Mixed electroosmotic-pressure driven multi-fluid flow in microchannels

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Mixed Electroosmotic-Pressure Driven Multi-fluid Flow in Microchannels

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Abstract

Microfluidic and Lab-on-a-chip devices have attracted great interest over the last decade due to the advantages such as increased efficiency, throughput, portability, reduced analysis time, reagent consumption and cost. Comparing with a number of techniques (e.g., pressure, centrifuge, and thermal gradient driven flow), electroosmosis has been used to induce electroosmotic flow to transport, separate, and mix samples. This thesis addresses the following four topics: (1) Three-fluid flow in straight microchannels based on the combined effect of hydrodynamic and electroosmosis with planar interface; (2) Modeling of tunable optofluidic lens based on the combined effect of hydrodynamic and electroosmosis; (3) Semi-analytical model of mixed electroosmotic/pressure-driven two immiscible fluids with curved interface; (4) The linear stability of two-fluid flow in a microchannel and a parallel electric field.

Firstly, multi-fluid flow in straight microchannels is analytically and experimentally investigated. The analytical part presents two mathematical models (steady model and transient model) to describe a three-fluid electroosmotic focusing/pumping techniques, in which a non-conducting fluid is focused and delivered by the combined interfacial viscous force of two conducting fluids and pressure gradient. The two conducting fluids are driven by electroosmosis and pressure gradient. The electric potential in the two conducting fluids and the velocity distribution of the steady and transient three-fluid electroosmotic stratified flow in a rectangular microchannel are presented by assuming planar interfaces between the three immiscible fluids. Analytical solutions are obtained by solving the linear Poisson-Boltzmann equation and the Navier-Stokes equation together. Due to the presence of the surface free charge densities at the liquid-liquid interfaces, the bulk electroosmotic forces in the diffuse layers and the forces exerted at the interfaces are accounted in this model. The velocity profiles and the flowrates are obtained analytically by the methods of Laplace transformation and separation of variable. The effects of viscosity ratio, electroosmosis and pressure gradient on velocity profile and flowrate are analyzed to show the potential feasibility of this technique. The experiments are completed in Polymethylmethacrylate (PMMA) microchannel. Experimental results show that the width and location of the sample flow can be controlled accurately using the electroosmotic effect, rather than the conventional “flow-rate-ratio” method. Under the fixed flowrates, the sample flow can be delivered to the desired outlet ports by electroosmosis. The electroosmotic effect under

different flowrates, different viscosity of the non-conducting fluid, and different aspect ratio were investigated. The comparison between the measured data (width of the non-conducting fluid and the location of the sample stream) and the analytical models show good agreement.

Secondly, modeling of tunable optofluidic lens based on the combined effect of hydrodynamic and electroosmosis are presented. A liquid-core liquid-cladding optofluidic lens is formed under the combined effect of hydrodynamic and electroosmosis in a circular micro chamber. In order for the lens to be tuned by a voltage, the cladding fluids are electrically conducting, while the core fluid is non-conducting. Under constant flowrates, mathematical models of two-dimensional dipole flow in a circular bounded domain and electric field outside the parallel-plate capacitor are used to predict the curvature of the interface. A test device with a circular chamber is fabricated in PMMA using thermal bonding method. Under the same inlet volumetric flowrates, the applied voltages are varied to tune the curvature of the interfaces between the cladding fluids and the core fluid. The interface shape is measured using the fluorescence imaging technique. The results show that the interfaces between the cladding fluids and the core fluid have optically smooth arc-shape. Under fixed cladding flowrates, the same voltages form symmetric biconvex lens only. Different voltages can form biconvex lens, plano-convex lens, and meniscus lens. The experimental results agree well with the analytical model.

Thirdly, a semi-analytical model is developed for predicting the two-fluid flow with a curved interface in rectangular stratified microchannels under the combined effect of hydrodynamic and electroosmosis. With the non-slip boundary conditions at the wall and the matching condition at the curved interface, the fully developed Navier-Stokes equation and Poisson-Boltzmann equation are solved analytically. The effects of interface shape on the electric potential distribution, electroosmotic velocity distribution, and flowrates are discussed; the results show that the interface shape influences the two-fluid flow in microchannel significantly.

Lastly, the electrohydrodynamic and stress instability of the interface between two viscous fluids with different electric properties under a parallel electric field in a microchannel is analytically and experimentally investigated. The linear Poisson-Boltzmann equation are solved using the separation of variable method for densities of bulk charge and surface charge; the electric field and fluid dynamic are coupled only at the interface through the stress balance equations. In the experiments, two immiscible fluids,

aqueous NaHCO_3 (conducting fluid) and silicone oil (non-conducting fluid) are pumped into a PMMA microchannel. The parallel electric field is added to the aqueous NaHCO_3 using a high voltage power supply. The results are recorded using a Charge Coupled Device (CCD) camera. The results show that the electric field can have either destabilizing or stabilizing effect depending on the ratio of viscosity of the two fluids. The flowrates, zeta potential, and dimension of the microchannel affect the growth rate of the perturbation. Both experimental and analytical results show good agreement.

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List of Symbols

c	Fluorescent concentration
D	The dielectric displacement
e	Elementary charge, $e = 1.602 \times 10^{-9}, [C]$
e_1, e_2, e_3	Liquid fractions
E	The electric field
E_s	The stream potential
$\mathbf{F}$	The electroosmotic body force
$\mathbf{G}$	Parameter measuring the electroosmotic force by external electric field
i	The imaginary unit
I	Intensity values
$I_\lambda(Kr)$	The modified Bessel functions of order λ of the first kind
$\mathbf{J}$	The transport flux
h	Height of the microchannel, [m]
$L_{microchannel}$	The total length of microchannel
L_{ref}	The length scale
M	Electrokinetic effect in the matching conditions
$\mathbf{n}$	The unit normal vector
n_0	Ionic number concentration in the bulk, $[m^{-3}]$
n_i	Ionic number concentration of the type-i in the bulk, $[m^{-3}]$
k_b	Boltzmann constant, $k = 1.381 \times 10^{-23} [JK^{-1}]$
K	Electrokinetic parameter
p	Pressure
Δp	Pressure gradient
q	Flowrate
Q	Laplace transform flowrate
r	The radius in the polar coordinate
r_p	The particle radius

$\mathbf{r}$	Position vector, [m]
R	The radius of the circular chamber
Re	Reynolds number
s	Laplace transform parameter
t	Time [s]
T	Temperature [K]
u	Velocity
U	Laplace transform velocity
U_{ref}	The velocity scale
v	Control volume
$\mathbf{v}$	The velocity vector
w	Half of width of the channel [m]
z_i	The valency of ion i

Greek symbols

α	Kinematic viscosity ratios
β	Dynamic viscosity ratios
χ	Aspect ratio
λ_b	The bulk conductivity
λ_D	The Debye length [m]
λ_s	The surface conductivity
ε	The permittivity of the dielectric
ε_0	The permittivity of the dielectric of vacuum
ε_r	The dielectric constant comparing with ε_0
ψ	The electric potential
ψ_0	The electric potential of surface
ϕ	The velocity potential
μ	Dynamic viscosity
μ_i	Chemical potential of ion i
$\bar{\mu}_i$	Electrochemical potential of ion i

ι	Any property in bulk
i	Volumetric production of the property in bulk
κ	Debye-Hückel parameter $[\text{m}^{-1}]$
τ	Shear stress
ν_E	The mobility of particles
ρ	The density
ρ_e	The net charge density
ρ_q^s	The surface charge density
ψ	Potential
ψ_0	Electrostatic potential
ξ	Zeta potential

Subscripts

ref	Reference quantity
1	Conducting fluid 1
2	Non-conducting fluid 2
3	Conducting fluid 3
q	Charge of bulk
w	Wall
wall	Surface of microchannel
max	The maximum value
min	The minimum value

Superscripts

0	Fundamental state
—	Dimensionless parameter
s	Surface or interface
p	Pressure driven
E	Electroosmotic effect
'	Derivative
I	Integrate
*	Dimensionless parameter

Operators

∇	Gradient
d	Ordinary differential
∂	Partial differential

Chapter 1 Introduction

1.1 Background and motivation

Micro electromechanical systems (MEMS) technologies, which can be adopted to fabricate miniature sensors, actuators and other systems, have been available since 1980s. MEMS technology is an enabling technology in various fields. For instance, this technology has been revolutionizing fluid flow manipulation in microscale. These microfluidic systems offer significant advantages comparing to their macro-scale counterparts. These devices are ideal for handling costly, difficult-to-obtain samples, reagents in medical and bio-analyze due to its small dimensions. A typical microfluidic structure requires sample and reagent of 100 nl and 10 nl. MEMS sensors for pressure, velocity, temperature and shear stress are at least one order of magnitude smaller than conventional sensors. The spatial and temporal resolutions of the measurement are improved by their small size, typically few microns and few microseconds, respectively. Because of the extreme miniaturization and low energy consumption, micro sensors are the ideal sensors for monitoring the flow state without appreciably affecting it.

The interface and the width of a focused stream are important for sorting, separation, reaction, mixing and other applications. Controlling the interface leads to controlling the stream width, this is useful for focusing sample fluid or flow switching. The flow focusing technique provides a particularly effective means of controlling the passage of chemical reagents or bio-samples in a microchannel network, and has been successfully demonstrated in a wide variety of applications such as cell handling and analysis [1], biomedical and biopower systems [2], clinical diagnosis [3], immunoassays [4], DNA [5], proteins [6], other bioassays [7], environmental hazards and gas analysis [8, 9].

Hydrodynamic and electroosmotic focusing techniques are two popular flow focusing techniques. Stiles et al. [10] proposed a simple method to focus the sample stream by using either a single suction pump or a capillary pump effect. The focused stream width was controlled by varying the relative resistances of the side and inlet channel flows. This work also used an analogous circuit to derive an expression for the relationship between the focused stream width and the length ratio of the inlet channel to

the side channel in microchannels with a Hele-Shaw limit. Precise control of the focused sample stream width is crucial for different applications.

Comparing with other controlling methods, the electroosmotic concept has many advantages. For example, the designs of pressure-driven flow require a higher flowrate ratio between the sheath and sample fluids to move the interface location or switch the sample fluid. Electroosmotic flow has a wide range of applications as it has a uniform velocity profile and no moving part. In microsystems, surface-to-volume ratio is large; electroosmosis will be more efficient than the ordinary pressure-driven flow. But it is limited to polar solvents and may lead to sample damage due to Joule heating under certain conditions.

Models of stratified flow are needed for predicting the flow characteristics, such as pressure drop and in-situ holdup, and are often used as a starting point in modeling flow pattern transitions. The common assumption is that the interface separating the phases is planar [11-14]. Comparing with macro and mini channels, the effect of interface curvature in microchannels is very significant but little work has been reported in this area. In microchannel, the gravity effect is low [15-19] while the surface tension is strong, causing the Bond number to approach to zero.

In microfluidic devices, the interfacial instability can produce a variety of flow patterns or even develop into a turbulent motion. The interfacial instability of the liquid flows can enhance mixing. In the microchannel, viscosity dominates and mixing by natural inertia driven turbulence is impossible; various groups explored the possibility of using this instability for efficient and non-mechanical micro-mixing [20-22]. On the other hand, stable flow is a crucial factor during pumping or switching of the immiscible liquid [13, 23-25]. The effects of parameters to the interfacial instability are necessary to control flow pattern effectively.

After a broad review of the currently available literature, most of the previous studies focused on flow focusing and mixing either by pressure-driven, or electroosmosis independently. There is no investigation on the coupled effect between pressure-driven and electroosmosis for multi-fluid flow with planar or curved interfaces in straight or circular chamber microchannels under stable or unstable condition.

1.2 Objective and scope

The objective of this report is to investigate mixed electroosmotic and pressure-driven multi-fluid flow in microchannels. The scopes of the proposed project are

(1) To study and characterize the three-fluid stratified pressure-driven flow with electroosmotic effect, including steady state and transient state, especially, the interfacial balance conditions, which provides a benchmark for further study.

(2) To control the width and position of the focused fluid, control the location of interfaces, and switching a non-conducting sample stream or droplets to the desired outlets by changing the electric field and pressure gradient.

(3) To provide a new design of a circular chamber to realize an optically smooth lens curvature; the curvature can be adjusted using the combined effect of pressure driven and electroosmosis. This is a model of tunable optofluidic lens.

(4) To study and characterize the two-fluid laminar-laminar stratified flow under the combined effect of pressure-driven and electroosmosis where the interface shapes are circular arcs with constant curvature.

(5) To study the effect of parallel electric field on the instability of two viscous fluids under the constant flowrates in microchannels.

1.3 Outline of the report

There are eight chapters in this thesis:

Chapter 1 gives an introduction to the system of the multi-fluid pressure-driven flow with electroosmotic effect and a description of the objective and the scope of this report.

In chapter 2, literature review is provided to focus on the two or multi-fluid flow in microchannels, interface control, switching technique, tunable microlens, multi-fluid flow in microchannels with curved interface, electrohydrodynamic instability of interface in microchannels and fabrication techniques.

In chapter 3, the governing equations about electric potential, momentum equations and continuity equations are introduced. The dimensionless equations are also formulated in this chapter.

Chapter 4 theoretically and experimentally investigated the electroosmotic effect in three-fluid pressure-driven flow. In the theoretical part, this report solves the Poisson-Boltzmann equation and the Navier-Stokes equations for the three-fluid pressure-driven flow with the electroosmotic effect. The methods of separation of variables and Laplace transformation are used to solve the steady state equations and the transient equations, respectively. The results provide insight into the characteristics of such flow. In the experimental part, the applications of electroosmotic flow, including control the interface location and the width, and switching a non-conducting sample stream or droplets are presented.

Chapter 5 presents the modeling and experimental results of three-fluid pressure-driven flow in a circular micro chamber with the electroosmotic effect.

In Chapter 6, the characteristics of two-fluid flow in microchannel under the combined effect of pressure-driven and electroosmosis with curved interface are studied.

Chapter 7, the electrohydrodynamic instability of two-fluid flow in microchannels for parallel electric field is analyzed using the linear perturbation theory. The effects of flow conditions and fluid properties to the instability of the flow are investigated.

Finally, Chapter 8 presents the main conclusions from this study and makes recommendation for future work.

Chapter 2 Literature review

This chapter reviews the related literature. The first part covers the fundamental aspect of the multi-fluid flow, including the background of the electric double layer (EDL), electrokinetic phenomena in microchannels. The following part covers the application of electroosmotic flow, e.g. interface control, flow switching, and tunable microlens. In the third part of this chapter, the electrohydrodynamic instability of the interface in microchannels under normal and parallel electric field is reviewed. Lastly, the manufacturing method of microchannels is reviewed.

2.1 Electrokinetic

2.1.1 Electric double layer

Stern combined the Helmholtz and the Gouy-Chapman models [26, 27] and introduced the electric double layer (EDL) first time in 1924. Generally, if substances contact with a polar medium, under the effect of electrostatics, surface electric charges are acquired around most surfaces of substances. Ionization, ion adsorption, and ion dissolution are the charging mechanisms. The distribution of nearby ions in the electrolyte solution will be influenced by the effect of any charge surfaces; it is shown in Fig. 2.1. The surface attracts the ions of opposite charge, and repels the ions with same charge at the same time. Because of the interaction of ions with different charges, the counter-ions concentration near the charged surface is higher than that in the bulk liquid far away from the surface charge. Under steady state, the distribution of ions by rearrangement is called the electric double layer (EDL). The electric double layer (EDL) closes to the charge surface, in this region, counter ions are preferentially distributed in the aqueous medium. It is divided into the compact layer (stern layer) and the diffuse double layer. Generally, the compact layer is about 0.5 nm thick. In the compact layer, the counter ions are strongly attracted by the charges of the wall surface. In this layer, the ions are immobile. In the diffuse double layer, the ions are less attracted by the charges of the wall and the ions are mobile. In this layer, the ions spread out as a result of thermal energy. The thickness of the diffuse layer ranges from several to a few hundreds of

nanometers, depending upon the bulk ionic concentration, pH value, the properties of the surface, and the other properties of the liquid.

Between the compact layer and diffusion layer, there is an assumed plane, it is called stern plane. The potential (ψ) at this plane is close to the electrokinetic potential or zeta potential (ξ). The zeta potential is defined as the potential at the shear plane between the electrolyte solution and the charged surface. The shear surface, also named shear plane, closes to the solid surface, but the fluid between this surface and the wall is stationary. Because of this, it can be characterized as the fluid flow past the charged surface. Fig. 2.2 shows the schematic ions distribution and the shear plane when the glass and an aqueous electrolyte liquid contact each other [28].

The net charge density ρ_e is not zero within the diffuse layer. The electric potential distribution at the EDL, particularly in the diffuse layer, is influenced by the net charge density. When an electric field is applied parallel to a charge surface, the electric field will exert a body force on the charge in the diffuse layer. Under the influence of applied electric field, the ions within the diffuse layer will move and pull the surrounding fluid of them, this flow called electroosmotic flow, as shown in Fig. 2.3. Thus, solution injection, mixing or displacing can be achieved by applying an external electric field.

2.1.2 Electrokinetic phenomena

There are four basic types for the electrokinetic effects, they are named as electrophoresis, electroosmosis, streaming potential and sedimentation potential, respectively [26]. Electrophoresis and sedimentation potential describe the phenomena about the charged particles in quiescent liquid. Electroosmosis and streaming potential describe the motion that the flow of fluid past a stationary charged surface. According to the driving force and response, these effects can be classified again. Electrophoresis and electroosmosis involve the particle or fluid flow, respectively, but they need an external electric field. For sedimentation potential or streaming potential, an external pressure gradient is imposed; they can generate an electric potential or current.

When an electric field is applied to one fluid, such as a liquid or gas, and the second phase is suspended as particles of solids or liquids in this fluid, the particles can be induced to move because of the electric field. The movement of these charged colloidal

particles is termed electrophoresis. The sedimentation potential is another electric potential which generated by the forced movement of charged colloidal particles in a liquid due to gravitation. If the solid is in the form of capillary or porous plug and remains stationary, and the liquid moves in this kind of solid because of the external electric field, the movement of the liquid is called electroosmosis. By a pressure gradient, the liquid can be forced through a capillary or porous plug; if there are the excess charges near the wall, these charges will be carried along by the liquid. When they accumulate in the downstream of the liquid, an electric field will be generated. This electric field can introduce an electric current against the direction of the liquid flow. This potential difference, which caused by the accumulation of the charges, is called the streaming potential. The driving pressure and the electric field applied to the neighborhood of the wall are related to this potential.

Zeta potential (ξ) is one of the most important characterization parameters when studying electrokinetic phenomena. It is the potential at the electrokinetic shear plane, and generally, this potential cannot be measured directly. By changing the strategy of measurement, the zeta potential can be measured from electrokinetic measurements [26, 29].

An experimental procedure that monitors the electric current in electroosmotic flow through a capillary, for determining zeta potential and surface conductance was shown by Arulanandam et al. [30]. Huang [31] presented another method to measure the zeta potential by measuring the average velocity based on current monitoring in electroosmotic flow. If the zeta potential is assumed to be small, $\xi < 25\text{mV}$, according to the Debye-Hückel approximation, the average electroosmotic velocity (u^E) can be presented as $u^E = \frac{E\varepsilon\xi}{\mu}$ where E is the external electric field, ε is the permittivity of fluid, and μ is the viscosity of the fluid. Erickson [32] improved the method of current monitoring [30, 31], and used the slope-intercept method to determine the zeta potential and surface conductance. Previously, the method that monitors the electric current in electroosmotic flow through a capillary is the main method to determine the zeta potential and surface conductance.

In the past few years, electrokinetic effects on the viscosity, heat transfer, velocity profile and zeta potential were reported by numerous papers using the methods of analytical modeling, experiment and numerical simulation.

The electrokinetic effects on pressure-driven fluid flow in rectangular microchannels were investigated by Yang et al. [33]. Using a finite-difference scheme, Yang et al. [33] solved the two-dimensional nonlinear Poisson-Boltzmann equation governing the EDL by numerical simulation, and an exact solution of motion equation was obtained by employing the Green function formulation. The potential distribution in an overlapped EDL region cannot be evaluated by the classical theory, because the misuse of the Boltzmann equation and the boundary conditions are being defined. The electric potential and ionic concentration distribution in an overlapped EDL field were determined by Qu et al. [34]. Without using the Boltzmann equation, Qu et al. [34] derived new governing equations for the overlapped EDL fields, its results showed that the modified model predicts a lower electric potential in the EDL field than the classical theory. Using the different models, Dutta [35, 36] presented the electroosmotic transport through rectangular channels with small and large zeta potentials by Taylor-Aris dispersivity, respectively. The results showed that if zeta potential was large enough that there were overlapped EDL fields, the mean velocity and the slug dispersivity can vary as much as an order of magnitude comparing with small zeta potential.

Experimentally, an setup for studying the electrokinetic potential and the surface conductivity of flat surface was developed by Werner et al. [37]. In the setup of Werner et al. [37], the slit channel was formed by varying the distance of the two parallel flat sample surfaces, when different aqueous electrolyte flow through the slit, the zeta potential was calculated. If the distance is sufficient, using the setup of Werner et al. [37], the surface conductivity can be determined from the extrapolation of the channel conductance values.

Electroosmotic flows in complicated geometries were simulated by a numerical scheme which was developed by Patankar et al. [38]. In this work, the electroosmotic injection characteristics of a cross channel device for capillary electrophoresis were studied. The results showed that the injection by high electric field intensities makes the

desired rectangular shape of the sample plug at the intersection of the cross channel to be remained.

In intersecting channels with T-shaped configuration, the steady electroosmotic flow was studied through numerical simulation by Hu et al. [39]. First, the finite difference method was employed to solve the nonlinear Poisson-Boltzmann equation, and then, the modified Navier-Stokes equation was solved numerically by using the SIMPLER algorithm. The results showed that liquid leakage from the floating side channel was affected by the hydrodynamic effects significantly. The length and the location of the channel affect the magnitude of the leakage. The leakage decreased when the length of the side channel increased, or the side channel was moved downstream from the entrance to the outlet of the main channel.

The characteristics of electroosmotic flow in rectangular microchannels was presented by Arulanandam et al. [40]. The electric double layer field and the flow field were described by the two dimensional Poisson-Boltzmann equation and the two dimensional momentum equation, respectively. Arulanandam et al. [40] also investigated the velocity profile of electroosmotic flow in the rectangular microchannel by numerical simulation. Tsao et al. [41] showed the velocity profile when electroosmotic flow through an annulus by solving the Stokes equation, and presented the distribution of electric potential by solving the Poisson-Boltzmann equation.

2.1.3 Measurement of electroosmotic velocity

As the development of electroosmotic flow in microsystems, the accurate measurement of electroosmotic velocity in microsystems is important. The electroosmotic velocity can be predicated by different models and can be measured using direct and indirect methods.

The electroosmotic velocity has been predicated by different methods. Many research works have been published for single conducting fluid electroosmotic flow in cylindrical capillaries [42, 43] slit channels [44], rectangular channels [40, 45], irregular shape microchannel [46] and arbitrary shaped microchannel [47].

Fig. 2.4 shows the model of the current-monitoring method [172]. The same electrolyte solutions with different concentrations, c_1 and c_2 , are contained into

reservoirs 1 and 2 respectively. Note that $c_1 = 70\%c_2$. The two reservoirs are connected using a microchannel. Initially, the solution of concentration c_1 fills with the connecting microchannel. After an electric field is applied along the channel, electroosmotic flow is generated. Gradually, the solution of higher concentration c_2 from the reservoir 2 displaces the solution of lower concentration c_1 causes the flow towards the reservoir 1. The total displacement time $t_{\max}$ can be determined from measurement via the recorded current-time relationship (Fig. 2.4 (b)). The average electroosmotic flow velocity can be calculated using the following equation:

$$u^E = \frac{L_{\text{microchannel}}}{t_{\max}} \quad (2.1)$$

where u^E is the average electroosmotic velocity, $L_{\text{microchannel}}$ is the total length of the connecting microchannel. This indirect method has been developed by a number of researchers [31, 48-50]. The disadvantages of the indirect method are: (1) two solutions with the same electrolyte and different ionic concentration are needed; the difference between ionic concentrations of two solutions must be small to ensure the same measurement conditions; (2) the microchannel must have uniform cross section to determine the average electroosmotic velocity. The advantage of this method is its simplicity.

The direct method visualizes the flow fields using trace amounts of a caged fluorescent dye; the velocity can be calculated by comparing flow fields in different time. The direct method requires a specialized, caged fluorescent dye. The fluorescent dye flows follow the flow field and visualizes it. The image of free fluorescent dye at the different time instance is imaged using classical fluorescence techniques. Velocity data are then calculated. This method can present the whole velocity profiles and the details of velocity. The disadvantage of the direct method is that it requires specialized caged dyes and hardware; the process of measurement is complex. Micro Tagging Velocimetry (MTV) technique is one of the popular direct measurement methods [51, 52]. Micro-scale Particle Image Velocimetry (μ PIV) is an optical, non-intrusive whole-field method which is introduced by Santiago et al. [53]. The principle of measurement for μ PIV is that: particle distributions are recorded at two instant of time. The flow motion is

determined by the change of the particle distribution over time. This method is used to measure velocity fields by a number of researchers [54-57]. Fig. 2.5 shows the setup and principle of μ PIV method.

2.1.4 Measurement of zeta potential

The zeta potential is one important parameter to determine the characteristics of electrickinetic effect [58]. The zeta potential influences a number of applications including microfluidic [38, 59, 60], membrane efficiency [61, 62], blood cells [63], electrokinetic transport of particles [64], and biomedical polymers [65, 66]. The value of zeta potential can be measured indirectly based on one of the three electrokinetic effects: electrophoresis, electroosmosis, and the stream potential [58].

Electrophoresis can be applied to determine the zeta potential by measuring the mobility of the particles (v_E). When the particle is placed in an electric field, the zeta potential of the interface (ξ) can be calculated using the Smoluchowski equation [67]

$$v_E = 4\pi\epsilon_0\epsilon_r \frac{\xi}{6\pi\mu} (1 + \kappa r_p) \quad (2.2)$$

In Eq. (2.2), ϵ_0 and ϵ_r are the electric permittivity of a vacuum and the relative dielectric constant respectively, μ is the solution viscosity, r_p is the particle radius and $\kappa = (2n_0 z^2 e^2 / \epsilon_r \epsilon_0 k_b T)^{1/2}$ is the Debye-Hückel parameter, k_b is Boltzmann constant, T is the absolute temperature, z_0 is the valence of the ions, e is elementary charge, and n_0 is the reference value of the ion concentration. The mobility of particles can be measured using suitable method [68, 69], and then the zeta potential of surface is calculated by Eq. (2.2).

The streaming potential technique is another method to determine the zeta potential. For steady incompressible and laminar flow, a streaming potential can be induced by the downstream convection of ions via pressure-driven flow. The streaming potential (E_s), pressure gradient (Δp) and zeta potential (ξ) has a relationship as follows

$$\frac{E_s}{\Delta p} = \frac{\epsilon_0\epsilon_r\xi}{\mu} \frac{1}{(\lambda_b + 2\lambda_s/h)} \quad (2.3)$$

where λ_b is the bulk conductivity, λ_s is the surface conductivity, h is the channel height. The difficulty of this method is to measure streaming potential and the surface conductivity. It has been used to measure zeta potential of flat surface [70-73], the detailed model of streaming potential technique has been approved by Erickson et al. [74].

Electroosmotic flow has been applied to transport solutions in microchannels [58]. It also can be used to calculate zeta potential using the following equation

$$u^E = \frac{\varepsilon_r \varepsilon_0 \xi}{\mu} E \quad (2.4)$$

where E is the applied electric field strength. The average electroosmotic velocity can be measured by various methods [31, 48-57].

Comparing with electrophoresis and streaming potential techniques, the electroosmotic technique is relatively simple and easy [75].

2.2 Two-fluid and multi-fluid flow in microchannels

2.2.1 Model of pump

Electroosmotic (EO) pump has been widely used to drive conducting fluids and non-conducting fluid in Lab-on-a-chip devices. It can be classified into three types: direct pump, indirect pump, and hybrid pump.

In direct pump, electrodes are in direct contact with a conducting buffer. The electroosmotic force is generated in the conducting buffer, and the conducting buffer is driven by this electroosmotic force. It is called the electroosmotic flow. Direct current (DC) and alternate current (AC) electric field can be used to drive the conducting fluids [28, 76-79].

The DC direct pumps are usually constructed using silica or glass. The electric field is applied along the channel length. When the conducting buffer connects with the wall of the channel, electric double layer (EDL) is formed. The conducting buffer can be pumped according the Helmholtz-Smoluchowski equation [67]. The thickness of the EDL is thinner comparing with the size of the microchannel; and the DC direct micropump can be represented by Poisson-Boltzmann equation [28]. When the dimension of the

microchannel has the same order of magnitude with the thickness of the EDL, the Poisson-Boltzmann equation cannot describe the characteristic of the pump. Hu and Chao [80] developed a model to predicate the characteristic of the pump at this operating region.

Two main problems occur frequently during the operation of the DC direct micropump. First, when the high electric field is added to the conducting fluid, the bubble generates and blocks microchannels due to the gas from the conducting buffer. This problem has been solved by incorporating a bubble-release mechanism in the downstream. Another problem is that the stall pressure of the DC direct pump with a single simple channel is very low to influence the use of the pump. This problem has been solved using various methods [81-85]. Chen et al. [81] designed a electroosmotic (EO) pump based on monolithic silica using a sol-gel process [86-89]; the stall pressure of the pump achieved as high as 400kPa. The packed beads were used to incorporate with surface of microchannel to increase the interfacial area available for electroosmotic flow, and to increase the maximum pressure of the pump [83-90]. Chen et al. [82] designed a novel EO pump with multi-stage. The same voltage was added to each porous silica-packed segment. The stall pressure using this method can achieve 10 MPa.

AC electric field can also be used to pump the conducting fluids using the asymmetric electrodes [91]. The advantage of the AC electric field is that: the operation voltage is very low (< 10 V) [92, 93]. Fig. 2.6 schematically shows the mechanism of AC EO pump. Electrodes A and B are adjacent. When the anode and cathode of AC electric field are added to A and B respectively, the electric fields at the electrode surface have both tangential and normal components. The normal electric field induces EDL and the tangential electric field induces the movement of ions. The fluids can be dragged by the viscosity due to the movement of ions. Capacitive charging and the Faradaic charging are two main methods to polarize the electrodes. The polarization caused by capacitive charging cannot exceed the equilibrium charge density on the electrode side (electrode A), while the Faradaic charging can induce charge density higher than the equilibrium charge density. Capacitive charging and the Faradaic charging can induce two major vortices close to the inner edges (vortices D and E) and two minor vortices at the outer edges (vortices C and F). If the voltages of electrodes A and B are symmetric, the main vortices

D and E are symmetric and there is no net flow. If the electrodes are asymmetric, the main vortices D and E are asymmetric. The suitable electrodes or method can cause main vortices flow in the same direction (vortices D_d and E), and the flow is pumped by AC electric field. Usually, the velocity due to capacitive charging is proportional to V^2 (V is the voltage applied to the electrodes), while the velocity due to the Faradaic charging is proportional to e^V . Capacitive charging is the main mechanism when the applied voltage is low while the Faradaic charging dominates at higher voltage. The detailed mechanism of AC pump is shown by Wu [91]. Urbanski et al. [94] deposited asymmetric metal layer on top of a symmetric array of electrodes to induce asymmetric vortices. Garc ía-S ánchez et al. [95] used traveling-wave to break up the symmetry of the electrodes. The combined effect of AC and DC electric fields induces main vortices flow in the same direction.

Another type of EO pump is indirect EO pump. In contrast to direct contact between electrode and conducting buffer of direct EO pump, in indirect pump, the electrodes are separated by some barriers from the conducting buffer where the EO flow take places. When the indirect EO pump operates, the ions can pass the barriers from surface of electrodes to bulk fluids but the bulk fluids cannot pass through the barriers. Morf et al. [77] used an ion-exchange membrane to allow only positive or negative ions to pass through only.

The last type of EO pump is the hybrid of the direct and indirect EO pump. It is usually achieved by two different fluids. The electrodes contact directly with the high conducting fluids. Hence, electroosmotic flow is induced in the high conducting fluids. The fluids with the low conducting can be dragged by the shear stress of the high conducting fluids. Fig. 2.7 shows the principle of this method. Brask et al.[23, 96] proposed an idea to use an electroosmotic flow (EOF) as a driving mechanism to drag another liquid, the concept was to pump a non-conducting fluid using a conducting fluid by viscous shear stress. By equivalent circuit theory and CFD simulations, Brask et al. [97] analyzed the performance of the pump. Gao et al. [98] presented a mathematical model to describe a two-fluid electroosmotic pump technique, in which an electrically non-conducting fluid is delivered by the interfacial viscous force of a conducting fluid; the latter is driven by electroosmotic flow. Gao et al. [11] solved the complete Poisson-Boltzmann equation and obtained the transient two-fluid electroosmotic flow in

microchannel with electric charges at the interface. Glawdel et al. [99] separated the working fluid and the secondary fluid using oil, and the secondary fluid can be pumped by EO flow of the working fluid. The different methods for separating working fluid and secondary fluid are presents by different researchers [100, 101].

2.2.2 Interface control and property

The fluid-fluid interface is important for separation, reaction and mixing of chemical. If the flows are immiscible, the interface control is useful for focusing sample fluid or flow switching. Comparing with other controlling method, the electroosmosis methods have many advantages [102], so electroosmosis methods on the microchips have been investigated intensively [103] and their effectiveness and usefulness have been demonstrated in many applications [104, 105]. By experiment, Wang et al. [12] showed a concept to control the interface location of a pressure-driven two-fluid flow in a microchannel using electroosmotic effect, adjusting the magnitude and direction of the electric field successfully controlled the interface position between the two liquids.

Gao et al. [106] presented a numerical scheme for stratified two-fluid electroosmotic flow. The simulation results showed that using the electroosmotic effects can control the interface location of a pressure-driven two-fluid flow. By controlling the location of the interface between the identical control and the test channel, Abkarian et al. [107] presented a high-speed microfluidic differential manometer for cellular-scale hydrodynamic by monitoring the interface location. Gao et al. [13] presented theoretical and experimental investigations of the pressure-driven two-fluid flow in microchannels with the DC electroosmotic effect, it showed that adjusting the external electric field could control the fluid interface position precisely.

Manipulating gas and liquids within networks of microchannels is crucial in the design and fabrication of microfluidic devices for applications in bioassays, micro reactors, and chemical and biological sensing [108]. Zhao et al. [109] showed that in microchannels, the maximum pressure of interface between gas and fluid was determined by the surface free energy of the liquid, the advancing contact angle of the liquid on the hydrophobic regions, and the channel depth. Lee et al. [110] presented the first experimental evidence on electroosmotic flow at a liquid-air interface, and the results

showed that the particle velocity at the liquid-air interface was significantly slower than the particle velocity in the bulk. Lee et al. [111] also studied the liquid velocity near the liquid-fluid interface, the experimental results showed that the liquid velocity near the liquid-fluid interface resembles a plug-like profile for both water-oil systems.

2.2.3 Flow switching

The theory that differential pressures between inlets can be used to guide sample into any desired outlets was shown by Blankenstein et al. [112]. Fig. 2.8 shows the experimental results about flow switching. Lee et al. [113] presented a simple theoretical model based on the “flow-rate-ratio” method, this method was proposed to predict the performance of the device. Using volume of fluid model, Reiyu et al. [114] presented a design of microfluidic flow switching. Fu et al. [115] developed a comprehensive theoretical model for flow switching using the electrokinetic force, it was more straight forward and general than the model proposed previously.

A fluidic oscillator employing a V-shaped fluidic circuit with feedback channel was demonstrated by Gebhard et al. [116], in this paper, a part of the output flow was fed back into the inlet region causing the main flow to be redirected to the other channel, then the flow could be switched between two output channels. The experiment of Blankenstein et al. [112] showed that a sample flow stream could be guided to a desired outlet port using hydrodynamic forces. The results showed that the sample fluid can be guided to one of the five outlet which was desired. Using the pre-focusing function, Lee et al. [113] presented experimental result to switch the sample flow to the discretionary outlets. Lee et al. [117] also showed that more than one sample flows can flow into desired outlets at the same time, respectively. Chein et al. [114] showed two specific flow switches, one with a guided fluid flowed to one of five desired outlets, and the other with a guided fluid flowed into one, two, or three outlets equally distributed along the outlet channel of a Y-shaped channel.

Simply applying hydrodynamic properties to guide flow into a desired outlet, Döring et al. [118] and Lemoff et al. [119] showed that activated structures can also be integrated with microchannels to form a flow switching. Döring et al. [118] showed that using a thermal bimorph cantilever, a laminar flow could be steered into one of two outlets,

Lemoff et al. [119] described an magneto hydrodynamic microfluidic switch using Lorentz force to pump an electrolytic solution. Lin et al. [120] presented a numerical simulation of electrokinetic control model, which allowed the volume of the sample fluid to be controlled. Fu et al. [121] employed the electrokinetic switching approach to develop an electrokinetically driven micro flow cytometer. The “electrokinetic forces” model was also applied to microfluidic applications such as continuous sample injection systems [115].

2.3 Tunable optofluidic microlens

Recently, the development of optofluidic lens has attracted interests from the microfluidic research community [122-124]. To enhance the optical performance, Dong et al. [125] formed an in-situ tunable liquid microlens using liquid-air interface of liquid droplets. To further demonstrate the potential of using the liquid microlenses, Liu et al. [126] formed microlens arrays with separate tunable microlenses. Unfortunately, complex structure and controlling procedures limited the application potential of this method. When liquids flow through a 90-degree curve in a microchannel, the centrifugal effect causes the fluidic interface to bend. Utilizing this effect, Mao et al. [127-129] designed normal refractive lens [127] and Gradient Refractive index (GRIN) lens [128, 129] respectively. The shape of the interface is difficult to control accurately using this method. Tang et al. [130] and Song et al. [131] designed liquid-core liquid-cladding optofluidic lenses based on three laminar streams in PDMS rectangular and circular chambers. Fig. 2.9 shows liquid-core liquid-cladding optofluidic lenses based on three laminar streams. The curvature of these lenses can be tuned by the flowrate ratios of the three streams [132, 133].

2.4 Stable two-fluid flow in straight microchannels with curved interface

For a general micro two-fluid system, the gravity effect is reduced and surface forces become important; the basic in situ configuration is stratified layers with a curved

interface. In a stratified flow pattern, the interface shape is one of crucial factor on the flow pressure drop.

The effect of interface curvature has been studied in macro or mini channels by a number of researchers. Bentwich [134] investigated the two-phase flow with interface defined by a circular arc or an eccentric circle using analytical method. The interfacial tension, capillary force and gravity are included in this model. Ong et al. [135] solved the characteristics of straight liquid-liquid flow with curved interface using numerical method. Brauner et al. [136] analytically solved the stratified two-phase flow characteristics for curved interface with the interface curvature as a prescribed parameter. Brauner et al. [15, 16] also provided a predictive tool for determining the characteristic interface curvature in two-phase system. The steady interfacial curvature is shown to correspond to the interface configuration for which the total system energy is at its minimum. Brauner et al. [15, 16] assumed that the interface is an approximate circular arc. Gorelik et al. [17] obtained an exact analytical solution for interface shape between two immiscible fluids in unidirectional axial laminar pipe flow. The flat interface and the interface with constant curvature also can be calculated using this model. By solving Young-Laplace equation, Ng et al. [18] presented a different exact interface shapes model for various Bond numbers, contact angles and holdups. The results showed that for small Bond numbers, surface tension is dominant and the interface approaches a circular arc shape. Using the Boundary Element Method Ng et al. [137] also calculated the volumetric flowrates, the interface and wall shear stresses and the velocity profiles on the interface and through the cross-section of the pipe using actual interface shape.

In microchannel, the gravity effect is weak [19] while the surface tension is strong, causing the Bond number approaches to zero. The results of previous works [15-18] showed that the interface is a circular arc with constant curvature in microchannels.

2.5 Unstable flow in microchannels

The unstable flow in microchannels is shown in Fig. 2.10. The background of electrohydrodynamic instability theory used to analyze interfacial instability in microchannels stems from the work of Taylor and Melcher on the leaky dielectric model

[24] for the fluids. In literature there are two modeling approaches to investigate the interface instability.

2.5.1 Instability of interface between miscible fluids flow in a normal electric field

In the first approach, the bulk coupled model, assumes a conductivity gradient in a thin diffusion layer between the two fluids, resulting in an electric body force on the fluid. The interaction between the conductivity gradient and the high electric field [138-142] is one of the critical role to generate the interfacial instability. This model is suitable to calculate the instability of interface between miscible fluids. Comparing with the work of Oddy et al. [20], Chen et al. [138] provided a more systematic investigation to establish the critical importance of conductivity gradient and high electric field strength in inducing the instability. The results show that the conductivity gradient and their associated bulk charge accumulation are crucial for instabilities. Lin et al. [139] studied temporal instability in a T-junction merging two fluid streams of differing conductivities via linear stability analysis, experiments, and numerical simulations. All the methods showed that there are thresholds where the electric body force is too great for diffusion to quench. In this flow, the electric field is normal to the conductivity gradient. Chen et al. [140] used experiments and linear stability analysis to study a convective instability occurring in an electroosmotic flow in a similar configuration presented in previously [139]. Using the method of numerical simulations, Storey [141] showed the generation of turbulent flow at low Reynolds number and evaluate the effects of different assumptions in boundary conditions of nonlinear behavior and mixing. Kang et al. [142] studied the initial growth of electrohydrodynamic instability via numerical simulation. The results showed that the molecular diffusion has dual role in the onset and development of instability. The two-fluid system is unstable to high electric fields even without any external disturbances.

The second approach, the surface coupled model, considers a jump in electric conductivity at the interface of the two fluids. In this method, the bulk electric force is vanished; the discontinuity of the electric properties of the fluids across the interface affects the force balance at the fluid-fluid interface, which may either stabilize or

destabilize the interface in question. This model is suitable to calculate the instability of the interface between immiscible flows. Mohamed et al. [143] analyzed the stability of the Couette-Poiseuille flow. The results showed that the velocity stratification affects the stability of fluids; the normal electric field is greatly reduced by the increase of the thickness of the non-conducting fluid layer. Abdella and Rasmussen [144] analyzed Couette flow in an unbounded domain subjected to a normal electric field. They developed a model of two viscous fluids with different electric and fluid properties; the model includes ten parameters. Two special cases, the electrohydrodynamic free-charge case and the electrohydrodynamic polarization charge configuration, were analyzed using Ariy functions [144] and Ariy integrals [144]. Thaokar et al. [145] linearly analyzed the stability of the interface between two dielectric fluids confined between parallel plates subjected to a normal electric field in the limit of zero Reynolds number. The results indicated that the interface becomes unstable when the electric field exceeds a critical value, and the critical value are influenced by the ratio of dielectric constants, ratio of viscosities, ratio of thickness, and surface tension. The results also showed that for small wave-number limit, the critical potential increases proportional to wave number k ; for large wave number limit, the critical potential increases proportional to square root of k . Ozen et al. [146] analyzed the linear stability of the interface between two immiscible fluids; they analyzed the effect of electric fields and the effect of mechanical properties to the instability of the interface. Moatimid [147] investigated the linear surface wave's instability between two finite fluid layers; the fluid layers have different electric properties and subjected to an electric field normal to their interface. It is found that surface tension has a stabilizing influence, while the streaming velocity is strictly destabilizing. A combination of the dielectric constants and the finite conductivities makes a dual role for the electric field.

2.5.2 Instability of interface between immiscible fluids flow in parallel electric field

The stability of the interface between the conducting fluid and non-conducting fluid is critical to achieve the pumping effect. Under the combined effect of hydrodynamic and electroosmosis, the electrohydrodynamic instability occurs at the interface [24]. The

linear stability of the micro electroosmotic pump has both fundamental and practical interest. On the practical side, the breakup phenomenon of a micro pump has become a key technology to keep the accuracy of the pump. On the fundamental side, the stability mechanisms are influenced by the surface tension, the viscosity of the fluids, and the electric properties of the fluids.

