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Joule heating and induced charge effects on electroosmotic entry flows in microfluidic devices

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JOULE HEATING AND INDUCED CHARGE EFFECTS ON ELECTROOSMOTIC ENTRY FLOWS IN MICROFLUIDIC DEVICES

A Thesis
Presented to
the Graduate School of
Clemson University

In Partial Fulfillment
of the Requirements for the Degree
Master of Science
Mechanical Engineering

by
Rama Aravind Prabhakaran
August 2016

Accepted by:
Dr. Xiangchun Xuan, Committee Chair
Dr. Phanindra Tallapragada
Dr. Gang Li
ABSTRACT

Microfluidic devices have become a powerful tool for chemical and biological applications in the past two decades due to their several advantages over the conventional benchtop counterparts such as smaller size, reduced reagent consumption and higher efficiency. Manipulations of samples and electrolytes, for instance, sorting, trapping, and mixing etc., are important in these applications. Electrokinetics is an efficient method for performing these manipulations by the application of electric field. It is the preferred method of sample transport over the traditional pressure driven flow due to the essentially plug-like velocity profile that has smaller dispersion effects. However, the majority of the previous works on electrokinetic phenomena is limited to inside the microchannel. Very few studies have been reported on electroosmotic fluid entry from reservoir to microchannel. Due to the mismatch in sizes of the macro-scale reservoir and the micro-scale channel, several electrokinetic phenomena such as dielectrophoresis (DEP), Joule heating effects and induced charge electroosmosis (ICEO) are prominent at their interface. This thesis tries to provide a fundamental insight on the effects of Joule heating and ICEO on electroosmotic entry flows through experimental and numerical studies.

Joule heating is inevitable in an electrokinetic system especially when the solution’s electrical conductivity is high and/or a low-thermal-conductivity material such as polydimethylsiloxane (PDMS) is used for manufacturing the device. In such cases the electrolyte temperature increases which in turn affects the temperature dependent fluid properties. This leads to the formation of electrothermal flows which under certain
conditions can disrupt the flow. We performed an experimental and numerical study of Joule heating effects on electroosmotic flow at the entrance of the microchannel. We observed that two counter rotating circulations can be formed under appropriate conditions. We also did a parametric study and found that the vortex becomes stronger with increasing AC to DC voltage ratio. Moreover we developed a two dimensional depth-averaged numerical model which is able to predict the Joule heating and its effects on electrokinetic phenomenon with reasonable accuracy. This model does not require the high memory or processing requirement as a three dimensional model, nor does it have any unrealistic assumptions as the previous two dimensional model.

ICEO is due to the leakage of electric field through the substrate around the microchannel. It is strong in low-conductivity solutions wherein Joule heating effects can be neglected. We conducted a combined experimental and numerical study of ICEO effects on electroosmotic entry flow in such a situation. We found that, similar to electrothermal flows, ICEO has two counter rotating circulations but are rotating in opposite directions. We also pointed out that a regular two dimensional model cannot accurately predict the flow field in this phenomenon. We therefore developed a depth averaged two-dimensional model, which predicted the experimentally obtained flow field with a good agreement. This numerical model was used to explain the formation of induced zeta potential on the channel surface due to electric field leakage. The effects of ICEO on fluid flow and particle motion at the reservoir-microchannel junction are discussed. Moreover, parametric study is done to further understand the effects by changing the AC to DC ratio, relative permittivity of polymer substrate and the channel width.
DEDICATION

I dedicate my thesis work to my family and many friends. I thank my mother and my father for their kind words of encouragement and love throughout my life. My brother has always believed in me and never left my side. I would also like to dedicate this work to my late grandfather who would have felt proud to see me earn a Master’s degree.
ACKNOWLEDGMENTS

I sincerely thank my advisor Dr. Xiangchun Xuan for providing me the opportunity to work in this field and guiding me throughout the graduate studies. The wonderful meetings I had with him has helped me to gain more technical knowledge than I could have learnt from anywhere else. I learned on how to lead a successful professional career from him. I also thank him and the Mechanical Engineering Department of Clemson University for the Assistantships provided to fund my living expenses. I thank my committee members Dr. Phanindra Tallapragada and Dr. Gang Li for their valuable inputs. I thank my professors from CIT and BVB for helping me to find my interests.

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I am grateful to all my family members, my cousins and their family who have always loved me and encouraged me to perform to my fullest.
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CHAPTER ONE
INTRODUCTION

1.1 Objective and Motivation

Microfluidics-based lab-on-a-chip (LOC) devices integrate several necessary laboratory components and functions onto small chips of few centimeters in size. The latest development in the photolithography techniques have enabled to manufacture these devices faster and at low costs [1]. These devices can be built on a thin glass plate with a network of microchannel, electrodes and circuits. Elastomer materials like polydimethylsiloxane (PDMS) have been found to be a better replacement to the traditional glass and silicon based devices. The significant advantages of LOC over the conventional benchtop devices include the minimized reagent consumption, smaller analysis times, higher throughput by parallel operation, safer for biological and chemical studies, better process control and lower manufacturing costs [2-4]. LOC devices has been applied to various fields such as clinical diagnostics of bacteria and virus, chemistry, biotechnology, food technology and water treatment [5-7].

Microfluidics is the transport and control of fluids and samples (e.g., ions, molecules, viruses and cells) in microchannels in LOC devices [8]. The manipulation of samples inside these devices has been achieved by trapping, focusing, sorting, filtering and mixing samples through several force fields such as magnetic [9,10], optical [11,12], acoustic [13,14], electrokinetic [15-17], hydrodynamic [18-20], and inertial [21] forces. Among these various methods, electrokinetics (DC [22] and/or AC [23,24]) is an efficient
technique and the preferred mode for transport of samples due to the smaller dispersion and a scope to integrate with other electronic sensors in microfluidic devices [25-28]. It transports fluids by electroosmosis and (charged) samples by electrophoresis through microchannels. To date electrokinetic transport phenomena have been investigated on a variety of microchannel structures including straight, curved and network of channels [29-31]. However, these studies have been almost entirely focused upon the fluid and sample transport inside the microchannels. Very little attention has been given to the fluid and sample entry from the inlet reservoir (i.e., the well that supplies liquids and samples) to the microchannel [32,33]. At this interface there exists a significant mismatch in the size of the macro scale reservoir (typically a few millimeters in diameter and depth) and the micro scale channel (typically tens of micrometers in width and depth). Strong electric field gradients are therefore inherently induced at the reservoir-microchannel junction [34], which have been recently demonstrated by our group to focus, trap and sort particles via reservoir-based dielectrophoresis (rDEP) [35,36] and/or induced charge electroosmosis [37].

Joule heating is an inevitable phenomenon accompanying electrokinetic fluid and sample transport in microfluidic devices [38]. It causes temperature rises and gradients in the fluid, and in turn disrupts the preferred plug like velocity profile. The PDMS used in microfluidic devices is not completely insulating. It has a very small but a non-negligible value of electrical permittivity. So the electric field leaks from the fluid and causes polarization of PDMS [39] and especially near the microchannel corners that significantly affects the zeta potential on these surfaces. This phenomenon causes the so called induced
charge electroosmosis (ICEO) [40] that can disrupt the electroosmotic fluid flow. This thesis attempts to provide a detailed experimental, computational and theoretical explanation of these two phenomena of Joule heating and ICEO on the electroosmotic entry flow at the interface of the reservoir and microchannel in microfluidic devices.

1.2 Background

Electrokinetics, as the name suggests, is the manipulation of fluids and samples with the application of electric field. As mentioned earlier electrokinetic flow has an essentially plug like velocity profile and so the velocity at any given cross section of the channel has the same value. So the concentration of samples is uniform over the entire width of channel. But in a pressure driven flow the parabolic velocity profile causes sample dispersion due to the gradients in velocity at the cross section of microchannel. Hence electrokinetics is the preferred mode of sample transport. The electric field is supplied by the application of voltage drop using external electrodes. A single voltage source can be used to power other electrical sensing devices. This section provides a brief background explanation of electrokinetic phenomenon such as electroosmosis, electrophoresis, dielectrophoresis, Joule heating and ICEO required for the investigation done in this thesis.

1.2.1 Electrical Double Layer (EDL)

Understanding electrokinetic flow lies within the concept of electrical double layer (EDL). The electrolyte used is always electrically neutral in the bulk of solution away from the surfaces surrounding it. But near a solid surface, the solid surface obtains a specific value of charge. This charge will attract the oppositely charged ions (counter ions) in electrolyte towards the surface and repels the like charged ions (co-ions) away from it.
Thus the higher concentration of counter ions causes a diffusive effect into the fluid away from the wall. Due to the electrostatic attraction and diffusion, an equilibrium concentration of counter ions is created normal to the wall. The concentration of ions is high near the walls slowly decreases as we move away from wall and thus the charge concentration reduces to zero at the bulk of fluid [22]. Due to this charge distribution, the equilibrium potential will be high near the wall and reduce to zero in the bulk of fluid. This region of two oppositely charged ions near the wall is called as the electrical double layer. Fig. 1.1 shows the region near walls and distribution of charges and electric potential.

**Fig. 1.1** Distribution of electrical potential and charges near the surface of walls. The electrical potential is plotted as a function of normal $n$ to the walls and the equilibrium zeta potential at the slip plane are shown.
1.2.2 Electroosmosis

Electroosmotic flow is the motion of electrolyte tangential to a charged surface under the application of external electric field. When the electric field is applied the charges in the electrolyte experience a force. In the bulk of the electrolyte the force is zero but this force is non zero inside the electrical double layer because of nonzero charge density. This causes the ions in this region to move towards the oppositely charged electrode. This motion drags the bulk of the fluid in this direction due to viscous drag. The Navier Stokes equation can be solved with the body force term equal to the Coulomb force, \( \rho_e \mathbf{E} \). The mathematics for this derivation can be referred elsewhere [23]. Due to the very small thickness of EDL, the electroosmotic flow is considered with negligible thickness of EDL with a slip velocity of,

\[
\mathbf{u}_{eo} = -\frac{\varepsilon}{\eta} \zeta_w \mathbf{E}
\]

\[
\mu_{eo} = -\frac{\varepsilon}{\eta} \zeta_w
\]

where \( \mathbf{u}_{eo} \) is the electroosmotic slip velocity [m/s], \( \mu_{eo} \) is the electroosmotic mobility [m^2 V/s], \( \varepsilon \) is the electrical permittivity of fluid [F/m], \( \eta \) is the viscosity of fluid [Pa·s], \( \zeta_w \) is the equilibrium wall zeta potential [V] and \( \mathbf{E} \) is the applied electric field.

It is notable in Fig. 1.2 that the velocity profile for electroosmotic flow is plug like in the bulk of fluid. The EDL can be assumed to be a slip plane with the appropriate slip velocity eq. (1.1). This slip velocity allows the numerical simulation to avoid the small
EDL and resolve only the bulk of fluid and hence saving a lot of computational resources.

Equation (1.1) is well known as the Smoluchowski slip velocity.

**FIG. 1.2** Schematic representation of electroosmotic flow induced due to Coulomb force on EDL and its velocity profile across a straight channel. It is noted that velocity variation is present only in the EDL. The electrophoresis velocity of particle is opposite in direction to electroosmosis.

### 1.2.3 Electrophoresis

Electrophoresis is the movement of charged particles inside an electrolyte due to the external electric field. This thesis work deals with manipulation of charged particles and hence electrophoresis is discussed here. Similar to the walls, particles obtain a surface charge when they come in contact with electrolyte. Hence these charges attract the counter ions of fluid around them forming EDL. So under the application of electric field these particles experience a similar Coulomb force which is in opposite direction when compared to electroosmosis. Hence electroosmosis and electrophoresis are always opposite in direction to each other. The value of electrophoresis velocity, $u_{ep}$ is given by

$$u_{ep} = \frac{\varepsilon}{\eta} \zeta_p E$$

(1.3)

$$\mu_{ep} = \frac{\varepsilon}{\eta} \zeta_p$$

(1.4)
where $\zeta_p$ is the particle zeta potential and $\mu_{ep}$ is the electrophoretic mobility.

The vector sum of electroosmosis and electrophoresis is called as the electrokinetic velocity, $\mathbf{u}_{ek}$ given by

$$
\mathbf{u}_{ek} = \frac{\varepsilon}{\eta} (\zeta_p - \zeta_w) \mathbf{E}
$$

(1.5)

$$
\mu_{ek} = \frac{\varepsilon}{\eta} (\zeta_p - \zeta_w)
$$

(1.6)

where $\mu_{ek}$ is called as electrokinetic mobility. From Eq. (1.6) it is clear that direction of motion of particles depends on difference between the particle and wall zeta potential.

1.2.4 Dielectrophoresis

Dielectrophoresis (DEP) is the response of polarizable particles either charged or uncharged to a non-uniform electric field. DEP is a powerful tool in manipulation of particles and can be used for trapping, sorting and filtering of particles in an electrolyte. Insulator based DEP (iDEP) and reservoir based DEP (rDEP) have been found to be effective and cheaper ways of manipulation of particles [35,36,41,42]. The particle experience a DEP force which depends on the strength of electric field, properties and size of particle and frequency of AC electric field. The DEP force for a spherical particle is given by

$$
\mathbf{F}_{DEP} = \frac{\pi}{4} \varepsilon a^3 f_{CM} \nabla|\mathbf{E}|^2
$$

(1.7)

here $a$ is the diameter of the particle, $f_{CM}$ is the Clausius-Mossotti factor. When an uncharged particle is placed in a medium experiencing a non-uniform electric field, the particle will move due to this force. In the absence of any electroosmosis and
electrophoresis the particle will start moving and its velocity can be calculated by equating the DEP force to that of Stokes drag. From this expression the velocity of particle is calculated as,

\[
U_{DEP} = \frac{\varepsilon a^2}{12\eta} f_{CM} \nabla |E|^2
\]  

(1.8)

The Clausius-Mossotti factor depends on the difference between the electrical conductivity of particle and the electrolyte. \(f_{CM}\) for low AC voltage frequencies is expressed as \(f_{CM} = (\sigma_p - \sigma_f)/(\sigma_p + 2\sigma_f)\), where \(\sigma_p\) and \(\sigma_f\) are the electrical conductivity of particle and electrolyte solution respectively. If it a positive value (i.e. when the conductivity of particles is greater than that of electrolyte), then it is called as positive DEP and if it is lesser than zero it is called as negative DEP. In case of positive DEP, the particles move from regions of lower electric field to higher electric field and it is the opposite for negative DEP. The total velocity of particles is given by the sum of fluid velocity, electrophoresis velocity and the DEP velocity,

\[
U_p = u + u_{ep} + U_{DEP}
\]  

(1.9)

This method is called as Lagrangian tracking method. This is suitable for small particles where the influence of the size and shape of particles on the electric field is negligible. The velocity particles used in this thesis are calculated from this expression.

