microchannels were fabricated by photolithography technique for the experiment. Kim *et al* [17] in their study of two fluid flow field measurement in microchannel used PDMS microchannel to carry out the experiment. A T-shaped microchannel was fabricated to investigate the convective mixing in microchannel by Sato *et al* [18].

Laser ablation is another techniques for plastic microfluidics fabrication. Local temperatures on the polymer surface during ablation can be very high during the ablation process. In this high-energy process, a shock wave is produced and particles are ejected from the substrate thus creating a void. Small particulates can be seen on the surface of the substrate material after laser ablation. Micromachining using laser ablation may be achieved by exposing the polymer substrate to the laser through a mask that defines the area to be ablated. In this case, the mask is made from a material that does not have significant absorption at the laser wavelength, e.g. some metals. Alternatively, channels and other structures can be defined and micromachined using a direct-write, maskless process. In this process, the polymer substrate material is placed on a moveable (preferably programmable) stage, and the substrate is moved under the focused laser beam to create the desired structure. The direct-write micromachining process is advantageous in

Chapter 2 Literature Review

that a mask does not have to be created to change the design of the microchannel network; therefore, channel design can be changed rapidly during the prototyping process. The disadvantage to this approach is that parts are made in a sequential manner thereby limiting the ability to mass produce devices for commercial applications.

Roberts *et al* [19] reported a UV laser photoablation method for the production of miniaturized liquid-handling systems on polymer substrate chips. The fabrication of fluid channel and reservoir networks was accomplished by firing 200 mJ pulses from an UV excimer laser at substrates moving in predefined computer-controlled patterns. This method was used for producing channels in polystyrene, polycarbonate, cellulose acetate, and poly(ethylene terephthalate). The photoablated surfaces showed an increase in their hydrophilicity and rugosity as a group compared to the original polymer samples.

Johanson *et al* [20] reported on the design of a new type of passive micromixing device. A preformed T-microchannel imprinted in polycarbonate was later modified with a pulsed UV excimer laser (KrF, 248 nm) to create a series of slanted wells at the junction cite. The presence of the wells leads to a high degree of lateral transport within the channel and rapid mixing of two confluent streams undergoing electroosmotic flow. Several mixer designs were fabricated and investigated. All designs were relatively successful at low flow rates (0.06 cm/s, greater than or equal to 75% mixing), but had varying degrees of success at high flow rates (0.81 cm/s, 45-80% mixing).

A CO₂-laser system was used to direct-write on plastic substrates for rapidly producing polymer microfluidic structures by the research group from Denmark [21, 22]. The depth and width of laser-cut channels depends on the laser beam power and on the number of passes of the beam along the same channel. In their experiments the laser beam power was varied between 0 and 40 W and the passes were varied in the range of 1 to 7 times. Typical channel depths were obtained between 100 and 300 μm , while the channels were typically 250 μm wide. Several bonding methods for microstructured

Chapter 2 Literature Review

PMMA poly(methyl methacrylate) parts were investigated, such as solvent-assisted gluing, melting, laminating and surface activation using a plasma asher. A solvent-assisted thermal bonding method proved to be the most time-efficient one. Using laser micro-machining together with bonding, a three-layer polymer microstructure with included optical fibers was fabricated within two days. These systems are especially useful in microfluidic prototyping due to the very short cycle time of production. Using the same system and techniques, they developed an integrated polymer microfluidic manifold for the determination of phosphorus [23]. The fabrication parameters, the cutting sequence, the number of cut passes, the laser beam velocity and the laser radiant flux, which affect the channel depth of microchannels were examined [24]. The laser system has proven to be a flexible and rapid tool for the production of polymer microfluidic prototypes.

Another low-cost rapid prototyping technique is the plastic lamination [25]. The basic idea underneath the lamination technique is: microfluidics network or structure can be built of several layers of simple fluidic circuit. Each layer of fluidic circuit can be cut from the CAD drawing easily using CO₂ laser system. For a single channel structure, 3 layers, top layer, bottom layer and a middle fluidic channel layer are needed. If more layers are laminated complex structure could be possible [26].

Wu *et al* [27] studied the nonlinear diffusive mixing in microchannels theoretically and experimentally. In their experiment, the microchannel was built from two layers of PMMA plates (75mm × 25 mm) and a layer of double sided adhesive tape. The channel structure was cut on the adhesive tape using a CO₂ laser system. Afterwards, the PMMA plates were bonded as a cover and a bottom through the adhesive tape, while the cut-through channel of the tape serves as a microfluidic channel. A mixing channel with 850 μm width and 50 μm height was fabricated for the experiments.

Other techniques to make plastic microfluidic device include: hot embossing [28] or imprinting technique [29], and injection molding [30–32] techniques.

2.2 Two-fluid and multi-fluid flow in microfluidics

The Reynolds number (Re) of a fluid flow describes its flow regime: laminar or turbulent. The Reynolds number can be calculated by $Re = \frac{\rho u D_h}{\mu}$, where ρ is the fluid density, u is the characteristic velocity of the fluid, μ is the fluid viscosity, and D_h is the hydraulic diameter. The hydraulic diameter is a computed value that depends on the channel's cross-sectional geometry.

$Re < 2300$, as calculated by the above formula, generally indicates a laminar flow. As Re approaches 2300, the fluid begins to show signs of turbulence, and as Re greater than 2300 the flow is considered to be turbulent.

In microfluidics, due to the small geometry of the microstructure, the Reynolds numbers are usually smaller than 100. Here is an example from textbook [33]: for a microchannel with hydraulic diameter $D_h=100 \mu\text{m}$, average flow velocity $u=1 \text{ cm s}^{-1}$, the calculated Reynolds number is about 1.

2.2.1 Mixing in microfluidics

If two or more streams of fluids, which are miscible to each other, flow inside a microchannel, the flows are in the laminar flow regime. One consequence of laminar flow is that two or more streams flowing in contact with each other will not mix except by diffusion. Mixing is often crucial to the effective functioning of microfluidic devices [34]. Often the objective is rapid mixing between two initially segregated streams in a minimal amount of space. An example involves interspersing two streams (as in the rapid mixing of macromolecular solutions for chip-based molecular diagnostics) as in the case of biochemical sensing techniques such as immunoassays and hybridization analysis, which require the rapid, homogeneous mixing of macromolecular solutions, such as DNA or globular proteins. One has to find suitable ways to mix fluid streams in small-scale geometries in acceptable time-scales [35].

Chapter 2 Literature Review

Mixing between the adjacent laminar streams in a straight, smooth-walled microchannel occurs only by diffusion. The diffusion through the phases is a function of diffusion coefficient and distance [36]. The mixing time is proportional to the square of the distance. There are two general strategies for generating such transverse flows [37]: (a) passive methods in which transverse flows result from the interaction of the externally driven flow (e.g., pressure-driven or electro-osmotic) with the fixed channel geometry, and (b) active methods in which transverse flows are generated by oscillatory forcing (mechanical or electrical) within the channel.

The challenge in designing passive micromixers is to generate laminar chaos in an accessible geometry. A simple design that satisfies this criterion uses obliquely oriented grooves on one wall of the channel to generate transverse components in steady flows [20, 38]. Burghilea *et al* [39] showed that chaotic flow can be generated in a smooth microchannel of a uniform width at arbitrarily low Re , if a small amount of flexible polymers is added to the working liquid. Chang *et al* [40] presented a numerical investigation of electrokinetically driven flow mixing in microchannels with various numbers of incorporated patterned rectangular blocks. Several pieces of work used electrokinetic flow to enhance mixing [5–7].

2.2.2 Diffusion-based microfluidics

At other times, the objective is to prevent mixing and maintain segregation, for example, having two streams co-flowing side by side and controlling or monitoring processes occurring at the interface between the two fluids. Because only diffusion occurs between different streams of flow, it is used for extraction or separation in the biological analysis [25, 41].

Diffusion-based microfluidic devices, such as the T-sensor and the H-filter have been developed for commercial use by Micronics, Inc [42]. The early work of diffusion-based microfluidic originates from the research group led by Paul Yager in University of

Chapter 2 Literature Review

Washington [43–45].

Figure 2.2: A T-sensor.

T-Sensor in its simplest embodiment has two input ports and one outlet port, allowing two fluids to be brought into contact such that they flow side by side, as shown in Figure 2.2¹. Two fluids enter through input channels, merging at the stagnation point (sp). In the Figure 2.2, the fluid on the right contains a diffusible analyte (gray) that spreads across the D -dimension as flow proceeds along the channel length. Typical measurements are made by fluorescence detection along the optical axis, denoted with large arrows in the y -direction. Because the Reynolds number is small, usually much less than 1, the flow of the two streams is completely laminar and no convective mixing between the streams occurs. Thus, the only means by which molecules in opposite streams can mix is by molecular diffusion across the interface of the two-fluid streams. The chemical binding or other reaction events than occur along this centerline produce a measurable signal, usually fluorescence, which can be used to calculate a parameter of interest for the analyte, such as concentration or diffusion coefficient.

The principle of the H-Filter platform relies on the absence of turbulent mixing

¹This figure is from <http://faculty.washington.edu/yagerp/microfluidicstutorial/tsensor/tsensor.htm>

Chapter 2 Literature Review

within a microfluidic channel. Two flow streams can be brought together and run side by side in the center channel of the H-filter with laminar flow characteristics producing absolutely no turbulent mixing. Because there is no mixing, the only transport of molecules from one flow stream to the other is by diffusion. The major factors affecting the rate of diffusion are (a) the size and shape of the particles; (b) the viscosity of the solution; and (c) the temperature of the medium. In the Figure 2.3², small molecules will diffuse quickly from a sample stream to the buffer stream, while very large molecules and particles will remain indefinitely in the sample stream because of their much larger size and much decreased diffusion rate.

Figure 2.3: An H-filter.

Hatch *et al* [44] have developed a rapid diffusion immunoassay that allows measurement of small molecules down to sub-nano-molar concentrations in less than 1 minute based on the T-sensor. This competitive assay is based on measuring the distribution of a labelled probe molecule after it diffuses for a short time from one region into another region containing antigen-specific antibodies.

The pressure-driven velocity profile in the duct-shaped device influences diffusive transport in ways that affect the use of the T-sensor to measure molecular properties. Kamholz [46] studied the time-dependent evolution of analyte distribution using a combination of one- and two-dimensional models.

²This figure is from <http://www.micronics.net/products/access.php>

Chapter 2 Literature Review

Kamholz *et al* [47] presented a theoretical analysis of the scaling laws for analyte diffusion in a microfluidic chemical analysis device, the T-sensor. Because the flow is pressure-driven, the velocity profile is non-uniform, inducing a distribution in residence time among analyte molecules. They compared the two-dimensional model with a simple one dimensional model. Experimentally, demonstrated the rapid measurement of diffusion coefficients of large and small molecules in the T-sensor, by means of conventional epifluorescence microscopy. Diffusion coefficients were measured for four fluorescently labeled molecules: fluorescein-biotin, insulin, ovalbumin, and streptavidin. The resulting values differed from accepted results by an average of 2.4%. Other similar work includes: integration of a neutral ionophore-based ion pair extraction reaction onto a glass microchip [41] and microscale continuous ion exchanger [48].

2.2.3 Droplets in microfluidics

Monodisperse droplets were traditionally used in the fields of food science, cosmetics, and pharmaceuticals. Recently, The formation of micro-droplets in microchannels attracts the interest of microfluidics and lab-on-a-chip community [49]. The main application of this phenomenon is micro-reaction technology [50]. The water droplets serve as well confined micro-reactors, in which chemical reaction can take place; while the inert carrier fluid can serve as a transport and a protection medium. Micro-droplets have been used for DNA analysis, protein crystallization [51], analysis of human physiological fluids, encapsulation [52], and production of polymeric micro beads [53].

Most of the recently reported works are based on droplets formed between immiscible fluids. Formation of droplets can be prepared using a simple T-junction [10] or a cross junction [9]. In the paper by Thorsen *et al* [10], they performed a study in a system using tetradecane and water as the two working immiscible fluids. Their experiments showed the existence of complex patterns. Their aim was to generate liquid droplets from liquid, whose size is controlled accurately. They showed that instability

Chapter 2 Literature Review

can develop as a competition between shear forces and surface tension in a microfluidic device. It was shown that self-assembly of the vesicles into patterns depends on channel geometry and relative fluid pressure, enabling the production of motifs ranging from monodisperse droplets to helices and ribbons. The microchannels have a cross configuration in the experiment by Dreyfus *et al* [9], were etched in glass, and covered by a silicon wafer, using anodic bonding. They used tetradecane and water as the two immiscible fluids. Their study revealed the existence of a rich variety of patterns, depending on the flow-rates, and the injection configuration.

Song *et al* [54] experimentally studied the scaling of chaotic mixing in a droplet moving through a winding microfluidic channel. A T-junction microchannel was used. Two aqueous streams of different colors were introduced through the side channel. The carrier fluid broke them into droplets. While these droplets were moving through the microchannel, the chaotic mixing occurred because of the stretching and folding inside the droplets. Ismagilov's group extended this concept for the application of functional and structural characterization of proteins [50, 51, 55–57]. These applications include: droplet-based microfluidic system for evaluation protein crystallization conditions with on-chip x-ray diffraction [51], millisecond kinetics on a microfluidic chip using nanoliters of reagents [57], dynamics of drosophila embryonic patterning network perturbed in space and time with microfluidics [55].