The electrohydrodynamic instability of the interface under a parallel electric field are investigated from 1960s [148]. Nayyar and Murty [148] reported the electrohydrodynamic instability in the presence of a parallel electric field in perfect dielectric fluids for the first time. Raco [149] and Taylor [24] experimentally studied the stabilizing of strong parallel electric field. The results showed that the parallel electric field increases the stability of the flow for the perfect dielectric fluids. Elshehawey and Mahmoud [150] investigated the electrohydrodynamic Kelvin-Helmholtz instability under a parallel electric field. The paper indicated that under the suitable choice of the streaming and the parallel electric field, the system is always stable for a given wave number because the parallel electric field supplies a stabilizing effect to the system. Brask et al. [96] analyzed the stability of the interface in a novel electroosmotic pump for pumping non-conducting liquid. The results showed the interface is stabilized under the applied electric field in microchannels. Uguz et al. [151] formulated the linear stability analysis of a two-fluid layer flowing in a channel and subjected to a parallel electric field. The results showed that the electric field is destabilizing over a range of parameter values, which is broader for a normal electric field than for a parallel electric field. The work of Elcoot [152] showed that for leaky dielectric fluids, the parallel electric field has a dual effect, stability or instability, depending on the permittivity and conductivity ratios of the fluids.

2.6 Fabrication of microchannels

Microchannel can be fabricated from a variety of materials because of the rapid development of microfluidic. Based on the fabrication techniques from the microelectronics industry, silicon has been used extensively to fabricate microchannels. Due to the semiconductor industry and the chromatography industry can characterize the surface of silicon, silicon fabrication has been dominant since the introduction of “lab-on-

chip”. Nguyen [153] showed the general procedures of silicon microfabrication techniques.

Lee [154] investigated electrokinetic micromixers fabricated on silicon. The property of the wall surface of the microchannel can be modified by the mature microelectronic fabrication techniques. The zeta potential variation of the channel wall surface is the basic characteristic of the electrokinetic micromixer. In micro-machined silicon microchannels, reversibly adsorbing and removing Avidin from a hydrophobic self-assembled monolayer, was used to demonstrate the reconfigurable hydrophobic-hydrophilic surfaces in [155]. Dreyfus [156] and Thorsen [157] also fabricated microstructures from silicon for the studies of multi-phase flow within microchannel.

Although complex systems can be manufactured out of silicon, it is not the ideal material for microfluidic applications because of some drawbacks of silicon fabrication. For example, expensive processes, difficulty in component integration, and cannot be used for biological applications. In addition, the specialized skills, equipments, and facilities are required for silicon micromachining techniques. However, when the experiment require strong solvents, high temperature, or chemically stable surfaces, silicon and glass-based microfluidic devices are suitable.

Hence, plastic fabrication methods have been proposed. Comparing with silicon and glass, the structures of plastic materials have many benefits that include reduced cost and simplified manufacturing procedures. The wide range of available materials is another advantage of the plastic fabrication methods. It allows the manufacturer to choose material properties suitable for their specific applications, and it is fast to develop prototypes.

Poly-dimethylsiloxane (PDMS) and soft-lithography are getting more attention. Comparing with the conventional methods, fabrication of microfluidic devices in PDMS by soft-lithography is faster and cheaper. The soft-lithographic methods are based on rapid prototyping and replica molding. Because the devices in PDMS do not need to be fabricated in a clean room, it is more accessible to chemists and biologists working under bench-top conditions than the microelectronics-driven methods.

Fig. 2.11 described the method of fabrication of PDMS by soft-lithography. A liquid pre-lithography is poured onto a master. Cured PDMS layer can subsequently be peeled

off the master. As PDMS is elasticity and low surface free energy, this material allows the release from masters without damaging the master or itself. There are many methods for fabricating the master, such as standard microelectronics fabrication technique. Photolithography is one of the common methods. The microstructures are designed using CAD software, and the 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 the structures are transferred into SU-8 resist which serves as a master for fabricating PDMS moulds [158].

Whitesides' research group [159] at Harvard University originally used of PDMS as a material for microfluidic applications. Ichikawa [160] made two kinds of rectangular microchannels, one used glass capillary and the sizes of channel were about $50\text{--}100\text{ }\mu\text{m}$ square. The other kind used PDMS by photolithography technique for the experiment. In the study of two-fluid flow field measurement in microchannel, Kim [161] used PDMS microchannel to conduct the experiment. Sato [162] also designed a T-shaped microchannel using PDMS to investigate the convective mixing in a microchannel.

Another technique for plastic fabrication for microfluidics is using laser ablation. During the ablation process, local temperature on the polymer surface is very high. This high-energy process produces a shock wave and particles are ejected from the substrate thus creating a void. After laser ablation, small particulates can be seen on the surface of the substrate. When using laser ablation, the polymer substrate is exposed to the laser through a mask that defines the area to be ablated. In this process, the mask is made from some materials which cannot be ablated by the laser. Using a direct-write, maskless process, the channels or other structures can be defined. In this process, the polymer substrate material is placed on a moveable stage, and the substrate is moved under the focused laser beam to create the desired structure. One of the advantages of direct-write micromachining process is that the mask does not have to be created to change the design of the microchannel network. Therefore, during the prototyping process, channel design can be changed rapidly. There are also some disadvantages about this method; such as,

parts are made in a sequential manner, thereby limiting the ability to mass produce devices for commercial application.

A UV laser photo-ablation method was used by Roberts [163] for producing the miniaturized liquid-handling systems on polymeric substrate. The fabrication of channel networks and reservoir was accomplished by firing 200 mJ pulses from an UV exciter laser at substrates moving in predefined computer-controlled patterns. The channel in polystyrene, polycarbonate, cellulose acetate, and poly (ethylene terephthalate) can be produced by this method. Comparing with the original polymer sample, the photo-ablated surface showed an increase in their hydrophilicity and rugosity.

Johnson [164] modified a preformed T-microchannel imprinted on polycarbonate using a pulsed UV exciter laser. This microchannel was used for a new type of passive micromixing. In this experiment, a high degree of lateral transport within the channel and rapid mixing of two confluent streams under the effect of electroosmotic flow were presented. Using the UV laser photo-ablation method, several mixer designs were fabricated and investigated. All designs were relatively successful at low flowrates (0.06 cm/s, greater than or equal 75% mixing), but had varying degree of success at high flowrates (0.81 cm/s, 45-80% mixing).

The research group from Denmark [165] directly write on plastic substrates for rapidly producing polymer microfluidic structures using a CO₂-laser. In this method, the laser beam power and the number of passes of the beam along the same channel determined the depth and width of the microchannel. In their experiment, 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 the channel depths were obtained between 100 and 300 μm , and the typical width of the channel was about 250 μm . Several bonding methods for microstructured PMMA (poly methyl methacrylate) parts, such as solvent-assisted gluing, melting, laminating and surface activation using a plasma asher were investigated. Comparing with other bonding methods, the solvent-assisted thermal bonding method is the most time-efficient method. Using laser micromachining together with bonding, a three-layer polymer microstructure with optical fibers is fabricated within two days. Because of the very short cycle time of the production, these systems are especially useful in microfluidic prototyping. Using the same system and technique, Bowden [166]

developed an integrated polymeric microfluidic manifold for the determination of phosphorus. It is noted that the cutting sequence, the number of passes, the laser beam velocity and the laser power, affect the channel depth of microchannels, Snakenborg [167] measured the effect of these parameters. The laser system is a flexible and rapid tool for the production of polymeric microfluidic prototypes.

Weigl [168] showed a method of plastic lamination, it is another low-cost rapid prototyping. The basic idea underneath the lamination technique is about that: several layers of simple fluidic circuit can form microfluidic network or structure. After drawing with CAD software, the CO₂ laser system can cut each layer of fluidic circuit easily. Three layers, top layer, bottom layer and a middle fluidic channel layer, made up of a single channel structure, using more layers, Ismagilov [169] showed that complex structure can be laminated.

Using two layers of PMMA (75mm×25mm) and a layer of double sided adhesive tape, Wu [170] built a new type of microchannels for studying the nonlinear diffusive mixing in microchannels theoretically and experimentally. Using a CO₂ laser system, the channel structure was cut on the adhesive tape, and then the PMMA plates were bonded as a cover and a bottom through the adhesive tape, the cut-through channel for the tape serves as microchannel. A mixing channel with 850 μm width and 50 μm height was fabricated for the experiment.

2.7 Summary

From the literature review, we can conclude that the multi-fluid flow in microchannels has received significant attention over the past decades. Significant efforts have been put into the development of micropump and flow focusing/switching techniques. In addition, the review shows that most of previous works focus on techniques either by pressure-driven or by electroosmosis only, there are few investigations on the coupled effect between pressure-driven flow and electroosmotic flow for multi-fluid system.

Therefore, this report presents theoretical and experimental investigations on electroosmotic control of stream width in hydrodynamic focusing. This new technique is

applied to switch a non-conducting sample stream or droplets. The effects of curved interface and the instability of the two-fluid interface under the coupled effect between pressure-driven flow and electroosmotic flow will be presented.

Fig. 2.1 Schematic representation of ion distribution near contact surface [26]

Fig. 2.2 Schematic of the structure of the EDL. Glass acquires a negative surface charge upon contact with an aqueous solution. The zeta potential (ξ) is the potential at the shear plane and the Debye length (λ_D) is the characteristic thickness of the double layer [28].

Fig. 2.3 Schematic of electroosmotic flow in a cylindrical capillary [28]

Fig. 2.4 A schematic describing of the indirect method for measuring electroosmotic velocity [171]

Fig. 2.5 (top) Schematic of a μ PIV setup. (lower left): Typical setup with inverted microscope (Zeiss). (lower right): Exemplary image pair from flow in curved T-joint with corresponding velocity field. (AHD, TU Delft) [54]

Fig. 2.6 Mechanism of AC electroosmotic pump [91]

Fig. 2.7 Mechanism of hybrid electroosmotic pump [96]

Fig. 2.8 Experimental results about switching sample flow [172]

Fig. 2.9 The principle of liquid-core liquid-cladding optofluidic lenses based on three laminar streams [131]

Fig. 2.10 The flow patterns under different normal electric fields [139]

Fig. 2.11 Schematic of PDMS by soft-lithography fabrication [158]

Chapter 3 Conservation equations

3.1 Electric potential and species

According to the model of Gouy [173], one layer of charge is assumed to be smeared out uniformly over a planar surface immersed in an electrolyte solution. This surface has an electrostatic potential, ψ_0 . The compensating ions are regarded as point charges immersed in a continuous dielectric medium. The fundamental electrostatic equation for the system is the Poisson's equation:

$$\nabla \cdot \mathbf{D} = \rho_e \quad (3.1)$$

where $\mathbf{D}$ is the dielectric displacement, and ρ_e is the density of charge.

The relationship between the dielectric displacement and the electric field is

$$\mathbf{D} = \varepsilon \mathbf{E} \text{ and } \mathbf{E} = -\nabla \psi \quad (3.2)$$

where ε is the permittivity of the dielectric, $\mathbf{E}$ is the electric field, and ψ is the electrostatic potential.

Integrated (3.1) and (3.2), leads to

$$\nabla^2 \psi = -\frac{\rho_e}{\varepsilon} \quad (3.3)$$

For the electric field under steady-state condition, the electrochemical potential $\bar{\mu}_i$ must be constant everywhere, usually this potential is composed by electrostatic potential ($z_i e \psi$) and chemical potential (μ_i) [26], so

$$\bar{\mu}_i = \mu_i + z_i e \psi \quad (3.4)$$

Eq. (3.4), leads to

$$\nabla \bar{\mu}_i = \nabla \mu_i + \nabla z_i e \psi \quad (3.5)$$

For steady state, the electrochemical potential $\bar{\mu}_i$ must be the same everywhere:

$$\nabla \bar{\mu}_i = 0 \quad (3.6)$$

then

$$\nabla \mu_i = -\nabla z_i e \psi \quad (3.7)$$

The chemical potential is defined as

$$\mu_i = \mu_i^0 + k_b T \ln n_i \quad (3.8)$$

where n_i is the ionic number concentration in the bulk.

From Eq.(3.8), we can obtain

$$\nabla \mu_i = \nabla \mu_i^0 + \nabla k_b T \ln n_i \quad (3.9)$$

Because $\nabla \mu_i^0 = 0$, from Eq. (3.9) we can obtain

$$\nabla \mu_i = \nabla k_b T \ln n_i \quad (3.10)$$

Integrating Eq. (3.7) and Eq. (3.10), results in:

$$\nabla k_b T \ln n_i = -\nabla z_i e \psi \quad (3.11)$$

For one dimension, Eq. (3.11) becomes

$$\frac{d}{dx} k_b T \ln n_i = -\frac{d}{dx} z_i e \psi \quad (3.12)$$

$$\frac{d(\ln n_i)}{dx} = \frac{1}{n_i} \frac{dn_i}{dx} = -\frac{z_i e}{k_b T} \frac{d\psi}{dx} \quad (3.13)$$

The solution of Eq. (3.13) is

$$n_i = n_i^0 \exp(-z_i e \psi / k_b T) \quad (3.14)$$

Because

$$\rho_e = \sum n_i z_i e \quad (3.15)$$

Integrating Eq. (3.14) and Eq. (3.15), Eq. (3.15) changes to

$$\rho_e = -2z_0 e n_0 \sinh(z_i e \psi / k_b T) \quad (3.16)$$

Hence, Eq. (3.3) becomes

$$\nabla^2 \psi = \frac{2z_0 e n_0}{\varepsilon} \sinh\left(\frac{z_i e \psi}{k_b T}\right) \quad (3.17)$$

This is the famous Poisson-Boltzmann equation describing the distribution of potential in the diffuse layer.

3.2 Momentum equation

The momentum equation for an incompressible Newtonian liquid is given by the incompressible Navier-Stokes equation as

$$\frac{\partial(\rho \mathbf{v})}{\partial t} + \nabla \bullet (\rho \mathbf{v} \mathbf{v}) = -\nabla p + \mathbf{F} + \mu \nabla^2 \mathbf{v} \quad (3.18)$$

In this equation, $\mathbf{v}$ is the velocity vector, p is the pressure, ρ and μ are the density and dynamic viscosity of the fluid. When an external electric field ($\mathbf{E} = -\nabla \psi$) is applied, the external electric field interacts with the electric double layers in the conducting fluids and creates the electroosmotic body force, $\mathbf{F}$, on the bulk conducting fluid, whereas there is no such electroosmotic body force in the non-conducting fluid because of the absence of electric double layer.

To evaluate the electrokinetic effects, our model assumes that the flow is formed by three simple immiscible Newtonian liquids with constant viscosities, which are independent of shear rate and the local electric field strength. The model assumes:

(1) All the fluids are Newtonian and incompressible.

(2) The properties of the liquids are independent of local electric field and ion concentration. The electric field strength and ion concentration may affect the properties of the conducting fluids. In the current study, these effects are neglected [174].

(3) The liquid's properties are independent of temperature. Joule heating is neglected for dilute electrolytes and low field strength [175].

(4) The flow is fully developed with the non-slip boundary condition. The second term on the left-hand side of the Eq.(3.18), $\nabla \bullet (\rho \mathbf{v} \mathbf{v})$, will vanish.

The pressure gradient is assumed to be uniform along the channel, and the pressure gradients along y and z directions are both zero.

For the conducting fluids, the momentum equation is

$$\frac{\partial(\rho \mathbf{v})}{\partial t} = -\nabla p + \rho_e \mathbf{E} + \mu \nabla^2 \mathbf{v} \quad (3.19)$$

and the momentum equation for the non-conducting fluid is

$$\frac{\partial(\rho \mathbf{v})}{\partial t} = -\nabla p + \mu \nabla^2 \mathbf{v} \quad (3.20)$$

3.3 Continuity equation

The fluids in the microchannel are treated as continuous isotropic matter. A way of defining deviations from the continuum description is through the Knudsen number

$$Kn = \frac{\lambda}{l} \quad (3.21)$$

where λ is the mean-free-path of the molecules and l is a characteristic length scale. l should be chosen to include gradients in the velocity. For liquids λ is approximately the intermolecular length L_{mod} (bond length) and can be described as

$$\lambda = L_{\text{mod}} = \left(\frac{M_{\text{mol}}}{\rho N_A} \right)^{\frac{1}{3}} \quad (3.22)$$

where ρ is the density, M_{mol} is the molar mass, and N_A is Avogadro's number. For water $\lambda \approx 0.31 \text{ nm}$. For this thesis, the characteristic of microchannel is larger than $50 \mu\text{m}$. According Eq. (3.21), $Kn < \frac{0.31}{50000} \approx 6.2 \times 10^{-6}$. According previous work, for $0 < Kn < 0.01$, the continuum approximation is applicable and the Navier-Stokes equation usable. This means that in our case, the continuum approximation is applicable and the Navier-Stokes equation usable.

Consider a control volume, ν , with boundary, $\partial\nu$ in a continuum. For any properties, ι , the evolution of which satisfies the following conservation equation

$$\frac{\partial(\rho\iota)}{\partial t} + \nabla \cdot [\mathbf{J} + \rho\iota\mathbf{v}] = \rho i \quad (3.23)$$

where ρ is the density, $\mathbf{v}$ is the velocity of the continuum, $\mathbf{J}$ is the transport flux. For conservation of mass, the property is taken as $\iota = 1$, the flux $\mathbf{J} = 0$ and the mass production $i = 0$. Eq. (3.23) becomes

$$\frac{\partial\rho}{\partial t} + \nabla \cdot (\rho\mathbf{v}) = 0 \quad (3.24)$$

This is the equation of continuity. For an incompressible Newtonian fluid, the continuity equation becomes

$$\nabla \cdot \mathbf{v} = 0 \quad (3.25)$$

3.4 Dimensionless equations

In the previous section, the governing equation for potential and the Navier-Stokes equations are

$$\nabla^2 \psi = \frac{2 z_0 e n_0}{\varepsilon} \sinh\left(\frac{z_i e \psi}{k_b T}\right) \quad (3.26)$$

$$\rho_e = -2 z_0 e n_0 \sinh(z_i e \psi / k_b T) \quad (3.27)$$

$$\nabla \cdot \mathbf{v} = 0 \quad (3.28)$$

$$\frac{\partial(\rho \mathbf{v})}{\partial t} = -\nabla p + \rho_e \mathbf{E} + \mu \nabla^2 \mathbf{v} \quad (3.29)$$

With reference parameters L_{ref} and U_{ref} for the length and the velocity scale, respectively, the variables in the above equations are normalized as

$$\mathbf{r} = L_{ref} \bar{\mathbf{r}} \quad (3.30)$$

$$t = \frac{L_{ref}}{U_{ref}} \bar{t} \quad (3.31)$$

$$\mathbf{v} = U_{ref} \bar{\mathbf{v}} \quad (3.32)$$

$$p = \rho_{ref} U_{ref}^2 \bar{p} \quad (3.33)$$

$$n_i = n_0 \bar{n}_i \quad (3.34)$$

$$\psi = \frac{k_b T}{z_0 e} \bar{\psi} \quad (3.35)$$

$$\rho_e = 2 z_0 e n_0 \bar{\rho}_e \quad (3.36)$$

$$\mu = \mu_{ref} \bar{\mu} \quad (3.37)$$

Eqs. (3.26)-(3.29) are normalized as

$$\nabla^2 \bar{\psi} = K^2 \sinh(\bar{\psi}) \quad (3.38)$$

$$\bar{\rho}_e = -\bar{\psi} \quad (3.39)$$

$$\nabla \cdot \bar{\mathbf{v}} = 0 \quad (3.40)$$

$$\frac{\partial(\bar{\rho} \bar{\mathbf{v}})}{\partial \bar{t}} = -\nabla \bar{p} + \mathbf{G} \bar{\rho}_e + \frac{1}{\text{Re}} \bar{\mu} \nabla^2 \bar{\mathbf{v}} \quad (3.41)$$

where

$$K = L_{ref} \kappa \quad (3.42)$$

$$\frac{1}{\kappa} = \left(\frac{\varepsilon k_b T}{2 z_0^2 e^2 n_0} \right)^{\frac{1}{2}} \quad (3.43)$$

$$\mathbf{G} = \frac{2z_0 en_0 L_{ref} \mathbf{E}}{\rho_{ref} U_{ref}^2} \quad (3.44)$$

$$\text{Re} = \frac{\rho_{ref} U_{ref} L_{ref}}{\mu_{ref}} \quad (3.45).$$

Chapter 4 Three-fluid flow in straight microchannels with planar interface

4.1 Introduction

The model of a three-fluid flow in microchannels under the combined effect of pressure driven and electroosmosis is presented in Fig. 4.1. Two fluids (fluid 1 and fluid 3) are conducting fluids with high electroosmotic mobility, while the focused fluid (fluid 2) is non-conducting fluid with low electroosmotic mobility. For a given pressure gradient, different electric fields are applied across the conducting fluids, electroosmotic forces will be generated and the velocities of conducting fluids can be regulated depending on the directions and the strength of the applied electric fields. The fluid with low electroosmotic mobility (non-conducting fluid) is focused and delivered by the interfacial viscous force of the conducting fluids and pressure gradient.

To analyze the system proposed above, a Cartesian coordinate system (x, y, z) is used where the origin point, O, is set to be at the center of the non-conducting fluid 2 and the symmetric line is shown in Fig. 4.1. Planar interfaces are assumed. The heights of the conducting fluids and of the non-conducting fluid are denoted as h_1 , h_3 and $2h$, respectively. Half of the width of the channel is denoted by w . The aspect ratio is defined as $\chi = (h_1 + h_3 + 2h)/2w$, the liquid fractions of conducting fluids 1 and 3, and non-conducting fluid 2 are

$$e_1 = \frac{\bar{h}_1}{\bar{h}_1 + 2\bar{h} + \bar{h}_3} \quad (4.1)$$

$$e_3 = \frac{\bar{h}_3}{\bar{h}_1 + 2\bar{h} + \bar{h}_3} \quad (4.2)$$

$$e_2 = \frac{2\bar{h}}{\bar{h}_1 + 2\bar{h} + \bar{h}_3} \quad (4.3)$$

respectively. As a result of surface charges, electric double layers (EDLs) form next to the two liquid-liquid interfaces and the channel walls that are in contact with the conducting fluids. For a more general situation, the walls of the microchannel may be

made of different materials, so that the zeta potentials at the bottom and top walls are denoted as ξ_1 , ξ_4 , and at the side walls as ξ_2 , ξ_5 , and zeta potentials at the interfaces are ξ_3 and ξ_6 , respectively. The electroosmotic flows are along the x direction. Due to symmetry, only half of the cross section ($z > 0$) of the rectangular channel is considered.

The three fluids are driven by the combined pressure and electroosmotic body forces. When the three-fluid flow is fully developed, the velocities of the three liquids, u_1 , u_2 and u_3 along the channel are independent of x . The subscripts, 1, 2 and 3, denote the conducting fluid 1, the non-conducting fluid 2 and the conducting fluid 3, respectively.

4.2 Analytical model

4.2.1 Electric potential

Linearization and solution

The electric potential in the conducting fluids is first considered. Assuming that the electric charge density is not affected by the external electric field due to thin EDLs and the small fluid velocity, the charge convection can be ignored, hence the electric field equation and the fluid flow equation are decoupled [26, 176]. Based on the assumption of local thermodynamic equilibrium, for a small zeta potential, the electric potentials due to the charged wall are described by the linear Poisson-Boltzmann equation which can be written in terms of dimensionless variables as

$$\nabla^2 \bar{\psi} = K^2 \bar{\psi} \quad (4.4)$$

where $K = L_{ref} \kappa$ is the ratio of the length scale L_{ref} to the characteristic double-layer thickness $1/\kappa$. For the case of this section, the reference length is chosen as $L_{ref} = w$. Here, κ is the Debye-Hückel parameter,

$$\frac{1}{\kappa} = \left(\frac{\epsilon k_b T}{2 z_0^2 e^2 n_0} \right)^{\frac{1}{2}} \quad (4.5)$$

where ε is the permittivity of the conducting fluid. Based on the linear approximation, the dimensionless volumetric charge density is given by

$$\bar{\rho}_e = -\bar{\psi}(\bar{z}, \bar{y}) \quad (4.6)$$

Due to the symmetry of the EDL fields in the rectangular channel, Eq. (4.4) is subjected to the following boundary conditions:

$$\text{(conducting fluid 1)} \left\{ \begin{array}{ll} \partial \bar{\psi}_1 / \partial \bar{z} = 0 & \text{at } \bar{z} = 0 \\ \bar{\psi}_1 = \bar{\xi}_2 & \text{at } \bar{z} = \bar{w} \\ \bar{\psi}_1 = \bar{\xi}_3 & \text{at } \bar{y} = -\bar{h} \\ \bar{\psi}_1 = \bar{\xi}_1 & \text{at } \bar{y} = -\bar{h}_1 - \bar{h} \end{array} \right. \quad (4.7)$$

$$\text{(conducting fluid 3)} \left\{ \begin{array}{ll} \partial \bar{\psi}_3 / \partial \bar{z} = 0 & \text{at } \bar{z} = 0 \\ \bar{\psi}_3 = \bar{\xi}_5 & \text{at } \bar{z} = \bar{w} \\ \bar{\psi}_3 = \bar{\xi}_6 & \text{at } \bar{y} = \bar{h} \\ \bar{\psi}_3 = \bar{\xi}_4 & \text{at } \bar{y} = \bar{h}_3 + \bar{h} \end{array} \right. \quad (4.8)$$

The solutions to the Poisson-Boltzmann equation subjected to the above boundary conditions are obtained as

$$\begin{aligned} \bar{\psi}_1 = & \sum_{j=0}^{\infty} \frac{4\bar{\xi}_1}{(2j+1)\pi} (-1)^{j+1} \frac{\sinh[(\bar{h} + \bar{y})]}{\sinh(B_j \bar{h}_1)} \cos(\lambda_j \bar{z}) \\ & - \sum_{j=0}^{\infty} \frac{4\bar{\xi}_3}{(2j+1)\pi} (-1)^{j+1} \times \frac{\sinh[B_j(\bar{h}_1 + \bar{h} + \bar{y})]}{\sinh[B_j \bar{h}_1]} \cos(\lambda_j \bar{z}) \\ & + \sum_{p=1}^{\infty} \frac{2\bar{\xi}_2}{p\pi} [(-1)^p - 1] \frac{\cosh(A_{p1} \bar{z})}{\cosh(A_{p1} \bar{w})} \sin\left[\frac{p\pi}{\bar{h}}(\bar{y} + \bar{h})\right] \end{aligned} \quad (4.9)$$

for conducting fluid 1, and

$$\begin{aligned} \bar{\psi}_3 = & - \sum_{j=0}^{\infty} \frac{4\bar{\xi}_4}{(2j+1)\pi} \times (-1)^{j+1} \times \frac{\sinh[B_j(\bar{y} - \bar{h})]}{\sinh(B_j \bar{h}_3)} \cos(\lambda_j \bar{z}) \\ & + \sum_{j=0}^{\infty} \frac{4\bar{\xi}_6}{(2j+1)\pi} \times (-1)^{j+1} \times \frac{\sinh[B_j(\bar{y} - \bar{h}_3 - \bar{h})]}{\sinh[B_j \bar{h}_3]} \cos(\lambda_j \bar{z}) \\ & - \sum_{p=1}^{\infty} \frac{2\bar{\xi}_5}{p\pi} [(-1)^p - 1] \frac{\cosh(A_{p3} \bar{z})}{\cosh(A_{p3} \bar{w})} \sin\left[\frac{p\pi}{\bar{h}_3}(\bar{y} - \bar{h})\right] \end{aligned} \quad (4.10)$$

for conducting fluid 3, where

$$\begin{cases} \lambda_j = \frac{(2j-1)\pi}{2\bar{w}} \\ B_j = \sqrt{K^2 + \lambda_j^2} \\ A_{p1} = \sqrt{K^2 + \left(\frac{p\pi}{\bar{h}_1}\right)^2} \\ A_{p3} = \sqrt{K^2 + \left(\frac{p\pi}{\bar{h}_3}\right)^2} \end{cases} \quad (4.11)$$

In the above discussion of electroosmosis, the charge state of the surface is described in terms of surface potential at the shear plane, which is identified by the zeta potential [27]. This surface potential is related to the charge density at the surface [177]. From electrostatics, the normal component of the gradient of the electric potential, ψ , jumps by an amount proportional to the surface charge density, ρ_e^s . That is

$$\rho_e^s = -\varepsilon \frac{\partial \psi}{\partial y} \quad (4.12)$$

It is assumed that the gradient of electric potential in the non-conducting fluid vanishes. Using the reference surface charge density as $(\varepsilon k_b T) / (L_{ref} z_0 e)$, we obtain the dimensionless surface charge densities at the liquid-liquid interface as

$$\begin{aligned} \bar{\rho}_{e1}^s(\bar{z}) &= -\frac{\partial \bar{\psi}_1(-\bar{h}, \bar{z})}{\partial \bar{y}} \\ &= -\sum_{j=1}^{\infty} \frac{4(-1)^{j+1} B_j}{(2j+1)\pi} \left[\frac{\bar{\xi}_1}{\sinh(B_j \bar{h}_1)} - \frac{\bar{\xi}_3}{\tanh(B_j \bar{h}_1)} \right] \cos(\lambda_j \bar{z}) \\ &\quad - \sum_{p=1}^{\infty} \frac{2\bar{\xi}_2}{\bar{h}_1} [(-1)^p - 1] \frac{\cosh(A_{p1} \bar{z})}{\cosh(A_{p1} \bar{w})} \end{aligned} \quad (4.13)$$

for the surface charge at interface 1-2, and

$$\begin{aligned} \bar{\rho}_{e3}^s(\bar{z}) &= -\frac{\partial \bar{\psi}_3(\bar{h}, \bar{z})}{\partial \bar{y}} \\ &= \sum_{j=1}^{\infty} \frac{4(-1)^{j+1} B_j}{(2j+1)\pi} \left[\frac{\bar{\xi}_4}{\sinh(B_j \bar{h}_3)} - \frac{\bar{\xi}_6}{\tanh(B_j \bar{h}_3)} \right] \cos(\lambda_j \bar{z}) \\ &\quad + \sum_{p=1}^{\infty} \frac{2\bar{\xi}_5}{\bar{h}_3} [(-1)^p - 1] \frac{\cosh(A_{p3} \bar{z})}{\cosh(A_{p3} \bar{w})} \end{aligned} \quad (4.14)$$

for the surface charge at interface 2-3.

It should be mentioned that the surface charge at the interface 1-2, $\bar{\rho}_{e1}^s$, and interface 2-3, $\bar{\rho}_{e3}^s$, are opposite in sign to the volumetric net charge in the EDL region at the vicinity of the interface. From Eq.(4.12), the surface charge density at the top/bottom walls, $\bar{\rho}_{e3w1}^s$ and $\bar{\rho}_{e1w1}^s$, and the side walls, $\bar{\rho}_{e1w2}^s$ and $\bar{\rho}_{e3w2}^s$, can be obtained. W_1 means the top/bottom wall and W_2 means the side walls. Here, for a large value of EDL parameter K (thin EDL), the electric neutrality conditions are held for the conducting fluid 1 (Eq.(4.15)) and conducting fluid 3 (Eq.(4.16)):

$$\int_0^{\bar{w}} [\bar{\rho}_{q1}^s(\bar{z}) + \bar{\rho}_{q1w1}^s(\bar{z})] d\bar{z} + \int_{-\bar{h}-\bar{h}_1}^{-\bar{h}} \bar{\rho}_{q1w2}^s(\bar{y}) d\bar{y} + \int_0^{\bar{w}} \int_{-\bar{h}-\bar{h}_1}^{-\bar{h}} \bar{\rho}_{q1}(\bar{z}, \bar{y}) d\bar{z} d\bar{y} = 0 \quad (4.15)$$

$$\int_0^{\bar{w}} [\bar{\rho}_{q3}^s(\bar{z}) + \bar{\rho}_{q3w1}^s(\bar{z})] d\bar{z} + \int_{\bar{h}}^{\bar{h}+\bar{h}_3} \bar{\rho}_{q3w2}^s(\bar{y}) d\bar{y} + \int_0^{\bar{w}} \int_{\bar{h}}^{\bar{h}+\bar{h}_3} \bar{\rho}_{q3}(\bar{z}, \bar{y}) d\bar{z} d\bar{y} = 0 \quad (4.16)$$

The solutions of Eq. (4.13) and (4.14) show that the contributions of zeta potential at the top/bottom walls, $\bar{\xi}_4$ and $\bar{\xi}_1$, are relatively small and the contributions of the side walls, $\bar{\xi}_2$ and $\bar{\xi}_5$ are also relatively small except when z approaches to w . The volumetric net charge density (Eq.(4.6)) and the interface charge density (Eq. (4.13)) are required to determine the electrostatic force caused by the presence of zeta potential. The bulk electrostatic force is considered as an additional body force exerting on the conducting fluid in the conventional Navier-Stokes equation. Therefore, the conducting fluids are under the action of pressure gradient, electrostatic force and the viscous shear force at the interface. Similarly, the non-conducting fluid flows as a result of the pressure gradient and the external electrostatic force due to the electrokinetic charge density at the interface, which will be discussed in the following section.

Electric potential distribution in the aqueous solution

The dimensionless parameter K is defined as $K = \kappa L_{ref}$ to evaluate parameters affecting EDL profiles. $1/\kappa$ refers to the characteristic thickness of the EDL. As the Debye-Hückel parameter $\kappa = (2z_0^2 e^2 n_0 / \varepsilon_0 \varepsilon_r k_b T)^{1/2}$ is proportional to the square root of the bulk ionic concentration n_0 , the variation of the ionic concentration will alter the

EDL thickness. In this analysis, the concentration of the two conducting fluids is in the range of $10^{-6} \sim 10^{-5}$ M, therefore, the bulk concentration $n_0 = 6.022 \times 10^{20} \sim 6.022 \times 10^{21} \text{ m}^{-3}$ and the EDL dimension parameter $K = 87 \sim 275$. The EDL profiles are shown in Fig. 4.2 where $K = 87$ ($1/\kappa \approx 300\text{nm}$) and $K = 275$ ($1/\kappa \approx 97\text{nm}$). It shows that the value of K controls the dimensionless EDL thickness: a larger value of K corresponds to a thinner EDL.

4.2.2 Momentum equations of three-liquid flow

Chapter 3 showed the details of dimensionless process for momentum equation and continuity equation. The dimensionless momentum equation for an incompressible Newtonian liquid is given by

$$\frac{\partial(\bar{\rho} \bar{\mathbf{v}})}{\partial \bar{t}} + \nabla \cdot (\bar{\rho} \bar{\mathbf{v}} \bar{\mathbf{v}}) = -\nabla \bar{p} + \mathbf{G} \bar{\rho}_e + \frac{1}{\text{Re}} \bar{\mu} \nabla^2 \bar{\mathbf{v}} \quad (4.17)$$

Because EDLs only form in the conducting fluids, the momentum equations of the three liquids reduce to

$$\frac{\partial(\bar{u}_1)}{\partial \bar{t}} = \frac{1}{\text{Re}} \left(\frac{\partial^2 \bar{u}_1}{\partial \bar{y}^2} + \frac{\partial^2 \bar{u}_1}{\partial \bar{z}^2} \right) + G_{x1} \bar{\rho}_{e1} - \frac{d\bar{p}}{d\bar{x}} \quad (4.18)$$

for conducting fluid 1,

$$\bar{\rho}_3 \frac{\partial(\bar{u}_3)}{\partial \bar{t}} = \frac{\bar{\mu}_3}{\text{Re}} \left(\frac{\partial^2 \bar{u}_3}{\partial \bar{y}^2} + \frac{\partial^2 \bar{u}_3}{\partial \bar{z}^2} \right) + G_{x3} \bar{\rho}_{e3} - \frac{d\bar{p}}{d\bar{x}} \quad (4.19)$$

for the conducting fluid 3, and

$$\bar{\rho}_2 \frac{\partial(\bar{u}_2)}{\partial \bar{t}} = \frac{\bar{\mu}_2}{\text{Re}} \left(\frac{\partial^2 \bar{u}_2}{\partial \bar{y}^2} + \frac{\partial^2 \bar{u}_2}{\partial \bar{z}^2} \right) - \frac{d\bar{p}}{d\bar{x}} \quad (4.20)$$

for the non-conducting fluid 2, where $G_{x1} = \frac{2z_0 en_0 L_{ref} E_{x1}}{\rho_{ref} U_{ref}^2}$ and $G_{x3} = \frac{2z_0 en_0 L_{ref} E_{x3}}{\rho_{ref} U_{ref}^2}$. Here,

the reference viscosity and the density are those of the conducting fluid 1 as $\mu_{ref} = \mu_1$ and

$\rho_{ref} = \rho_1$. Thus, $\beta_1 = \bar{\mu}_1 = 1$, $\beta_2 = \bar{\mu}_2 = \frac{\mu_2}{\mu_1}$ and $\beta_3 = \bar{\mu}_3 = \frac{\mu_3}{\mu_1}$ are the dynamic viscosity

ratios. The continuity conditions of the velocities at the liquid-liquid interfaces are:

$$\bar{u}_1 = \bar{u}_2, \quad \text{at} \quad \bar{y} = -\bar{h} \quad (\text{interface 1-2}) \quad (4.21)$$

$$\bar{u}_2 = \bar{u}_3, \quad \text{at} \quad \bar{y} = \bar{h} \quad (\text{interface 2-3}) \quad (4.22)$$

The shear stress balances, which jumps abruptly at the interface due to the presence of a certain surface charge density,

$$\bar{\mu}_1 \frac{\partial \bar{u}_1}{\partial n} + \bar{E}_{x1} \bar{\rho}_{e1}^s = \bar{\mu}_2 \frac{\partial \bar{u}_2}{\partial n}, \quad \text{at} \quad \bar{y} = -\bar{h} \quad (\text{interface 1-2}) \quad (4.23)$$

$$\bar{\mu}_3 \frac{\partial \bar{u}_3}{\partial n} + \bar{E}_{x3} \bar{\rho}_{e3}^s = \bar{\mu}_2 \frac{\partial \bar{u}_2}{\partial n}, \quad \text{at} \quad \bar{y} = \bar{h} \quad (\text{interface 2-3}) \quad (4.24)$$

Here n is the direction normal to the interface of the two liquids. As planar interface is assumed, the normal direction of interface is along the y direction. The dimensionless matching conditions become

$$(\text{interface 1-2}) \left\{ \begin{array}{l} \bar{u}_1(\bar{z}, -\bar{h}, \bar{t}) = \bar{u}_2(\bar{z}, -\bar{h}, \bar{t}) \\ \frac{\partial \bar{u}_1(\bar{z}, -\bar{h}, \bar{t})}{\partial \bar{y}} = \beta_2 \frac{\partial \bar{u}_2(\bar{z}, -\bar{h}, \bar{t})}{\partial \bar{y}} - M_1 \bar{\rho}_{e1}^s(\bar{z}) \end{array} \right. \quad (4.25)$$

$$(\text{interface 2-3}) \left\{ \begin{array}{l} \bar{u}_3(\bar{z}, \bar{h}, \bar{t}) = \bar{u}_2(\bar{z}, \bar{h}, \bar{t}) \\ \frac{\partial \bar{u}_3(\bar{z}, \bar{h}, \bar{t})}{\partial \bar{y}} = (\beta_2 / \beta_3) \frac{\partial \bar{u}_2(\bar{z}, \bar{h}, \bar{t})}{\partial \bar{y}} - M_3 \bar{\rho}_{e3}^s(\bar{z}) \end{array} \right. \quad (4.26)$$

$$\text{where } M_1 = \frac{\varepsilon k_b T E_{x1}}{z_0 e U_{ref} \mu_{ref}} \quad \text{and} \quad M_3 = \frac{\varepsilon k_b T E_{x3}}{z_0 e U_{ref} \mu_{ref}}.$$

In the rectangular-cross-section channel, the dimensionless boundary conditions for fluids 1, 2 and 3 are respectively

$$(\text{conducting fluid 1}) \left\{ \begin{array}{ll} \frac{\partial \bar{u}_1}{\partial \bar{z}} = 0 & \text{when } \bar{z} = 0 \\ \bar{u}_1 = 0 & \text{when } \bar{z} = \bar{w} \\ \bar{u}_1 = 0 & \text{when } \bar{y} = -\bar{h} - \bar{h}_1 \\ \bar{u}_1 = 0 & \text{when } \bar{t} = 0 \end{array} \right. \quad (4.27)$$

$$(\text{non-conducting fluid 2}) \left\{ \begin{array}{ll} \frac{\partial \bar{u}_2}{\partial \bar{z}} = 0 & \text{when } \bar{z} = 0 \\ \bar{u}_2 = 0 & \text{when } \bar{t} = 0 \end{array} \right. \quad (4.28)$$

$$\left. \begin{array}{l} \text{(conducting fluid 3)} \end{array} \right\} \begin{cases} \frac{\partial \bar{u}_3}{\partial \bar{z}} = 0 & \text{when } \bar{z} = 0 \\ \bar{u}_3 = 0 & \text{when } \bar{z} = \bar{w} \\ \bar{u}_3 = 0 & \text{when } \bar{y} = \bar{h}_3 + \bar{h} \\ \bar{u}_3 = 0 & \text{when } \bar{t} = 0 \end{cases} . \quad (4.29)$$

Due to linearity, the velocities of the fluids (Eqs. (4.18) - (4.20)) can be decomposed into two parts:

$$\bar{u} = \bar{u}^P + \bar{u}^E \quad (4.30)$$

where $\bar{u}^E$ corresponds to the velocity driven by electroosmotic force, and $\bar{u}^P$ is the velocity driven by pressure gradient. The final velocities are the superposition of these electroosmotic and pressure-driven components

$$\left\{ \begin{array}{l} \bar{u}_1 = \bar{u}_1^P + \bar{u}_1^E \\ \bar{u}_2 = \bar{u}_2^P + \bar{u}_2^E \\ \bar{u}_3 = \bar{u}_3^P + \bar{u}_3^E \end{array} \right. \quad (4.31)$$

4.2.3 Steady state three-liquid pressure-driven flow under electroosmotic effect

Analytical solutions

For the steady state fully developed flow, the dimensionless velocity for the conducting fluid 1, when Eq.(4.18) is combined with Eq.(4.6), becomes

$$\left(\text{conducting fluid 1} \right) \frac{\partial^2 \bar{u}_1^E}{\partial \bar{z}^2} + \frac{\partial^2 \bar{u}_1^E}{\partial \bar{y}^2} = \text{Re } G_{x1} \bar{\psi}_1(\bar{z}, \bar{y}) \quad (4.32)$$

corresponding to the velocity component driven by the electroosmotic force and

$$\left(\text{conducting fluid 1} \right) \frac{\partial^2 \bar{u}_1^P}{\partial \bar{z}^2} + \frac{\partial^2 \bar{u}_1^P}{\partial \bar{y}^2} = \text{Re } \frac{d\bar{p}}{d\bar{x}} \quad (4.33)$$

corresponding to the velocity component driven by the pressure gradient. Similarly, the velocities of the non-conducting fluid 2 and the conducting fluid 3 are written in two components as

$$(\text{non-conducting fluid 2}) \quad \frac{\partial^2 \bar{u}_2^E}{\partial \bar{z}^2} + \frac{\partial^2 \bar{u}_2^E}{\partial \bar{y}^2} = 0 \quad (4.34)$$

and

$$(\text{conducting fluid 3}) \quad \frac{\partial^2 \bar{u}_3^E}{\partial \bar{z}^2} + \frac{\partial^2 \bar{u}_3^E}{\partial \bar{y}^2} = \frac{\text{Re}}{\beta_3} G_{x3} \bar{\psi}_3(\bar{z}, \bar{y}) \quad (4.35)$$

corresponding to the velocity component influenced by the electroosmotic flow and

$$(\text{non-conducting fluid 2}) \quad \frac{\partial^2 \bar{u}_2^P}{\partial \bar{z}^2} + \frac{\partial^2 \bar{u}_2^P}{\partial \bar{y}^2} = \frac{\text{Re}}{\beta_2} \frac{d\bar{p}}{d\bar{x}} \quad (4.36)$$

$$(\text{conducting fluid 3}) \quad \frac{\partial^2 \bar{u}_3^P}{\partial \bar{z}^2} + \frac{\partial^2 \bar{u}_3^P}{\partial \bar{y}^2} = \frac{\text{Re}}{\beta_3} \frac{d\bar{p}}{d\bar{x}} \quad (4.37)$$

corresponding to the velocity component driven by the pressure gradient.