1.2.4 Joule Heating

Joule Heating is inevitable in electrokinetic flows. When an electric field is applied on an electrolyte, Joule heating is generated due to the electrical resistance offered by the
electrolyte to electric field. It causes temperature rise and forms temperature gradients in the fluid, and in turn draws non-uniformities into the temperature dependent fluid properties (e.g., electric conductivity, dielectric permittivity, and dynamic viscosity) [43, 44]. As such, the favored plug-like velocity profile of electroosmotic flows can be distorted, which affects both the accuracy and the efficiency of electrokinetic sample manipulation [45,46]. These Joule heating effects can become significant especially when a buffer solution of high electric conductivity (which is often necessary for bio-samples [47]) and/or a polymer-based [e.g., polydimethylsiloxane (PDMS)] microchip of low thermal conductivity is used [48].

Previous studies on Joule heating and its effects in electroosmotic flows have been limited to inside either stand-alone capillaries or on-chip microchannels [38,43-46]. In capillary electrophoresis, Joule heating has been long known to generate transverse gradients in fluid temperature within the capillary, which increases the hydrodynamic dispersion of sample species via non-uniform diffusion and electrophoresis [49-51]. Axial temperature gradients can occur in thermostated capillary electrophoresis because of the dissimilar heat transfer conditions inside and outside the thermostated cartridge [52]. They can also occur near the capillary ends due to the liquid cooling effects from the reservoirs [53]. In either circumstance the axial temperature gradients have been demonstrated to induce a pressure-driven flow inside the capillary, leading to a reduced plate height in capillary electrophoresis [52-54].

The amount of Joule heating is directly proportional to electrical conductivity of electrolyte and the square of magnitude of electric field. It is given by the expression,
\[ Q = \sigma (E \cdot E) \tag{1.10} \]

where \( Q \) is the heat generated per unit volume [W/m\(^3\)], \( \sigma \) is electrical conductivity of electrolyte [S/m]. In electrokinetic microfluidic chips, Joule heating extends from within the fluid to the whole device due to thermal diffusion [55,56], which significantly complicates the problem of the coupled fluid and thermal transport [57-59]. The Joule heating in the microchannel is not uniform due to the varying cross section of the channel. This causes non uniform heating and thus results in temperature gradients throughout the device that results in a non-zero value of the so called electrothermal body force. The electrothermal body force term, \( f_{et} \) is given by [60],

\[ f_{et} = \rho_e E - 0.5E^2\nabla \varepsilon \tag{1.11} \]

where \( \rho_e \) is the volume charge density [C/m\(^3\)] given by \( \rho_e = \nabla \cdot (\varepsilon E) \). The first term on right hand side of this equation is the Coulomb force term that is dominant at low frequencies (\( \ll 10\text{MHz} \)) and the second term is Dielectric force, dominant at higher frequencies (\( \gg 10\text{MHz} \)). Under appropriate conditions this electrothermal body force can disrupt the plug like velocity profile of electroosmosis at the entrance of the channel (at microchannel reservoir interface).

**1.2.5 Induced Charge Electroosmosis**

Many of the microfluidic devices are manufactured using PDMS due to its several advantages as stated earlier. Most of the previous works in electrokinetics consider PDMS to be electrically insulating. But the PDMS has a small but a non-negligible value of dielectric constant. When an external electric field is applied to the device, it is leaked into
the PDMS. Though this leakage is negligible in most of the microchip, it cannot be neglected near sharp corners and rapidly changing cross sections of microchannel. The leaked electric field polarizes the opposite surfaces of PDMS [39]. This induces a zeta potential or charge that is different from the equilibrium zeta potential (which is formed when electrolyte comes in contact with the wall). This forms an “induced” electrical double layer that yields to induced-charge electroosmotic flow [40]. This induced flow has a nonlinear dependence on electric field and consists of two counter rotating circulations [61-63]. The induced zeta potential depends on the electric field leaked into the substrate which in turn depends on electrical permittivity of the substrate. Thamida and Chang [64] observed a nonlinear ejection flow from sharp corners in electrokinetic flows while using dielectric materials. Fig 1.3 shows a schematic diagram of the polarization of opposite surfaces of dielectric surfaces and hence its effect on the flow near the corner.
FIG. 1.3 Schematic representation of polarization of dielectric material under the application of external electric field. This polarization causes a locally generated electroosmotic flow that is directed towards the corner. This ejection flow disrupts the local flow field.

This phenomenon has been extensively studied around conducting surfaces of metal electrodes and particles that can either be electrically activated [65-67] or left floating [68-70]. This phenomenon is also observed near sharp corners [71-73] and at T-shaped or L-shaped [74-76] channels between narrow and wide channels where either one or a pair of vortices are found. At very low conductivities of electrolyte (when Joule heating is negligible) and at appropriate microchannel structures, ICEO distorts the plug like velocity
profile of electroosmosis. At the interface of reservoir and microchannel region ICEO would be strong at certain AC and DC voltages and can disrupt the electroosmotic flow at the microchannel entrance similar to Joule heating. This thesis provides a detailed experimental and numerical study of these phenomenon and how they impact the flow of sample and electrolyte at the reservoir microchannel interface.

### 1.3 Thesis structure

The remainder of this thesis is organized into three chapters. In the second chapter, we discuss the experimental and numerical study on the effects of Joule heating on electroosmotic flow at the entrance of microchannel. The origin and the growth of electrothermal body force and how it influences the flow field is explained. In the following chapter, the experimental and numerical study of ICEO at the entrance of microchannel is presented. Parametric study is performed to understand the effect ICEO on the flow field. More importantly two dimensional depth averaged analysis is done to simulate these two effects on electroosmotic entry flows. This depth averaged analysis can predict these two phenomena with good accuracy. Thus the costly 3D modelling could be replaced with a simpler depth averaged model that can be performed with much lesser computational resources. The detailed steps in arriving at the depth averaged governing equations are presented in the Appendix. In the last chapter we summarize the conclusions from the previous two chapters and discuss the potential future works.
CHAPTER TWO

JOULE HEATING EFFECTS ON ELECTROOSMOTIC ENTRY FLOW

2.1 Introduction

In electrokinetic microfluidic chips, Joule heating extends from within the fluid to the whole device due to thermal diffusion [55,56], which significantly complicates the problem of the coupled fluid and thermal transport [57-59]. Moreover, under appropriate conditions, the action of electric field on the thermally induced gradients in fluid properties can generate a pair of counter-rotating fluid vortices inside on-chip microchannels [77]. This so-called electrothermal flow [78,79] was first predicted by Hawkins et al. [80] to occur near a two-dimensional (2D) constriction in channel depth of insulator-based dielectrophoresis (DEP) systems. It was later experimentally verified by Sridharan et al. [77] using a rectangular microchannel with a widthwise constriction in the middle. Moreover, a 2D numerical model was developed by Sridharan et al. [77] to simulate the effects of Joule heating on electroosmotic flows in such a microfluidic chip. However, an unrealistically large convective heat transfer coefficient had to be assumed because most of the heat generated in the fluid was actually rejected along the depth-wise direction [48]. Hence a three-dimensional (3D) full-scale numerical model was later developed by Kale et al. [81] to capture the heat dissipation in all directions without any assumption, which, however, is computationally very expensive.

We present in this work the first experimental study of electroosmotic fluid entry from reservoir to microchannel in a PDMS-based microchip. More importantly, we
develop a 2D depth-averaged numerical model to study the effects of Joule heating on electroosmotic entry flows. This full-scale simulation in the horizontal plane of the microchip accounts for the influences of the top and bottom microchannel walls on the heat and fluid transfer via additional terms in the standard transport equations. It requires neither the assumption of high convective heat transfer coefficient in a regular 2D model [77] nor the consumption of expensive computational resource in a 3D model [81].

2.2 Experiment

2.2.1 Microfluidic chip fabrication

Fig. 2.1A shows a top-view picture of the microfluidic chip used in our experiments, which was fabricated with PDMS using the standard soft lithography technique. The detailed fabrication procedure can be referred to our previous paper [82] and is skipped here. The chip has a 2 mm thick PDMS slab on top and a 1 mm thick glass slide at the bottom with a 10 µm thin PDMS film sandwiched between. This three-layer structure is schematically illustrated by the cross-sectional view in Fig. 2.1B. At the bottom side of the PDMS slab is the 25 µm deep microchannel, which is composed of a long straight section of 400 µm wide in the middle and a tapered 40 µm wide, 1 mm long constriction at each end. The total length of the microchannel is 1 cm. At the far end of each constriction is a 5 mm-diameter reservoir for the fluid and sample supply. The PDMS-PDMS configuration of the microchannel ensures uniform and identical surface properties for the walls that surround the fluid.
FIG. 2.1. Top-view picture (A, the channel and reservoirs are filled with green food dye for clarity) and schematic cross-sectional view (B, not to scale) of the microfluidic chip used in experiments. The block arrow in (A) shows the direction of electroosmotic fluid flow.

2.2.2 Particle solution preparation and experimental methods

In order to study Joule heating effects on the fluid flow pattern, 520 nm-diameter fluorescent polystyrene microspheres (Bangs Laboratories, Fishers, IN) were used as tracers and re-suspended in 5 mM phosphate buffer solution. DC-biased AC electric voltages were applied through 0.5 mm-diameter platinum electrodes in the inlet and outlet reservoirs (see Fig. 2.1A). They were supplied using a function generator (33220A, Agilent Technologies, Santa Clara, CA) that was connected with a high-voltage amplifier (609E-6, Trek, Medina, NY). The AC voltage frequency was fixed at 1 kHz in experiments. Particle motion at the inlet/outlet reservoir-microchannel junctions was monitored using an inverted microscope imaging system (Nikon Eclipse TE2000U, Nikon Instruments) and recorded with a CCD camera (Nikon DS-Qi1Mc) at a rate of about 15 frames/s. The captured digital images were processed using Nikon imaging software (NIS-Elements AR 2.30, Nikon Instruments, Lewisville, TX). Prior to every test, the liquid heights in the two
reservoirs were carefully balanced to minimize the effects of pressure-driven flow. Also, each test was run for no more than 1 min to avoid significant backflow.

2.3 Theory

In this section, we first present the 2D depth-averaged governing equations for the coupled electric charge, energy and fluid transport involved in on-chip electroosmotic flows. Next the computational domain for the microfluidic chip under consideration is described along with boundary conditions necessary for solving the governing equations. Finally we present the numerical method with a table of parameters needed for modelling.

2.3.1 Governing equations

2.3.1.1 Electric field

As PDMS can be considered to be electrically insulating, electric field is confined inside the fluid part and governed by the quasi-electrostatic equation [83],

\[ \nabla \cdot (\sigma \mathbf{E} + i \omega \mathbf{D}) = 0 \]  \hspace{1cm} (2.1)

where \( \sigma \) is the electric conductivity of the fluid, \( \mathbf{E} = -\nabla \phi \) is the electric field with \( \phi \) as the electric potential, \( i \) is the imaginary unit, \( \omega = 2\pi f \) is the angular frequency of the applied electric voltage with \( f \) as the normal frequency, and \( \mathbf{D} = \varepsilon \mathbf{E} \) is the electric field displacement with \( \varepsilon \) as the fluid’s electric permittivity. Note that the convection current has been assumed to be negligible when compared with the conduction current, i.e., \( \sigma \mathbf{E} \) in Eq. (2.1) [83]. We are using DC-biased AC electric fields with the AC frequency, \( f \) (of 1 KHz), being much less than the charge relaxation frequency, \( \sigma / 2\pi \varepsilon \) (of order MHz [83]).
This results in negligible effects of frequency on the electric field displacement, and hence Eq. (2.1) reduces to

\[ \nabla \cdot (\sigma \mathbf{E}) = 0 \]  

(2.2)

We follow the same analysis as in our previous papers [77,81,82] to solve only the DC component of electric field, \( \mathbf{E}_{DC} \). The method used to incorporate the AC component into the energy and flow equations will be discussed later. A depth-averaged analysis for Eq. (2.2) was done by the use of an asymptotic method described in Lin et al. [84] (see the Appendix for the detailed derivation). The resulting 2D equation for the DC electric field preserves its original form in 3D and is given by

\[ \nabla_H \cdot (\sigma \mathbf{E}_{DC}) = 0 \]  

(2.3)

where \( \nabla_H \) is the gradient along the \( x \) and \( y \) directions in the horizontal plain, and \( \mathbf{E}_{DC} = -\nabla \phi_{DC} \) with \( \phi_{DC} \) being the applied DC potential.