A double emulsion (also referred to as a multiple emulsion) can be defined as a multiple-phase dispersion in which droplets enclosing finer droplets are suspended in a continuous liquid phase. Okushima *et al* [52] demonstrated the production of double emulsions using microfluidic devices having a hydrophobic junction and a hydrophilic junction positioned serially. In their study, the monodisperse double emulsions were prepared using a two-step method of droplet formation in microchannel networks. The microchannel networks consists two T-junctions, Figure 2.4. For a Water in Oil in Water (W/O/W) emulsion, the aqueous drops to be enclosed are formed periodically upstream

Chapter 2 Literature Review

at the hydrophobic T-junction; then, in a continuing series, organic droplets enclosing the aqueous droplets are formed downstream at the hydrophilic T-junction. In this technique, the wetting of the channel has a significant effect on the type of dispersion that can be prepared. Channels fabricated on hydrophobic material are suitable for dispersing water drops in an organic phase, whereas hydrophilic channels are suited to oil-in-water dispersions.

Figure 2.4: Basic concept for preparing double emulsions (W/ O/W) using T-shaped microchannels [52].

Figure 2.5: Microcapillary geometry for generating double emulsions from coaxial jets. (A) Schematic of the coaxial microcapillary fluidic device. (B-H) Various form of double emulsions by varying the diameters of both the outer and inner drops and the number of inner droplets [58].

Recently, Utada *et al* [58] report another method to produce monodisperse double emulsions using microcapillary device (Figure 2.5). They described a fluidic device that

Chapter 2 Literature Review

generates double emulsions in a single step, allowing precision control of the outer and inner drop sizes as well as the number of droplets encapsulated in each larger drop. The device consists of cylindrical glass capillary tubes nested within a square glass tube. The innermost fluid is pumped through a tapered cylindrical capillary tube, and the middle fluid is pumped through the outer coaxial region, which forms a coaxial flow at the exit of the tapered tube. The outermost fluid is pumped through the outer coaxial region from the opposite direction, and all fluids are forced through the exit orifice formed by the remaining inner tube. A variety of double emulsions can be produced by varying the flow rates of the fluids, the geometry of the microfluidics devices. They demonstrated that the microfluidic device can produce uniform double emulsions, in which each drop contains a single internal droplet, or emulsions with multiple internal droplets.

2.3 Electrokinetics

2.3.1 Electric double layer

The electrical double layer model was first introduced by Stern in 1924 by combining the Helmholtz and the Gouy-Chapman models [2, 59]. Generally, most substances acquire a surface electric charge when brought into contact with an aqueous (polar) medium. Some of the charging mechanisms include ionization, ion adsorption, and ion dissolution. The effect of any charged surface in an electrolyte solution will influence the distribution of nearby ions in the solution, as shown in Figure 2.6. Ions of opposite charge (counterions) to that of the surface are attracted toward the surface while ions of like charges (co-ions) are repelled from the surface. Because of the electrostatic interaction, the counter-ions concentration near the charged surface is higher than that in the bulk liquid far away from the charged surface. This attraction and repulsion and the subsequent rearrangement of the charges on the charged surface and the balancing charges in the liquid is called the electrical double layer (EDL). The electrical double

Chapter 2 Literature Review

Figure 2.6: Schematic representation of ion distribution near contact surface [2].

layer (EDL) is a region close to the charged surface where counterions and coions in the aqueous (polar) medium are preferentially distributed. It is composed of the compact layer (stern layer) and the diffuse double layer. The compact layer is usually about 0.5 nm thick. In the compact layer, the ions are strongly attracted to the wall surface and are immobile. In the diffuse double layer, the ions are less affected by the electrical field and are mobile. They spread out as a result of thermal energy. The thickness of the diffuse EDL generally ranges from several to a few hundreds of nanometers, depending upon the bulk ionic concentration and other properties of liquid.

The plane that divides the EDL into two layers is called Stern plane. The potential at this plane is close to the electrokinetic potential or zeta (ζ) potential. The zeta potential is defined as the potential at the shear surface between the charged surface and the electrolyte solution. The shear surface, also known as shear plane, is an imaginary surface, which is considered to lie close to the solid surface and within which the fluid is stationary. It can be characterized as the plane at which the mobile portion of the diffuse layer can ‘slip’ or flow past the charged surface. In the case of contact between glass (or silica) and an aqueous electrolyte liquid, the schematic ions distribution and shear plane

Chapter 2 Literature Review

Figure 2.7: Schematic of the structure of the EDL: Glass acquires a negative surface charge upon contact with an aqueous solution. The surface charge attracts dissolved counter-ions and repels co-ions, resulting in a charge separation. The zeta potential(ζ) is the potential at the shear plane and the Debye length (λ_D) is the characteristic thickness of the double layer. A layer of immobile positive ions immediately adjacent to the wall is called the Stern layer [60].

Figure 2.8: Schematic of electroosmotic flow in a cylindrical capillary.

is shown in Figure 2.7 [60].

Within the diffuse layer, the net charge density, ρ_e is not zero. The net charge density influences the electrical potential distribution at the EDL, particularly at the diffuse layer. If an electric field is applied tangentially along a charged surface then the electric field will exert a body force on the charge in the diffuse layer. This movement of ions, under the influence of applied electrical field, will pull the surrounding fluid with them, resulting in electroosmotic flow, as shown in Figure 2.8³. Thus, solution injection, mixing or displacing can be achieved by applying an applied electrical field.

³This figure is from http://www.stanford.edu/~dlaser/electrokinetics_and_eof/electrokinetics_and_eof.htm

Chapter 2 Literature Review

2.3.2 Electrokinetic phenomena

The electrokinetic effects can be classified into four basic types, namely electrophoresis, electroosmosis, streaming potential and sedimentation potential [2]. Electrophoresis and sedimentation potential involve the motion of charged particles in quiescent liquid. Electro-osmosis and streaming potential involve the flow of fluid past a stationary charged surface. These effects can also be classified according to the driving force and response. An electric field is applied in electrophoresis and electro-osmosis, thus particle or fluid flow is generated. An external pressure gradient is imposed, in sedimentation potential or streaming potential experiments, to generate an electrical potential or current.

If one phase consists of a liquid or gas in which the second phase is suspended as particles of solids or liquids, the particles can be induced to move by applying an electric field across the system. The movement of charged colloidal particles in an electric field is termed electrophoresis. Forced movement of charged solid particles in a liquid, e.g., due to gravitation induces a difference of electric potentials, the sedimentation potential. When the solid remains stationary and the liquid moves in response to an applied electric field, usually, this occurs when the solid is in the form of capillary or porous plug. The movement of the liquid caused by the electric field is called electroosmosis. The liquid can be forced through a capillary or porous plug under a pressure gradient. The excess charges near the wall are carried along by the liquid and their accumulation downstream causes the buildup of an electric field which drives an electrical current against the direction of the liquid flow. A steady state is quickly established. The potential difference is called the streaming potential. It is related to the driving pressure and to the potential in the neighborhood of the wall.

In studying electrokinetic phenomena, one of the most important characterization parameters is zeta potential, ζ , which is the potential at the electrokinetic shear plane. In particular, it is a powerful concept to relate the macroscopic measurable quantities

Chapter 2 Literature Review

to the microscopic parameters characterizing this state. This potential, however, cannot be measured directly. In many practical situations, it is difficult to obtain a reliable estimate of ζ . An alternative strategy is to interpret zeta potential from electrokinetic measurements [2, 61].

Arulanandam and Li [62] provided an experimental procedure for determining zeta potential and surface conductance by monitoring the electrical current in electroosmotic flow through a capillary. A known method of measuring the average velocity based on current monitoring [63] in electroosmotic flow was used by Arulanandam to evaluate ζ and surface conductance. By applying the Debye-Hückel approximation where the value of electrical potential, ζ , is assumed to be small $\zeta < 25mV$, the zeta potential can be expressed as an explicit function of the average velocity.

An improved method based on current monitoring [62, 63] was employed by Erickson [64] to determine the zeta potential and surface conductance λ_s by employing the slope-intercept method. Previously, these two parameters were obtained by monitoring the electrical current in electroosmotic flow through a capillary.

Numerous pieces of research have been done on the electrokinetic effects, analytically, numerically or experimentally during the past few years. The research involved investigations of viscosity effects, heat transfer effects, velocity characteristics, zeta potential and etc.

Yang *et al* [65] investigated the electrokinetic effects on pressure-driven liquid flows in rectangular microchannels. The two-dimensional nonlinear Poisson-Boltzmann equation governing the EDL is numerically solved with the use of a finite-difference scheme. In the equation of motion, an exact solution is obtained by employing the Green function formulation. Qu *et al* [66] proposed a model to determine the electrical potential and ionic concentration distributions in an overlapped EDL field. Two infinitely large plates were considered. It was found out that using the classical theory to evaluate the potential distribution in an overlapped EDL region will lead to an inaccurate

Chapter 2 Literature Review

description due to the misuse of the Boltzmann's equation and the boundary conditions that are being defined. Hence they derived new governing equations for the overlapped EDL fields without using the Boltzmann's equation. The results show that the classical theory predicts a higher electrical potential in the EDL field than the modified model.

Experimentally, Werner *et al* [67] developed an experimental setup to study the electrokinetic potential and the surface conductivity of flat surfaces. The device accommodates the variability of the distances between two parallel flat sample surfaces forming a slit channel. Different aqueous electrolyte solutions were used. The zeta potential was calculated at different channel geometries and the surface conductivity was determined from the extrapolation of the channel conductance values gained at a number of sufficiently small distances. This experiment setup also allows the hydrodynamic thickness to be determined within the adsorption layers.

Patankar *et al* [68] developed a numerical scheme to simulate electroosmotic flows in complicated geometries. In the work, they studied the electroosmotic injection characteristics of a cross-channel device for capillary electrophoresis. They found that the desired rectangular shape of the sample plug at the intersection of the cross-channel could be obtained when the injection was carried out at high electric field intensities. The shape of the sample plug can also be controlled by applying an electric potential or a pressure at the side reservoirs. Flow induced from the side channels into the injection channel squeezes the streamlines at the intersection, thus giving a less distorted sample plug.

Hu *et al* [69] presented a numerical study for steady electrokinetic flow in intersecting channels in a T-shaped configuration. The nonlinear Poisson-Boltzmann equation was solved numerically by employing the finite difference method. Thereafter, the modified Navier-Stokes and the continuity equation was solved numerically by employing the SIMPLER algorithm. The results showed that the hydrodynamic effect affects the liquid leakage from the floating side channel significantly. The magnitude of this

Chapter 2 Literature Review

leakage depends on two factors, the length and the location of the channel. The longer the side channel is, the less the fluid leaks out of it. The amount of leakage decreased when the side channel was moved downstream from the entrance to the outlet of the main channel. The velocity profile in the floating side channel is shown to be parabolic as the driving force is due to pressure only. However in the active channels, the velocity profile is a combination of the electroosmotic flow and Poiseuille flow.

Arulanandam *et al* [70] investigated characteristics of electroosmotic flow in rectangular microchannels. A two dimensional Poisson-Boltzmann equation and the two dimensional momentum equation were used to model the electric double layer field and the flow field. The velocity profile of electro-osmotic flow (EOF) in rectangular microchannels has been investigated numerically. Tsao *et al* [71] studied electroosmotic flow through an annulus. He linearized the Poisson-Boltzmann equation to solve the electric potential and used the Stokes equation to obtain the velocity profile. Ren *et al* [72] discussed theoretical and experimental studies of electroosmotic flow in a capillary with one electrolyte solution displacing another solution.

2.4 Experimental techniques for microfluidics

Many of the traditional techniques of measuring fluid behavior have been adapted or replaced due to the small size and the planar format of these systems [37]. Most other measurements of microflows have been performed with optical microscopes. Santiago *et al* [73] developed a micron-resolution particle image velocimetry (micro-PIV) system to measure instantaneous and ensemble-averaged flow fields in micron-scale fluidic devices. The system utilized an epifluorescent microscope, 100-300 nm diameter seed particles, and an intensified CCD camera to record high-resolution particle-image fields. Velocity vector fields can be measured with spatial resolutions down to $6.9 \times 6.9 \times 1.5 \mu\text{m}$. The vector fields are analyzed using a double-frame cross-correlation algorithm. They used micro-PIV technique to measure velocities in a Hele-Shaw flow around a 30

Chapter 2 Literature Review

μm (major diameter) elliptical cylinder, with a bulk velocity of approximately $50 \mu\text{m s}^{-1}$.

Devasenathipathy *et al* [74] applied particle tracking techniques to obtain spatially resolved velocity measurements in electrokinetic flow devices. Both micrometer-resolution particle image velocimetry (micro-PIV) and particle tracking velocimetry (PTV) techniques have been used to quantify and study flow phenomena in electrokinetic systems applicable to microfluidic bioanalytical devices. They performed a series of seed particle calibration experiments. First, to measure the electroosmotic wall mobility of a borosilicate rectangular capillary (40 by $400 \mu\text{m}$) using current monitoring. Afterwards, PTV was applied to determine the electrophoretic mobilities of more than 1000 fluorescent microsphere particles in aqueous buffer solutions. Particles from this calibrated particle/ buffer mixture were then introduced into two electrokinetic flow systems for particle tracking flow experiments. In these experiments, they measured the velocity field of the flow using micro-PIV.