Using the separation variables method and substituting the solution of $\bar{\psi}_1$ from Eq. (4.9), the analytical velocity profile corresponding to the electroosmotic force, $\bar{u}_1^E$, and velocity profile corresponding to the pressure gradient, $\bar{u}_1^P$, are obtained from Eqs. (4.32) and (4.33), respectively. They are

$$\begin{aligned} \bar{u}_1^E(\bar{y}, \bar{z}) = & \sqrt{\frac{2}{\bar{w}}} \sum_{j=0}^{\infty} \{ \{ \tanh[\lambda_j(\bar{h} + \bar{h}_1)] \cosh(\lambda_j \bar{y}) + \sinh(\lambda_j \bar{y}) \} b_{1j}^E \\ & - \frac{\phi_{1j}(-\bar{h} - \bar{h}_1) \cosh(\lambda_j \bar{y})}{\cosh[\lambda_j(\bar{h} + \bar{h}_1)]} + \phi_{1j}(\bar{y}) \} \cos(\lambda_j \bar{z}) \end{aligned} \quad (4.38)$$

$$\begin{aligned} \bar{u}_1^P(\bar{y}, \bar{z}) = & \sqrt{\frac{2}{\bar{w}}} \sum_{j=0}^{\infty} \{ \frac{1}{\lambda_j^3} \text{Re} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} (-1)^j \{ \frac{\cosh(\lambda_j \bar{y})}{\cosh[\lambda_j(\bar{h}_1 + \bar{h})]} - 1 \} \\ & + \{ \tanh[\lambda_j(\bar{h}_1 + \bar{h})] \cosh(\lambda_j \bar{y}) + \sinh(\lambda_j \bar{y}) \} b_{1j}^P \} \cos(\lambda_j \bar{z}) \end{aligned} \quad (4.39)$$

The dimensionless velocity profiles for the non-conducting fluid 2 ($\bar{u}_2^E$, $\bar{u}_2^P$), and the conducting fluid 3 ($\bar{u}_3^E$, $\bar{u}_3^P$) are also obtained respectively as

$$\bar{u}_2^E = \sqrt{\frac{2}{\bar{w}}} \sum_0^{\infty} [a_{2j}^E(\bar{y}) \cosh(\lambda_j \bar{y}) + b_{2j}^E(\bar{y}) \sinh(\lambda_j \bar{y})] \cos(\lambda_j \bar{z}) \quad (4.40)$$

$$\bar{u}_2^P = \sqrt{\frac{2}{\bar{w}}} \sum_{j=1}^{\infty} \{ a_{2j}^P \cosh(\lambda_j \bar{y}) + b_{2j}^P \sinh(\lambda_j \bar{y}) - \frac{(-1)^j}{\beta_2 \lambda_j^3} \text{Re} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} \} \times \cos(\lambda_j \bar{z}) \quad (4.41)$$

$$\begin{aligned}\bar{u}_3^E(\bar{y}, \bar{z}) = & \sqrt{\frac{2}{\bar{w}}} \sum_{j=0}^{\infty} \{ \{-\tanh[\lambda_j(\bar{h} + \bar{h}_3)] \cosh(\lambda_j \bar{y}) + \sinh(\lambda_j \bar{y})\} b_{3j}^E \\ & - \frac{\phi_{3j}(\bar{h} + \bar{h}_3)}{\cosh[\lambda_j(\bar{h} + \bar{h}_3)]} \cosh(\lambda_j \bar{y}) + \phi_{3j}(\bar{y}) \} \cos(\lambda_j \bar{z})\end{aligned}\quad (4.42)$$

$$\begin{aligned}\bar{u}_3^P(\bar{y}, \bar{z}) = & \sqrt{\frac{2}{\bar{w}}} \sum_{j=0}^{\infty} \{ \frac{1}{\lambda_j^3} \frac{\text{Re } d\bar{p}}{\beta_3} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} (-1)^j \{ \frac{\cosh(\lambda_j \bar{y})}{\cosh[\lambda_j(\bar{h}_3 + \bar{h})]} - 1 \} \\ & + \{-\tanh[\lambda_j(\bar{h}_3 + \bar{h})] \cosh(\lambda_j \bar{y}) + \sinh(\lambda_j \bar{y})\} b_{3j}^P \} \cos(\lambda_j \bar{z})\end{aligned}\quad (4.43)$$

where the parameter functions $\phi_{1j}(\bar{y})$ and $\phi_{3j}(\bar{y})$, dependent on j and $\bar{y}$ are defined by

$$\begin{aligned}\phi_{1j}(\bar{y}) = & \sqrt{\frac{2}{\bar{w}}} \frac{4 \text{Re } G_{x1} (-1)^{j+1}}{(2j+1)\pi \sinh[B_j \bar{h}_1]} \\ & \times \{ \bar{\xi}_1 \frac{\sinh[B_j(\bar{h} + \bar{y})]}{(B_j^2 - \lambda_j^2)} - \bar{\xi}_3 \frac{\sinh[B_j(\bar{h}_1 + \bar{h} + \bar{y})]}{(B_j^2 - \lambda_j^2)} \} \\ & + \sqrt{\frac{2}{\bar{w}}} \sum_{p=1}^{\infty} \frac{2 \text{Re } G_{x1} \bar{\xi}_2 \lambda_j}{p\pi(A_{p1}^2 + \lambda_j^2)} [(-1)^p - 1] \times (-1)^{j+1} \frac{\sin[\frac{p\pi}{h_1}(\bar{h} + \bar{y})]}{[(\frac{p\pi}{h_1})^2 + \lambda_j^2]}\end{aligned}\quad (4.44)$$

and

$$\begin{aligned}\phi_{3j}(\bar{y}) = & -\sqrt{\frac{2}{\bar{w}}} \sum_{j=1}^{\infty} \frac{4G_{x3} \frac{\text{Re}}{\beta_3} (-1)^{j+1}}{(2j+1)\pi \sinh[B_j \bar{h}_3](B_j^2 - \lambda_j^2)} \\ & \times \{ \bar{\xi}_4 \sinh[B_j(\bar{y} - \bar{h})] - \bar{\xi}_6 \sinh[B_j(\bar{y} - \bar{h}_3 - \bar{h})] \} \\ & - \sqrt{\frac{2}{\bar{w}}} \sum_{p=1}^{\infty} \frac{2\bar{\xi}_5 G_{x3} \frac{\text{Re}}{\beta_3} \lambda_j [(-1)^p - 1] (-1)^{j+1}}{p\pi} \times \frac{\sin[\frac{p}{h_3} \pi(\bar{y} - \bar{h})]}{(A_{p3}^2 + \lambda_j^2) [(\frac{p\pi}{h_3})^2 + \lambda_j^2]}\end{aligned}\quad (4.45)$$

The matching conditions which given in Eqs. (4.25)-(4.26) can also be decomposed

as

$$\text{(interface 1-2) } \begin{cases} \bar{u}_1^E(\bar{z}, -\bar{h}) = \bar{u}_2^E(\bar{z}, -\bar{h}) \\ \frac{\partial \bar{u}_1^E(\bar{z}, -\bar{h})}{\partial \bar{y}} = \beta_2 \frac{\partial \bar{u}_2^E(\bar{z}, -\bar{h})}{\partial \bar{y}} - M_1 \bar{\rho}_{e1}^s(\bar{z}) \end{cases} \quad (4.46)$$

$$\text{(interface 1-2) } \begin{cases} \bar{u}_1^p(\bar{z}, -\bar{h}) = \bar{u}_2^p(\bar{z}, -\bar{h}) \\ \frac{\partial \bar{u}_1^p(\bar{z}, -\bar{h})}{\partial \bar{y}} = \beta_2 \frac{\partial \bar{u}_2^p(\bar{z}, -\bar{h})}{\partial \bar{y}} \end{cases} \quad (4.47)$$

and

$$\text{(interface 2-3) } \begin{cases} \bar{u}_3^E(\bar{z}, \bar{h}) = \bar{u}_2^E(\bar{z}, \bar{h}) \\ \frac{\partial \bar{u}_3^E(\bar{z}, \bar{h})}{\partial \bar{y}} = (\beta_2 / \beta_3) \frac{\partial \bar{u}_2^E(\bar{z}, \bar{h})}{\partial \bar{y}} - M_3 \bar{\rho}_{e3}^s(\bar{z}) \end{cases} \quad (4.48)$$

$$\text{(interface 2-3) } \begin{cases} \bar{u}_3^p(\bar{z}, \bar{h}) = \bar{u}_2^p(\bar{z}, \bar{h}) \\ \frac{\partial \bar{u}_3^p(\bar{z}, \bar{h})}{\partial \bar{y}} = (\beta_2 / \beta_3) \frac{\partial \bar{u}_2^p(\bar{z}, \bar{h})}{\partial \bar{y}} \end{cases} \quad (4.49)$$

At interface 1-2, substitute Eqs. (4.38) and (4.40) into Eq. (4.46) and substitute Eqs. (4.39) and (4.41) into Eq. (4.47); at interface 2-3, substitute Eqs. (4.40) and (4.42) into Eq. (4.48) and substitute Eqs. (4.41) and (4.43) into Eq. (4.49), we can obtain the constants

$$a_{2j}^E = Jb_{1j}^E + Kb_{3j}^E + L \quad (4.50)$$

$$b_{3j}^E = \frac{b_{2j}^E}{B} + I \quad (4.51)$$

$$b_{1j}^E = \frac{b_{2j}^E}{A} + H \quad (4.52)$$

$$b_{2j}^E = \frac{CI + DH + G}{1 - \frac{C}{B} - \frac{D}{A}} \quad (4.53)$$

where

$$A = \tanh[\lambda_j(\bar{h} + \bar{h}_1)] \cosh(\lambda_j \bar{h}) \sinh(\lambda_j \bar{h}) - \sinh(\lambda_j \bar{h}) \sinh(\lambda_j \bar{h}) \\ - \frac{\cosh(\lambda_j \bar{h})}{\beta_2} \tanh[\lambda_j(\bar{h} + \bar{h}_1)] \sinh(\lambda_j \bar{h}) + \frac{\cosh(\lambda_j \bar{h}) \cosh(\lambda_j \bar{h})}{\beta_2} \quad (4.54)$$

$$B = - \frac{\cosh(\lambda_j \bar{h}) \tanh[\lambda_j(\bar{h} + \bar{h}_3)] \sinh(\lambda_j \bar{h})}{\beta_3} + \frac{\cosh(\lambda_j \bar{h}) \cosh(\lambda_j \bar{h})}{\beta_3} \\ + \tanh[\lambda_j(\bar{h} + \bar{h}_3)] \cosh(\lambda_j \bar{h}) \sinh(\lambda_j \bar{h}) - \sinh(\lambda_j \bar{h}) \sinh(\lambda_j \bar{h}) \quad (4.55)$$

$$C = - \frac{\sinh(\lambda_j \bar{h}_3)}{2 \cosh[\lambda_j(\bar{h} + \bar{h}_3)] \sinh(\lambda_j \bar{h})} \quad (4.56)$$

$$D = -\frac{\sinh(\lambda_j \bar{h}_1)}{2 \cosh[\lambda_j (\bar{h} + \bar{h}_1)] \sinh(\lambda_j \bar{h})} \quad (4.57)$$

$$G = -\frac{\phi_{3j}(\bar{h} + \bar{h}_3) \cosh(\lambda_j \bar{h})}{2 \sinh(\lambda_j \bar{h}) \cosh[\lambda_j (\bar{h} + \bar{h}_3)]} + \frac{\phi_{3j}(\bar{h})}{2 \sinh(\lambda_j \bar{h})} \\ + \frac{\phi_{1j}(-\bar{h} - \bar{h}_1) \cosh(\lambda_j \bar{h})}{2 \sinh(\lambda_j \bar{h}) \cosh[\lambda_j (\bar{h} + \bar{h}_1)]} - \frac{\phi_{1j}(-\bar{h})}{2 \sinh(\lambda_j \bar{h})} \quad (4.58)$$

$$H = \frac{\sinh(\lambda_j \bar{h})}{A} \left\{ \frac{\phi_{1j}(-\bar{h} - \bar{h}_1)}{\cosh[\lambda_j (\bar{h} + \bar{h}_1)]} \cosh(\lambda_j \bar{h}) - \phi_{1j}(-\bar{h}) \right\} \\ - \frac{\cosh(\lambda_j \bar{h})}{A \beta_2} \left\{ \frac{\phi'_{1j}(-\bar{h})}{\lambda_j} + \frac{M_1 \bar{\rho}_{q1}^s(\bar{z})}{\lambda_j} + \frac{\phi_{1j}(-\bar{h} - \bar{h}_1)}{\cosh[\lambda_j (\bar{h} + \bar{h}_1)]} \sinh(\lambda_j \bar{h}) \right\} \quad (4.59)$$

$$I = \frac{\cosh(\lambda_j \bar{h}) \sinh(\lambda_j \bar{h}) \phi_{3j}(\bar{h} + \bar{h}_3)}{B \beta_3 \cosh[\lambda_j (\bar{h} + \bar{h}_3)]} - \frac{\cosh(\lambda_j \bar{h})}{B \beta_3 \lambda_j} \phi'_{3j}(\bar{h}) \\ - \frac{\cosh(\lambda_j \bar{h}) M_3 \bar{\rho}_{e3}^s(\bar{z})}{B \beta_3 \lambda_j} - \frac{\phi_{3j}(\bar{h} + \bar{h}_3)}{B \cosh[\lambda_j (\bar{h} + \bar{h}_3)]} \cosh(\lambda_j \bar{h}) \sinh(\lambda_j \bar{h}) \\ + \frac{\phi_{3j}(\bar{h}) \sinh(\lambda_j \bar{h})}{B} \quad (4.60)$$

$$J = \frac{1}{2} \{ \tanh[\lambda_j (\bar{h} + \bar{h}_1)] - \tanh(\lambda_j \bar{h}) \} \quad (4.61)$$

$$\phi'_{1j}(\bar{y}) = \sqrt{\frac{\bar{w}}{2}} \frac{4 \operatorname{Re} G_{x1} B_j (-1)^{j+1}}{(2j+1) \pi \sinh[B_j h_1]} \\ \times \left\{ \bar{\xi}_1 \frac{\cosh[B_j (\bar{h} + \bar{y})]}{(B_j^2 - \lambda_j^2)} - \bar{\xi}_3 \frac{\cosh[B_j (\bar{h}_1 + \bar{h} + \bar{y})]}{(B_j^2 - \lambda_j^2)} \right\} \\ + \sqrt{\frac{2}{\bar{w}}} \sum_{p=1}^{\infty} \frac{2 \operatorname{Re} G_{x1} \bar{\xi}_2 \lambda_j}{\bar{h}_1 (A_{p1}^2 + \lambda_j^2)} [(-1)^p - 1] \\ \times (-1)^{j+1} \frac{\cos[\frac{p\pi}{\bar{h}_1} (\bar{h} + \bar{y})]}{[(\frac{p\pi}{\bar{h}_1})^2 + \lambda_j^2]} \quad (4.62)$$

$$\begin{aligned}
\phi'_{3j}(\bar{y}) = & -\sqrt{\frac{2}{\bar{w}}} \sum_{j=1}^{\infty} \frac{4G_{x3}B_j \frac{\text{Re}}{\beta_3} (-1)^{j+1}}{(2j+1)\pi \sinh[B_j \bar{h}_3](B_j^2 - \lambda_j^2)} \\
& \times \{ \bar{\xi}_4 \cosh[B_j(\bar{y} - \bar{h})] - \bar{\xi}_6 \cosh[B_j(\bar{y} - \bar{h}_3 - \bar{h})] \} \\
& - \sqrt{\frac{2}{\bar{w}}} \sum_{p=1}^{\infty} \frac{2\bar{\xi}_5 G_{x3} \frac{\text{Re}}{\beta_3} \lambda_j [(-1)^p - 1] \times (-1)^{j+1}}{\bar{h}_3} \\
& \times \frac{\cos[\frac{p}{h_3} \pi(\bar{y} - \bar{h})]}{(A_{p3}^2 + \lambda_j^2)[(\frac{p\pi}{h_3})^2 + \lambda_j^2]}
\end{aligned} \tag{4.63}$$

and

$$b_{2j}^p = \frac{O - \frac{RT}{S} - \frac{PU}{V}}{1 - \frac{R}{S} - \frac{P}{V}} \tag{4.64}$$

$$b_{1j}^p = \frac{b_{2j}^p}{S} - \frac{T}{S} \tag{4.65}$$

$$b_{3j}^p = \frac{b_{2j}^p}{V} - \frac{U}{V} \tag{4.66}$$

$$a_{2j}^p = Nb_{3j}^p + Qb_{1j}^p + M \tag{4.67}$$

where

$$\begin{aligned}
M = & \frac{1}{\lambda_j^3} \text{Re} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} \times (-1)^j \times \left\{ \frac{1}{2\beta_3 \cosh[\lambda_j(\bar{h}_3 + \bar{h})]} - \frac{1}{2 \cosh(\lambda_j \bar{h}) \beta_3} \right. \\
& \left. + \frac{1}{2 \cosh[\lambda_j(\bar{h}_1 + \bar{h})]} - \frac{1}{2 \cosh(\lambda_j \bar{h})} + \frac{1}{\cosh(\lambda_j \bar{h}) \beta_2} \right\}
\end{aligned} \tag{4.68}$$

$$N = -\frac{\sinh(\lambda_j \bar{h}_3)}{2 \cosh(\lambda_j \bar{h}) \cosh[\lambda_j(\bar{h}_3 + \bar{h})]} \tag{4.69}$$

$$Q = \frac{\sinh(\lambda_j \bar{h}_1)}{2 \cosh(\lambda_j \bar{h}) \cosh[\lambda_j(\bar{h}_1 + \bar{h})]} \tag{4.70}$$

$$O = \frac{1}{\lambda_j^3} \text{Re} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} (-1)^j \times \left\{ \frac{\coth(\lambda_j \bar{h})}{2\beta_3 \cosh[\lambda_j(\bar{h}_3 + \bar{h})]} - \frac{1}{2 \sinh(\lambda_j \bar{h}) \beta_3} - \frac{\coth(\lambda_j \bar{h})}{2 \cosh[\lambda_j(\bar{h}_1 + \bar{h})]} + \frac{1}{2 \sinh(\lambda_j \bar{h})} \right\} \quad (4.71)$$

$$P = -\frac{\sinh(\lambda_j \bar{h}_3)}{2 \sinh(\lambda_j \bar{h}) \cosh[\lambda_j(\bar{h}_3 + \bar{h})]} \quad (4.72)$$

$$R = -\frac{\sinh(\lambda_j \bar{h}_1)}{2 \sinh(\lambda_j \bar{h}) \cosh[\lambda_j(\bar{h}_1 + \bar{h})]} \quad (4.73)$$

$$S = \frac{\sinh(\lambda_j \bar{h}) \sinh(\lambda_j \bar{h}_1)}{\cosh[\lambda_j(\bar{h}_1 + \bar{h})]} + \frac{\cosh(\lambda_j \bar{h}) \cosh(\lambda_j \bar{h}_1)}{\beta_2 \cosh[\lambda_j(\bar{h}_1 + \bar{h})]} \quad (4.74)$$

$$T = \frac{1}{\lambda_j^3} \text{Re} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} (-1)^j \times \left\{ \frac{\sinh(\lambda_j \bar{h}) \cosh(\lambda_j \bar{h})}{\cosh[\lambda_j(\bar{h}_1 + \bar{h})]} - \sinh(\lambda_j \bar{h}) - \frac{\cosh(\lambda_j \bar{h}) \sinh(\lambda_j \bar{h})}{\beta_2 \cosh[\lambda_j(\bar{h}_1 + \bar{h})]} + \frac{\sinh(\lambda_j \bar{h})}{\beta_2} \right\} \quad (4.75)$$

$$U = \frac{1}{\lambda_j^3} \frac{\text{Re} d\bar{p}}{\beta_3 d\bar{x}} \sqrt{\frac{2}{\bar{w}}} (-1)^j \times \left\{ -\frac{\cosh(\lambda_j \bar{h}) \sinh(\lambda_j \bar{h})}{(\beta_2 / \beta_3) \cosh[\lambda_j(\bar{h}_3 + \bar{h})]} + \frac{\sinh(\lambda_j \bar{h}) \cosh(\lambda_j \bar{h})}{\cosh[\lambda_j(\bar{h}_3 + \bar{h})]} - \sinh(\lambda_j \bar{h}) + \frac{\sinh(\lambda_j \bar{h})}{(\beta_2 / \beta_3)} \right\} \quad (4.76)$$

$$V = \frac{\cosh(\lambda_j \bar{h}) \cosh(\lambda_j \bar{h})}{(\beta_2 / \beta_3) \cosh[\lambda_j(\bar{h}_3 + \bar{h})]} + \frac{\sinh(\lambda_j \bar{h}) \sinh(\lambda_j \bar{h}_3)}{\cosh[\lambda_j(\bar{h}_3 + \bar{h})]} \quad (4.77)$$

The contribution of surface charges, $\bar{\rho}_{ej1}^s$ and $\bar{\rho}_{ej3}^s$, can be obtained by applying Fourier transform to $\bar{\rho}_{e1}^s$ and $\bar{\rho}_{e3}^s$, these are

$$\bar{\rho}_{ej1}^s = -\sqrt{\frac{2}{\bar{w}}} \frac{4(-1)^{j+1} B_j}{(2j+1)\pi} \left[\frac{\bar{\xi}_1}{\sinh(B_j \bar{h}_1)} - \frac{\bar{\xi}_3}{\tanh(B_j \bar{h}_1)} \right] - \sqrt{\frac{2}{\bar{w}}} \times \sum_{p=1}^{\infty} \frac{2\bar{\xi}_2}{\bar{h}_1} [(-1)^p - 1] \times \frac{(-1)^j \lambda_j}{A_{p1}^2 + \lambda_j^2} \quad (4.78)$$

$$\begin{aligned}\bar{\rho}_{ej3}^s = & \sqrt{\frac{\bar{w}}{2}} \frac{4(-1)^{j+1} B_j}{(2j+1)\pi} \left[\frac{\bar{\xi}_4}{\sinh(B_j \bar{h}_3)} - \frac{\bar{\xi}_6}{\tanh(B_j \bar{h}_3)} \right] \\ & + \sum_{p=1}^{\infty} \frac{2\bar{\xi}_5}{\bar{h}_3} [(-1)^p - 1] \times (-1)^j \times \frac{\lambda_j}{A_{p3}^2 + \lambda_j^2}\end{aligned}\quad (4.79)$$

The dimensionless volumetric flowrates through the rectangular-cross-section channel can be defined as $\bar{q}_1 = \bar{q}_1^E + \bar{q}_1^P = q_1 / (L_{ref}^2 U_{ref})$, $\bar{q}_2 = \bar{q}_2^E + \bar{q}_2^P = q_2 / (L_{ref}^2 U_{ref})$ and $\bar{q}_3 = \bar{q}_3^E + \bar{q}_3^P = q_3 / (L_{ref}^2 U_{ref})$, the dimensionless flowrates are given as

$$\bar{q}_1^E = 2 \int_{-\bar{h}-\bar{h}_1}^{-\bar{h}_1} \int_0^{\bar{w}} \bar{u}_1^E(\bar{y}, \bar{z}) d\bar{z} d\bar{y} \quad (4.80)$$

$$\bar{q}_1^P = 2 \int_{-\bar{h}-\bar{h}_1}^{-\bar{h}} \int_0^{\bar{w}} \bar{u}_1^P(\bar{y}, \bar{z}) d\bar{z} d\bar{y} \quad (4.81)$$

$$\bar{q}_2^E = 2 \int_{-\bar{h}}^{\bar{h}} \int_0^{\bar{w}} \bar{u}_2^E(\bar{y}, \bar{z}) d\bar{z} d\bar{y} \quad (4.82)$$

$$\bar{q}_2^P = 2 \int_{-\bar{h}}^{\bar{h}} \int_0^{\bar{w}} \bar{u}_2^P(\bar{y}, \bar{z}) d\bar{z} d\bar{y} \quad (4.83)$$

and

$$\bar{q}_3^E = 2 \int_{\bar{h}}^{\bar{h}+\bar{h}_3} \int_0^{\bar{w}} \bar{u}_3^E(\bar{y}, \bar{z}) d\bar{z} d\bar{y} \quad (4.84)$$

$$\bar{q}_3^P = 2 \int_{\bar{h}}^{\bar{h}+\bar{h}_3} \int_0^{\bar{w}} \bar{u}_3^P(\bar{y}, \bar{z}) d\bar{z} d\bar{y} \quad (4.85)$$

Substituting $\bar{u}_1^E$, $\bar{u}_1^P$, $\bar{u}_2^E$, $\bar{u}_2^P$, $\bar{u}_3^E$ and $\bar{u}_3^P$ into Eqs. (4.80)-(4.85), respectively, yields the dimensionless volumetric flowrates as

$$\begin{aligned}\bar{q}_1^E = & \sqrt{\frac{2}{\bar{w}}} \sum_{j=0}^{\infty} \frac{2 \sin(\lambda_j \bar{w})}{\lambda_j} \left\{ \frac{1}{\lambda_j} \tanh[\lambda_j (\bar{h} + \bar{h}_1)] \times [\sinh(\lambda_j (\bar{h} + \bar{h}_1)) - \sinh(\lambda_j \bar{h})] \right. \\ & + \frac{1}{\lambda_j} [\cosh(\lambda_j \bar{h}) - \cosh(\lambda_j (\bar{h} + \bar{h}_1))] \} b_{1j}^E \\ & + \sqrt{\frac{2}{\bar{w}}} \sum_{j=0}^{\infty} \frac{2 \sin(\lambda_j \bar{w})}{\lambda_j} \times \left\{ \frac{1}{\lambda_j} \frac{\phi_{1j}(-\bar{h} - \bar{h}_1)}{\cosh[\lambda_j (\bar{h} + \bar{h}_1)]} \right. \\ & \times [\sinh(\lambda_j \bar{h}) - \sinh(\lambda_j (\bar{h} + \bar{h}_1))] + \phi_{1j}'(-\bar{h}) - \phi_{1j}'(-\bar{h} - \bar{h}_1) \} \end{aligned}\quad (4.86)$$

$$\begin{aligned}\bar{q}_1^p &= \sum_{j=0}^{\infty} \frac{4}{\bar{w}} \sin(\lambda_j \bar{w}) \frac{\text{Re } d\bar{p}}{\lambda_j^4 d\bar{x}} (-1)^j \left\{ \frac{\sinh(\lambda_j(\bar{h} + \bar{h}_1)) - \sinh(\lambda_j \bar{h})}{\lambda_j \cosh[\lambda_j(\bar{h}_1 + \bar{h})]} - \bar{h}_1 \right\} \\ &+ \sum_{j=0}^{\infty} 2\sqrt{\frac{2}{\bar{w}}} \frac{\sin(\lambda_j \bar{w})}{\lambda_j^2} \{ \tanh[\lambda_j(\bar{h}_1 + \bar{h})][\sinh(\lambda_j(\bar{h} + \bar{h}_1)) - \sinh(\lambda_j \bar{h})] \\ &+ [\cosh(\lambda_j \bar{h}) - \cosh(\lambda_j(\bar{h} + \bar{h}_1))] \} b_{1j}^p\end{aligned}\quad (4.87)$$

$$\bar{q}_2^E = 4\sqrt{\frac{2}{\bar{w}}} \sum_{j=0}^{\infty} \frac{a_{2j}^E \sinh(\lambda_j \bar{h}) \sin(\lambda_j \bar{w})}{\lambda_j^2} \quad (4.88)$$

$$\bar{q}_2^p = 4\sqrt{\frac{2}{\bar{w}}} \sum_{j=0}^{\infty} [a_{2j}^p \sinh(\lambda_j \bar{h}) - \frac{(-1)^j}{\beta_2 \lambda_j^2} \text{Re } \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} \bar{h}] \frac{\sin(\lambda_j \bar{w})}{\lambda_j^2} \quad (4.89)$$

$$\begin{aligned}\bar{q}_3^E &= 2\sqrt{\frac{2}{\bar{w}}} \sum_{j=0}^{\infty} \left\{ \frac{1}{\lambda_j} \{ -\tanh[\lambda_j(\bar{h} + \bar{h}_3)][\sinh(\lambda_j(\bar{h} + \bar{h}_3)) - \sinh(\lambda_j \bar{h})] \right. \\ &+ \cosh(\lambda_j(\bar{h} + \bar{h}_3)) - \cosh(\lambda_j \bar{h}) \} b_{3j}^E - \frac{1}{\lambda_j} \frac{\phi_{3j}(\bar{h} + \bar{h}_j)}{\cosh[\lambda_j(\bar{h} + \bar{h}_3)]} \\ &\times [\sinh(\lambda_j(\bar{h} + \bar{h}_3)) - \sinh(\lambda_j \bar{h})] + \phi_{3j}'(\bar{h} + \bar{h}_3) - \phi_{3j}'(\bar{h}) \} \frac{\sin(\lambda_j \bar{w})}{\lambda_j}\end{aligned}\quad (4.90)$$

$$\begin{aligned}\bar{q}_3^p &= 2\sqrt{\frac{2}{\bar{w}}} \sum_{j=0}^{\infty} \left\{ \frac{1}{\lambda_j^4} \frac{\text{Re } d\bar{p}}{\beta_3 d\bar{x}} \sqrt{\frac{2}{\bar{w}}} (-1)^j \left\{ \frac{1}{\lambda_j} \frac{\sinh(\lambda_j(\bar{h} + \bar{h}_3)) - \sinh(\lambda_j \bar{h})}{\cosh[\lambda_j(\bar{h}_3 + \bar{h})]} - \bar{h}_3 \right\} \right. \\ &\times \sin(\lambda_j \bar{w}) + 2\sqrt{\frac{2}{\bar{w}}} \{ -\tanh[\lambda_j(\bar{h}_3 + \bar{h})][\sinh(\lambda_j(\bar{h} + \bar{h}_3)) - \sinh(\lambda_j \bar{h})] \\ &+ [\cosh(\lambda_j(\bar{h} + \bar{h}_3)) - \cosh(\lambda_j \bar{h})] \} b_{3j}^p \} \frac{\sin(\lambda_j \bar{w})}{\lambda_j^2}\end{aligned}\quad (4.91)$$

where the parameter functions ϕ_{1j}' , ϕ_{3j}' are defined as

$$\begin{aligned}\phi_{1j}'(\bar{y}) &= \sqrt{\frac{\bar{w}}{2}} \frac{4 \text{Re } G_{x1} (-1)^{j+1}}{(2j+1)\pi B_j \sinh[B_j \bar{h}_1]} \\ &\times \left\{ \bar{\xi}_1 \frac{\cosh[B_j(\bar{h} + \bar{y})]}{(B_j^2 - \lambda_j^2)} - \bar{\xi}_3 \frac{\cosh[B_j(\bar{h}_1 + \bar{h} + \bar{y})]}{(B_j^2 - \lambda_j^2)} \right\} \\ &- \sqrt{\frac{2}{\bar{w}}} \sum_{p=1}^{\infty} \frac{2 \text{Re } G_{x1} \bar{\xi}_2 \lambda_j \bar{h}_1}{(p\pi)^2 (A_{p1}^2 + \lambda_j^2)} [(-1)^p - 1] \times (-1)^{j+1} \frac{\cos[\frac{p\pi}{\bar{h}_1}(\bar{h} + \bar{y})]}{[(\frac{p\pi}{\bar{h}_1})^2 + \lambda_j^2]}\end{aligned}\quad (4.92)$$

$$\begin{aligned}
\phi'_{3j}(\bar{y}) = & -\sqrt{\frac{2}{\bar{w}}} \sum_{j=1}^{\infty} \frac{4G_{x3} \frac{\text{Re}}{\beta_3} (-1)^{j+1}}{(2j+1)\pi B_j \sinh(B_j \bar{h}_3) \times (B_j^2 - \lambda_j^2)} \\
& \times \{ \bar{\xi}_4 \cosh[B_j(\bar{y} - \bar{h})] - \bar{\xi}_6 \cosh[B_j(\bar{y} - \bar{h}_3 - \bar{h})] \} \\
& + \sqrt{\frac{2}{\bar{w}}} \sum_{p=1}^{\infty} \frac{2\bar{\xi}_5 G_{x3} \frac{\text{Re}}{\beta_3} \bar{h}_3 \lambda_j [(-1)^p - 1] \times (-1)^{j+1}}{(p\pi)^2} \times \frac{\cos[\frac{p}{h_3} \pi(\bar{y} - \bar{h})]}{(A_{p3}^2 + \lambda_j^2) [(\frac{p\pi}{h_3})^2 + \lambda_j^2]}
\end{aligned} \tag{4.93}$$

Results of the steady state model

In Fig. 4.3, the three-fluid, (two conducting fluids and a non-conducting fluid) are introduced through a constant pressure source. When electric fields are applied across the conducting fluids, the external electric field interacts with the net charges within the electric double layers and creates electroosmotic forces within the bulk conducting fluids. If the applied electric field varies, such applied electroosmotic body forces will change accordingly. As a result, for a given pressure gradient, the velocities and flowrates of the three liquids depend on the applied electroosmotic force.

From Eq. (4.30), the velocity $\bar{u}$ of three-fluid can be decomposed into two parts, $\bar{u}^p$ and $\bar{u}^E$. $\bar{u}^p$ corresponds to velocity driven by pressure gradient and $\bar{u}^E$ corresponds to velocity driven by electroosmotic effect. With the proposed analytical model, we investigate the following cases. (a) With zero pressure gradient is applied across the microchannel, the flow is simply a three-fluid electroosmotic flow with the flow velocity $\bar{u}^E$. (b) With zero applied electric fields, the flow is simply driven by pressure gradient with the flow velocity $\bar{u}^p$. (c) When both the pressure gradient and the electric field are applied across the microchannel, the three-fluid flow is driven by the combined electroosmotic force and pressure gradient with the flow velocity $\bar{u}$.

In the analysis, the reference potential and the applied electric field are taken at 25 °C as $\xi_{ref} = k_b T / (z_0 e) \approx -25.7 \text{ mV}$ and $E_{x1} = E_{x3} = 1000 \text{ V/m}$. The viscosity and density of the NaCl solution is $\mu_{ref} = 10^{-3} \text{ Pa s}$ and $\rho_{ref} = 10^3 \text{ kg/m}^3$, respectively. With these reference potential and viscosity, the Helmholtz-Smoluchowski electroosmotic velocity is

chosen as the reference velocity $U_{ref} = \frac{E_{x1} \varepsilon \xi_{ref}}{\mu_{ref}} = 5.3 \times 10^{-5} m/s$. The corresponding

Reynolds number is $Re \approx 0.0021$.

(a) Three-fluid electroosmotic flow

In the following, the results show the velocity at the center line of microchannel $z = 0$. Fig. 4.4 shows the dimensionless velocity profiles, $\bar{u}^E$, at the symmetric line, at zero pressure gradient, $E_{x1} = E_{x3} = 1000 V/m$, $\beta_1 = \beta_3 = 1$, and different viscosity ratios $\beta_2 = 0.5, 1, 2, 3$. When electric fields are applied across the conducting fluids, the conducting fluids 1 and 3 are driven by electroosmosis, which drag the non-conducting fluid 2 by the hydrodynamic shear force. The three-fluid flow is affected by viscosity ratios, the strength of the external electric fields and electroosmotic characteristics of the conducting fluids. By this way, the non-conducting fluid can be delivered by electroosmosis.

The flow characteristics depend on the coupled effects of the three-fluid which involve the electroosmotic driving force in the conducting fluids and the interfacial phenomenon. The interfacial phenomenon is obtained from the balance of the modified stress force as shown in Eqs. (4.25)-(4.26), which involves the opposite electrostatic force exerted on the interface and the hydrodynamic shear stress at the liquid-liquid interface. The velocities at the liquid-liquid interface must match, i.e. the conducting fluids and the non-conducting fluid velocities must be the same and the forces must be balanced at the interface. To investigate the effect of viscosity ratio between the three fluids, the value of $\beta_3 = 1$, whereas, β_2 are chosen to have different values.

The electric body force is resulted from the interaction of the applied electric fields and the net charge density. This driving force exists only within the non-neutral charge region — the electric double layer (EDL) in the conducting fluids 1 and 3. Liquid outside the EDL regions is set in motion passively due to the frictional stresses originating from the liquid viscosity. The velocity profile of the non-conducting fluid 2 is passive. It is purely due to the interfacial shear stresses dragged by the conducting fluids on the non-conducting fluid. The results indicate that the velocity profiles of the conducting fluids

are strongly dependent on the viscosity ratio, β_2 . Because the viscosity ratio is small, the flow resistance of the non-conducting fluid decrease with the decreasing in β_2 . Thus, the non-conducting fluid can be driven with less flow resistance as shown in Fig. 4.4. 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 conducting fluids.

For an extreme case, the infinite viscosities of μ_2 and μ_3 make the flows of the non-conducting fluid 2 and the conducting fluid 3 resemble that of the channel wall. Hence the flow of the conducting fluid 1 resembles the single-fluid electroosmotic flow (EOF), through which we can compare the results of the proposed analytical model with those of the previous works. If the channel width is much larger than the channel height, in the fully developed state, the velocity profile at the symmetric section in the channel is identical with the flow between two infinite parallel flat plates. If the zeta potentials are assumed as the same between two parallel plates, Patankar et al. [38] presented an analytical result of the stream wise velocity,

$$\bar{u}(\bar{y}) = \bar{\xi}_1 \left[\frac{\cosh(K\bar{y} - K\bar{h}_1/2)}{\cosh(K\bar{h}_1/2)} - 1 \right] \quad (4.94)$$

Fig. 4.5 compares the velocity profile of the conducting fluid 1 from the three-fluid model when $\beta_2 = \beta_3 = 10^4$ with the exact solution from Eq.(4.94). It clearly presents that the results from the two different models are identical.

(b) Three-fluid flow driven by electroosmosis and pressure gradient

From Fig. 4.6, when both pressure gradient and electric field are applied, the three liquids are driven by electroosmotic body force and pressure gradient. For a given pressure gradient, the velocities and flowrates of the three liquids depend only on the applied electroosmotic force.

Figs. 4.6 to 4.8 show the dimensionless velocity profiles at the symmetric line of the three-fluid flow driven by the combined effect of electroosmosis and pressure gradient. In

the analysis, $e_1 = e_3 = 0.29$, $e_2 = 0.42$ and equal applied electric fields $E_{x1} = E_{x3}$ are specified. e_1 , e_2 , and e_3 are the liquid fractions defined in Eqs. (4.1)-(4.3).

The effect of applied electric fields on the dimensionless velocity profile is shown in Fig. 4.6. The flow parameters are $\beta_1 = \beta_2 = \beta_3 = 1$, $d\bar{p}/d\bar{x} = 1000$. The results indicate that for a given pressure gradient, the velocity profile $\bar{u}$ of the three-fluid flow strongly depends on the applied electric field; hence the volumetric flowrates of the three-fluid flow can be controlled. With zero electric field, $E_{x1} = E_{x3} = 0$ V/m, the flow is in fact a pressure-driven flow of a single liquid, showing the parabolic velocity profile, $\bar{u}^p$. We can compare the analytical solution of the three-fluid model with the one-dimensional, fully developed Navier-Stokes equation under the steady state condition [178]. It is clearly seen that the results from the two different models are identical.

The relationship between $\bar{u}^p$, $\bar{u}^E$ and $\bar{u}$ for $d\bar{p}/d\bar{x} = 1500$, $E_{x1} = E_{x3} = 15000$ V/m and $\beta_3 = \beta_2 = 1$ is shown in Fig. 4.7. The result shows that the velocity profile, $\bar{u}$ of a three-fluid flow driven by the combined electroosmosis and pressure gradient is the superposition of the solutions of a three-fluid electroosmotic velocity profile $\bar{u}^E$ and a pressure-driven parabolic profile, $\bar{u}^p$.

Fig. 4.8 shows the dimensionless velocity profiles $\bar{u}$ at the symmetric line when $\beta_2 = 5$, $\beta_1 = \beta_3 = 1$, $d\bar{p}/d\bar{x} = 1000$. Comparing Fig. 4.8 ($\beta_2 = 5$) and Fig. 4.6 ($\beta_2 = 1$), the results indicate that the velocity profiles of the conducting fluids are strongly dependent on the viscosity ratio, β_2 . For a given pressure gradient, a higher viscosity ratio β_2 and consequently a higher flow resistance of the non-conducting fluid result in a steeper velocity gradient at the interface of the conducting fluids.

Fig. 4.9 shows the effect of applied pressure gradient on the dimensionless velocity profile at the symmetric line of the three-fluid driven by the combined electroosmosis and pressure gradient. In the analysis, $e_1 = e_3 = 0.29$, $e_2 = 0.42$, $\beta_2 = 5$, $\beta_1 = \beta_3 = 1$ and equal applied electric fields $E_{x1} = E_{x3} = 1000$ V/m are specified. The results indicate that for a given applied electric field, the velocity profile $\bar{u}$ of the three-fluid flow strongly depends on the applied pressure gradient. With zero pressure gradient ($d\bar{p}/d\bar{x} = 0$), the

flow is simply a three-fluid electroosmotic flow $\bar{u}^E$. The velocity $\bar{u}$ increases with the applied pressure gradient as shown by the linear combination of the electroosmotic profile $\bar{u}^E$, and the pressure-driven profile $\bar{u}^P$. At a high pressure gradient, the electroosmotic flow effect becomes weaker.

The flow characteristics depend on the coupling effect of the three-fluid flow which involves the electroosmotic phenomenon of the conducting fluids and the interfacial stresses at the interface of the adjacent fluids. To investigate the effect of viscosity ratio between the three fluids, the value of $\beta_3 = 1$ is set to be a constant value, whereas, β_2 are chosen to vary. Fig. 4.10 shows the dimensionless velocity profiles at the symmetric line at $d\bar{p}/d\bar{x} = 1000$, $E_{x1} = E_{x3} = 1000$ V/m, and different viscosity ratios β_2 of 0.5, 1, 2 and 3 respectively. The velocity profiles of the three-fluid flow strongly depend on the viscosity ratio, β_2 . For a given pressure gradient and applied electric fields, if the viscosity ratio β_2 decreases, the non-conducting fluid can be driven with less flow resistance.

The comparison between theoretical analysis and the published two-fluid experimental data [12] is shown in Fig. 4.11. To simulate the flow, a infinite viscosity of $\bar{\mu}_3$, is assumed which make the conducting fluid 3 resemble that of the channel wall. Hence we can compare our theoretical analysis with the two-fluid data. Our results agree well with the published experimental data.

Fig. 4.12 shows the influence of the viscosity ratio β_2 on the volumetric flowrate. For a given pressure gradient and the applied electric fields, volumetric flowrates of the three-fluid flow increase with decreasing viscosity ratio β_2 . A rapid increase in the flowrate occurs when the viscosity ratio β_2 decrease from 10 to 0.1.

(c) Effect of the applied electric fields and pressure gradient on flowrate

The model shows that the volumetric flowrates increase linearly with the applied pressure gradient (Fig. 4.13). The volumetric flowrates also increase linearly with the

applied electric fields (Fig. 4.14). The flowrates of the conducting fluids are higher due to the presence of electroosmotic forces.