**2.3.1.2 Temperature field**

The applied electric field across the microchannel induces Joule heating in the fluid, especially strong inside the two constriction regions due to the locally amplified electric field. This heat generated within the fluid is diffused in all directions through the PDMS and glass substrates in three modes: (1) conduction from the bottom wall of the channel to the PDMS film and then the glass substrate (see Fig. 2.1B); (2) conduction from the top and side walls of the channel to the PDMS slab, which is then removed by the natural
convection on the top and side surfaces of the PDMS slab; (3) natural convection on the top surface of the fluid in each reservoir. Hence temperature gradients are formed throughout the microfluidic chip. The steady-state time-averaged temperature field, $T$, in the fluid is governed by the energy equation [83],

$$\rho C_p (\mathbf{u} \cdot \nabla T) = \nabla \cdot (k \nabla T) + \sigma \langle \mathbf{E}^2 \rangle$$

(2.4)

where $\rho$, $C_p$ and $k$ are the mass density, heat capacity and thermal conductivity, respectively, of the fluid, and $\mathbf{u}$ is the fluid velocity. The last term on the right-hand side of Eq. (2.4) is the time-averaged heat generation due to Joule heating (present only inside the fluid). By the use of dimensional analysis, we find that the thermal Peclet number, $Pe = Re Pr$, is smaller than 0.1 because the Reynolds number is $Re \leq 0.01$ and the Prandtl number is $Pr < 10$ in typical electrokinetic microfluidic systems [83, 85]. Therefore, the advection term in Eq. (2.4) can be dropped out leaving a purely heat conduction equation,

$$0 = \nabla \cdot (k \nabla T) + \sigma \langle \mathbf{E}^2 \rangle$$

(2.5)

An asymptotic analysis of Eq. (2.5) yields the following 2D depth-averaged energy equation in the fluid (see the Supporting Information for the detailed derivation),

$$0 = k \nabla^2_H T + \sigma E_{DC}^2 (1 + r^2) - \frac{T-T_\infty}{d_{ch}} \left( \frac{1}{R_{us}} + \frac{1}{R_{ls}} \right)$$

(2.6a)

$$R_{us} = \frac{t_{us,PDMS}}{k_{PDMS}} + \frac{1}{h}$$

(2.6b)

$$R_{ls} = \frac{t_{ls,PDMS}}{k_{PDMS}} + \frac{t_{glass}}{k_{glass}}$$

(2.6c)
where $r$ is the ratio of root-mean-squared (RMS) AC field to DC field (equivalent to the ratio of RMS AC voltage to DC voltage). The last term on the right-hand-side of Eq. (2.6a) accounts for the heat dissipation through the upper and lower substrates to the fluid (see Fig. 2.1B), where $T_\infty$ is the room temperature, $d_{ch}$ is the depth of the microchannel, and $R_{us}$ and $R_{ls}$ are the equivalent thermal resistances offered by the upper and lower substrates to the fluid, respectively [86]. In the definitions of thermal resistances in Eq. (2.6b) and Eq. (2.6c), $t_{us,PDMS}$ is the thickness of the PDMS slab (see Fig. 2.1B), $k_{PDMS}$ is the thermal conductivity of PDMS, $h$ is the natural convection coefficient, $t_{ls,PDMS}$ is the thickness of the PDMS film (see Fig. 2.1B), and $t_{glass}$ and $k_{glass}$ are the thickness and thermal conductivity of glass. Note that natural convection is considered only for the upper-substrate thermal resistance while an isothermal condition (at room temperature) is used in the lower-substrate thermal resistance. This is attributed to the fact that the bottom surface of the glass slide is rested on the microscope stage. It is also important to note that the fluid inside the reservoirs in the horizontal plain experiences a different upper-substrate thermal resistance, $R_{us}$, from that in Eq. (2.6b), where the thermal conductivity of PDMS, $k_{PDMS}$, should be replaced with the fluid thermal conductivity, $k$

### 2.3.1.3. Flow field

As noted earlier, the Reynolds number is much smaller than 1 [83,85], so the flow field is governed by the continuity and modified Stokes equations for an incompressible fluid [83],
\[ 0 = -\nabla p + \nabla \cdot (\eta \nabla u) + \langle f_e \rangle \] (2.7a)

\[ \nabla \cdot u = 0 \] (2.7b)

where \( p \) is the pressure, \( \eta \) is the dynamic viscosity of the fluid, and \( \langle f_e \rangle \) is the time-averaged electrothermal body force that is equal to the sum of Coulomb force and dielectric force terms given by [87],

\[ \langle f_e \rangle = \langle \rho_e E - 0.5E^2 \nabla \varepsilon \rangle \] (2.8)

where \( \rho_e \) is the free charge density given by Poisson’s equation, \( \rho_e = \nabla \cdot (\varepsilon E) \). A linear asymptotic analysis for Eq. (2.7a) and Eq. (2.7b) leads to the following 2D depth-averaged flow equations (see the Appendix for the detailed derivation),

\[ \nabla_H \cdot u = 0 \] (2.9a)

\[ 0 = -\nabla_H p + \nabla_H \cdot (\eta \nabla_H u) + f_e - 3(\eta u + \varepsilon \zeta_w E_{DC})/d_{ch}^2 \] (2.9b)

\[ f_e = (1 + r^2)[\nabla_H \cdot (\varepsilon E_{DC})E_{DC} - 0.5E_{DC}^2 \nabla_H \varepsilon] \] (2.9c)

Note that the effects of AC voltage on the electrothermal body force has been considered by the use of the AC to DC field ratio, \( r \), in Eq. (2.9c) [77,81,82]. This force is non zero due to the presence of gradients in fluid properties such as electric conductivity [through \( E_{DC} \) in Eq. (2.3)] and permittivity, which are caused by the Joule heating induced temperature gradients. The last term of Eq. (2.9b) is the additional body force that arises due to the depth-averaging with \( \zeta_w \) being the wall zeta potential [84].
As mentioned earlier, the fluid flow field is visualized by tracking the motion of small fluorescent particles. The trajectories of these tracing particles are simulated by calculating their velocity, \( \mathbf{U}_P \), as a vector sum of fluid velocity, \( \mathbf{u} \), and electrophoretic particle velocity, \( \mathbf{U}_{EP} \).

\[
\mathbf{U}_P = \mathbf{u} + \mathbf{U}_{EP} \tag{2.10a}
\]

\[
\mathbf{U}_{EP} = \varepsilon \zeta_p \mathbf{E}_{DC} / \eta \tag{2.10b}
\]

where \( \zeta_p \) is the particle zeta potential. The influence of DEP on particle velocity has been neglected in Eq. (2.10a) because it is found in our simulation to be two orders of magnitude smaller than the fluid velocity. Other contributions to particle velocity due to, for example, Brownian, inertial and gravitational motions are also neglected [85].

2.3.2 Computational domain and boundary conditions

![Diagram](image)

FIG. 2.2. Computational domain for the 2D depth-averaged modeling of the electrokinetic microfluidic chip with important boundary conditions being highlighted.

The coupled depth-averaged electric, temperature and flow field equations, i.e., Eqs. (2.3), (2.6a), (2.9a) and (2.9b), are solved simultaneously in the horizontal plain of the
microfluidic chip that cuts the microchannel by half along its depth. Fig. 2.2 shows the 2D computational domain that covers only one half of the chip due to symmetry. It has the fluid and PDMS subdomains where the latter is, however, solved only for the temperature field. Due to their high thermal and electric conductivities, platinum electrodes are considered to remain at constant temperature and electrical potential during the experiment and hence simply treated as holes with appropriate boundary conditions in the 2D simulation. The four boundaries of the computational domain in Fig. 2.2 are the microchannel sidewalls, electrode surfaces, outer walls of PDMS, and the symmetry line. The boundary conditions required to solve the coupled equations are summarized below except for the symmetry ones.

2.3.2.1. Electric field

Electric potential, $\phi_{DC} = \phi_{DC}^0$, is applied on the inlet electrode surface where $\phi_{DC}^0$ is the applied DC voltage. On the other electrode surface $\phi_{DC} = 0$ (ground) is applied. Since the electric field is confined within the fluid subdomain, an electrically insulating condition, $\mathbf{n} \cdot (\sigma \nabla \phi_{DC}) = 0$, is imposed on all sidewalls of the microchannel where $\mathbf{n}$ denotes the unit normal vector.

2.3.2.2. Temperature field

Due to the electrode’s high thermal conductivity, it can be assumed that there is no temperature drop inside them. So on the electrode surface, an isothermal condition, $T = T_{\infty}$, is applied. The condition of temperature continuity is applied by default in COMSOL.
at the interface of the fluid and PDMS subdomains. The outer walls of PDMS are exposed to atmosphere and hence experience a natural convection.

2.3.2.3 Flow field

On the surface of electrodes, a no-slip condition, $\mathbf{u} = 0$, is applied because the local electric field is normal to its surface. As pressure driven flow is absent, an equal pressure needs to be applied to each of the reservoir. In our model we choose one point each on the most left and right ends of the fluid subdomain (that lies on the line of symmetry) and apply the condition, $p = 0$. Since the electric double layer is very small (on the order of few nanometers in our experimental conditions) compared to the width of the microchannel, fluid motion can be modelled by using the well-accepted Smoluchowski electroosmotic slip velocity [22,23],

$$\mathbf{u} = -\varepsilon \zeta_w \mathbf{E}_{DC} / \eta,$$

on the sidewalls of both the microchannel and the two reservoirs.

2.3.3 Numerical method and material properties

Commercial finite element software package, COMSOL Multiphysics 5.1 (Burlington, MA) was used to carry out the numerical simulation. The 2D geometry in Fig. 2.2 was created using the ‘Geometry’ feature. The modules of ‘Electrical currents’, ‘Heat transfer in fluids’, ‘Heat transfer in solids’, and ‘Creeping flow’ were coupled together to solve the governing equations, i.e., Eqs. (2.3), (2.6a), (2.9a) and (2.9b). The function of ‘Streamline’ was used to plot the streaklines of tracing particles via the velocity in Eq. (2.10a). In order to avoid singularities, sharp corners were filleted. The domain was meshed
using approximately 120,000 free triangular elements. The number of elements was varied from 5,000 to 120,000 and we found that the results were grid independent after approximately 60,000 elements. The constriction region was meshed extremely fine (with a maximum element size of 2 µm) compared to the mesh size at other regions because of the occurrence of high electric field, temperature and fluid property gradients in this region. The model was solved using a nonlinear, segregated, iterative solver with appropriate damping factors to achieve a faster convergence. The whole process for one run took about 10 minutes in a standardly configured laptop.

The temperature dependences of the fluid properties including electric conductivity, permittivity and viscosity were evaluated in our model using the following expressions [43,46]

\[
\sigma = \sigma_\infty [1 + \beta (T - T_\infty)] \tag{2.11a}
\]

\[
\varepsilon = \varepsilon_\infty [1 + \alpha (T - T_\infty)] \tag{2.11b}
\]

\[
\eta = 2.761 \times 10^{-6} \exp(1713/T) \tag{2.11c}
\]

where \( \sigma_\infty \) and \( \varepsilon_\infty \) are the electric conductivity and permittivity of the fluid at room temperature \( T_\infty \), and \( \beta \) and \( \alpha \) are their respective temperature coefficients. The values of these constants along with other parameters used in the model are given in Table 2.1. Note that the wall zeta potential, \( \zeta_w \), was determined from the electroosmotic fluid velocity that was measured using the electric current-monitoring method in a straight uniform
microchannel [88]. The particle zeta potential, $\zeta_p$, was calculated from the electrokinetic particle velocity that was measured by tracking single particle motions in a straight uniform microchannel [34,35,36].

TABLE 2.1. Summary of parameters and material properties used for modeling Joule heating effects [89,90].

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Value</th>
<th>Unit</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_\infty$</td>
<td>293</td>
<td>K</td>
<td>Room Temperature</td>
</tr>
<tr>
<td>$\sigma_\infty$</td>
<td>0.1</td>
<td>S/m</td>
<td>Fluid electric conductivity at room temp</td>
</tr>
<tr>
<td>$\varepsilon_\infty$</td>
<td>$7.09 \times 10^{-10}$</td>
<td>F/m</td>
<td>Fluid electric permittivity at room temp</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>$-0.0046$</td>
<td>1/K</td>
<td>Temperature coefficient of fluid permittivity</td>
</tr>
<tr>
<td>$\beta$</td>
<td>0.02</td>
<td>1/K</td>
<td>Temperature coefficient of fluid conductivity</td>
</tr>
<tr>
<td>$k$</td>
<td>0.61</td>
<td>W/(m\cdot K)</td>
<td>Thermal conductivity of fluid</td>
</tr>
<tr>
<td>$k_{PDMS}$</td>
<td>0.15</td>
<td>W/(m\cdot K)</td>
<td>Thermal conductivity of PDMS</td>
</tr>
<tr>
<td>$k_{glass}$</td>
<td>1.38</td>
<td>W/(m\cdot K)</td>
<td>Thermal conductivity of glass</td>
</tr>
<tr>
<td>$d_{ch}$</td>
<td>25</td>
<td>$\mu$m</td>
<td>Depth of microchannel</td>
</tr>
<tr>
<td>$t_{us,PDMS}$</td>
<td>2</td>
<td>mm</td>
<td>Thickness of PDMS slab</td>
</tr>
<tr>
<td>$t_{ls,PDMS}$</td>
<td>10</td>
<td>$\mu$m</td>
<td>Thickness of PDMS film</td>
</tr>
<tr>
<td>$t_{glass}$</td>
<td>1</td>
<td>mm</td>
<td>Thickness of glass slide</td>
</tr>
<tr>
<td>$h$</td>
<td>20</td>
<td>W/(m$^2$\cdot K)</td>
<td>Natural convection coefficient</td>
</tr>
<tr>
<td>$\zeta_w$</td>
<td>$-50$</td>
<td>mV</td>
<td>Zeta potential of walls</td>
</tr>
<tr>
<td>$\zeta_p$</td>
<td>$-80$</td>
<td>mV</td>
<td>Zeta potential of particles</td>
</tr>
<tr>
<td>$\rho$</td>
<td>1000</td>
<td>kg/m$^3$</td>
<td>Mass density of fluid</td>
</tr>
</tbody>
</table>
2.4 Results and Discussion

Fig. 2.3 shows the comparison of experimentally obtained superimposed images and numerically predicted particle streaklines at the interface of the inlet reservoir and microchannel for various 20 V DC-biased AC voltages. In each case the flow pattern is established soon after the electric voltage is supplied. At 450 V AC, the tracing particles mostly follow electric field lines and flow smoothly from reservoir to microchannel. Only those that travel near the channel centerline exhibit a slight bending, which is correctly captured by our simulation in Fig. 2.3A (bottom half). This phenomenon is not expected for a pure electroosmotic flow with uniform properties due to the similarity of flow and electric fields therein [91]. It is a consequence of electrothermal flow that arises from the action of electric field on Joule heating-induced fluid inhomogeneities. This flow grows with the increase of AC voltage, leading to the formation of two small counter-rotating vortices near the centerline of the channel entrance at 500 V AC (see Fig. 2.3B). Further increasing the AC voltage significantly enhances the fluid circulations as demonstrated in Fig. 2.3C for 600 V AC and Fig. 2.3D for 700 V AC. The experimentally observed variations in the size and location of electrothermal circulations with AC voltage are predicted with a good agreement for every case in Fig. 2.3.
FIG. 2.3. Comparison of experimentally obtained images (top half) and numerically predicted streaklines (bottom half) of 520-nm tracing particles at the inlet reservoir-microchannel junction under various 20 V DC-biased AC voltages: (A) 450 V AC (RMS), (B) 500 V AC, (C) 600 V AC, and (D) 700 V AC. The arrowed line in D indicates the direction of the electrothermal flow circulation. The direction of electroosmotic fluid flow is from left to right in all images while that of the tracing particles is opposite due to the dominance of particle electrophoresis over fluid electroosmosis.

