ELECTROKINETIC FLOW DIAGNOSTICS USING MICRO PARTICLE IMAGE VELOCIMETRY

By

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The members of the Committee appointed to examine the dissertation of Keisuke Horiuchi find it satisfactory and recommend that it be accepted.

(Chair)

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ELECTROKINETIC FLOW DIAGNOSTICS

USING MICRO PARTICLE IMAGE VELOCIMETRY

Abstract

By Keisuke Horiuchi, Ph.D. Washington State University December 2005

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Electrokinetic based microflows are studied both experimentally and theoretically. Soft lithography technique is used to form microchannels on poly-di-methyl siloxane (PDMS), while a glass slide or thin PDMS layer is used to cover the microchannels. For microflow quantification, an in-house micro scale particle image velocimetry (μ PIV) system is developed from an existing PIV system.

The first part of the experimental work focuses on quantifying electrokinetic microflows in trapezoidal microchannels. In this investigation, pressure, electroosmotic, and mixed electroosmotic-pressure driven cases are considered. Experimental results obtained from μ PIV are compared with three-dimensional numerical models; the results show an excellent agreement between experimental work and simulation. Trapezoidal microchannels provide tapered-cosine velocity profiles if there is pressure gradient in the streamwise direction. The experimental results verify that velocity distribution in mixed flow can be decomposed into pressure and electroosmotic driven components.

In the second part of the experimental work, a field-effect transistor, which locally modifies the surface charge condition, is developed to control flow in microfluidic chips. By applying a gate voltage to one side of the wall, zeta potential on the controlled surface is altered which results in a secondary electroosmotic flow in the lateral direction. Flow control is observed both quantitatively and qualitatively at relatively low voltage (in the order of 10¹ [V]) having leakage current through the interface between PDMS and glass layers. A leakage

capacitance theory is introduced to estimate the zeta potential at the straight channel wall, and the predicted zeta potential agrees with the experimental results.

Finally, a theoretical study is presented to investigate the microflow behavior in mixed non-uniform electroosmotic flow and pressure driven flow due to step change in zeta potential. A biharmonic equation in terms of stream functions is solved with double-sided Laplace transformation, and the explicit analytical expressions for the entire velocity field and pressure distribution in a two-dimensional straight microchannel are found. Possibilities of recirculation and separation of the flow, which leads to hydrodynamic dispersion, band broadening, and deformation of the band shape, are examined.

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Dedication

This thesis is dedicated to my mother and father who provided their emotional and financial support.

CHAPTER 1

BACKGROUND

1-1 : LITERATURE SURVEY

The following review is devoted to a general historical summary of microfluidics, electrokinetics, electroosmotic flow in microgeometries, flow control in microgeometries, and electroosmosis with variable zeta potential. The primary emphasis of this project involves and connects all of the above topics, as will be shown later on in this writing in much greater detail.

Recent developments in microfluidics, a study of flow behavior on a micro scale, are greatly impacting approaches to molecular chemical analysis (Fredrickson et al. 2001), biological sensing (Mabeck et al., 2005), drug delivery (Date et al., 2006), amplification/synthesis of nucleic acids (Peytavi et al., 2005a; Cady et al., 2005), and environmental monitoring (Marle and Greenway, 2005). Microfluidics is one of the emerging research areas in microelectromechanical systems (MEMS) and Lab-on-a-chip devices, carrying not only practical applications in life science (Figeys, 2002; Wang, 2002; Stone et al., 2004) but also bringing attention to unique fluid phenomena in the realm of physics (Koch et al., 2000). In the history of microfluidics, ink-jet print heads initially accelerated the evolution in the respective industry (Siewill et al., 1985). Ink-jet print heads made of a thinfilm and 3-D electroformed nickel nozzle for ink ejection. The ultimate goal for microfluidic system is to integrate within a small size/area functions such as mixing, separation, and dilution in order to give birth to micro-Total Analysis Systems (µ-TAS), since power consumption, quantity of reagents, and required time for analysis are shortened by reducing the size of a chip. Examples of integrated systems include separation by Capillary Electrophoresis (CE) (Siangproh et al., 2005) and followed by Nuclear Magnetic Resonance

(NMR) (Webb, 2005) for analysis or Polymerase Chain Reaction (PCR) (Dolnik and Liu, 2005) for amplification. Such devices are being used in recent years primarily in biomedical experimentations among other areas such as gene analysis for genomic study (Pilarski et al., 2005), proteins (Lionello et al., 2005), drug delivery (Goettsche et al., 2005) and cell studies (Ho et al., 2005). Dealing with small amounts of fluid is particularly a big advantage compared with macro scale devices when the reagents are either very expensive or hazardous. In analytical chemistry, analysis time clearly needs to be small in order to quickly compile huge amounts of data. On a micro scale, the time required for analysis is much shorter (typically in the order of tens of minutes (Peytavi et al., 2005b)) than that of regular scale experiments (normally a few days). Due to high cost performance in terms of sample volume and analysis time, many companies and governmental organizations have increasingly considered the possibility of unveiling microfluidic technologies to the public. Microfluidic technologies involve the miniaturization of microfluidic components, including micropumps and actuators with/without valves, nozzles, and diffusers. Laser and Santiago (2004) conducted a survey which dealt with information regarding micropumps compiled in the last 25 years; they found from the survey results that electroosmotic micropumps are promising in a variety of applications requiring high pressures (in the order of 100kPa). As a valveless and non-mechanical pump, working principle of electroosmotic flow is academically remarkable as well.

Electroosmotic flow is one of the electrokinetic phenomena, which has been studied since the 18th century. Reuss (1809) first demonstrated both electroosmosis and electrophoresis experimentally by showing migration of water through porous clay diaphragms towards the cathode. This is due to the existence of electric double layer (EDL). When a polar liquid and solid are placed in contact with one another, a difference in potential between them results in the formation of thin layers on the liquid side due to either ionization, ion adsorption, or ion absorption. The theory of transporting phenomena based on the double-layer was formally introduced by von Helmholtz (1879). The related electrostatic charge and potential distribution at interfaces was first explicitly modeled by Perrin (1904), according to Overbeek (1952). However, it had long been recognized as an inadequate representation of the phenomena because liquid has diffusive characteristics unlike metals. The theory for such a diffuse double layer was introduced by Guoy (1910) and by Chapman (1913). A more elaborate theory, proposed by Stern (1924), notes that the inner layer of adsorbed ions interacts specifically with the wall. For dilute ionic solutions, Debye and Hückel (1923) approximated the ion energy distribution. Under the condition of having this electric double layer, fluid can be migrated when an external electric field is applied in micro geometries. Because the resultant velocity profile is almost uniform, this unique transporting phenomenon has received a great deal of attention among researchers in recent times. Basic sensor and actuator technologies have contributed advanced fabrication techniques to microfluidic society (Yao et al., 2005); a number of research groups introduced electroosmotic microflows occurring within microstructures of various shapes (Chen and Santiago, 2002; Tripp et al., 2004). As mentioned above, the utilization of the micropump is one of the most important applications.

The combination of pressure driven flow and electroosmotic flow is expected when electroosmotic pumping action is desired. Even if electroosmotic flow is used for other bioanalytical applications aside from micropumps, local pressure gradient may be induced unintentionally (Dutta and Leighton, 2001). The resulting pressure gradient may distort the plug-like velocity distribution and may result in large Taylor dispersion. Thus, it is extremely important to observe flow characteristics when both electroosmosis and Poiseuille flows coexist. Analytical work is available for a cylindrical channel (Maynes and Webb, 2003) and

a rectangular channel (Dutta and Beskok, 2001). The prediction of dispersion effects may be possible by solving their analytical solutions.

On the other hand, in order to maximize or minimize dispersion effects for mixing and drug delivery applications, there are two broad approaches: passive and active flow control. In both ways, the ability to control electroosmotic flow behavior is important, especially in the field of mixing and drug delivery applications. For instance, electroosmotic flow within capillary zone electrophoresis is one of the conventional techniques used to distinguish chemical species because electroosmotic velocity profile is plug-like which is suitable to avoid distortion of moving sample bands (Ghosal, 2004). The passive flow control method uses geometric configuration without applying external energy. An example of passive flow control is the optimized minimal dispersion channel using serpentine channels with partially reduced cross-sections (Mohammadi et al., 2000). Mohammadi et al. utilized CAD to design electroosmotic microfluidic channels with a curvature and realized minimal geometrical dispersion at 90- and 180- degrees. In contrast, active flow control utilizes external energy, resulting in stronger and variable flow control. Electroosmotic flow can be controlled electronically through a mechanism modeled as an electric circuit (Lee et al. 1990, 1991; Hayes et al., 1993b), as we are already aware that zeta potential can be expressed in terms of electronic capacitances (Hunter, 1981). Recently, local flow control in silicon/glass-base microgeometries has been developed by a number of research groups and entities (Karnik et al., 2005; van der Wouden et al., 2005).

The field-effect flow control method obviously changes zeta potential. Nonuniform zeta potential may also exist on the wall due to fabrication defects. If the velocity distribution is mathematically expressed, it is quite useful to predict some of the dispersion effects prior to the experiments. The first theoretical approach of nonuniform zeta potential on electroosmotic flow was done within capillaries (Anderson and Idol, 1985). Since then, a

couple of groups have extended analytical solutions to velocity field in a rectangular channel containing wavy zeta potential (Adjari, 1995, 1996; Long et al., 1999). Recent development showed that sudden changes in either zeta potential or cross section causes distortion of plug-like velocity even in hydraulically fully developed region (Christopher and Davis, 2004). There are a number of numerical works for continuously changing boundary conditions (Dutta et al., 2002; Ren and Li, 2001).

1-2 : RESEARCH OBJECTIVES

The main goal of this project is to characterize the combined electroosmotic and Poiseuille flow along a microchannel in which the zeta potential on the wall is locally modulated. The mixed electroosmotic and pressure driven flow is very often seen in the application of micropump due to placement of valves, nonuniform zeta potential, etc. On the other hand, modification of the zeta potential is also a common topic because it happens either unintentionally due to uncertainty of microfabrication techniques or intentionally by coating or activating the wall from outside for local flow control.

To start with, a poly-di-methyl siloxane microchannel possessing embedded electrodes is fabricated. This transparent channel is suitable for the micro particle image velocimetry detection system. Here, deposited electrodes guide electrons to generate a desired electric field between the anode and the cathode so that the magnitude of electroosmotic flow can be controlled. The resulting polymeric channel has a trapezoidal cross section which is common in the society of microfabrication, but has yet to be visualized.

Second, micro particle image velocimetry (μ PIV) is developed in order to observe velocity field in the fabricated channels. All optical components are built separately and assembled based on the knowledge of conventional macro PIV. The original post processing data analysis software was developed specifically for our study of background noise

reduction, ensemble averaging, and subtraction of the electrophoretic velocity of seeding particles.

To analyze the effects of the mixed electroosmotic and pressure driven flow, a simple straight trapezoidal channel without having modulation of zeta potential is explored experimentally. Most analytical or numerical studies are conventionally based on either circular or rectangular microchannels (Probstein, 1994; Newman, 2004), but in reality, micromachined channels are typically formed with trapezoidal cross-sections (Bianchi et al, 2001; Qu et al., 2000). Flow quantification in silicon based trapezoidal channels has been revealed to the public in recent years (Wu and Cheng, 2005; Hao et al., 2005), although most microchannels have been approximated as rectangular channels for quite a long time. It is remarkable to note that the microchannels made purely by PDMS have never been tested by other people using μ PIV, because researchers normally use cover glass due to limited small depth of field. Long-working distance microscopes are utilized to address this problem and show the channel material dependence of the electroosmotic flow.

Next, a field-effect control in a straight channel as well as a T-channel is investigated. By applying another voltage (gate voltage) from outside of the channel, the fluid flow behavior can be locally controlled for many applications such as minimizing sample dispersion at T-junction in practice (Takhistov et al., 2003). Here, the field-effect-control system with PDMS base capacitor is fabricated. This capacitor shows leakage current. The analysis of leakage current has never been rigorously performed by researchers as of yet, so theoretical prediction of change in zeta potential based on the equivalent electric circuit is developed. Nobody has considered polymeric material as the boundary for altering zeta potential. On top of that, only straight and/or curved channel configuration has been studied so far by other researchers (Lee et al., 2004b). In practice, in order to discriminate and extract a specific sample from a liquid buffer, a branch channel is often used. Thus, local flow control at the nearby junction of the two channels should be investigated. Use of PIV and even measurement of velocity profile inside a field-effect flow system has also not been attempted by any entities so far. The proposed field-effect control flow is observed experimentally and verified with our prediction.

Lastly, the combination of the two above mentioned scenarios is considered analytically. In other words, mixed electroosmotic and pressure-driven flow passes through the section in which the zeta potential is locally modulated in this case. Treatment of the step change in zeta potential is mathematically challenging in solving the biharmonic equation based on Navier-Stokes equation in terms of stream functions. By using the step function and the double-sided Laplace transformation technique, explicit analytical solutions of both velocity and pressure fields in a straight channel are found. The distorted shape of sample inside this microchannel is predicted by changing the relative values of zeta potential and net flow rate.

More rigorous motivations are reemphasized at the introduction of each chapter.

1-3 : SUMMARY OF DOCUMENT

In Chapter 2, the fundamentals of electroosmotic flow with electric double layers are emphasized. Driving mechanisms of electroosmotic flow and its theoretical aspects are presented. Other electrokinetic phenomena such as electrophoresis, streaming potential, and sedimentation potential are also described. The knowledge of electrophoretic flow will be used when seeding particles dynamics becomes important in the PIV section (Chapter 5 and 6).

Chapter 3 reviews the characteristics of PDMS material as well as metal deposition techniques on PDMS. Understanding of material properties has served to greatly aid the development of optically based detection systems. Because there are not many literatures

discussing a coating of thin film deposition on top of polymeric material, available techniques are summarized in this chapter. Our original recipe is documented at the end of this chapter.

Chapter 4 presents the experimental setup. The basic concept of the PIV algorithm is described first, followed by an explanation of the original hardware and software for the μ PIV system.

Chapter 5 quantifies mixed electroosmotic and pressure driven flow inside a trapezoidal channel. This is one of our main contributions to the scientific community, because nobody has visualized and analyzed the flow inside a trapezoidal channel despite the fact that it is common to see both mixed flow and trapezoidal channel cross sections in practice.

Chapter 6 introduces a novel field-effect control system in order to locally control the flow in microgeometries. As mentioned in the previous section, local flow control can be achieved by changing zeta potential. The leakage current theory has also been investigated in this chapter.

Chapter 7 discusses the theoretical study of step change in zeta potential in mixed electroosmotic and pressure driven flow. This chapter essentially covers potential techniques of application of the material presented in Chapters 5 and 6. Both mixed flow and zeta potential modulation are possible experimentally in previous chapters, so this chapter claims that distortion of a sample such as protein and DNA is caused by the step change zeta potential.

Finally, in Chapter 8, conclusions, suggestions, and recommendations for future work are presented.