4.2.4 Transient three-liquid pressure-driven flow under electroosmotic effect

Analytical solutions

The Laplace transform method is applied to solve the transient momentum equations Eqs. (4.18)-(4.20). This report assumes that the zeta potentials are not affected by the external electric field.

$$\text{(conducting fluid 1)} \left\{ \begin{array}{l} \text{Re} \frac{\partial \bar{u}_1^E}{\partial \bar{t}} = \frac{\partial^2 \bar{u}_1^E}{\partial \bar{z}^2} + \frac{\partial^2 \bar{u}_1^E}{\partial \bar{y}^2} + \text{Re} G_{x1} \bar{\rho}_{e1} \\ \text{Re} \frac{\partial \bar{u}_1^P}{\partial \bar{t}} = \frac{\partial^2 \bar{u}_1^P}{\partial \bar{z}^2} + \frac{\partial^2 \bar{u}_1^P}{\partial \bar{y}^2} - \text{Re} \frac{d\bar{p}}{d\bar{x}} \end{array} \right. \quad (4.95)$$

$$\text{(non-conducting fluid 2)} \left\{ \begin{array}{l} \frac{\text{Re}}{\alpha_2} \frac{\partial \bar{u}_2^E}{\partial \bar{t}} = \frac{\partial^2 \bar{u}_2^E}{\partial \bar{z}^2} + \frac{\partial^2 \bar{u}_2^E}{\partial \bar{y}^2} \\ \frac{\text{Re}}{\alpha_2} \frac{\partial \bar{u}_2^P}{\partial \bar{t}} = \frac{\partial^2 \bar{u}_2^P}{\partial \bar{z}^2} + \frac{\partial^2 \bar{u}_2^P}{\partial \bar{y}^2} - \frac{\text{Re}}{\beta_2} \frac{d\bar{p}}{d\bar{x}} \end{array} \right. \quad (4.96)$$

$$\text{(conducting fluid 3)} \left\{ \begin{array}{l} \frac{\text{Re}}{\alpha_3} \frac{\partial \bar{u}_3^E}{\partial \bar{t}} = \frac{\partial^2 \bar{u}_3^E}{\partial \bar{z}^2} + \frac{\partial^2 \bar{u}_3^E}{\partial \bar{y}^2} + \frac{\text{Re}}{\beta_3} G_{x3} \bar{\rho}_{e3} \\ \frac{\text{Re}}{\alpha_3} \frac{\partial \bar{u}_3^P}{\partial \bar{t}} = \frac{\partial^2 \bar{u}_3^P}{\partial \bar{z}^2} + \frac{\partial^2 \bar{u}_3^P}{\partial \bar{y}^2} - \frac{\text{Re}}{\beta_3} \frac{d\bar{p}}{d\bar{x}} \end{array} \right. \quad (4.97)$$

where $\alpha_2 = \frac{\bar{\mu}_2/\bar{\rho}_2}{\bar{\mu}_1/\bar{\rho}_1}$, $\alpha_3 = \frac{\bar{\mu}_3/\bar{\rho}_3}{\bar{\mu}_1/\bar{\rho}_1}$.

The boundary conditions and initial conditions are shown using these equations:

$$\text{(conducting fluid 1)} \left\{ \begin{array}{ll} \frac{\partial \bar{u}_1}{\partial \bar{z}} = 0 & \text{when } \bar{z} = 0 \\ \bar{u}_1 = 0 & \text{when } \bar{z} = \bar{w} \\ \bar{u}_1 = 0 & \text{when } \bar{y} = -\bar{h} - \bar{h}_1 \\ \bar{u}_1 = 0 & \text{when } \bar{t} \leq 0 \end{array} \right. \quad (4.98)$$

$$(\text{non-conducting fluid 2}) \begin{cases} \frac{\partial \bar{u}_2}{\partial \bar{z}} = 0 & \text{when } \bar{z} = 0 \\ \bar{u}_2 = 0 & \text{when } \bar{t} \leq 0 \end{cases} \quad (4.99)$$

$$(\text{conducting fluid 3}) \begin{cases} \frac{\partial \bar{u}_3}{\partial \bar{z}} = 0 & \text{when } \bar{z} = 0 \\ \bar{u}_3 = 0 & \text{when } \bar{z} = \bar{w} \\ \bar{u}_3 = 0 & \text{when } \bar{y} = \bar{h}_3 + \bar{h} \\ \bar{u}_3 = 0 & \text{when } \bar{t} \leq 0 \end{cases} \quad (4.100)$$

We substitute the local volumetric net charge density, Eq.(4.12), into momentum equation Eq. (4.95), for conducting fluid 1, the Laplace transform of Eq. (4.95) as

$$(\text{conducting fluid 1}) \begin{cases} s \operatorname{Re} \bar{U}_1^E(\bar{z}, \bar{y}, s) = \frac{\partial^2 \bar{U}_1^E(\bar{z}, \bar{y}, s)}{\partial \bar{z}^2} + \frac{\partial^2 \bar{U}_1^E(\bar{z}, \bar{y}, s)}{\partial \bar{y}^2} - \frac{\operatorname{Re} G_{x1} \bar{\psi}_1(\bar{z}, \bar{y})}{s} \\ s \operatorname{Re} \bar{U}_1^P(\bar{y}, \bar{z}, s) = \frac{\partial^2 \bar{U}_1^P(\bar{y}, \bar{z}, s)}{\partial \bar{z}^2} + \frac{\partial^2 \bar{U}_1^P(\bar{y}, \bar{z}, s)}{\partial \bar{y}^2} - \frac{\operatorname{Re}(d\bar{p}/d\bar{x})}{s} \end{cases} \quad (4.101)$$

with the boundary conditions of

$$\begin{cases} \frac{\partial \bar{U}_1^E(\bar{z}, \bar{y}, s)}{\partial \bar{z}} = 0 & \text{when } \bar{z} = 0 \\ \bar{U}_1^E(\bar{z}, \bar{y}, s) = 0 & \text{when } \bar{z} = \bar{w} \\ \bar{U}_1^E(\bar{z}, \bar{y}, s) = 0 & \text{when } \bar{y} = -\bar{h} - \bar{h}_1 \end{cases} \quad (4.102)$$

and

$$\begin{cases} \frac{\partial \bar{U}_1^P(\bar{z}, \bar{y}, s)}{\partial \bar{z}} = 0 & \text{when } \bar{z} = 0 \\ \bar{U}_1^P(\bar{z}, \bar{y}, s) = 0 & \text{when } \bar{z} = \bar{w} \\ \bar{U}_1^P(\bar{z}, \bar{y}, s) = 0 & \text{when } \bar{y} = -\bar{h} - \bar{h}_1 \end{cases} \quad (4.103)$$

where s is the Laplace transform parameter.

For non-conducting fluid 2, the Laplace transform of Eq. (4.96) is

$$(\text{non-conducting fluid 2}) \begin{cases} \frac{s \operatorname{Re}}{\alpha_2} \bar{U}_2^E(\bar{z}, \bar{y}, s) = \frac{\partial^2 \bar{U}_2^E(\bar{z}, \bar{y}, s)}{\partial \bar{z}^2} + \frac{\partial^2 \bar{U}_2^E(\bar{z}, \bar{y}, s)}{\partial \bar{y}^2} \\ \frac{s \operatorname{Re}}{\alpha_2} \bar{U}_2^P(\bar{z}, \bar{y}, s) = \frac{\partial^2 \bar{U}_2^P(\bar{z}, \bar{y}, s)}{\partial \bar{z}^2} + \frac{\partial^2 \bar{U}_2^P(\bar{z}, \bar{y}, s)}{\partial \bar{y}^2} - \frac{\operatorname{Re}(d\bar{p}/d\bar{x})}{s\beta_2} \end{cases} \quad (4.104)$$

with the boundary conditions of

$$\begin{cases} \frac{\partial \bar{U}_2^E(\bar{z}, \bar{y}, s)}{\partial \bar{z}} = 0 & \text{when } \bar{z} = 0 \\ \bar{U}_2^E(\bar{z}, \bar{y}, s) = 0 & \text{when } \bar{z} = \bar{w} \end{cases} \quad (4.105)$$

and

$$\begin{cases} \frac{\partial \bar{U}_2^P(\bar{z}, \bar{y}, s)}{\partial \bar{z}} = 0 & \text{when } \bar{z} = 0 \\ \bar{U}_2^P(\bar{z}, \bar{y}, s) = 0 & \text{when } \bar{z} = \bar{w} \end{cases} \quad (4.106)$$

For conducting fluid 3, the Laplace transform of Eq. (4.97) is

$$\text{(conducting fluid 3)} \begin{cases} \frac{s \operatorname{Re}}{\alpha_3} \bar{U}_3^E(\bar{z}, \bar{y}, s) = \frac{\partial^2 \bar{U}_3^E(\bar{z}, \bar{y}, s)}{\partial \bar{z}^2} + \frac{\partial^2 \bar{U}_3^E(\bar{z}, \bar{y}, s)}{\partial \bar{y}^2} - \frac{\operatorname{Re}}{s \beta_3} G_{x3} \bar{\psi}_3(\bar{z}, \bar{y}) \\ \frac{s \operatorname{Re}}{\alpha_3} \bar{U}_3^P(\bar{z}, \bar{y}, s) = \frac{\partial^2 \bar{U}_3^P(\bar{z}, \bar{y}, s)}{\partial \bar{z}^2} + \frac{\partial^2 \bar{U}_3^P(\bar{z}, \bar{y}, s)}{\partial \bar{y}^2} - \frac{\operatorname{Re}(d\bar{p}/d\bar{x})}{s \beta_3} \end{cases} \quad (4.107)$$

with the boundary conditions of

$$\begin{cases} \frac{\partial \bar{U}_3^E(\bar{z}, \bar{y}, s)}{\partial \bar{z}} = 0 & \text{when } \bar{z} = 0 \\ \bar{U}_3^E(\bar{z}, \bar{y}, s) = 0 & \text{when } \bar{z} = \bar{w} \\ \bar{U}_3^E(\bar{z}, \bar{y}, s) = 0 & \text{when } \bar{y} = \bar{h} + \bar{h}_3 \end{cases} \quad (4.108)$$

and

$$\begin{cases} \frac{\partial \bar{U}_3^P(\bar{z}, \bar{y}, s)}{\partial \bar{z}} = 0 & \text{when } \bar{z} = 0 \\ \bar{U}_3^P(\bar{z}, \bar{y}, s) = 0 & \text{when } \bar{z} = \bar{w} \\ \bar{U}_3^P(\bar{z}, \bar{y}, s) = 0 & \text{when } \bar{y} = \bar{h} + \bar{h}_3 \end{cases} \quad (4.109)$$

The matching conditions are

$$\text{(interface 1-2)} \begin{cases} \bar{U}_1^E(\bar{z}, -\bar{h}, s) = \bar{U}_2^E(\bar{z}, -\bar{h}, s) \\ \frac{\partial \bar{U}_1^E(\bar{z}, -\bar{h}, s)}{\partial \bar{y}} = \beta_2 \frac{\partial \bar{U}_2^E(\bar{z}, -\bar{h}, s)}{\partial \bar{y}} - \frac{M_1 \bar{\rho}_{e1}^s(\bar{z})}{s} \end{cases} \quad (4.110)$$

$$\text{(interface 1-2)} \begin{cases} \bar{U}_1^P(\bar{z}, -\bar{h}, s) = \bar{U}_2^P(\bar{z}, -\bar{h}, s) \\ \frac{\partial \bar{U}_1^P(\bar{z}, -\bar{h}, s)}{\partial \bar{y}} = \beta_2 \frac{\partial \bar{U}_2^P(\bar{z}, -\bar{h}, s)}{\partial \bar{y}} \end{cases} \quad (4.111)$$

$$(\text{ interface 2-3 }) \left\{ \begin{aligned} \bar{U}_3^E(\bar{z}, \bar{h}, s) &= \bar{U}_2^E(\bar{z}, \bar{h}, s) \\ \frac{\partial \bar{U}_3^E(\bar{z}, \bar{h}, s)}{\partial \bar{y}} &= (\beta_2/\beta_3) \frac{\partial \bar{U}_2^E(\bar{z}, \bar{h}, s)}{\partial \bar{y}} - \frac{M_3 \bar{\rho}_{e3}^s(\bar{z})}{s} \end{aligned} \right. \quad (4.112)$$

$$(\text{ interface 2-3 }) \left\{ \begin{aligned} \bar{U}_3^P(\bar{z}, \bar{h}, s) &= \bar{U}_2^P(\bar{z}, \bar{h}, s) \\ \frac{\partial \bar{U}_3^P(\bar{z}, \bar{h}, s)}{\partial \bar{y}} &= (\beta_2/\beta_3) \frac{\partial \bar{U}_2^P(\bar{z}, \bar{h}, s)}{\partial \bar{y}} \end{aligned} \right. \quad (4.113)$$

Using the separation variables method, the solutions of Eq. (4.101) with the boundaries of Eq. (4.102) and Eq. (4.103) are given as

$$\begin{aligned} \bar{U}_1^E(\bar{z}, \bar{y}, s) &= \sqrt{\frac{2}{\bar{w}}} \sum_{j=0}^{\infty} \{ \{ \tanh[D_{1j}(\bar{h} + \bar{h}_1)] \cosh(D_{1j}\bar{y}) + \sinh(D_{1j}\bar{y}) \} b_{1j}^E \\ &\quad - \frac{\phi_{1j}(-\bar{h} - \bar{h}_1, s)}{\cosh[D_{1j}(\bar{h} + \bar{h}_1)]} \cosh(D_{1j}\bar{y}) + \phi_{1j}(\bar{y}, s) \} \cos(\lambda_j \bar{z}) \end{aligned} \quad (4.114)$$

$$\begin{aligned} \bar{U}_1^P(\bar{y}, \bar{z}, s) &= \sqrt{\frac{2}{\bar{w}}} \sum_{j=0}^{\infty} \left\{ \frac{\text{Re}}{s \lambda_j D_{1j}^2} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} (-1)^j \left\{ \frac{\cosh(D_{1j}\bar{y})}{\cosh[D_{1j}(\bar{h}_1 + \bar{h})]} - 1 \right\} \right. \\ &\quad \left. + \{ \tanh[D_{1j}(\bar{h}_1 + \bar{h})] \cosh(D_{1j}\bar{y}) + \sinh(D_{1j}\bar{y}) \} b_{1j}^P \right\} \cos(\lambda_j \bar{z}) \end{aligned} \quad (4.115)$$

Same as before, the solutions for non-conducting fluid 2 and for conducting fluid 3 are

$$\bar{U}_2^E(\bar{z}, \bar{y}, s) = \sqrt{\frac{2}{\bar{w}}} \sum_{j=0}^{\infty} [a_{2j}^E \cosh(E_{2j}\bar{y}) + a_{2j}^E \sinh(E_{2j}\bar{y})] \cos(\lambda_j \bar{z}) \quad (4.116)$$

$$\begin{aligned} \bar{U}_2^P(\bar{z}, \bar{y}, s) &= \sum_{j=1}^{\infty} \{ a_{2j}^P \cosh(E_{2j}\bar{y}) + b_{2j}^P \sinh(E_{2j}\bar{y}) \\ &\quad - \text{Re} \frac{d\bar{P}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} \frac{(-1)^j}{s \beta_2 \lambda_j E_{2j}^2} \} \cos(\lambda_j \bar{z}) \end{aligned} \quad (4.117)$$

and

$$\begin{aligned} \bar{U}_3^E(\bar{z}, \bar{y}, s) &= \sum_{j=0}^{\infty} \{ \{ -\tanh[E_{3j}(\bar{h} + \bar{h}_3)] \cosh(E_{3j}\bar{y}) + \sinh(E_{3j}\bar{y}) \} b_{3j}^E \\ &\quad - \frac{\phi_{3j}(\bar{h} + \bar{h}_3, s)}{\cosh[E_{3j}(\bar{h} + \bar{h}_3)]} \cosh(E_{3j}\bar{y}) + \phi_{3j}(\bar{y}, s) \} \cos(\lambda_j \bar{z}) \end{aligned} \quad (4.118)$$

$$\begin{aligned}\bar{U}_3^P(\bar{z}, \bar{y}, s) = & \sqrt{\frac{2}{\bar{w}}} \sum_{j=0}^{\infty} \left\{ \frac{1}{E_{3j}^2} \frac{\text{Re}}{\lambda_j s \beta_3} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} (-1)^j \left\{ \frac{\cosh(E_{3j} \bar{y})}{\cosh[E_{3j}(\bar{h}_3 + \bar{h})]} - 1 \right\} \right. \\ & \left. + \{-\tanh[E_{3j}(\bar{h}_3 + \bar{h})] \cosh(E_{3j} \bar{y}) + \sinh(E_{3j} \bar{y})\} b_{3j}^P \right\} \cos(\lambda_j \bar{z})\end{aligned}\quad (4.119)$$

where

$$D_{1j} = \sqrt{(\lambda_j^2 + s\text{Re})} \quad (4.120)$$

$$E_{2j} = \sqrt{\lambda_j^2 + s\text{Re}/\alpha_2} \quad (4.121)$$

$$E_{3j} = \sqrt{\lambda_j^2 + s\text{Re}/\alpha_3} \quad (4.122)$$

Integrate the matching conditions of Eqs. (4.110)-(4.113), the coefficients $b_{1j}^E, a_{2j}^E,$

$b_{2j}^E, b_{3j}^E, b_{1j}^P, a_{2j}^P, b_{2j}^P,$ and b_{3j}^P can be obtained as

$$b_{1j}^E = \frac{b_{2j}^E}{A} + H \quad (4.123)$$

$$a_{2j}^E = Jb_{1j}^E + Kb_{3j}^E + L \quad (4.124)$$

$$b_{2j}^E = Cb_{3j}^E + Db_{1j}^E + G \quad (4.125)$$

$$b_{3j}^E = \frac{b_{2j}^E}{B} + I \quad (4.126)$$

$$b_{1j}^P = \frac{b_{2j}^P}{S} - \frac{T}{S} \quad (4.127)$$

$$a_{2j}^P = Nb_{3j}^P + Qb_{1j}^P + M \quad (4.128)$$

$$b_{2j}^P = \frac{O - \frac{RT}{S} - \frac{PU}{V}}{1 - \frac{R}{S} - \frac{P}{V}} \quad (4.129)$$

$$b_{3j}^P = \frac{b_{2j}^P}{V} - \frac{U}{V} \quad (4.130)$$

where

$$\begin{aligned}
A = & \tanh[D_{1j}(\bar{h} + \bar{h}_1)] \cosh(D_{1j}\bar{h}) \sinh(E_{2j}\bar{h}) - \sinh(D_{1j}\bar{h}) \sinh(E_{2j}\bar{h}) \\
& - \frac{D_{1j}}{\beta_1 E_{2j}} \tanh[D_{1j}(\bar{h} + \bar{h}_1)] \sinh(D_{1j}\bar{h}) \cosh(E_{2j}\bar{h}) \\
& + \frac{D_{1j}}{\beta_1 E_{2j}} \cosh(D_{1j}\bar{h}) \cosh(E_{2j}\bar{h})
\end{aligned} \tag{4.131}$$

$$\begin{aligned}
B = & -\frac{E_{3j}}{(\beta_2 / \beta_3) E_{2j}} \tanh[E_{3j}(\bar{h} + \bar{h}_3)] \sinh(E_{3j}\bar{h}) \cosh(E_{2j}\bar{h}) \\
& + \frac{E_{3j}}{(\beta_2 / \beta_3) E_{2j}} \cosh(E_{3j}\bar{h}) \cosh(E_{2j}\bar{h}) \\
& + \tanh[E_{3j}(\bar{h} + \bar{h}_3)] \cosh(E_{3j}\bar{h}) \sinh(E_{2j}\bar{h}) - \sinh(E_{3j}\bar{h}) \sinh(E_{2j}\bar{h})
\end{aligned} \tag{4.132}$$

$$C = -\frac{\sinh(E_{3j}\bar{h})}{2 \cosh[E_{3j}(\bar{h} + \bar{h}_3)] \sinh(E_{2j}\bar{h})} \tag{4.133}$$

$$D = -\frac{\sinh(D_{1j}\bar{h})}{2 \cosh[D_{1j}(\bar{h} + \bar{h}_1)] \sinh(E_{2j}\bar{h})} \tag{4.134}$$

$$\begin{aligned}
G = & -\frac{\phi_{3j}(\bar{h} + \bar{h}_3, s) \cosh(E_{3j}\bar{h})}{2 \sinh(E_{2j}\bar{h}) \cosh[E_{3j}(\bar{h} + \bar{h}_3)]} + \frac{\phi_{3j}(\bar{h}, s)}{2 \sinh(E_{2j}\bar{h})} \\
& + \frac{\phi_{1j}(-\bar{h} - \bar{h}_1, s) \cosh(D_{1j}\bar{h})}{2 \sinh(E_{2j}\bar{h}) \cosh[D_{1j}(\bar{h} + \bar{h}_1)]} - \frac{\phi_{1j}(-\bar{h}, s)}{2 \sinh(E_{2j}\bar{h})}
\end{aligned} \tag{4.135}$$

$$\begin{aligned}
H = & \frac{\phi_{1j}(-\bar{h} - \bar{h}_1, s)}{A \cosh[D_{1j}(\bar{h} + \bar{h}_1)]} \sinh(E_{2j}\bar{h}) \cosh(D_{1j}\bar{h}) - \frac{\sinh(E_{2j}\bar{h}) \phi_{1j}(-\bar{h}, s)}{A} \\
& - \frac{M_1 \bar{\rho}_{q1}^s(\bar{z}) \cosh(E_{2j}\bar{h})}{A \beta_1 E_{2j}} - \frac{\phi'_{1j}(-\bar{h}, s) \cosh(E_{2j}\bar{h})}{A \beta_1 E_{2j}} \\
& - \frac{D_{1j}}{A \beta_1 E_{2j}} \frac{\phi_{1j}(-\bar{h} - \bar{h}_1, s)}{\cosh[D_{1j}(\bar{h} + \bar{h}_1)]} \sinh(D_{1j}\bar{h}) \cosh(E_{2j}\bar{h})
\end{aligned} \tag{4.136}$$

$$\begin{aligned}
I = & \frac{E_{3j} \cosh(E_{2j}\bar{h})}{B(\beta_2 / \beta_3) E_{2j}} \frac{\phi_{3j}(\bar{h} + \bar{h}_3, s)}{\cosh[E_{3j}(\bar{h} + \bar{h}_3)]} \sinh(E_{3j}\bar{h}) - \frac{\phi'_{3j}(\bar{h}, s) \cosh(E_{2j}\bar{h})}{B(\beta_2 / \beta_3) E_{2j}} \\
& - \frac{M_3 \bar{\rho}_{q3}^s(\bar{z}) \cosh(E_{2j}\bar{h})}{B(\beta_2 / \beta_3) E_{2j}} + \frac{\sinh(E_{2j}\bar{h}) \phi_{3j}(\bar{h}, s)}{B} \\
& - \frac{\phi_{3j}(\bar{h} + \bar{h}_3, s)}{B \cosh[E_{3j}(\bar{h} + \bar{h}_3)]} \sinh(E_{2j}\bar{h}) \cosh(E_{3j}\bar{h})
\end{aligned} \tag{4.137}$$

$$J = \frac{\tanh[D_{1j}(\bar{h} + \bar{h}_1)] \cosh(D_{1j}\bar{h}) - \sinh(D_{1j}\bar{h})}{2 \cosh(E_{2j}\bar{h})} \quad (4.138)$$

$$K = \frac{-\tanh[E_{3j}(\bar{h} + \bar{h}_3)] \cosh(E_{3j}\bar{h}) + \sinh(E_{3j}\bar{h})}{2 \cosh(E_{2j}\bar{h})} \quad (4.139)$$

$$L = -\frac{\phi_{3j}(\bar{h} + \bar{h}_3, s) \cosh(E_{3j}\bar{h})}{2 \cosh(E_{2j}\bar{h}) \cosh[E_{3j}(\bar{h} + \bar{h}_3)]} + \frac{\phi_{3j}(\bar{h}, s)}{2 \cosh(E_{2j}\bar{h})} \\ - \frac{\phi_{1j}(-\bar{h} - \bar{h}_1, s) \cosh(D_{1j}\bar{h})}{2 \cosh(E_{2j}\bar{h}) \cosh[D_{1j}(\bar{h} + \bar{h}_1)]} + \frac{\phi_{1j}(-\bar{h}, s)}{2 \cosh(E_{2j}\bar{h})} \quad (4.140)$$

$$M = \frac{\text{Re}}{\lambda_j s} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} (-1)^j \left\{ \frac{\cosh(E_{3j}\bar{h})}{2E_{3j}^2 \beta_3 \cosh(E_{2j}\bar{h}) \cosh[E_{3j}(\bar{h}_3 + \bar{h})]} \right\} \\ + \frac{\text{Re}}{s\lambda_j} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} (-1)^j \left\{ \frac{\cosh(D_{1j}\bar{h})}{2D_{1j}^2 \cosh(E_{2j}\bar{h}) \cosh[D_{1j}(\bar{h}_1 + \bar{h})]} \right\} \\ - \frac{\text{Re}}{s\lambda_j} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} (-1)^j \left\{ \frac{1}{2E_{3j}^2 \beta_3 \cosh(E_{2j}\bar{h})} + \frac{1}{2D_{1j}^2 \cosh(E_{2j}\bar{h})} \right\} \\ + \text{Re} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} \frac{(-1)^j}{s\beta_2 \lambda_j E_{2j}^2 \cosh(E_{2j}\bar{h})} \quad (4.141)$$

$$N = -\frac{\sinh(E_{3j}\bar{h}_3)}{2 \cosh(E_{2j}\bar{h}) \cosh[E_{3j}(\bar{h}_3 + \bar{h})]} \quad (4.142)$$

$$O = \frac{\text{Re}}{\lambda_j s} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} (-1)^j \left\{ \frac{\cosh(E_{3j}\bar{h})}{2E_{3j}^2 \beta_3 \sinh(E_{2j}\bar{h}) \cosh[E_{3j}(\bar{h}_3 + \bar{h})]} \right\} \\ - \frac{\text{Re}}{s\lambda_j} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} (-1)^j \left\{ \frac{\cosh(D_{1j}\bar{h})}{2D_{1j}^2 \sinh(E_{2j}\bar{h}) \cosh[D_{1j}(\bar{h}_1 + \bar{h})]} \right\} \\ - \frac{\text{Re}}{s\lambda_j} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} (-1)^j \left\{ \frac{1}{2E_{3j}^2 \beta_3 \sinh(E_{2j}\bar{h})} - \frac{1}{2D_{1j}^2 \sinh(E_{2j}\bar{h})} \right\} \quad (4.143)$$

$$P = -\frac{\sinh(E_{3j}\bar{h}_3)}{2 \sinh(E_{2j}\bar{h}) \cosh[E_{3j}(\bar{h}_3 + \bar{h})]} \quad (4.144)$$

$$Q = \frac{\sinh(D_{1j}\bar{h}_1)}{2 \cosh(E_{2j}\bar{h}) \cosh[D_{1j}(\bar{h}_1 + \bar{h})]} \quad (4.145)$$

$$R = -\frac{\sinh(D_{1j}\bar{h}_1)}{2 \sinh(E_{2j}\bar{h}) \cosh[D_{1j}(\bar{h}_1 + \bar{h})]} \quad (4.146)$$

$$S = \frac{\sinh(D_{1j}\bar{h}_1)\sinh(E_{2j}\bar{h})}{\cosh[D_{1j}(\bar{h}_1 + \bar{h})]} + \frac{D_{1j} \cosh(D_{1j}\bar{h}_1)\cosh(E_{2j}\bar{h})}{\beta_2 E_{2j} \cosh[D_{1j}(\bar{h}_1 + \bar{h})]} \quad (4.147)$$

$$\begin{aligned} T = & \frac{Re}{s\lambda_j D_{1j}^2} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} (-1)^j \left\{ \frac{\cosh(D_{1j}\bar{h})\sinh(E_{2j}\bar{h})}{\cosh[D_{1j}(\bar{h}_1 + \bar{h})]} - \sinh(E_{2j}\bar{h}) \right\} \\ & - \frac{Re}{s\beta_2 \lambda_j D_{1j} E_{2j}} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} (-1)^j \frac{\sinh(D_{1j}\bar{h})\cosh(E_{2j}\bar{h})}{\cosh[D_{1j}(\bar{h}_1 + \bar{h})]} \\ & + Re \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} \frac{(-1)^j \sinh(E_{2j}\bar{h})}{s\beta_2 \lambda_j E_{2j}^2} \end{aligned} \quad (4.148)$$

$$\begin{aligned} U = & \frac{1}{E_{3j}} \frac{Re}{\lambda_j s \beta_1 E_{2j}} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} (-1)^j \frac{\sinh(E_{3j}\bar{h})\cosh(E_{2j}\bar{h})}{\cosh[E_{3j}(\bar{h}_3 + \bar{h})]} \\ & - \frac{1}{E_{3j}^2} \frac{Re}{\lambda_j s \beta_3} \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} (-1)^j \left\{ \frac{\cosh(E_{3j}\bar{h})\sinh(E_{2j}\bar{h})}{\cosh[E_{3j}(\bar{h}_3 + \bar{h})]} - \sinh(E_{2j}\bar{h}) \right\} \\ & - Re \frac{d\bar{p}}{d\bar{x}} \sqrt{\frac{2}{\bar{w}}} \frac{(-1)^j \sinh(E_{2j}\bar{h})}{s\beta_2 \lambda_j E_{2j}^2} \end{aligned} \quad (4.149)$$

$$V = \frac{\sinh(E_{2j}\bar{h})\sinh(E_{3j}\bar{h}_3)}{\cosh[E_{3j}(\bar{h}_3 + \bar{h})]} + \frac{E_{3j} \cosh(E_{3j}\bar{h}_3)\cosh(E_{2j}\bar{h})}{(\beta_2 / \beta_3) E_{2j} \cosh[E_{3j}(\bar{h}_3 + \bar{h})]} \quad (4.150)$$

$$\begin{aligned} \phi_{1j}(\bar{y}, s) = & \sqrt{\frac{\bar{w}}{2}} \frac{4 \operatorname{Re} G_{x1} (-1)^{j+1}}{(2j+1)s\pi \sinh[B_j \bar{h}_1]} \\ & \times \left\{ \bar{\xi}_1 \frac{\sinh[B_j(\bar{h} + \bar{y})]}{(B_j^2 - D_{1j}^2)} - \bar{\xi}_3 \frac{\sinh[B_j(\bar{h}_1 + \bar{h} + \bar{y})]}{(B_j^2 - D_{1j}^2)} \right\} \\ & + \sqrt{\frac{2}{\bar{w}}} \sum_{p=1}^{\infty} \frac{2 \operatorname{Re} G_{x1} \bar{\xi}_2 \lambda_j}{p\pi s(A_{p1}^2 + \lambda_j^2)} [(-1)^p - 1] \times (-1)^{j+1} \frac{\sin[\frac{p\pi}{h_1}(\bar{h} + \bar{y})]}{[(\frac{p\pi}{h_1})^2 + D_{1j}^2]} \end{aligned} \quad (4.151)$$

$$\begin{aligned} \phi'_{1j}(\bar{y}, s) = & \sqrt{\frac{\bar{w}}{2}} \frac{4 \operatorname{Re} G_{x1} B_j (-1)^{j+1}}{(2j+1)s\pi \sinh[B_j \bar{h}_1]} \\ & \times \left\{ \bar{\xi}_1 \frac{\cosh[B_j(\bar{h} + \bar{y})]}{(B_j^2 - D_{1j}^2)} - \bar{\xi}_3 \frac{\cosh[B_j(\bar{h}_1 + \bar{h} + \bar{y})]}{(B_j^2 - D_{1j}^2)} \right\} \\ & + \sqrt{\frac{2}{\bar{w}}} \sum_{p=1}^{\infty} \frac{2 \operatorname{Re} G_{x1} \bar{\xi}_2 \lambda_j}{\bar{h}_1 s(A_{p1}^2 + \lambda_j^2)} [(-1)^p - 1] \times (-1)^{j+1} \frac{\cos[\frac{p\pi}{h_1}(\bar{h} + \bar{y})]}{[(\frac{p\pi}{h_1})^2 + D_{1j}^2]} \end{aligned} \quad (4.152)$$

$$\begin{aligned}
\phi_{3j}(\bar{y}, s) = & \sum_{j=1}^{\infty} \frac{4ReG_{x3}(-1)^{j+1}}{(2j-1)s\pi\beta_3 \sinh[B_j\bar{h}_3](B_j^2 - \lambda_j^2 - s)} \\
& \times \{ \bar{\xi}_4 \sinh[B_j(\bar{y} - \bar{h})] - \bar{\xi}_6 \sinh[B_j(\bar{y} - \bar{h}_3 - \bar{h})] \} \\
& + \sum_{p=1}^{\infty} \frac{4\bar{\xi}_2(-1)^{j+1}\lambda_j}{p\pi\bar{w}s(A_{p3}^2 + \lambda_j^2)[(\frac{p\pi}{\bar{h}_3})^2 + \lambda_j^2 + s]} \\
& \times [(-1)^{p+1} + 1] \times \sin[\frac{p}{\bar{h}_3}\pi(\bar{y} - \bar{h})]
\end{aligned} \tag{4.153}$$

$$\begin{aligned}
\phi'_{3j}(\bar{y}, s) = & \sum_{j=1}^{\infty} \frac{4ReB_jG_{x3}(-1)^{j+1}}{(2j-1)s\pi\beta_3 \sinh[B_j\bar{h}_3](B_j^2 - \lambda_j^2 - s)} \\
& \times \{ \bar{\xi}_4 \cosh[B_j(\bar{y} - \bar{h})] - \bar{\xi}_6 \cosh[B_j(\bar{y} - \bar{h}_3 - \bar{h})] \} \\
& + \sum_{p=1}^{\infty} \frac{4\bar{\xi}_2(-1)^{j+1}\lambda_j}{\bar{w}\bar{h}_3s(A_{p3}^2 + \lambda_j^2)[(\frac{p\pi}{\bar{h}_3})^2 + \lambda_j^2 + s]} \\
& \times [(-1)^{p+1} + 1] \times \sin[\frac{p}{\bar{h}_3}\pi(\bar{y} - \bar{h})]
\end{aligned} \tag{4.154}$$

The transient volumetric flowrates for three liquids are defined as

$$\bar{q}_1^E(\bar{t}) = 2 \int_{-\bar{h}-\bar{h}_1}^{-\bar{h}} \int_0^{\bar{w}} \bar{u}_1^E(\bar{y}, \bar{z}, \bar{t}) d\bar{z} d\bar{y} \tag{4.155}$$

$$\bar{q}_1^P(\bar{t}) = 2 \int_{-\bar{h}-\bar{h}_1}^{-\bar{h}} \int_0^{\bar{w}} \bar{u}_1^P(\bar{y}, \bar{z}, \bar{t}) d\bar{z} d\bar{y} \tag{4.156}$$

$$\bar{q}_2^E(\bar{t}) = 2 \int_{-\bar{h}}^{\bar{h}} \int_0^{\bar{w}} \bar{u}_2^E(\bar{y}, \bar{z}, \bar{t}) d\bar{z} d\bar{y} \tag{4.157}$$

$$\bar{q}_2^P(\bar{t}) = 2 \int_{-\bar{h}}^{\bar{h}} \int_0^{\bar{w}} \bar{u}_2^P(\bar{y}, \bar{z}, \bar{t}) d\bar{z} d\bar{y} \tag{4.158}$$

$$\bar{q}_3^E(\bar{t}) = 2 \int_{\bar{h}}^{\bar{h}+\bar{h}_3} \int_0^{\bar{w}} \bar{u}_3^E(\bar{y}, \bar{z}, \bar{t}) d\bar{z} d\bar{y} \tag{4.159}$$

$$\bar{q}_3^P(\bar{t}) = 2 \int_{\bar{h}}^{\bar{h}+\bar{h}_3} \int_0^{\bar{w}} \bar{u}_3^P(\bar{y}, \bar{z}, \bar{t}) d\bar{z} d\bar{y} \tag{4.160}$$

The Laplace transform of Eqs.(4.155)-(4.160) are

$$\bar{Q}_1^E(s) = 2 \int_{-\bar{h}-\bar{h}_1}^{-\bar{h}} \int_0^{\bar{w}} \bar{U}_1^E(\bar{y}, \bar{z}, s) d\bar{z} d\bar{y} \quad (4.161)$$

$$\bar{Q}_1^P(s) = 2 \int_{-\bar{h}-\bar{h}_1}^{-\bar{h}} \int_0^{\bar{w}} \bar{U}_1^P(\bar{y}, \bar{z}, s) d\bar{z} d\bar{y} \quad (4.162)$$

$$\bar{Q}_2^E(s) = 2 \int_{-\bar{h}}^{\bar{h}} \int_0^{\bar{w}} \bar{U}_2^E(\bar{y}, \bar{z}, s) d\bar{z} d\bar{y} \quad (4.163)$$

$$\bar{Q}_2^P(s) = 2 \int_{-\bar{h}}^{\bar{h}} \int_0^{\bar{w}} \bar{U}_2^P(\bar{y}, \bar{z}, s) d\bar{z} d\bar{y} \quad (4.164)$$

$$\bar{Q}_3^E(s) = 2 \int_{\bar{h}}^{\bar{h}+\bar{h}_1} \int_0^{\bar{w}} \bar{U}_3^E(\bar{y}, \bar{z}, s) d\bar{z} d\bar{y} \quad (4.165)$$

$$\bar{Q}_3^P(s) = 2 \int_{\bar{h}}^{\bar{h}+\bar{h}_1} \int_0^{\bar{w}} \bar{U}_3^P(\bar{y}, \bar{z}, s) d\bar{z} d\bar{y} \quad (4.166)$$

Substituting Eqs. (4.114) - (4.119) into Eqs. (4.161) - (4.166), respectively, the dimensionless volumetric flowrates are obtained as

$$\begin{aligned} \bar{Q}_1^E(s) = & \sum_{j=0}^{\infty} \frac{2 \sin(\lambda_j \bar{w})}{\lambda_j} \left\{ \frac{1}{D_{1j}} \times \tanh[D_{1j}(\bar{h} + \bar{h}_1)] \right. \\ & \times [\sinh(D_{1j}\bar{h}) - \sinh(D_{1j}(\bar{h} + \bar{h}_1))] \\ & + \frac{1}{D_{1j}} [\cosh(D_{1j}(\bar{h} + \bar{h}_1)) + \cosh(D_{1j}\bar{h})] \} b_{1j}^E \\ & + \sum_{j=0}^{\infty} \frac{2 \sin(\lambda_j \bar{w})}{\lambda_j} \{ \phi_{1j}'(-\bar{h} - \bar{h}_1, s) - \phi_{1j}'(-\bar{h}, s) \\ & + \frac{1}{D_{1j}} \frac{\phi_{1j}'(-\bar{h} - \bar{h}_1, s)}{\cosh[D_{1j}(\bar{h} + \bar{h}_1)]} [\sinh(D_{1j}\bar{h}) - \sinh(D_{1j}(\bar{h} + \bar{h}_1))] \} \end{aligned} \quad (4.167)$$

$$\begin{aligned}
\bar{Q}_1^p(s) = & \sum_{j=0}^{\infty} \frac{2}{\lambda_j} \sin(\lambda_j \bar{w}) \frac{\text{Re}}{s D_{1j}^2} \frac{d\bar{p}}{d\bar{x}} \frac{4 \times (-1)^{j+1}}{(2j-1)\pi} \frac{1}{D_{1j}} \\
& \times \left\{ \frac{\sinh(D_{1j} \bar{h}) - \sinh(D_{1j}(\bar{h} + \bar{h}_1))}{\cosh[D_{1j}(\bar{h}_1 + \bar{h})]} + \bar{h}_1 \right\} \\
& + \sum_{j=0}^{\infty} \frac{2}{\lambda_j} \sin(\lambda_j \bar{w}) \frac{\text{Re}}{s D_{1j}^2} \frac{d\bar{p}}{d\bar{x}} \frac{4 \times (-1)^{j+1}}{(2j-1)\pi} \frac{1}{D_{1j}} \\
& \times \{ \tanh[D_{1j}(\bar{h}_1 + \bar{h})][\sinh(D_{1j} \bar{h}) - \sinh(D_{1j}(\bar{h} + \bar{h}_1))] \\
& + [\cosh(D_{1j}(\bar{h} + \bar{h}_1)) - \cosh(D_{1j} \bar{h})] \} b_{1j}^p
\end{aligned} \tag{4.168}$$

$$\bar{Q}_2^E(s) = 4 \sum_{j=0}^{\infty} \frac{a_{2j}^E \sinh(E_{2j} \bar{h}) \sin(\lambda_j \bar{w})}{\lambda_j E_{2j}} \tag{4.169}$$

$$\bar{Q}_2^p(s) = 4 \sum_{j=0}^{\infty} \left[a_{2j}^p \sinh(E_{2j} \bar{h}) - \frac{1}{E_{2j}^2} \frac{\text{Re}}{s \beta_2} \frac{4 \times (-1)^{j+1}}{(2j-1)\pi} \bar{h} \right] \frac{\sin(\lambda_j \bar{w})}{\lambda_j E_{2j}} \tag{4.170}$$

$$\begin{aligned}
\bar{Q}_3^E(s) = & 2 \sum_{j=0}^{\infty} \left\{ \frac{1}{E_{3j}} \{ -\tanh[E_{3j}(\bar{h} + \bar{h}_3)][\sinh(E_{3j}(\bar{h} + \bar{h}_3)) - \sinh(E_{3j} \bar{h})] \right. \\
& + \cosh(E_{3j}(\bar{h} + \bar{h}_3)) - \cosh(E_{3j} \bar{h}) \} b_{3j}^E \\
& + 2 \sum_{j=0}^{\infty} \frac{\sin(\lambda_j \bar{w})}{\lambda_j} \{ \phi_{3j}^I(\bar{h} + \bar{h}_3, s) - \phi_{3j}^I(\bar{h}, s) \\
& \left. - \frac{1}{E_{3j}} \frac{\phi_{3j}(\bar{h} + \bar{h}_3, s)}{\cosh[E_{3j}(\bar{h} + \bar{h}_3)]} [\sinh(E_{3j}(\bar{h} + \bar{h}_3)) - \sinh(E_{3j} \bar{h})] \} \right\}
\end{aligned} \tag{4.171}$$

$$\begin{aligned}
\bar{Q}_3^p(s) = & 2 \sum_{j=0}^{\infty} \left\{ \frac{\sin(\lambda_j \bar{z})}{\lambda_j E_{3j}} \frac{1}{E_{3j}^2} \frac{\text{Re}}{s \beta_3} \frac{d\bar{p}}{d\bar{x}} \frac{4 \times (-1)^{j+1}}{(2j-1)\pi} \right. \\
& \times \frac{1}{E_{3j}} \left\{ \frac{\sinh(E_{3j}(\bar{h} + \bar{h}_3)) - \sinh(E_{3j} \bar{h})}{\cosh[E_{3j}(\bar{h}_3 + \bar{h})]} - \bar{h}_3 \right\} \\
& + \{ -\tanh[E_{3j}(\bar{h}_3 + \bar{h})][\sinh(E_{3j}(\bar{h} + \bar{h}_3)) - \sinh(E_{3j} \bar{h})] \\
& \left. + [\cosh(E_{3j}(\bar{h} + \bar{h}_3)) - \cosh(E_{3j} \bar{h})] \} b_{3j}^p \right\}
\end{aligned} \tag{4.172}$$

To obtain the final results, it is required to applied inversions of the Laplace transform on velocities $\bar{U}^E(\bar{y}, \bar{z}, s)$, $\bar{U}^p(\bar{y}, \bar{z}, s)$ and flowrates $\bar{Q}^E(s)$, $\bar{Q}^p(s)$. Due to the complex nature of those forms, the numerical inversion of Laplace transforms method-Gaver-Stehfest method with double precision is used in this report [179].