Micro-PIV was used to measure the motion of nominally $1\text{-}\mu\text{m}$ diameter polystyrene particles suspended in a water/sugar solution in a micro-device under AC electric field [75]. Particle motion is induced by an applied electric field through the dielectrophoretic (DEP) forces and viscous interactions with the fluid. The motion of the fluid is governed by electrothermal forces arising from temperature-induced gradients in the electrical properties of the fluid near the electrodes and through the viscous interaction with the particles. Comparisons between finite element calculations and experimental micro-PIV results confirm quantitatively that electrothermal forces play a major role in the motion of the particles.

In the study of Kim *et al* [17], micro-PIV characterization of two-fluid flow in a microchannel was analysed. The effect of differences in the refractive indices of the two fluids on the accuracy of the PIV data was presented. In the study, they first analyzed the objective-imaging system used for two-fluid flow measurement, and then derived the

Chapter 2 Literature Review

precondition for measurement of a valid velocity profile across the two-fluid interface. A micro PIV experimental system was set up to measure the two-fluid flow inside a Y-shaped microchannel.

Other experimental studies using micro-PIV technique includes liquid flow in microchannel [76, 77], flow development of co-flowing streams in rectangular microchannel [78] and transition from laminar to turbulent flow in liquid filled microtubes [79].

Most of the mixing experiment in microchannel utilizes the optical microscope for the measurement. The mixing performance in a T-mixer was characterized using a blue dye and a colourless liquid [4]. The results were further verified by the hydrolysis reaction of dichloroacetyl phenol red. In the experiment of nonlinear diffusive mixing [27], the images obtained using a microscope were analyzed using a customized MATLAB program. The measured dimensionless intensity is assumed to be equal to the dimensionless concentration of the fluorescent dye. A measurement technique using fluorescent dye in combination with micro-resolution particle image velocimetry (micro-PIV) has been devised to investigate convective mixing in microspace was developed by Sato *et al* [18]. Tris(bipyridine)ruthenium(II), whose fluorescent intensity when excited by ultraviolet light is strongly temperature dependent, was applied to the bottom surface of a cover glass, that served as the upper boundary surface of a flow channel. They realized a two-dimensional temperature measurement of the flow in microchannel. Other electrokinetic mixing experiments also used microscope to characterize the mixing performance [5, 80–82].

2.5 Remarks on previous works

Concerning the stratified two-fluid flow, the literature review shows that most of the research work focused on single fluid electroosmosis flow, or pressure-driven two-fluid flow, or mixing using electroosmosis effect. The survey shows that so far, no work is done to study the stratified two-fluid flow in microchannel under the combined effect of

Chapter 2 Literature Review

pressure and electroosmosis. One aim of this work is to study the pressure-driven two-fluid flow in microchannel with electroosmosis. Experiments were performed to study the velocity field and the interface position of the two fluids. Theoretical analysis of the velocity field was solved.

With respect to droplets in microfluidics, the flow field inside the droplets causes chaotic advection and improves mixing. Although this effect was well known and widely used in several applications, understanding of the flow field inside the droplet were limited by speculation [50]. So far, there is no experimental measurement of the flow field inside a moving micro-droplet. Thus, the second aim of this project is to measure the flow field inside the micro-droplet and examine the recirculation. Mixing of two water droplets during coalescence was also measured by using the fluorescence imaging technique.

Chapter 3

Two-fluid stratified flow under pressure and electroosmosis: modelling

The schematic of the two-fluid flow in a microchannel is shown in Figure 3.1. Fluid 1 and fluid 2 are conducting and non-conducting fluids respectively. Electrical double layer forms near the channel walls in contact with the conducting fluid and the fluid-fluid interface. When an external electric field is applied along the channel longitudinal direction, there will be an electroosmotic flow of the conducting fluid 1. The non-conducting fluid will be dragged by fluid 1 because of the viscous force at the interface.

Figure 3.1: Schematic of two fluids flow in microchannel and the coordinates used for mathematical formulation.

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

A cartesian orthonormal coordinate system (x, y, z) is used for mathematical formulation. The origin is at the intersection of the interface and the symmetrical line. The widths of fluid 1 and fluid 2 are w_1 and w_2 respectively. Due to the symmetry, only half of the cross section of the channel is considered. EDLs only form near the channel walls in contact with the conducting fluid and fluid-fluid interface. The zeta potentials at the side walls and top wall and the fluid-fluid interface are ζ_1 , ζ_2 , and ζ_3 respectively. The driving forces and electroosmotic flow are along the x -direction.

The closed form analytical solution of the two-fluid flow with electroosmosis effect is presented. The pressure effect is also included. The assumptions made for the mathematical formulation are

- The flow is fully developed and laminar.
- The fluids are incompressible.
- The fluids are immiscible.
- No-slip boundary conditions at the walls.
- The interface is planar and smooth.

3.1 Electrical potential distribution in microchannel

Fundamental equations governing the electrostatic potential and fluid flow are essential to the understanding of the electroosmotic flow. The relation between the potential (or the electric field) ψ at any point and the number of charges is given by Poisson's equation. The equation is with such assumptions that: the solvent is continuous, ions are point charges, and the solvent is characterized by a constant permittivity. The two dimensional Poisson equation:

$$\frac{\partial^2 \psi}{\partial y^2} + \frac{\partial^2 \psi}{\partial z^2} = -\frac{\rho_e}{\epsilon_r \epsilon_0} \quad (3.1)$$

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

where ρ_e is the net volumetric charge density, ϵ_r is the dimensionless dielectric constant of the solution and ϵ_0 is the permittivity of the vacuum. The ionic concentration can be described using the Boltzmann equation, as

$$n_i = n_{i0} \exp\left(-\frac{z_i e \Psi}{k_b T}\right) \quad (3.2)$$

where n_{i0} and z_i are the bulk concentration and the valence of type- i ions, respectively; e is the elementary charge; k_b is the Boltzmann constant and T is the absolute temperature.

For simplicity, a symmetric solution with univalent charge is analysed. The cations and anions are identified with $+$ and $-$, thus $n_+ = -n_- = n_0$ and $z_+ = -z_- = z_0$, where n_0 is the ionic concentration in the bulk solution. The net volumetric charge density, ρ_e , is proportional to the concentration difference between symmetric cations and anions,

$$\rho_e = z_0 e (n_+ - n_-) = -2z_0 e n_0 \sinh\left(\frac{z_0 e \Psi}{k_b T}\right) \quad (3.3)$$

Substituting Eqn (3.3) to Eq. (3.1) leads to the Poisson-Boltzmann equation

$$\frac{\partial^2 \Psi}{\partial y^2} + \frac{\partial^2 \Psi}{\partial z^2} = \frac{2n_0 z_0 e}{\epsilon_r \epsilon_0} \sinh\left(\frac{z_0 e \Psi}{k_b T}\right) \quad (3.4)$$

By defining the Debye-Huckel parameter $\kappa = (2z_0^2 e^2 n_0 / \epsilon_r \epsilon_0 k_b T)^{1/2}$, and hydraulic diameter $D_h = [4(w_1 + w_2)h] / (w_1 + w_2 + 2h)$, and by introducing non-dimensional parameters $Y = y/D_h$, $Z = z/D_h$, $\Psi = \frac{z_0 e \Psi}{k_b T}$, and $K = \kappa D_h$, The non-dimensional equation can be written as

$$\frac{\partial^2 \Psi}{\partial Y^2} + \frac{\partial^2 \Psi}{\partial Z^2} = K^2 \sinh \Psi \quad (3.5)$$

Debye length, $1/\kappa$, is the characteristic thickness of the electrical double layer [2]. The dimensionless group κD_h measures the relative channel diameter, compared to the EDL thickness. It is often referred as the electrokinetic diameter, and defined as $K = \kappa D_h$.

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

Due to the symmetry of the rectangular channel, the boundary conditions are

$$Y = 0, \quad \Psi = \bar{\zeta}_3 \quad (3.6a)$$

$$Y = -w_1/D_h = W_1, \quad \Psi = \bar{\zeta}_1 \quad (3.6b)$$

$$Z = h/D_h = H, \quad \Psi = \bar{\zeta}_2 \quad (3.6c)$$

$$Z = 0, \quad \frac{\partial \Psi}{\partial Z} = 0 \quad (3.6d)$$

where the non-dimensional zeta potentials are defined as $\bar{\zeta}_1 = z_0 e \zeta_1 / k_b T$, $\bar{\zeta}_2 = z_0 e \zeta_2 / k_b T$, and $\bar{\zeta}_3 = z_0 e \zeta_3 / k_b T$.

For small values of zeta potential (the Debye-Hückel approximation, which means physically that the electrical potential is small compared with the thermal energy of ions; i.e., $|ze\psi| < kT$), the Poisson-Boltzmann equation can be linearized as,

$$\frac{\partial^2 \Psi}{\partial Y^2} + \frac{\partial^2 \Psi}{\partial Z^2} = K^2 \Psi \quad (3.7)$$

This is a linear partial differential equation with nonhomogeneous boundary conditions. The nonhomogeneous boundary conditions have to be transformed to homogeneous. It is assumed that the solution is in the form

$$\Psi(Y, Z) = \Psi_1(Y, Z) + \Psi_2(Y, Z) + \Psi_3(Y, Z) \quad (3.8)$$

And the three subset partial differential equations have following boundary conditions

$$\frac{\partial^2 \Psi_1}{\partial Y^2} + \frac{\partial^2 \Psi_1}{\partial Z^2} = K^2 \Psi_1 \quad (3.9a)$$

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

$$\Psi_1(0, Z) = 0 \quad \Psi_1(W_1, Z) = \bar{\zeta}_1 \quad (3.9b)$$

$$\frac{\partial \Psi_1}{\partial Z}(Y, 0) = 0 \quad \Psi_1(Y, H) = 0 \quad (3.9c)$$

$$\frac{\partial^2 \Psi_2}{\partial Y^2} + \frac{\partial^2 \Psi_2}{\partial Z^2} = K^2 \Psi_2 \quad (3.10a)$$

$$\Psi_2(0, Z) = \bar{\zeta}_3 \quad \Psi_2(W_1, Z) = 0 \quad (3.10b)$$

$$\frac{\partial \Psi_2}{\partial Z}(Y, 0) = 0 \quad \Psi_2(Y, H) = 0 \quad (3.10c)$$

$$\frac{\partial^2 \Psi_3}{\partial Y^2} + \frac{\partial^2 \Psi_3}{\partial Z^2} = K^2 \Psi_3 \quad (3.11a)$$

$$\Psi_3(0, Z) = 0 \quad \Psi_3(W_1, Z) = 0 \quad (3.11b)$$

$$\frac{\partial \Psi_3}{\partial Z}(Y, 0) = 0 \quad \Psi_3(Y, H) = \bar{\zeta}_2 \quad (3.11c)$$

If Ψ_1 , Ψ_2 and Ψ_3 are solved respectively, the solution of Eq. (3.7) will be obtained using Eq. (3.8). Eq. (3.9a) is solved by the method of separation of variables. The solution is

$$\Psi_1(Y, Z) = \sum_{n=1}^{\infty} \frac{4\bar{\zeta}_1 (-1)^{n+1} \sinh \left[\sqrt{K^2 + \frac{(2n-1)^2 \pi^2}{4H^2}} Y \right]}{(2n-1)\pi \sinh \left[\sqrt{K^2 + \frac{(2n-1)^2 \pi^2}{4H^2}} W_1 \right]} \cos \left[\frac{(2n-1)\pi}{2H} Z \right] \quad (3.12)$$

Eq. (3.10a) can be solved in a similar way using separation of variables techniques.

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

Introducing $\bar{Y} = W_1 - Y$, as for

$$\frac{\partial \Psi_2}{\partial \bar{Y}} = \frac{\partial \Psi_2}{\partial Y} \frac{\partial Y}{\partial \bar{Y}} = -\frac{\partial \Psi_2}{\partial Y}$$

and

$$\frac{\partial^2 \Psi_2}{\partial \bar{Y}^2} = \frac{\partial Y}{\partial \bar{Y}} \frac{\partial}{\partial Y} \left(\frac{\partial \Psi_2}{\partial \bar{Y}} \right) = \frac{\partial^2 \Psi_2}{\partial Y^2}$$

Eq. (3.10a) can be transformed into

$$\frac{\partial^2 \Psi_2}{\partial \bar{Y}^2} + \frac{\partial^2 \Psi_2}{\partial Z^2} = K^2 \Psi_2 \quad (3.13a)$$

$$\Psi_2(W_1, Z) = \bar{\zeta}_3 \quad \Psi_2(0, Z) = 0 \quad (3.13b)$$

$$\Psi_2(\bar{Y}, 0) = 0 \quad \Psi_2(\bar{Y}, H) = 0 \quad (3.13c)$$

which has exactly the same form as Eq. (3.9a). Therefore, the solution to Eq. (3.13a) is

$$\Psi_2(\bar{Y}, Z) = \sum_{m=1}^{\infty} \frac{4\bar{\zeta}_3(-1)^{m+1} \sinh \left[\sqrt{K^2 + \frac{(2m-1)^2 \pi^2}{4H^2}} \bar{Y} \right]}{(2m-1)\pi \sinh \left[\sqrt{K^2 + \frac{(2m-1)^2 \pi^2}{4H^2}} W_1 \right]} \cos \left[\frac{(2m-1)\pi}{2H} Z \right]$$

In (Y, Z) coordinates the final solution of $\Psi_2(Y, Z)$ would be

$$\Psi_2(Y, Z) = \sum_{m=1}^{\infty} \frac{4\bar{\zeta}_3(-1)^{m+1} \sinh \left[\sqrt{K^2 + \frac{(2m-1)^2 \pi^2}{4H^2}} (W_1 - Y) \right]}{(2m-1)\pi \sinh \left[\sqrt{K^2 + \frac{(2m-1)^2 \pi^2}{4H^2}} W_1 \right]} \cos \left[\frac{(2m-1)\pi}{2H} Z \right] \quad (3.14)$$

Similarly, Eq. (3.11a) will be solved using separation of variables method as well.