The generation of electrothermal fluid circulations in electroosmotic entry flow can be explained by the numerically predicted fluid property and field contours in Fig. 2.4 for the case of 20 V DC-biased 600 V AC. The size mismatch between the reservoir and the microchannel (more precisely, the constriction section of the channel; see Fig. 2.1A) amplifies the electric field inside the microchannel as illustrated in Fig. 2.4A. This draws strong Joule heating in the constriction region and hence raises the local fluid temperature. However, as viewed from Fig. 2.4B, the fluid inside the reservoir remains slightly above room temperature. The resulting steep temperature gradients lead to highly non-uniform fluid properties at the reservoir-microchannel junction, e.g., electric permittivity decreases by approximately 10% (Fig. 2.4C) while electric conductivity increases by almost 50% (Fig. 2.4D) in the constriction. The action of electric field (both DC and AC) on these gradients in fluid properties induces the electrothermal body force, i.e., $f_e$ in Eq. (2.9c), at
the channel entrance region. The magnitude of this force is higher near the channel walls (than the centerline, see Fig. 2.4E) and hence able to reverse the local fluid velocity (towards reservoir), yielding the electrothermal fluid circulations in Fig. 2.4F.

FIG. 2.4. Illustration of numerically predicted contours of electric field (A), temperature field (B), electric permittivity (C), electric conductivity (D), electrothermal body force (E), and fluid velocity magnitude (F, arrows are velocity vector plots) at the inlet reservoir-microchannel junction under 20 V DC-biased 600 V AC.

The growth of electrothermal fluid circulations at a higher AC voltage in Fig. 2.3 is attributed to the increase in Joule heating-induced temperature gradients (and in turn fluid property gradients). This is demonstrated in Fig. 2.5, where the temperature variations at four points of the microfluidic chip (see their locations in the inset) are plotted for various AC voltages: (1) 200 µm ahead of the constriction inside the Reservoir, (2) center of the Constriction, (3) center of the Microchannel, and (4) 200 µm above the constriction inside the PDMS. The fluid temperature at the center of the constriction increases in a parabolic nature with increase in AC voltage. In contrast, the fluid
temperature rise inside the reservoir is almost linear and much less than that in the constriction. This difference in temperature variation causes significantly increased temperature gradients at the reservoir-microchannel junction for higher AC voltages. However, the temperatures at the center of the microchannel and the PDMS both experience a very small increase with AC voltage. This indicates that Joule heating in the fluid is primarily rejected along the channel depth direction, especially through the glass slide at the bottom [48,81].

**FIG. 2.5.** Numerically predicted temperature variations at four points of the computational domain (see their locations in the inset, not drawn to scale; refer also to Fig. 2.2) for various AC voltages with a fixed 20 V DC offset.

Electrothermal flow also occurs in electroosmotic exit flow from the microchannel to the outlet reservoir due to the same mechanism as noted above. The experimentally and numerically obtained particle streaklines at the outlet reservoir-microchannel junction are displayed in Fig. 2.6 for various 20 V DC-biased AC voltages. Similar to the electroosmotic entry flow in Fig. 2.3, fluid circulations appear at 500 V AC in the exit flow and become
more prominent with the further increase in AC voltage. Moreover, the electrothermal flows in both locations point away from the channel at the near-wall region due to the local strong electrothermal body force (see Fig. 2.4E) and circulate back in the bulk fluid region (see the arrowed lines in Fig. 2.3D and Fig. 2.6D). However, the centers of the two fluid vortices are shifted from near the channel centerline in the entry flow (Fig. 2.3) to near the channel walls in the exit flow (Fig. 2.6). These phenomena are all reasonably simulated by the 2D depth-averaged model.

**FIG. 2.6.** Comparison of experimentally (top) and numerically (bottom) obtained particle streaklines in electroosmotic exit flow from the microchannel to the outlet reservoir under various 20 V DC-biased AC (RMS) voltages: (A) 450 V AC, (B) 500 V AC, (C) 600 V AC, and (D) 700 V AC. The arrowed line in D indicates the direction of the electrothermal flow circulation. The direction of electroosmotic fluid flow is from left to right in all images.
2.5 Summary

This chapter presents the first experimental and numerical studies of Joule heating effects on electroosmotic fluid entry from reservoir to microchannel in a polymer microfluidic chip. Electrothermal fluid circulations are observed to form at the reservoir-microchannel junction due to the action of the locally amplified electric field upon the Joule heating-induced gradients in fluid properties. Their influences on the electroosmotic entry flow grow with the increasing AC voltage when the DC voltage is fixed. Moreover, a depth-averaged theoretical analysis is performed for the transport equations, based on which a 2D numerical model is developed to understand the effects of Joule heating on fluid temperature and flow fields in electrokinetic microfluidic chips. This model takes into the account the heat dissipation and electroosmotic slip from/on the top and bottom microchannel walls without any assumption. It is found sufficient to predict the temperature field in both the fluid and the substrate subdomains of the entire microchip. More importantly, it simulates accurately the size and position of the experimentally observed fluid vortices in electroosmotic flow at both the inlet and the outlet reservoir-microchannel junctions under various AC voltages. Due to the simple 2D nature, this modeling can be easily carried out in a laptop, which enables quick and efficient parametric studies for design and operation optimizations.
CHAPTER THREE

INDUCED CHARGE EFFECTS ON ELECTROOSMOTIC ENTRY FLOW

3.1 Introduction

ICEO is a nonlinear electrokinetic phenomenon that is prominent at low electrolyte conductivities and especially near sharp corners of the microchannel under appropriate electric fields. The leakage of electric field into a polarizable substrate polarizes its opposite surfaces and hence results in a change of zeta potential over the surface. This induced zeta potential disrupts the electroosmotic flow (due to equilibrium zeta potential) and is called as induced charge electroosmosis flow. The induced charge effect on conducting spheres and particles [39,40] and on dielectric materials inside the channel [96] have been extensively done. The induced charge effects would be prominent near the reservoir microchannel region due to the rapidly changing cross-section. This results in the formation of two counter rotating circulations.

In this chapter we would be presenting the results of both experimental and numerical modelling of ICEO effects. For the numerical modelling both a regular 2D and 2D Depth averaged analysis are presented and it is seen that use of the depth averaged model is better and can be used to get comparable results with experiments. The numerical model is used to explain the electric field leakage into the PDMS and its effect on the zeta potential. To explain the variation of ICEO effects, a parametric study is done such as: the effects of AC Voltage, effect of channel width and effect of using different materials to make the channels (variation of the relative permittivity of substrate).
3.2 Experiment

3.2.1 Microchip Fabrication

Fig. 3.1 shows a picture of the microchannel used in the experiments that was fabricated using the standard soft lithography technique. The detailed procedure can be referred from our previous works [81,82]. Similar to the previous chapter, the microchip has a 2 mm thick PDMS on top of 1 mm thick glass slide. We do not have any PDMS film between the in between them (a 10 μm film PDMS was sandwiched in the previous experiment). The bottom side of PDMS slab is the microchannel of 25 μm deep. It consists of a 3.3 cm long straight channel of 500 μm wide between two fluid reservoirs. Reservoirs are the well that supply the microchannel with fluid and samples and are 5 mm in diameter. Near the inlet reservoir, a 180 μm long constriction with two different widths are used in the experiments. The widths of the channel were 35 μm and 60 μm. The inset in fig. 3.1 shows the detailed schematic of the constriction region.
FIG. 3.1. Picture of microchannel (the channel and reservoirs are filled with green food dye for clarity). The direction of electroosmotic fluid flow is shown in the block arrow. The inset shows the zoomed in schematic of the constriction region.

3.2.2 Particle solution preparation and experimental methods

1 µm polystyrene microspheres (Bangs Laboratories, Fishers, IN) were used to study the ICEO effects. The particles were re suspended in 0.01 mM phosphate buffer solution (used as the electrolyte). The low concentration of the buffer solution results in low electrical conductivity of electrolyte. The electrolyte’s conductivity was measured to be 6 µS/cm. This ensures that Joule Heating is very low and hence the effects of Joule heating phenomenon can be neglected. DC biased AC voltage was supplied through the 0.5 mm-diameter platinum electrodes in the inlet and outlet reservoir. The AC voltage was supplied at a constant frequency of 1Khz. Voltage was supplied using a function generator (33220A, Agilent Technologies, Santa Clara, CA) that was connected with a high-voltage
amplifier (609E-6, Trek, Medina, NY). Particle motion at the reservoir-microchannel junctions was monitored using an inverted microscope imaging system (Nikon Eclipse TE2000U, Nikon Instruments) and recorded with a CCD camera (Nikon DS-Qi1Mc) at a rate of about 15 frames/s. The captured digital images were processed using Nikon imaging software (NIS-Elements AR 2.30, Nikon Instruments, Lewisville, TX). To avoid any pressure driven flow, the fluid in each reservoir was carefully levelled before the start of each experiment. The experiments were not run for more than 1 min to avoid any backflow.

3.3 Theory

In this section, the governing equations for electric field and flow field involved in on-chip ICEO flows for both the regular 2D and 2D depth averaged models. Next the 2D computational domain for the microfluidic chip under consideration is described along with the necessary boundary conditions for solving the governing equations. Finally we present the numerical method with a table of parameters used in modelling.

3.3.1 Governing equations

3.3.1.1. Electric field

The PDMS used is weakly polarizable and has a small value of dielectric constant. The electric field inside the fluid, PDMS and the glass parts are governed by the quasi-electrostatic equation [83],

\[ \nabla \cdot (\sigma \mathbf{E} + i\omega \mathbf{D}) = 0 \]

(3.1)

where \( \sigma \) is the electric conductivity, \( \mathbf{E} = -\nabla \phi \) is the electrical field with \( \phi \) as the electrical potential, \( i \) is the imaginary unit, \( \omega = 2\pi f \) is the angular frequency of the applied electric
voltage with $f$ as the normal frequency, and $D = \varepsilon E$ is the electric field displacement with $\varepsilon$ as the electric permittivity. Note that the convection current has been assumed to be negligible in Eq. (3.1) [83]. Since the electrical conductivity of electrolyte is very small ($\sigma = 6 \ \mu\text{S/cm}$) and conduction current can be assumed to be zero for PDMS and glass substrates. Hence the conduction current can also be neglected. Eq. (3.1) reduces to a simple Laplace equation for both AC and DC voltage cases [73,86] (for both inside the fluid and PDMS). The equations for regular 2D and depth averaged analysis are,

$$\nabla^2 \phi_f = 0 \quad (3.2a)$$

$$\nabla^2 \phi_w = 0 \quad (3.2b)$$

with $\phi_f$ and $\phi_w$ as the electric potentials inside the fluid and the PDMS (or glass) substrates respectively.

### 3.3.1.2. Flow field

The Flow field is governed by the steady state continuity and momentum equations for incompressible fluid given by,

$$\nabla \cdot \mathbf{u} = 0 \quad (3.3a)$$

$$\rho (\mathbf{u} \cdot \nabla \mathbf{u}) = -\nabla p + \eta \nabla^2 \mathbf{u} \quad (3.3b)$$

where $p$ is the pressure, $\eta$ is the dynamic viscosity of the fluid. Due to the very low electrical conductivity of fluid, the Joule heating in the device is negligible. Hence the
properties such as electrical conductivity, permittivity and viscosity do not vary throughout the entire domain. The governing flow equation for 2D depth averaged analysis will be different from Eq. (3.3b). We follow the same asymptotic analysis as the previous chapter [84]. In this case the electrothermal body force terms are zero and thus simplifying the analysis. The depth averaged flow equations for are,

\[
\nabla_H \cdot \mathbf{u} = 0 \tag{3.4a}
\]

\[
0 = -\nabla_H p + \nabla_H \cdot (\eta \nabla_H \mathbf{u}) - 3 (\eta \mathbf{u} + \epsilon \zeta \omega \mathbf{E}_1) / d_{ch}^2 \tag{3.4b}
\]

Here \( \nabla_H \) is the gradient along the \( x \) and \( y \) directions, \( \mathbf{E}_1 \) is the electric field due to the DC supply. The last two terms in Eq. (3.4b) are the additional terms due to depth averaging (See Appendix for more information).