Results of the transient three-liquid flow model

In this section, the fluid fractions for conducting fluid 1 and conducting fluid 3 are set at 0.3, respectively, and the non-conducting fluid holds the middle part of the channel, the fraction is 0.4. Many methods for determining the zeta potentials at the wall and at the interface were proposed [26]. The zeta potentials at the channel walls, $\bar{\xi}_{wall}$ ($\bar{\xi}_1, \bar{\xi}_2, \bar{\xi}_4, \bar{\xi}_5$) depend on the material properties of the wall and the ionic properties of the fluid [32]. We choose the zeta potentials of the walls to be the same at $\bar{\xi}_{wall}$ ($\bar{\xi}_1, \bar{\xi}_2, \bar{\xi}_4, \bar{\xi}_5$) = -25.7 mV as the default value. The zeta potential between two immiscible liquids, $\bar{\xi}_3$ and $\bar{\xi}_6$, does not only depend on the ionic properties of two fluids, but also on the pH value and the concentration of the electrolyte [180, 181]. The dimensionless parameter K is defined as $K = \kappa L_{ref}$ to evaluate parameters affecting EDL profiles. $1/\kappa$ refers to the characteristic thickness of the EDL. As the Debye-Hückel parameter in Eq. (4.5) is proportional to the square root of the bulk ionic concentration n_0 , the variation of the ionic concentration will alter the EDL thickness. In this analysis, the concentration of the two conducting fluids is in the range of $10^{-6} \sim 10^{-5}$ M, therefore, the bulk concentration $n_0 = 6.022 \times 10^{20} \sim 6.022 \times 10^{21} \text{ m}^{-3}$ and the EDL dimension parameter $K = 87 \sim 275$.

Fig. 4.15 shows the potential distribution along the symmetrical line when the interface zeta potential $\bar{\xi}_3 = \bar{\xi}_6 = \bar{\xi}_{wall}$. The EDL only exists in the two conducting fluids. The interface zeta potential induces an EDL at the liquid-liquid interface, where the electric body forces are present. The result shows that the interface zeta potential affects the electric potential distribution dramatically.

Points A, B, C are indicated on Fig. 4.15. Point A is within the EDL at $\bar{y} = 0.9925$, point B corresponds to flow of conducting fluid 1, outside the EDL at $\bar{y} = 0.85$. Point C is in the non-conducting fluid region. The dynamic response of the flow in points A, B and C will be discussed in Fig. 4.21.

From Eq. (4.31), the velocity $\bar{u}$ of three-fluid flow can be decomposed into two parts, $\bar{u}^p$ and $\bar{u}^E$. $\bar{u}^p$ corresponds to velocity driven by pressure gradient and $\bar{u}^E$ corresponds to velocity driven by electroosmotic effect. With the proposed transient model, we investigate the following cases: (a) With zero pressure gradient is applied

across the microchannel, the flow is simply a three-fluid electroosmotic flow with the flow velocity $\bar{u}^E$. (b) When both the pressure gradient and the electric field are applied across the microchannel, the three-fluid flow is driven by the combined electroosmotic force and pressure gradient with the flow velocity $\bar{u}$.

Case (a) Three-fluid electroosmotic flow

Figs. 4.16 - 4.17 show the time evolution of the $\bar{u}^E$ velocity profiles, at the symmetric line when the flow parameters are : $d\bar{p}/d\bar{x} = 0$, $E_{x1} = E_{x3} = 3000 \text{ V/m}$, $K = 130$, $\alpha_1 = \alpha_2 = \alpha_3 = 1$, $\beta_1 = \beta_2 = \beta_3 = 1$, and interface zeta potential at $\bar{\xi}_{wall}$ and 0, respectively. The viscosity and density of the NaCl solution is $\mu_{ref} = 10^{-3} \text{ Pa s}$ and $\rho_{ref} = 10^3 \text{ kg/m}^3$, respectively. With these reference potential and viscosity, the Helmholtz-Smoluchowski electroosmotic velocity is chosen as the reference velocity

$$U_{ref} = \frac{E_{x1} \varepsilon \bar{\xi}_{ref}}{\mu_{ref}} = 1.59 \times 10^{-4} \text{ m/s}. \text{ The corresponding Reynolds number is } Re \approx 0.0063.$$

Fig. 4.16 shows the time evolution of the velocity profile when the interface zeta potentials $\bar{\xi}_3 = \bar{\xi}_6 = \bar{\xi}_{wall}$. It can be seen that upon the application of the electric fields, the conducting fluids are activated in regions close to the channel walls and the interfaces. The velocities of the two conducting fluids increase rapidly from zero in the same direction within the EDL regions close to the walls and the interfaces. But the effects of the force from the free charges of the interface cause the interface velocity increases rapidly in the opposite direction. This is because the interface charges are equal to the total net charges within the EDL regions, close to the interface. The velocity at the interface region is significantly influenced by the surface force during the initial transient state. As time elapses, the liquid inside the EDLs exerts hydrodynamic shear stress on its adjacent liquid; the liquid outside the EDL regions may be considered as ‘passive’ flow caused by shear viscous forces.

For the non-conducting fluid, during the initial transient time, it moves in the opposite direction. As time elapse, because the EDLs exert the hydrodynamic shear stress

on the adjacent conducting fluids, the negative velocity reduces. The characteristic of the non-conducting liquid depends on the matching conditions between the three liquids, which involve the external electric fields, the viscosity ratio of the liquids, the interface phenomenon, and the charge density in the conducting liquids.

Fig. 4.17 shows the velocity profile when the interface zeta potential $\bar{\xi}_3 = \bar{\xi}_6 = 0$. Comparing Fig. 4.16 and Fig. 4.17, we find that: the velocity profile is completely different in the absence of the surface charge. Upon the application of the electric field, the flow is activated only in the EDL regions near the channel walls in the conducting liquids. In the non-conducting liquid, because of the absence of EDLs, there is no opposite force exerted on the interface; the non-conducting liquid is driven by the hydrodynamic viscous forces at the interfaces only. These figures show that a lower interface zeta potential would result in a higher velocity of the non-conducting liquid.

Fig. 4.18 shows the steady state ($\bar{t} > 10^{-2}$) dimensionless velocity profiles, $\bar{u}^E$, at the symmetric line, when $d\bar{p}/d\bar{x} = 0$, $E_{x1} = E_{x3} = 3000$ V/m, $\beta_1 = \beta_3 = 1$ and different viscosity ratios $\beta_2 = 1, 2, 3$. When electric fields are applied across the conducting fluids, the conducting fluids 1 and 3 are driven by electroosmosis, which drag the non-conducting fluid 2 by the hydrodynamic shear force. The results indicate that the velocity profiles of the conducting fluids are strongly dependent on the viscosity ratio, β_2 . The flow resistance of the non-conducting fluid decreases with the decreasing in β_2 . Thus, the non-conducting fluid can be driven with less flow resistance as shown in Fig. 4.18. 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 conducting fluids.

Case (b) Three-fluid flow driven by electroosmosis and pressure gradient

When both pressure gradient and electric field are applied, the three liquids are driven by electroosmotic body force and pressure gradient. For a given pressure gradient, the velocities and flowrates of the three liquids depend only on the applied electroosmotic force.

Figs. 4.19 - 4.20 show the time evolution of the velocity profiles ($\bar{u}$, $\bar{u}^p$ and $\bar{u}^E$) when $K = 130$, $E_{x1} = E_{x3} = 3000$ V/m, $d\bar{p}/d\bar{x} = 50000$, $\alpha_1 = \alpha_2 = \alpha_3 = 1$, $\beta_1 = \beta_2 = \beta_3 = 1$. The interface zeta potential of Figs. 4.19 and 4.20 are $\bar{\xi}_3 = \bar{\xi}_6 = \bar{\xi}_{wall}$, and $\bar{\xi}_3 = \bar{\xi}_6 = 0$ respectively. When $\bar{t} = 0$, the fluid velocity anywhere in the channel is at rest and hence the velocity equals zero. It can be seen that upon the application of the electric field and pressure gradient, the flow begins close to the channel walls and the liquid-liquid interfaces, the velocity increases rapidly at the wall and interface to its peak value within the EDL regions. This reveals the unique feature of the electroosmotic flow as the flow is driven by the electric body force due to the interaction of the applied electric field and the net charge density, such driving force occurs only within the EDL regions. The velocity profile of the non-conducting liquid depends on the interface shear stress and the density charge in the conducting liquids. During this early stage of the evolution, electroosmosis effect dominates, the velocity profile of $\bar{u}$ and $\bar{u}^E$ are similar as shown in Fig. 4.19 (a) and Fig. 4.20 (a).

At the steady state, the velocity profile $\bar{u}^E$ exhibits a plateau, resembling a plug-like flow pattern in the conducting fluids. The velocity profiles of $\bar{u}^p$ exhibit a parabolic profile due to the presence of the pressure gradient. The combined velocity profile, $\bar{u}$ is the superposition of the solutions of $\bar{u}^E$ and $\bar{u}^p$.

To study the dynamic responses of the flow under the applications of both constant electric field and pressure gradient, flow velocity at locations within the EDL region and outside the EDL are analyzed. Fig. 4.21 (a) shows the time-dependent velocity response within the EDL at $\bar{y} = 0.9925$ (point A in Fig. 4.15), whereas Fig. 4.21 (b) shows the velocity response outside the EDL at $\bar{y} = 0.85$ (point B in Fig. 4.15), and Fig. 4.21 (c) shows the velocity response of the non-conducting fluid region at $\bar{y} = 0.5$ (point C in Fig. 4.15). The flow conditions are: $K = 130$, $E_{x1} = E_{x3} = 3000$ V/m, $d\bar{p}/d\bar{x} = 50000$, $\alpha_1 = \alpha_2 = \alpha_3 = 1$, $\beta_1 = \beta_2 = \beta_3 = 1$, $\bar{\xi}_3 = \bar{\xi}_6 = \bar{\xi}_{wall}$.

The local velocity, $\bar{u}^E$, $\bar{u}^p$ at points A, B and C are normalized by defining dimensionless variables as

$$\bar{u}^{E*} = \frac{\bar{u}^E}{\bar{u}_{\text{steady state}}^E} \quad (4.173)$$

$$\bar{u}^{P*} = \frac{\bar{u}^P}{\bar{u}_{\text{steady state}}^P} \quad (4.174)$$

where $\bar{u} = \frac{u}{U_{\text{ref}}}$, and $\bar{u}_{\text{steady state}} = \frac{u_{\text{local steady state velocity}}}{U_{\text{ref}}}$ is the local steady state velocity.

It is noted from Fig. 4.21 (a) that, within the EDL region (point A), the velocity response due to pressure gradient is always behind that applied electric field. Though near the wall, the velocity is affected by the no-slip boundary condition, however, within the EDL region the flow is driven by the electric body force resulting from the interaction of the electric field and the net charge density. As time elapse, the liquid inside the EDL exerts viscous shear stress to the adjacent liquid and fluids outside the EDL (point B) is set in motion layer by layer, finally extending to the entire region of the conducting liquid, in point A, electroosmosis effect dominate.

At the bulk of the conducting liquid region (region away from EDL, at point B), the liquid is driven by the viscous shear diffusion as the result of (i) the faster fluid layers at the EDL region and (ii) the faster lamina fluid layers immediately closer to the center. i.e. the combined electric field and pressure gradient which pull the fluid at the bulk of the conducting liquid region. The results show that both $\bar{u}^E$ and $\bar{u}^P$ achieve the steady state at $\bar{t} = 10^{-5}$.

Fig. 4.21 (c) shows the flow in the non-conducting fluid region. The velocity response due to the pressure gradient is faster than that of the applied electric field. In this non-conducting fluid region, pressure gradient effect dominates. The liquid at point C is driven by: i) the faster lamina immediately closer to the center due to pressure gradient, and ii) the interfacial shear stress. Same as before, both $\bar{u}^E$ and $\bar{u}^P$ achieve the steady state at $\bar{t} = 10^{-5}$.

Volumetric flowrate

The dimensionless volumetric flowrates of the three-fluid electroosmotic flow, $\bar{q}_1$, $\bar{q}_2$ and $\bar{q}_3$, where zero pressure gradient ($d\bar{p}/d\bar{z} = 0$), $E_{x1} = E_{x3} = 3000 \text{ V/m}$, $K = 130$,

$\rho_1 = \rho_2 = \rho_3$, are plotted in Fig. 4.22 for different values of interface zeta potentials (Fig. 4.22 (a)), dynamic viscosity ratio (Fig. 4.22 (b)), β_2 , and kinematic viscosity ratio (Fig. 4.22 (c)), α_2 .

In Fig. 4.22 (a), the dynamic viscosity and density of three liquids are identical. It can be seen that higher interface zeta potential will induce larger flowrate of the conducting liquids ($\bar{q}_1$ and $\bar{q}_3$), but the flowrate of the non-conducting liquid ($\bar{q}_2$) is lower. In the presence of free charges at the interface, the non-conducting liquid will flow in the opposite direction. Gao [11] called this phenomena “back flow”. The back flowrate increases as the increase of the interface zeta potential. As shown in Fig. 4.22 (a), if the interface zeta potentials are larger than the wall zeta potential, the flowrate of the non-conducting liquid may be negative at steady state.

Fig. 4.22 (b) shows the effect of different viscosity ratio, β_2 , on the volumetric flowrate. In this figure, $\bar{\xi}_3 = \bar{\xi}_6 = 0.5\bar{\xi}_1$, $\bar{\rho}_1 = \bar{\rho}_2 = \bar{\rho}_3$, $\beta_1 = \beta_3 = 1$. The results show that the liquid with a lower viscosity ratio, can be driven with a lower flow resistance, in another word, the volumetric flowrate is higher under the same electric fields. Hence the three-fluid electroosmotic pump method is feasible for the relatively small interface zeta potentials and viscosity ratio, β_2 .

Fig. 4.22 (c) shows the volumetric flowrates for different density of the non-conducting fluid. In this figure, $\bar{\xi}_3 = \bar{\xi}_6 = 0.5\bar{\xi}_1$, $\beta_1 = \beta_2 = \beta_3 = 1$, $\rho_1 = \rho_3$. Eqs. (4.114) to (4.119) show that fluid density does not affect the velocity profile but affects the developing time. The kinematic viscosity ratio, $\alpha_2 = (\bar{\mu}_2/\bar{\rho}_2)/(\bar{\mu}_1/\bar{\rho}_1)$, evaluates the developing time. For a large value of α_2 , a shorter time is required to approach the steady state.

Fig. 4.23 shows the dimensionless volumetric flowrates of the three-fluid flow for case (a) $E_{x1} = E_{x3} = 3000 \text{ V/m}$, $d\bar{p}/d\bar{x} = 0$, and case (b) $E_{x1} = E_{x3} = 3000 \text{ V/m}$, $d\bar{p}/d\bar{x} = 10000$. As shown, the effect of pressure gradient does not affect the developing time. Volumetric flowrate is higher under the combined electroosmotic and pressure gradient.

4.3 Experimental investigations on electroosmotic control of width and position of liquid streams in hydrodynamic flow

4.3.1 Analytical solution of interface control

In section 4.2, the analytical solutions of three-fluid flow driven by pressure gradient and electroosmosis are obtained. From Eqs. (4.86) - (4.91), the flowrates of the three-fluid flow are a function of dimensionless width, e_1, e_2, e_3 , and the pressure gradient, $d\bar{p}/d\bar{x}$,

$$\begin{cases} \bar{q}_1 = f(e_1, e_2, e_3, d\bar{p}/d\bar{x}) \\ \bar{q}_2 = f(e_1, e_2, e_3, d\bar{p}/d\bar{x}) \\ \bar{q}_3 = f(e_1, e_2, e_3, d\bar{p}/d\bar{x}) \\ e_1 + e_2 + e_3 = 1 \end{cases} \quad (4.175)$$

If the flowrates are specified, the system of equations can be solved. Here, the Newton-Raphson method is used to solve the dimensionless widths (e_1, e_2, e_3) and pressure gradient as follows:

- (1) The initial values of e_1, e_2, e_3 are assumed;
- (2) From the initial values of e_1, e_2, e_3 the electroosmotic flowrates, $\bar{q}^E$, are calculated using Eqs. (4.86), (4.88) and (4.90);
- (3) From $\bar{q}^p = \bar{q} - \bar{q}^E$, the pressure-driven flowrates, $\bar{q}^p$, are calculated;
- (4) As $(d\bar{p}/d\bar{x})_i = f(\bar{q}_i^p, e_1, e_2, e_3)$, the values of $(d\bar{p}/d\bar{x})_i$ are calculated;
- (5) Compare the values of $(d\bar{p}/d\bar{x})_i$; if they are same, the values of e_1, e_2, e_3 are the fractions of flows; if they are different, modify the values of e_1, e_2, e_3 . Using the modified values of e_1, e_2, e_3 , repeat steps (2)-(5) until $(d\bar{p}/d\bar{x})_i$ have the same value.

4.3.2 Electroosmotic control of width and position of fluid streams in hydrodynamic flow

The interface and width of the focused stream are important for separation, reaction and mixing of chemical. Controlling the interface leads to control the stream width, this is useful for focusing sample fluid and switching of a sample stream. This section presents experimental results of the pressure-driven three-fluid flow in an H-shaped microchannel with electroosmotic effect.

Material and fabrication

Fig. 4.24 shows the H-shaped microchannel used in the experiments. The adhesive lamination technique [170] is used to fabricate the microchannel. In this method, two polymethylmethacrylate (PMMA) plates (50 mm × 25 mm) are bonded by one or multiple layers of double-sided adhesive tape. The channel height can be adjusted by the number of the adhesive layer. The channel structures are cut into the adhesive tape (Arclad 8102 transfer adhesive, Adhesives Research Inc.) using CO₂ laser. As shown in Fig. 4.24, the three-fluid flow side by side in the straight microchannel in the direction from left to right. The flows at the inlets A, B and C are driven by syringe pumps. The outlets D, E and F are connected to a waste reservoir. Electric fields are applied by inserting two pairs of platinum electrodes into A, D and C, F, respectively.

Experimental setup

Fig. 4.25 shows a schematic of the experimental setup. Fluorescence imaging technique is used in our experiments. The setup consists of four main components: an illumination system, an optical system, a coupled charge device (CCD) camera and a personal computer (PC) based control system. A mercury lamp is used as the illumination source for the fluorescence measurement. The optical system is a Nikon inverted microscope (Model ECLIPSE TE2000-S) with a set of epi-fluorescent attachments. The filter cube consists of three optical elements: excitation filter, dichroic mirror and emission filter. Emission filters are used in the measurements to select the specific emission wavelength of the sample and to remove traces of excitation light. The image acquisition device consists of an interline transfer CCD camera (HiSense MkII), a

FlowMap system hub with last-in-first-out (LIFO) capability. The resolution of the CCD camera is 1,344×1,024 pixels with 12 bits grayscale.

A microchannel with a cross section of 1000 μm ×50 μm and a length of 2 cm is used in the experiment. The width of inlet is 333 μm . Three identical syringes (5 ml gastight, Hamilton) are driven by a single syringe pump (Cole-Parmer, 74900-05, 0.2 $\mu\text{l/h}$ to 500 l/h, accuracy of 0.5%) to introduce the same volumetric flowrates of the three inlet flows, NaCl solution (concentration 7×10^{-4} M), aqueous glycerol (volume concentration 10%), and NaCl solution (concentration 7×10^{-4} M) into the inlets A, B and C, respectively.

A high voltage power supply (Model PS350 power supply from Stanford Research System, Inc) is used to provide the controlling electric fields. The power supply is capable of producing up to 5000 V and changing the polarity of the voltage.

Results of electroosmotic control of width and position of liquid streams

Fluorescence imaging measurement

Rhodamine B ($\text{C}_{28}\text{H}_{31}\text{N}_2\text{O}_3\text{Cl}$) is used as the fluorescent dye in our experiment. For the purpose of image collection, the fluorescent dye is added to the NaCl solution. Rhodamine B is a water-solute fluorescent dye, which can be excited by green light and emit red light (the maximum wavelength is 610 nm), the fluorescent image is recorded by an epi-fluorescent attachment of type Nikon G-2E/C. The interface positions between the three fluids are determined using the recorded images and a customized image processing program written in MATLAB [170]. The program removes the noise in the collected images with an adaptive noise-removal filter. The program also determines the pixel intensity values across the channel, which is normalized using $I^* = \frac{I - I_{\min}}{I_{\max} - I_{\min}}$, where

$I_{\max}$ and $I_{\min}$ are the maximum and minimum intensity values, respectively.

A linear relationship between the light intensity and the concentration of the fluorescent dye is assumed [162]. Because of the mixing effect, an inter-diffusion region

exists at the interface. This report assumes that the inner diffusion of dye between the two fluids is a Gaussian distribution. A MATLAB program is developed to determine the interface position. The program calculated the position (y) based on the derivative of concentration of fluorescence c with respect to the distance y , dc/dy can be taken by the formula of $(c_2 - c_1)/(y_2 - y_1)$. The interface position is taken as the position where dc/dy its maximum value has.

Fig. 4.26 shows the flow images at a flowrate of 0.05 ml/h when $E_{x1} = E_{x3} = 200 \text{ V/cm}$ and $E_{x1} = E_{x3} = -200 \text{ V/cm}$, respectively. The corresponding normalized concentration profiles for different electric fields are shown in Fig. 4.27. A planar interface between NaCl solution and aqueous glycerol is assumed. Fig. 4.28 shows the cross sectional view of the three-fluid flow in microchannel. The dimensionless widths of the NaCl solution are:

$$\begin{aligned} e_1 &= \frac{h_1}{h_1 + 2h + h_3} \\ e_3 &= \frac{h_3}{h_1 + 2h + h_3} \end{aligned} \quad (4.176)$$

The dimensionless width of aqueous glycerol is

$$e_2 = \frac{2h}{h_1 + 2h + h_3} \quad (4.177)$$

The aspect ratio, χ , is defined as [13]

$$\chi = \frac{(2h + h_1 + h_3)}{2w} \quad (4.178)$$

The negative wall potential affects the distribution of free ions in the NaCl solution and the electric double layers (EDL) near the wall. An EDL can hardly form in the glycerol solution because only few free ions are available [26]. This fact is reflected in the conductivity of the NaCl solution of $86.6 \mu\text{S/cm}$, and the conductivity of pure glycerol of $0.064 \mu\text{S/cm}$ [182]. Thus, the electroosmosis has little effect on the aqueous glycerol flow. If two positive electric fields are applied between A and D, and between C and F (A and C are the positive electrodes, D and F are the negative electrodes), the NaCl solution is dragged in the same direction as the pressure-driven flow. When two negative

electric fields are applied between A and D, and between C and F (A and C are the negative electrodes, D and F are the positive electrodes), the NaCl solution is dragged in the opposite direction as the pressure-driven flow.

(b) Interface control and flow focusing

Three liquids (two conducting liquids with NaCl and aqueous glycerol as the non-conducting liquid with viscosity ratio $\beta_1 = \beta_3 = 1$ and $\beta_2 \approx 1.46$) are introduced through the inlets by the syringe pump. The volumetric flowrates of all three liquids are maintained at the same value.

When no electric field is applied, the flow is simply driven by a pressure gradient. As the aqueous glycerol ($\mu_2 = 1.24 \times 10^{-3} \text{ Nsm}^{-2}$) is more viscous than the NaCl solution ($\mu_1 = 0.85 \times 10^{-3} \text{ Nsm}^{-2}$), the NaCl solutions occupy two smaller portions of the channel, Fig. 4.29 (a). When negative electric fields are applied along the conducting liquids, the electroosmotic fluids act against the pressure-driven flow. Consequently, the NaCl solutions encounter a larger resistance. Apparently, the NaCl solutions appear to be more “viscous” due to the electroosmotic effect. Due to the same pressure drop along the microchannel, in order to achieve the same volumetric flowrate, the more viscous fluid has to spread over a larger area, thus occupy a larger portion of the channel. Fig. 4.29 (b) shows that when $E_{x1} = E_{x3} = -200 \text{ V/cm}$, the dimensionless widths of NaCl increase symmetrically, $e_1 = e_3$. e_1 and e_3 increase with the increase of the negative electric fields. At different electric fields ($E_{x1} = -50 \text{ V/cm}$, $E_{x3} = -200 \text{ V/cm}$), e_1 and e_3 , increase differently, Fig. 4.29 (c). If positive electric fields ($E_{x1} = 200 \text{ V/cm}$, $E_{x3} = 200 \text{ V/cm}$) are applied to the sheath flows, e_1 and e_3 decrease, Fig. 4.29 (d). Since the electroosmotic flow is in the same direction as the pressure-driven flow, the NaCl solutions flow faster. When a positive electric field ($E_{x1} = 50 \text{ V/cm}$) and a negative electric field ($E_{x3} = -200 \text{ V/cm}$) are applied, e_1 decreases, and e_3 increases, as shown in Fig. 4.29 (e). The above results show that the location of the two interfaces can be controlled by applying different electric fields on the sheath flows.

(c) Comparing of theoretical analysis and experimental results

Fig. 4.30 compares the measured and analytical results under different applied electric field strengths. A reasonable agreement is obtained. The reference velocity is $U_{\text{ref}} = 1.77 \times 10^{-3} \text{ m/s}$, and the reference length is $L_{\text{ref}} = 1 \text{ mm}$. The viscosities of the NaCl solution and aqueous glycerol are taken as $0.85 \times 10^{-3} \text{ Nsm}^{-2}$ and $1.24 \times 10^{-3} \text{ Nsm}^{-2}$ respectively. Under these conditions, the dimensionless flowrates are 0.019, 0.057, 0.095, and 0.19 for 0.1 ml/h, 0.3 ml/h, 0.5 ml/h and 1 ml/h, respectively. The zeta potential at the PMMA channel and NaCl solution is -25.7 mV as measured by the electroosmotic mobility method [26]. The zeta potential at the interface between the aqueous glycerol and the NaCl solution is taken as 0 mV.

Fig. 4.30 shows the relationship between the focused flow width e_2 and the applied voltage for the same volumetric flowrates of the three-fluid. Fig. 4.30 indicates that the focused width e_2 changes according to the magnitude and the polarity of the electric field. When no electric field is applied along the channel, the flow is simply a pressure-driven three-fluid flow where the more viscous glycerol occupies a larger portion of the channel. Without electric field, e_2 is about 0.53.

The experimental results of Fig. 4.30 also show that the electroosmotic effect on the pressure-driven flow becomes weaker with the increase of the flowrates of the three-fluid. At a flowrate of 0.1 ml/h, e_2 increases from 0.47 to 0.73, if the voltage varies from -1200 V to 1200 V. At a flowrate of 1 ml/h, the dimensionless focused width increases from 0.51 to 0.537.

Fig. 4.31 shows the ability of manipulating the interface position by the externally applied electric field for different viscosity ratios. In these cases, $q_1 = q_2 = q_3 = 0.1 \text{ ml/h}$, $\beta_1 = \beta_3 = 1$, and $\chi = 20$ are used. The experimental results show that, the lower the viscosity ratio of β_2 , i.e. as $\beta_2 = 1.46$, the larger span is e_2 for the same range of applied electric field. Comparing the experiment results of different β_2 and the theoretical analysis, a reasonable agreement is obtained.

Fig. 4.32 shows the variation of the focused width e_2 with the applied electric fields for different aspect ratios when $q_1 = q_2 = q_3 = 0.1$ ml/h and $\beta_2 = 1.46$, $\beta_1 = \beta_3 = 1$. The results show that the rate of change with applied electric field of e_2 is higher at higher aspect ratios. An increase of e_2 from 0.4 to 0.75 has been successfully obtained for $\chi = 20$. The comparison of measured data and theoretical results for different aspect ratios shows that the proposed model is capable to predict the experimental results reasonable well.

Fig. 4.33 shows the variation of pressure gradient with the applied electric field for different flowrates. Under large flowrates, where the pressure gradient is higher, larger applied electric fields are required to control the interface position.

4.3.3 Flow switching of a sample stream

In section 4.3.2, spreading and focusing sample flow either by pressure-driven or by electroosmosis are experimentally presented. This section focuses on switching the sample flow or droplets to the desired outlets using the electroosmotic effect under the fixed flowrates. It includes two parts: the first part presents experimental results of valveless flow switching in a Y-shaped Polydimethylsiloxane (PDMS) microchannel under the coupled effect of pressure-driven and electroosmosis. Fig. 4.34 (a) schematically shows the microchannel system used for flow switching of a non-conducting sample stream. Similar with section 4.3.2, liquid fractions are investigated by fluorescence imaging technique. Fig. 4.1 shows the cross section view of the model of valveless flow switching in a Y-shaped microchannel. Experimental results are compared with the proposed analytical model (section 4.2) using the method presented in section 4.3.2. The second part presents droplet switching in a T-shaped Polymethylmethacrylate (PMMA) microchannel under the coupled effect of pressure-driven and electroosmosis. Fig. 4.34 (b) shows flow switching with droplet generation in the non-conducting fluid. Generation of these water droplets can be achieved by shearing the water stream at a T-junction using a continuous mineral oil stream.

Design and fabrication of test devices

Fig. 4.35 (a) shows the layout and the detailed geometries of the microchannels used in the experiments. The test section is a simple straight channel with five output ports. All structures have the same depth of $50\text{ }\mu\text{m}$. The widths of the inlet channel and outlet channel are 0.17 mm and 0.1 mm , respectively. Two sheath streams are injected into the microchannel from the inlet ports of I_1 and I_3 , the sample stream is injected into the microchannel from the inlet port of I_2 . The sample stream are focused and delivered to the desired outlet ports, Fig. 4.35 (b).

Soft lithography technique was used to fabricate the microchannel in polydimethylsiloxane (PDMS) [183]. The layout shown in Fig. 4.35 (a) is designed using AutoCAD and printed on a transparency film using a laser printer with a resolution of 8000 dpi. We used rectangular channels for the experiments as shown in Fig. 4.35. A $50\text{ }\mu\text{m}$ thick SU-8 layer was first spin-coated on a silicon wafer after soft bake (65°C , 30 min) and hard bake (95°C , 5 min), the SU-8 layer was exposed and developed. The SU-8 master mold was washed in DI water and IPA, and then dried in an oven. PDMS (Dow Corning Corporation, USA) was mixed from the two components (base and curing agent) with a weight ratio of 10:1. The mixed PDMS was placed in a vacuum oven for 20 minutes to remove air bubbles. The bubble-free PDMS mixture was poured into the SU-8 mold. The PDMS part was then kept at room temperature for 36 hours to cure. Subsequently, the cured PDMS was peeled off from the master mold. Eight ports with a diameter of 0.75 mm were punched into the PDMS part. Finally, another cured flat PDMS slab and the PDMS with the microchannels were treated by oxygen plasma and bonded together.

The main problem of a PDMS device is swelling when exposed to silicone oil. For the long duration of the experiments of droplet switching, PDMS is not suitable as a substrate material. Therefore, polymethylmethacrylate (PMMA) was used as the device material for the droplet switching device. Two PMMA plates were cut by laser-machining and thermally bonded. The laser machined structures were designed by the software of Corel DRAW. The width of the channel is controlled by the width of the drawn lines. The depth of the microchannel can be controlled by the power and speed of the laser beam. The thermal bonding process was achieved using a hot press (CARVER

manual press 4386). First, the temperature of the hot plate increases to 165 °C. Second, the temperature is kept at 165 °C for 30 minutes. Third, the temperature is reduced to 80 °C. Fourth, the temperature is kept at 80 °C for 30 minutes. Finally, the whole system is allowed to cool down to the room temperature. Fig. 4.36 (a) shows the layout and the detailed geometries of the microchannel used in our experiments. The test section includes a T-junction channel and a simple straight channel with five output ports. All structures have the same depth of 50 μm . According the experiments, the curved channel is of advantage for a stable flow especially if the viscosity of the fluid is high.

Materials

In the experiments of valveless flow switching, de-ionized (DI) water (MILLIPAK[®] EXPRESS, Millipore, 0.22 μm) and silicone oil (Sigma-Aldrich) work as sheath streams and sample stream, respectively. Rhodamine B ($\text{C}_{28}\text{H}_{31}\text{N}_2\text{O}_3\text{Cl}$) (Sigma-Aldrich) was added to the DI water as the fluorescent dye to achieve a distinct interface with fluorescent microscopy. The concentration of the fluorescence is 1 g/L. NaCl was diluted in DI water with a concentration of 7×10^{-4} M. The conductivity and viscosity of the NaCl solution are 86.6 $\mu\text{S}/\text{cm}$ and $0.85 \times 10^{-3} \text{ Nsm}^{-2}$, respectively. The viscosity of silicone oil as sample stream is $4.565 \times 10^{-3} \text{ Nsm}^{-2}$. The details of the flow conditions are shown in Table 4.1.

In the experiments of droplet switching, de-ionized (DI) water was used to form liquid droplets at a T-junction by a continuous mineral oil stream with 2% w/w surfactant Span 80 (Sigma-Aldrich). In order to display the droplets, Rhodamine B was added to the DI water. The concentration of the fluorescence is 1g/L. The continuous fluid is mineral oil (Sigma-Aldrich). Glycerol was mixed with DI water ($V_{\text{glycerol}} / (V_{\text{DI water}} + V_{\text{glycerol}}) = 0.72$) as sheath streams. NaCl was diluted in aqueous glycerol with a concentration of 7×10^{-4} M. The conductivity and viscosity of the NaCl solution are 86.6 $\mu\text{S}/\text{cm}$ and $2.6 \times 10^{-2} \text{ Nsm}^{-2}$, respectively. The viscosity of mineral oil is $2.69 \times 10^{-2} \text{ Nsm}^{-2}$.

Fluorescence imaging measurement

The detail of the Fluorescence imaging measurement is shown in part of 4.3.2. Fig. 4.37 (d) shows the cross sectional view of the three fluids in the microchannel, h_1 , $2h$ and h_3 are denoted as the width of sheath flow, sample stream and sheath flow, respectively. Fig. 4.37 (e) shows the cross sectional view of the channel for droplet switching.

Results and discussion

(a) Switching and focusing under electroosmotic effect

Fig. 4.38 shows a series of pictures for sample stream injection into specific outlet ports. The electroosmotic effect for switching is confirmed experimentally. The sample stream can be delivered into any desired output ports under the coupled effect of hydrodynamic effect and electroosmosis.

Two sheath streams q_1 and q_3 (conducting liquids of aqueous NaCl), and the sample stream q_2 (silicon oil, dimethylsiloxane 200®fluid. Viscosity 5 cSt), are introduced through the inlets by the syringe pumps. The volumetric flowrate of sample stream (q_2) is 0.1 ml/h, and the volumetric flowrates of sheath flow (q_1 and q_3) are 0.3 ml/h. The mean velocity (U) of the sample stream is about 0.003 m/s. The surface tension (γ) of silicon oil/DI water is 19.7 mN/m, and the capillary number $Ca \approx 7 \times 10^{-4}$. These immiscible fluids flow side by side in a straight rectangular microchannel.

Fig. 4.38 (a)-(f) show that at fixed flowrates, the sample stream can be delivered to the desired outlet ports according to the magnitude and the polarity of the electric fields. The results also show that when $V_{x1} = V_{x3}$, the sample stream cannot be switched to other ports except the center port, O_3 . When the magnitudes or polarities of applied two electric fields are different ($V_{x1} \neq V_{x3}$), the sample flow are shifted away from the center port.

When a negative electric field is applied along the conducting sheath stream 1 (q_1 , Fig. 4.38 (a), $V_{x1} = -1700V$), the electroosmotic flow acts against the pressure-driven flow. Consequently, q_1 encounters a larger resistance and appears to be more “viscous”

due to the additional electroosmotic effect. Because of the same pressure drop along the microchannel and the fixed volumetric flowrate forced by the syringe pump, the more viscous fluid has to spread over a larger area, thus occupy a larger portion (e_1) of the channel. When a positive electric field is applied along the conducting sheath stream 3 (q_3 , Fig. 4.38 (a), $V_{x3} = 1700V$), the electroosmotic effect acted along the pressure driven flow, and q_3 is dragged in the same direction as the pressure-driven flow. As a result, q_2 flows faster and occupies a smaller width (e_2) of the channel. The results show that when $V_{x1} = V_{x3} = -300V$ (Fig. 4.38 (d)) and $V_{x1} = V_{x3} = -700V$ (Fig. 4.38 (c)) the liquid fraction of NaCl increase symmetrically ($e_1 = e_3$), and the sample stream is delivered to the center port O_3 . The fractions e_1 and e_3 increase with the increase of negative electric fields, hence the focused width e_2 varies with the applied electric fields. In (Fig. 4.38 (b)), when a positive electric field ($V_{x3} = 300V$) and a negative electric field ($V_{x1} = -1000V$) are applied, e_3 decrease, and e_1 increase, the sample stream is delivered to port O_4 . (Fig. 4.38 (a, e, f) show that the sample stream can be guided to the desired outlet port by adjusting the magnitudes and the polarities of the electric fields.

Fig. 4.39 shows that droplets can be delivered to the desired outlet ports according to the magnitudes and the polarities of the electric fields. The effect of electric fields is similar to that shown in Fig. 4.38.

The above results show that a negative electric field could ‘push’ the sample stream (droplet), while a positive electric field could ‘pull’ the sample stream (droplet) in the span-wise direction of the microchannel. Hence the position of the interface or of the droplet could be controlled.

(b) Comparison between theoretical analysis and experiment

Fig. 4.40 (a) compares the measured and mathematical results under different applied electric field strengths. For the mathematical solutions, a reference velocity of $U_{ref} = 1.16 \times 10^{-3} \text{ m/s}$, a reference length of $L_{ref} = 500 \mu\text{m}$, and respective viscosities of NaCl solution and aqueous glycerol of $0.85 \times 10^{-3} \text{ Nsm}^{-2}$ and $4.565 \times 10^{-3} \text{ Nsm}^{-2}$ were

used. The zeta potentials of PDMS and silicon oil with NaCl solution are $\xi_{\text{PDMS}} = -85 \text{ mV}$ [110], $\xi_{\text{oil}} = -40 \text{ mV}$ [184].

Fig. 4.40 (a) shows the relationship between applied voltages V_{x1} , and V_{x3} for flow switching into desired outlet ports at a fixed flowrates $q_1 = q_3 = 0.3 \text{ ml/h}$, $q_2 = 0.1 \text{ ml/h}$. It is clearly seen that the proposed model gives reasonable agreement with the measurement. In general, for guiding the sample stream into output ports other than the center port, a combination of the positive and negative electric fields is needed. The results further suggest that the most sensitive switching can be achieved by setting the two applied electric fields to have the same magnitudes but opposite in polarities, $V_{x1} = -V_{x3}$. This manipulation scheme can be accomplished by using only one high voltage power supply.

Fig. 4.40 (b) shows that the voltage required for flow switching depends on the fluid viscosity ratio. The theoretical results indicate that it is more difficult to switch the sample stream with higher viscosity, because a higher voltage is needed. The switching response becomes slower with the increase of the viscosity of the sample stream.

Fig. 4.41 shows the influence of the applied voltage when $V_{x1} = -V_{x3}$ on the sample stream position in the microchannel. The location of the sample stream for flow switching into desired outlet ports has also been compared with the analytical model. A rapid increase in the sample stream position occurs when the voltages decrease from 1000 V to -1000 V . At fixed flowrates, the switching response under the electroosmotic effect becomes slower as the sample stream is far from the center of the channel.

Experimental results are compared with the analytical results in Fig. 4.40 and Fig. 4.41. These results agree within a 95% confidence level. The discrepancy between the mathematical model and the experiment is possibly caused by the assumption of flat interfaces and the fluctuation of the pressure gradient provided by the syringe pumps.

Table 4.1 The details of flow conditions for experiments

Exp	Fluids		Viscosity	Conductivity	Channel
Switching Sample fluid	Conducting fluids	Aqueous NaCl + Rhodamine B	$0.85 \times 10^{-3} \text{ Nsm}^{-2}$	$86.6 \mu\text{S/cm}$	PDMS
	Non -conducting fluid	Silicon oil	$4.565 \times 10^{-3} \text{ Nsm}^{-2}$		
Switching droplets	Conducting fluids	Aqueous glycerol + NaCl	$2.6 \times 10^{-2} \text{ Nsm}^{-2}$	$86.6 \mu\text{S/cm}$	PMMA
	Non -conducting fluid	Mineral oil	$2.69 \times 10^{-2} \text{ Nsm}^{-2}$		
	droplets	DI water + Rhodamine B	$0.85 \times 10^{-3} \text{ Nsm}^{-2}$

Fig. 4.1 Schematic of the phenomenon and coordinate system

Fig. 4.2 EDL profiles at the symmetric line in the two conducting fluids

Fig. 4.3 Schematic representation of the three-fluid electroosmotic stratified flow

Fig. 4.4 Dimensionless velocity distribution at the symmetric line for different values of viscosity ratio, β_2 ($E_{x1} = E_{x3} = 1000$ V/m, $d\bar{p}/d\bar{x} = 0$, $\beta_1 = \beta_3 = 1$)

Fig. 4.5 Comparison of the analytical solutions between three-fluid model and single-fluid model [38]

Fig. 4.6 Dimensionless velocity distribution at the symmetric line for different values of electric field E_{x1} and E_{x3} ($d\bar{p}/d\bar{x}=1000$, $\beta_1=1$, $\beta_2=1$, $\beta_3=1$)

Fig. 4.7 Dimensionless velocity distribution of $\bar{u}^P$ ($d\bar{p}/d\bar{x}=1500$, $E_{x1}=E_{x3}=0$ V/m), $\bar{u}^E$ ($d\bar{p}/d\bar{x}=0$, $E_{x1}=E_{x3}=15000$ V/m), and $\bar{u}$ ($d\bar{p}/d\bar{x}=1500$, $E_{x1}=E_{x3}=15000$ V/m), when $\beta_1=1$, $\beta_2=1$, $\beta_3=1$

Fig. 4.8 Dimensionless velocity distribution at the symmetric line for different values of electric field ($d\bar{p}/d\bar{x} = 1000$, $\beta_1 = 1$, $\beta_2 = 5$, $\beta_3 = 1$)

Fig. 4.9 Dimensionless velocity distribution at the symmetric line for different pressure gradients $\frac{d\bar{p}}{d\bar{x}}$ ($E_{x1} = E_{x3} = 1000$ V/m, $\beta_1 = 1$, $\beta_2 = 5$, $\beta_3 = 1$)

Fig. 4.10 Dimensionless velocity distribution at the symmetric line for different dynamic viscosity ratios, β_2 ($E_{x1} = E_{x3} = 1000$ V/m, $d\bar{p}/d\bar{x} = 1000$, $\beta_1 = \beta_3 = 1$)

Fig. 4.12 Dimensionless volumetric flowrates versus dynamic viscosity ratio β_2
 ($E_{x1} = E_{x3} = 1000 \text{ V/m}$, $d\bar{p}/d\bar{x} = 1000$, $\beta_1 = \beta_3 = 1$)

Fig. 4.13 Dimensionless volumetric flowrates versus dimensionless pressure gradient
 ($E_{x1} = E_{x3} = 1000 \text{ V/m}$, $\beta_1 = \beta_3 = 1$, $\beta_2 = 5$)

Fig. 4.14 Dimensionless volumetric flowrates versus applied electric fields ($E_{x1} = E_{x3}$, $d\bar{p}/d\bar{x} = 1000$, $\beta_1 = \beta_3 = 1$, $\beta_2 = 5$)

Fig. 4.15 Potential distributions along the symmetrical line when $\bar{\xi}_3 = \bar{\xi}_6 = \bar{\xi}_{wall}$

Point A is within the EDL at $\bar{y} = 0.9925$;

Point B is outside the EDL at $\bar{y} = 0.85$;

Point C is in the non-conducting fluid region.