1-4 : NOMENCLATURE

A area of capacitance(s)

- c_{∞} concentration in the bulk region
- $C_{\rm EDL}$ capacity of the EDL (diffuse layer)
- C_{wall} capacitance of the wall

 C_{total} total capacitance

- D molecular diffusivity
- D_p denominator (characteristic equation) of ϕ_p
- D_{ψ} denominator (characteristic equation) of ϕ_{ψ}
- \vec{E} applied electric field
- *E* electric field component in the streamwise direction
- F Faraday's constant
- $d_{diff \infty}$ theoretical diameter of a diffraction-limited point source of light
- d_{eff} effective particle diameter
- d_p particle diameter
- e electron charge
- $f^{\#}_{\infty}$ f-number for infinite-corrected optics
- *H* half channel height
- H_h hydraulic diameter

HETP theoretical plate height

- I current
- *K* pressure gradient in far field, K = -dP/dx
- k_B Boltzmann constant
- *L* channel length
- *M* magnification of objective lens

 M_{EO} electroosmotic mobility

- M_{PH} electrophoretic mobility
- NA numerical aperture
- N_P numerator of ϕ_P

 N_{ψ} numerator of ϕ_{ψ}

- *n* refractive index of the immersion medium
- n_0 average number of ions in the buffer

P pressure

R universal gas constant

R resistance

- *s* roots of characteristic equation
- t thickness of the wall between main channel and sub channel
- *T* absolute temperature
- u_{HS} Helmholtz-Smoluchowski velocity
- u_{B} Brownian motion velocity
- u_{PH} electrophoretic velocity

 \vec{V} velocity field

- V_x velocity component in the streamwise direction
- V_{v} velocity component in the cross-stream direction

 $\langle V_x \rangle$ mean velocity

Re Reynolds number, $\text{Re} = u_{HS} D/v$

u streamwise velocity component

 u_{HS} Helmholtz-Smoluchowski velocity, $u_{HS} = -\zeta \varepsilon E_x/\eta$

- u_{PH} Electrophoretic velocity, $ze/3\pi d_P \eta$
- \vec{V} velocity field, $\vec{V} = (u, v)$
- W channel width
- x streamwise coordinate system
- y cross flow coordinate system
- z valence

Greek letters

- δz thickness of the measurement plane
- ε permitivity of the medium, $\varepsilon = \varepsilon_b \varepsilon_0$
- ε_b relative permittivity of the buffer solution
- ε_{w} relative permittivity of the channel material
- ε_0 permittivity of free space
- ζ zeta potential
- ε dielectric constant
- φ external electric potential
- ϕ_{P} near-field pressure in Laplace space
- ϕ_{ψ} near-field stream function in Laplace space
- η dynamic viscosity
- λ_D Debye length
- λ_{ex} excitation wavelength of light in vacuum
- λ_{em} emission wavelength of light in vacuum
- μ_{eo} electroosmotic mobility

- σ electrical conductivity of the buffer fluid
- $\vec{\Omega}$ vorticity
- ω z-component of vorticity
- ξ non-dimensional streamwise distance
- ψ electrokinetic potential
- ψ stream function
- $\vec{\Psi}$ stream function vector
- ρ resistivity
- ρ_{e} electric charge density
- ρ_f fluid density
- *v* kinematic viscosity
- ζ zeta potential

Subscripts

- max maximum value
- ds drain-source
- gs gate- source
- leak value(s) at current leakage area

Superscripts

- * near-field solutions
- variables at upstream (x < 0)
- + variables at downstream $(x \ge 0)$

Symbols

- ^ nondimensional solutions for discontinuous zeta potential variation
- \sim nondimensional solutions for continuous zeta potential variation

CHAPTER 2

DESCRIPTION OF ELECTROKINETICS

2-1: INTRODUCTION

Electrokinetic flow has been in the academic spotlight recently because it does not require pressure head or mechanical valves inside a microchannel. Moreover, the velocity profile is almost uniform across the channel, so it is one of the important methods used to transport samples such as DNA, RNA, and protein with minimum distortion. This chapter deals with *electrokinetic phenomena* which relate an *electric double layer* and an external electric field to a hydrodynamic flow.

The electric double layer is known as the separation of charge that occurs at the interface between solid and liquid (Hunter, 1981). This double layer namely consists of two layers: Stern layer and diffuse layer, in liquid phase. Most surfaces acquire net electric charge when they come in contact with a polar medium via ionization, ion adsorption, or ion dissolution. The charged surface attracts the counter-ions in the solution due to powerful electrostatic forces (10^7 to 10^9 V/cm) (Lyklema et al., 1998) while co-ions are repelled away from the solid. The resultant layer with typical thickness of a few ionic diameters is called the Stern layer, or the stagnant layer. Unlike solid surfaces, liquid has both electric forces and thermal diffusive forces. Thus, outside of the Stern layer, thermal movement of ions takes an active role and forms the diffuse layer that contains both co-ions and counter-ions. Here, the plane separating the Stern layer and the diffuse layer is known as the shear plane, and the electric potential at the shear plane is called *zeta potential* ζ . Such a diffuse double layer theory was developed by Gouy (1910) and Chapman (1913), and uses Poisson's equation expressing the electrokinetic potential distribution inside liquid.

When the electric double layer interacts with the tangential electric field, the

movement of ionic solution occurs near the charged interfaces, also known as electrokinetic phenomena. There are, in principle, four kinds of the electrokinetic effects called electroosmosis, electrophoresis, streaming potential, and sedimentation potential. Each phenomenon is described in the following sections.

2-2 : ELECTROOSMOSIS

Electroosmosis is the movement of a polar medium relative to a stationary charged solid by an applied electric field. Here, the solid refers to either a capillary, microchannel, or porous plug that is charged spontaneously. When an electrolyte comes into contact with a dielectric surface such as PDMS, glass, or acrylic, the surface generally acquires net charges due to ionization, ion adsorption, or ion absorption. These surface charges influence the distribution of counter ions close to the surface, and form an electric double layer (EDL) which adjoins to the surface. The extent of EDL depends on the ion concentration in the electrolytes, and it is normally characterized by the Debye layer thickness (λ). For example, ion concentration of 1 and 100 mM corresponds to Debye length thickness of 10 and 1 nm, respectively. If an external electric field is applied along the channel surface, there will be net movement of electrolytes due to the formation of an electrokinetic body force. In the case of a silicon base channel, a fraction of the surface silanol group (Si[°]OH) results in either net negative potential (SiO[°]) or positive potential (SiOH²⁺) depending on the pH of the buffer solution. As the liquid inside the EDL migrates in response to the field, ions drag the bulk region of liquid with them.

Let us estimate the electroosmotic velocity formed in a rectangular microchannel by a uniform electric field applied along the axis as shown in Fig.2-1. The channel walls contain net negative charges in this particular example, which can be represented in terms of a reallife situation when a glass surface is immersed into water. The charges drawn in the figure indicate net charge, so the electric double layer has net positive charges in Fig.2-1. The thickness of the electric double layer is typically in the order of up to a few tens of nanometers depending on the ionic concentration. When an external electric field is applied in the streamwise direction, positive ions in the electric double layer are mobilized toward the cathode. Due to the viscous drag, movement in the electric double layer results in net movement of ionized fluid in the direction of the electric field. Therefore, the bulk region of the solution moves in the same direction with flat velocity profile whereas the velocity in the electric double layer decreases rapidly due to a sudden drop electrokinetic potential.



Figure 2-1 : Schematic diagram of the electroosmotic flow inside a rectangular microchannel. The origin of the coordinate system is placed at the inlet and the center of the channel. The flow direction is the same as the streamwise electric field when the wall surface is negatively charged.

At steady state, the electrokinetic potential, ψ , can be obtained from the Poisson-Boltzmann equation as (Hunter, 1981)

$$\nabla^2 \psi = -\frac{\rho_e}{\epsilon} = -\frac{e(n^+ z^+ + n^- z^-)}{\epsilon}$$
(2.1)

where ρ_e is the electric charge density and ε is the permittivity of the medium, $e=1.6 \times 10^{-19}$ C is the electron charge, z is the valence, and n is the ion number concentration. For a symmetrical, dilute, and univalent electrolyte $(z^+ = z^- = z)$, the electric charge density can be found as (Probstein, 1994)

$$\rho_e = -2n_o ez \sinh\left(\frac{ez\psi}{k_B T}\right) \tag{2.2}$$

where n_0 is the average number of positive or negative ions in the buffer, k_B is the Boltzmann constant, and T is the absolute temperature. The electroosmotic flow is governed by the modified Navier-Stokes equations as (Probstein, 1994)

$$\rho_f \left\{ \frac{\partial \vec{V}}{\partial t} + \left(\vec{V} \cdot \nabla \right) \vec{V} \right\} = -\nabla P + \eta \nabla^2 \vec{V} + \rho_e \vec{E}$$
(2.3)

where ρ_f is the fluid density, \vec{E} is the applied electric field, P is the pressure, η is the dynamic viscosity, and $\vec{V} = (u, v)$ is the velocity field subjected to no slip and no penetration boundary conditions at the surface. The final term, $\rho_e \vec{E}$, represents electrokinetic body force due to formation of the electric double layer next to the surface. For pure electroosmotic flow at steady state in a wide and shallow microchannel, Eqs.(2.1) and (2.3) are combined and simplified to the next differential equation.

$$0 = \eta \frac{\partial^2 u}{\partial y^2} - \varepsilon \frac{\partial^2 \psi}{\partial y^2} E_x$$
(2.4)

where the inertial term in Eq.(2.3) is negligible in comparison to viscous term due to the fact
that the associated Reynolds number of electroosmotic flow in a microchannel is very small because the field of microfluidics deals with flow in geometries with dimensions of less than 1 mm. By solving this governing equation with boundary conditions (u = 0 and $\psi = \zeta$ at $y = \pm H$), the axial velocity component can be obtained as

$$u = -\frac{\varepsilon E_x}{\eta} (\zeta - \psi) = u_{HS} (1 - \frac{\psi}{\zeta})$$
(2.5)

where electoroosmotic velocity in the bulk region is known as the Helmholtz-Smoluchowski velocity, $u_{HS} = -\zeta \varepsilon E_x / \eta = \text{const}$ (Probstein, 1994). Here, $\varepsilon = 6.9 \times 10^{-10} \text{ C}^2/(\text{J m})$, $\zeta \sim -75 \text{mV/cm}$, and $\mu = 8.95 \times 10^{-4} \text{ kg/(m s)}$ if the electrolyte is DI water. For a straight microchannel, Eqs.(2.1) and Eq.(2.2) yield electrokinetic potential as (Dutta and Beskok, 2001)

$$\Psi = \frac{4k_BT}{ez} \tanh^{-1} \left[\tanh\left(\frac{ez\zeta}{4k_BT}\right) \exp\left\{-e z \sqrt{\frac{8\pi n_0 D}{k_BT}} \left(1 - \left|\frac{y}{D}\right|\right)\right\} \right]$$
(2.6)

A recent study shows that the active region of this electrokinetic potential is extremely small (approximately $\delta_{99} \approx 0.04$ H), so the velocity profile is almost flat except at the region very close to the wall. This plug-like velocity profile is useful for transporting immersed or solvated substances with minimal hydraulic dispersion (Cummings et al., 1999). The main simplifying assumptions used to obtain above equations are:

(a) The fluid is Newtonian, and microflow is laminar.

(b) Fluid properties such as viscosity, permittivity, and conductivity are independent of local and overall electric field strength.

(c) Temperature variation due to Joule heating is negligible, hence fluid properties are assumed to be constant.

(d) Ions are point charges, and ionic diffusion effects are much higher than ionic-convection.

(e) The solvent is continuous.

2-3 : ELECTROPHORESIS

Electrophoresis is the movement of charged ions with attached materials relative to stationary liquid by an applied electric field. Although the electrophoretic phenomenon has been known for over 185 years (Mosher et al., 1992), the usefulness of the phenomenon in the analysis of complex protein mixtures was first introduced by Tiselius in the 1930's (Tiselius, 1940). Since then, the electrophoresis has become an important technique in bioanalytical chemistry, especially for deoxyribonucleic acid (DNA) and protein separation. The experimental setup is very similar to the one used for observation of electroosmosis, except that the channel material is electrically neutral. A wide range of molecules may be naturally charged, including, but not limited to, DNA, ribonucleic acid (RNA), and protein molecules. The charged molecules can be separated depending upon the size, shape, and respective molecular charges under the influence of an electric field. Generally, the smaller and strongly charged molecules move faster than the others. Figure 1-2 shows an example of applications using electrophoresis. Here, two different proteins are separated because each protein has a different isoelectric point (pI) where the net charge becomes zero. In this particular example, pH gradient is required along the channel in order to allow each molecule be charged before applying the electric field.



Figure 2-2: Schematic of protein separation based on electrophoresis. The technique shown in this figure is called *isoelectric focusing* since it uses pH gradient along the channel. Protein A and B have different isoelectric points (pl) in which the net charge becomes zero, so the separation of these two proteins can be achieved by applying electric field.

In order to estimate the electrophoretic velocity, consider a spherical particle of radius r_p , and let the origin of a spherical coordinate system be placed at the center of this particle as shown in Fig.1-3. The electrical force is equal to the Stokes drag on the particle (Probstein, 1994)

$$qE_x = 6\pi\eta u_{PH}r_P \tag{2.7}$$

where q = ez is the net charge between the particle and the concentric spherical double layer of predominantly opposite charge and u_{PH} is the electrophoretic velocity.



Figure 2-3: Electrophoretic motion of a charged spherical particle.

In the figure, λ_D is the Debye length which is defined as the length of the diffuse layer; it is assumed to be large compared with the radius of the particle r_p . The zeta potential ζ_p is calculated as the superposition of two potentials: one arising from a positive charge + q on the particle surface, and the other arising from an opposite charge - q on an imaginary sphere of radius $r_p + \lambda_D$ (Probstein, 1994).

$$\zeta_{P} = \frac{(+q)}{4\pi\varepsilon r_{P}} + \frac{(-q)}{4\pi\varepsilon(r_{P} + \lambda_{D})}$$
(2-8)

From Eqs.(2-7) and (2-8), the electrophoretic velocity in terms of zeta potential is

$$u_{PH} = \frac{2}{3} \frac{\zeta_P \varepsilon E_x}{\eta} \left(1 + \frac{r_P}{\lambda_D} \right)$$
(2-9)

When r_p / λ_D is neglected, Eq.(2-9) becomes the Huckel equation.

2-4 : STREAMING POTENTIAL AND SEDIMENTATION POTENTIAL

The streaming potential and the sedimentation potential are not quite as common in comparison with electroosmosis and electrophoresis among four electrokinetic phenomena. The fundamental difference between the streaming potential and electroosmosis is input and output. Instead of applying the electric field in order to bring forth net movement, liquid is made to flow along a stationary charged surface in order to create the electric field from the accumulation of charge at the downstream in case of the streaming potential. Here, it is noted that the bulk of the moving liquid carries no net charge, so only the movement near the charged wall boundary is important. Because it requires a pressure gradient to force the liquid through, the resultant electric field (electric current) is related to the driving pressure and to the potential in the neighborhood of the wall. In Fig.2-4(a), the movement of ions near the wall surface results in accumulation, which produces a streaming current, Is, and the However, some ions experience conduction/diffusion and/or potential difference. electroosmotic flow through the liquid. Therefore, as indicated in Fig.2-4(b), the conduction current in the reverse direction Ic is generated and balanced with the streaming current Is. When an equilibrium condition is attained, the measured potential difference, Es, is the streaming potential.