The final solution is

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

$$\Psi_3(Y, Z) = \sum_{p=1}^{\infty} \frac{2\bar{\zeta}_2 [1 + (-1)^{p+1}] \cosh \left(\sqrt{K^2 + \frac{p^2 \pi^2}{W_1^2}} Z \right)}{p\pi \cosh \left(\sqrt{K^2 + \frac{p^2 \pi^2}{W_1^2}} H \right)} \sin \left(\frac{p\pi}{W_1} Y \right) \quad (3.15)$$

Substituting Eqs. (3.12), (3.14) and (3.15) into Eq. (3.8), the analytical solution of the electrical potential distribution is

$$\begin{aligned} \Psi(Y, Z) = & 4\bar{\zeta}_1 \sum_{n=1}^{\infty} \frac{(-1)^{n+1} \sinh \left[\sqrt{K^2 + \frac{(2n-1)^2 \pi^2}{4H^2}} Y \right]}{(2n-1)\pi \sinh \left[\sqrt{K^2 + \frac{(2n-1)^2 \pi^2}{4H^2}} W_1 \right]} \cos \left[\frac{(2n-1)\pi}{2H} Z \right] \\ & + 4\bar{\zeta}_3 \sum_{m=1}^{\infty} \frac{(-1)^{m+1} \sinh \left[\sqrt{K^2 + \frac{(2m-1)^2 \pi^2}{4H^2}} (W_1 - Y) \right]}{(2m-1)\pi \sinh \left[\sqrt{K^2 + \frac{(2m-1)^2 \pi^2}{4H^2}} W_1 \right]} \cos \left[\frac{(2m-1)\pi}{2H} Z \right] \\ & + 2\bar{\zeta}_2 \sum_{p=1}^{\infty} \frac{[1 + (-1)^{p+1}] \cosh \left(\sqrt{K^2 + \frac{p^2 \pi^2}{W_1^2}} Z \right)}{p\pi \cosh \left(\sqrt{K^2 + \frac{p^2 \pi^2}{W_1^2}} H \right)} \sin \left(\frac{p\pi}{W_1} Y \right) \end{aligned} \quad (3.16)$$

3.2 The flow field of two-fluid flow

For stratified flow of water and oil under steady conditions with a constant cross sectional area, inertial terms in the Navier-Stokes equation are zero, the Navier-Stokes equation reduces to

$$\frac{\partial^2 u_1^*}{\partial y^2} + \frac{\partial^2 u_1^*}{\partial z^2} = -\frac{1}{\mu_1} E_x \rho_e(y, z) - \left(\frac{dp}{dx} \right)_1 \quad (3.17)$$

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

$$\frac{\partial^2 u_2^*}{\partial y^2} + \frac{\partial^2 u_2^*}{\partial z^2} = -\left(\frac{dp}{dx}\right)_2 \quad (3.18)$$

where subscript 1 denotes the fluid 1, i.e. water phase, and subscript 2 denotes fluid 2, i.e. oil phase. E_x is the externally applied electrical field along the x direction.

The right sides of the Eqs. (3.17) and (3.18) indicate that the flow is caused by electroosmosis and pressure gradient effects. Therefore, these two linear partial differential equations can be split into two sets of equations to account for the electroosmosis effect and pressure gradient effect respectively, as

$$\frac{\partial^2 u_1}{\partial y^2} + \frac{\partial^2 u_1}{\partial z^2} = -\frac{1}{\mu_1} E_x \rho_e(y, z) \quad (3.19)$$

$$\frac{\partial^2 u_2}{\partial y^2} + \frac{\partial^2 u_2}{\partial z^2} = 0 \quad (3.20)$$

and

$$\frac{\partial^2 u_3}{\partial y^2} + \frac{\partial^2 u_3}{\partial z^2} = -\left(\frac{dp}{dx}\right)_1 \quad (3.21)$$

$$\frac{\partial^2 u_4}{\partial y^2} + \frac{\partial^2 u_4}{\partial z^2} = -\left(\frac{dp}{dx}\right)_2 \quad (3.22)$$

The flow field induced by electroosmosis and pressure gradient are to be presented in the following sections. Afterwards, the solution of u_1^* and u_2^* will be obtained as

$$u_1^* = u_1 + u_3$$

and

$$u_2^* = u_2 + u_4$$

3.3 The effect of the electroosmosis

To nondimensionalize Eqs. (3.19) and (3.20), some reference quantities are introduced: $U_{ref} = E_x \epsilon_r \epsilon_0 k_v T / (ze_0 \mu_{ref})$ is the Helmholtz-Smoluchowski electroosmotic velocity

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

and $\mu_{ref} = \mu_1$, $\rho_{ref} = \rho_1$. Defining the reference Reynolds number

$$Re_0 = \frac{\rho_{ref} D_h U_{ref}}{\mu_{ref}}$$

and introducing dimensionless parameters

$$Y = \frac{y}{D_h} \quad X = \frac{x}{D_h Re_0}$$

$$U_1 = \frac{u_1}{U_{ref}} \quad U_2 = \frac{u_2}{U_{ref}}$$

Substituting the electrical potential described in Eq. (3.16) the non-dimensional equations corresponding to Eqs. (3.19) and (3.20) are

$$\frac{\partial^2 U_1}{\partial Y^2} + \frac{\partial^2 U_1}{\partial Z^2} = K^2 \Psi(Y, Z) \quad (3.23)$$

$$\frac{\partial^2 U_2}{\partial Y^2} + \frac{\partial^2 U_2}{\partial Z^2} = 0 \quad (3.24)$$

The symmetric and no-slip boundary conditions apply for the conducting fluid as

$$\text{at } Z = 0, \quad \frac{\partial U_1}{\partial Z} = 0 \quad (3.25a)$$

$$\text{at } Z = h/D_h = H, \quad U_1 = 0 \quad (3.25b)$$

$$\text{at } Y = -w_1/D_h = W_1, \quad U_1 = 0 \quad (3.25c)$$

And the boundary conditions for the non-conducting fluid are

$$\text{at } Z = 0, \quad \frac{\partial U_2}{\partial Z} = 0 \quad (3.26a)$$

$$\text{at } Z = h/D_h = H, \quad U_2 = 0 \quad (3.26b)$$

$$\text{at } Y = w_2/D_h = W_2, \quad U_2 = 0 \quad (3.26c)$$

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

At the interface of the two fluids, matching conditions exist. The continuity of velocity and hydrodynamic shear stress are presented by

$$U_1(0, Z) = U_2(0, Z) \quad (3.27)$$

$$\frac{\partial U_1}{\partial \tau}|_{Y=0} = \beta \frac{\partial U_2}{\partial \tau}|_{Y=0} \quad (3.28)$$

where τ is the tangential direction along the interface between the two fluids, $\beta = \mu_2/\mu_1$ is the dynamic viscosity ratio.

3.3.1 The non-conducting fluid flow

Using the separation of variables method, The solution for Eq. 3.20 can be written as

$$U_2(Y, Z) = \sum_{i=1}^{\infty} \left[\sinh(\lambda_i Y) - \tanh(\lambda_i W_2) \cosh(\lambda_i Y) \right] b_i \cos(\lambda_i Z) \quad (3.29)$$

where

$$\lambda_i = \frac{(2i-1)\pi}{2H} \quad i = 1, 2, 3 \dots$$

The coefficient b_i is an unknown, which will be determined later using the matching conditions between the two fluids.

3.3.2 The conducting fluid flow

The eigenfunctions of the associated eigenvalue problem of Eq. (3.23) is

$$\cos\left(\frac{2j-1}{2H}\pi Z\right)$$

in which $j = 1, 2, 3 \dots$. The solution is assumed to have the form

$$U_1(Y, Z) = \sum_{j=1}^{\infty} U_{1j}(Y) \cos\left(\frac{2j-1}{2H}\pi Z\right) \quad (3.30)$$

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

where $U_{1j}(Y)$ are to be determined. The boundary conditions of (3.25a) and (3.25b) are satisfied by Eq. (3.30).

Applying the separation of variables techniques, The solution of $U_1(Y, Z)$ can now be written as

$$U_1(Y, Z) = \sum_{j=1}^{\infty} \left\{ [\sinh(\lambda_j Y) - \tanh(\lambda_j W_1) \cosh(\lambda_j Y)] b_j - \frac{\cosh(\lambda_j Y)}{\cosh(\lambda_j W_1)} \Phi_j(W_1) + \Phi_j(Y) \right\} \cos(\lambda_j Z) \quad (3.31)$$

where the function $\Phi_j(Y)$ as

$$\Phi_j(Y) = \frac{1}{\lambda_j} \int_0^Y \sinh[\lambda_j(Y - \tau)] F_j(\tau) d\tau \quad (3.32)$$

and

$$F_j(Y) = \frac{2}{H} \int_0^H K^2 \Psi(Y, Z) \cos\left(\frac{2j-1}{2H} \pi Z\right) dZ \quad (3.33)$$

b_j will be determined using the matching conditions (3.27) and (3.28). The evaluation $\Phi_j(Y)$ can be determined at a later stage.

The j in Eq. (3.31) and i in Eq. (3.29) are dummy variables, $i = j = 1, 2, 3, \dots$, thus the coefficient of b_j and b_i can be replaced by b_{1j} and b_{2j} . The solution of $U_1(Y, Z)$ and $U_2(Y, Z)$ can be written as

$$U_1(Y, Z) = \sum_{j=1}^{\infty} \left\{ [\sinh(\lambda_j Y) - \tanh(\lambda_j W_1) \cosh(\lambda_j Y)] b_{1j} - \frac{\cosh(\lambda_j Y)}{\cosh(\lambda_j W_1)} \Phi_j(W_1) + \Phi_j(Y) \right\} \cos(\lambda_j Z) \quad (3.34)$$

and

$$U_2(Y, Z) = \sum_{j=1}^{\infty} [(\sinh(\lambda_j Y) - \tanh(\lambda_j W_2) \cosh(\lambda_j Y)] b_{2j} \cos(\lambda_j Z) \quad (3.35)$$

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

where $b1_j$ and $b2_j$ are to be determined from the matching conditions (3.27) and (3.28).

From Eqs. (3.34) and (3.20), we have the following

$$\begin{aligned} \frac{\partial U_1(Y, Z)}{\partial Y} = \sum_{j=1}^{\infty} \left\{ [\lambda_j \cosh(\lambda_j Y) - \lambda_j \tanh(\lambda_j W_1) \sinh(\lambda_j Y)] b1_j \right. \\ \left. + \frac{\lambda_j \sinh(\lambda_j Y)}{\cosh(\lambda_j W_1)} \Phi_j(W_1) + \Phi'_j(Y) \right\} \cos(\lambda_j Z) \end{aligned} \quad (3.36)$$

and

$$\frac{\partial U_2(Y, Z)}{\partial Y} = \sum_{j=1}^{\infty} [\lambda_j \cosh(\lambda_j Y) - \lambda_j \tanh(\lambda_j W_2) \sinh(\lambda_j Y)] b2_j \cos(\lambda_j Z) \quad (3.37)$$

The continuity of velocity at the interface, as in Eq. (3.27) leads to

$$\tanh(\lambda_j W_1) b1_j + \frac{\Phi_j(W_1)}{\cosh(\lambda_j W_1)} - \Phi_j(0) = \tanh(\lambda_j W_2) b2_j \quad (3.38)$$

The continuity of shear stress at the interface, as in Eq. (3.28) gives

$$\lambda_j b1_j + \Phi'_j(0) = \beta \lambda_j b2_j \quad (3.39)$$

From Eqs. (3.38) and (3.39), the coefficients of $b1_j$ and $b2_j$ can now be obtained

$$b1_j = \beta \frac{\frac{\Phi_j(W_1)}{\cosh(\lambda_j W_1)} - \frac{\Phi'_j(0)}{\lambda_j} \tanh(\lambda_j W_1) - \Phi_j(0)}{\tanh(\lambda_j W_2) - \beta \tanh(\lambda_j W_1)} - \frac{1}{\lambda_j} \Phi'_j(0) \quad (3.40)$$

$$b2_j = \frac{\frac{\Phi_j(W_1)}{\cosh(\lambda_j W_1)} - \frac{\Phi'_j(0)}{\lambda_j} \tanh(\lambda_j W_1) - \Phi_j(0)}{\tanh(\lambda_j W_2) - \beta \tanh(\lambda_j W_1)} \quad (3.41)$$