The velocity of particles, \( \mathbf{U}_p \) used in the experiments is equal to the sum of fluid velocity, \( \mathbf{u} \), electrophoretic velocity, \( \mathbf{U}_{EP} \) and dielectrophoretic velocity \( \mathbf{U}_{DEP} \) as follows,

\[
\mathbf{U}_p = \mathbf{u} + \mathbf{U}_{EP} + \mathbf{U}_{DEP} \tag{3.5a}
\]

\[
\mathbf{U}_{EP} = \epsilon \zeta_p \mathbf{E}_1 / \eta \tag{3.5b}
\]

\[
\mathbf{U}_{DEP} = \epsilon d^2 f_{CM} \nabla (E_1^2 + E_2^2) / 12 \eta \tag{3.5c}
\]

where \( \zeta_p \) is the particle zeta potential, \( f_{CM} \) is the Clausius-Mossotti (CM) factor, \( d \) is the diameter of the particle used. The value of the CM factor is calculated from the expression \( f_{CM} = (\sigma_p - \sigma_f) / (\sigma_p + 2\sigma_f) \), where \( \sigma_p \) and \( \sigma_f \) are the electrical conductivity of particle
and electrolyte solution respectively. The particle conductivity can be calculated as \( \sigma_p = 4K_s/d \) with \( K_s = 1 \) nS as the recommended value of surface conductance for polystyrene particles [92]. For 1 μm particles, \( \sigma_p = 40 \) μS/cm while \( \sigma_f = 6 \) μS/cm and so the value of \( f_{CM} = 0.65 \) [37]. Thus the particles experience positive DEP.

### 3.3.2 Computational domain and boundary conditions

![Diagram showing a computational domain for depth averaged modelling.](image)

**FIG. 3.2.** Computational domain for performing the depth averaged modelling. The boundary conditions have been highlighted. The electrodes have been considered as holes with appropriate boundary conditions.

Equations (3.2) and (3.3) are solved for the computational domain consisting of both the fluid and the PDMS (as shown in Fig. 3.2). The platinum electrodes can be considered as holes with appropriate boundary conditions due to their high electrical conductivity. The boundaries in the device are the sidewalls separating the fluid domain and PDMS, electrode surfaces, the outer walls of PDMS. The boundary conditions for both the 2D and the 2D depth averaged analysis are the same and so the conditions presented in
this section is common for both the modelling. The boundary conditions used on each of the boundaries are explained below.

### 3.3.2.1 Electric field - DC

Electric potential, $\phi_{1,f} = \phi_{DC}$, is applied on the inlet electrode surface where $\phi_{DC}$ is the applied DC voltage. On the other electrode surface $\phi_{1,f} = 0$ (ground) is applied. On the surfaces separating the fluid and PDMS (or glass) domains, the following boundary conditions are applied [73],

$$\frac{\partial \phi_{1,f}}{\partial n} = 0 \quad (3.6a)$$

$$\phi_{1,w} + \lambda \frac{\varepsilon_w}{\varepsilon_f} \frac{\partial \phi_{1,w}}{\partial n} = \phi_{1,f} + \zeta_w \quad (3.6b)$$

Here $\phi_{1,f}$ and $\phi_{1,w}$ are electric potentials inside the fluid and PDMS substrates for the DC case. $\varepsilon_w$ and $\varepsilon_f$ are the electrical permittivity of PDMS and fluid domain, $\lambda$ is the Debye length and $\zeta_w$ is the equilibrium wall zeta potential. Electric field inside the fluid is for DC is $E_1 = -\nabla \phi_{1,f}$.

### 3.3.2.2 Electric field - AC

Similar to the DC case, the AC voltage is applied on the inner side of one electrode $\phi_{2,f} = \phi_{AC}$ and ground is applied on the other. On the interface between the fluid and PDMS, the following sets of boundary conditions are applied [86],
\[ \frac{\varepsilon_w}{\varepsilon_f} \frac{\partial \phi_{2,w}}{\partial n} = \frac{\gamma^2}{\gamma^2 - 1} \frac{\partial \phi_{2,f}}{\partial n} \]  

(3.7a)

\[ \phi_{2,f} - \phi_{2,w} = \frac{\lambda}{\gamma^2} \frac{\varepsilon_w}{\varepsilon_f} \frac{\partial \phi_{2,w}}{\partial n} \]  

(3.7b)

Here \( \gamma^2 = 1 + j(2\pi f) \lambda^2/D_f \), with \( \lambda^2/D_f \) as the diffusion time and \( D_f \) is the diffusivity of the electrolyte and \( f \) is frequency of AC signal. Due to the low AC frequency used in our experiments, the imaginary part of \( \gamma^2 \) is very small. Electric field inside the fluid is for AC is \( \mathbf{E}_2 = -\nabla \phi_{2,f} \). Insulating boundary conditions, \( \mathbf{n} \cdot (\nabla \phi) = 0 \) are applied on the outer surfaces of PDMS for both DC and AC fields. The subscripts 1 and 2 in \( \phi \) and \( \mathbf{E} \) denote the electric potential and electric field for DC and AC case respectively.

### 3.3.2.3 Flow field

No slip condition, \( \mathbf{u} = 0 \) is applied on the surface of the electrode since the electric field is normal to it. Due to the absence of pressure driven flow, equal pressures of \( p = 0 \) is applied at two points at inlet and outlet reservoir at their far ends. The electrical double layer is very small compared to the dimension of the channel. Hence the well-accepted Smoluchowski electroosmotic slip velocity [22,23], \( \mathbf{u} = -\varepsilon \zeta \mathbf{E}/\eta \) is applied on the interface between fluid and PDMS. Here the zeta potential \( \zeta \) is the difference between the electric potentials in the wall and the fluid for both DC and AC cases. It is the sum of induced and equilibrium zeta potentials. Hence the slip velocity is given by,

\[ \mathbf{u} = -\varepsilon \left[ (\phi_{1,w} - \phi_{1,f})\mathbf{E}_1 + (\phi_{2,w} - \phi_{2,f})\mathbf{E}_2 \right]/\eta \]  

(3.8)
3.3.3 Numerical method and material properties

Commercial finite element software package, COMSOL Multiphysics 5.1 (Burlington, MA) was used to carry out the numerical simulation. The 2D geometry in Fig. 3.2 was created using the ‘Geometry’ feature. The modules ‘Electrostatics’ and ‘Laminar flow’ were used to solve the governing equations. Several functions like ‘Streamline’ and ‘Surface’ were used to post process the data acquired and present the results. Unlike our previous works on Joule heating [77,81,82] the temperature dependence of fluid properties need not be considered since the it is almost negligible in these experiments. The sharp corners were filleted and to avoid any singularities. The mesh independence in the results were tested by varying the total number of elements from approximately 6000 elements to 110000 elements and the model was grid independent after approximately 30000 elements. The constriction region in channel was meshed very fine with a maximum element size of 2 µm. The PDMS domain was meshed with a lesser mesh quality when compared to the mesh inside the fluid domain. The model was solved by a nonlinear, segregated, iterative solver with appropriate damping factors for a faster convergence. The 2D modelling has low memory and resource requirements and so can be solved in a standard PC. It took only a few minutes to solve the model. The wall zeta potential, $\zeta_w$ was determined from the electroosmotic fluid velocity that was measured using the electric current-monitoring method in a straight uniform microchannel [88]. The particle zeta potential, $\zeta_p$ was calculated from the electrokinetic particle velocity that was measured by tracking single particle motions in a straight uniform microchannel under the application of a small DC Voltage [34-36]. Table 3.1 provides all the values of parameters used in the modelling. The
Debye length has been assumed to be 100 nm. To perform the parametric study on the relative permittivity of wall, the values used were between 2 and 10 as they are the acceptable values of permittivity that can be used for making microchannels.

### TABLE 3.1. Summary of parameters and material properties used for modeling ICEO effects [89,90].

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Value</th>
<th>Unit</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\varepsilon_f)</td>
<td>(7.09 \times 10^{-10})</td>
<td>F/m</td>
<td>Fluid electric permittivity</td>
</tr>
<tr>
<td>(\varepsilon_w)</td>
<td>4</td>
<td>F/m</td>
<td>Relative electric permittivity of wall</td>
</tr>
<tr>
<td>(d_{ch})</td>
<td>25</td>
<td>(\mu)m</td>
<td>Depth of microchannel</td>
</tr>
<tr>
<td>(\zeta_w)</td>
<td>-100</td>
<td>mV</td>
<td>Zeta potential of walls</td>
</tr>
<tr>
<td>(\zeta_p)</td>
<td>-80</td>
<td>mV</td>
<td>Zeta potential of particles</td>
</tr>
<tr>
<td>(\rho)</td>
<td>1000</td>
<td>kg/m³</td>
<td>Mass density of fluid</td>
</tr>
<tr>
<td>(f_{CM})</td>
<td>0.65</td>
<td>-</td>
<td>Clausius-Mossotti factor</td>
</tr>
<tr>
<td>(\lambda)</td>
<td>100</td>
<td>nm</td>
<td>Debye length</td>
</tr>
<tr>
<td>(D_f)</td>
<td>(1.03 \times 10^{-10})</td>
<td>m²/s</td>
<td>Diffusivity of electrolyte</td>
</tr>
<tr>
<td>(f)</td>
<td>1</td>
<td>KHz</td>
<td>Frequency of AC signal</td>
</tr>
</tbody>
</table>

The depth averaged analysis done in this thesis has an assumption that the channel depth is much smaller than the channel width. Also, the model used in this chapter cannot predict the electric field leakage through the top and bottom walls of the channel (i.e. through the PDMS and glass substrates at the top and bottom sides respectively). So the model presented has an assumption of electrically insulating conditions on the top and bottom surfaces of microchannel. The model used in this thesis does not consider the convective terms in energy equation (chapter two) and nonlinear terms in flow equation (chapter two and three) due to the very small Peclet and Reynolds numbers. So our future goal would be to develop a complete model that can overcome these limitations.
3.4 Results and Discussion

In this section first we will discuss the numerical (both regular 2D and 2D depth averaged model) and experimental comparison of ICEO effects at the constriction region. Then we will explain the effects of electric field leakage and how the electroosmosis and electrophoresis trajectories are affected by the ICEO effects. Then we will discuss the effects due to electric field by changing the AC to DC voltage ration, followed by the effect of relative permittivity of PDMS substrate on particle trajectories. At last we will study the effects of the constriction width on the particle streaklines (35 µm and 60 µm). We will also present how the induced zeta potential is varies for different conditions.
FIG. 3.3. The numerical and experimental images of ICEO effect on particle trajectories at the channel entrance of 35 µm wide constriction channel at 10 V DC and 200 V AC voltage: (A) Numerically predicted particle streaklines obtained from the regular 2D modelling, (B) Particle streaklines from depth averaged modelling, (C) Snapshot of particles trapped inside the ICEO vortices after a few seconds of supplying the electric field, (D) Superimposed image showing particle streaklines. The block arrow shows the direction of fluid and particle motion. The scale bar represents 50 µm.

Fig. 3.3 shows the numerical and experimental images of particles streaklines obtained at a constant DC voltage of 10 V and AC voltage of 200V for the 35 µm channel. Fig. 3.3A is the numerically obtained particle streak line from the pure 2D modelling. Fig. 3.3B is obtained from the depth averaged numerical modelling. Fig3.3C and Fig3.3D are the experimentally obtained snapshots and superimposed images of particle streaklines. We observe two counter rotating circulations on either side at the entrance of microchannel. The direction of rotation is shown with red arrows in Fig. 3.3C. The direction of rotations
of these two circulations are opposite in direction of the electrothermal flow (See Fig. 2.3D). It is observed that the particles are trapped inside the two circulations. The particles also experience positive DEP and hence some of the particles accumulate near the corners. The regular 2D model (see Fig. 3.3A) predicts a much larger and a stronger circulation near the entrance. The vortex is much bigger in size and its full size is not visible in the figure. This size of the circulation is not comparable with the superimposed image obtained (Fig 3.3D). But the numerically predicted depth averaged model has a vortex size much comparable to the experimental image. The location and size predicted by this model is fairly accurate. It can be concluded that the pure 2D model cannot be used to predict the ICEO phenomenon and hence we use only the depth averaged model for the numerical simulations results in the following sections.
FIG. 3.4. Effects of electric field leakage on fluid and electrophoresis velocity on the flow field, induced charge for 35 µm wide constriction channel at 10 V DC and 200 V AC voltage: (A) Electric field lines inside the fluid (blue lines) and PDMS (red line), (B) streamlines and velocity contours of only the fluid flow, (C) streaklines and velocity contours of electrokinetic flow of particles, (D) induced zeta potential (due to both DC and AC electric field leakage) along the surface of the constriction. The zeta potential is plotted from the entrance at reservoir to the end of constriction. The color bar corresponds to the value of velocity contour plots of (B) and (C).