Figure 2-4: The mechanism of streaming potential.

The relationship between the sedimentation potential and the electrophoresis is very similar to the one between the streaming potential and the electroosmosis. When charged particles are migrated by external forces such as the force of gravity and/or centrifugal force, a potential difference dubbed "sedimentation potential" is generated. The shear stress near the particle causes the surface current density to flow from the back of the particle to the front (Newman, 2004). This current must then flow through the liquid from the front to the back of the particle as shown in Fig.2-5. Like Fig.2-3, the dotted line and the solid line in Fig.2-5 represent the electric double layer and the particle surface, respectively. The arrows in the figure indicate the current flow. A backflow of opposite ions occurs, and the steady state is eventually established after balancing with induced currents like the conduction current described in Fig.2-4(b). The appreciable sedimentation potential is difficult to observe experimentally, so the sedimentation potential is not often studied in great detail by many researchers unlike the other electrokinetic phenomena.



Figure 2-5: Schematic picture of current flow which generates the sedimentation potential.

CHAPTER 3

FABRICATION AND MATERIALS

3-1: INTRODUCTION

Without successful developments in micromachining technologies, the field of microfluidics would not have come to be. As MEMS technology develops and matures, the interdisciplinary nature of both micromachining techniques and their applications can lead to existing synergies, including Lab-on-a-chip devices. The use of lithographics for patterning and other fundamental microfabrication technologies such as the addition, subtraction, and modification to create miniaturized sensors, actuators, and structures has become really popular since the late 1960's (Kovacs, 1998). These techniques were typically developed with silicon and other semiconductors because they were originally developed for the discrete and integrated electronic circuit industry (Campbell, 1996).

Micromachining can be roughly categorized into "bulk" and "surface" micromachining. Bulk micromachining involves etching, doping, or the carrying out of an oxidation process, while surface micromachining includes patterning and/or the building up of 2-D layers of thin-film. Etching refers to the removal of selected material and is mainly divided into "wet" and "dry" etching. Wet etching is normally chemical etching characterized by the existence of isotropic behavior, (etching occurs in all directions of the material at the same rate). In dry etching, smaller patterns and higher aspect ratio structures can be achieved by physical/chemical etching. An excellent summary of both wet and dry etching processes can be found in a paper by Williams and Muller (1996). Thin film deposition is used for electric contacts, masking, or isolation purposes. MEMS deposition technology can be categorized into two groups: (a) using chemical reaction and (b) using physical reaction. The chemical deposition technique includes chemical vapor deposition

(CVD), electrodeposition (electroplating), epitaxy, and thermal oxidation, while physical vapor deposition (PVD), sputtering, and casting are categorized as physical deposition techniques (Jaeger, 2002).

The patterning of structures is called "photolithography." Photolithography patterning techniques are widely used in surface micromachining processes. The process begins by spin-coating the substrate with a photosensitive material (photoresist). The desired thickness of coating will depend on the resist viscosity as well as the spinning rate. Next, the coated substrate is baked and exposed to ultraviolet (UV) light. After exposure, the photoresist is developed, and the desired pattern is left on the substrate. Here, if the exposed material is etched away by the developer and the unexposed region remains, the material is called a positive resist. Contrarily, if the exposed material is resilient to the developer and the unexposed region is washed away, it is known as a negative resist.

If the procedure of molding and embossing is added, it is then known as "softlithography." This technique employs soft and flexible materials such as polymers and gels. The process makes it possible to reuse the mold many times, which reduces fabrication costs and time (Quake and Scheret, 2000; Becker and Heim, 2000). Some of the advantages of this technique include easy multilayer fabrication, short fabrication time, and cost (Unger et al., 2000). Bonding is also needed if complex 3-D structures are desired via the building up of 2-D layers. There are well-known methods such as anodic bonding, silicon fusion bonding, and plasma bonding. These are described by Kovacs in a short paragraph (Kovacs, 1998).

The choice of material and soft lithographic techniques for fabrication coincides with the desire for inexpensive, efficient prototyping. Poly-Di-Methyl Siloxane (PDMS) is chosen as channel material and used for soft lithography techniques. We developed our own original technique of thin metal deposition on PDMS channel. Our achievements (original recipes) are recorded in the Appendix. The soft lithography is well described by its pioneering group (Xia and Whitesides, 1998a), but there are not many papers which discuss how to deposit metal on polymeric material. Thus, this chapter focuses on two sections: (i) choice of material and (ii) available metal deposition techniques.

Historically, silicon based materials have been the major resources employed for micromachining in MEMS applications simply because most of the fabrication techniques originated from the electronic circuit industry. However, polymeric materials have gained significance nowadays due to issues of cost, ease of disposal, and biocompatibility. To the best of our knowledge, nobody has rigorously studied velocity profiles of the polymeric trapezoidal channel. Additionally, while electroosmotic flow under the existence of the pressure gradient has been studied theoretically in the past, up until recently there were no actual experiments conducted by research groups. Therefore, characterizing the mixed flow will be one of the important tasks in this study. For the purpose of comparison with numerical solutions, a single material (PDMS) is used to form a microchannel.

3-2 : POLY-DI-METHYL SILOXANE

Initially, silicon based materials have been the major resources employed for micromachining in MEMS applications. However, among all available materials in current technology, Poly-Di-Methyl Siloxane (PDMS) has been attracting attention because of its unique advantages (Duffy et al., 1998; Armani, 1999; Becker and Locascio, 2002). The PDMS material is bioand chemically compatible, can be used while performing the soft lithography technique, possesses adjustable stiffness, and is transparent down to 280 nm which is suitable for optical based detection systems (Armani, 1999). Furthermore, polymers are less expensive than glass and silicon. Material properties of PDMS are shown by Becker and Locascio (2002) in their paper, and glass transition temperature is given by Bischoff and Cray (1999). Table 4-1

lists the specification of PDMS manufactured by Dow Corning. The properties include the low glass transition temperature, low surface energy, high permeability to gases, good insulating properties, and very good thermal stability. The softlithography technique in PDMS is straightforward (Xia and Whitesides, 1998a; Chan et al., 1999). It is well known that elastomeric polymers such as PDMS have excellent adhesion to a wide variety of substrate materials (PDMS itself, glass, silicon, silicon oxide, quartz, silicon nitride, polyethylene, polystyrene, and glassy carbon) by plasma oxidation (Becker and Locascio, 2002; McDonald et al., 2000; Chudhury and Whitesides, 1991; Ismagilov et al., 2001; Anderson et al., 2000) based on the nature of the siloxane bond shown in Fig.3-1. One problem with PDMS is its poor mechanical properties. To improve the mechanical stability of PDMS, one can reinforce the material with silica and then vulcanize it (Anderson et al., 2000), but even then the tensile strength of PDMS is relatively low compared to other elastomers. PDMS is cured by an organometallic crosslinking reaction. The siloxane base oligomers contain vinyl groups (CH₂=CH⁻). The cross-linking oligomers contain at least 3 silicon hydride bonds each. The curing agent contains a proprietary platinum-based catalyst that catalyzes the addition of the SiH bond across the vinyl groups, forming Si⁻CH²-CH²-Si linkages. The multiple reaction sites on both the base and crosslinking oligomers allow for three-dimensional crosslinking; see Fig.3-2.

Property	Value
Color	Clear
Viscosity	3900 mPa·s
Specific gravity	1.08
Glass transition temperature	150 K**
Thermal conductivity	0.18 W/m°K
Shelf life	24 months

Table 3-1: Product Specification Dow Corning Sylgard 184*

*: Taken from: http://www.dowcorning.com

**: Theoretical value for linear chains of infinite length (Bischoff and Cray, 1999)



Figure 3-1 : siloxane bond



Figure 3-2 : Cross-linking of dimethylsiloxane (Picture taken from: http://mrsec.wisc.edu/)

One advantage of this type of addition reaction is that no waste products such as water are generated. If the ratio of curing agent to base is increased, a harder, more cross-linked elastomer results. Heating will also accelerate the crosslinking reaction. During curing, the elastomer shrinks somewhat.

The primary result that oxygen plasma treatment has on a PDMS surface is that it renders the surface hydrophilic. The Si-OH groups at the surface of the PDMS slabs can then come into contact with one another in order to form a covalent Si-O-Si bond between them. If the treated samples are left to age in air, the hydrophobicity recovers. This recovery is due to the low molecular weight PDMS that migrates to the surface and not by contamination through adsorption from the air (Hillborg et al., 2000). When PDMS is exposed to oxygen plasma, this leads not only to oxidation, but also to chain scission, crosslinking and formation of a silica-like layer, according to Hillborg and his colleagues. The thickness of the oxidized surface layer is approximately 130-160 nm and consists of SiOx i.e. silicon bonded to three or four oxygen atoms. If one uses prolonged plasma treatment (>30 seconds), the oxidized layer becomes thinner and starts to show evidence of cracking; Hillborg et al. showed this via the use of scanning electron microscopy (SEM).

3-3 : METAL DEPOSITION

There are various kinds of metal deposition techniques, but only physical vapor deposition (PVD), chemical vapor deposition (CVD), atomic layer deposition (ALD), and electroplating are considered to be the main and preferred techniques. Park et al. (2003) have fabricated resistance temperature detectors (RTD) inside a microchannel by sputtering, which is one of the PVD techniques as shown in Fig.4-3. However, their channel consists of glass and silicon, which is more easily sputtered than polymeric materials. In recent years, specific deposition technology has been developed for the fabrication of thin metal layers on polymeric material as well.



Figure 3-3 : Photographs of deposited Platinum (Park et al., 2003)

Several types of methods have been developed such as laser-assisted chemical vaporphase deposition (LCVD) (Haba et al., 1994), laser-induced forward transfer (LIFT) (Bohandy et al., 1988; Toth et al., 1993; Mogyorosi et al., 1989; Kantor et al., 1994), pulsedlaser deposition (PLD) (Cillesen et al., 1993; van Ingen et al., 1994), and so on. These techniques cover a good range of metals to be deposited on insulators, but their disadvantages are the necessity of vacuum/low-pressure gas systems, and dangerous compounds (in LCVD).

Considering the yield of the laser-enhanced metallization process, two processes can be distinguished. In laser-induced direct metallization, the deposited metal pattern is created in its final form, which is continuous, conductive, adhesive, regular, and requires no further treatments (Bali et al., 1993a; Kordás et al., 1999; Bali et al., 1993b; Kordás et al., 2000; Montgomery and Mantei, 1986; Yokoyama et al., 1984). In the non-direct method, the surface (polymers, ceramics) is simply activated with a laser pretreatment (Shafeev and Hoffmann, 1999; Dolgaev et al., 1996) or with the deposition of thin metal layers or metal seeds upon the substrate from liquid precursors (Schrott et al, 1995). The final metal coating is applied using chemical or electrolytic methods (due to its simplicity, usually a chemical method is applied) (Furness, 1968; Loewenheim, 1978; Schrott et al, 1995).

Processes that deposit metal layers onto different surfaces without external electricalcurrent sources are called electro(de)less plating or chemical deposition of the considered metal (Pearlstein, 1974).

In the microelectronics industry, especially in packaging technology, the metallization of insulator surfaces like polymers or ceramics is an important step of the manufacturing process. Each of the above mentioned techniques has their own specific area of application in which they perform their purpose in the quickest and/or most cost-effective way.

Coating PDMS with metal or ink has been available recently for the purposes of microcontact printing (μ CP) technology. For example, high-resolution elastomeric PDMS stamps coated with Au have already been used for nanoscale transfer printing (Loo et al., 2002).

For EOF micropump applications, only small numbers of researchers have tried the metallization of insulator surfaces in order to make electrodes or sensors. Some researchers deposit electrodes at the middle of a microchannel (Kenis et al., 1999; Studer et al., 2002) as shown in Figs. 4-4 and 4-5. This type of feature, which consists of a metal layer inside the channel, may disturb or change the flow.



Figure 3-4 : Schema of metal deposition at the middle of a microchannel (Studer et al., 2002)



Figure 3-5 : Micrograph of metal deposition at the middle of a microchannel (Kenis et al., 1999)

In order to prevent disturbing flow by metal inside the channel, other researchers deposited electrodes into the center of the reservoirs (Yun et al., 2002) as shown in Fig. 4-6.



Figure 3-6 : Photographs of deposited Platinum (Yun et al., 2002)

3-4 : OUR MICROFABRICATION SCHEME

In this study, two different kinds of microchannels (PDMS-glass and PDMS-PDMS) are fabricated as shown in Fig.3-7. Both PDMS and glass materials are favorable to electrokinetic flow, compatible with a variety of bio-analytical processes, and suitable for optical-based detection techniques. Here, microchannel structures are formed on PDMS using a soft lithography technique.

Basic soft lithography sequences are well known (Xia and Whitesides 1998a), but following procedure is used to form microchannels with embedded electrodes. First, positive photoresist (AZP4620, AZ Electronic Materials, NJ USA) is spincoated on a silicon wafer at 2000 rpm for 19 sec. in order to form a 10 µm thick layer. After 60 sec. of ultraviolet (UV) light exposure, patterned positive relief is prepared by dissolving the exposed regions with corresponding developer (AZ400, AZ Electronic Materials, NJ). Meanwhile, PDMS prepolymer and curing agent (Sylgard 184, Dow Corning Inc., MI USA) are mixed at a ratio of 10:1. The mixed liquid elastomer is then degassed for 2 hrs at 0.001 Torr and cast onto the prepared substrate after placing capillaries at the inlet and outlet reservoir positions. After curing for 6 hrs at 80^oC in a hot kiln, the solidified PDMS is carefully peeled off and the capillaries are removed (revealing a pair of holes) on PDMS. The resulting slab is the top layer of the microchannel. For PDMS-glass microchannels, a cover glass is used as the bottom layer, while for PDMS-PDMS channels, another layer of thin flat PDMS is formed.

Next, 5 nm of titanium tungsten (TiW) is sputtered on the bottom layer followed by 95 nm of gold (Au). Here, titanium tungsten is deposited as a binding agent since the bonding strength between gold and glass/PDMS is very weak. In order to minimize flow disturbances, the total electrode thickness is restricted to 100 nm. The precise patterning of photoresist (AZ4620) on this thin metal layer is carried out by identical photolithography, which is used to prepare the top layer. Gold that is uncoated by photoresist is etched and then

rinsed in DI water, and then agitated in 30% hydrogen peroxide for 40 sec. in order to remove the titanium tungsten layer. Cleaning with acetone and IPA washes away photoresist, so a patterned metal layer is formed on the bottom layer. Oxygen RF plasma (PDC-32G, Harrick Scientific Co., NY USA) is utilized for 20 sec. in order to temporally activate the PDMS surface before bonding the two layers. This step is employed to enhance the bonding strength between the layers as well as to make PDMS surfaces hydrophilic for the sake of an easy loading process.



Figure 3-7: Fabrication schemes used to form PDMS-glass and PDMS-PDMS microchannels. The following steps are shown: (a)cleaning of the substrate, (b)photoresist spincoating, (c)UV exposure, (d)develop the exposed photoresist and placement of capillaries, (e)PDMS casting, (f)top layer, (g)PDMS spincoating and curing, (h)sputtering TiW, (i)sputtering Au, (j)photolithography and metal etching, (k)bottom layer, (l)alignment of two layers, (m)bonding two layers.