3.3.3 The formulation of $\Phi_j(Y)$

The solution of $\Psi(Y, Z)$ has been obtained in Eq. (3.16). The function of $F_j(Y)$ is also known in Eq. (3.33), the expression of $\Phi_j(Y)$ in Eq. (3.32), after integration, is

$$\begin{aligned} \Phi_j(Y) = & -\frac{4K^2\bar{\zeta}_1(-1)^{1+j}\sinh(B_jY)}{(2j-1)\pi(\lambda_j^2-B_j^2)\sinh(b_jW_1)} \\ & -\frac{4K^2\bar{\zeta}_3(-1)^{1+j}\sinh[B_j(W_1-Y)]}{(2j-1)\pi(\lambda_j^2-B_j^2)\sinh(B_jW_1)} \\ & -\sum_{p=1}^{\infty}\frac{4K^2\bar{\zeta}_2[1+(-1)^{1+p}]\lambda_j(-1)^{1+j}}{pH\pi(A_p^2+\lambda_j^2)(\gamma_p^2+\lambda_j^2)}\sin(\gamma_pY) \end{aligned} \quad (3.42)$$

where

$$\begin{aligned} A_p &= \sqrt{K^2 + \frac{p^2\pi^2}{W_1^2}} \\ \gamma_p &= \frac{p\pi}{W_1} \\ B_j &= \sqrt{K^2 + \frac{(2j-1)^2\pi^2}{4H^2}} \\ \lambda_j &= \frac{(2j-1)\pi}{2H} \end{aligned}$$

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

And

$$\begin{aligned}\Phi'_j(Y) = & -\frac{4K^2\bar{\zeta}_1(-1)^{1+j}B_j \cosh(B_j Y)}{(2j-1)\pi(\lambda_j^2 - B_j^2) \sinh(b_j W_1)} \\ & + \frac{4K^2\bar{\zeta}_3(-1)^{1+j}B_j \sinh[B_j(W_1 - Y)]}{(2j-1)\pi(\lambda_j^2 - B_j^2) \sinh(B_j W_1)} \\ & - \sum_{p=1}^{\infty} \frac{4K^2\bar{\zeta}_2[1 + (-1)^{1+p}]\lambda_j(-1)^{1+j}\gamma_p \cos(\gamma_p Y)}{pH\pi(A_p^2 + \lambda_j^2)(\gamma_p^2 + \lambda_j^2)}\end{aligned}\quad (3.43)$$

3.3.4 Volumetric flow rate due to electroosmosis

After the velocity field of the two fluids are found, the volumetric flow rates can be obtained by integrating the velocity field over the cross section. The dimensionless volumetric flow rate through the rectangular channel can be defined as $Q_1 = q_1/(D_h^2 U_{ref})$ and $Q_2 = q_2/(D_h^2 U_{ref})$ (where q_1 and q_2 are the volumetric flow rate for the conducting and non-conducting fluids, respectively). The dimensionless flow rate are given by

$$Q_1 = 2 \int_0^H \int_{W_1}^0 U_1(Y, Z) dY dZ \quad (3.44)$$

and

$$Q_2 = 2 \int_0^H \int_0^{W_2} U_2(Y, Z) dY dZ \quad (3.45)$$

Substituting Eqs. (3.34) and (3.35) into Eqs. (3.44) and (3.45), the dimensionless volumetric flow rates are

$$\begin{aligned}Q_1 = 2 \sum_{j=1}^{\infty} \left\{ \left[1 - \frac{1}{\cosh(\lambda_j W_1)} \right] \frac{b1_j}{\lambda_j} + \frac{\Phi_j(W_1) \sinh(\lambda_j W_1)}{\lambda_j \cosh(\lambda_j W_1)} \right. \\ \left. + I_j^{\Phi}(0) - I_j^{\Phi}(W_1) \right\} \frac{\sinh(\lambda_j H)}{\lambda_j}\end{aligned}\quad (3.46)$$

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

$$Q_2 = 2 \sum_{j=1}^{\infty} \left[\frac{1}{\cosh(\lambda_j W_2)} - 1 \right] \frac{b 2_j}{\lambda_j^2} \sin(\lambda_j H) \quad (3.47)$$

where

$$\begin{aligned} I_j^{\Phi}(Y) = & -\frac{4K^2 \bar{\zeta}_1 (-1)^{1+j} \cosh(B_j Y)}{(2j-1)\pi(\lambda_j^2 - B_j^2) B_j \sinh(B_j W_1)} \\ & + \frac{4K^2 \bar{\zeta}_3 (-1)^{1+j} \sinh[B_j(W_1 - Y)]}{(2j-1)\pi(\lambda_j^2 - B_j^2) B_j \sinh(B_j W_1)} \\ & - \sum_{p=1}^{\infty} \frac{4K^2 \bar{\zeta}_2 [1 + (-1)^{1+p}] \lambda_j (-1)^{1+j}}{pH\pi(A_p^2 + \lambda_j^2)(\gamma_p^2 + \lambda_j^2)\gamma_p} \cos(\gamma_p Y) \end{aligned} \quad (3.48)$$

3.4 The effect of pressure gradient

If the effect of pressure gradient is considered, the velocity due to the pressure gradient can be written in dimensionless form as,

$$\frac{\partial^2 U_3}{\partial Y^2} + \frac{\partial^2 U_3}{\partial Z^2} = \left(\frac{dP}{dX} \right)_1 \quad (3.49)$$

$$\frac{\partial^2 U_4}{\partial Y^2} + \frac{\partial^2 U_4}{\partial Z^2} = \left(\frac{dP}{dX} \right)_2 \quad (3.50)$$

where the dimensionless pressure gradients are defined as

$$\left(\frac{dP}{dX} \right)_1 = \frac{D_h^2}{\mu_1 U_{ref}} \left(-\frac{dp}{dx} \right)_1 \quad (3.51)$$

and

$$\left(\frac{dP}{dX} \right)_2 = \frac{D_h^2}{\beta \mu_1 U_{ref}} \left(-\frac{dp}{dx} \right)_2 \quad (3.52)$$

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

with boundary conditions, for conducting fluid, as

$$\text{at } Z = 0, \quad U_3 = 0 \quad (3.53a)$$

$$\text{at } Z = h/D_h = H, \quad U_3 = 0 \quad (3.53b)$$

$$\text{at } Y = -w_1/D_h = W_1, \quad U_3 = 0 \quad (3.53c)$$

and for non-conducting fluid, as

$$\text{at } Z = 0, \quad \frac{\partial U_4}{\partial Z} = 0 \quad (3.54a)$$

$$\text{at } Z = h/D_h = H, \quad U_4 = 0 \quad (3.54b)$$

$$\text{at } Y = w_2/D_h = W_2, \quad U_4 = 0 \quad (3.54c)$$

The same techniques are used to solve Eqs. (3.49) and (3.50). The solution of $U_3(Y, Z)$ can be written as

The solution of $U_3(Y, Z)$ can be written as

$$U_3(Y, Z) = \sum_{j=1}^{\infty} \left\{ \left[\sinh(\lambda_j Y) - \tanh(\lambda_j W_1) \cosh(\lambda_j Y) \right] b_{3j} - \frac{\cosh(\lambda_j Y)}{\cosh(\lambda_j W_1)} \Phi_{3j}(W_1) + \Phi_{3j}(Y) \right\} \cos(\lambda_j Z) \quad (3.55)$$

where the function $\Phi_{3j}(Y)$ is

$$\Phi_{3j}(Y) = \frac{1}{\lambda_j} \int_0^Y \sinh[\lambda_j(Y - \tau)] F_{3j}(\tau) d\tau \quad (3.56)$$

and

$$F_{3j}(Y) = \frac{2}{H} \int_0^H \left(\frac{dP}{dX} \right)_1 \cos\left(\frac{2j-1}{2H} \pi Z\right) dZ = \frac{4 \times (-1)^{(1+j)}}{(2j-1)\pi} \left(\frac{dP}{dX} \right)_1 \quad (3.57)$$

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

Similarly, the solution of $U_4(Y, Z)$ are found to be

$$U_4(Y, Z) = \sum_{j=1}^{\infty} \left\{ \left[\sinh(\lambda_j Y) - \tanh(\lambda_j W_2) \cosh(\lambda_j Y) \right] b_{4j} - \frac{\cosh(\lambda_j Y)}{\cosh(\lambda_j W_2)} \Phi_{4j}(W_2) + \Phi_{4j}(Y) \right\} \cos(\lambda_j Z) \quad (3.58)$$

where the function $\Phi_{4j}(Y)$ is

$$\Phi_{4j}(Y) = \frac{1}{\lambda_j} \int_0^Y \sinh[\lambda_j(Y - \tau)] F_{4j}(\tau) d\tau \quad (3.59)$$

and

$$F_{4j}(Y) = \frac{4 \times (-1)^{(1+j)}}{(2j-1)\pi} \left(\frac{dP}{dX} \right)_2 \quad (3.60)$$

where, the coefficients b_{3j} and b_{4j} are to be determined from the matching conditions (3.27) and (3.28).

The derivatives of (3.55) and (3.58) are

$$\begin{aligned} \frac{\partial U_3(Y, Z)}{\partial Y} = \sum_{j=1}^{\infty} \left\{ \left[\lambda_j \cosh(\lambda_j Y) + \lambda_j \tanh(\lambda_j W_1) \sinh(\lambda_j Y) \right] b_{3j} + \frac{\lambda_j \sinh(\lambda_j Y)}{\cosh(\lambda_j W_1)} \Phi_{3j}(W_1) + \Phi'_{3j}(Y) \right\} \cos(\lambda_j Z) \end{aligned} \quad (3.61)$$

and

$$\begin{aligned} \frac{\partial U_4(Y, Z)}{\partial Y} = \sum_{j=1}^{\infty} \left\{ \left[\lambda_j \cosh(\lambda_j Y) + \lambda_j \tanh(\lambda_j W_2) \sinh(\lambda_j Y) \right] b_{4j} + \frac{\lambda_j \sinh(\lambda_j Y)}{\cosh(\lambda_j W_2)} \Phi_{4j}(W_2) + \Phi'_{4j}(Y) \right\} \cos(\lambda_j Z) \end{aligned} \quad (3.62)$$

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

The continuity of velocity at the interface, as in Eq. (3.27) leads to

$$-\tanh(\lambda_j W_1) b_{3j} - \frac{\Phi_{3j}(W_1)}{\cosh(\lambda_j W_1)} + \Phi_{3j}(0) = -\tanh(\lambda_j W_2) b_{4j} - \frac{\Phi_{4j}(W_2)}{\cosh(\lambda_j W_2)} + \Phi_{4j}(0) \quad (3.63)$$

The continuity of shear stress at the interface, as in Eq. (3.28) gives

$$\lambda_j b_{3j} + \Phi'_{3j}(0) = \beta \lambda_j b_{4j} + \beta \Phi'_{4j}(0) \quad (3.64)$$

From Eqs. (3.63) and (3.64), the coefficients of b_{3j} and b_{4j} can now be obtained

$$b_{3j} = \beta \frac{\frac{\Phi_{3j}(W_1)}{\cosh(\lambda_j W_1)} - \Phi_{3j}(0) - \frac{\Phi_{4j}(W_2)}{\cosh(\lambda_j W_2)} + \Phi_{4j}(0) - \frac{\tanh(\lambda_j W_2)}{\beta \lambda_j} [\Phi'_{3j}(0) - \Phi'_{4j}(0)]}{\tanh(\lambda_j W_2) - \beta \tanh(\lambda_j W_1)} \quad (3.65)$$

$$b_{4j} = \frac{b_{3j}}{\beta} - \frac{1}{\beta \lambda_j} [\Phi'_{3j}(0) - \Phi'_{4j}(0)] \quad (3.66)$$

3.4.1 Evaluation of $\Phi_{3j}(Y)$ and $\Phi_{4j}(Y)$

From Eqs. (3.57) and (3.56), we have

$$\begin{aligned} \Phi_{3j}(Y) &= \frac{4 \times (-1)^{(1+j)}}{(2j-1)\pi} \left(\frac{dP}{dX} \right)_1 \frac{1}{\lambda_j} \int_0^Y \sinh[\lambda_j(Y-\tau)] d\tau \\ &= \frac{4 \times (-1)^{(1+j)}}{\lambda_j^2 (2j-1)\pi} \left(\frac{dP}{dX} \right)_1 [\cosh(\lambda_j Y) - 1] \end{aligned} \quad (3.67)$$

$$\Phi'_{3j}(Y) = \frac{4 \times (-1)^{(1+j)}}{\lambda_j (2j-1)\pi} \left(\frac{dP}{dX} \right)_1 \sinh(\lambda_j Y) \quad (3.68)$$

Similarly, $\Phi_{4j}(Y)$ and $\Phi'_{4j}(Y)$ are

$$\Phi_{4j}(Y) = \frac{4 \times (-1)^{(1+j)}}{\lambda_j^2 (2j-1)\pi} \left(\frac{dP}{dX} \right)_2 [\cosh(\lambda_j Y) - 1] \quad (3.69)$$

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

and

$$\Phi'_{4j}(Y) = \frac{4 \times (-1)^{(1+j)}}{\lambda_j(2j-1)\pi} \left(\frac{dP}{dX} \right)_2 \sinh(\lambda_j Y) \quad (3.70)$$