(a) $\bar{t} = 1 \times 10^{-6}$

(b) $\bar{t} = 1 \times 10^{-5}$

(c) $\bar{t} = 1 \times 10^{-4}$

(d) $\bar{t} = 1 \times 10^{-2}$

Fig. 4.16 Time evolution of the three-fluid electroosmotic flow (a) $\bar{t} = 1 \times 10^{-6}$; (b) $\bar{t} = 1 \times 10^{-5}$; (c) $\bar{t} = 1 \times 10^{-4}$; (d) $\bar{t} = 1 \times 10^{-2}$ ($K = 130$, $E_{x1} = E_{x3} = 3000$ V/m, $d\bar{p}/d\bar{x} = 0$, $\alpha_1 = \alpha_2 = \alpha_3 = 1$, $\beta_1 = \beta_2 = \beta_3 = 1$, $\bar{\xi}_3 = \bar{\xi}_1 = \bar{\xi}_4 = \bar{\xi}_6 = \bar{\xi}_{wall}$)

(a) $\bar{t} = 1 \times 10^{-6}$

(b) $\bar{t} = 1 \times 10^{-5}$

(c) $\bar{t} = 1 \times 10^{-4}$

(d) $\bar{t} = 1 \times 10^{-2}$

Fig. 4.17 Time evolution of the three-fluid electroosmotic flow (a) $\bar{t} = 1 \times 10^{-6}$; (b) $\bar{t} = 1 \times 10^{-5}$; (c) $\bar{t} = 1 \times 10^{-4}$; (d) $\bar{t} = 1 \times 10^{-2}$ ($K = 130$, $E_{x1} = E_{x3} = 3000$ V/m, $d\bar{p}/d\bar{x} = 0$, $\alpha_1 = \alpha_2 = \alpha_3 = 1$, $\beta_1 = \beta_2 = \beta_3 = 1$, $\bar{\xi}_3 = \bar{\xi}_6 = 0$)

Fig. 4.18 Dimensionless steady state velocity distribution at the symmetric line for different values of dynamic viscosity ratio, β_2 ($E_{x1} = E_{x3} = 3000 \text{ V/m}$, $d\bar{p}/d\bar{x} = 0$, $\beta_1 = \beta_3 = 1$)

(a) $\bar{t} = 10^{-6}$

(b) $\bar{t} = 10^{-5}$

Fig. 4.19 Time evolution of $\bar{u}^p$, $\bar{u}^E$ and $\bar{u}$ (a) $\bar{t} = 10^{-6}$; (b) $\bar{t} = 10^{-5}$; (c) Steady state;
 ($d\bar{p}/d\bar{x} = 50000$, $E_{x1} = E_{x3} = 3000$ V/m , $\beta_1 = \beta_2 = \beta_3 = 1$, $\alpha_1 = \alpha_2 = \alpha_3 = 1$;
 $\bar{\xi}_3 = \bar{\xi}_6 = \bar{\xi}_{wall}$)

(a) $\bar{t} = 10^{-6}$

(b) $\bar{t} = 10^{-5}$

(c) Steady state

Fig. 4.20 Time evolution of $\bar{u}^p$, $\bar{u}^E$ and $\bar{u}$. (a) $\bar{t} = 10^{-6}$; (b) $\bar{t} = 10^{-5}$; (c) Steady state;
 ($d\bar{p}/d\bar{x} = 50000$, $E_{x1} = E_{x3} = 3000$ V/m, $\beta_1 = \beta_2 = \beta_3 = 1$, $\alpha_1 = \alpha_2 = \alpha_3 = 1$; $\bar{\xi}_3 = \bar{\xi}_6 = 0$)

(a)

(b)

Fig. 4.21 Time dependent velocity, $\bar{u}^E$ and $\bar{u}^P$

- (a) Within the EDL region (point A of Fig. 4.15);
- (b) Outside the EDL region (point B of Fig. 4.15);
- (c) In non-conducting fluid 2 (point C of Fig. 4.15)

(a) Effect of interface zeta potential

(b) Effect of dynamic viscosity

(c) Effect of kinetic viscosity

Fig. 4.22 Dimensionless volumetric flowrates for different parameters
 ($E_{x1} = E_{x3} = 3000 \text{ V/m}$, $d\bar{p}/d\bar{x} = 0$, $K=130$)

Fig. 4.23 Dimensionless volumetric flowrates under the effect of electroosmosis and pressure gradient ($\beta_1 = \beta_2 = \beta_3 = 1$, $\alpha_1 = \alpha_2 = \alpha_3 = 1$, $\bar{\xi}_3 = \bar{\xi}_6 = \bar{\xi}_{wall}$)

Fig. 4.24 H-shaped microchannel: (a) the fabricated device used in the experiment; (b) Schematic representation of the three-fluid microchannel

Fig. 4.26 Measurement results at a flowrate of $q_1 = q_2 = q_3 = 0.05$ ml/h :

(a) $E_{x1} = E_{x3} = 200$ V/cm; (b) $E_{x1} = E_{x3} = -200$ V/cm;

Fig. 4.27 Normalized concentration distribution of the fluorescent dye across the channel width under different applied voltages at a flowrate of $q_1 = q_2 = q_3 = 0.05$ ml/h : (a) $E_{x1} = E_{x3} = 200$ V/cm; (b) $E_{x1} = E_{x2} = -200$ V/cm

Fig. 4.28 Cross sectional view of the three-fluid flow in the main channel

Fig. 4.29 Interface control and flow focusing of three-fluid flow ($q_1 = q_2 = q_3 = 0.05 \text{ ml/h}$): (a) Purely hydrodynamic focusing; (b) Combined electroosmotic/hydrodynamic focusing; (c) Focusing and switching to the bottom; (d) Combined electroosmotic/hydrodynamic extension; (e) Extension and switching to the bottom.

Fig. 4.30 Relationship between focused width e_2 and the applied voltage for different flowrates, ($\beta_1 = \beta_3 = 1$, $\beta_2 = 1.24/0.85 \approx 1.46$ and $\chi = 20$)

Fig. 4.31 Relationship between e_2 and the applied voltage for different viscosity ratios, when $\beta_1 = \beta_3 = 1$, $q_1 = q_2 = q_3 = 0.1$ ml/h and $\chi = 20$.

Fig. 4.32 Relationship between e_2 and the applied voltage for different aspect ratio χ when $q_1 = q_2 = q_3 = 0.1 \text{ ml/h}$, $\beta_1 = \beta_3 = 1$, $\beta_2 = 1.24/0.85 \approx 1.46$

Fig. 4.33 Dependence of the pressure gradient on the externally applied electric field for different flowrates, when $\beta_1 = \beta_2 = \beta_3 = 1$ and $\chi = 20$

Fig. 4.34 Schematic representation of three-fluid electroosmotic switching: (a) switching without droplets; (b) Switching with droplets

Fig. 4.35 Flow switching: (a) Layout of the PDMS microchannel; (b) Measured result of three-fluid electroosmotic flow switching (color).

Fig. 4.36 Droplet switching: (a) layout of the PMMA microchannel for droplet switching; (b) measured result (color).

Fig. 4.37 Process of defining the liquid fraction: (a) The location (y - y) of the measurement; (b) Normalized concentration distribution of the fluorescent dye across the channel width; (c) Liquid fractions of the three flows; (d) Cross sectional view of the three fluids in the main channel; (e) Cross sectional view of the switching droplets system in the main channel.

Fig. 4.38 Flow focusing and switching based on the combined effect of hydrodynamic and electroosmosis (color coded) ($q_1 = q_3 = 0.3 \text{ ml/h}$; $q_2 = 0.1 \text{ ml/h}$): (a) $V_{x1} = -1700(\text{V})$, $V_{x3} = 1700(\text{V})$; (b) $V_{x1} = -1000(\text{V})$, $V_{x3} = 300(\text{V})$; (c) $V_{x1} = -700(\text{V})$, $V_{x3} = -700(\text{V})$; (d) $V_{x1} = -300(\text{V})$, $V_{x3} = -300(\text{V})$; (e) $V_{x1} = 300(\text{V})$, $V_{x3} = -1200(\text{V})$; (f) $V_{x1} = 1700(\text{V})$, $V_{x3} = -1700(\text{V})$.

Fig. 4.39 Droplet switching based on the combined effect of hydrodynamic and electroosmosis (color coded) ($q_1 = q_2 = q_3 = 0.1 \text{ ml/h}$): (a) $V_{x1} = -1500(\text{V})$, $V_{x3} = 1500(\text{V})$; (b) $V_{x1} = -1200(\text{V})$, $V_{x3} = 1200(\text{V})$; (c) $V_{x1} = 0(\text{V})$, $V_{x3} = 0(\text{V})$; (d) $V_{x1} = 1200(\text{V})$, $V_{x3} = -1200(\text{V})$; (e) $V_{x1} = 1500(\text{V})$, $V_{x3} = -1500(\text{V})$.

(a)

(b)

Fig. 4.40 Relationship between the location of the sample fluid and the applied voltages V_{x1} and V_{x3} . ($\beta_1 = \beta_3 = 1$, $q_1 = q_3 = 0.3 \text{ ml/h}$, $q_2 = 0.1 \text{ ml/h}$; $\xi_{\text{PDMS}} = -85 \text{ mV}$, $\xi_{\text{oil}} = -40 \text{ mV}$): (a) $\beta_2 = 1$; (b) $\beta_2 = 5.43$.

Fig. 4.41 Relationship between the location of the sample stream and the applied voltages ($\beta_1 = \beta_3 = 1$, $\beta_2 = 5.43$, $q_1 = q_3 = 0.3\text{ml/h}$, $q_2 = 0.1\text{ml/h}$, $V_{x3} = -V_{x1}$, $\xi_{\text{PDMS}} = -85\text{ mV}$, $\xi_{\text{oil}} = -40\text{ mV}$)

Chapter 5 Modeling of tunable optofluidic lens based on the combined effect of hydrodynamic and electroosmosis

5.1. Introduction

Microlenses are widely integrated into lab-on-a-chip (LOC) systems to improve the portability of optical components and make the system cost-effective [185, 186]. However, the focal length of the solid lens is not tunable and the smoothness of interface of the solid lens depends on the fabrication process. Micro optofluidic, which combines the fields of microfluidic and micro-optics, is implemented by controlling liquid interfaces in microchannels. Fluids have unique properties that cannot be found in solid equivalents, these properties can be used to design novel optical components. These properties are: 1) the optical property can be changed by simple replacing one fluid with another; 2) interface between two immiscible fluids is smooth; 3) the miscible fluids can create gradient in optical properties by diffusion [187]; 4) finer features are possible to be achieved in the composed fluid medias [187]. In microfluidic devices, the flows are always laminar due to the small size of the devices [168]. Under laminar condition in a micro device, the interfaces between different fluids are stable and smooth, thus ideal for designing optical components such as waveguides, lenses and mirrors [188]. This chapter presents a new design of a circular chamber to realize an optically smooth lens curvature; the curvature can be adjusted using the combined effect of pressure-driven and electroosmosis. Fig. 5.1 shows the schematic concept of this optofluidic lens. The two cladding streams (cladding fluid 1 and cladding fluid 3) are electrically conducting with high electroosmotic mobility, while the core stream (core fluid 2) is non-conducting with low electroosmotic mobility. At a given pressure gradient and voltages applied along the conducting cladding fluids, electroosmotic forces control the curvature of the interfaces (interface 1-2 and interface 3-2) between the conducting cladding fluids and the core fluid. The curvatures of the interfaces depend on the directions and magnitudes of the applied voltages. This chapter focuses on the fluidic aspect of the lens design under the

combined effect of pressure-driven flow and electroosmotic flow. It includes numerical model and experimental investigations.

5.2. Numerical simulation

5.2.1. Governing equations

Fig. 5.1 (a) shows the schematic of the problem. The main part of the system is circular chamber with diameter of 1 mm and thickness of 0.05 mm. The channel has the uniform thickness of 0.05 mm. Fig. 5.1 (b) shows the zone of simulation in numerical simulation. The circular chamber is filled with three immiscible fluids (two high electroosmotic mobility fluids and one low electroosmotic mobility fluid). The high electroosmotic mobility fluids 1 and 3 are introduced through the inlet 1 and inlet 3 respectively; the low electroosmotic mobility of fluid 2 is introduced through the inlet 2. To illustrate the concept, the volumetric flow rate of the fluids are fixed, electric fields (V_{x1} and V_{x3}) are applied along the high electroosmotic mobility fluids 1 and 3, respectively. The flow is affected by the combined effect of pressure gradient and electroosmotic effect.

According Eq. (3.24) and Eq. (3.18), the steady state, incompressible continuity and momentum equations for three-Newtonian fluid flow under the combined effect of pressure gradient and electric field in Cartesian coordinate system can be written as

$$\nabla \cdot (\rho_i \mathbf{v}) = 0 \quad (5.1)$$

$$\nabla \cdot (\rho_i \mathbf{v} \mathbf{v}) - \mu_i \nabla^2 \mathbf{v} = -\nabla p + \mathbf{E} \rho_e \quad (5.2)$$

where $\mathbf{v}$ is the velocity vector, p is the pressure, ρ and μ are the density and viscosity appropriate for the phase occupying the particular spatial location, ρ_e is the net electric charge density, $\mathbf{E}$ is the electric field intensity, and the subscript of i means the fluids. The electric field, $\mathbf{E}$, in Eq. (5.2) is related to the electric potential, ϕ , through

$$\mathbf{E} = -\nabla \phi \quad (5.3)$$

The electric potential, ϕ , can be decomposed into two parts for small Debye length and small zeta potential; these two parts are the potential due to the charge acquired at the wall ψ , and the potential due to the external electric field φ [38]

$$\phi = \psi + \varphi \quad (5.4)$$

Furthermore, the potential due to the charge acquired at the wall ψ , is governed by Poisson equation as follows

$$\nabla^2 \psi = -\frac{\rho_e}{\varepsilon} \quad (5.5)$$

and the potential due to the external electric field is governed by Laplace equation as

$$\nabla^2 \varphi = 0 \quad (5.6)$$

where ε is the electric permittivity of the fluids.

According the definition of Eq. (3.30) to Eq. (3.37), the dimensionless form of the governing equations can be described as

$$\begin{cases} \nabla \cdot (\bar{\rho}_i \bar{\mathbf{v}}) = 0 \\ \nabla \cdot (\bar{\rho}_i \bar{\mathbf{v}} \bar{\mathbf{v}}) = -\nabla \bar{p} + \mathbf{G} \cdot \bar{\rho}_e + \frac{1}{\text{Re}} \bar{\mu}_i \nabla^2 \bar{\mathbf{v}} \\ \nabla^2 \bar{\varphi} = 0 \\ \nabla^2 \bar{\psi} = K^2 \bar{\psi} \\ \bar{\rho}_e = -\bar{\psi} \end{cases} \quad (5.7)$$

where $\mathbf{G}$ is the non-dimensional effect of electric field defined as $\mathbf{G} = \frac{2z_0 en_0 L_{ref} \mathbf{E}}{\rho_{ref} U_{ref}^2}$, Re is

the Reynolds number defined as $\text{Re} = \frac{\rho_{ref} U_{ref} L_{ref}}{\mu_{ref}}$, K is the electrical double layer

parameter defined as $K = \frac{L_{ref}}{\kappa_D}$, where $\frac{1}{\kappa} = \left(\frac{\varepsilon k_b T}{2z_0^2 e^2 n_0} \right)^{\frac{1}{2}}$.

5.2.2 Boundary conditions

The boundary conditions of velocities and electric potential are presented in Fig. 5.2. They can be described as follows

$$\text{(at the inlet 1)} \begin{cases} u = 0 \\ \phi = V_{1,in} \\ Q_{1,in} = v_{1,in} \cdot w \end{cases} \quad (5.8)$$

$$\text{(at the inlet 2)} \begin{cases} Q_{2,in} = u_{2,in} \cdot h \\ v = 0 \\ \phi = 0 \end{cases} \quad (5.9)$$

$$\text{(at the inlet 3)} \begin{cases} u = 0 \\ Q_{3,in} = v_{3,in} \cdot w \\ \phi = V_{3,in} \end{cases} \quad (5.10)$$

$$\text{(at the wall)} \begin{cases} u = 0 \\ v = 0 \\ \frac{\partial \phi}{\partial n} = 0 \end{cases} \quad (5.11)$$

$$\text{(at the outlet 1)} \begin{cases} Q_{1,out} = Q_{1,in} \\ v = 0 \\ \phi = V_{1,out} \end{cases} \quad (5.12)$$

$$\text{(at the outlet 2)} \begin{cases} Q_{2,out} = Q_{2,in} \\ v = 0 \\ \phi = 0 \end{cases} \quad (5.13)$$

$$\text{(at the outlet 3)} \begin{cases} Q_{3,out} = Q_{3,in} \\ v = 0 \\ \phi = V_{3,out} \end{cases} \quad (5.14)$$

where u is the velocity in x -axis and v is the velocity in y -axis, $u_{2,in}$ is the average velocity of inlet 2, $v_{1,in}$ and $v_{3,in}$ are the average velocities in inlet 1 and inlet 3 respectively, $V_{1,in}$ and $V_{3,in}$ are the electric voltages in inlet 1 and inlet 3 respectively, $Q_{1,in}$, $Q_{2,in}$ and $Q_{3,in}$ are the volume flow rates of inlets respectively, $Q_{1,out}$, $Q_{2,out}$ and $Q_{3,out}$ are the volume flow rates of outs respectively, w is the width of inlet 1 and inlet 3, h is the height of inlet 2 (Fig. 5.1(b)).

5.2.3 Interface definition

The level-set method is applied to treat the interfaces between different fluids [189].

The level-set function is defined as

$$\mathcal{G} = \begin{cases} -d & \mathbf{r} \in \Omega_{\text{high electrical mobility fluids}} \\ 0 & \mathbf{r} \in \Gamma_{\text{interface}} \\ d & \mathbf{r} \in \Omega_{\text{low electrical mobility fluids}} \end{cases} \quad (5.15)$$

where $\mathbf{r}$ is the position vector in the computational domain Ω , Γ is the interface, and d is the shortest normal distance from the interface Γ .

The governing equation for the evolution of the level-set function is described as

$$\mathbf{v} \cdot \nabla \mathcal{G} = 0 \quad (5.16)$$

In the solution of Eq. (5.16), any convenient reference value can be assigned to the interface. The value of $\mathcal{G}$ at all node points can be calculated based on the reference value at the interface. The level-set function $\mathcal{G}$ is the normal distance from the interface and satisfies

$$|\nabla \mathcal{G}| = 1 \quad (5.17)$$

Eq. (5.15) shows that the value of $\mathcal{G}$ is set to zero at an interface and $\mathcal{G}$ has opposite signs in adjacent liquids. In order to work properly, $\mathcal{G}$ must remain the function of Eq. (5.17). However, the function of Eq. (5.17) can only be ensured at the beginning of the iteration process where the location of the interface is assumed and the values of $\mathcal{G}$ at all nodes are specified. During the iteration process, the values of $\mathcal{G}$ are calculated using Eq. (5.16). Although the interfaces are still represented by the reference values, the other values of $\mathcal{G}$ may not be the shortest distance to the interface. In order to maintain the properties of level-set function, another scalar variable is defined and solved. This variable must be a distance function and has the same interface value as $\mathcal{G}$. This scalar variable is defined as

$$\frac{\partial \zeta}{\partial \tilde{t}} = \text{sign}(\mathcal{G})(1 - |\nabla \mathcal{G}|) \quad (5.18)$$

where $\tilde{t}$ is a pseudo time for the variable ζ . The value of sign is given by

$$\text{sign}(\mathcal{G}) = \frac{\mathcal{G}}{\sqrt{\mathcal{G}^2 + |\nabla \mathcal{G}|^2 (\Delta x)^2}} \quad (5.19)$$

where Δx is the grid size. The steady state solution of ς always satisfies the properties of level-set function. The initial condition of Eq. (5.18) is

$$\varsigma(\mathbf{r}, 0) = \mathcal{G}(\mathbf{r}) \quad (5.20)$$

Although the variable ς satisfies the level-set function, it suffers a significant drawback that the conservation of mass for the various phase cannot be ensured. To ensure the mass conservation at each cross section, a local mass correction factor is defined and an additional equation is solved as [190]

$$\frac{\partial \varsigma'}{\partial \tilde{t}} = \dot{m}_{cor} \quad (5.21)$$

In Eq. (5.21), $\tilde{t}$ is pseudo-time and can be different from the pseudo-time of $\tilde{t}$, $\dot{m}_{cor}$ is the mass conservation factor. The mass conservation factor is calculated using the following equation

$$\dot{m}_{cor} = \text{sign}(\mathcal{G}) \frac{(\dot{m}_d - \dot{m}_c)}{\dot{m}_d} \quad (5.22)$$

where $\dot{m}_d$ is the desired mass flow rate and $\dot{m}_c$ is the most current local mass flow rate of the reference phase. The mass flow rate of the reference phase can be calculated using the following equation

$$\dot{m} = \begin{cases} \sum \rho_{ref} (1 - H_\gamma) u \Delta A & H_{r,ref} = 0 \\ \sum \rho_{ref} H_\gamma u \Delta A & H_{r,ref} = 1 \end{cases} \quad (5.23)$$

where ΔA is the cross-section of one grid and the summation is performed over a cross-section. In this paper, the desired mass flow rate is calculated using the inlet conditions.

5.2.4 Properties

The density ρ , the dynamic viscosity μ and the electric permittivity ε are calculated using

$$\alpha = (1 - H_r) \alpha_{\text{low electrical mobility fluid}} + H_r \alpha_{\text{high electrical mobility fluid}} \quad (5.24)$$

where α means the density ρ , the dynamic viscosity μ and the electric permittivity ε . The Heaviside function H_γ is related to the normal distance from the interface and is calculated [189]

$$H_\gamma = \begin{cases} 0 & \text{for } \mathcal{G} < -\gamma \\ \frac{\mathcal{G} + \gamma}{2\gamma} + \frac{1}{2\pi} \sin\left(\frac{\pi\mathcal{G}}{\gamma}\right) & \text{for } |\mathcal{G}| \leq \gamma \\ 1 & \text{for } \mathcal{G} > \gamma \end{cases} \quad (5.25)$$

where $\gamma = 1.5\Delta x$ [189].

5.2.5 Depth average method

The size of model along x direction and y direction is larger than that of z direction. The velocity profile in z direction is shown in Fig. 5.3. The electroosmotic velocity of cross section along z direction is plug profile [13, 14, 38], it is a function of electric field $\bar{u}^E = \int u^E dz = f(E)$; the pressure driven velocity is parabolic profile, it is a function of pressure gradient $\bar{u}^P = \int u^P dz = f(p)$. The depth average method [191] is used in this chapter to reduce the 3D model to 2D model.

5.2.6 Solution procedure

The solution procedure can be summarized as follows

- (1) Guess the locations of the interfaces.
- (2) Calculate the value of $\mathcal{G}$ for all nodes for the interface.
- (3) Calculate the properties for all nodes using Eqs. (5.24) and (5.25).
- (4) Solve the distribution of electric potentials using Eqs. (5.7).
- (5) Calculate the average velocity using the depth average method.
- (6) Solve the continuity and momentum equations given by Eqs. (5.7).
- (7) Calculate the value of $\mathcal{G}$ using Eq. (5.16) according the velocity distribution of step (6).
- (8) Calculate the value of ς according the solution of step (7).
- (9) Calculate the value of ς' using Eq. (5.21) according the solution of step (8).

(10) Set $\mathcal{G}(\mathbf{r}) = \zeta'(\mathbf{r})$

(11) Repeat steps (3)-(10) until the solution converges.

5.2.7 Numerical method

The continuity equation, momentum equation, electric potential equations, and level-set equations can be described using the following equations

$$\rho \frac{\partial \Phi}{\partial t} + \rho u_j \frac{\partial \Phi}{\partial x_j} = \frac{\partial}{\partial x_j} \left(\Gamma \frac{\partial \Phi}{\partial x_j} \right) + S \quad (5.26)$$

where Φ , ρ , Γ and S are the dependent variables, density, diffusion coefficient and source term respectively. Eq. (5.26) is solved using the finite-volume method of Patankar [192]. A staggered grid is used in this article. The scalar variables are stored at the centers of the control volumes, and the velocities are located at the control volume faces. In this paper, the power-law is used to calculate the combined convection-diffusion effect in the momentum equations. The convection of the level-set equations is modeled by the first-order unwind scheme. The velocity-pressure coupling is resolved using SIMPLER algorithm. The transient term is discretized using the fully implicit scheme. The resulting algebraic equations are solved using the TriDiagonal Matrix Algorithm.

5.3 Experiment

5.3.1 Materials and fabrication

The fabrication technique of the microchannel is similar to that of the previous microchannel (section 4.3.2). An optofluidic lens with a chamber diameter of 1 mm and channel width of 100 μm was fabricated. As shown in Fig. 5.1, the three fluids flow side by side in the straight microchannel in the direction from left to right. The flows at the inlets are driven by syringe pumps. The outlets are connected to a waste reservoir. Voltages are applied by inserting two pairs of platinum electrodes.

5.3.2 Chemical and experimental setup

In the experiment, the cladding liquid is the aqueous NaCl solution (7×10^{-4} M). Rhodamine B is added into the aqueous NaCl to allow flow visualization of the interface shapes. The core fluid is silicone oil (Sigma-Aldrich), the viscosity and the density of the silicone oil is 1cSt and 0.82 g/ml.

5.4 Validation of the numerical scheme

The interface shape and positions in the micro chamber using various computational grids are shown in Fig. 5.4. Uniform grid is used in the simulations.

In the simulations, the width of the inlets (w) 1 and 3 are $100 \mu\text{m}$; the height of the inlet 2 is $100 \mu\text{m}$; the diameter of the circular chamber (D) is 1 mm. The voltages added to cladding fluids are $V_{x1} = V_{x3} = 800 \text{ V}$. The flowrates of cladding fluids and core fluid are $q_1 = q_2 = q_3 = 0.05 \text{ ml/h}$. The properties of the fluid 1 are reference quantities. The permittivity of conduction fluid is $6.9 \times 10^{-1} (\text{C}^2 \text{N}^{-1} \text{m}^{-2})$ [13]. All fluids have the same viscosity $\mu_1 = \mu_2 = \mu_3 = 0.85 \times 10^{-3} \text{ Ns/m}^2$. The reference length is $L_{ref} = D$; the reference velocity is the average velocity of fluid 1 of $U_{ref} = Q_{1,in} / w$ and the Reynolds number is about 3×10^{-4} .

The results presented in Fig. 5.4 show the decay of discretization error with increased the number of grid. It can be seen that the exact solution [193] is reproduced accurately when the grid number in circular chamber exceeds 17000^2 . It means that the interface evolution between immiscible fluids is well captured and the numerical method is suitable. In the following simulation, the grid number in circular chamber is 17000^2 .

5.5 Results and discussion

In the experiment and simulation, the flowrates of cladding fluids and core fluid are kept constant ($q_1 = q_2 = q_3 = 0.05 \text{ ml/h}$); the voltages (V_{x1} and V_{x3}) are regulated using the high voltage power supply. The zeta potential between PMMA and aqueous solution is 24.7 mV [194] and the permittivity of conduction fluid is $6.9 \times 10^{-1} (\text{C}^2 \text{N}^{-1} \text{m}^{-2})$ [195]. The method to determine the interface has been introduced in section 4.3.2.

5.5.1 Fluid-fluid lens under the combined effect of pressure-driven and electroosmosis

We define the field of the core fluid 2 as a region, the interface 1-2 and interface 3-2 are the boundaries of the region, and the curvature of the interface is defined as the divergence of the normal of the interface. The symbol of the curvatures can be defined as $\kappa > 0$ for the convex region and $\kappa < 0$ for the concave region (Fig. 5.5).

The curvature of interface 1-2 (κ_{1-2}) and the curvature of the interface 3-2 (κ_{3-2}) can be calculated using the following formula (Fig. 5.6):

$$\begin{cases} \kappa_{1-2} = -\frac{2B_2}{B_2^2 + R^2} \\ \kappa_{3-2} = \frac{2A_2}{A_2^2 + R^2} \end{cases} \quad (27)$$

where R is the radius of the interface.

Fig. 5.7 compares the measured interface shapes with the numerical simulation. The agreement between the extracted interface shape and numerical simulation confirms that the interface shapes in the circular chamber have a optically smooth arc-shape. The combined effect of hydrodynamic and electroosmosis can be used to control the optofluidic lens. The results also show that numerical model, and experimental results have good agreement with each other.

Fig. 5.8 shows a series of different optofluidic lenses by numerical simulations. Fig. 5.8 (a)-(f) shows that at a fixed flowrate, the curvatures of the interfaces (interfaces 1-2 and 3-2) can be modified according to the magnitudes and the polarities of the voltages.

When negative voltages are added to the cladding fluid 1 and the cladding fluid 3 simultaneously (Fig. 5.8(a)), the electroosmotic flows act against the pressure-driven flow. Consequently, q_1 and q_3 encounter a larger resistance. q_1 and q_3 appear to be more “viscous” due to the electroosmotic effect. Because of the same pressure drop along the microchannel and the fixed volumetric flowrate forced by the syringe pump, the more viscous fluid has to spread over a larger area, thus decreases the curvature of the interface. If positive voltages are added to the cladding fluid 1 and the cladding fluid 3 simultaneously respectively (Fig. 5.8(b)), the electroosmotic velocity has the same

direction with the flow direction and the cladding fluids flow faster. Due to the fixed volumetric flowrate forced by the syringe pump, the cladding fluids have to spread over a smaller area, thus increase the curvature of the interface. Figs. 5.8 (a) and (b) show that a symmetric biconvex lens can be achieved when symmetric voltages ($V_{x1} = V_{x3}$) are added to the cladding fluids.

When the applied voltages are different ($V_{x1} \neq V_{x3}$), the curvatures of the interface 1 and the interface 3 have different values (Fig. 5.8 (c)-(f)). Fig. 5.5 defines the symbol of curvature and the type of the lens. When a negative voltage ($V_{x1} = -600V$), and a positive voltage ($V_{x3} = 600V$) are applied (Fig. 5.8(c)), the curvature of the interface 1 is negative and the curvature of the interface 3 is positive. Under the combined effect of a positive voltage and a negative voltage, a meniscus lens is formed (Fig. 5.8 (c) and (f)). At moderate negative voltage strength, this technique allows the curvatures of both interfaces to be positive, hence a biconvex lenses or a plano-convex lens can be achieved (Fig. 5.8(e))

The above results show that a negative voltage could decrease the curvature of the interface, while a positive voltage can increase the curvature of the interface. Under constant flowrates, the same voltage achieves symmetric biconvex lens only. Different voltages can form biconvex lens, plano-convex lens, and meniscus lens.

5.5.2 Symmetric biconvex lens

First, the symmetric voltages are added to the cladding fluids ($V_{x1} = V_{x3}$). Under the constant flowrates ($q_1 = q_2 = q_3 = 0.05\text{ml/h}$) and the symmetric voltage, symmetric biconvex lenses are achieved. Fig. 5.9 compares the dimensionless lens thickness ($\bar{T}_{hl}$) and the numerical results when symmetric voltages are applied. The dimensionless lens thickness ($\bar{T}_{hl}$) is defined in Fig. 5.6. A reasonable agreement is obtained.

Fig. 5.9 shows that symmetric voltages can be used to regulate the lens thickness. The dimensionless lens thickness $\bar{T}_{hl}$ decreases with the decrease of the voltages. The rate of change of $\bar{T}_{hl}$ is dependent on the polarity of the voltages. When positive voltages are added to the cladding fluids, the rate of change in the dimensionless lens thickness increase rapidly when the applied voltage increases from 0 to 1000 V; hence symmetric

voltages can control the symmetric biconvex lens sensitively. When negative voltages ($-1000 \text{ V} < V_{x1} < 0 \text{ V}$) are added to the cladding fluids, the rate of change in dimensionless lens thickness $\bar{T}_{h1}$ decreases very slowly. The sensitivity can be improved by reducing the diameter of the circular chamber.

5.5.3 Biconvex lens, plan-convex lens, and meniscus lens

Fig. 5.10 compares the measured curvature of the interface with the numerical results when the voltages have opposite polarities ($V_{x1} = -V_{x3}$). The results clearly show that the proposed models agree well with the measurement. Fig. 5.10 shows that under the constant flowrates, different optofluidic lenses can be achieved but a combination of the positive and negative voltages is needed. If there is no voltage, the flow is purely pressure driven. If the cladding flows have the same flowrates, a symmetric biconvex lens is achieved. When different voltages are added to the cladding flows, κ_{1-2} and κ_{3-2} have different values. If the magnitude of the voltage is not high enough (e.g. $-200 \text{ V} < V_{x1} < 200 \text{ V}$), κ_{1-2} and κ_{3-2} are positive at the same time forming a biconvex. κ_{1-2} or κ_{3-2} decreases with the increase of voltages. A plano-convex lens is formed when $\kappa_{1-2} = 0$ or $\kappa_{3-2} = 0$ (e.g. $V_{x1} = -200 \text{ V}$ or $V_{x3} = -200 \text{ V}$). When κ_{1-2} or κ_{3-2} is negative with an increasing magnitude of voltages (e.g. $|V_{x1}| > 200 \text{ V}$), a meniscus lens is formed. The above results show that under a constant flowrate, a biconvex lens, a plano-convex lens, and a meniscus lens can be accomplished. The control voltage can be further reduced with a lower flowrate.

Fig. 5.11 shows that the dimensionless thickness of the optofluidic lens ($\bar{T}_{h1}$) varies with the different voltage. $\bar{T}_{h1}$ is defined in Fig. 5.6. The experiment results are measured using the recording results by CCD camera. Under the same flowrates, the dimensionless thickness has the maximum value without the voltage; it decreases with the increasing voltage. The numerical results have a good agreement with the experimental results.

5.5.4. Velocity fields of center line for different lens model

The detail of velocity fields in circular chamber of biconvex lens and a meniscus lens are shown in Fig. 5.12(a) and Fig. 5.13(a) respectively. The velocities at the inlet and

outlet of the circular chamber are highest due to its smallest cross section. While Fig. 5.12(b) and 5.13(b) show the corresponding velocity u at the center of the chamber. u^p is the velocity component corresponding to hydrodynamic velocity; u^E is the velocity component corresponding to electroosmotic velocity.

Fig. 5.12(b) shows that the velocity distribution is symmetric due to the symmetrical of the biconvex lens. In the analysis, $\mu_1 = \mu_2 = \mu_3$. The flow characteristics depend on the coupling effects of the cladding fluids and core fluid 2 which involve the electrokinetics driving force in the cladding fluids and the interfacial phenomenon. The component of velocity v is zero at the center line of the chamber. u^p is plug profile because the viscosity of fluids has the same values and the aspect ratio is very large [196]. For u^E , when electric fields are applied across the cladding fluids, the cladding fluids 1 and 3 are driven by electroosmosis, which drags the core fluid 2 by the hydrodynamic shear force. The velocity field is affected by viscosity ratios, the strength of the external electric fields and electroosmotic characteristics of the cladding fluids. The electroosmotic velocity profile (u^E) of the core fluid 2 is negative. It is due to the interfacial shear stress dragged by the cladding fluids on the core fluid 2. u is the superposition of the solutions of u^p and u^E .

Figs. 5.13(b) shows the velocity profile of the meniscus lens. u^E changes dramatically due to the different applied electric field. Under the negative electric fields, the electroosmotic velocity is negative. Hence, the velocity profile is asymmetrical. The result indicates that flow fields can be modified by electric fields under same flow rates.

Fig. 5.1 (a) Schematic representation of the optofluidics lens under the combined effect of hydrodynamic and electroosmosis; (b) the domain of simulation

Fig. 5.2 The boundary conditions of simulation

Fig. 5.3 The velocity profile in z direction

Fig. 5.4 the effect of grid resolutions on errors between analytical solution [193] and numerical simulation

Fig. 5.5 The definition of the symbol of curvature and the model of lens. The core fluid is regarded as a region. Convex region has $\kappa > 0$, concave region has $\kappa < 0$, and a plan has $\kappa = 0$. (a) Biconvex lens; (b) Meniscus lens; (c) Plano-convex lens; (d) Meniscus lens.

Fig. 5.6 The interface positions of a lens

Fig. 5.7 The shape of the interface under the different electric fields

Fig. 5.8 The fluid-fluid lenses based on the difference electric fields ($q_1 = q_2 = q_3 = 0.05\text{ml/h}$)

(a) $V_{x1} = -100V$; $V_{x3} = -100V$; (b) $V_{x1} = 600V$; $V_{x3} = 600V$;

(c) $V_{x1} = 600V$; $V_{x3} = -600V$; (d) $V_{x1} = -300V$; $V_{x3} = 300V$;

(e) $V_{x1} = -600V$; $V_{x3} = 600V$; (f) $V_{x1} = 600V$; $V_{x3} = -600V$;

Fig. 5.9 Thickness of the lens verse applied voltage ($q_1 = q_2 = q_3 = 0.05\text{ml/h}$, $L_{\text{ref}} = 1\text{ mm}$, $V_{x1}=V_{x3}$)

Fig. 5.10 A_2 and B_2 with different voltages ($q_1 = q_2 = q_3 = 0.05\text{ml/h}$, $V_{x1} = -V_{x3}$)

Fig. 5.11 Thickness of the lens with different voltages ($q_1 = q_2 = q_3 = 0.05\text{ml/h}$, $L_{\text{ref}} = 1$ mm, $V_{x1} = -V_{x3}$)

(a)

(b)

Fig. 5.12(a) velocity field of the circular chamber; (b) velocity profile of center line of circular chamber ($q_1 = q_2 = q_3 = 0.05 \text{ ml/h}$; $V_{x1} = -100 \text{ V}$; $V_{x3} = -100 \text{ V}$)

(a)

(b)

Fig. 5.13 (a) velocity field of the circular chamber; (b) velocity profile of center line of circular chamber ($q_1 = q_2 = q_3 = 0.05\text{ml/h}$; $V_{x1} = 600\text{V}$; $V_{x3} = -600$)

Chapter 6 Semi-analytical model of mixed electroosmotic/pressure-driven two immiscible fluids with curved interface

6.1. Introduction

In chapter 4, the model of three-fluid flow in microchannel under the combined effect of pressure gradient and electroosmosis with planar interface is presented. In contrast to macro and mini channels, the effect of interface curvature in microchannels is very significant. In microchannel, the gravity effect is neglected [19] while the surface tension is significant causing the Bond number to approach to zero. The results of [15-18] showed that the interface is a circular arc with constant curvature in microchannels.

The aim of chapter 6 is to present a precise calculation method for two-fluid laminar-laminar stratified flow under the combined effect of pressure-driven and electroosmosis where the interface shape is a circular arc with a constant curvature. Fig. 6.1 shows the general configuration of the fluids in microchannels with curved interface. The fluid with high electrical mobility is labeled as fluid 1, while the fluid with low electrical mobility is labeled as fluid 2. The flows are assumed to be at a steady state, fully developed, and therefore unidirectional.

6.2. Mathematical model

6.2.1 Description of the model

In order to analyze the system shown in Fig. 6.1, a Cartesian coordinate system (x, y, z) is used where the origin point, O , is set to be at the centre of the interface curvature. Curved interface with constant curvature is assumed. h_1 , h_2 , and h_s are defined in Fig. 6.2. The relationship between them is $h_1 + h_2 + h_s = h$ where h_s is the height of the curved interface measured from the origin and h is the height of the microchannel. The width of the channel is denoted as $2w$. The contact angle θ_c is the angle at which the

fluid 1 meets the wall. As a result of surface charges, electric double layers (EDLs) form next to the liquid-liquid interface and the channel walls that are in contact with the high electrical mobility fluid. For a more general situation, the walls of the microchannel may be made of different materials, so that the zeta potentials at the bottom wall is denoted as ξ_1 , and at the side walls as ξ_2 , ξ_4 , respectively. The zeta potential at the curved interface is ξ_3 . The flows are along the z direction.

The two fluids are driven by the combined pressure and electroosmotic body force. When the two-fluid flow is fully developed, the velocities of the two liquids, u_1 and u_2 at position (x, y) along the channel are independent of z . The subscripts of 1 and 2 denote the fluid 1 and fluid 2, respectively.

The reference values have been defined using Eqs. (3.30)-(3.37). The interface is concave with a contact angle $\theta_c < 90^\circ$ (Fig. 6.2 (a)) and convex with a contact angle $\theta_c > 90^\circ$ (Fig. 6.2 (c)). Fig. 6.2 (b) corresponds to the planar interface with $\theta_c = 90^\circ$. There are two restrictive conditions as shown in Fig. 6.3: one is that the interface contacts the bottom wall (Fig. 6.3 (a)) and the other is that the interface contacts the top wall (Fig. 6.3 (b)). In our study, the interface is located between the two extreme cases shown in Fig. 6.3 (a) and Fig. 6.3 (b).

The following derivation of analytical model is completed in polar coordinate. The two parameters are defined as follows

$$r = \sqrt{\bar{x}^2 + \bar{y}^2} \quad (6.1)$$

and

$$\theta = \begin{cases} 0 & (\bar{x} \geq 0, \bar{y} = 0) \\ \tan^{-1} \frac{\bar{y}}{\bar{x}} & (\bar{x} > 0, \bar{y} > 0) \\ \frac{\pi}{2} & (\bar{x} = 0, \bar{y} > 0) \\ \pi - \tan^{-1} \frac{\bar{y}}{|\bar{x}|} & (\bar{x} < 0, \bar{y} > 0) \\ \pi & (\bar{x} < 0, \bar{y} = 0) \\ \pi + \tan^{-1} \left| \frac{\bar{y}}{\bar{x}} \right| & (\bar{x} < 0, \bar{y} < 0) \\ \frac{3\pi}{2} & (\bar{x} = 0, \bar{y} < 0) \\ 2\pi - \tan^{-1} \left| \frac{\bar{y}}{\bar{x}} \right| & (\bar{x} < 0, \bar{y} < 0) \end{cases} \quad (6.2)$$

where r and θ are the radius and angle in the polar coordinate. In the derivation, r and θ are used to replace x and y . $\bar{x}$ and $\bar{y}$ are the dimensionless position of an arbitrary point in x -axis and y -axis, respectively.

6.2.2 Electric potential distributions

In cylindrical coordinates, the Poisson-Boltzmann equation of Eq. (4.4) is specified as

$$\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \bar{\psi}}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 \bar{\psi}}{\partial \theta^2} = K^2 \bar{\psi} \quad (6.3)$$

Eq. (6.3) is the modified Bessel's equation. Using the Separated Variable Method in polar coordinate, the solution of Eq. (6.3) can be described as

$$\bar{\psi}(r, \theta) = \sum_{\lambda=0}^{\infty} I_{\lambda}(Kr) [A_{1\lambda} \cos(\lambda\theta) + A_{2\lambda} \sin(\lambda\theta)] \quad (6.4)$$

The boundary condition of the zeta potential can be defined as

$$\bar{\psi}(r_i, \theta_i) = Z_i \quad (6.5)$$

where the subscript i is the serial number of the boundary condition. Using the description of Eq. (6.4), Eq. (6.5) transforms into

$$\sum_{\lambda=0}^J I_{\lambda}(Kr_i) [A_{1\lambda} \cos(\lambda\theta_i) + A_{2\lambda} \sin(\lambda\theta_i)] = \bar{\psi}(r_i, \theta_i) \quad (6.6)$$

where $I_\lambda(Kr)$ is the modified Bessel functions of order λ of the first kind. The real coefficients of $A_{I\lambda}$ and $A_{2\lambda}$ can be numerically determined from the Dirichlet boundary conditions of the specified value of zeta potential on the channel walls and interface, which can be calculated using the least square method in Appendix A and Appendix B.

6.2.3 Velocity driven by pressure gradient ($\bar{u}_1^p$ and $\bar{u}_2^p$)

Due to linearity, the velocity of the high electrical mobility fluid 1 and of the low electrical mobility fluid 2 can be decomposed into two parts:

$$\bar{u} = \bar{u}^p + \bar{u}^E \quad (6.7)$$

where $\bar{u}^E$ corresponds to the velocity driven by electroosmotic force, and $\bar{u}^p$ is the velocity driven by pressure gradient.