CHAPTER 4

MICRO PARTICLE IMAGE VELOCIMETRY

4-1: INTRODUCTION

Scientists have sought to understand fluid flow inside a tiny chip, but detection techniques for microfluidic devices did not develop until later in comparison to the developed techniques of microfabrication. As one of the successful detection systems, micron resolution particle image velocimetry (µPIV) has been recently receiving an increasing amount of attention among those belonging to the society of microfluidics because it has advantages of being a non-contacting technique, having a large number of vectors, and high resolution. The PIV is a flow-diagnostic technique that provides velocity fields both qualitatively and quantitatively. Since the 1980s, PIV on a macro scale has become a popular method for measuring spatially resolved velocity fields (Adrian and Yao, 1985; Adrian, 1986). The basic idea of the PIV system is to correlate two image files, acquire the mean displacement within a small region (the so-called "interrogation window") and divide the mean displacement by the separation time between the first and second pulsed lasers.

Micro particle image velocimetry (μ PIV) is an excellent technique which allows us to acquire velocity profile inside a microchannel. The main differences between macro and micro scale PIV are (i) capture of fluorescent light from particles rather than scattering light; (ii) consideration of Brownian motion of particles, and (iii) volume illumination instead of the formation of a light sheet. Although conventionally, two-dimensional PIV involves the illumination of a single plane of the flow with a light sheet with a thickness that is less than the depth of field of the image recording system, volume illumination is an alternative approach, whereby the test section is illuminated by a volume of light (Willert et al., 1992; Okamoto et al., 1995; Gray and Greated, 1993; Hnsch, 1995). Volume illumination may be needed for obtaining two-dimensional measurements in microscale geometries because alignment of the light sheet is difficult (Meinhart et al., 2000b). In μ PIV, seeding particles must be small enough to exactly follow the flow without clogging the device. At the same time, particles must be large enough in order to be adequately imaged and to dampen the effects of Brownian motion (Santiago et al., 1998). Santiago et al. first attempted to obtain a bulk velocity of 50 μ m/s using 300nm diameter polystyrene particles in a spatial resolution of 6.9 μ m×6.9 μ m×1.5 μ m by considering the use of epifluorescent microscopes equipped with an intensified CCD camera in 1998. In that experiment, they utilized a continuousillumination mercury arc lamp as a light source. Continuous mercury lamps give off low-light levels which are suitable in enabling us to view flows around living microorganisms. However, large time intervals between image exposures limits the PIV system to relatively low velocities. Thus, Meinhart et al. (1999) developed a new idea in order to deal with 5ns pulsed Nd:YAG lasers with ~1mJ of light intensity.

Before studying the theoretical aspect of micro particle image velocimetry (μ PIV), we made sure to understand why micro resolution PIV detection system is suitable in this study. A quick comparison of the commercially available diagnostic techniques is illustrated in Table 4-1. It is clear that each technique has its own advantages and disadvantages. The μ PIV is selected because the limitations of PIV can be overcome by using a transparent microchannel composed of PDMS and glass. Reynolds number in this project is very small (always less than 1), so the time resolution problem is not a big factor.

In section 4-2, the PIV algorithm, particularly the correlation method, is reviewed. Based on knowledge of general PIV theory, we built an original source code specifically for micro scale diagnostics considering ensemble averaging methods in section 4-3. A brief explanation of experimental setup as well as its reliability is also described in section 4-3.

Detection Techniques	Advantages	Limitations		
Hot Wire Anemometry (HWA)	 Good spatial resolution (a few microns) Good time resolution (hundreds kHz range) Relatively inexpensive No particle needed 	 Intrusive Non-linear response to physical quantity Point measurement Influence of other valuables 		
Laser-Doppler Anemometry (LDA)	 Good time resolution (ten or a few kHz range) Linear response to physical quantity 	 Limited spatial resolution (typically 100 microns) (fewer fringes increases the uncertainty) Point measurement Use of tracer particles Requires optically transparent test piece Velocity direction ambiguity 		
Particle Tracking Velocimetry (PTV)	 Flow-field measurement Good spatial resolution (Down to a few microns using objective lens) High accuracy Linear response to physical quantity 	 Low time resolution (typically 15 Hz sampling rate depending on the camera cost) Sparse velocity information located randomly Use of tracer particles Requires optically transparent test piece 		
Particle Image Velocimetry (PIV)	 Flow-field measurement Good spatial resolution (Down to a few microns using objective lens) High accuracy Linear response to physical quantity 	 Low time resolution (typically 15 Hz sampling rate depending on the camera cost) Use of tracer particles Requires optically transparent test piece 		
Laser Speckle Velocimetry (LSV)	 Flow-field measurement Good spatial resolution (Down to a few microns using objective lens) Linear response to physical quantity 	 Low time resolution (typically 15 Hz sampling rate depending on the camera cost) Use of tracer particles Requires optically transparent test piece Velocity direction ambiguity 		
Image Correlation Velocimetry (ICV) (Shadow image & PIV)	Flow-field measurementNo particle neededNon-intrusive	 Far from being able to fully characterize a flow Requires optically transparent test piece 		
Doppler Ultrasonic Velocimetry	 No particle needed No optically transparent test piece needed Non-intrusive 	Requires opaque test piecePoint measurement		
Scalar Image Velocimetry (SIV)	 Flow-field measurement Good spatial resolution (Down to a few microns using objective lens) Non-physical access 	 Use of fluorescent or phosphorescent Not suitable for unsteady or developing flow Requires optically transparent test piece Velocity direction ambiguity 		

Table 4-1 : Comparison of commonly used velocity measurement

Use of tracer particles may disturb the flow, so they should be carefully chosen based upon the following factors. First of all, the particles should obviously be able to fit inside the microchannel. The concentration of particles needs to be 5~10 particles per interrogation spot in order to accurately calculate the flow velocity using the correlation method. On the other hand, if the size of the particle is too small, the intensity of fluorescent light will be invisible. Moreover, Brownian motion cannot be negligible as the size of the tracer particles become smaller. Specific gravity is desired to be as close to 1 as possible in order to cause the particles to follow the flow behavior exactly. In this study, seeding particles are purchased from a specialized vendor (Molecular Probes, OR USA), and its specific gravity in pure water at 20°C is approximately 1.06 in order to minimize particle lag and maximize fluorescent emission signal. The properties of the tracer particles used in this study are listed in Table 4-2. It is assumed that their presence does not alter the flow properties.

Material	Modified Carboxylate
Particle Size	1.0±0.032 μm
Absorption Maximum	540 nm
Emission Maximum	561 nm
Relative Quantum Yield	0.2
Charge	0.0148 meq/g
Density	1.055 g/cm^3
Optimized Concentration	0.4113% v/v

Table 4-2 : Properties of Fluorescent Particles

4-2 : BASIC CONCEPT AND THEORY

The experimental setup is the same among the techniques of Particle Tracking Velocimetry (PTV), Particle Image Velocimetry (PIV), and Laser Speckle Velocimetry (LSV) even though the concentration of seeding particles will vary. The densities of tracer particles for PTV, PIV,

and LSV are low, medium, and high, respectively. For PTV, a small number of particles are tracked individually (typically under long continuous exposure to light). On the other hand, PIV and LSV do not use tracks of individual particles, but local fluid velocity can be measured by statistically measuring the fluid displacement (particle displacement assuming that particles follow the fluid motion) using two images (typically under short pulsed exposures) and dividing that displacement by the separation time between the exposures. The recorded photographs are divided into small regions called "interrogation spots." These interrogation spots are commonly overlapped with their neighbors in order to increase the number of vectors maintaining the same resolution. The statistical method used in PIV algorithm is the correlation technique. There are primarily two types of correlations. One is called *autocorrelation*, and the other is called *cross-correlation*.

4-2-1 : UNDERSTANDING AUTOCORRELATION

The idea of autocorrelation is explained as follows. For preparatory purposes of understanding, let us first define the location of particles within an interrogation region of a particle image as an example. Figure 4-1(a) is an input image and Fig.5-1(b) is the list of particle centroid locations corresponding to the image.



Figure 4-1 : Identification of Particles within an Interrogation Spot for the purposes of preparation for understanding Autocorrelation. (a) Particle image (b) Location of each particle.

From Fig.4-1(b), the distances between the particles are tabulated in Fig.4-2. The numbers within the Table indicate the number of counts for the same displacements. Obviously, no displacement (δx , δy)=(0.0, 0.0) occurs at all particles, so the corresponding number is (6) colored in red in Fig.4-2 because there are 6 particles. The data is then plotted on a 2-D graph as shown in Fig.4-3. The diameter of the circle in Fig.4-3 represents the number of counts of each displacement occurrence in Fig.4-2. The most common displacement count (6) indicates particles correlating with themselves at zero displacement. This biggest peak located at (δx , δy)=(0.0, 0.0) does not contain any velocity information unlike the other peaks.

<mark>(</mark> δx,δy	#1	#2	#3	#4	#5	#6
#1	6	3	2	2	2	1
#2	3	6	1	2	1	2
#3	2	1	6	3	1	1
#4	2	2	3	6	2	1
#5	2	1	1	2	6	3
#6	1	2	1	1	3	6
1	(0.1, 0.5), (-0.7, 0.0), (-0.5, 0.3), (0.7, 0.0), (0.2, 0.4), (0.5, 0.4), (0.5, 0.4)					
	(0.2, 0.3), (0.5, -0.3), (-0.2, -0.4), (-0.1, -0.5), (-0.5, -0.4), (-0.2, -0.3)					
2	(-0.4, 0.1)	., 0.1) 2 (-0.1, 0.2)			2 (-0.2, 0.4)
2	(0.4, -0.1)		2 (0.1,	-0.2)	2 (0.2, -0.4)
3	(-0.3, -0.1)		3 (0.3,	0.1)	6 (0.0, 0.0)

Figure 4-2 : Number of Counts at the Same Displacements (δx , δy)



Figure 4-3 : Autocorrelation Graph

The autocorrelation graph can be also explained by shifting the image given by Fig.4-1(a) into all other directions. In Fig.4-4, the same autocorrelation function is obtained by the summation at each offset counting overlapped particles. When the two images are overlaid with no displacement, all of the particle images are aligned so that the zero peak is the strongest correlation possible. However, because the resultant autocorrelation function is symmetrically distributed about the origin, the autocorrelation is not suitable for the flow diagnostics when the user does not know the obvious flow direction. Due to the ambiguity of directional information, long exposure to light is preferred in order to allow for the tracing of bulk movement. Transferring one frame after taking a picture and keeping a shutter open for a long period of time does not require expensive experimental setup, so the autocorrelation technique remains a useful method and has yet to become obsolete.



Most Common Displacement Zero Peak = Correlating themselves

Figure 4-4 : Autocorrelation Generated by Counting Overlapped Particles

4-2-2: UNDERSTANDING CROSS-CORRELATION

Cross-correlation technique does not have ambiguity of direction information unlike autocorrelation, because the image shift is biased intentionally by the user. There are normally two kinds of cross-correlations: (i)1-frame cross-correlation with double/multiple exposure and (ii) 2-frames cross-correlation with a single exposure. Of course, the combination of multiple-frames cross-correlation with either a single/multiple exposure is also available, but at this point it is rare and beyond our interests. 1-frame cross-correlation uses a double exposure light on a single frame within a short period of time and by having the image shift using the typical method of rotating mirrors (Landreth et al., 1988a) or electro-optical techniques (Landreth et al., 1988b; Lourenco, 1993). If a camera is not high speed but lasers can be used as a light source, this technique may be applicable. Since there is no ambiguity in which unnecessary peaks exist out of the flow path, the signal-to-noise ratio (S/N) is increased, in contrast to the autocorrelation method. The resulting correlation function is drawn in Fig.4-5.



Figure 4-5 : One-Frame Cross-correlation Graph for Double Exposure Light

Historically, 2-frames cross-correlation with a single exposure was developed later than other techniques because it requires both high speed camera and lasers, both of which are expensive. This technique uses double frame within a short amount of time, so it is not easy to synchronize the image-acquiring device and the light source. If the user can afford to develop such an experimental setup, the highest S/N ratio is afforded because zero peak is eliminated from the correlation graph by using two different pictures as shown in Fig.4-6.



Cross-correlation Graph

Figure 4-6: Double-Frame Cross-correlation Graph for Single Exposure Light

4-2-3 : MATHEMATICAL TREATMENT OF CORRELATION

The discrete cross-correlation function of small windows is as follows:

$$C(\delta X, \delta X) = \sum_{i=1}^{N} \sum_{j=1}^{N} \{ f_1(X_i, Y_i) \} \{ f_2(X_i + \delta X, Y_i + \delta Y) \}$$
(4-1)

where f_1 and f_2 are the light intensity of frame 1 and frame 2, respectively, N is the number of the particle within the interrogation spots, (X, Y) is the coordinate system in image plane, and δ indicates displacement. Equation (4-1) looks simple in nature, but it is sensitive to changes in intensity within f_1 and f_2 . To overcome this drawback, Eq.(4-1) is normalized as (Huang et al., 1993)

$$C_{n}(\delta X, \delta X) = \frac{\sum_{i=1}^{N} \sum_{j=1}^{N} \left\{ f_{1}(X_{i}, Y_{i}) - \bar{f}_{1} \right\} \left\{ f_{2}(X_{i} + \delta X, Y_{i} + \delta Y) - \bar{f}_{2} \right\}}{\sqrt{\sum_{i=1}^{N} \sum_{j=1}^{N} \left\{ f_{1}(X_{i}, Y_{i}) - \bar{f}_{1} \right\}^{2}} \sqrt{\sum_{i=1}^{N} \sum_{j=1}^{N} \left\{ f_{2}(X_{i} + \delta X, Y_{i} + \delta Y) - \bar{f}_{2} \right\}^{2}}}$$
(4-2)

where \bar{f}_1 and \bar{f}_2 represent the mean values of frame 1 and frame 2, respectively. The displacement in real plane $(\delta x, \delta y)$ can be found by dividing the image plane displacement $(\delta X, \delta Y)$ by the magnification of the objective lens *M*. The characteristic velocity can be calculated by simply dividing $(\delta x, \delta y)$ by the separation time between the two images $(u, v) = (\delta x / \Delta t, \delta y / \Delta t)$.

4-3: INSTALLATION

Because the velocity within a microchannel cannot be detectable using conventional PIV setup, a specific experimental setup for μ PIV has to be integrated into the system. In macro scale PIV, a laser light sheet is typically generated by using two cylindrical lenses. In the case of μ PIV setup, volume illumination using an objective lens is utilized instead due to the limitation of physical access to the microgeometries. As for software, ensemble averaging and background noise reduction schemes are needed for μ PIV because the signal to noise ratio is relatively small due to the fact that seeding particles are very tiny. In addition to that, object recognition tools should be incorporated to avoid unphysical vectors outside of the complex geometries of microchannels. This section consists of three parts: hardware setup, software setup, and the validation of our experimental setup.