From Eqs. (3.67) to (3.70), we have

$$\Phi_{3j}(0) = \Phi'_{3j}(0) = \Phi_{4j}(0) = \Phi'_{4j}(0) = 0 \quad (3.71)$$

Substituting Eq. (3.71) into Eqs. (3.65) and (3.66), b_{3j} and b_{4j} can be simplified to

$$b_{3j} = \beta \frac{\frac{\Phi_{3j}(W_1)}{\cosh(\lambda_j W_1)} - \frac{\Phi_{4j}(W_2)}{\cosh(\lambda_j W_2)}}{\tanh(\lambda_j W_2) - \beta \tanh(\lambda_j W_1)} \quad (3.72)$$

and

$$b_{4j} = \frac{\frac{\Phi_{3j}(W_1)}{\cosh(\lambda_j W_1)} - \frac{\Phi_{4j}(W_2)}{\cosh(\lambda_j W_2)}}{\tanh(\lambda_j W_2) - \beta \tanh(\lambda_j W_1)} \quad (3.73)$$

3.4.2 Volumetric flow rate due to pressure gradient

The volumetric flow rates due to the pressure gradient can be found as

$$Q_3 = 2 \int_0^H \int_{W_1}^0 U_3(Y, Z) dY dZ \quad (3.74)$$

and

$$Q_4 = 2 \int_0^H \int_0^{W_2} U_4(Y, Z) dY dZ \quad (3.75)$$

Substituting Eqs. (3.55) and (3.55) into Eqs. (3.74) and (3.75), the dimensionless volumetric flow rates are

$$\begin{aligned} Q_3 = 2 \sum_{j=1}^{\infty} \left\{ \left[1 - \frac{1}{\cosh(\lambda_j W_1)} \right] \frac{b_{3j}}{\lambda_j} + \frac{\Phi_{3j}(W_1) \sinh(\lambda_j W_1)}{\lambda_j \cosh(\lambda_j W_1)} \right. \\ \left. + I_{3j}^{\Phi_3}(0) - I_{3j}^{\Phi_3}(W_1) \right\} \frac{\sinh(\lambda_j H)}{\lambda_j} \end{aligned} \quad (3.76)$$

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

$$Q_4 = 2 \sum_{j=1}^{\infty} \left\{ \left[\frac{1}{\cosh(\lambda_j W_2)} - 1 \right] \frac{b_{4j}}{\lambda_j} - \frac{\Phi_{4j}(W_2) \sinh(\lambda_j W_2)}{\lambda_j \cosh(\lambda_j W_2)} - I_{4j}^{\Phi_4}(0) + I_{4j}^{\Phi_4}(W_2) \right\} \frac{\sinh(\lambda_j H)}{\lambda_j} \quad (3.77)$$

where

$$I_{3j}^{\Phi_3}(Y) = \frac{4 \times (-1)^{(1+j)}}{\lambda_j^2 (2j-1) \pi} \left(\frac{dP}{dX} \right)_1 \left[\frac{1}{\lambda_j} \sinh(\lambda_j Y) - Y \right] \quad (3.78)$$

$$I_{4j}^{\Phi_4}(Y) = \frac{4 \times (-1)^{(1+j)}}{\lambda_j^2 (2j-1) \pi} \left(\frac{dP}{dX} \right)_2 \left[\frac{1}{\lambda_j} \sinh(\lambda_j Y) - Y \right] \quad (3.79)$$

3.5 Results and discussion

For the calculation, an aqueous electrolyte (e.g. NaCl) with univalent charge is used as the conducting fluid. The non-conducting fluids can be oil, which can vary in the viscosity for a large range. The dimensions of the micro-channel for calculation is $100\mu\text{m}$ in width and $100\mu\text{m}$ in height. The conducting and non-conducting fluids both occupy half the width of the channel. Thus, $w_1 = w_2 = 50\mu\text{m}$. The normalized zeta potentials ζ_1 , ζ_2 and ζ_3 , depend on the concentration of the solution, the material properties of the wall and the fluid ionic properties. The zeta potential between two immiscible liquids not only depends on the ionic properties of two fluids but also depends on the pH value and the concentration of the electrolyte.

3.5.1 Electric potential distribution

Debye length, $1/\kappa$, is the characteristic thickness of the electrical double layer [2]. The electrokinetic diameter, $K = \kappa D_h$ measures the relative channel diameter, compared to the EDL thickness. The effect of K on the electric potential distribution in the conducting fluid is shown in Figure 3.2. As $\kappa = (2z_0^2 e^2 n_0 / \epsilon_r \epsilon_0 k_b T)^{1/2}$, the value of K increases with solution concentration. The values of K for $10^{-4}M$ and $10^{-6}M$ are 1030 and 103 respectively. The larger the K value, the more the EDL is compressed. For Figure 3.2,

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

Figure 3.2: Dimensionless electrical potential distribution at the symmetrical line for different values of K , ($K = 103$ for $10^{-6}M$, and $K = 1030$ for $10^{-4}M$), $\zeta_1 = \zeta_2 = -25$ mV, $\zeta_3 = 0$.

it is seen that the EDL rise is much sharper and more rapid for $10^{-4}M$ solution. The electrical potential decreases rapidly to zero in a short distance away from the channel wall. This agrees well with previous study [83].

3.5.2 Effect of viscosity ratio, β , on the velocity profile

The velocity field depends on the viscosity ratio between the two fluids, because of the coupling effect between the two fluids. At the interface of the two fluids, the velocities and stresses must match. To see the effect of the viscosity ratio, different values of β , 1, 10, 100, 1000 and 10000 are used. The dimensionless velocity distributions at the symmetrical line are shown in Figures 3.3-3.4.

The electrical body force exists only in the electrical double layer region. As shown in the figure, the velocity increases rapidly in the EDL region near the wall and EDL region near the liquid liquid interface. Liquid outside the EDL region is set in motion

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

Figure 3.3: Dimensionless velocity distribution for different viscosity ratio ($\zeta_1=\zeta_2=-25$ mV, $\zeta_3=0$), β : (a) $\beta = 1$, (b) $\beta = 10$, (c) $\beta = 100$, (d) $\beta = 1000$.

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

Figure 3.4: Dimensionless velocity distribution at the symmetrical line for different viscosity ratio ($\zeta_1=\zeta_2=-25$ mV, $\zeta_3=0$), β : (a) $\beta = 1$, (b) $\beta = 10$, (c) $\beta = 100$, (d) $\beta = 1000$.

Chapter 3 Two-fluid stratified flow under pressure and electroosmosis: modelling

due to the hydrodynamic shear stress.

There are strong dependence between the velocity profiles and viscosity ratio β . When the viscosity ratio is higher, the flow resistance of the non-conducting fluid is higher, resulting in a steeper velocity gradient at the interface of the two fluids. For smaller viscosity ratio, the non-conducting can be dragged into motion in less effort. Consequently, the overall velocity magnitude is higher. The values of the maximum velocities for case $\beta = 1$ and $\beta = 1000$ differ by 1 order. For an extreme case, when of the viscosity of the non-conducting fluid approaches infinity, the flow of the conducting fluid resembles to a single fluid electroosmotic flow. Figure 3.5 shows the velocity profile for viscosity ratio 10000. The flow is almost an single fluid electroosmotic flow. For the non-conducting fluid, the velocity is zero. The velocity profile of the conducting fluid is plug-like.

Figure 3.5: Dimensionless velocity distribution for $\beta = 10000$: (a) three-dimensional view, (b) at the symmetrical line.

Chapter 4

Two-fluid stratified flow under pressure and electroosmosis: experiment

In this chapter, the experimental study of two-fluid stratified flow under pressure and electroosmosis effect is presented. The fabrication of microchannel, experimental method will be explained. In the first part of the experiment, the interface location between the two fluids, aqueous NaCl and aqueous glycerol, was investigated by fluorescence imaging technique. Secondly, the velocity profile measurement of the two fluids was carried out using micro-PIV techniques. The velocity field was measured quantitatively to study the effect of electroosmosis effect. The velocity field obtained from experiment is compared with the theoretical analysis from **Chapter 3**.

4.1 Fabrication of the microchannel

The fabrication method used in this project follows the adhesive lamination techniques described by Wu *et al* [27]. In this method, two polymethylmethacrylate (PMMA) plates (75 mm $\times$ 50 mm) are bonded by a layer or layers of double-sided adhesive tapes to form a closed microfluidic channel with inlet and outlet holes. The channel structure was cut through the adhesive tape (Adhesives Research Inc., Arclad 8102 transfer adhesive). Thus the adhesive tapes define the depth of the channel. The PMMA plates and adhesive tape are shown in Figure 4.1. The top layer has 4 access holes for inlet and outlet flow,

Figure 4.1: PMMA plates and adhesive tape drawings for the fabrication: (a) top layer PMMA plate, (b) bottom layer PMMA plate, (c) the middle layer adhesive tape.

3 alignment holes and 1 alignment cross marker. The bottom layer has 3 alignment holes, 1 alignment cross marker and 1 ruler for the purpose of locating measurement position. The channel structure cut through the adhesive tape is shown in Figure 4.1(c). An H-shaped microchannel is shown in Figure 4.2.

4.2 Experimental setup

The experimental setup can be used for fluorescence imaging and micro-PIV measurement. It consists of four main components: an illumination system, an optical system, a coupled charge device (CCD) camera and a control system. The control system consisting of a peripheral component interface (PCI) card, and its corresponding software, is implemented in a personal computer. The personal computer can control and synchronize all actions related to illumination and image recording. A schematic of the setup is shown in Figure 4.3.

Chapter 4 Two-fluid stratified flow under pressure and electroosmosis: experiment

Figure 4.2: H-shaped microchannel: (a) the fabricated device used in the experiment, (b) schematic drawing of the channel configuration (the main channel is between E and F with a cross section of $910\mu\text{m} \times 100\mu\text{m}$), in which two fluids flow side by side (electric field from A to B: +ve, from B to A: -ve).

Two different light sources were used for the two measurements. For the fluorescence imaging measurement, a single mercury lamp was used for the illumination. Because of the ability of precise timing and intensity control, a laser beam was used for the micro-PIV (micro particle image velocimetry) measurement. In our system, a double pulsed Q-switched (quality switched) Nd:YAG laser was used. By including a Q-switch inside the cavity the laser can work in a triggered mode. The laser has a wavelength of 532 nm and a maximum energy of 160 mJ. The two-laser-head system allows the realization of two laser pulses with a very small delay. The system can work in different modes: single exposure in one frame, double exposure in one frame, and double exposure in double frames. In our experiments, the mode of double exposures in double frames was used because of the high signal-to-noise ratios and the better quality of the

Chapter 4 Two-fluid stratified flow under pressure and electroosmosis: experiment

cross-correlation technique.

Figure 4.3: Schematic of the experimental setup for fluorescence imaging and micro-PIV measurements.

The optical system was a Nikon inverted microscope (Model ECLIPSE TE2000-S) with a set of epi-fluorescent attachments. There are three optical elements in a filter cube: excitation filter, dichroic mirror and emission filter. Emission filters are used in both measurements to select more specifically the emission wavelength of the sample and to remove traces of excitation light.

An interline transfer CCD camera (Sony ICX 084) was used for recording the images. The resolution of the camera is $640 \text{ pixels} \times 480 \text{ pixels}$, with 12 bits gray scale. The active area of the CCD sensor is $6.3 \text{ mm} \times 4.8 \text{ mm}$. The minimum inter-frame transfer time, and thus the fastest time delay for the two PIV images, is 300 ns. To ensure that the CCD camera is working at its optimum temperature of -15°C , a cooling system is integrated in the CCD camera. In the mode of double exposure in double frames, the camera can record two frames of the flow fields and then digitizes them in the same image buffer.

4.3 Results and discussion

4.3.1 Fluorescence imaging measurement

For the study of interface location between the two fluids, a microchannel with the cross section of $910\mu\text{m} \times 100\mu\text{m}$ and the length of 5 mm was used. The aqueous NaCl solution (concentration $7 \times 10^{-4}\text{M}$) and aqueous glycerol (volume concentration 14%) were introduced through inlets A and C through syringes (Hamilton) and syringe pump (Cole-Parmer, 74900-05, $0.2\mu\text{L/h}$ to 500 mL/h , accuracy of 0.5%) system, as shown in Figure 4.2. The same volumetric flow rates of the two inlet flows were ensured by using two identical syringes driven by the syringe pump.

Fluorescent dye ($\text{C}_{20}\text{H}_{10}\text{Na}_2\text{O}_5$, also called Acid Yellow 73) was added in the NaCl solution for the purpose of images collection. When the fluorescein was illuminated by a mercury lamp, an epi-fluorescent attachment of type Nikon B-2A was used (excitation filter for 450-490 nm, dichroic mirror for 505 nm and an emission filter for 520 nm) to take the measurement. After recording the images, the interface location between the two fluids was determined by a customized algorithm written in MATLAB program [27]. The algorithm removes the noise in the collected images, and then determines the dimensionless pixel intensity across the channel. It is assumed that the proportional relationship exists between the light intensity and the concentration of the fluorescent dye. The interface position between the two fluids can be determined from the evaluation of the concentration gradient. The typical images taken in the experiment and the concentration profiles are shown in Figure 4.4.

The parameters studied in the experiments are the inlets volumetric flow rates, the electric voltage applied between A and B and the viscosity ratio β . For different operating conditions, the measurement was taken when the flows was stabilized. The holdup of the NaCl solution, e_1 , was obtained by normalizing its width to the whole channel width, given as $\frac{w_1}{w_1 + w_2}$. Similarly the holdup of the aqueous glycerol is $e_2 = 1 - e_1$.