The steady state Navier-Stokes equation with pressure driven can be described as

$$\nabla^2 \bar{u}_1^p = \text{Re} \frac{d\bar{p}}{d\bar{z}} \quad (\text{fluid 1}) \quad (6.8)$$

and

$$\nabla^2 \bar{u}_2^p = \frac{\text{Re}}{\beta} \frac{d\bar{p}}{d\bar{z}} \quad (\text{fluid 2}) \quad (6.9)$$

where β is the dynamic viscosity ratio $\beta = \mu_2 / \mu_1$, Re is the Reynolds number,

$\text{Re} = \frac{\rho_{\text{ref}} U_{\text{ref}} L_{\text{ref}}}{\mu_{\text{ref}}}$, $\frac{d\bar{p}}{d\bar{z}}$ is the pressure gradient along flow direction. In polar coordinates,

Eq. (6.8) is specified as

$$\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \bar{u}_1^p}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 \bar{u}_1^p}{\partial \theta^2} = \text{Re} \frac{d\bar{p}}{d\bar{z}} \quad (6.10)$$

In order to convert Eq. (6.10) to a Laplace equation, the following transformation is used

$$\bar{u}_1^p = \text{Re} \frac{d\bar{p}}{d\bar{z}} \left(\frac{r^2}{4} - \bar{u}_1^{p*} \right) \quad (6.11)$$

Substituting Eq. (6.11) into Eq. (6.10), it results in

$$\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \bar{u}_1^{p*}}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 \bar{u}_1^{p*}}{\partial \theta^2} = 0 \quad (6.12)$$

Eq. (6.12) is the modified Bessel's equation. Using the Separated Variable Method in polar coordinate, the solution of Eq. (6.12) can be described as

$$\bar{u}_1^{p*} = a_{10}^p + \sum_{j=1}^J r^j (a_{1j}^p \cos(j\theta) + b_{1j}^p \sin(j\theta)) \quad (6.13)$$

According Eq. (6.11)

$$\bar{u}_1^p = \text{Re} \frac{dp}{dz} \left(\frac{r^2}{4} - a_{10}^p - \sum_{j=1}^J r^j (a_{1j}^p \cos(j\theta) + b_{1j}^p \sin(j\theta)) \right) \quad (6.14)$$

The non-slip boundary conditions can be used as

$$a_{10}^p + \sum_{j=1}^J r^j (a_{1j}^p \cos(j\theta) + b_{1j}^p \sin(j\theta)) = \frac{r_i^2}{4} \quad (\text{for fluid 1}) \quad (6.15)$$

The parameters of a_{10}^p , a_{1j}^p , b_{1j}^p are constants for specified boundary conditions.

Following the same approach, the hydrodynamic velocity of fluid 2 is

$$\bar{u}_2^p = \frac{\text{Re}}{\beta} \frac{dp}{dz} \left(\frac{r^2}{4} - a_{20}^p - \sum_{j=1}^J r^j (a_{2j}^p \cos(j\theta) + b_{2j}^p \sin(j\theta)) \right) \quad (6.16)$$

The non-slip boundary conditions can be expressed as

$$a_{20}^p + \sum_{j=1}^J r^j (a_{2j}^p \cos(j\theta) + b_{2j}^p \sin(j\theta)) = \frac{r_i^2}{4} \quad (\text{for fluid 2}) \quad (6.17)$$

The matching conditions at the interface can be described as

$$\begin{cases} \bar{u}_1^p(r_i, \theta_i) = \bar{u}_2^p(r_i, \theta_i) \\ \frac{\partial \bar{u}_1^p(r_i, \theta_i)}{\partial n} = \beta \frac{\partial \bar{u}_2^p(r_i, \theta_i)}{\partial n} \end{cases} \quad (\text{at the interface}) \quad (6.18)$$

According Eqs. (6.15) and (6.17), the non-slip boundary conditions can be described using the following equations

$$\begin{cases} a_{10}^p + 0 \cdot a_{20}^p + \sum_{j=1}^J r_i^j (a_{1j}^p \cos(j\theta) + b_{1j}^p \sin(j\theta) + a_{2j}^p \cdot 0 + b_{2j}^p \cdot 0) = \frac{r_i^2}{4} & (\text{fluid 1}) \\ 0 \cdot a_{10}^p + a_{20}^p + \sum_{j=1}^J r_i^j (a_{1j}^p \cdot 0 + b_{1j}^p \cdot 0 + a_{2j}^p \cos(j\theta) + b_{2j}^p \sin(j\theta)) = \frac{r_i^2}{4} & (\text{fluid 2}) \end{cases} \quad (6.19)$$

The matching conditions of Eq. (6.18) can be described using Eq. (6.20)

$$\beta \cdot a_{10}^p - a_{20}^p + \sum_{j=1}^J \left(\beta \cdot r_i^j \cdot \cos(j\theta_i) + C_1^p(j) \right) \cdot a_{1j}^p + \left(\beta \cdot r_i^j \cdot \sin(j\theta_i) + S_1^p(j) \right) b_{1j}^p = (\beta - 1) \cdot \frac{r_i^2}{4} \quad (6.20)$$

where $C_1^p(j)$ and $S_1^p(j)$ can be described as

$$C_1^p(j) = r_i^{j-1} \cdot \cos(j\theta_i) \cdot j \cdot \cos\theta_i + r_i^{(j-2)} \cdot \sin(j\theta_i) \cdot j \cdot \sin\theta_i \quad (6.21)$$

$$S_1^p(j) = r_i^{j-1} \cdot \sin(j\theta_i) \cdot j \cdot \cos\theta_i - r_i^{(j-2)} \cdot \cos(j\theta_i) \cdot j \cdot \sin\theta_i \quad (6.22)$$

The real coefficients of a_{10}^p , a_{20}^p , a_{1j}^p , b_{1j}^p , a_{2j}^p , and b_{2j}^p in Eq. (6.19) can be calculated using the least square method in Appendix A and Appendix C, and then the velocity profile due to pressure gradient can be described using Eqs. (6.14) and (6.16).

6.2.4 Velocity driven by electroosmotic force ($\bar{u}_1^E$ and $\bar{u}_2^E$)

The steady state Navier-Stokes equation under the external electric field can be described as

$$\mu_1 \nabla^2 u_1^E = -\rho_e E \quad (6.23)$$

Combining the Poisson equation of Eq. (3.3) and Eq. (6.23), the Navier-Stokes equation transforms into

$$\mu_1 \nabla^2 u_1^E = \varepsilon_0 \varepsilon_r \cdot \nabla^2 \psi \cdot E \quad (6.24)$$

The dimensionless form of Eq. (6.24) is

$$\frac{1}{\text{Re}} \bar{\mu}_1 \nabla^2 \bar{u}_1^E = \bar{E} \cdot \nabla^2 \bar{\psi} \quad (6.25)$$

where $\bar{E} = \frac{k_b T \varepsilon_0 \varepsilon_r E}{L_{ref} U_{ref}^2 \rho_{ref} z_0 e}$. Eq. (6.25) can be rewritten as

$$\nabla^2 \left(\bar{u}_1^E - \frac{\text{Re} \cdot \bar{E}}{\bar{\mu}_1} \cdot \bar{\psi} \right) = 0 \quad (6.26)$$

A new velocity is defined as

$$\bar{u}_1^{E*} = \bar{u}_1^E - \frac{\text{Re} \cdot \bar{E}}{\bar{\mu}_1} \cdot \bar{\psi} \quad (6.27)$$

Substituting Eq. (6.27) into Eq. (6.26), it results in:

$$\nabla^2 \bar{u}_1^{E*} = 0 \quad (6.28)$$

In polar coordinates, Eq. (6.28) is specified as

$$\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \bar{u}_1^{E*}}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 \bar{u}_1^{E*}}{\partial \theta^2} = 0 \quad (6.29)$$

The solution of Eq. (6.29) can be described using the following equation

$$\bar{u}_1^{E*} = a_{10}^E + \sum_{j=1}^J r^j (a_{1j}^E \cos(j\theta) + b_{1j}^E \sin(j\theta)) \quad (6.30)$$

From Eq. (6.27), the distribution of the electroosmotic velocity is

$$\bar{u}_1^E(r, \theta) = a_{10}^E + \frac{\text{Re} \cdot \bar{E}}{\bar{\mu}_1} \cdot \bar{\psi}(r, \theta) + \sum_{j=1}^J r^j (a_{1j}^E \cos(j\theta) + b_{1j}^E \sin(j\theta)) \quad (6.31)$$

The non-slip boundary conditions can be expressed as

$$a_{10}^E + \sum_{j=1}^J r_i^j (a_{1j}^E \cos(j\theta_i) + b_{1j}^E \sin(j\theta_i)) = -\frac{\text{Re} \cdot \bar{E}}{\bar{\mu}_1} \cdot \bar{\psi}(r_i, \theta_i) \quad (6.32)$$

Using similar method, the electroosmotic velocity of fluid 2 is

$$\bar{u}_2^E = a_{20}^E + \sum_{j=1}^J r^j (a_{2j}^E \cos(j\theta) + b_{2j}^E \sin(j\theta)) \quad (6.33)$$

The non-slip boundary conditions can be defined as

$$a_{20}^E + \sum_{j=1}^J r_i^j (a_{2j}^E \cos(j\theta) + b_{2j}^E \sin(j\theta)) = 0 \quad (6.34)$$

The matching conditions at the interface can be described as

$$\begin{cases} \bar{u}_1^E(r_i, \theta_i) = \bar{u}_2^E(r_i, \theta_i) \\ \frac{\partial \bar{u}_1^E(r_i, \theta_i)}{\partial n} = \beta \frac{\partial \bar{u}_2^E(r_i, \theta_i)}{\partial n} - M \bar{\rho}_e^s \end{cases} \quad (6.35)$$

where M is the electrokinetic effect in the matching conditions, $M = \frac{\varepsilon_0 \varepsilon_r k_b T E}{z_0 e U_{ref} \mu_{ref}}$, $\bar{\rho}_e^s$ is

the density of the surface charge at the interface, $\bar{\rho}_e^s = -\frac{\partial \bar{\psi}}{\partial n}$. From Eqs.(6.32), and (6.34),

the non-slip boundary conditions can be described using the following equations

$$\begin{cases} a_{10}^E + 0 \cdot a_{20}^E + \sum_{j=1}^J r_i^j (a_{1j}^E \cos(j\theta_i) + b_{1j}^E \sin(j\theta_i)) + 0 \cdot a_{2j}^E + 0 \cdot b_{2j}^E = -\frac{\text{Re} \cdot \bar{E}}{\bar{\mu}} \cdot \bar{\psi}(r_i, \theta_i) & \text{(for fluid 1)} \\ 0 \cdot a_{10}^E + a_{20}^E + \sum_{j=1}^J r_i^j (0 \cdot a_{1j}^E + 0 \cdot b_{1j}^E + a_{2j}^E \cos(j\theta) + b_{2j}^E \sin(j\theta)) = 0 & \text{(for fluid 2)} \end{cases} \quad (6.36)$$

The matching conditions of Eq. (6.35) can be described as

$$\begin{aligned}
a_{10}^E - a_{20}^E + \sum_{j=1}^J \left[r_i^j \cos(j\theta_i) + C_1^E(j) \right] a_{1j}^E + \left[r_i^j \sin(j\theta_i) + S_1^E(j) \right] b_{1j}^E \\
- \left[r_i^j \cos(j\theta_i) + \beta C_2^E(j) \right] a_{2j}^E - \left[r_i^j \sin(j\theta_i) + \beta S_2^E(j) \right] b_{2j}^E \\
= -\frac{\text{Re} \cdot \bar{E}}{\bar{\mu}_1} \cdot \bar{\psi}(r_i, \theta_i) - M \bar{\rho}_e^s - \frac{\text{Re} \cdot \bar{E}}{\bar{\mu}_1} \cdot \frac{\partial \bar{\psi}(r_i, \theta_i)}{\partial n}
\end{aligned} \quad (6.37)$$

where, $C_1^E(j)$, $S_1^E(j)$, $C_2^E(j)$, and $S_2^E(j)$ can be described as

$$C_1^E(j) = C_2^E(j) = r_i^{j-1} \cdot \cos(j\theta_i) \cdot j \cdot \cos \theta_i + r_i^{(j-2)} \cdot \sin(j\theta_i) \cdot j \cdot \sin \theta_i \quad (6.38)$$

$$S_1^E(j) = S_2^E(j) = r_i^{j-1} \cdot \sin(j\theta_i) \cdot j \cdot \cos \theta_i - r_i^{(j-2)} \cdot \cos(j\theta_i) \cdot j \cdot \sin \theta_i \quad (6.39)$$

Similar to the velocity driven by pressure gradient, the real coefficients of a_{10}^E , a_{20}^E , a_{1j}^E , b_{1j}^E , a_{2j}^E , and b_{2j}^E in Eq. (6.36) can also be calculated using the method in Appendix A and Appendix D, and the velocity profile driven by electroosmotic force can be described using Eqs. (6.31) and (6.33).

6.2.5 Volumetric flowrates

The dimensionless volumetric flowrates through the rectangular-cross-section channel can be defined as $\bar{q}_1 = \bar{q}_1^E + \bar{q}_1^P = q_1 / (L_{ref}^2 U_{ref})$ and $\bar{q}_2 = \bar{q}_2^E + \bar{q}_2^P = q_2 / (L_{ref}^2 U_{ref})$.

The dimensionless flowrates are given as

$$\bar{q}_1^E = \iint \bar{u}_1^E(r, \theta) dr d\theta \quad (6.40)$$

$$\bar{q}_1^P = \iint \bar{u}_1^P(r, \theta) dr d\theta \quad (6.41)$$

$$\bar{q}_2^E = \iint \bar{u}_2^E(r, \theta) dr d\theta \quad (6.42)$$

$$\bar{q}_2^P = \iint \bar{u}_2^P(r, \theta) dr d\theta \quad (6.43)$$

Substituting $\bar{u}_1^E$, $\bar{u}_1^P$, $\bar{u}_2^E$, and $\bar{u}_2^P$ into Eqs. (4.80)-(6.43), the dimensionless volumetric flowrates can be calculated.

6.3. Convergence and comparison with previous results

Before the analysis of the results obtained via the analytical model, it is necessary to confirm the accuracy of the solution scheme. A convergence test has been performed to

determine the accuracy of the solution with respect to the number of boundary (N) and with respect to the highest order of Bessel' function (J).

Fig. 6.4 (a) shows the detail comparisons of the fully developed velocity profile ($x = 0$) obtained using various computational grids with the exact solution when $J = 17$. The following parameters are used in the calculation: $K = 13$, $\theta_c = 90^0$, $h_1 = 0.5$, $h_2 = 0.5$, $\bar{\xi}_1 = \bar{\xi}_2 = \bar{\xi}_4 = 1$, $\bar{\xi}_3 = 0$, $\mu_1 = \mu_2$. The results presented in Fig. 6.4 (a) show the decay of discretization error with increased the total grid number N. It can be seen that the exact solution is reproduced accurately. Fig. 6.4 (b) shows the comparisons of the fully developed velocity profiles obtained using four values of J with the exact solution when the total special boundary condition number is $N = 280$. J is the highest order of modified Bessel functions. Under the same total number of boundary condition N, a large value of J corresponds to a smaller error of calculation, the velocity profile becomes more accurate.

From all of above results, we can conclude that the interface between the two fluids is well captured, and this semi-analytical method is suitable. In the following simulations, $N = 280$ and $J = 17$ are used.

The validity of the analytical model with curved interface has been checked by comparing with experimental results available in the literature. For the comparison, the interface is planar ($h_s = 0$, $h_1 = 0.5$, $h_2 = 0.5$, $\theta_c = 90^0$), $N = 280$ and $J = 17$. Fig. 6.5 shows the comparison of the velocity profile between the semi-analytical model and the experimental results of data [12] at the symmetric line ($x = 0$) of a rectangular microchannel. While Fig. 6.6 shows the comparison between the analytical solution [13] and the semi-analytical solution for the two-fluid electroosmotic flow in rectangular microchannels. Our results agree well with the published data.

6.4. Results and discussion

To investigate the effects of interface shape on the electric potential and velocity distributions, we fix the liquid factions of the two fluids. The liquid fractions of fluid 1 and fluid 2 are defined as follows:

$$e_1 = \frac{\left\{ \left(\frac{w^2 + h_s^2}{2h_s} \right)^2 \cdot \arctan \left(\frac{w}{\left(\frac{w^2 + h_s^2}{2h_s} \right) - h_s} \right) - w \cdot \left(\left(\frac{w^2 + h_s^2}{2h_s} \right) - h_s \right) + 2wh_1 \right\}}{2wh} \quad (\text{convex})$$

$$e_1 = \frac{h_1}{h} \quad (\text{planar})$$

$$e_1 = \frac{\left\{ |h_s| \cdot 2w - \left(\frac{w^2 + h_s^2}{2 \cdot |h_s|} \right)^2 \cdot \arctan \left(\frac{w}{\left(\frac{w^2 + h_s^2}{2 \cdot |h_s|} \right) - |h_s|} \right) + w \cdot \left(\left(\frac{w^2 + h_s^2}{2 \cdot |h_s|} \right) - |h_s| \right) + 2wh_1 \right\}}{2wh} \quad (6.44)$$

(concave)

and

$$e_2 = 1 - e_1 \quad (6.45)$$

where e_1 and e_2 are the liquid fractions of fluid 1 and fluid 2, $|h_s|$ is the absolute value of the interface height measured from the origin. In the following calculation, the liquid fractions are $e_1 = e_2 = 0.5$. When h_s is fixed, h_1 can be calculated using Eq. (6.44), and $\bar{h}_2 = 1 - \bar{h}_s - \bar{h}_1$.

6.4.1 Electric potential

Figs. 6.7 (a, b, c) show the 2-D electric potential distributions in the high electric mobility fluid 1 for the concave interface ($h_s = 0.2$, $\theta_c = 50^\circ$), the planar interface ($h_s = 0$, $\theta_c = 90^\circ$) and the convex interface ($h_s = -0.2$, $\theta_c = 130^\circ$), respectively. While Fig. 6.6 shows the corresponding electric potential at the symmetric line of the rectangular microchannel. The other parameters are $K = 13$, $e_1 = e_2 = 0.5$, $\bar{\xi}_1 = \bar{\xi}_2 = \bar{\xi}_3 = \bar{\xi}_4 = 1$, $\bar{w} = 0.5$, $\bar{h} = 1$, $N = 280$ and $J = 17$.

The results indicate that the interface shape influences the electric potential distribution of the two-fluid flow in the rectangular microchannel. When two interfaces (wall and liquid interface) are sufficiently close to each other, the electric double layers will interact [46]. The effects of the EDL interaction are important to the electrokinetic flows within the microchannel.

Fig. 6.8 shows the interface shapes of the concave, planar and the convex interfaces at the same liquid fraction of 0.5. Sections a-a, b-b and c-c are indicated in Fig. 6.8. Sections a-a, b-b and c-c are located at $x = 0$, $x = 0.3$ and $x = 0.4$ respectively. The potential and velocity distributions in these sections will be investigated.

Figs. 6.9 (a, b, c) compares the electric potential distribution for the different cases: concave, planar and convex interfaces at the same liquid fraction of various sections a-a, b-b and c-c respectively. In the calculation, $h_s = 0.2$ for the concave interface, $h_s = 0$ for the planar interface, and $h_s = -0.2$ for the convex interface, $K = 13$, $h_1 = 0.5 - h_s$, $\bar{\xi}_1 = \bar{\xi}_2 = \bar{\xi}_3 = \bar{\xi}_4 = 1$, $\bar{w} = 0.5$, $\bar{h} = 1$, $N = 280$ and $J = 17$, $e_1 = e_2 = 0.5$, h_1 can be calculated using Eq. (6.44), and then $\bar{h}_2 = 1 - \bar{h}_s - \bar{h}_1$. Due to surface charges, EDLs form at the liquid-liquid interface and channel walls that are in contact with the conducting fluid 1. In general, the electric potential decreases rapidly from the channel wall/liquid-liquid interface to the bulk region of the conducting liquid. The results show that the electric potential and the net charge density are highest for the concave interface due to the closest distance between the liquid-liquid interface and the channel walls. The concave interface exhibits sharp corner (region A in Fig. 6.7) at the liquid-wall interface where the EDL overlaps (the calculated EDL thickness is $\frac{1}{\kappa} \approx 100nm$). This overlapped EDL results in higher values of the electric potential and net charge density.

6.4.2 Velocity distributions and flowrates

From Eq. (6.7), the velocity u of two-fluid can be decomposed into two parts, u^p and u^E . u^p corresponds to velocity driven by pressure gradient and u^E corresponds to velocity driven by electroosmotic effect. To investigate the influences of interface shape on the velocity distributions, we analyze the following cases: (1) Two-fluid

electroosmotic flow, u^E with zero pressure gradient; (2) The two-fluid is driven by the combined electroosmotic force and pressure gradient.

Two-fluid electroosmotic flow

With zero pressure gradient applied across the microchannel, the flow is mainly a two-fluid electroosmotic flow.

Figs. 6.10 (a, b, c) show the comparison of the dimensionless velocity profiles, u^E for the concave, planar and convex interfaces at sections a-a, b-b, c-c respectively. In the simulation, we fix the liquid fraction ($e_1 = e_2 = 0.5$) of the two fluids and the applied electric field $E_x = 1000$, $\bar{\mu}_1 = \bar{\mu}_2 = 1$, $\bar{\rho}_1 = \bar{\rho}_2 = 1$, the other parameters are the same as presented in section 6.4.1.

When electric field is applied across the conducting fluid, the conducting fluid 1 is driven by electroosmosis, which drags the non-conducting fluid 2 by the hydrodynamic shear force. By this way, the non-conducting fluid can be delivered by electroosmosis. Comparison of the velocity distributions at different sections (a-a, b-b and c-c) of the graphs shows that the concave interface produces the highest electroosmotic velocity. This is due to the larger value of the electric potential and net charge density induced by the concave interface. The velocity profile of the non-conducting fluid is passive.

Fig. 6.11 shows the influence of the interface shape on the dimensionless volumetric flowrate. For a given liquid fraction ($e_1 = e_2 = 0.5$) and applied electric field $E_x = 1000$, the volumetric flowrate of the conducting fluid increases with decreasing in contact angle (θ_c).

The results show that the volumetric electroosmotic flowrate of the conducting fluid with the concave interface it is about 1.5 times larger than that of planar interface when the contact angle is about 0° which is due to the larger value of the electric potential and hence higher electroosmotic velocity.

Two-fluid flow driven by electroosmosis and pressure gradient

When both pressure gradient and electric field are applied, the flow is driven by electroosmotic body force and pressure gradient. In the analysis, we fix the liquid fraction

($e_1 = e_2 = 0.5$), electric field $E_x = 1000$, pressure gradient $d\bar{p}/d\bar{z} = 50000$, and viscosities $\bar{\mu}_1 = \bar{\mu}_2 = 1$.

Figs. 6.12 (a, b, c) show the comparison of the dimensionless velocity profiles, u , for the concave, planar and convex interfaces at sections a-a, b-b, c-c respectively. The result shows that the velocity profile, $\bar{u}$ is the superposition of the solutions of the electroosmotic velocity profile $\bar{u}^E$ and the pressure-driven parabolic profile, $\bar{u}^P$.

As $\bar{\mu}_1 = \bar{\mu}_2 = 1$, $\bar{\rho}_1 = \bar{\rho}_2 = 1$, the pressure driven velocity $\bar{u}^P$ exhibits a parabolic velocity profile. Comparison of u^P , at different sections (a-a, b-b, c-c) shows that near the wall (section c-c) the velocity is affected by the non-slip boundary condition hence resulted in a lower velocity. The results also show u^P is independent of the interface shape when $\bar{\mu}_1 = \bar{\mu}_2 = 1$.

Comparison of the velocity profiles at different sections a-a and c-c illustrate that at section a-a, the pressure driven velocity $\bar{u}^P$ is distinctive larger than electroosmotic velocity $\bar{u}^E$. At section a-a (region far away from EDL and the side wall), the fluid is influenced by (i) the faster lamina fluid layer immediately closer to the center due to the applied pressure gradient and (ii) the interfacial shear stress. Hence $\bar{u}^P$ dominates.

At section c-c, the fluid is influenced by the no-slip boundary condition and the viscous shear diffusion due to (i) the faster lamina fluid layers at the EDL region and (ii) the faster lamina fluid layers immediately closer to the center. Hence in this section the electroosmosis effect dominates.

Fig. 6.1 Schematic of the cross section of two-fluid flow in microchannel with curved interface and coordinate system

(a) Concave ($\theta_c < 90^\circ$, $h_s > 0$)

(b) Planar ($\theta_c = 90^\circ$, $h_s = 0$)

(c) Convex ($\theta_c > 90^\circ$, $h_s < 0$)

Fig. 6.2 Various interface shapes in a rectangular microchannel

Fig. 6.3 The extreme cases of the interface location

(a) Various N and fixed $J = 17$

(b) Various J and fixed $N = 280$

Fig. 6.4 Electroosmotic velocity profiles for various value of N and J

Fig. 6.5 Comparison of the velocity profile between the semi-analytical model and experimental data [12]

Fig. 6.6 Dimensionless electric potential distribution at the symmetric line in the conducting fluid 1

Fig. 6.7 The electric potential distributions of conducting fluid 1

Fig. 6.8 Schematic illustration of the concave, planar and convex interfaces with liquid fraction of 0.5 ($e_1 = e_2 = 0.5$)

(a) Dimensionless electric potential distributions at section a-a

(b) Dimensionless electric potential distributions at section b-b

(c) Dimensionless electric potential distributions at section c-c

Fig. 6.9 Comparison of the dimensionless electric potential distribution at various sections ($e_1 = e_2 = 0.5$, $E_x = 1000$, $\bar{\mu}_1 = \bar{\mu}_2 = 1$)

(a) Dimensionless electroosmotic velocity distribution at section a-a

(b) Dimensionless electroosmotic velocity distribution at section b-b

(c) Dimensionless electroosmotic velocity distribution at section c-c

Fig. 6.10 Dimensionless electroosmotic velocity distribution at various sections

($e_1 = e_2 = 0.5$, $E_x = 1000$, $d\bar{p}/d\bar{z} = 0$, $\bar{\mu}_1 = \bar{\mu}_2 = 1$)

Fig. 6.11 (a) Variation of the interface shapes with the contact angle ($e_1 = e_2 = 0.5$)

Fig. 6.11 (b) Dimensionless volumetric flowrate of conducting fluid versus contact angle ($e_1 = e_2 = 0.5$, $E_x = 1000$, $\bar{\mu}_1 = \bar{\mu}_2 = 1$)

(a) Dimensionless velocity distribution at section a-a

(b) Dimensionless velocity distribution at section b-b

(c) Dimensionless velocity distribution at section c-c

Fig. 6.12 Dimensionless velocity distribution at various sections ($e_1 = e_2 = 0.5$, $E_x = 1000$, $d\bar{p}/d\bar{z} = 50000$, $\bar{\mu}_1 = \bar{\mu}_2 = 1$)

Chapter 7 The linear stability of two-fluid flow in a microchannel for parallel electric field

7.1 Introduction

7.1.1 Introduction of instability

A dynamical system is said to be unstable if a small perturbation of its equilibrium state increases exponentially in time; the system is stable if the perturbation die out in time. Chapters 4-6 analyze the exact steady state solution of three-fluid flow in microchannel. However, these stationary solutions exist only within stable range. Within this stable range any fluctuation imposed on the stationary flow will decay in time leaving the steady solution unaltered. Outside the range, the small fluctuations which are always present within the flow will destabilize the flow and the stationary solution will cease to exist. The outside regime is termed unstable. Such transitions from stable to unstable regimes are the stability of system.

In a micro total analysis systems (μ TAS), flow stability is a critical condition for many electronics and biological applications. The stability of the interface between a conducting fluid and a non-conducting fluid is critical to the operating limit and the accuracy of a μ TAS. Under the combined effect of hydrodynamics and electroosmosis, electrohydrodynamic instability occurs at the interface. The linear stability of the interface has both fundamental and practical interest. On the practical side, the interface between two immiscible liquids is subjected to flow disturbances. On the fundamental side, the stability mechanisms are influenced by the interplay of transport phenomena between the immiscible liquids.

In the previous chapters, the stable electroosmotic phenomenon in microchannels is studied. In the following chapter, the instability of electroosmotic phenomenon is shown. The instability of the two-fluid interface is the basic phenomenon responsible for either efficient mixing or droplet formation; it is also a critical effect for electroosmotic pump. By now, it is still far from being understood due to the strong coupling between the fluid

dynamic and the electrostatic problem [151]. The aim of this chapter is to study the effect of parallel electric fields on two viscous fluids under the constant flowrates in microchannels. Analytical model and experimental methods are shown.

7.1.2 Methodology and approach

The usual procedure in linear stability analysis can follow six basic steps:

(1). Introduce general governing equations and general boundary conditions. The governing equations control the initial and stationary flow conditions in the velocity field $\mathbf{V}_0$. The governing equations include the continuity equation, the momentum equations, the Maxwell equations and the Poisson-Boltzmann equation.

(2). The perturbation of $\mathbf{v}'$ is introduced and superposed to the stationary velocity $\mathbf{V}_0$. The velocity field changes into $\mathbf{v} = \mathbf{v}_0 + \mathbf{v}'$. Substituting the equation into governing equations and boundary conditions, the equations governing the perturbation are obtained.

(3). Linearized the equations governing the perturbation, the terms with quadratic and higher terms in $\mathbf{v}'$ are neglected.

(4). The perturbation quantities $\mathbf{v}'$ are further expressed in terms of normal modes. A spatially and temporally periodic perturbation can be assumed in the form of $\mathbf{v}'_{\mathbf{k}}(\mathbf{r}, t) = v_{\mathbf{k}}(z) \exp(-i\omega_{\mathbf{k}} t) \exp(i(\mathbf{k} \cdot \mathbf{r}))$

(5). Substituting equation of $\mathbf{v}'_{\mathbf{k}}(\mathbf{r}, t) = v_{\mathbf{k}}(z) \exp(-i\omega_{\mathbf{k}} t) \exp(i(\mathbf{k} \cdot \mathbf{r}))$ into governing equations, the temporal stability can be solved. The complex solution for $\omega_{\mathbf{k}}$ can be described as $\omega_{\mathbf{k}} = \text{Real}(\omega_{\mathbf{k}}) + i \text{Im}(\omega_{\mathbf{k}})$.

(6). The freely evolving waves are spatially periodic disturbances of infinite spatial extent which travel with a phase velocity, $c_r \equiv \omega_r / k$ and grow or decrease in amplitude with a temporal growth rate, $\text{Im} \omega_{\mathbf{k}}$. The system is considered to be linearly unstable to infinitesimal disturbances for $\text{Im} \omega_{\mathbf{k}} > 0$. The stability regimes can be distinguished as

$$\text{follows: } \text{Im}(\omega_{\mathbf{k}}) \begin{cases} > 0 \longrightarrow \text{instability} \\ = 0 \longrightarrow \text{neutral} \\ < 0 \longrightarrow \text{stability} \end{cases} .$$

7.2 The physical and mathematical model

7.2.1 General equations of motion

Fig. 7.1 (a) shows the stability model of the interface between two immiscible fluids under the combined effect of hydrodynamic and electroosmosis. Fluid 1 is conducting with high electroosmotic mobility, and fluid 2 is non-conducting with low electroosmotic mobility. Both fluids have different properties. The flows are driven by pressure and electric field. The two fluids have different properties. The applied electric field is parallel to the unperturbed interface. Fig. 7.1 (b) shows the cross sectional view of the two fluids in the rectangular microchannel, h_1 and h_2 are denoted as the fraction of conducting fluid 1 and non-conducting fluid 2, respectively.

The following notation will be used: superscripts (1) and (2) denote quantities pertaining to fluid 1 and fluid 2. The superscript ' indicates a perturbed variable; subscript 0 denotes the unperturbed variables, other subscripts are used for vector component.

The momentum equation for an isotropic incompressible Newtonian liquid is given by Eq. (3.41); the continuity equation of incompressible liquid is described using Eq. (3.40); the governing equations for the free charge can be described as Eq. (3.38).

The governing equations for the electric field within the dielectrics are

$$\nabla \cdot \bar{E} = 0 \quad (7.1)$$

Since large currents are not present in this flow, the effect of magnetic induction is negligible. Hence, the electric field is irrotational

$$\nabla \times \bar{E} = 0 \quad (7.2)$$

7.2.2 General boundary conditions

On the wall of the microchannel, the non-slip condition is assumed for the velocities

$$\mathbf{v}^{(1)} = 0; \quad \text{at } z = -h_1 \quad (7.3)$$

$$\mathbf{v}^{(2)} = 0; \quad \text{at } z = h_2 \quad (7.4)$$

When the electric fields are added to the fluids, the boundary conditions of the electric field at the wall can be described as

$$\mathbf{n} \cdot \mathbf{E}^{(1)} = 0 \quad \text{at } z = -h_1 \quad (7.5)$$

and

$$\mathbf{n} \cdot \mathbf{E}^{(2)} = 0 \quad \text{at } z = h_2 \quad (7.6)$$

where $\mathbf{n}$ is the normal direction of the wall.

Considering the effect of a small wave disturbance on the interface $z = 0$, the surface disturbance is assumed as

$$z = \zeta(x, y, t) \quad (7.7)$$

where ζ is the vertical displacement from the equilibrium position $z = 0$. Any deformed surface is characterized by a unit normal vector $\mathbf{n}$:

$$\mathbf{n} = \frac{1}{\sqrt{\left(\frac{\partial \zeta}{\partial x}\right)^2 + \left(\frac{\partial \zeta}{\partial y}\right)^2 + 1}} \left(-\frac{\partial \zeta}{\partial x}, -\frac{\partial \zeta}{\partial y}, 1\right) \quad (7.8)$$

The linear of Eq. (7.8) is

$$\mathbf{n} = \left(-\frac{\partial \zeta}{\partial x}, -\frac{\partial \zeta}{\partial y}, 1\right)$$

f is defined as the surface function of the two immiscible fluids. The change in any function $f(x, y, z, t)$ across the interface is defined by f

$$f = f(x, y, \zeta^-, t) - f(x, y, \zeta^+, t) = (f^{(1)} - f^{(2)})_{z=\zeta} \quad (7.9)$$

where the superscripts of $-$ and $+$ mean the side of interface nearing fluid 1 and fluid 2 respectively. At the interface $z = \zeta$, the continuity of the velocities are

$$u = v = w = 0 \quad (7.10)$$

$$w^{(1)} = w^{(2)} = \frac{d\zeta}{dt} = \frac{\partial \zeta}{\partial t} + u \frac{\partial \zeta}{\partial x} \quad (7.11)$$

where u, v, w are the velocities along x, y, z directions respectively.

The continuity of parallel electric fields at the interface can be described as

$$\mathbf{n} \cdot \varepsilon \mathbf{E} = 0 \quad (7.12)$$

$$\mathbf{n} \times \mathbf{E} = 0 \quad (7.13)$$

The stress balance at any part on the interface can be described using the following equation

$$-p \quad n_i + \left[\tau_{ik} + T_{ik}^M \right] n_k + \rho_e^s \cdot E_i = \sigma \nabla_s^2 \zeta n_i \quad (7.14)$$

where p is the pressure, τ_{ik} is the viscous stress, T^M is the Maxwell stress tensor, ρ_e^s is the surface charge, σ is the surface tension, $\nabla_s^2 \zeta$ describes the surface curvature for small deformations.

7.2.3 Perturbation equations

The base state solution

In the base state, the velocity field and the continuity equation can be described by the unperturbed Eqs. (3.41) and (3.40) respectively. Under unperturbed condition, Eqs. (3.41) and (3.40) transform into

$$\frac{\partial(\bar{\rho} \bar{\mathbf{u}}_0)}{\partial \bar{t}} = -\nabla \bar{p} + \bar{\mathbf{E}} \bar{\rho}_e + \frac{1}{\text{Re}} \bar{\mu} N^2 \bar{\mathbf{u}}_0 \quad (7.15)$$

$$\nabla \cdot \bar{\mathbf{u}}_0 = 0 \quad (7.16)$$

Under unperturbed condition, the detail of velocity profile and pressure gradient has been described in chapter 4.

The vector $\mathbf{n}$ normal to the unperturbed interface is given by

$$\mathbf{n}_0 = (0, 0, 1) \quad (7.17)$$

The base state solution in each region $i = 1, 2$, are

$$p_0^{(i)} = P_0^{(i)} \quad (7.18)$$

$$\mathbf{u}_0^{(i)} = \mathbf{U}_0 \quad (7.19)$$

$$E_{x0}^{(i)} = E_{x0}^{(i)} \quad (7.20)$$

where $P_0^{(i)}$ and $\mathbf{U}_0$ are pressure gradient and velocity profile calculated from the base state equations (7.15) and (7.16); $E_{x0}^{(i)}$ is the external electric field. According the explanation of Goranovic [19], the stress balance at the interface can be calculated using following method:

In the case of isotropic and linear media, the stress tensor σ_{ik}^M is the symmetric tensor and can be described as

$$\sigma_{ik}^M = -P_0 \delta_{ik} - \frac{1}{2} \varepsilon_r \varepsilon_0 \left[1 - \frac{\rho}{\varepsilon_r} \left(\frac{\partial \varepsilon_r}{\partial \rho} \right)_T \right] E^2 \delta_{ik} + \varepsilon_r \varepsilon_0 E_i E_k \quad (7.21)$$

At each side of the interface, the forces are equal and opposite, can be described as

$$\left[\left[\sigma_{ik}^M \right] \right] n_k = 0 \quad (7.22)$$

In the homogeneous dielectric liquid, the pressure can be redefined by electrostriction term $-\frac{1}{2} \cdot \left[\left[\varepsilon E_{x0}^2 \right] \right]$ [19]. The pressure is the combined effect of electrostriction and pressure gradient. The terms of pressure can be described as

$$P_{0 \text{ } nx} = P_{0 \text{ } ny} = 0 \quad (7.23)$$

$$P_{0 \text{ } nz} = -\frac{1}{2} \cdot \left[\left[\varepsilon E_{x0}^2 \right] \right] + P \quad (7.24)$$

where the subscripts of nx , ny and nz mean three components of normal vector in x , y and z directions respectively.

Linearization of the perturbed equations

The analysis involves the assumption that the perturbations to the base state are infinitesimally small. To carry out a linear stability analysis, all high-order terms in the perturbation quantities are neglected. The expanding solutions are

$$\mathbf{u} = \mathbf{u}_0 + \mathbf{u}' + \dots \quad (7.25)$$

$$p = p_0 + p' + \dots \quad (7.26)$$

Under perturbed condition, Eqs. (3.41) and (3.40) transform into

$$\frac{\partial (\bar{\rho}(\bar{\mathbf{u}}_0 + \bar{\mathbf{u}}'))}{\partial \bar{t}} = -\nabla (\bar{p}_0 + \bar{p}') + \bar{\mathbf{E}} \bar{\rho}_e + \frac{1}{\text{Re}} \bar{\mu} \nabla^2 (\bar{\mathbf{u}}_0 + \bar{\mathbf{u}}') \quad (7.27)$$

$$\nabla \cdot (\bar{\mathbf{u}}_0 + \bar{\mathbf{u}}') = 0 \quad (7.28)$$

In this thesis, the changes in density and viscosity are not noticed since in each region they are assumed constant [19]. When subtracting the base state solutions of Eqs. (7.15)-(7.16) from Eqs. (7.27)-(7.28), the equations reduce to

$$\rho \frac{\partial \bar{\mathbf{u}}'}{\partial t} = -\nabla \bar{p}' + \frac{1}{\text{Re}} \mu \nabla^2 \bar{\mathbf{u}}' \quad (7.29)$$

$$\nabla \cdot \bar{\mathbf{u}}' = 0 \quad (7.30)$$

Combining Eq. (7.29) and Eq. (7.30), leads to:

$$\nabla^2 \bar{p}' = 0 \quad (7.31)$$

For a constant charge density, the electric field can be described as

$$\nabla^2 \bar{E}_x' = 0 \quad (7.32)$$

As the electric field is irrotational, $\bar{\mathbf{E}}'$ is

$$\nabla \times \bar{\mathbf{E}}' = 0 \quad (7.33)$$

Linearization of the perturbed boundary conditions of rigid boundaries

The boundary of the wall under perturbed conditions can be linearized as follows

$$w^{(1)'} = u^{(1)'} = v^{(1)'} = 0 \quad \text{at } z = -h_1 \quad (7.34)$$

$$w^{(2)'} = u^{(2)'} = v^{(2)'} = 0 \quad \text{at } z = h_2 \quad (7.35)$$

$$\mathbf{n}_0 \cdot \bar{\mathbf{E}}^{(1)'} = 0 \quad \text{at } z = -h_1 \quad (7.36)$$

$$\mathbf{n}_0 \cdot \bar{\mathbf{E}}^{(2)'} = 0 \quad \text{at } z = h_2 \quad (7.37)$$

Linearization of the perturbed boundary conditions of interface

Since ζ is assumed to be small, the first order approximation is used to present the

conditions at $z = 0$. Then, the approximation is $E_y'(0 + \zeta) \approx E_y'(0) + \frac{\partial E_y'}{\partial z}(0) \cdot \zeta$. Because ζ

is assumed to be small, hence

$$E_y'(0 + \zeta) = E_y'(0) + \frac{\partial E_y'}{\partial z}(0) \cdot \zeta + \dots \approx E_y'(0) \quad (7.38)$$

is valid if both E_y' and ζ are small. In Eq. (7.38), $E_y'(0)$ is the perturbation of electric field in y direction at $z = 0$ and $E_y'(0 + \zeta)$ is the perturbation of electric field in y direction at $z = \zeta$. The normal vector is now

$$\mathbf{n} = \mathbf{n}_0 + \mathbf{n}' \quad (7.39)$$

where $\mathbf{n}'$ is

$$\mathbf{n}' = \left(-\frac{\partial \zeta}{\partial x}, -\frac{\partial \zeta}{\partial y}, 0\right) \quad (7.40)$$

The boundary conditions for perturbation at $z = \zeta$ are

$$u' = v' = w' = 0 \quad (7.41)$$

$$w^{(1)'}(0) = w^{(2)'}(0) = \frac{\partial \zeta}{\partial t} + u \frac{\partial \zeta}{\partial x} \quad (7.42)$$

The electric fields are

$$\mathbf{E} = \left(E_{x0} + E'_x, E'_y, E'_z\right) \quad (7.43)$$

The conditions at the interface are

$$E'_x = 0 \quad (7.44)$$

$$\left[-\varepsilon E_{x0} \frac{\partial \zeta}{\partial x} + \varepsilon E'_z \right] = 0 \quad (7.45)$$

Linearized stresses

In a microsystem, the effect of gravity is ignored. Substituting Eq. (7.22) into Eq. (7.14), the stress conditions of the interface can be described

$$-p' n_i - P n_i + \left[\tau_{ik} + T_{ik}^M \right] n_k + \rho_e^s \cdot E_i = \sigma \nabla_s^2 \zeta n_i \quad (7.46)$$

Eq. (7.46) can be transformed into the following equations in x, y, z directions

$$-\left(\left[\varepsilon E_{x0}^2 \right] - P \right) \cdot \frac{\partial \zeta}{\partial x} + \left[\mu \left(\frac{\partial u'}{\partial z} + \frac{\partial w'}{\partial x} \right) \right] + \rho_e^s \cdot \left[E'_x \right] + \left[\varepsilon E_{x0} E'_z \right] = 0 \quad (7.47)$$

$$P \cdot \frac{\partial \zeta}{\partial y} + \rho_e^s \cdot \left[E'_y \right] + \left[\mu \left(\frac{\partial v'}{\partial z} + \frac{\partial w'}{\partial y} \right) \right] = 0 \quad (7.48)$$

$$-p' + 2\mu \frac{\partial w'}{\partial z} + \rho_e^s \cdot \left[E'_z \right] + \left[-\varepsilon E_{x0} E'_x \right] = \sigma \nabla_s^2 \zeta \quad (7.49)$$

Expansion in normal modes

The perturbation can be expanded into the normal modes using the following equations

$$\zeta = \hat{\zeta} \exp \left[i \left(k_x x + k_y y \right) + nt \right] \quad (7.50)$$

$$\mathbf{u}' = \hat{\mathbf{u}}(z) \exp \left[i \left(k_x x + k_y y \right) + nt \right] \quad (7.51)$$

$$p' = \hat{p}(z) \exp \left[i \left(k_x x + k_y y \right) + nt \right] \quad (7.52)$$

$$E'_x = \hat{E}_x(z) \exp \left[i \left(k_x x + k_y y \right) + nt \right] \quad (7.53)$$

where $n = -i\omega$, ω is the complex phase velocity of the disturbance, $k^2 = k_x^2 + k_y^2$, k is the magnitude of the total wave number, i is the standard imaginary unit. Using Eqs. (7.50)-(7.53), the stress balance at the interface (Eq. (7.47)) is transformed into

$$\left[\varepsilon E_{x0}^2 \right] \cdot k_x^2 \hat{\zeta} - P \cdot k^2 \hat{\zeta} - \left[\mu \left(\frac{d^2}{dz^2} + k^2 \right) \hat{w} \right] - \rho_e^s \cdot \left[\frac{d\hat{E}_z}{dz} \right] + \left[\varepsilon E_{x0} \cdot \frac{d\hat{E}_x}{dz} \right] = 0 \quad (7.54)$$