4-3-1 : HARDWARE SETUP

The µPIV system is converted from the conventional PIV setup. Figure 4-7 illustrates the optical path. Two Nd:YAG lasers (Powerlite, Continuum, CA USA) are utilized as a light source. Two-laser-configuration allows any pulse separation from very short to long with the full power of each laser, but careful alignment is required within the laser box prior to the PIV experiment. Parameters of Q-switch and flashlamp voltage are set at 180 [µs] and 1.21 [kV], respectively in order to have enough pulse energy to illuminate submicron particles. The pulse duration is 10 [nm], which is short enough to freeze the motion of fast flows (in this study velocity is on the order of a few microns up to mm/s). The neutral density filter (FSR-OD80, Newport Corp, CA USA) is used to reduce the intensity from 70[mJ] to 14[mJ]. Mirrors (02 MLQ009/025, Melles Griot, CA USA) with fine adjustments are used to guide the light into the inverted fluorescent microscope (CKX41, Olympus Co., Tokyo Japan). The Galilean type beam expander, composed of two lenses (01LPK007/078 and 0.LAO256/078, Melles Griot, CA USA), is placed to not only reduce the energy to 10[mJ], but also to form the uniformly illuminated image plane.



Figure 4-7: Schematic of Light Path for μ PIV Setup. Here Galilean type beam expander is used by using plano-concave and achromat convex lenses.

Figure 4-8 shows the two types of beam expanders. The Keplerian type consists of two positive lenses which are positioned with their focal points nominally coincident. On the other hand, the Galilean type consists of a negative diverging lens, followed by a positive collimating lens. In both cases, the overall length of the optical system is

$$L = f_1 + f_2 \tag{5-3}$$

and the magnification is given by

$$M_{beam} = \frac{f_2}{f_1}$$
(5-4)

In this study, the Galilean beam expander is utilized, and f_1 and f_2 are -30mm and 300mm, respectively - purchased from Melles Griot (CA). In Fig.5-7, an achromatic lens is used to prevent chromatic aberration.



Figure 4-8: Two Main Types of Beam Expanders

Inside the microscope, the light passes the excitation filter (532RDC, Chromatechnology, VT USA), dichroic mirror (Z532/10, Chromatechnology, VT USA), objective lens (10x/20x/40x, Olympus Co., Tokyo Japan), test piece (microchannel), the objective lens again, and the emission filter (HQ575/50, Chromatechnology, VT USA). Inside the microchannel, 500 nm orange fluorescent (540 nm excitation and 560 nm emission) polystyrene microspheres (Molecular Probes, OR USA) are filled in the buffer solution. Finally, the cooled CCD camera (PIVCAM13-8, TSI Inc., MN USA) captures the image synchronizing with the timing of the pulsing laser with the camera. The timing diagram of the PIV camera triggered double exposure frame straddle mode is drawn in Fig.4-9.



Figure 4-9 : PIVCAM13-8 (1.3 mega pixels, 1280×1024 pixels; 8Hz, 125ms single image repetition rate) Timing Diagram Triggered Double Exposure Frame Straddle Mode with Two Nd:YAG Lasers with Pulse Duration of 10 [nm]

Experimental setup of our system is depicted in Fig.4-10. It consists of a Nd:YAG laser, a CCD camera, optical components (neutral density filter, beam expander, dichroic mirror, color filters, and objective lens), a synchronizer, and a data acquisition system; both camera and laser are synchronized as mentioned above.



Figure 4-10: Experimental Setup for μ PIV technique. The modified carboxyl seeding tracers are excited at 540 nm and emit light at 560 nm of wavelength. The cooled CCD camera with 1280 × 1024 pixels provides 1280 instantaneous vectors. For a 40x magnification objective lens, the spatial resolution of our PIV system is 5.36 μ m × 5.36 μ m with a depth of field δz =6.73 μ m.

The theoretical diameter of a diffraction-limited point source of light, obtained from an infinitely corrected optical system onto a CCD chip is given by

$$d_{diff-\infty} = 2.44 f^{\#}_{\infty} M \lambda_{em} = 1.22 M \lambda_{em} \sqrt{(n/NA)^2 - 1}$$
(5-5)

where $f^{\#_{\infty}}$ is the f-number for infinite-corrected optics, *M* is the magnification of the objective lens, λ_{em} is the wavelength of emission light, *NA* is the numerical aperture, and n is the refractive index of the immersion medium. The theoretical diameter of diffraction-limited point source ($d_{diff-\infty}$) identifies the smallest feature a µPIV system can obtain. For our µPIV system (M=20, n=1, NA=0.4, and $\lambda_{em} = 555 nm$), the theoretical diameter has been estimated as 31.31 µm. The effective particle diameter recorded on the CCD chip is the convolution of the geometric image with the point spread function. If the Gaussian distribution is assumed for both geometric and diffraction limited images, the effective particle diameter can be given as

$$d_{eff} = \sqrt{(Md_P)^2 + d_{diff - \infty}^2}$$
(5-6)

where d_P is the diameter of the particle. According to Meinhart et al. (1993), a measurement uncertainty can be calculated as $\delta x \approx d_{eff} / (10M)$, if a particle image diameter is resolved by 4 pixels. The thickness of the measurement plane can be determined as (Meinhart et al. 2000b)

$$\delta z = \frac{3n\lambda_{ex}}{NA^2} + \frac{2.16nd_P}{NA} + d_P$$
(5-7)

where λ_{ex} is the excitation wavelength of light in vacuum. Inertial effects of particles are not so significant in this study because the response time of our particles is in the order of 10⁻⁸ sec. For an optical magnification M=20, particle diameter d_p=500 nm, excitation wavelength $\lambda_{ex} = 532 \text{ nm}$, refractive index n=1 and numerical aperture NA=0.4, the depth of field is calculated as 13.18 µm. Since the depth of field (δ_z) is larger than channel height (H), the µPIV results presented here are the averages of all the values depth-wise. Thus, our µPIV system is very similar to that of Kim and Kihm (2004), where they have used Ar ion lasers to image in 7 μ m deep microchannels). Since in electroosmotic flow, EDL varies between 1~10 nm, our μ PIV system cannot capture the flow physics within the EDL. In this study, interrogation window size is set at 32 x 32 pixels with 50% overlapping. The concentration of fluorescent micro-spheres is optimized to have 10 particles per interrogation volume (32 x 6.7/M)² x $\delta z \mu m^3$ where the system resolution is (32 x 6.7/M).

Depending on the width of the channel, 20x and 40x magnification objective lenses are chosen for wide and narrow channels, respectively. Table 6-1 lists the above-mentioned important parameters of our μ PIV system. A syringe pump is utilized to validate our experimental data. The error was estimated within 5% for the velocity range studied here (10 μ m/s~1000 μ m/s).

Table 4-3: List of effective particle diameters, measurement uncertainties, and depth of measurements for different objective lens used in this study.

М	п	NA	W.D.(mm)	$f^{\#}{}_{\infty}$	$d_{diff-\infty}$ (µm)	$d_{eff}(\mu m)$	$\delta x(nm)$	$\delta z (\mu m)$
20	1	0.40	3.0	1.1456	31.31	32.87	164	13.18
40	1	0.60	2.5	0.6667	36.44	41.57	104	6.73
4-3-2 : SOFTWARE DEVELOPMENT

Original post processing data analysis software is developed for this project written in Matlab. All results have good matching with commercial software (Insight version 3.2, TSI Incorporation). Some features of this code are listed below.

- Image background subtraction (TSI software does not have this).
- Cross correlation in FFT domain (fast calculation).
- Developed a moviemaker written in my original Matlab code. This creates not only simple movies with x-y axes but also advanced movies with vectors (TSI software does not have this).
- Object recognition to extract error vectors surrounding boundaries. This feature will be extremely useful when vectors are desired to be smoothened in complex geometries. (TSI software does not have this).
- Filtering techniques (window filter, standard-deviation filter, median filter, and smooth filter).
- Three different kinds of ensemble averaging techniques in case of steady flow (TSI software does not have this).
 - 1. Average Velocity Method.
 - 2. Average Image Method.
 - 3. Average Correlation Method.
- Controllable resolution (interrogation spot size) with recursive check. This enables super-resolution PIV with high accuracy. (TSI software does not have this).
- Interpolation of vectors to fill holes (for sparse vector fields in case of steady flow).
 - 1. Linear interpolation.
 - 2. Cubic spline interpolation.

3. Kriging interpolation.

Computational speed of Matlab may be not be high, but it can be increased by converting formats to 'C language' or generating 'dll files'. What is more important is that our original software can be modified at any time for the purposes of future work rather than having to rely on the black box developed by TSI.

Figure 4-11 shows three different ensemble averaging techniques for steady state flow. Averaging of each velocity data file (Average Velocity Method) is good enough if and only if all the instantaneous measurements are reliable. Average Image Method is used when the particle image density is low so that even small signals can be adequate in order to obtain proper realistic velocity fields. The third method is called the Average Correlation Method, which calculates the average of correlation functions. This method has a much higher signalto-noise ratio than instantaneous correlation functions. Window filter is used to make all tails of instantaneous correlation function at the same level so that there will be no discontinuous averaging. All of these ensemble techniques can eliminate unphysical vectors, but each method yields a slightly different maximum velocity.

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Figure 4-11(a) : Instantaneous Data before Averaging

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Figure 4-11(b) : Average Velocity Method

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Figure 4-11(c) : Average Image Method



Figure 4-11(d) : Average Correlation Method

Smoothing filter is also available in our program when a hole of a vector needs to be filled. In the case of TSI software, the area of interest should be rectangular in order to utilize the smoothing function. Therefore, it will be very difficult to deal with complicated geometry using TSI software. Our original software is capable of recognizing the object by itself so that the smoothing filter is still useful for all types of geometries. Figure 4-12 shows the velocity field after using smoothing filter on the data shown in Fig.4-11(c). It is remarkable that the velocities at the center of the straight channel are relatively larger than those near the wall, and that the profile is symmetrical. This is well-known behavior of the pressure driven flow.



Figure 4-12 : Straight Channel with Smoothing Filter

4-3-3 : VALIDATION OF μ PIV RESULTS

The developed software and experimental setup is tested by pressure-driven flow facilitated by a syringe pump. Because the channel cross sectional area is known, flow rate is set in correspondence to the desired velocity. For instance, 1 mm/s of the mean velocity leads to 10.8 μ l/hr of the flow rate. All of data points fit within a percentage of error at 6% as shown in Fig.4-13. Each data point consists of 10 experimental runs. The experimental method is appropriable both quantitatively and qualitatively as long as a tolerance of 6% is acceptable.



Figure 4-13 : Error of Measurement Data Compared to Expected Mean Velocity

CHAPTER 5

MIXED ELECTROOSMOTIC AND PRESSURE DRIVEN FLOWS IN A TRAPEZOIDAL MICROCHANNEL

5-1: INTRODUCTION

In the past two decades, microchannels have attracted increasing attention in "lab-on-a-chip" devices (Erickson and Li 2004). Unlike capillaries, microchannels can be formed to build complicated geometries. Commonly used microchannel fabrication techniques are physical etching, chemical etching, soft lithography, or a combination of all of the above. Among them, soft lithography is the most successful technology for the formation of complex microchannels on polymeric materials due to its potential advantages such as flexibility, reproducibility, reliability, simplicity, and cost-effectiveness (Xia and Whitesides 1998a). In soft lithography, microchannel is fabricated through replica molding of polymeric material after patterning positive relief on a substrate using photolithography. Depending on the type of photoresist used in the photolithography process, the resulting microchannel may not have a perfect rectangular shape in cross-section. Figure 5-1 shows the profile of two microchannels (Channel1: 300 µm base and Channel2: 140 µm base) fabricated on poly-dimethyl siloxane (PDMS) using soft lithography, where AZP4620 (AZ Electronic Materials, NJ USA) is used as photoresist. Both channels do not have sharp edges due to an imperfect developing process. As a matter of fact, for small aspect (depth to width) ratio channels, trapezoidal cross sections are more likely to be seen rather than perfect rectangular profiles if the soft lithography technique is applied.

Flow studies in microchannels have been reported by several research groups. Liquid flow visualization techniques in microgeometries can be divided into scalar-based and particle-based methods. There exist a number of scalar image velocimetries (SIV) such as laser-induced fluorescence (Erickson, et al., 2003), molecular tagging velocimetry (Maynes and Webb, 2002), and photo-bleached fluorescence (Paul, et al., 1998). In SIV, the velocity profile is quantified by tracing transported dye (normally fluorescent or phosphorescent). However, this technique suffers from serious limitations if the flow is not steady and fully developed. For quantification of velocity distribution across a microgeometry, particle-based methods are appropriate. Although laser Doppler velocimetry (Tieu et al. 1995) and particle tracking velocimetry (Devasenathipathy et al. 2002) are available on a micro scale, particle image velocimetry (PIV) is the most common microscale flow analysis technique nowadays. This is primarily due to the ability of PIV to acquire velocity distribution with high spatial resolution. The first work of PIV on a micro scale was presented by Santiago et al. (1998). They utilized a continuous mercury arc lamp, so their system is suitable for the slow motion of photosensitive biological molecules. Later on, Meinhart et al. (1999) introduced pulsed laser as a light source in order to relax the limitation of measured velocity range because the laser can effectively freeze the particle motion. This group also introduced an algorithm to reduce the Brownian motion of seeding particles using temporal and/or spatial averaging techniques (Meinhart et al. 2000a). Recently, a number of research groups have contributed to the understanding of depth of focus (Olsen and Adrian 2000; Wereley et al. 2002; Bourdon et al. 2004), and developed high resolution µPIV system (Keane et al., 1995; Takehara et al., 2000). Cummings (2000) first visualized electrokinetic flows using the μ PIV technique. Later on, Kim et al. (2002) presented both experimental and numerical work in a grooved microchannel. In all these experimental studies, either pressure or electrokinetic driven flow is obtained in a rectangular microchannel, and none of them have considered trapezoidal shape geometry. Moreover, no previous experimental works addressed mixed electroosmotic and pressure driven flow in microgeometries where an external pressure head is superimposed upon electroosmotically-driven flows.



(a)



Figure 5-1: Profiles of microchannel fabricated using soft lithography technique. Here (a) Channel1: 300 μm base and (b) Channel2: 140 μm base.

In order to quantify velocity distribution in trapezoidal microchannels, the μ PIV system is utilized. Here, an external pressure is imposed upon an electroosmotically-driven mechanism for the purposes of studying the combined flow phenomena in the realm of physics. This chapter is arranged as follows. In the next section, theory and governing equations for electrokinetic microflow in a trapezoidal channel are provided. This is followed by the numerical scheme and its validation. Next, the fabrication techniques and μ PIV setup developed specifically for this chapter are introduced. Then the results of electrophoretic, pressure, electroosmotic and mixed electroosmotic-pressure driven flow velocities are presented. Finally, the experimental results are discussed.