In order to understand the mechanism of the formation of the ICEO circulations, a numerical study of the electric field leakage, fluid and electrophoresis velocities, and the induced zeta potential values are plotted near the reservoir - constriction region. Fig. 3.4A shows the electric field lines inside the fluid (denoted by blue lines) and PDMS (denoted
by red lines) domains. The electric field lines leaked into the PDMS follow a path of least resistance. The electric field lines that rejoin the fluid domain will polarize the PDMS surface at that point and thus induces a zeta potential on that surface. This leakage causes an ejection flow at the reservoir and constriction (See Fig. 1.3) and that results in the formation of circulations. When only the fluid velocities are plotted (Fig 3.4B) we can observe that the fluid seems to be undisturbed and follow a smooth electroosmotic flow. Any vortex that might be created due to ICEO has been overwhelmed by the strong electroosmotic flow (maximum velocity of 1.7 mm/s). But when the particle’s electrokinetic streaklines plotted (in Fig 3.4C), the vortex is prominently visible. It can also be observed that the electrokinetic velocity magnitude is much smaller when compared to the fluid velocity (maximum magnitude of 0.5 mm/s). So the particles are moving in a much smaller velocity than the fluid flow. Hence the particles experience the ICEO effects for a longer period of time and a strong vortex is formed. When the particle’s DEP velocity is added, the final streaklines of particles is obtained (See Fig. 3.3B). It can be understood from these plots that if particles move slow then the ICEO effects become more prominent due to the fact that ICEO vortices have smaller magnitude in velocity. So as the particle’s zeta potential approaches the wall equilibrium potential, the particles experience stronger ICEO. Fig 3.4D is a plot of induces zeta potential along the length of constriction. The induced potential is plotted along the arc length starting from the entrance of constriction near the reservoir to the end of constriction. The induced zeta potential varies from negative values to positive values along the arc length, suggesting that the PDMS surfaces experiences polarization due electric field leakage. The first polarization occurs near the
fillet curve at the reservoir - constriction interface and the second polarization occurs at the end of constriction near the constriction – microchannel interface. Due to this polarization we find vortices both at the entrance of constriction and at its exit (not shown in the images). The vortices formed due to ICEO depends on the magnitude of zeta potential induced. This value increases either with increase in electric field, with the permittivity of PDMS or with the sharpness of the corner. The effects of these parameters will be discussed in the following sections. Though the ICEO vortex becomes more prominent when the particles are moving slowly, it should be noted that the induced zeta potential does not depend on the particle’s electrokinetic mobility.
FIG. 3.5. Comparison of numerically predicted and experimentally obtained particle streaklines for the 35 µm wide constriction channel at 10 V DC voltage and varying AC voltages of: (A) 50V, (B) 150 V, (C) 250 V. The ICEO effects are very weak at 50V AC and it becomes strong with increasing AC voltage. The fluid and particle flow is from left to right.

Fig. 3.5 shows the dependence of ICEO flow on the AC voltage. For low values of AC voltage such as 50 V (Fig. 3.5A) the Electroosmotic flow is very strong when compared to the ICEO effects. The induced zeta potential near the corner is not large enough for vortices to be formed. We also do not see any DEP effects at this condition. When the AC voltage is increased, both the induced Charge and DEP effects increase. At 150 V AC (Fig.
there is a weak vortex formed at the entrance of the constriction that captures the particles. At 250 V AC voltage (Fig. 3.5C) the vortex grows bigger in size. We find two separate vortices on either side at the entrance. The numerical model is able to predict the size and position of this vortex fairly accurate. At 50V AC, EO is very strong and at 250 V AC ICEO is dominant. There seems to exist a competition between the EO flow and ICEO effects at 150 V AC and hence we are neither able to see a clear stable vortex nor a smooth EO flow. Few of the streaklines in the numerical plots at 150 V and 250 V AC end at the fillet. This shows the particles are experiencing a strong positive DEP. Experimentally we observe that few particles are accumulated in the corner. The size of the two vortices are not equal because the two corners in the constriction are not exactly symmetrical.

**FIG. 3.6.** Plot of numerically predicted maximum induced zeta potential along the surface of reservoir for different AC voltages for the 35 µm wide constriction channel. The DC voltage is kept at a constant value of 10V. At 0 V AC voltage, only the DC voltage is applied and so the induced zeta potential is a little higher than 0 mV. The induced zeta potential increases almost linearly with increase in AC voltage.
With increasing AC voltage, the size of ICEO vortex increases (Fig. 3.5) because the induced zeta potential increases near the reservoir – constriction region. Fig. 3.6 shows a plot of maximum induced zeta potential for the 35 µm wide constriction channel at 10 V DC and varying AC voltages. At 0 V AC, the induced zeta potential is only due to the DC electric field. As the AC voltage is increased the induced zeta potential increases almost linearly. It is known that slip velocity at the fluid wall interface depends linearly on the product of zeta potential and electric field. Since the induced zeta potential is increasing linearly, the slip velocity becomes a function of the second power of electric field. Hence we observe a much bigger increase in size of ICEO vortices with increasing electric field. It should be noted that increasing DC electric field will increase the EO flow simultaneously (due to the equilibrium zeta potential). If the DC electric field is increased a similar effect might not be possible since the equilibrium zeta potential is several times larger than the induced zeta potential.
FIG. 3.7. Variation of ICEO vortices with the relative permittivity of wall material for 35 µm wide constriction channel at 10 V DC and 200V AC. The three images on top row are the particle streaklines at varying relative wall permittivity values of: (A) 2, (B) 4, (C) 6. The circulations become larger and stronger with increasing wall permittivity. (D) Plot of maximum magnitude of induced zeta potentials for different relative wall permittivity. The electric field is kept constant.

When other polymer based materials are used for the manufacture of micro devices instead of PDMS their relative permittivity values would change. This has a direct impact on induced zeta potential and hence on the flow field. The usual relative permittivity values of such materials vary from 2 to 10. Hence we perform a numerical study of varying the relative permittivity values of the substrate and study its effects on particles. The 35 µm
wide constriction channel is used and an electric field of 10 V DC and 200V AC is applied. At a small relative permittivity of 2 (Fig. 3.7A) the particles seem to experience only DEP as some of the streaklines stop at the fillet. The bulk of the particles do not experience any ICEO effects. At a relative permittivity of 4, the vortex are stable and they are formed at the entrance region (similar to Fig. 3.3). At a relative permittivity of 6, the vortex is much stronger and is very large. The variation of the induced zeta potential with permittivity is found to be linear as plotted in Fig. 3.7D. The plot consists for both the 35 and 60 µm wide constriction channels. The 35 µm channel has a higher induced zeta potential for all values of permittivity than the 60 µm channel. This effect is reasonable since the 35 µm will be having a steeper change in cross section of channel than the other. So the electric field leakage would be higher for this channel. Hence the induced zeta potential is higher. So we might expect that the 60 µm channel would be having a weaker vortex when compared to 35 µm channel. In order to verify this phenomenon, we compare the numerically predicted and experimentally obtained images for both these channels at 10 V DC and 250 V AC. Fig. 3.8 shows the study of effects of constriction width on ICEO flow.
FIG. 3.8. Comparison of numerically predicted and experimentally obtained flow field for (A) 35 µm and (B) 60 µm wide constriction channels. (1) numerically predicted particle streaklines, (2) experimentally obtained snapshots after a few second the electric field is supplied, (3) experimentally obtained superimposed particle streaklines. 10 V DC voltage and 250 V AC voltage was applied for this case. The red arrow shows the direction of rotation of the particles.

Fig. 3.8A and Fig. 3.8B shows the particle trajectories and snapshots of both numerically and experimentally obtained for 35 µm and 60 µm channels respectively at 10V DC and 250 V AC voltages. The numerically obtained images (Fig. 3.8.1) suggest that the wider channel has a larger effect of ICEO than the narrow channel. This phenomenon agrees with the experimentally obtained snapshots and superimposed images. More particles are trapped in the 60 µm channel because of the larger and stronger vortex. This phenomenon seems to contradict the plot in Fig. 3.7D. The explanation could be drawn from the velocity contour plots for both the channels. Though the 35 µm channel has a higher induced zeta potential, the narrower constriction region will have a higher electric field which in turn results in stronger EO flow. The wider channel has a much weaker EO
flow. The maximum velocity in 60 µm channel is 0.9 mm/s while for the 35 µm constriction it is 1.4 mm/s. Since the particles are moving slowly in the wider channel they experience much stronger ICEO effects than the particles in narrow channel. Hence the vortex is bigger than the narrow constriction channel. There might be a critical value of constriction width where the induced zeta potential would be too low and the narrow channel might have a stronger vortex. This study is left to be carried out in the future. In the absence of DC voltage, i.e. under the application of a pure AC voltage, the narrower channel will always experience stronger ICEO vortices. The application of DC biased AC electric field makes the ICEO effects more interesting.

3.5 Summary

Thus we performed a combined experimental and numerical study of induced charge effects at the entrance of a microchannel. It was found that at certain electric fields the induced charge causes two counter rotating rolls that could trap particles. We also find that a regular 2D modelling of the effects would over predict the size and strength of ICEO effects. Hence we carry out a depth averaged analysis and use the modified governing equations for depth averaged modelling which had a very good agreement with the experimental data. The effects of electric field leakage on induced zeta potential along the constriction was presented from the numerical model. We used the depth averaged numerical model to show how the fluid streamline and particle’s electrokinetic streaklines behave under the induced charge effects. A parametric study on the AC electric field (both experimental and numerical study) and the electrical permittivity of the solid substrate around the fluid (only numerical study) was done. The ICEO vortices grew stronger with
increasing wall permittivity and AC voltage. The induced zeta potential was found to vary linearly with these two parameters. The effect of constriction width was also studied (both numerical and experimentally) and it was found that the wider channel had a stronger ICEO circulations even though the induced zeta potential in this channel is lesser than the narrow channel. This was due to the stronger EO flow in narrower channel. It was shown that the DC voltage in the electric field supply plays a significant role in ICEO vortex. Similar to the previous chapter, the depth averaged model is good in predicting the ICEO phenomenon and it can solve the model in just a few minutes and thus avoiding the necessity to resort for computationally more expensive 3D modelling.
CHAPTER FOUR

CONCLUSION AND FUTURE WORK

4.1 Conclusion

This thesis provides a combined experimental and numerical study of Joule heating and induced charge effects in electrokinetic flows. Electrokinetics is the preferred method of particle manipulation in microfluidic devices and so a fundamental study is necessary in understanding the characteristics of these two flow phenomena. Importance was given to the electroosmotic fluid entry from reservoir to microchannel since most of the previous works have been limited to inside of a microchannel. A straight microchannel with constrictions at the reservoir–microchannel interface was used in our studies. Also, a two dimensional depth-averaged modeling was created to simulate these two phenomena. This model was able to predict the experimentally obtained results with reasonable accuracy. This depth averaged model has a potential to replace the more computationally expensive three dimensional model.

The first chapter provides the motivation and objective to carry out this thesis. We understand that Joule heating and ICEO are inevitable in electrokinetic flows. So to understand them, a basic background of electric double layer, electroosmosis and electrophoresis is provided. With this background a brief introduction and theory for DEP, Joule heating and ICEO are provided that are necessary in understanding the thesis. Joule heating is the heat generated in the electrolyte when an electric field is passed through it and induced charge is the surface charge resulting from the leakage of electric field through
the substrate of the microchannel. The effects of these two phenomena on electroosmotic flow field at the entrance of microchannel were analyzed in the following chapters.

The governing equations and boundary conditions for Joule heating effects on electrokinetics were provided in chapter two. The channel used for performing the experiments and the simulations was explained. It was found that Joule heating produces two counter rotating circulations at the reservoir-microchannel junction. These electrothermal circulations become stronger with increasing AC electric field because of the increasing Joule heating. Moreover a depth averaged two-dimensional numerical model was developed to understand the electrothermal flow. This model was able to successfully predict the heat dissipated through the top and bottom surfaces of channel. It was found that most of the heat generated inside the channel is rejected through the depth direction because of the higher thermal resistance in the horizontal plane, which cannot be captured by a regular two dimensional model. The effects of Joule heating on electric field, flow field and fluid properties were explained using the numerical model. Due to high temperature gradients in the horizontal plane the electrothermal body force is strong which causes the vortex. Also the flow field at the exit of microchannel was also observed to have two counter rotating circulations in both the experiment and simulation.

In chapter three, the equations and boundary conditions governing ICEO was explained followed by the experimental methods. Similar to Joule heating, ICEO was found to have two counter rotating circulations at the entrance of microchannel. But the directions of rotation of these circulations were opposite to those of electrothermal flows. It was also shown that a regular 2D model would significantly over predict the size of ICEO
circulations. So we used a depth averaged analysis and its prediction had a fairly good agreement with the experimentally obtained images. This numerical model was used to understand how the electric field leaks into the substrate. The induced zeta potential along the length of constriction is plotted. The effects of ICEO on the contours and streamlines of fluid and tracing particle velocities were numerically plotted. It was seen that the ICEO circulations were overwhelmed by the strong EO flow and so the fluid streamlines did not experience any visible vortices. However, the streamlines (or more accurately, the particle streaklines) of the tracing particles can exhibit the vortices because their electrokinetic velocity is comparable to the ICEO velocity. The AC electric field increases the ICEO vortex strength due to the increase in induced zeta potential. Similarly it was also numerically seen that as relative permittivity of wall increases, the magnitude of induced zeta potential and the strength of ICEO vortex increases. Finally it was also seen that a wider constriction of the microchannel had a larger ICEO vortex. Though a narrower constriction can induce a larger zeta potential and in turn a stronger ICEO, the EO flow near the constriction also gets amplified that suppresses the ICEO circulations.

4.2 Future work

Joule heating and induced charge phenomenon are very strong at two limiting conditions, i.e. Joule heating is strong at higher electrical conductivities of electrolyte and ICEO is strong at very low conductivities. In the future we would like to study both the phenomena at conditions in between these two limiting cases. It would be interesting to see how these two phenomena behave and there could be a possibility where both these effects can possibly cancel out each other and the flow field might be undisturbed like a pure EO
flow. It would also be interesting to find if there exists any condition where these two phenomena could co-exist with each other. A combined model solving for the appropriate boundary conditions would be needed to fully understand these two effects. We would like to build a depth averaged two-dimensional model for the same, if possible, due to its lesser computational requirements. This model is anticipated to be a powerful tool in predicting Joule heating and ICEO effects on electroosmotic flows in microfluidic devices at any user defined conditions.
APPENDIX
DERIVATION OF DEPTH AVERAGED GOVERNING EQUATIONS

The three governing equations, electric field, energy and flow field equations are

\[ \nabla \cdot (\sigma \mathbf{E}) = 0 \]  \hspace{1cm} (A-1)

\[ 0 = \nabla \cdot (k \nabla T) + \sigma \langle \mathbf{E}^2 \rangle \]  \hspace{1cm} (A-2)

\[ 0 = -\nabla p + \nabla \cdot (\eta \nabla \mathbf{u}) + \nabla \cdot (\varepsilon \mathbf{E})\mathbf{E} - 0.5\mathbf{E}^2 \nabla \varepsilon \]  \hspace{1cm} (A-3)

\[ \nabla \cdot \mathbf{u} = 0 \]  \hspace{1cm} (A-4)

Here the electric field \( \mathbf{E} \) is represented as \( \mathbf{E} = -\nabla \phi \). The governing equations have the following boundary conditions at the top and bottom surfaces of the microchannel.