Chapter 4 Two-fluid stratified flow under pressure and electroosmosis: experiment

Figure 4.4: Measurement results at a flow rate of 0.6 mL/h: (a) no electric field, original image; (b) under -0.8 kV, original image; (c) the normalized light intensity image for 0 kV; (d) the normalized light intensity image for -0.8 kV; (e) normalized concentration distribution of the fluorescent dye across channel width under different applied voltages, from which NaCl solution holdup is about 35% for 0 kV applied voltage, 47% for -0.8 kV applied voltage.

Chapter 4 Two-fluid stratified flow under pressure and electroosmosis: experiment

Effect of electric field

As shown in Figure 4.5(a), as the electric field changes in magnitude and direction, the holdup of NaCl solution changes accordingly. When no voltage is applied across A and B, the flow is simply a pressure-driven two-fluid flow. As the aqueous glycerol is more viscous than the NaCl solution by about 1.5 times, the less viscous NaCl solution occupies a smaller portion of the channel and it has holdup of 0.35 when no externally applied voltage is applied, as shown in Figure 4.5(a). When a negative electric field is applied across A and B, the holdup of the NaCl solution increases. The electroosmotic flow is directed against the pressure-driven flow under a negative electric field. Hence more resistance force will be encountered to drive the NaCl solution. With the effect of electroosmosis effect, the "apparent viscosity" of NaCl solution increases and thus occupies a larger channel width. The holdup of the NaCl solution increases with the increase in the negative electric field. Due to the same pressure drop across E and F, in order to achieve the same volumetric flow rates, the more viscous fluid has to spread to a larger width, i.e. a higher liquid holdup. In another case where the positive electric field is applied, the NaCl solution has apparently a smaller "viscosity", since the electroosmotic flow is in the same direction with the pressure-driven flow. In another words, electroosmotic flow aids the flow of the NaCl solution. Figure 4.5a also shows that, as the inlet volumetric flow rates of the two fluids increase, the electroosmotic flow effect on the pressure-driven flow becomes weaker. At the flow rate of 1.2 mL/h, it seems that the holdup of NaCl, e_1 , remains constant though the voltage varies from -0.8 kV to +0.6 kV.

For typical electroosmotic flows, in which hundreds of volts per centimeter of the electric field is applied, the resulted velocity is of order 0.1 to a few mm/s [33]. But for the pressure-driven flow in microchannel, the velocity can be controlled in a wider range. In this experiment, when the flow rate is at 0.4 mL/h, the average velocity of NaCl through the channel with no externally applied electric field is 3.17 mm/s, the

Chapter 4 Two-fluid stratified flow under pressure and electroosmosis: experiment

Figure 4.5: Relationship between NaCl solution holdup and (a) different applied voltage for the same volumetric flow rates of the two fluids or (b) volumetric flow rate under the same applied voltage. (The viscosity ratio between the two fluids is $\beta = 1.5$)

magnitude of which is comparable with that from the electroosmotic flow. Figure 4.5a shows that by adjusting the magnitude and direction of electric field the interface position between the two fluids, i.e. the variation of the NaCl holdup from 0.25 to 0.50, has been successfully controlled. When the flow rate is set at 1.2 mL/h, the average velocity due to pressure gradient is about 9.52 mm/s for NaCl solution. The pressure driven flow and the electroosmotic velocities are of different orders, which makes EOF effect less significant.

Chapter 4 Two-fluid stratified flow under pressure and electroosmosis: experiment

Effect of flow rate

The relationship between the NaCl holdup e_1 at different flow rates under a fixed electric field is shown in Figure 4.5(b). Holdup remains the same (0.35) for different volumetric flow rates in the absence of an externally applied electric field. This because the volumetric flow rates ratio between the two fluids is kept unchanged, 1:1. This agrees very well with a previous theoretical and numerical study reported in literature [84]. Figure 4.5(b) shows that as flow rate increases, holdup converges to a constant value $e_1 = 0.35$, which is the value without the externally applied electric field. The reason is that the higher pressure-driven velocity makes the electroosmosis effect almost insignificant.

Effect of viscosity ratio, β

Figure 4.6: The holdup of NaCl, e_1 , for different β value, (channel size = $910\mu\text{m} \times 50\mu\text{m}$)

To investigate the effect of viscosity ratio ($\beta = \mu_2/\mu_1$), aqueous glycerol solution of two different viscosity, were used in the experiment. The values of the viscosity ratio

Chapter 4 Two-fluid stratified flow under pressure and electroosmosis: experiment

Figure 4.7: The holdup of NaCl, e_1 , for different β value, (channel size = $910\mu\text{m} \times 100\mu\text{m}$)

β are 1.5 and 2 respectively. The interface position for different β values is shown in Figure 4.6, in which the channel used has the cross section $910\mu\text{m} \times 100\mu\text{m}$. From Figure 4.6, it is seen that for the case of pressure driven two fluid flow, i.e in the absence of applied electric field, the holdup of NaCl, e_1 , is about 0.35 and 0.23 for $\beta = 1.5$ and $\beta = 2$ respectively. The holdup of the NaCl solution decreases with the increase of the viscosity ratio. This is because the more viscous aqueous glycerol spreads to a larger width. By comparing the slope of the interface position (NaCl holdup) change with respect to the voltage applied, we can see the gradient of $\beta = 1.5$ is steeper than that of $\beta = 2$. The interface position of the two fluids depend on flow rate, viscosity ratio and the applied electric field. For a given applied electric field, the difference between the "apparent viscosity" μ_{app} and the actual viscosity is $\Delta\mu_1 = \mu_{app} - \mu_1$. The relative change of viscosity μ_1 with respect to the fluid 2, i.e. glycerol, is $\frac{\Delta\mu_1}{\mu_2} = \frac{\Delta\mu_1}{\beta\mu_1}$. Thus for a larger β value, the relative change of μ_1 will be smaller. The interface position can

Chapter 4 Two-fluid stratified flow under pressure and electroosmosis: experiment

be adjusted more effectively for the two fluids when they have comparable viscosities, i.e. β close to 1. In Figure 4.7, the holdup e_1 changes with respect to applied voltage is shown, in which channel has cross section of $910\mu\text{m} \times 100\mu\text{m}$. At the flow rate of 0.8 mL/h, there seems no holdup change, though the applied voltage varies from 0 till -0.8 kV for $\beta = 2$. But for $\beta = 1.5$, the holdup still changes at the flow rate 0.8 mL/h. That means, for smaller viscosity ratio, the electroosmosis effect can still be effective to control the holdup for a larger flow rate range.

Summary

Through the parameters investigated, it is found that

- The larger the applied electric field, the more the holdup changes.
- The larger the flow rate, the less significant the EOF effect is.
- The larger the viscosity ratio, the less the EOF effect is.

4.3.2 Micro-PIV measurement

In our experiments, the seeding particles used were Duke red particles (Duke scientific Co.). The particles have a maximum excitation wavelength of 540 nm (green, very close to the the characteristic wavelength of Nd: YAG) and a maximum emission wavelength of 610 nm (red). The diameter of the particles can be chosen from several hundreds of nanometers to several microns. The PIV-measurement uses an epi-fluorescent attachment of type Nikon G-2E/C (excitation filter for 540 nm, dichroic mirror for 565 nm and an emission filter for 605 nm). Both filters in the attachment have a bandwidth of 25 nm.

Chapter 4 Two-fluid stratified flow under pressure and electroosmosis: experiment

Figure 4.8: Micro-PIV measurements for 0.1 ml/h flow rate under different electric fields, (a) in the absence of external electric field: (a1) PIV image, (a2) velocity vector; (b) -1.0 kV voltage electric field: (b1) PIV image, (b2) velocity vector; (c) the streamwise velocity profile over the channel width.

Chapter 4 Two-fluid stratified flow under pressure and electroosmosis: experiment

The measurement reported in this experiment was carried out with a $4\times$ objective lens. With a CCD sensor size of $6.3\text{ mm} \times 4.8\text{ mm}$, the size of an image pixel is $2.475\text{ }\mu\text{m}$ and the size of the measured area is $1584\text{ }\mu\text{m} \times 1188\text{ }\mu\text{m}$. Fluorescent particles with a diameter of $3\text{ }\mu\text{m}$ were used to trace the flow. A microchannel with the cross section of $910\text{ }\mu\text{m} \times 50\text{ }\mu\text{m}$ and the length of 5 mm was used. The fluids used in the experiment were the aqueous NaCl solution (concentration 10^{-4} M) and aqueous glycerol (volume concentration 24%). The viscosity ratio between the two fluids is $\beta = 2$. The fluorescent particles were diluted in the two fluids. Syringe pump was used to pump the two fluids through the two inlets, while electric field was applied across the electrodes on the NaCl solution side.

Two 30 mJ laser pulses with a delay time of 3.5 mS were used as illumination sources. The integration area is $32\text{ pixels} \times 32\text{ pixels}$. Previous studies show that the entry length of two fluids flow in microchannel was very short [17, 27]. The measurement was taken at 1 mm downstream of the entrance, thus the stable velocity field was obtained. Figure 4.8 shows two typical results of the micro-PIV measurements. The original images are shown in (a1) and (b1). The respective vector plots of the velocity field are shown in (a2) and (b2). The streamwise velocities across the channel are shown in Figure 4.8(c). The operating conditions under which measurement was taken are both inlet flow rates at 0.1 ml/h and no applied voltage for (a1). For (b1), the flow rates are 0.1 ml/h for both inlets, and applied voltage is -1.0 kV .

Effect of electric field

The velocity profiles under different electric fields are shown in Figure 4.9. Without externally applied voltage, the NaCl (which is on the left side in the figure) flows faster. When negative electric field is present, the velocity of NaCl decreases, whereas the velocity of aqueous glycerol increases. The NaCl and aqueous glycerol have about the same velocity when the applied voltage is -0.4 kV . That means, the two fluid have the

Chapter 4 Two-fluid stratified flow under pressure and electroosmosis: experiment

same average residence time. If the applied voltage continues to increase to -0.8 kV, the NaCl flows at smaller velocity and spreads more in the channel.

Figure 4.9: The velocity profile under different applied voltage, (flow rate= 0.1 mL/h, the left side is NaCl).

Effect of the volumetric flow rate

To investigate the effect of electroosmosis under different flow rates, the dimensionless streamwise velocity profiles are shown in Figure 4.10. The velocity profile for different flow rates, 0.08 mL/h and 0.15 mL/h, are compared under same externally applied electric field. The dimensionless velocity was obtained by normalizing the velocity to the average velocity through the channel. From Figure 4.10(a), in which no electric field was applied, it can be seen that the dimensionless velocity profiles are very similar for both flow rates 0.08 mL/h and 0.15 mL/h. Once the electric field is applied, the velocity profile changes. Figures 4.10(b), (c) and (d) show that the effect of electroosmosis affects the flow velocity more significantly under a lower flow rate condition. This agrees well with the interface position change in the previous section 4.3.1. Under lower flow rate operating condition, the interface position can be adjusted easily.

Chapter 4 Two-fluid stratified flow under pressure and electroosmosis: experiment

Figure 4.10: Comparison of the dimensionless velocity for two different flow rate 0.08 mL/h and 0.15 mL/h, (a) no applied electric voltage, (b) -0.2 kV applied electric voltage, (c) -0.6 kV applied electric voltage, (d) -1.0 kV applied electric voltage.

Chapter 4 Two-fluid stratified flow under pressure and electroosmosis: experiment

Figure 4.11: Comparison between PIV measurements and theoretical analysis at flow rate 0.1 mL/h, under various applied electric voltage. (a) under no applied electric voltage, (b) under -0.2 kV applied electric voltage, (c) under 0.2 kV applied electric voltage, (d) under 0.4 kV applied electric voltage.

4.4 Comparison of PIV measurement and theoretical model

The comparison between theoretical and experimental is shown in Figure 4.11. The flow rate is 0.1 mL/h. The comparison between measurement and theoretical analysis under different electric voltage shows there exist reasonable agreement. In the theoretical prediction, a relatively sharp transverse velocity gradient occurs at the interface as the model assumes two immiscible liquids. In fact, glycerol is miscible in water; therefore, there exists an interfacial region in the measured velocity profiles.

Chapter 5

Flow field inside water droplet: experiment

In this chapter, the experimental measurement of the flow field inside a moving droplet is presented. The droplets are formed by two immiscible liquids, water and oil. The oil acts as carrier fluid to break the water stream into droplets. Tracing particles were seeded in the water droplet for micro-PIV measurement. The result show that the outer recirculation flow inside the droplet has the same direction as the droplet motion. This fact is counterintuitive and opposes common speculation that the shear stress on the droplet interface acts in a direction opposite to the droplet motion [50]. The details of the experiment are presented in the following sections.

5.1 Experimental setup

5.1.1 Microfluidic device

The microfluidic device was fabricated by deep reactive ion etching (DRIE) in silicon. The fluidic access was etched through the silicon wafer. A glass wafer was bonded on the etched silicon wafer to seal the microchannel and to act as the optical access to the formed microdroplets. The microfluidic network has a T-junction configuration and consists of a large carrier channel of $150\text{ }\mu\text{m} \times 150\text{ }\mu\text{m}$ cross section and a small injection

Chapter 5 Flow field inside water droplet: experiment

Figure 5.1: The silicon/glass chip and the adapter used in our experiment. The inserted figure shows the concept of droplet formation investigated in our experiments.

channel of $50\ \mu\text{m} \times 150\ \mu\text{m}$ cross section. The aqueous liquid with tracing particles enters through the injection channel, while an immiscible carrier liquid is introduced into the carrier channel. The droplets form at the T-junction. The size of the silicone/glass chip is $10\ \text{mm} \times 10\ \text{mm}$. To interface with the macro world, such as the microscope and micropump. The silicon chip is mounted in a adapter made of Perplex glass. Syringe needles (19G, Terumo) and tubing ($1/16'' \times 1/8''$, Cole-Parmer) were used as fluidic interconnects for the adapter. Figure 5.1 shows the silicon/glass chip and the Perplex adapter. The inserted figure explains the concept of droplet formation at the T-junction.