5-2 : NUMERICAL SCHEME AND VALIDATION

The electrokinetic velocity field can be obtained by solving equations (2-1, 2-2, 2-3) with appropriate boundary conditions. However, the direct solution of the coupled system will be extremely (computationally) expensive, especially for 3-D geometry, due to the different length scales associated with microfluidic devices. Generally microfluidic channels are 1~10 microns thick, 10~500 microns wide, and 1~5 cm long. In addition to the diverse spatial length scales, the EDL thickness ranges from 1 and 10 nm. The complete numerical solution of 3D electrokinetic flows is presented in Yang and Li (1998) and Patankar and Hu (1998) based on Debye-Huckel approximation. Later on, Dutta et al. (2002) presented the steady electroosmotic flow in planar geometries using the spectral element method. In their study, EDL is resolved for dilute electrolytes where the channel height was two order magnitudes higher than the Debye length (λ). However, in most microfluidic devices, the ratio of channel height to Debye length ranges from 10³ to 10⁵. Moreover, for device level (3-D) simulation, significantly longer channel width and length needs to be considered. The computational effort of 3D electrokinetic flow can be minimized considerably by placing an

effective slip velocity at the channel wall to capture the thin EDL effects on the fluid flow. The slip velocity concept has been reported recently in a number of numerical works to study (Santiago, 2001; Sadr et al., 2004; Hlushkou et al. 2005).

In this chapter, the finite volume scheme (Patankar, 1980) is utilized in order to solve the Navier-Stokes equations without any electrokinetic body forces. For electroosmotic and mixed flow, the velocity at the slip plane is specified as

$$\vec{\mathbf{v}} = -\left(\frac{\varepsilon\zeta\nabla\phi}{\eta}\right) = -M_{eo}\nabla\phi \tag{5-1}$$

where ζ is the zeta potential and M_{eo} is the electroosmotic mobility. For purposes of simplicity, electroosmotic mobility is assumed to be constant along the channel wall; electroosmotic mobility has been obtained from the experiments. In solving electroosmotic velocity field, pressure is maintained at the atmospheric level at both inlet and exit reservoirs. The external electric field distribution is obtained from $\nabla \cdot [\sigma \nabla \phi] = 0$, which is subjected to insulating boundary conditions ($\nabla \phi \cdot \vec{n} = 0$) on the channel surfaces.

Figure 5-2 shows the numerical results for pressure driven flows in rectangular microchannels with a pressure gradient of 9313 N/m³. Here, velocity results are averaged in the depth-wise direction. Two different channel sizes are considered in this numerical work, where (a) 300 μ m X 12 μ m and (b) 140 μ m X 8 μ m. If the pressure gradient applied to both channels is the same, the thinner channel predicts lower flow velocity. These numerical results are also compared with the analytical results of Ebadian and Dong (1998), and an excellent agreement is obtained throughout the computational domain.



(b)

Figure 5-2: Validation of numerical results for pressure driven flows (dP/dx=9319 N/m^3) in rectangular microchannel. Here (a) 300 μ m X 12 μ m and (b) 140 μ m X 8 μ m microchannels. Analytical solution of pressure-driven flow for rectangular channel by Ebadian and Dong (1998) is used for validation.

5-3: DISCUSSION OF RESULTS

Two kinds of microchannel materials (PDMS-glass and PDMS-PDMS) and two different channel sizes (Channel1: 300 µm base and Channel2: 140 µm base) are tested to characterize the flow behavior in trapezoidal microchannels. Figure 5-3 shows the photograph of the microchannels. Embedded electrodes are used to apply an electric field along the channel, while a pressure gradient is formed along the channel by creating head difference between inlet and exit reservoirs. The applied pressure gradient can be calculated as $\rho_{fg}\Delta h/L - C_{Loss}\rho_{f}V_{m}^{2}/(2L)$ where C_{Loss} accounts for entrance and exit losses, $\rho_{f} = 998 \text{ kg/m}^{3}$ is fluid density, and L=2 cm is channel length. The height difference Δh is varied from 10 to 30 mm. Since $C_{Loss}V_{m}^{2}/2$ is extremely small compared to $g\Delta h$, effects of entrance and exit losses are minor in this experiment. In this study, DI water (pH=7.5) and 100 mM borate buffer (pH=9.2) are used as working fluids.

Instantaneous measured velocity field obtained from μ PIV system can be decomposed into several terms as

$$\vec{u}_{measure} = \vec{u}_{flow} + \vec{u}_B + \vec{u}_{PH}$$
(5-2)

where \vec{u}_{flow} is the flow velocity, \vec{u}_B is the velocity contribution due to Brownian motion, and \vec{u}_{PH} is the electrophoretic velocity. The flow velocity may contain electroosmotic, pressure driven, or mixed electroosmotic-pressure driven components depending upon the pumping technique applied along the channel. Detailed discussions of Brownian velocity (\vec{u}_B) and electrophoretic velocity (\vec{u}_{PH}) are presented in the next sections.



Figure 5-3: Photograph of a fabricated microchannel on poly-di-methyl siloxane along with embedded (TiW-Au) electrodes and reservoirs. Capillaries are used to have pressure head difference between inlet and outlet, while electrodes are used to apply electric field.

5-3-1 : ESTIMATION OF BROWNIAN MOTION EFFECTS

Brownian motion of seeding particles may cause fluctuation of measurement velocities when the size of the particle is less than a few micro meters. The diffusivity of seeding particles can be estimated as (Friedlander 1977)

$$D = \frac{k_B T}{3\pi\eta d_P} \tag{5-3}$$

Based on the diffusivity, Brownian motion velocity can be calculated as (Einstein 1956)

$$u_B = \sqrt{\frac{2D}{\Delta t}}\vec{e}_r \tag{5-4}$$

where Δt is the desired separation time between pulses of laser and \vec{e}_r is the random unit vector. Since Brownian motion occurs in all directions, the error due to this random motion can be dramatically reduced by ensemble-averaging techniques if the flow is steady (Meinhart et al. 2000a). Among three major averaging techniques (average velocity method, average image method, and average correlation method), averages of correlation functions over 40 experimental-runs demonstrated the best result in our particular experiment, and fluctuation of velocity due to Brownian motion is significantly reduced.

5-3-2: ESTIMATION OF ELECTROPHORETIC VELOCITY

Although seeding particles are neutrally buoyant in both working fluids (DI water and 100 mM borate buffer), they acquire negative charges at pH > 7 due to the presence of carboxyl group (-COOH) at their surfaces. This fact introduces electrophoretic mobilization of seeding particles when electric field is applied to drive the flow. The theory of electrophoretic velocity is already described in Chapter 2-2. The electrophoretic velocity of seeding particles under an applied electric field can be estimated as

$$\vec{u}_{PH} = \frac{2}{3} \frac{\zeta_{P} \varepsilon \vec{E}}{\eta} = M_{PH} \vec{E}$$
(5-5)

where M_{PH} is the electrophoretic mobility. In order to isolate electrophoretic velocity from observed velocity, the wall surface is coated with methylcellulose (1.1% w/w MC) before buffer solution and tracer particles are loaded into the channel. This dynamic coating can considerably suppress the electroosmotic flow (Cui et al. 2005). The mean velocity of the particles is then measured by the µPIV detection technique, although electrophoresis has been classified as a particulate flow phenomena. Perhaps particle tracking velocimetry (PTV) has more physical meaning for the measurement of electrophoretic velocities of each particle because PIV detection system will result in mean electrophoretic velocity field rather than distinguishing each particle. However, this approach is more appropriate since electroosmotic flow field will be obtained by subtracting the electrophoretic velocity from the observed velocity field ($u_{measure}$). The mean electrophoretic velocities of two different buffer solutions (DI water and 100 mM borate buffer) are illustrated in Fig. 5-4. The direction of the flow in both cases is from cathode to anode, indicated by a negative sign of velocity or mobility (opposite direction from the electric field). The electrophoretic velocity in the borate buffer is higher than that of DI water. This is due to the fact that seeding particles acquire higher negative charges at high pH. The observed velocity is linearly proportional to the applied electric field, and the slope of the curve represents the electrophoretic mobility. However, the linearity of the curve will be broken at high electric fields (> 300 V/cm) due to Joule heating effects. Joule heating increases the fluid temperature which changes dynamic viscosity and mobility. Note that neither channel materials (PDMS-glass and PDMS-PDMS) nor channel size (300 μ m base channel and 140 μ m base channel) changed the value of the electrophoretic velocity.



Figure 5-4: Mean electrophoretic velocity of modified carboxyl seeding particles. The estimated mobilities of DI water (pH=7.5) and 100mM borate buffer (pH=9.2) are -0.3742×10^{-4} cm²/V s and -1.3965×10^{-4} cm²/V s, respectively.

5-3-3 : PURE PRESSURE DRIVEN FLOW

Figure 5-5 shows the pressure-driven velocity profile across the microchannel. Here, symbols represent experimental results, while the solid line represents numerical prediction. Numerical results obtained from 3-D simulation are also averaged in the depth-wise direction. Two different channel heights depict two different magnitudes, but similar trends are visible in the velocity profile. Numerical results show that (depthwise average) the velocity profile in the rectangular channel is uniform across the channel, except at regions very close to the side wall. However, in trapezoidal microchannels the average velocity has a tapered-cosine shape (containing two single inflection points placed symmetrically around the centerline). This is due to the fact that trapezoidal microchannel side walls are not perpendicular to the base. It is clearly seen that both numerical and experimental results manifest the tapered-cosine velocity profile for trapezoidal microchannels. The experimental results show good agreement with numerical predictions except at the tip of the tapered region. There are two possibilities behind poor agreement at the tip of tapered region. First, the focal plane, achieved by objective lens, is not parallel to the side wall of trapezoidal channel. Optically, it is not possible to observe all lights emitted by seeding particles although the camera captures images from the bottom of the channel. Second, the concentration of particles near the side walls is lot less than at the center of the channel. This causes fewer signals from each particle. Third, modified carboxyl seeding tracers tend to adhere to the channel material partly due to PDMS surface roughness and partly due to electrical attraction between the particles and native PDMS.



Figure 5-5: Depth-wise average velocity profile in pressure-driven flow for (a) Channel 1 and (b) Channel 2 presented in figure 5-1. Numerical results for rectangular microchannels are also presented for comparison.

5-3-4 : ELECTROOSMOTIC FLOW

In electroosmotic flow, an external electric field is applied along the channel by using electrodes. The embedded electrodes are 1mm wide, 100 nm thick, and 2 cm apart. Simulation results (not shown here) indicate that this configuration of electrodes provides uniform electric field along the channel. All experimental data is captured 30 sec. after initiation of the electric field, and the height of the liquid surface is kept at the same level to prevent pressure-driven flow by fabricating 1 cm diameter inlet and outlet reservoirs.

The electroosmotic velocities for PDMS-glass and PDMS-PDMS are exhibited in Fig. 5-6 for an electric field range of $50 \sim 300$ V/mm. Pure electroosmotic flow velocity is calculated by subtracting the estimated electrophoretic velocity (u_{PH}) from measured velocity $(u_{measure})$ of the µPIV system. Like the electrophoretic velocity results, electroosmotic velocity is also a linear function of the electric field. In all cases, the fluid travels toward the cathode, which means that both glass and PDMS surfaces are negatively charged. Because the electroosmotic flow profile is almost uniform, there was no dependence on channel size or geometries (300 µm base channel and 140 µm base channel). However, experimental results show that the channel material influences the magnitude of electroosmotic flow. If PDMS is used as a channel material, one should take special consideration in how the channel surface is treated. Plasma is utilized to bond the top (PDMS) layer and the bottom (PDMS or glass) layer in the fabrication process as described in Chapter 4. Plasma treatment affects the hydrophobicity on both glass and PDMS surfaces. The oxidized PDMS-PDMS channel surface is highly hydrophilic so that the relatively high electroosmotic mobility is achieved as shown in Fig.6-9. However, the PDMS surface quickly returns to its original hydrophobic feature when the microchannel is flushed three times with buffer solution. The hydrophobic PDMS is known as native PDMS. The electroosmotic flow is stable in native PDMS for a

longer period of time (40 days of observation for this work), even though their mobility values are much smaller. Unlike other studies, other chemical solutions (such as sodium hydroxide) are not loaded into the channel; the surface chemistry prior to the experiment was unaltered. The glass material has substantially higher electroosmotic mobility than the native PDMS. For this reason, the hybrid PDMS-glass channel yields higher electroosmotic velocity than that of the native PDMS. This agrees with the findings of other researchers (Duffy et al. 1998; Devasenathipathy et al. 2002).



Figure 5-6 : Mean electroosmotic velocity of 100 mM borate buffer inside PDMS-glass and PDMS-PDMS microchannels. The estimated mobilities of modified PDMS-PDMS, untreated PDMS-glass, and native PDMS-PDMS are $3.5658 \times 10^{-4} \text{ cm}^2/\text{V} \text{ s}$, $2.3890 \times 10^{-4} \text{ cm}^2/\text{V} \text{ s}$, and $1.1202 \times 10^{-4} \text{ cm}^2/\text{V} \text{ s}$, respectively.

5-3-5 : MIXED ELECTROOSMOTIC AND PRESSURE DRIVEN FLOW

In this section, the applied electric field is maintained at 100 V/cm, and the head difference of the liquid in inlet and outlet capillaries is varied between 10 and 30 mm. Both favorable $(\Delta P/L < 0)$ and adverse $(\Delta P/L > 0)$ pressure gradient mixed flow cases are studied here. In case of favorable pressure gradient mixed flow, the flow direction of pressure-driven flow and electroosmotic flow is the same. This may take place in two scenarios. First, if the electric field becomes enhanced due to the stacking of charged molecules or seeding particle on the channel surface, pressure is induced locally. Charged seeding particles may stick to the wall due to geometric complexity or roughness of the channel surface. Second, favorable pressure gradient mixed flow can occur by maintaining a pumping head along the channel by connecting the microchannel with a syringe pump or pressure regulator whilst electric field is applied for the purpose of separation or mixing of the species. On the other hand, adverse pressure gradient arises more often in practice when electroosmotic pumps are desired. Due to placement of the valve or an increase in head of the liquid in the outlet reservoir, the velocity at the center of the channel tends to decrease or may even become a negative value (reverse flow). Mixed flow cases are also predicted using numerical techniques and then compared with experimental results. It is important to note that our µPIV system cannot resolve electric double layer regions. Hence, in numerical scheme, slip velocity boundary conditions on the channel surface are specified, where slip velocity is calculated from electroosmotic mobility and applied electric field.

Mixed flow under favorable pressure gradient is illustrated in Fig. 5-7(a). Obviously, as the pressure gradient becomes steeper, the pressure driven flow becomes more dominant. Two inflection points, placed symmetrically around the centerline, are characteristics of pressure-driven flow (depthwise average) velocity as discussed in the section discussing pressure-driven flow. Experimental data (symbols) match well with numerical prediction

(solid lines). The deviation of profile is the largest if the flow is purely electroosmotic and the maximum deviation is estimated to be ~ 9 μ m/s. This deviation can be explained from the following facts. In flow velocity estimation, the mean electrophoretic velocity has been subtracted from the observed velocity under the assumption that seeding particles are distributed uniformly and have identical charges and sizes. In practice, each particle experiences slightly different electrophoretic migration. Moreover, since our microchannel is not pretreated with a concentrated sodium hydroxide, there may be non-uniform zeta potential distribution inside the channel. As compared to pure pressure driven flow (Fig. 5-5), the mixed velocity (experimental) near the side walls is slightly higher than expected. This can be attributed to relatively stronger surface interaction (adsorption) between seeding particles and the channel due to applied electric field.