For the electric field equation,

\[ \left( \sigma \frac{\partial \phi}{\partial z} \right) = 0 \]  \hspace{1cm} (A-5)

For Energy equation,

\[ \frac{\partial T}{\partial z} = -\frac{1}{k} \frac{(T-T_\infty)}{R_{us}} \] at the top surface. \hspace{1cm} (A-6a)

\[ \frac{\partial T}{\partial z} = \frac{1}{k} \frac{(T-T_\infty)}{R_{is}} \] at the bottom surface. \hspace{1cm} (A-6b)

For the flow equation, (assuming that the slip velocity is same for both the top and bottom surface at any location \((x,y)\))

\[ \mathbf{u} = -\varepsilon \frac{\zeta_w \mathbf{E}}{\eta} \text{ and } \mathbf{w} = 0 \]  \hspace{1cm} (A-7)
To carry out the depth averaged asymptotic analysis we follow the method of Lin et al. [84] the three governing are non-dimensionalized using following scales for the respective variables,

\[
[x, y] = H; \quad [z] = d; \quad [\phi] = \phi_0 = E_0 H; \quad [\sigma] = \sigma_\infty; \quad [\varepsilon] = \varepsilon_\infty
\]

\[
[u, v] = U_{ev} = \varepsilon_\infty E_0^2 d^2 / \eta_\infty H; \quad [w] = U_{ev} d / H; \quad [T] = T_\infty; \quad [\eta] = \eta_\infty
\]

Here \( H \) is half the width of channel (in \( y \) –direction) and \( d \) is half the depth of channel (in \( z \)-direction). The reference electric field \( E_0 \) is taken as the value of electric field applied inside the channel at 20V DC and 400 V AC. \( \sigma_\infty, \varepsilon_\infty \) and \( \eta_\infty \) are the reference electrical conductivity, permittivity and viscosity of electrolyte at room temperature. The reference for horizontal value of velocity is taken as \( U_{ev} \) such that the viscous forces balance the body force in Eq. (A-3) [84,93-95]. However the choice of this velocity scale does not affect our analysis. The reference velocity along \( z \) direction is calculated from the continuity equation. The reference value of pressure is also set by maintaining the same order as that of the viscous forces. Room temperature, \( T_\infty \) is chosen as the reference for \( T \).

The shallowness of the channel is defined by parameter, \( \delta = d / H \). The horizontal plane gradient is denoted as \( \nabla_H \) (gradients along \( x \) and \( y \) directions only).

Using these scales, the dimensionless governing equations are,

\[
\delta^2 \nabla_H \cdot (\sigma \nabla_H \phi) + \frac{\partial}{\partial z} \left( \sigma \frac{\partial \phi}{\partial z} \right) = 0 \quad (A-8)
\]

\[
0 = \delta^2 \nabla_H^2 T + \frac{\partial^2 T}{\partial z^2} + Z \sigma \left( \nabla_H \phi \nabla_H \phi + \frac{1}{\delta^2} \left( \frac{\partial \phi}{\partial z} \right)^2 \right) \quad (A-9)
\]
\[ 0 = -\nabla_H p + \delta^2 \nabla_H \cdot (\eta \nabla_H \mathbf{u}) + \frac{\partial}{\partial z} \left( \eta \frac{\partial u}{\partial z} \right) + \left( \nabla_H \cdot \left( \varepsilon \nabla_H \phi \right) + \frac{1}{\delta^2} \frac{\partial}{\partial z} \left( \varepsilon \frac{\partial \phi}{\partial z} \right) \right) \nabla_H \phi - \]

\[ \frac{1}{2} \left( \nabla_H \phi \nabla_H \phi + \frac{1}{\delta^2} \left( \frac{\partial \phi}{\partial z} \right)^2 \right) \nabla \varepsilon \tag{A-10a} \]

\[ 0 = -\frac{\partial p}{\partial z} + \delta^4 \nabla_H \cdot (\eta \nabla_H \mathbf{w}) + \delta^2 \frac{\partial}{\partial z} \left( \eta \frac{\partial w}{\partial z} \right) + \left( \nabla_H \cdot \left( \varepsilon \nabla_H \phi \right) + \frac{1}{\delta^2} \frac{\partial}{\partial z} \left( \varepsilon \frac{\partial \phi}{\partial z} \right) \right) \frac{\partial \phi}{\partial z} - \]

\[ \frac{1}{2} \left( \nabla_H \phi \nabla_H \phi + \frac{1}{\delta^2} \left( \frac{\partial \phi}{\partial z} \right)^2 \right) \frac{\partial \varepsilon}{\partial z} \tag{A-10b} \]

\[ \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} = 0 \tag{A-11} \]

Here \( Z = \frac{\sigma_\infty d^2 \phi_0^2}{H^2 k T_\infty} \) is a dimensionless constant. Eq. (A-10a) is the dimensionless in the horizontal plane (x and y) and Eq. (A-10b) is the dimensionless along z direction.

On carrying out the dimensionless analysis for (A-5) to (A-7) we obtain,

\[ \frac{\partial \phi}{\partial z} = 0 \text{ at } z = \pm 1 \tag{A-12} \]

\[ \frac{\partial T}{\partial z} = -\frac{d}{k R_{us}} (T - 1) = q_u \text{ at } z = +1 \tag{A-13} \]

\[ \frac{\partial T}{\partial z} = \frac{d}{k R_{ls}} (T - 1) = q_l \text{ at } z = -1 \tag{A-14} \]

\[ \mathbf{u} = \mathbf{u}_{e_0} = \left( \frac{\xi_u H}{E_0 d^2} \right) \frac{\varepsilon}{\eta} \nabla_H \phi \text{ at } z = \pm 1 \tag{A-15} \]

Asymptotic analysis is performed for Eq. (A-8) to Eq. (A-11) assuming \( \delta \ll 1 \). The variables are expanded as,

\[ f = f_0 + \delta f_1 + \delta^2 f_2 + \cdots \tag{A-16} \]
Our goal is to perform a two dimensional depth-averaged analysis up to the second order in $\delta$ (first order in $\delta$ for electric field). In our analysis we find that the variation of fluid properties such as $\sigma$, $\varepsilon$ and $\eta$ along $z$ direction to be small when compared to the variation along the horizontal plane. So we consider these properties to be a function of $(x, y)$. This analysis also avoids solving for the nonlinear equations. The depth averaged function for a variable can be defined as,

$$\bar{f} = \frac{1}{2} \int_{-1}^{1} f \, dz$$  \hspace{1cm} (A-17)

**1. Electric field equation**

Performing a $\delta^0$ order balance for Eq. (A-8):

$$\frac{\partial}{\partial z} \left( \sigma \frac{\partial \phi_0}{\partial z} \right) = 0$$  \hspace{1cm} (A-18)

Considering $\sigma = \sigma(x, y)$ only,

$$\frac{\partial}{\partial z} \left( \frac{\partial \phi_0}{\partial z} \right) = 0$$  \hspace{1cm} (A-19)

Integrating Eq. (A-19) and applying the boundary conditions on top and bottom surfaces of the channel,

At $z = \pm 1$, $\left( \frac{\partial \phi_0}{\partial z} \right) = 0$  \hspace{1cm} (A-20)

Hence Eq. (A-19) becomes, $\left( \frac{\partial \phi_0}{\partial z} \right) = 0$  \hspace{1cm} (A-21)

Thus $\phi_0 = \phi_0(x, y)$ only.
Performing a $\delta^1$ balance order on Eq. (A-8),

$$\frac{\partial}{\partial z} \left( \sigma \frac{\partial \phi_1}{\partial z} \right) = 0 \quad (A-22)$$

Integrating Eq. (A-22) and applying the boundary conditions for $\phi_1$ we obtain

$$\left( \frac{\partial \phi_1}{\partial z} \right) = 0 \quad (A-23)$$

Hence, $\phi_1 = \phi_1(x, y)$ only.

Similarly for a $\delta^2$ order balance of Eq. (A-8):

$$\nabla_H \cdot (\sigma \nabla_H \phi_0) + \frac{\partial}{\partial z} \left( \sigma \frac{\partial \phi_2}{\partial z} \right) = 0 \quad (A-24)$$

Depth averaging the above equation and applying the boundary conditions for $\phi_2$

$$\nabla_H \cdot (\sigma \nabla_H \phi_0) = 0 \quad (A-25)$$

From Eq. (A-24) and Eq. (A-25), we obtain

$$\left( \frac{\partial \phi_2}{\partial z} \right) = 0 \quad (A-26)$$

For $\delta^3$ order balance of Eq. (A-8):

$$\nabla_H \cdot (\sigma \nabla_H \phi_1) + \frac{\partial}{\partial z} \left( \sigma \frac{\partial \phi_3}{\partial z} \right) = 0 \quad (A-27)$$

Depth averaging Eq. (A-27) and applying boundary condition for $\phi_3$,

$$\nabla_H \cdot (\sigma \nabla_H \phi_1) = 0 \quad (A-28)$$
To obtain the depth averaged the equation in electric field we perform (A-25) + $\delta \times$ (A-28) and obtain the fooling depth averaged equation

$$\nabla_H \cdot (\sigma \nabla_H \bar{\phi}) = 0$$

(A-29)

2. Energy Equation

Performing a $\delta^0$ order balance for Eq. (A-9):

$$0 = \frac{\partial^2 T_0}{\partial z^2} + Z \sigma \nabla_H \phi_0 \nabla_H \phi_0$$

(A-30)

Depth averaging the above equation,

$$0 = \frac{1}{2} \int_{-1}^{1} \frac{\partial^2 T_0}{\partial z^2} + \frac{1}{2} Z \int_{-1}^{1} \sigma \nabla_H \phi_0 \nabla_H \phi_0$$

(A-31)

$$0 = \frac{1}{2} \left[ \frac{\partial T_0}{\partial z} \right]_{z=-1}^{1} + Z \sigma \nabla_H \phi_0 \nabla_H \phi_0$$

(A-32)

$$0 = \frac{\bar{q}_{u0} - \bar{q}_{l0}}{2} + Z A$$

(A-33)

Here $A = \sigma \nabla_H \phi_0 \nabla_H \phi_0$ can be considered as a constant for this equation since $\phi_0$ and $\sigma$ are independent of $z$ direction.

Performing a $\delta^1$ order balance for Eq. (A-9):

$$0 = \frac{\partial^2 T_1}{\partial z^2} + Z(\sigma \nabla_H \phi_0 \nabla_H \phi_1 + \sigma \nabla_H \phi_1 \nabla_H \phi_0)$$

(A-34)

Since $\phi_1$ is also independent of $z$ direction, Eq. (A-34) can be written as,

$$0 = \frac{\partial^2 T_1}{\partial z^2} + Z B$$

(A-35)
Here $B = \sigma \nabla H \phi_0 \nabla H \phi_1 + \sigma \nabla H \phi_1 \nabla H \phi_0$ \hfill (A-36)

Depth averaging Eq. (A-35) and following the steps in Eq. (A-31) to Eq. (A-33), we obtain,

$$0 = \frac{q_{u_1} - q_{l_1}}{2} + ZB$$ \hfill (A-37)

Performing a $\delta^2$ order balance for Eq. (A-9):

$$0 = \nabla_H^2 T_0 + \frac{\partial^2 T_2}{\partial z^2} + Z(\sigma \nabla H \phi_0 \nabla H \phi_2 + \sigma \nabla H \phi_2 \nabla H \phi_0 + \sigma \nabla H \phi_1 \nabla H \phi_1)$$ \hfill (A-38)

The six terms inside the brackets in Eq. (A-38) are again independent of $z$ direction. So Eq. (A-38) can be written as,

$$0 = \nabla_H^2 T_0 + \frac{\partial^2 T_2}{\partial z^2} + ZC$$ \hfill (A-39)

Here $C = \sigma \nabla H \phi_0 \nabla H \phi_2 + \sigma \nabla H \phi_2 \nabla H \phi_0 + \sigma \nabla H \phi_1 \nabla H \phi_1$ \hfill (A-40)

Depth averaging Eq. (A-39) we obtain,

$$0 = \nabla_H^2 \bar{T}_0 + \frac{q_{u_2} - q_{l_2}}{2} + ZC$$ \hfill (A-41)

Thus the final depth averaged energy equation is obtained by (A-33) $+ \delta \times (A-37) + \delta^2 \times$ (A-41)

$$0 = \delta^2 \nabla_H^2 \bar{T}_0 + \frac{q_{u_0} - q_{l_0}}{2} + \delta \frac{q_{u_1} - q_{l_1}}{2} + \delta^2 \frac{q_{u_2} - q_{l_2}}{2} + Z(A + \delta B + \delta^2 C)$$ \hfill (A-42)

The simplification of Eq. (A-42) leads to

$$0 = \delta^2 \nabla_H^2 \bar{T} + \frac{q_{u} - q_{l}}{2} + Z(\sigma \nabla H \bar{\phi} \nabla H \bar{\phi})$$ \hfill (A-43)
On arriving at the Eq. (A-43) we have arbitrarily added or dropped terms of $o(\delta^2)$ without losing the asymptotic consistency [84].