5.1.2 Materials

In our experiments, oil with a viscosity of $6.52 \times 10^{-2}\ \text{Pa}\cdot\text{s}$ enters the larger carrier channel, while the aqueous liquid with tracing particles (Duke red particles, 930 nm in diameter, Duke scientific Co.) joins through the smaller injection channel. The aqueous liquid is DI-water with a viscosity of approximately $10^{-3}\ \text{Pa}\cdot\text{s}$. The two liquids were delivered by a precision micro-syringe pump (Lomir Biomedical Inc.). A 500- μl syringe and a 5-ml syringe (Hamilton) were used for the aqueous liquid and oil, respectively.

Chapter 5 Flow field inside water droplet: experiment

Since both syringes are driven by the same stepper motor system, the flow rate ratio between the carrier liquid and the aqueous liquid is kept constant at 10:1. The total flow rate mentioned in this report is the sum of both flow rates of oil and the aqueous liquid.

The micro-PIV measurement equipment is the same as described in section 4.3.2. The measurement reported in this experiment was carried out with a 10 $\times$ objective lens. With a CCD sensor size of 6.3 mm $\times$ 4.8 mm, the size of an image pixel is 0.495 μ m and the size of the measured area is 316.8 μ m $\times$ 237.6 μ m. The delay times between two PIV images were selected as shown in Table 5.1.

Flow rate (μ L/h)	55	110	165	220	275	330	440
Delay time (μ sec)	700	400	250	200	150	150	100

Table 5.1: PIV delay times used for different total flow rates.

5.2 Results and discussion

5.2.1 Size and shape of the droplets

The original PIV images of the droplets at different flow rates are shown in Figure 5.2. We can observe as the total flow rate increases, the droplet size decreases. At low flow rate, the droplet length is larger than the channel width, as in Figure 5.2(a)(b)(c), thus the droplet possess a cylindrical plug shape. At high flow rate, the droplet length decreases, and the droplet reduces to a sphere shape, Figure 5.2(f). The droplet length at different flow rates is shown in Figure 5.3. This agrees with the theoretical analysis by Nguyen *et al* [85].

The droplets have different curvatures at the advancing and the receding edges of the droplet. The advancing edge has a larger curvature. At low flow rate, this gives the droplet a bullet-like shape. As the flow rate increases, the curvature difference decreases; the droplet decreases in size and becomes sphere-shaped if the flow rate is high enough.

Chapter 5 Flow field inside water droplet: experiment

Figure 5.2: Original PIV images of the droplets at different flow rates (the droplets flow from right to left). (a) 55 $\mu\text{L/h}$, (b) 165 $\mu\text{L/h}$, (c) 220 $\mu\text{L/h}$, (d) 275 $\mu\text{L/h}$, (e) 330 $\mu\text{L/h}$, (f) 440 $\mu\text{L/h}$.

Chapter 5 Flow field inside water droplet: experiment

Figure 5.3: The droplet length at different flow rates.

5.2.2 Velocity field inside the droplets

After recording the PIV images, we used commercial software PIVview1.7 (PIVTEC GmbH, Germany) to evaluate the velocity fields of the particles. The interrogation algorithm used is multiple-pass interrogation (number of pass is 4) with an interrogation window of $64 \text{ pixels} \times 64 \text{ pixels}$. The measurement of velocity was taken at the middle plane of the droplet, Figure 5.4. The velocity at the middle line of the droplet is shown in Figure 5.5. Clearly seen, the velocity at the edge of droplet moves at a larger velocity than that at the center.

Subsequently, the droplet velocity vector was calculated by averaging the particle velocity over the droplet area. Subtracting the droplet velocity vector from the Eulerian particle velocity field results in the Lagrangian velocity field inside the droplet. Clear recirculation flow was observed inside the droplet, Figure 5.6.

From Figure 5.6, we can see that the outer recirculation flow inside the droplet has the same direction as the droplet motion. This fact is counterintuitive and opposes the common speculation that the shear stress on the droplet interface acts in a direction opposite to the droplet motion [50]. If the shear stress on the droplet is caused by contact between the droplet and a immobile film of carrier liquid on the wall, the Lagrangian velocity at the interface should have the same magnitude and a opposite direction as the Eulerian velocity vector of the droplet. Measurements at different flow rates reveal the consistent results, that the Lagrangian velocity at the interface is about $1/5$ of the Eulerian droplet velocity and has the same direction, Figure 5.7. The results also show that the Eulerian droplet velocity is slightly less than the mean velocity calculated based on the total flow rate of the two liquids and the channel cross section of $150 \mu\text{m} \times 150 \mu\text{m}$.

The observed Lagrangian velocity field can be explained by the existence of the mobile carrier liquid around the droplet as shown in the inserted figure in Figure 5.7. The plug-shaped droplet does not possess a square shape in cross section to occupy the

Chapter 5 Flow field inside water droplet: experiment

Figure 5.4: Velocity field inside the droplets at different flow rates (the droplets flow from right to left). (a) 55 $\mu\text{L/h}$, (b) 165 $\mu\text{L/h}$, (c) 220 $\mu\text{L/h}$, (d) 275 $\mu\text{L/h}$, (e) 330 $\mu\text{L/h}$, (f) 440 $\mu\text{L/h}$.

Chapter 5 Flow field inside water droplet: experiment

Figure 5.5: The velocity profile at the middle line of the droplet.

Chapter 5 Flow field inside water droplet: experiment

Figure 5.6: Recirculation flow inside the droplets at different flow rates (the droplets flow from right to left). (a) $55 \mu\text{L/h}$, (b) $165 \mu\text{L/h}$, (c) $220 \mu\text{L/h}$, (d) $275 \mu\text{L/h}$, (e) $330 \mu\text{L/h}$, (f) $440 \mu\text{L/h}$.

Chapter 5 Flow field inside water droplet: experiment

Figure 5.7: Velocities of the droplet. The circles ○ are the mean velocity of the droplet evaluated from the micro-PIV measurement. The dashed line — depicts the averaged flow velocity in the microchannel calculated based on the total flow rate. The crosses × are measured Lagrangian velocity at the interface of the droplet.

whole channel. There exist gaps between the microchannel and the droplet at the corner region [86, 87]. We believe that the carrier liquid flows at a slight higher velocity than the droplet. However, due to the reduction in cross section area of the carrier liquid flow (about $\frac{4-\pi}{4}$ for the case of circular cross section), from a square to the hatched region shown in Figure 5.7, the mobile carrier liquid flows at a much higher local velocity through these gaps. These mobile carrier liquid flows at a velocity higher than the Eulerian droplet velocity, causing shear stress in the same direction at the droplet interface.

5.2.3 Velocity field during droplet formation

The velocity field inside the droplets at the formation stage were obtained using micro-PIV technique, Figure 5.8. At the earlier stage, there exists strong swirling flow inside the droplet, and the velocity magnitude is smaller; at later stage, the flow recirculation is weak and the velocity magnitude increases.

Chapter 5 Flow field inside water droplet: experiment

Figure 5.8: The velocity fields inside the water droplet during the droplet formation stage, (a) at $t = 0$ second, (b) at $t = 0.1$ second, (c) at $t = 0.2$ second, (d) at $t = 0.3$ second, (e) at $t = 0.4$ second, (f) at $t = 0.5$ second.

5.3 Summary

In summary, we have presented a the measurement of the Lagrangian velocity field inside a droplet. The droplet was formed in a microchannel network fabricated in silicon and glass using micro-technology. The measurement method is based on micro-PIV. Lagrangian velocity field was extracted from the measured Eulerian velocity field of tracing particles. The experimental results reveal that the recirculation flow at the droplet interface has the same direction as the droplet motion. This effect can be explained by the mobile carrier flow around the droplet through the gaps between the droplet and the microchannel wall.

Chapter 6

Mixing of droplets

From the experimental investigations presented in the previous chapter, we see the recirculation flow field inside the moving droplet. This recirculation flow contributes to the chaotic mixing inside the droplet. In this chapter, the mixing between two droplets during the coalescence is presented.

6.1 Concept

To study the mixing during the process of coalesce of two water droplet, another microfluidic device has been designed. The schematic of this device is shown in Figure 6.1. Two aqueous droplets, one being pure water while the other being water with dye, form and meet at the downstream junction. Once the two droplets merge to a big droplet, the dye concentration field is measured at different time instances to study the mixing.

6.2 Experiment and results

The microfluidic devices used in our experiment was based on the polymeric lamination technique, shown in Figure 6.2. This technique provides a fast way of prototyping. Polymer sheets commercially available as lamination porches are cut using a CO₂ laser machining machine. The channels and other structures was designed on a CAD program

Chapter 6 Mixing of droplets

Figure 6.1: Schematic of the microfluidic device for studying the coalesce and mixing of two droplets.

Figure 6.2: The fabricated device used in our experiment.

and transferred on the polymer sheets by a laser cutting machine. We used the Universal M-300 Laser Platform (Universal Laser Systems Inc.) with a maximum CO₂-laser power of 25 Watt and a maximum beam speed of about 640 mm/s. The lamination process was carried out using a commercial hot laminator (Aurora LM-450HC, laminating temperature of 120 °C, laminating speed of 0.3 m/min, maximum laminating thickness of 600 μm). The microchannel has a cross section of 100 $\mu\text{m} \times 600\mu\text{m}$ was used. Tubing connections are made to provide interface to a syringe pump (Cole-Parmer 74900-05, 0.2 $\mu\text{l/hr}$ to 500 ml/hr, accuracy of 0.5%).

Chapter 6 Mixing of droplets

Fluorescent dye (fluorescein disodium salt $C_{20}H_{10}Na_2O_5$, also called Acid Yellow 73) was added into one of the water streams to form a droplet with fluorescent dye. The flow rate ratio between the oil, DI water and DI water with dye is 20:1:1. The pure DI water droplet and water droplet with dye travel along the microfluidic network downstream, as shown in Figure 6.1.

The coalescence of the droplets was observed and recorded as video file for later analysis. When the fluorescein was illuminated by a mercury lamp, CCD camera (Nikon B-2A) was used to take the measurement. A program written in MATLAB was used to evaluate the light intensity distribution [27]. A proportional relationship is assumed between the light intensity and the concentration of the fluorescent dye. The coalescence and the concentration distribution across the droplets are shown in Figure 6.3. Once the two droplets merge, the front droplet is slowed down. Complete mixing can be achieved in 1/6-second time, (5 frames' time, video frequency is 30 Hz).

Chapter 6 Mixing of droplets

Figure 6.3: Concentration field of the dye during the coalescence of two droplets (channel size = $100\mu\text{m} \times 600\mu\text{m}$), total flow rate = $330\mu\text{L/h}$, flow rate ratio = 20:1:1.

Chapter 7

Conclusion and Future Works

7.1 Conclusion

The characteristics of the two-fluid flows in microchannel are studied theoretically and experimentally. A mathematical model was formulated based on the Poisson-Boltzmann equation and Navier-Stokes equation. Various parameters, viscosity ratio, and zeta potential, which affect the fluid flow behavior were investigated.

The experimental study utilized the fluorescein image techniques to investigate the liquid holdup, i.e. interface location between the two fluids, under the electroosmosis effect. It is noted that: the larger the applied electric field, the larger shift the interface position; the larger the flow rate, the less significant is the electroosmosis effect; the larger the viscosity ratio, the less is the electroosmosis effect. The experiment demonstrated an alternative method to control the interface location, rather than the conventional 'flow-rate-ratio' method. Without changing the flow rate, the interface location between the two fluids was successfully controlled. This could be useful in the application of multiple laminar flow streams in microchannel, which is often used in the biological separation, extraction and detection process.

The velocity field of the two fluids was quantitatively studied by the micro-PIV technique. The change of velocity fields indeed reflects the interface location shift. The experimental results were compared to that of the theoretical analysis. There exists good

Chapter 7 Conclusion and Future Works

agreement between them.

In the second part of the project, the flow field inside a moving droplet in microchannel has been studied. The experimental results reveal that the recirculation flow at the droplet interface has the same direction as the droplet motion. This effect can be explained by a mobile carrier flow around the droplet through the gaps between the droplet and the microchannel wall. By knowing the recirculation flow field inside a droplet, the mixing between two droplets during coalescence was investigated using fluorescence imaging technique.

7.2 Future works

It is shown in the experiment that the interface location can be controlled through the applied electric field. This could be used to realize flow switching in microfluidic platform to develop microfluidic flow switch. In the future work, it is recommended to develop microfluidic device based on this principle for biological application, such as diffusion-based bio-analysis.

The flow field inside a droplet is not only important to understand the mixing but also of interest to the physics involved. In the future work, it is recommended to study the underlying physics of the flow field inside the droplet, using analytical method or numerical method.

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