Figure 5-7(b) demonstrates the combined electroosmotic and adverse pressure-driven flow. Although all the values of velocities in this figure are positive, the observed velocity field before subtracting electrophoretic velocity did indicate reverse flow, especially at the center of the channel. The pure electroosmotic flow is identical to the one in Fig.5-7(a) at an electric field of 100 V/cm. If the pressure driven component were to be subtracted from the pure electroosmotic flow instead of being added in Fig. 5-8(a), the results would look more like Fig.5-7(b). This means that Fig.5-7(a) and (b) are almost symmetrical about the horizontal line of the pure electroosmotic flow velocity profile. The evidence of linear superposition of electrokinetic (electrophoretic velocity and electroosmotic velocity) and pressure driven flow indicates that calculation in PIV post-processing techniques, such as ensemble averaging, successfully eliminates Brownian motion effects. Mixed flow with adverse pressure gradient requires careful testing of erroneous vectors if the total flow rate happens to be zero, since ambiguity of the flow direction sometimes creates errors in PIV calculation. For this reason, three different magnitudes of pressure gradient are chosen in such a way that reverses flow at the center of the channel; the flow reversal is visible to the naked eye. It turns out that relatively small pressure gradient can generate reverse flow in this experiment. This implies that the difference in elevation between inlet and outlet is sensitive to the generation of pressure head if a micro scale pumping system is interconnected to meso scale devices.



Figure 5-7: Depth-wise average velocity distribution in trapezoidal microchannel (Channel1) for (a) favorable pressure gradient mixed flow and (b) adverse pressure gradient mixed flow. Here, experimental results are shown with symbols and numerical results with solid lines. The applied electric field is kept constant at 100 V/cm, while pressure head is varied.

5-4 : CONCLUSIONS

Pressure, electroosmotic, and mixed electroosmotic-pressure driven flow in a trapezoidal microchannel using μ PIV technique are demonstrated. The use of relatively low magnification and low NA objective lenses leads to a larger depth of focus compared to the depth of the microchannel, so the resultant flow velocity values are averaged in the depth-wise direction. The average correlation method successfully reduced Brownian motion effects using 40 ensemble data.

Electrophoretic velocity of modified carboxyl seeding particles is measured to accurately quantify electroosmotic and mixed flows. Dynamic coating of PDMS surfaces with methylcellulose (1.1% w/w MC) before loading the buffer solution effectively suppressed electroosmotic flow. Two different buffer solutions (DI water and 100 mM borate buffer) are tested, and it is observed that higher pH value yields higher electrophoretic mobility. The linear relationship between the electric field and electrokinetic velocity starts to break if the magnitude of electric field exceeds 300 V/cm.

Three-dimensional numerical results of pressure-driven flow in trapezoidal microchannels show that the averaged depth-wise velocity profile has two single inflection points placed symmetrically around the centerline. This unique characteristic of the pressure-driven velocity profile is experimentally verified by using an in-house μ PIV detection technique.

Electroosmotic velocity is obtained by subtracting pre-examined electrophoretic velocity from the observed velocity field. Electroosmotic velocity profiles are almost uniform and linearly proportional to the electric field regardless of channel dimensions. Modified PDMS surface activated by oxygen plasma enhances electroosmotic flow. Native PDMS, however, is more stable for longer period of time, although the electroosmotic mobility is very low.

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Finally, mixed electroosmotic and pressure driven flow is analyzed. Electroosmotic flow combined with favorable pressure gradient provides higher pumping capabilities. On the other hand, mixed flow with adverse pressure gradient showed reverse flow especially at the center of the channel. In both cases, numerical prediction matches with experimental data nicely, and the maximum deviation of velocity was 9 μ m/s. Experimental results verify that the observed flow can be decomposed into electrophoretic, electroosmotic, and pressure driven flows after applying ensemble averaging techniques.

CHAPTER 6

ELECTROOSMOTIC FLOW CONTROL IN MICROGEOMETRIES USING FIELD EFFECT

6-1: INTRODUCTION

In the last decade, microfluidic technology has experienced exponential growth in bioanalytical areas such as DNA sequencing, PCR amplification, protein separation and analysis, immunoassays, cell sorting and manipulation, and vitro fertilization (Fiorini and Chiu, 2005; Becker and Locascio, 2002). This is because microfluidic devices offer rapid separation, automation, minimum sample consumption, and online integration with other unit operations. Generally, complex structured microgeometries such as T-channel, cross-channel, and Ychannel are required for the above-mentioned process in order to carry out all the functionalities on a single Lab-on-the-chip device. Thus, a microchannel is preferred over a traditionally-used capillary tube since integrated microfluidic devices can be formed fairly easily with microchannels using existing microfabrication techniques.

In microfluidic devices, an electroosmotic pump is a more attractive choice in sample transportation over any other existing transport mechanism because (a) the pump can be integrated within the microchip, (b) the flow rate can be controlled very precisely with an applied electric field, and (c) there are no moving parts. Electroosmotic flow has been widely investigated in recent years due to its relative importance in microdevices. In electroosmotic flow, ionized liquid moves with respect to the stationary charged surface under the action of an electrokinetic body force. The magnitude of electroosmotic velocity in a microchannel can be controlled either by changing the external electric field applied along the flow direction or by modifying the electrokinetic potential at the stern plane (also known as zeta potential). It is relatively easy to control the discharge in a microchannel by adjusting the external electric

field applied in the flow direction. This type of control is generally global (throughout the electric field region) and cannot be applied to a local area of interest or in complex microgeometries such as in the case of T-junctions or cross-channel junctions. On the other hand, selective manipulation of zeta potential seems to be effective in controlling flow at the local level.

The exact manipulation of zeta potential within microchannels is quite challenging because it depends on solution pH, buffer concentration and surface charge density. Lee et al. first introduced the zeta potential control inside a capillary by applying a gate voltage from outside of the capillary (Lee et al. 1990, 1991). They also demonstrated a simple capacitor model in order to estimate the zeta potential inside the capillary wall. The thickness of the capillary wall in their experiment was 100 [µm], so the required gate voltage was in the order of 10³ [V] to obtain a distinct change in zeta potential. Control of zeta potential at relatively low gate voltage can be achieved by either reducing chemical ionization or enhancing the induced charge. Chemical ionization of surface silanol groups can be minimized using a dilute solution (Ghowsi, 1991) and/or a low pH buffer solution (Hayes et al. 1993a). However, utilization of low pH buffer restricts the applicability of this technique to acidic buffer only. This problem can be overcome to some degree by coating channel walls. Hayes demonstrated that the coating of capillary walls with large organo silanes allows for better control (Hayes, 1999), and can be used for a wide range of buffer pH. It is important to note that coating wall material with polymer itself changes zeta potential (Herr et al. 2000; Barker et al. 2000), so careful analysis of zeta potential is necessary when a combination of dynamic coating and the gate voltage approach is desired. On the other hand, in order to maximize the effects of induced charge via externally applied gate voltage, a very thin type of wall material (insulator or capacitor) is needed since the induced charge density on the capacitance is inversely proportional to the thickness of the wall (in case of rectangular channel). Schasfoort et al.

(1999) fabricated ultra thin silicon nitride layers (390 [nm]) and exhibited a low gate voltage flow control (in the order of 10 [V]) in a silicon microchip. They are the first group to report the field-effect control using a microchannel rather than capillaries. In their model, three kinds of capacitances (the capacitances of the channel wall, Stern layer, and EDL) were considered, and they pointed out that saturation occurs at high gate voltages due to the fact that the capacitance of EDL also increases with the applied secondary (perpendicular) electric field. Later on Buch et al. (2001) demonstrated field-effect flow control using a 2 [µm] thick silicon dioxide capacitor covered by a poly-di-methyl siloxane (PDMS) microchannel. Recently, Karnik et al. (2005) examined the effects of perpendicular electric field which originated from gate voltage in a nanofluidic channel, and found that not only zeta potential but also ionic concentration is influenced when the channel size is comparable to or smaller than the Debye length. Lately, field effects are utilized in silicon microchips to obtain nonuniform zeta potential for micromixing (Lee et al. 2004a) and localized dispersion control (Lee et al. 2004b).

So far, field-effect transistors or capacitors are formed either on glass or silicon wafers to control the flow in a microchannel; this has not been attempted on polymeric materials as of yet, although polymeric materials are extremely popular in lab-on-a-chip devices. Moreover, none of the above-mentioned literatures discussed leakage current of the capacitor assuming perfect insulation of the channel wall. In practice, all dielectric materials used in electrokinetic based microfluidic channel allow current leakage, especially at high electric fields. The main objective of this study is to demonstrate localized flow control in microgeometries using a novel field-effect transistor formed on PDMS using soft lithography techniques. A detailed theoretical model for leakage current to estimate the zeta potential at the Stern plane is also presented. Moreover, in the experiment, high resolution micro μ PIV technique is utilized in order to obtain detailed velocity distribution in the controlled region.

The resultant velocity distributions tell us potential applicability and limitations of the proposed field-effect-controlled mechanism inside a PDMS-glass microchannel.

6-2 : THEORY

6-2-1 : FIELD EFFECT TRANSISTOR DESIGN

The field effect controlled microchip, shown in Fig.6-1, consists of two perpendicular channels with a micron sized separation between them. The main (horizontal) channel between drain and source reservoirs is filled with a target liquid, and a net flow of the liquid can be achieved by applying an electric field along source and drain reservoirs. On the other hand, a stationary conductive liquid is placed inside the side/sub (vertical) channel. The side channel is formed to apply perpendicular electric field for the proposed field-effect transistor. The potential at the drain and gate reservoirs are set as V_d , and V_g , respectively, while the source reservoir is connected to the common ground. In this investigation, both main and side channels are filled with 100 [mM] borate buffer, although it is possible to fill this channel with any ionized liquid. When borate buffer comes into contact with the channel (PDMS) wall, the channel surface is negatively charged due to the ionization, ion absorption or ion adsorption. Counter ions from the liquid become attracted to the wall, forming an immobilized Stern layer (on set figure in Fig.6-1). The next layer is known as a diffuse layer where ions are subjected to both electrostatic potential energy and thermal kinetic energy. The thickness of the diffuse layer is generally quantified by a characteristics dimension called Debye length (λ). Debye length represents the distance from Stern plane where a balance between electrostatic and thermal energy occurs, and for symmetric ionic valences $(z_+ = -z_- = z)$ it is given by (Hunter 1981) the formula

$$\lambda = \sqrt{\frac{\varepsilon_b \varepsilon_0 RT}{2z^2 F^2 c_\infty}} \tag{6-1}$$

where ε_b is the relative permittivity of the buffer solution, ε_0 is the permittivity of free space, R is the universal gas constant, T is the absolute temperature, F is the Faraday's constant, and c_{∞} is the concentration in the bulk region. Stern layer and the diffuse layer form the electric double layer (EDL). The thickness of Stern layer is generally much lower than that of the diffuse layer. Hence, in this study, the effects of Stern layer are neglected in the EDL phenomena and capacitance estimation.



Figure 6-1: Schematic view of field effect controlled microfluidic device for straight channel application. The main (horizontal) channel is separated from the sub (vertical) channel by a micron-sized distance (d). The potential at drain and gate reservoirs are V_d and V_g, respectively, while the source reservoir is connected to a common ground (V_s=0). The length of the main channel (L_{main}) and the sub channel (L_{sub}), and controlled region (L_{control}), are 2 [cm], 2.5 [mm], and 1[mm], respectively. In this study, drain voltage is fixed at 50 [V], while the gate voltage is modulated between 5 [V] and 45 [V]. The onset figure shows the formation of EDL next to the channel surface.

Figure 6-2(a) illustrates the equivalent electric circuit of our field-effect flow controller. Due to the presence of two fluidic channels, there are three different capacitors in

this system: EDL capacitor of the side channel (C_{EDL1}) , wall capacitor (C_{wall}) , and EDL capacitor of the main channel (C_{EDL2}) . The capacitance of the EDL can be expressed as (Newman, 2004)

$$C_{EDL} = \frac{\varepsilon_b \varepsilon_0 A}{\lambda} \cosh\left(\frac{zF\zeta}{2RT}\right)$$
(6-2)

where A is the charged area and ζ is the zeta potential. In a straight microchannel, one can determine the value of ζ experimentally by measuring the electroosmotic velocity (u_{eo}) for an electric field (E) as

$$\zeta = -\frac{u_{eo}\mu}{E\varepsilon_b\varepsilon_0} \tag{6-3}$$

where μ is the viscosity of the fluid. On the other hand, the capacitance of the wall is given by

$$C_{wall} = \frac{A\varepsilon_w \varepsilon_0}{d} \tag{6-4}$$

where ε_w is the relative permittivity of the wall material and d is the separation distance between the main channel and the side channel. Since all three capacitors are in a series, the total capacitance can be calculated as

$$C_{total} = \left[C_{wall}^{-1} + C_{EDL1}^{-1} + C_{EDL2}^{-1}\right]^{-1}$$
(6-5)

In a particular microfluidic system, if the thickness of the wall is much higher than the Debye length, the order of magnitude of the C_{wall} is smaller than C_{EDL} . This implies that the value of C_{wall} dominates C_{total} in Eq.(6-5). The change in zeta potential at the inner wall due to applied voltage across the total capacitance (ΔV_c) is (Lee et al. 1991)

$$\Delta \zeta = \frac{C_{total}}{C_{EDL2}} \Delta V_C \tag{6-6}$$

This equation tells us that the use of diluted solution and/or ultra thin wall thickness (small separation distance between the main and side walls d) enhance the influence of ΔV_c on the change in zeta potential, $\Delta \zeta$.



Figure 6-2 : (a)Equivalent electric circuit of field effect controlled device shown in Fig.6-1. A leakage capacitance model is considered due to the presence of leakage current through the capacitor. The capacitance of Stern layer is neglected in the present study because diffuse layer dominates the EDL capacitance; (b) and (c) Linear decomposition of Fig.6-2(a) for drain voltage and gate voltage, respectively.

For no-leakage capacitor models used in earlier studies (Lee et al. 1991; Ghowsi, 1991; Hayes et al., 1993a; Hayes, 1999; Schasfoort et al., 1999; Buch et al., 2001; Karnik et al., 2005; Lee et al., 2004a; Lee et al., 2004b), the applied voltage across the total capacitor can be estimated

by viewing the difference in the gate voltage and the local fluid voltage. However, in this study, leakage current from the side channel to the main channel is observed due to improper bonding and/or finite surface roughness (~150 nm) of channel materials, and this is consistent with the observation of Hayes et al. (1993a). Therefore, the estimation of applied voltage across the total capacitance is not straightforward anymore, and it requires consideration of leakage current.