3. Flow equation

Performing a $\delta^0$ order balance for Eq. (A-10) and Eq. (A-11) we obtain,

$$0 = -\nabla_H p_0 + \frac{\partial}{\partial z} \left( \eta \frac{\partial u_0}{\partial z} \right) + \nabla_H \cdot (\epsilon \nabla_H \phi_0) \nabla_H \phi_0 - \frac{1}{2} (\nabla_H \phi_0 \cdot \nabla_H \phi_0) \nabla_H \epsilon \quad (A-44)$$

$$0 = -\frac{\partial p_0}{\partial z} \quad (A-45)$$

$$\nabla_H \cdot \mathbf{u}_0 + \frac{\partial w_0}{\partial z} = 0 \quad (A-46)$$

As mentioned earlier, we consider $\eta = \eta(x, y)$ only and $\epsilon = \epsilon(x, y)$ only. So while performing order analysis in Eq. (A-10a) and (A-10b) the gradients of $\phi_0, \phi_1, \phi_2, \eta$ and $\epsilon$ with respect to $z$ will be zero. From Eq. (A-45), $p_0$ is not a function of $z$. That is $p_0 = p_0(x, y)$.

Rearranging Eq. (A-44),

$$\eta \frac{\partial}{\partial z} \left( \frac{\partial w_0}{\partial z} \right) = \nabla_H p_0 - \nabla_H \cdot (\epsilon \nabla_H \phi_0) \nabla_H \phi_0 + \frac{1}{2} (\nabla_H \phi_0 \cdot \nabla_H \phi_0) \nabla_H \epsilon \quad (A-47)$$

The second and third terms on the right hand side of the Eq. (A-47) are all independent of $z$.

So

$$D = -\nabla_H \cdot (\epsilon \nabla_H \phi_0) \nabla_H \phi_0 + \frac{1}{2} (\nabla_H \phi_0 \cdot \nabla_H \phi_0) \nabla_H \epsilon.$$ 

Integrating Eq. (A-47) we obtain,
\[ \mathbf{u}_0 = \left( \frac{\nu H p_0 - D}{\eta} \right) \frac{z^2}{2} + c_1 z + c_2 \]  
(A-48)

Here \( c_1 \) and \( c_2 \) are constants of integration. Applying the boundary conditions \( \mathbf{u}_0 = \mathbf{u}_{eo} \) at \( z = \pm 1 \) to Eq. (A-48) and simplifying,

\[ \mathbf{u}_0 = \left( \frac{\nu H p_0 - D}{\eta} \right) \left( \frac{z^2}{2} - \frac{1}{2} \right) + \mathbf{u}_{eo} \]  
(A-49)

Depth averaging Eq. (A-49)

\[ \overline{\mathbf{u}}_0 = \left( \frac{\nu H p_0 - D}{\eta} \right) \left( -\frac{1}{3} \right) + \mathbf{u}_{eo} \]  
(A-50)

Here replacing the first term on right hand side of Eq. (A-50) with,

\[ \overline{\mathbf{U}}_0 = \left( \frac{\nu H p_0 - D}{\eta} \right) \left( -\frac{1}{3} \right) \]  
(A-51)

Substituting Eq. (A-50) and Eq. (A-51) in Eq. (A-49),

\[ \mathbf{u}_0 = \overline{\mathbf{u}}_0 + \overline{\mathbf{U}}_0 \left( \frac{1}{2} - \frac{3z^2}{2} \right) \]  
(A-52)

Depth averaging Eq. (A-46), and applying the boundary conditions for \( \mathbf{w}_0 \),

\[ \nabla_H \cdot \overline{\mathbf{u}}_0 + \frac{1}{2} \int_{-1}^{1} \frac{\partial \mathbf{w}_0}{\partial z} = 0 \]  
(A-53)

\[ \nabla_H \cdot \overline{\mathbf{u}}_0 = 0 \]  
(A-54)

To calculate \( \mathbf{w}_0 \) we integrate Eq. (A-46) and substitute Eq. (A-52),

\[ \mathbf{w}_0 = \nabla_H \cdot \overline{\mathbf{U}}_0 \left( \frac{z^3}{2} - \frac{z}{2} \right) \]  
(A-55)

Performing a \( \delta^4 \) order balance for Eq. (A-10) and Eq. (A-11)
\[ 0 = -\nabla_H p_1 + \eta \frac{\partial}{\partial z} \left( \frac{\partial u_1}{\partial z} \right) + \nabla_H \cdot (\epsilon \nabla_H \phi_0) \nabla_H \phi_1 + \frac{1}{2} (\nabla_H \phi_0 \cdot \nabla_H \phi_1) - \nabla_H \cdot (\epsilon \nabla_H \phi_1) \nabla_H \phi_0 \cdot \nabla_H \phi_1 \]  
\[ \nabla_H \phi_1 + \nabla_H \phi_1 \cdot \nabla_H \phi_0 \nabla_H \varepsilon \]  
\[ (A-56) \]

\[ 0 = -\frac{\partial p_1}{\partial z} \]  
\[ (A-57) \]

\[ \nabla_H \cdot u_1 + \frac{\partial w_1}{\partial z} = 0 \]  
\[ (A-58) \]

From Eq. (A-56), \( p_1 = p_1(x, y) \) only.

The last four terms in the right hand side of the Eq. (A-56) are independent of \( z \). Hence considering,

\[ E = \nabla_H \cdot (\epsilon \nabla_H \phi_0) \nabla_H \phi_1 + \nabla_H \cdot (\epsilon \nabla_H \phi_1) \nabla_H \phi_0 - \frac{1}{2} (\nabla_H \phi_0 \cdot \nabla_H \phi_1 + \nabla_H \phi_1 \cdot \nabla_H \phi_0) \nabla_H \varepsilon \]  
\[ (A-59) \]

Integrating Eq. (A-56) similar to Eq. (A-48) we get,

\[ u_1 = \left( \frac{\nabla_H p_1 - E}{\eta} \right) \frac{z^2}{2} + c_1 z + c_2 \]  
\[ (A-60) \]

Here \( c_1 \) and \( c_2 \) are constants of integration. Applying the boundary conditions \( u_1 = u_{e01} \) at \( z = \pm 1 \) to Eq. (A-60) and simplifying,

\[ \bar{u}_1 = \left( \frac{\nabla_H p_1 - E}{\eta} \right) \left( -\frac{1}{3} \right) + u_{e01} \]  
\[ (A-61) \]

Also depth averaging Eq. (A-58) and applying the boundary conditions for \( w_1 \) at \( z = \pm 1 \), we get

\[ \nabla_H \cdot \bar{u}_1 = 0 \]  
\[ (A-62) \]
Performing a \( \delta^2 \) order balance for Eq. (A-10) and Eq. (A-11)

\[
0 = -\nabla_H p_2 + \nabla_H \cdot (\eta \nabla_H \mathbf{u}_0) + \eta \frac{\partial}{\partial z} \left( \frac{\partial u_2}{\partial z} \right) + \nabla_H \cdot (\varepsilon \nabla_H \phi_0) \nabla_H \phi_2 + \nabla_H \cdot (\varepsilon \nabla_H \phi_2) \nabla_H \phi_0 + \\
\nabla_H \cdot (\varepsilon \nabla_H \phi_1) \nabla_H \phi_1 - \frac{1}{2} (\nabla_H \phi_0 \cdot \nabla_H \phi_2 + \nabla_H \phi_2 \cdot \nabla_H \phi_0 + \nabla_H \phi_1 \cdot \nabla_H \phi_1) \nabla_H \varepsilon
\]  
(A-63)

\[
0 = -\frac{\partial p_2}{\partial z} + \frac{\partial^2 w_0}{\partial z^2} \tag{A-64}
\]

\[
\nabla_H \cdot \mathbf{u}_2 + \frac{\partial w_2}{\partial z} = 0 \tag{A-65}
\]

The last six terms in Eq. (A-63) are independent of \( z \) direction. Hence we can consider

\[
F = \nabla_H \cdot (\varepsilon \nabla_H \phi_0) \nabla_H \phi_2 + \nabla_H \cdot (\varepsilon \nabla_H \phi_2) \nabla_H \phi_0 + \nabla_H \cdot (\varepsilon \nabla_H \phi_1) \nabla_H \phi_1 - \frac{1}{2} (\nabla_H \phi_0 \cdot \\
\nabla_H \phi_2 + \nabla_H \phi_2 \cdot \nabla_H \phi_0 + \nabla_H \phi_1 \cdot \nabla_H \phi_1) \nabla_H \varepsilon
\]
(A-66)

Hence Eq. (A-61) becomes,

\[
0 = -\nabla_H p_2 + \nabla_H \cdot (\eta \nabla_H \mathbf{u}_0) + \eta \frac{\partial}{\partial z} \left( \frac{\partial u_2}{\partial z} \right) + F + \tag{A-67}
\]

Substituting for \( w_0 \) from Eq. (A-55) in Eq. (A-64) and integrating, we get

\[
p_2 = \nabla_H \cdot \overline{\mathbf{U}_0} \left( \frac{3z^2}{2} \right) \tag{A-68}
\]

Depth averaging Eq. (A-68) we obtain,

\[
\overline{p_2} = \nabla_H \cdot \overline{\mathbf{U}_0} \left( \frac{1}{2} \right) \tag{A-69}
\]

From Eq. (A-68) and (A-69), we obtain

\[
p_2 = \overline{p_2} + \nabla_H \cdot \overline{\mathbf{U}_0} \left( \frac{3z^2}{2} - \frac{1}{2} \right) \tag{A-70}
\]
Using Eq. (A-70) and Eq. (A-52) in Eq. (A-67) and integrating it,

\[ \mathbf{u}_2 = \frac{1}{\eta} \left[ (\nabla_H \bar{p}_2 - \nabla_H \cdot (\eta \nabla_H \bar{u}_0) - F) \left( \frac{z^2}{2} - \frac{1}{2} \right) + \nabla_H \left[ \nabla_H \cdot \bar{u}_0 - \frac{z^4}{4} \right] \right] + c_1 z + c_2 \]  

(A-71)

Applying the boundary conditions for \( \mathbf{u}_2 = \mathbf{u}_{eo2} \) at \( z = \pm 1 \) to Eq. (A-71)

\[ \mathbf{u}_2 = \frac{1}{\eta} \left[ (\nabla_H \bar{p}_2 - \nabla_H \cdot (\eta \nabla_H \bar{u}_0) - F) \left( \frac{z^2}{2} - \frac{1}{2} \right) + \nabla_H \left[ \nabla_H \cdot \bar{u}_0 - \frac{z^4}{4} + \frac{1}{8} \right] \right] + \nabla_H \left[ \eta \nabla_H \bar{u}_0 \left( \frac{z^4}{8} - \frac{z^2}{4} + \frac{1}{8} \right) \right] + \mathbf{u}_{eo2} \]  

(A-72)

On depth averaging the above equation, coefficients that are as small as \( \frac{1}{15} \) are dropped as they are small compared to other terms. We obtain,

\[ \bar{u}_2 = \frac{1}{\eta} (\nabla_H \bar{p}_2 - \nabla_H \cdot (\eta \nabla_H \bar{u}_0) - F) \left( -\frac{1}{3} \right) + \mathbf{u}_{eo2} \]  

(A-73)

Hence to get the final depth averaged equation we perform (A-50) + \( \delta \times \text{(A-61)} + \delta^2 \times \text{(A-73)} \) to obtain

\[ \bar{u}_0 + \delta \bar{u}_1 + \delta^2 \bar{u}_2 = \left( -\frac{1}{3} \right) \left[ \frac{\nabla_H \bar{p}_0 - D}{\eta} \right] + \delta \left( \frac{\nabla_H \bar{p}_1 - E}{\eta} \right) + \delta^2 \left( \frac{\nabla_H \bar{p}_2 - \nabla_H \cdot (\eta \nabla_H \bar{u}_0) - F}{\eta} \right) \]  

(A-74)

\[ \mathbf{u}_{eo0} + + \delta \mathbf{u}_{eo1} + \delta^2 \mathbf{u}_{eo2} \]

Rearranging the equation we obtain

\[ 0 = (-\nabla_H \bar{p}_0 - \delta \nabla_H \bar{p}_1 - \delta^2 \nabla_H \bar{p}_2) + \delta^2 \nabla_H \cdot (\eta \nabla_H \bar{u}_0) + (D + \delta E + \delta^2 F) - 3 \left[ (\bar{u}_0 + \delta \bar{u}_1 + \delta^2 \bar{u}_2) - (\mathbf{u}_{eo0} + + \delta \mathbf{u}_{eo1} + \delta^2 \mathbf{u}_{eo2}) \right] \]  

(A-75)
Substituting for $D$, $E$ and $F$ and simplifying Eq. (A-75) we get the depth averaged flow equation as,

$$0 = -\nabla_H \bar{p} + \delta^2 \nabla_H \cdot (\eta \nabla_H \bar{u}) + \nabla_H \cdot (\varepsilon \nabla_H \bar{\phi}) \nabla_H \bar{\phi} - \frac{1}{2} (\nabla_H \bar{\phi} \cdot \nabla_H \bar{\phi}) \nabla_H \varepsilon - 3(\bar{u} - \bar{u}_{eo})$$

(A-76)

On arriving at the Eq. (A-76) we have arbitrarily added or dropped terms of $o(\delta^2)$ without losing the asymptotic consistency. We have also dropped some additional terms (for $\bar{u}_2$) as they are much smaller in magnitude than the other terms [84].

Equations (A-29), (A-43) and (A-76) are the depth averaged non dimensional electric, temperature and flow field equations respectively. The same method can be used for deriving the depth averaged governing equations used in chapter three. The steps used to derive them are straightforward and hence not presented here.
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