Solutions and linearized boundary conditions

The boundary conditions for velocity (Eqs. (7.3) and (7.4)) and electric field (Eqs. (7.5) and (7.6)) can be described using the following equations

$$\hat{w}^{(1)} = A \sinh k(z + h_1) \quad (7.55)$$

$$\hat{w}^{(2)} = B \sinh k(z - h_2) \quad (7.56)$$

$$\hat{E}_x^{(1)} = C_1 \exp(kz) + C_2 \exp(-kz) \quad (7.57)$$

$$\hat{E}_x^{(2)} = D_1 \exp(kz) + D_2 \exp(-kz) \quad (7.58)$$

$$\hat{E}_z^{(2)} = \frac{k}{ik_x} \left[C_1 \exp(kz) - C_2 \exp(-kz) \right] \quad (7.59)$$

$$\hat{E}_z^{(2)} = \frac{k}{ik_x} \left[D_1 \exp(kz) - D_2 \exp(-kz) \right] \quad (7.60)$$

Substituting Eqs. (7.55)-(7.60) into boundary conditions of Eqs. (7.34)-(7.37), the boundary conditions are transformed into

$$\hat{w}^{(1)}(-h_1) = 0 \quad (7.61)$$

$$\hat{w}^{(2)}(h_2) = 0 \quad (7.62)$$

$$\hat{E}_x^{(1)}(-h_1) = \hat{E}_y^{(1)}(-h_1) = \hat{E}_z^{(1)}(-h_1) = 0 \quad (7.63)$$

$$\hat{E}_x^{(2)}(h_2) = \hat{E}_y^{(2)}(h_2) = \hat{E}_z^{(2)}(h_2) = 0 \quad (7.64)$$

At the interface, the continuity of velocities of perturbation for both fluids has the same values; this condition is transformed into

$$\llbracket \hat{w}(0) \rrbracket = \llbracket \frac{d\hat{w}(0)}{dz} \rrbracket = 0 \quad (7.65)$$

Combining Eqs. (7.42), (7.50) and (7.55), we can obtain that

$$\hat{\zeta} = \frac{\hat{w}^{(1)}(0)}{(n + u \cdot ik_x)} = \frac{\hat{w}^{(1)}(0)}{m} \quad (7.66)$$

where

$$m = n + u \cdot ik_x \quad (7.67)$$

Eqs. (7.44)-(7.46) describing the stress balance at the interface are transformed into following forms:

$$\llbracket \hat{E}_x(0) \rrbracket = 0 \quad (7.68)$$

$$\llbracket -i\varepsilon k_x E_{x0} \frac{\hat{w}(0)}{m} + \varepsilon \hat{E}_z(0) \rrbracket = 0 \quad (7.69)$$

$$\begin{aligned} & \llbracket \left[-\rho n + \mu \left(\left(\frac{d}{dz} \right)^2 - 3k^2 \right) \right] \frac{d\hat{w}(0)}{dz} \rrbracket \\ & - k^2 \left(\rho_e^s \cdot \llbracket \hat{E}_z(0) \rrbracket + \llbracket -\varepsilon E_{x0} \hat{E}_x(0) \rrbracket + \sigma k^2 \frac{\hat{w}^{(0)}(0)}{m} \right) = 0 \end{aligned} \quad (7.70)$$

$$\begin{aligned} & \left(\llbracket \varepsilon E_{x0}^2 \rrbracket \cdot k_x^2 - P \cdot k^2 \right) \frac{\hat{w}^{(1)}(0)}{m} - \llbracket \mu \left(\left(\frac{d}{dz} \right)^2 + k^2 \right) \hat{w}(0) \rrbracket \\ & - q_s \cdot \llbracket \frac{d\hat{E}_z(0)}{dz} \rrbracket + \llbracket \varepsilon E_{x0} \frac{d\hat{E}_z(0)}{dz} \rrbracket = 0 \end{aligned} \quad (7.71)$$

7.2.4 The principle of exchange of stabilities

In Eq. (7.50), if $n < 0$, the disturbances decay exponentially to zero as time lapses; the disturbance cannot induce the instability of the interface and the two-fluid electroosmotic flow is stable. If $n > 0$, the disturbances advance exponentially to a large

value as time lapses; the disturbance can be enlarged to induce the instability of interface and the two-fluid electroosmotic flow is unstable. According the boundary conditions, Eqs. (7.55)-(7.58) reduce to

$$\hat{w}^{(1)} = A \sinh k(z + h_1) \quad (7.72)$$

$$\hat{w}^{(2)} = B \sinh k(z - h_2) \quad (7.73)$$

$$\hat{E}_x^{(1)} = C \sinh k(z + h_1) \quad (7.74)$$

$$\hat{E}_x^{(2)} = D \sinh k(z - h_2) \quad (7.75)$$

Substituting Eqs. (7.72)-(7.75) into Eqs. (7.68)-(7.71), the expression of n is:

$$n = \frac{k^2}{m} (T_1 - T_2 - T_3) \quad (7.76)$$

where

$$T_1 = \frac{2(\mu^{(1)} \coth kh_1 + \mu^{(2)} \coth kh_2)}{(\rho^{(1)} \coth kh_1 + \rho^{(2)} \coth kh_2)} \quad (7.77)$$

$$T_2 = \frac{\sigma k}{\rho^{(1)} \coth kh_1 + \rho^{(2)} \coth kh_2} \quad (7.78)$$

$$T_3 = - \frac{k_x^2 \cdot (\varepsilon^{(1)} E_{x0}^{(1)} - \varepsilon^{(2)} E_{x0}^{(2)})}{(\rho^{(1)} \coth kh_1 + \rho^{(2)} \coth kh_2) \cdot k^2 \cdot (\varepsilon^{(1)} \coth kh_1 + \varepsilon^{(2)} \coth kh_2)} \\ - \rho_e^s \cdot ik_x \frac{(\coth kh_1 + \coth kh_2) \cdot (\varepsilon^{(1)} E_{x0}^{(1)} - \varepsilon^{(2)} E_{x0}^{(2)})}{k^2 \cdot (\rho^{(1)} \coth kh_1 + \rho^{(2)} \coth kh_2) \cdot (\varepsilon^{(1)} \coth kh_1 + \varepsilon^{(2)} \coth kh_2)} \quad (7.79)$$

$$\frac{1}{m} = \frac{2(\mu^{(1)} - \mu^{(2)})}{\left[\left((\varepsilon^{(1)} E_{x0}^{(1)2} - \varepsilon^{(2)} E_{x0}^{(2)2}) \cdot \frac{k_x^2}{k^2} - (P_1 - P_2) \right) - \frac{(k^2 - \mu^{(1)}/\mu^{(2)})^2}{h_1/h_2} \right.} \\ \left. - \frac{k_x^2 \cdot (\varepsilon^{(1)} E_{x0}^{(1)} - \varepsilon^{(2)} E_{x0}^{(2)}) \cdot (\varepsilon^{(1)} E_{x0}^{(1)} \cdot \coth kh_1 + \varepsilon^{(2)} E_{x0}^{(2)} \cdot \coth kh_2)}{k^2 \cdot (\varepsilon^{(1)} \coth kh_1 + \varepsilon^{(2)} \coth kh_2)} \right] \quad (7.80)$$

$$k^2 = k_x^2 + k_y^2 \quad (7.81)$$

Term T_1 represents the effect of viscosity mismatch, term T_2 represents the effect of surface tension force, term T_3 represents the effect of the electric field. For a given set of

stability parameters, the temporal evolution of each disturbance mode is governed by the sign of n . $n = 0$, is the critical point of exchange of instability (marginal stable).

7.3 Materials and Methods

Fig. 7.2 shows the fabrication of the microchannel used in the experiments. The adhesive lamination technique [170] was used to fabricate the microchannel. The microchannel includes two polymethylmethacrylate (PMMA) plates ($40 \text{ mm} \times 50 \text{ mm}$) and multiple layers of double-sided adhesive tape. A microchannel with a cross section of $1 \text{ mm} \times 50 \text{ }\mu\text{m}$ was fabricated. The experimental set up is similar to that described in section 4.3.2.

The flows at the inlets I_1 and I_2 are driven by syringe pumps. The outlets O_1 and O_2 are connected to a waste reservoir. A high voltage power supply (Model PS350, Stanford Research System, Inc) is used to provide the controlling electric field. The power supply is capable of producing up to 5000 V and changing the polarity of the voltage. The controlling electric field is added to the fluids using platinum wires. The positive electrode is inserted into the inlet I_1 , and the negative electrode is inserted into the outlet O_1 directly. The applied electric field is parallel to the unperturbed interface.

Aqueous NaHCO_3 (10^{-7} M) is used as the conducting fluid. Rhodamine B ($\text{C}_{28}\text{H}_{31}\text{N}_2\text{O}_3\text{Cl}$) is added into the aqueous NaHCO_3 (0.1 g/ml) as the fluorescent dye to achieve a distinct interface with fluorescence microscopy. Surfactant of Span 20 is mixed with the aqueous NaHCO_3 (w/w is 2%) to reduce the surface tension. The viscosity and conductivity of NaHCO_3 are $0.85 \times 10^{-3} \text{ Ns/m}^2$ and $86.6 \text{ }\mu\text{S/cm}$, respectively. The viscosity of aqueous NaHCO_3 can be modified by mixing with glycerol (Sigma-Aldrich).

Different types of silicone oil (Sigma-Aldrich) are used as non-conducting fluid; the surfactant of Span 80 is added to the fluid (w/w is 0.2%) to reduce the surface tension. The conductivity of silicone oil is $0.064 \text{ }\mu\text{S/cm}$; their viscosities are 1.0 cSt, 5 cSt, and 10cSt, respectively.

7.4 Validation of the analytical model

For pressure-driven two-immiscible-fluid flow in microchannels without the application of electric field and surface tension, the growth rate of Eq. (7.76) can be reduced into

$$n = k^2 \frac{2\mu}{\rho \cdot m} \quad (7.82)$$

where

$$\frac{1}{m} = \frac{2(\mu^{(2)} - \mu^{(1)})}{\left[(P_1 - P_2) + \frac{(k^2 - \mu^{(1)}/\mu^{(2)})^2}{h_1/h_2} \right]} \quad (7.83)$$

A validation is performed by running cases using parameters reported by Khomami and Su [197]. The flow parameters are: $\mu^{(1)}/\mu^{(2)} = 0.203$, $h_1/h_2 = 4.875$. Fig. 7.3 compares the theoretically calculated growth rate n in Eq. (7.82) and experimentally measured by Khomami and Su [197], and the updated results by Khomami [198].

The results show that the proposed model can reasonably well predict the flow instability under the influence of viscosity.

7.5 Results and discussion

7.5.1 Phenomenon of instability

Fig. 7.4 shows the observed flow pattern at various values of electric field. Two immiscible fluids, aqueous NaHCO_3 (conducting fluid, $\mu^{(1)} = 0.85 \times 10^{-3} \text{ Ns/m}^2$) and silicone oil (non-conducting fluid, $\mu^{(2)} = 4.25 \times 10^{-3} \text{ Ns/m}^2$) are introduced into the microchannel at $q_1 = q_2 = 50 \text{ }\mu\text{l/h}$. The flow is stable when a positive electric field is added to the conducting fluid 1 (Fig. 7.4 (a)). When a negative electric field is added, we note a slightly unstable flow at -50 kV/m (Fig. 7.4 (c)). Unstable flow is observed at larger negative electric field (Fig. 7.4 (d), Fig. 7.4 (e)).

Fig. 7.5 shows that the growth rate depends on the relative magnitude of the relation in Eq. (7.76). The term of viscosity mismatch $\frac{(\mu^{(1)} \coth kh_1 + \mu^{(2)} \coth kh_2)}{(\rho^{(1)} \coth kh_1 + \rho^{(2)} \coth kh_2)}$ has a destabilizing effect as $T_1 > 0$ (Fig. 7.5). The viscosity mismatch induces a velocity mismatch at the perturbed interface which causes viscous instability [199].

The term of surface tension, $\frac{\sigma k}{(\rho^{(1)} \coth kh_1 + \rho^{(2)} \coth kh_2)}$, has a stabilizing effect as shown by a negative growth rate ($n < 0$) in Fig. 7.5.

The influence of the electric field and surface charge, which may either stabilize or destabilize the interface, depends on the sign of the term of $(\epsilon^{(1)} E_{x0}^{(1)} - \epsilon^{(2)} E_{x0}^{(2)})$. When this term becomes negative, the combined effect of electric field and surface charge becomes stabilizing and vice versa. The growth rate increases as the strength of the negative electric field increases.

The results clearly show that the orders of magnitude of the three terms are comparable. When the electric field reaches E_{critical} , the interface becomes perturbed; ripple wave arises at the interface. As the electric field increases, the destabilizing factor of the electric field increases.

7.5.2 Electrohydrodynamic instability of interface under the combined effect of electroosmosis and pressure gradient

Effect of the applied field

Fig. 7.6 shows the flow patterns for different values of electric field. The parameters of the experiments are $q_1 = q_2 = 0.05$ ml/h, $\epsilon_r^{(1)} = 80$, $w = 0.05$ mm, $\zeta_1 = \zeta_2 = -24.7$ mV, $\zeta_3 = -40.4$ mV, $\sigma = 1.8 \times 10^{-4}$ N/m, $\mu^{(2)} = 4.25 \times 10^{-3}$ Ns/m². The effect of electric field is displayed obviously in case 1 of Fig. 7.6 when $\mu^{(1)} = 0.85 \times 10^{-3}$ Ns/m². When a positive electric field is added to the conducting fluid 1, the width of the conducting fluid 1 decreases because of the electroosmotic effect [12, 13]. The interface is even and stable, the positive electric field cannot induce the instability of the interface under the range of

experimental conditions (Figs. 7.6 (1a-1c), (2a-2c), (3a-3c)). When a negative electric field is added, depending on the viscosity ratio and the magnitude of the applied electric field, instability occurs at a higher negative electric field. The results show that the negative electric field has a destabilizing effect.

In order to understand the effect of the parallel electric field on the instability of the interface, we fix the channel size, flowrates, electric properties and viscosity of fluid 2, and vary the electric field. Fig. 7.7 shows the growth rate n versus viscosity of conducting liquid 1 and the applied electric field. The flow conditions are $q_1 = q_2 = 0.05$ ml/h, $\varepsilon_r^{(1)} = 80$, $w = 0.05$ mm, $\xi_1 = \xi_2 = -24.7$ mV, $\xi_3 = -40.4$ mV, $\mu^{(2)} = 0.85 \times 10^{-3}$ Ns/m², $\sigma = 1.8 \times 10^{-4}$ N/m, and $\mu^{(1)}$ varying from 0.17×10^{-3} Ns/m² to 8.5×10^{-3} Ns/m². The width of fluid 1 and 2, (h_1 and h_2), and the pressure gradients, P_1 and P_2 , are determined from [13]. The results indicate that for a given channel geometry and $\mu^{(1)}$, the growth rate increases with increasing magnitude of the negative electric field. The negative electric field has a destabilizing effect to the flow while a positive electric field has a stabilizing effect. The analytical results are in agreement with the experimental results (Fig. 7.6).

Table 7.1 compares experimental results and analytical results for marginal stable conditions. The results show a discrepancy between experimental results and analytical results is about 30%. The reasons for discrepancy can be explained as follows. Except the effects of viscosity and electric field, the disturbance in the experiments also includes pressure perturbation, rough surface, heterogeneous properties of fluids and so on. These effects are not included in the analytical model. These undesired disturbances influence the instability and cannot be controlled. Furthermore, the marginal stable condition is difficult to determine accurately.

Effect of viscosity

Fig. 7.7 also shows the effect of viscosity on the growth rate. For a given channel geometry and applied electric field, the system is more stable with increasing viscosity of the conducting fluid 1. Comparing different viscosities (Figs. 7.6 (1f), (2f), (3f)), shows that the flow is unstable when $\mu^{(1)} = 0.85 \times 10^{-3}$ Ns/m², while the flow is stable when $\mu^{(1)} = 4.25 \times 10^{-3}$ Ns/m². Hence increasing the viscosity of the conducting fluid 1 has a stabilizing effect; this finding agrees well with Govindarajan [200].

The effect of the width (h) of the microchannel

Fig. 7.8 shows the effect of the width of the microchannel on growth rate over a range of $\mu^{(1)}$. We now fix the inlet flowrates ($q_1 = q_2 = 0.05$ ml/h), the applied electric field ($E_x = -150$ kV/m), the fluid properties ($\varepsilon_r^{(1)} = 80$, $\zeta_1 = \zeta_2 = -24.7$ mV, $\zeta_3 = 0$ mV, $\sigma = 1.8 \times 10^{-4}$ N/m, $\mu^{(2)} = 0.85 \times 10^{-3}$ Ns/m²), and vary the width of the microchannel from 50 μ m to 200 μ m. The results show that for a given inlet flow conditions and an applied electric field, the growth rate n decreases with increasing width of microchannel, thus the increase of the channel dimension has a stabilizing effect to the interface between the immiscible fluids. The reason is that the electroosmotic effect becomes weaker as width increases [13].

The effect of flowrates

Fig. 7.9 shows the effect of different flowrates on growth rate over a range of $\mu^{(1)}$. We now fix the applied electric field ($E_x = -150$ kV/m), the width of microchannel ($w = 50$ μ m), viscosity of fluid 2, $\mu^{(2)} = 0.85 \times 10^{-3}$ Ns/m², and the fluid properties ($\varepsilon_r^{(1)} = 80$, $\zeta_1 = \zeta_2 = 24.7$ mV, $\zeta_3 = 0$ mV, $\sigma = 1.8 \times 10^{-4}$ N/m), and vary the flowrates from 0.05 ml/h to 0.2 ml/h. The results show that for a given applied electric field and channel geometry, the growth rate n decreases with the increasing of the inlet flowrates, thus the increase of flowrates has a stabilizing effect to the interface between the immiscible fluids. The reason is that the inertia effect becomes higher with increasing flowrate [13].

7.6 Summary

This chapter studies the electrohydrodynamic and shear-stress instability of the interface between immiscible fluids under the combined effect of hydrodynamics and electroosmosis in a microchannel. The effect of parameters, e.g. electric field, viscosity, flowrate, and dimension of the channel are studied using an analytical model and experiments. The results show that negative electric field has a destabilizing effect to the interface between immiscible fluids. At the same time, the interface is more stable with increasing viscosity of conducting fluid. The effect of flowrates and size of channel are

dependent on the electroosmotic effect. Increasing flowrates and size of channel are stabilizing effects for the interface because the electroosmotic effect becomes insignificant.

Table 7.1 Comparing results between analytical results and experimental results

($q_1 = q_2 = 0.05$ ml/h, $\varepsilon_r^{(1)} = 80$, $w = 0.05$ mm, $\zeta_1 = \zeta_2 = -24.7$ mV, $\zeta_3 = -40.4$ mV, $\sigma = 1.8 \times 10^{-4}$ N/m, $\mu^{(2)} = 4.25 \times 10^{-3}$ Ns/m²)

	Conditions	Electric fields of marginal stable (v/m)	errors
Experiment (1)	$\mu^{(1)} = 0.85 \times 10^{-3}$ Ns/m ²	-50000	39%
Analytical model (1)		-82100	
Experiment (2)	$\mu^{(1)} = 1.7 \times 10^{-3}$ Ns/m ²	-130000	32.8%
Analytical model (2)		-193300

Fig. 7.1 (a) Two-fluid instabilities under the combined effect of hydrodynamic and electroosmosis; (b) Schematic of the phenomenon and coordinate systems

Fig. 7.2 The structure of the microchannel

Fig. 7.3 A comparison of theoretically calculated growth rate and experimentally measured by Khomami and Su [197], and new results by Khomami [198]

(a) Stable

(b) Stable

(c) Marginal stable

(d) Unstable

(e) Unstable

Fig. 7.4 The observed flow pattern at various values of electric field

Fig. 7.5 Relative magnitude of the relation in Eq. (7.76)

E_x (kV/m)	Case (1)	Case (2)	Case (3)
	$\mu^{(1)} = 0.85 \times 10^{-3} \text{ Ns/m}^2$	$\mu^{(1)} = 4.25 \times 10^{-3} \text{ Ns/m}^2$	$\mu^{(1)} = 8.5 \times 10^{-3} \text{ Ns/m}^2$
150
	1(a)	2(a)	3(a)
50
	1(b)	2(b)	3(b)
0
	1(c)	2(c)	3(c)
-50
	1(d)	2(d)	3(d)
-150
	1(e)	2(e)	3(e)
-200
	1(f)	2(f)	3(f)

Fig. 7.6 The observed flow pattern at various values of electric field and viscosity of fluid
 $1 \mu^{(1)}$ ($q_1 = q_2 = 0.05 \text{ ml/h}$, $\epsilon_r^{(1)} = 80$, $w = 0.05 \text{ mm}$, $\xi_1 = \xi_2 = -24.7 \text{ mV}$, $\xi_3 = -40.4 \text{ mV}$, $\sigma = 1.8 \times 10^{-4} \text{ N/m}$, $\mu^{(2)} = 4.25 \times 10^{-3} \text{ Ns/m}^2$)

Fig. 7.7 Effect of the applied voltage on the growth rate over a range of viscosity of fluid 1. The input parameters are ($q_1 = q_2 = 0.05$ ml/h, $\varepsilon_r^{(1)} = 80$, $w = 0.05$ mm, $\xi_1 = \xi_2 = -24.7$ mV, $\xi_3 = 0$ mV, $\sigma = 1.8 \times 10^{-4}$ N/m, $\mu^{(2)} = 0.85 \times 10^{-3}$ Ns/m²)

Fig. 7.8 Effect of the width of microchannel on the growth rate over a range of viscosity of fluid 1 ($q_1 = q_2 = 0.05$ ml/h, $\varepsilon_r^{(1)} = 80$, $\xi_1 = \xi_2 = -24.7$ mV, $\xi_3 = 0$ mV, $\sigma = 1.8 \times 10^{-4}$ N/m, $\mu^{(2)} = 0.85 \times 10^{-3}$ Ns/m², $E_x = -150$ kV/m)

Fig. 7.9 Effect of different flowrate on the growth rate over a range of viscosity of fluid 1 ($q_1 = q_2$, $\varepsilon_r^{(1)} = 80$, $\xi_1 = \xi_2 = -24.7 \text{ mV}$, $w = 0.05 \text{ mm}$, $\xi_3 = 0 \text{ mV}$, $\sigma = 1.8 \times 10^{-4} \text{ N/m}$, $\mu^{(2)} = 0.85 \times 10^{-3} \text{ Ns/m}^2$, $E_x = -150 \text{ kV/m}$)

Chapter 8 Conclusions and future work

8.1 Concluding remarks

Theoretical models of the pressure-driven three immiscible fluids in rectangular microchannels with electroosmotic effect, including steady state model and transient model, are presented. By considering the electroosmotic force as the body force in the conducting liquids, analytical solutions were obtained by solving the linear Poisson-Boltzmann equation and the Navier-Stokes equation together. Due to the presence of the surface free charge densities at the liquid-liquid interfaces, the bulk electroosmotic forces in the diffuse layers and the forces exerted at the interfaces are accounted in this model. The velocity profiles and the flowrates for two-dimensional three-liquid flow are obtained analytically by the methods of Laplace transformation method and separation of variable. The results indicate that pressure gradients, external electric field, and properties of fluids influence the velocity profiles and flowrates. The computational results also show that the three-liquid electroosmotic pump is feasible for the relative small interface zeta potential and viscosity ratio β_2 . The details insight of the flow characteristics of this flow configuration are provided by the velocity profile as time evolution in the transient model.

Experimental results show that the width and location of the sample flow can be controlled accurately using the electroosmotic effect, rather than the conventional “flow-rate-ratio” method. Under the fixed flowrates, the sample flow can be delivered to the desired outlet ports by electroosmosis. The electroosmotic effect under different flowrates, different viscosity of the non-conducting fluid, and different aspect ratio were investigated. The results indicate that the electroosmotic effect on the pressure-driven flow becomes weaker with the increase of the flowrates, and viscosity ratio β_2 ; the sample flow or droplets can be delivered to the desired outlets.

The comparison between the measured data (width of the non-conducting fluid and location of the sample stream) and the analytical models show good agreement.

An analytical model of a liquid-core liquid-cladding optofluidic lens tunable by the combined effect of hydrodynamic and electroosmosis are presented. In this lens, an electrically conducting liquid is used as the cladding fluids, and the core fluid is non-conducting. Voltages are added to the cladding fluids. Under the constant flowrates, the theory of two-dimensional dipole flow in a circularly bounded domain and the theory of the electric field outside the parallel plate capacitor are used to predict the curvature of the interface. The proposed analytical model is validated by experimental results. The results show that the interfaces between the cladding fluids and the core fluid are optically smooth arc-shape under the combined effect of hydrodynamic and electroosmosis. Under fixed cladding flowrates, the symmetric voltages form symmetric biconvex lens only, while asymmetric voltages form biconvex lens, plano-convex lens, and meniscus lens. The experimental results agree well with the analytical model.

A semi analytical model of the pressure driven two-fluid flow in rectangular microchannels with electroosmotic effect is developed. Different from the previous works, the curved interfaces between the immiscible fluids are used to replace the assumed planar interface.

The validity of the semi-analytical model with curved interfaces has been checked by comparing with the solution obtained with the results available in the literatures.

The results show that the interface shape influences the characteristics of the flow significantly. Especially in the sharp corner between interface and the wall, the electric potential is higher. Comparing with the planar and convex interfaces, the concave interface has the highest electric potential and electroosmotic velocity. Under the same liquid fractions, the maximum volumetric electroosmotic flowrate of the conducting fluid with the concave interface is about 1.5 times larger than that of planar interface.

A model is developed for the electrohydrodynamic and shear-stress instability of the interface between immiscible fluids under the combined effect of hydrodynamic and electroosmosis in a microchannel. The effect of parameters, e.g. electric field, viscosity, flowrate, and dimension of the channel are studied using an analytical model and experiments.

In the analytical analysis, the electric field and fluid dynamics are coupled at the interface through the tangential and normal interfacial stress balance equations under the

coupled effect of hydrodynamic and electroosmosis. In the experiments, two immiscible fluids, Aqueous NaHCO_3 (conducting fluid) and silicone oil (non-conducting fluid) are introduced into the PMMA microchannel using a syringe pump. The parallel electric field is added to the aqueous NaHCO_3 using a high voltage power supply. The results show that negative electric fields have a destabilizing effect to the interface between immiscible fluids. At the same time, the interface is more stable with increasing viscosity of the conducting fluid. The effects of flowrates and size of channel are dependent on the electroosmotic effect. Increasing flowrates and size of channel are stabilizing effects for the interface because the electroosmotic effect becomes insignificant.

8.2 Recommendations for future studies

Having performed the above investigations, some recommendations are made as following for future research in order to achieve better understanding of the electrokinetic phenomenon in multi-fluid flow.

Currently, our analytical model is only used to analyse the interface location at the symmetrical line of the circular chamber. The depth average method is used in the analytical model. It is suggested that by developing a three-dimensional numerical program for the simulation of three immiscible fluids in circular chamber or more complex channels, the numerical results would give the electric field and velocity profiles in the whole channel. Thus, the numerical program is very useful to show the details of flow conditions.

Micro lenses are useful in many applications and are integrated into lab-on-a-chip systems. It is suggested that future studies can examine the optical characteristics of this tunable lens by the combined effect of hydrodynamic and electroosmosis. Fluids with different reflection index are to be used to confirm the characterizations of optofluidic lens. The relationship between the external electric field and the properties of optofluidic lens would then be determined. Therefore the micro lenses can be integrated into lab-on-chip systems to confirm the characterizations.

Electrokinetic instability phenomenon can enhance the micro mixing. The model in the thesis can be further developed to study the effect of normal electric fields on two immiscible fluids under the coupled effect of pressure-driven and electroosmosis. In the

experiment, the stabilizing or destabilizing effects of the external electric field, geometry of microchannel, free surface charge, and properties of fluids can be investigated. The relationship between the flow pattern and external electric fields can be established.

Appendix A

The least-squares method for numerical calculation is introduced in this appendix.

Eqs. (6.6), (6.19), and (6.36) can be described using matrix form as follows

$$\mathbf{MB} = \mathbf{Z} \quad (\text{A.84})$$

In Eq. (A.84), $\mathbf{M}$ is the matrix for coefficients, $\mathbf{B}$ is the matrix for unknown parameters, $\mathbf{Z}$ is the matrix for boundaries. The parameter of $\mathbf{B}$ can be calculated numerically using the least-squares method that minimizes of $\|\mathbf{MB} - \mathbf{Z}\|$ to find the optimal values of $\mathbf{B}$. The details of $\mathbf{M}$, $\mathbf{B}$, and $\mathbf{Z}$ for Eqs. (6.6), (6.19), and (6.36) are shown in Appendix B, Appendix C, and Appendix D respectively.

Appendix B

The details of $\mathbf{M}$, $\mathbf{B}$, and $\mathbf{Z}$ for Eqs. (6.6) are as follows:

$$\mathbf{M} = \begin{pmatrix} I_0(Kr_1), & & I_J(Kr_1)\cos(J \cdot \theta_1), I_J(Kr_1)\sin(J \cdot \theta_1) \\ I_0(Kr_2), & \cdots & I_J(Kr_2)\cos(J \cdot \theta_2), I_J(Kr_2)\sin(J \cdot \theta_2) \\ \vdots & \ddots & \vdots \\ I_0(Kr_{N-1}), & \cdots & I_J(Kr_{N-1})\cos(J \cdot \theta_{N-1}), I_J(Kr_{N-1})\sin(J \cdot \theta_{N-1}) \\ I_0(Kr_N), & & I_J(Kr_N)\cos(J \cdot \theta_N), I_J(Kr_N)\sin(J \cdot \theta_N) \end{pmatrix}_{N \times (2J+1)} \quad (\text{A.85})$$

$$\mathbf{B} = \begin{pmatrix} A_{10} \\ \vdots \\ A_{1J} \\ A_{2J} \end{pmatrix}_{(2J+1) \times 1} \quad (\text{A.86})$$

$$\mathbf{Z} = \begin{pmatrix} \bar{\psi}(r_1, \theta_1) \\ \bar{\psi}(r_2, \theta_2) \\ \vdots \\ \bar{\psi}(r_{N-1}, \theta_{N-1}) \\ \bar{\psi}(r_N, \theta_N) \end{pmatrix}_{N \times 1} \quad (\text{A.87})$$

where N is the total number of the boundary conditions and J is the highest order of the Bessel functions chosen in this report.

Appendix C

The details of $\mathbf{M}$, $\mathbf{B}$, and $\mathbf{Z}$ for Eqs. (6.19) are as follows:

$$\mathbf{M} = \begin{pmatrix} \mathbf{M}_{11} \\ \mathbf{M}_{22} \\ \mathbf{M}_{33} \end{pmatrix} \quad (\text{A.88})$$

$$\mathbf{M}_{11} = \begin{pmatrix} 1, 0, r_1^1 \cos(\theta_1), r_1^1 \sin(\theta_1), & & r_1^J \cos(J \cdot \theta_1), r_1^J \sin(J \cdot \theta_1), 0, 0 \\ 1, 0, r_2^1 \cos(\theta_2), r_2^1 \sin(\theta_2), & \cdots & r_2^J \cos(J \cdot \theta_2), r_2^J \sin(J \cdot \theta_2), 0, 0 \\ \vdots & \ddots & \vdots \\ 1, 0, r_{N_1-1}^1 \cos(\theta_{N_1-1}), r_{N_1-1}^1 \sin(\theta_{N_1-1}), & \cdots & r_{N_1-1}^J \cos(J \cdot \theta_{N_1-1}), r_{N_1-1}^J \sin(J \cdot \theta_{N_1-1}), 0, 0 \\ 1, 0, r_{N_1}^1 \cos(\theta_{N_1}), r_{N_1}^1 \sin(\theta_{N_1}), & \cdots & r_{N_1}^J \cos(J \cdot \theta_{N_1}), r_{N_1}^J \sin(J \cdot \theta_{N_1}), 0, 0 \end{pmatrix}_{N_1 \times (4J+2)} \quad (\text{A.89})$$

$$\mathbf{M}_{22} = \begin{pmatrix} 0, 1, 0, 0, r_1^1 \cos(\theta_1), r_1^1 \sin(\theta_1), & & r_1^J \cos(J \cdot \theta_1), r_1^J \sin(J \cdot \theta_1) \\ 0, 1, 0, 0, r_2^1 \cos(\theta_2), r_2^1 \sin(\theta_2), & \cdots & r_2^J \cos(J \cdot \theta_2), r_2^J \sin(J \cdot \theta_2) \\ \vdots & \ddots & \vdots \\ 0, 1, 0, 0, r_{N_2-1}^1 \cos(\theta_{N_2-1}), r_{N_2-1}^1 \sin(\theta_{N_2-1}), & \cdots & r_{N_2-1}^J \cos(J \cdot \theta_{N_2-1}), r_{N_2-1}^J \sin(J \cdot \theta_{N_2-1}) \\ 0, 1, 0, 0, r_{N_2}^1 \cos(\theta_{N_2}), r_{N_2}^1 \sin(\theta_{N_2}), & \cdots & r_{N_2}^J \cos(J \cdot \theta_{N_2}), r_{N_2}^J \sin(J \cdot \theta_{N_2}) \end{pmatrix}_{N_2 \times (4J+2)} \quad (\text{A.90})$$

$$\mathbf{M}_{33} = \begin{pmatrix} \beta, -1, [\beta r_1^1 \cos(\theta_1) + C_1(1)], & & [-r_1^J \cos(J \cdot \theta_1) - C_1(J)], [-r_1^J \sin(J \cdot \theta_1) - S_1(1)] \\ \beta, -1, [\beta r_2^1 \cos(\theta_2) + C_1(1)], & \cdots & [-r_2^J \cos(J \cdot \theta_2) - C_1(J)], [-r_2^J \sin(J \cdot \theta_2) - S_1(1)] \\ \vdots & \ddots & \vdots \\ \beta, -1, [\beta r_{N_3-1}^1 \cos(\theta_{N_3-1}) + C_1(1)], & \cdots & [-r_{N_3-1}^J \cos(J \cdot \theta_{N_3-1}) - C_1(J)], [-r_{N_3-1}^J \sin(J \cdot \theta_{N_3-1}) - S_1(1)] \\ \beta, -1, [\beta r_{N_3}^1 \cos(\theta_{N_3}) + C_1(1)], & \cdots & [-r_{N_3}^J \cos(J \cdot \theta_{N_3}) - C_1(J)], [-r_{N_3}^J \sin(J \cdot \theta_{N_3}) - S_1(1)] \end{pmatrix}_{N_3 \times (4J+2)} \quad (\text{A.91})$$

$$\mathbf{B} = \begin{pmatrix} \mathbf{B}_{11} \\ \mathbf{B}_{21} \\ \mathbf{B}_{31} \end{pmatrix} \quad (\text{A.92})$$

$$\mathbf{B}_{11} = \mathbf{B}_{22} = \mathbf{B}_{33} = \begin{pmatrix} a_{10}^p \\ a_{20}^p \\ \vdots \\ a_{2J}^p \\ b_{2J}^p \end{pmatrix}_{(4J+2) \times 1} \quad (\text{A.93})$$

$$\mathbf{Z} = \begin{pmatrix} \mathbf{Z}_{11} \\ \mathbf{Z}_{22} \\ \mathbf{Z}_{33} \end{pmatrix} \quad (\text{A.94})$$

$$\mathbf{Z}_{11} = \begin{pmatrix} \frac{r_1^2}{4} \\ \vdots \\ \frac{r_{N_1}^2}{4} \end{pmatrix}_{N_1 \times 1} \quad (\text{A.95})$$

$$\mathbf{Z}_{22} = \begin{pmatrix} \frac{r_1^2}{4} \\ \vdots \\ \frac{r_{N_2}^2}{4} \end{pmatrix}_{N_2 \times 1} \quad (\text{A.96})$$

$$\mathbf{Z}_{33} = \begin{pmatrix} (\beta-1) \frac{r_1^2}{4} \\ \vdots \\ (\beta-1) \frac{r_{N_3}^2}{4} \end{pmatrix}_{N_3 \times 1} \quad (\text{A.97})$$

where N_1 is the total number of the boundary conditions for fluid 1, N_2 is the total number of the boundary conditions for fluid 2, N_3 is the total number of the matching conditions at the interface, $N = N_1 + N_2 + N_3$, and J is the highest order of the Bessel functions chosen in this report.

Appendix D

The details of $\mathbf{M}$, $\mathbf{B}$, and $\mathbf{Z}$ for Eqs. (6.36) are as follows:

$$\mathbf{M} = \begin{pmatrix} \mathbf{M}_{11} \\ \mathbf{M}_{22} \\ \mathbf{M}_{33} \end{pmatrix} \quad (\text{A.98})$$

$$\mathbf{M}_{11} = \begin{pmatrix} 1, 0, r_1^1 \cos(\theta_1), r_1^1 \sin(\theta_1), & & r_1^J \cos(J \cdot \theta_1), r_1^J \sin(J \cdot \theta_1), 0, 0 \\ 1, 0, r_2^1 \cos(\theta_2), r_2^1 \sin(\theta_2), & \cdots & r_2^J \cos(J \cdot \theta_2), r_2^J \sin(J \cdot \theta_2), 0, 0 \\ \vdots & \ddots & \vdots \\ 1, 0, r_{N_1-1}^1 \cos(\theta_{N_1-1}), r_{N_1-1}^1 \sin(\theta_{N_1-1}), & \cdots & r_{N_1-1}^J \cos(J \cdot \theta_{N_1-1}), r_{N_1-1}^J \sin(J \cdot \theta_{N_1-1}), 0, 0 \\ 1, 0, r_{N_1}^1 \cos(\theta_{N_1}), r_{N_1}^1 \sin(\theta_{N_1}), & \cdots & r_{N_1}^J \cos(J \cdot \theta_{N_1}), r_{N_1}^J \sin(J \cdot \theta_{N_1}), 0, 0 \end{pmatrix}_{N_1 \times (4J+2)} \quad (\text{A.99})$$

$$\mathbf{M}_{22} = \begin{pmatrix} 0, 1, 0, 0, r_1^1 \cos(\theta_1), r_1^1 \sin(\theta_1), & & r_1^J \cos(J \cdot \theta_1), r_1^J \sin(J \cdot \theta_1) \\ 0, 1, 0, 0, r_2^1 \cos(\theta_2), r_2^1 \sin(\theta_2), & \cdots & r_2^J \cos(J \cdot \theta_2), r_2^J \sin(J \cdot \theta_2) \\ \vdots & \ddots & \vdots \\ 0, 1, 0, 0, r_{N_2-1}^1 \cos(\theta_{N_2-1}), r_{N_2-1}^1 \sin(\theta_{N_2-1}), & \cdots & r_{N_2-1}^J \cos(J \cdot \theta_{N_2-1}), r_{N_2-1}^J \sin(J \cdot \theta_{N_2-1}) \\ 0, 1, 0, 0, r_{N_2}^1 \cos(\theta_{N_2}), r_{N_2}^1 \sin(\theta_{N_2}), & \cdots & r_{N_2}^J \cos(J \cdot \theta_{N_2}), r_{N_2}^J \sin(J \cdot \theta_{N_2}) \end{pmatrix}_{N_2 \times (4J+2)} \quad (\text{A.100})$$

$$\mathbf{M}_{33} = \begin{pmatrix} 1, -1, [r_1^1 \cos(\theta_1) + C_1^E(1)], & & [-r_1^J \sin(J \cdot \theta_1) - \beta S_2^E(J)] \\ 1, -1, [r_2^1 \cos(\theta_2) + C_1^E(1)], & \cdots & [-r_2^J \sin(J \cdot \theta_2) - \beta S_2^E(J)] \\ \vdots & \ddots & \vdots \\ 1, -1, [r_{N_3-1}^1 \cos(\theta_{N_3-1}) + C_1^E(1)], & \cdots & [-r_{N_3-1}^J \sin(J \cdot \theta_{N_3-1}) - \beta S_2^E(J)] \\ 1, -1, [r_{N_3}^1 \cos(\theta_{N_3}) + C_1^E(1)], & \cdots & [-r_{N_3}^J \sin(J \cdot \theta_{N_3}) - \beta S_2^E(J)] \end{pmatrix}_{N_3 \times (4J+2)} \quad (\text{A.101})$$

$$\mathbf{B} = \begin{pmatrix} \mathbf{B}_{11} \\ \mathbf{B}_{21} \\ \mathbf{B}_{31} \end{pmatrix} \quad (\text{A.102})$$

$$\mathbf{B}_{11} = \mathbf{B}_{22} = \mathbf{B}_{33} = \begin{pmatrix} a_{10}^E \\ a_{20}^E \\ \vdots \\ a_{1J}^E \\ B_{2J}^E \end{pmatrix}_{(4J+2) \times 1} \quad (\text{A.103})$$

$$\mathbf{Z} = \begin{pmatrix} \mathbf{Z}_{11} \\ \mathbf{Z}_{22} \\ \mathbf{Z}_{33} \end{pmatrix} \quad (\text{A.104})$$

$$\mathbf{Z}_{11} = \begin{pmatrix} -\frac{\text{Re} \cdot \bar{E}}{\bar{\mu}} \cdot \bar{\psi}(r_1, \theta_1) \\ \vdots \\ -\frac{\text{Re} \cdot \bar{E}}{\bar{\mu}} \cdot \bar{\psi}(r_{N_1}, \theta_{N_1}) \end{pmatrix}_{N_1 \times 1} \quad (\text{A.105})$$

$$\mathbf{Z}_{22} = \begin{pmatrix} 0 \\ \vdots \\ 0 \end{pmatrix}_{N_2 \times 1} \quad (\text{A.106})$$

$$\mathbf{Z}_{33} = \begin{pmatrix} -\frac{\text{Re} \cdot \bar{E}}{\bar{\mu}} \cdot \bar{\psi}(r_1, \theta_1) - M \bar{\rho}_q^s - \frac{\text{Re} \cdot \bar{E}}{\bar{\mu}_1} \cdot \frac{\partial \bar{\psi}(r_1, \theta_1)}{\partial n} \\ \vdots \\ -\frac{\text{Re} \cdot \bar{E}}{\bar{\mu}} \cdot \bar{\psi}(r_{N_3}, \theta_{N_3}) - M \bar{\rho}_q^s - \frac{\text{Re} \cdot \bar{E}}{\bar{\mu}_1} \cdot \frac{\partial \bar{\psi}(r_{N_3}, \theta_{N_3})}{\partial n} \end{pmatrix}_{N_3 \times 1} \quad (\text{A.107})$$

where N_1 is the total number of the boundary conditions for fluid1, N_2 is the total number of the boundary conditions for fluid2, N_3 is the total number of the matching conditions for interface, $N = N_1 + N_2 + N_3$, and J is the highest order of the Bessel functions chosen in this report.

Publications arising from this thesis

[1] **Haiwang Li**, T.N. Wong, N.T. Nguyen, Time-dependent model of mixed electroosmotic/pressure-driven three immiscible fluids in a rectangular microchannel, *International Journal of Heat and Mass Transfer*, 53 (2010) 772-785.

[2] **Haiwang Li**, T.N. Wong, N.T. Nguyen, Analytical model of mixed electroosmotic/pressure driven three immiscible fluids in a rectangular microchannel, *International Journal of Heat and Mass Transfer*, 52 (2009) 4459-4469.

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[4] **Haiwang Li**, T.N. Wong, N.T. Nguyen, Microfluidic switch based on combined effect of hydrodynamic and electroosmosis, *Microfluidic and Nanofluidics*, 10 (5) (2011) 965-976.

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[6] **Haiwang Li**, T.N. Wong, N.T. Nguyen, Tunable micro optofluidic lens based on combined effect of hydrodynamic and electroosmosis, *Microfluidic and Nanofluidics* 10 (5) (2011) 1033-1043.

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