6-2-2 : LEAKAGE CAPACITANCE MODEL

To fully understand the effects of leakage current in the zeta potential modification, we introduced a leakage capacitance model, considering the effective resistance parallel to the capacitors as shown in Fig.6-2 (a). Note that if there was no leakage current (so-called perfect capacitor), the leakage resistance would be infinity ($R_{leak} \rightarrow \infty$). For purposes of simplicity, the electrical circuit presented in Fig.6-2 (a) is decomposed into two separate circuits as shown in Fig.6-2 (b) and (c) using the superposition theorem (Temes and LaPatra 1977). Now we analyze the two separate circuits in order to determine applied voltage across the total capacitor. In Fig.6-2(b), the current through resistance R_{b1} along the main channel can be calculated as

$$I_{d} = \frac{V_{d}}{R_{b1} + (R_{b3} + R_{leak}) \parallel R_{b2}}$$
(6-7)

where the notation, ||, is employed to simplify the equivalent resistance in parallel (i.e. $R_A \parallel R_B \equiv R_A R_B / (R_A + R_B)$). The current passing through the resistance in the side channel (I_{c1}) is

$$I_{c1} = \frac{R_{b2}}{R_{b2} + (R_{b3} + R_{leak})} I_d$$
(6-8)

Also, the voltage at the center of the channel (V_1) can be analytically solved as

$$V_{1} = \frac{(R_{b3} + R_{leak}) || R_{b2}}{R_{b1} + (R_{b3} + R_{leak}) || R_{b2}} V_{d}$$
(6-9)

Similarly in Fig.6-2(c), the current through the side channel can be found as

$$I_{c2} = I_g = \frac{V_g}{(R_{b3} + R_{leak}) + R_{b1} \parallel R_{b2}}$$
(6-10)

and the potential at the controlled region (V_2) can be calculated as

$$V_{2} = \frac{R_{b1} \| R_{b2}}{(R_{b3} + R_{leak}) + R_{b1} \| R_{b2}} V_{g}$$
(6-11)

Note that the direction of the current I_{c1} is the opposite of I_{c2} . Under no control case, there is no net current through the leakage resistance ($I_{c1}=I_{c2}$), and the corresponding gate voltage (reference gate voltage) can be found as

$$V_{g0} = V_{g} \Big|_{I_{c1} = I_{c2}} = \frac{R_{b2}}{R_{b2} + (R_{b3} + R_{leak})} \frac{(R_{b3} + R_{leak}) + R_{b1} || R_{b2}}{R_{b1} + (R_{b3} + R_{leak}) || R_{b2}} V_{d}$$
(6-12)

Therefore, voltage drop across the capacitance for any gate voltage is

$$\Delta V_{C} = R_{leak} \left(I_{C2} - I_{C1} \right) = \frac{R_{leak} \left(V_{g} - V_{g0} \right)}{\left(R_{b3} + R_{leak} \right) + R_{b1} \parallel R_{b2}} = \frac{R_{leak}}{\left(R_{b3} + R_{leak} \right) + R_{b1} \parallel R_{b2}} \Delta V_{g}$$
$$= R_{leak} \left[\frac{V_{g}}{\left(R_{b3} + R_{leak} \right) + R_{b1} \parallel R_{b2}} - \frac{R_{b2}}{R_{b2} + \left(R_{b3} + R_{leak} \right)} \frac{V_{d}}{R_{b1} + \left(R_{b3} + R_{leak} \right) \parallel R_{b2}} \right]$$
(6-13)

where $\Delta V_g = V_g - V_{g0}$. The above relation shows that the voltage drop across the capacitor increases as the ΔV_g increases. Hence, in our leakage capacitance model, the zeta potential change can be estimated as

$$\Delta \zeta = \left(\frac{C_{total}}{C_{EDL2}}\right) \left(\frac{R_{leak}}{(R_{b3} + R_{leak}) + R_{b1} \parallel R_{b2}}\right) \Delta V_g$$
(6-14)

To obtain flow control in a straight microchannel, ΔV_g must be non-zero; for the sake of convenience all of the experimental results are presented for different values of ΔV_g .

6-3 : FABRICATION SCHEME

Using the soft lithography technique, a field-effect channel consisting of PDMS and glass is fabricated. The overall process is almost the same as each step in Fig.3-7. A disconnected sub-channel is also formed with a different mask as shown in Fig.6-3. Here, metal deposition is not needed since electrodes are directly inserted into each reservoir.


Figure 6-3 : Fabrication sequences for a glass-PDMS field-effect flow control device. The channel pattern shown above is a straight channel control system.

6-4 : RESULTS AND DISCUSSION

6-4-1 : PROBLEMS IN USING HIGH GATE VOLTAGE

Since electroosmotic flow velocity is directly related to the zeta potential (ζ), large flow control can be achieved by increaseing $\Delta \zeta$. However, Eqs.(6-2) and (6-14) indicate that the effect of gate voltage on the zeta potential change is not straightforward. As the gate voltage increases, the capacitance of EDL2 also increases, and saturation occurs at high gate voltage (Schasfoort et al., 1999). Moreover, at a very high gate voltage (on the order of 10³ [V]), an enormous amount of current passes through the PDMS wall because the breakdown voltage of PDMS is relatively low. Our separate experiment indicated that the dielectric strength of the PDMS is 21.18 [V/µm] (7.2 [kV] for 340 [µm]), which agrees with the specification of Dow Corning Corporation (http://www.dowcorning.com/). The resulting large current obtained from high electric field can easily open the disconnected region (capacitors in our model). The fluid then eventually leaks into the pathway. Application of this single-use-valve has been introduced by McDonald et al. (2001). However, at low electric field strength there is no fluid leakage, so our model is useful when the gate voltage difference is less than 100 [V].

6-4-2 : ORIGIN OF CURRENT LEAKAGE

Before starting the flow control by field effect, two separate experiments have been performed to make sure that there is no 'fluid leakage' between the main and sub channel. The overall setting of these experiments is the same as the one in Fig.6-1 where positive voltages are applied in gate and drain reservoirs, and the source reservoir is connected to the common ground. The first experiment is designed to make sure that there is no fluid leakage from the side channel to the main channel. In this experiment, dye (Rodamine) is mixed with the borate buffer solution inside the side channel, whereas the main channel is filled with 100 mM borate buffer only. For an applied gate voltage of 100[V] (corresponding electric field,

80 V/cm) and zero drain voltage for 5 hours, no fluid leakage into the main channel has been observed, although a measurable current of 20 [nA] is leaking from the side channel to the main channel. The current leakage is monitored using a precise current meter (Keithley Instruments, Inc., Model 6487) which is connected to the DC gate voltage source in series. These experiments are repeated for higher gate voltages of up to 300 [V]; no penetration of dyes to the main channel has been observed. The purpose of the second experiment is to confirm whether or not the current leakage behavior is universal in our microfluidic system or if it is only due to the presence of conductive liquid in the side channel. In order to prove this, the side channel is filled with a 2.5% w/w agarose gel instead of borate buffer. The gel is preheated at 80°C in water before loading. The entire chip is then cooled down to 2°C to solidify the gel for 3 [min]. The same current leakage phenomena is observed in this experiment as well, although the agarose gel required a slightly higher gate voltage (approximately 20[V] higher) compared to the field-effect control with borate buffer because of higher resistance. The results of these two experiments conclude that current passing takes place regardless of choice of conducting materials (borate buffer or agarose gel) used in the side channel, and that there is no fluid flow taking place through the 50 [µm] PDMS wall. This is in agreement with the results of others that PDMS material is not porous unless the surface is treated with laser or solid NaCl particles present in the buffer solution (Khorasani et al. 2005).

Next, the effects of gate voltage (V_g) on the leakage current behavior are observed. A separate experiment is conducted in order to determine the resistance of the leakage region by applying only control voltage (V_g) at the gate reservoir. In this experiment, both primary and side channels are filled with 100 mM borate buffer, the source reservoir is connected to the ground, and the electrode at the drain reservoir is kept electrically floating $(V_d=0)$ like in previous experiments. Using this configuration, there is no current through the resistance at

the first half of the main channel, $(R_{b1} \rightarrow \infty \text{ and } I_{ds}=0)$, and the current at the leakage resistance is recorded with a precision current meter. Experimental results show a linear relationship between the applied voltage and the measured current as

$$I_a = 0.2079 \, V_a - 1.0946 \tag{6-15}$$

where I_g is the measured current in nano amperes [nA] and V_g is the gate potential in volts [V]. Note that the inverse of slope of the linear relation presented in Eq.(6-15) is the total resistance ($R_{total} = 1/0.2079$ [GΩ]). Therefore, the resistance of leakage region (R_{leak}) can be estimated as

$$R_{leak} = R_{total} - (R_{b2} + R_{b3}) \tag{6-16}$$

where R_{b2} and R_{b3} are the resistances of the borate buffer in the main channel and sub channel, respectively. The values of R_{b2} and R_{b3} are calculated from the specific resistivity of borate buffer ($\rho_{Borate} = 1.83 [\Omega \cdot m]$) and channel geometries as 12.2 [M Ω] and 2.94 [M Ω] , respectively. Therefore, the resulting leakage resistance becomes 4.795 [G Ω]. Now if the PDMS wall is assumed to allow leakage current, the resistivity of the wall material for this experiment becomes $0.96 \times 10^6 \, [\Omega \cdot m]$. This value is significantly smaller compared to the bulk PDMS ($1.2 \times 10^{12} [\Omega \cdot m]$ Dow Corning Corporation, of the resistivity http://www.dowcorning.com/). It may be primarily due to the existence of nano-pores at the interface between PDMS and glass by knowing that PDMS is not acting like porous material although it is polymeric material (Khorasani et al. 2005). Right before bonding the two layers, the roughness of the plasma-treated PDMS and the coverslip are measured as 150 [nm] and 30 [nm], respectively by utilizing profilometer. The oxidized PDMS surface has relatively large roughness, so even after bonding with the other layer, tiny gaps are created which trap some portion of liquid when the buffer solution is loaded. This hypothesis explains why the

single-use-valve of McDonald et al. (2001) opens from the interface between the two layers. In other words, the interface has the weakest resistance against the flow of electrons. Each nano-pore seems to have a width of approximately 30 [nm]. Because the smallest thickness dominates the total value of the wall capacitance by considering Eqs.(6-4) and (6-5), we regard the effective thickness of the capacitor as the minimum size of these nano-pores rather than the thickness of the PDMS wall (i.e. use d = 30[nm] instead of $d = 50[\mu m]$) in this study.

6-4-3 : TIME DEPENDENCE CONSIDERATION

In this section, the time scale for the control voltage to come into steady state is analyzed. A finite amount of time will be required to establish the steady voltage across the capacitors since a leakage capacitance model is used in this study. To analyze the transient time scale, an equivalent electrical circuit corresponding to the gate voltage circuit (in Fig.6-2(b)) is presented in Fig.6-4. Here, another loop given by Fig.6-2(c) is neglected because the same drain voltage is maintained in order to see the effects of fluctuation of the gate voltage only. According to Thevenin's theorem, any linear circuit can be converted to an equivalent circuit of one voltage source, one series resistance, and a load component through a couple of simple steps (Williams 1973). In this study, the capacitor is first regarded as the load, and it is remove temporarily from the circuit to find Thevenin voltage. The voltage across the load terminal (Thevenin voltage) is identical to Eq.(6-13), and its value is approximately $V_{thev} = 0.998\Delta V_g$. On the other hand, Thevenin resistance is the effective resistance as seen from the load terminals without having all the power sources in the original circuit. By referring to Fig.6-4, the Thevenin resistance of this circuit is determined as

$$R_{\text{thev}} = (R_{b1} \parallel R_{b2} + R_{b3}) \parallel R_{\text{leak}}$$
(6-17)

The voltage level appears on the capacitor after the power supply is turned on and is given by Williams (1973) as:

$$\Delta V_C = V_{\text{thev}} \left(1 - e^{-t/(R_{\text{thev}}C_{total})} \right)$$
(6-18)

Table 6-1 shows the value of all resistances and capacitances for the straight channel experiments. At time $t = 4.8 R_{\text{thev}} C_{total} \approx 10.4 [\mu s]$, the voltage difference across the capacitor has quickly become 99.0% of the original voltage source where $C_{total} \approx 0.24 [\text{pF}]$. As the time passes, this voltage difference eventually reaches 99.8% of ΔV_{gs} according to Eq.18. Because all of the velocity data is acquired after 30 [sec] from initiation of the gate voltage change, the time dependency of all experimental data can be neglected.



Figure 6-4: Equivalent circuit of Fig. 6-2(c) considering Thevenin's theorem. The total capacitor is treated as the reactive component. Here, Thevenin voltage is $R_{leak} / \{(R_{b3} + R_{leak}) + R_{b1} || R_{b2}\} \Delta V_{gs}$, and Thevenin resistance is $(R_{b1} || R_{b2} + R_{b3}) || R_{leak}$.

R_{b1}	12.2 [MΩ]
R_{b2}	12.2 [MΩ]
R_{b3}	2.94 [MΩ]
R _{leak}	4.795 [GΩ]
R _{thev}	9.02 [MΩ]
C _{total}	1.22×10^{-10} [F]

Table 6-1 : Values of all resistances and capacitance for the straight channel

6-4-4 : FLOW CONTROL IN A STRAIGHT CHANNEL

The experimental set up for flow control in a straight channel is shown in Fig.6-1. Both the main channel and the sub channel have a width of 150 microns wide and a depth of 10 microns, while the length of the main and the side channels are 2 cm and 2.5 mm, respectively. To the best of our knowledge, we are the first group to investigate in great detail velocity distribution inside a field-effect-controlled microchannel using micro particle image velocimetry (μ PIV) technique. Since tracer particles used for the μ PIV detection system are negatively charged due to the presence of a carboxyl group (-COOH) at their surfaces, electrophoretic mobility of seeding particles is measured prior to the flow control experiment.

In order to estimate the electrophoretic velocity in our microfluidic system, the wall surface is coated with methylcellulose (1.1% w/w MC) before loading the buffer solution and tracer particles into the channel. It is noteworthy to mention that electrophoresis is a particulate flow phenomenon, so particle tracking velocimetry (PTV) has more physical meaning for the measurement of the electrophoretic velocities of each particle. However, this approach is more appropriate since the electroosmotic flow field is obtained by subtracting the electrophoretic velocity from the observed velocity field ($\vec{u}_{measure}$) obtained from µPIV. The PIV detection system provides a mean electrophoretic velocity field rather than

distinguishing each particle. In this experiment, the mean electrophoretic mobility of the seeding particles is determined as $M_{EP} = -1.3965 \times 10^{-4} \text{ [cm}^2/(\text{V} \cdot \text{s})\text{]}.$

Next, the electroosmotic mobility of 100 [mM] borate buffer is obtained in a glass-PDMS straight channel by subtracting the measured electrophoretic velocity from the observed flow field. The resulting electroosmotic mobility becomes $M_{EOF} = 2.3890 \times 10^{-4}$ [cm²/(V·s)]. Therefore, the Debye length and the zeta potential under no control conditions are calculated as $\lambda \approx 0.962$ [nm] and $\zeta \approx -35.2$ [mV], respectively, using the flow properties ($\varepsilon_0 = 8.8541878176 \times 10^{-12}$ [F/m], $\varepsilon_b \approx \varepsilon_{water} = 78.5$ [-], z = 1[-], R = 8.3144 [J/(mol·K)] , T = 25[C°] = 298.15[K] , F = 96485.3415[c/mol] , $c_{\infty} = 100$ [mM], $\mu = 1.025 \times 10^{-3}$ [Pa·s]).

For the field effect control experiment, both main and side channels are filled with 100 mM of borate buffer, and potentials are applied in the drain and the gate reservoirs using platinum electrodes. The controlled wall is placed at the right hand side (y=290 [µm] colored in purple in Fig.6-5(b)). The length of the control region is identical to the width of the side channel, although our results are shown only for 300 [microns] because of the field of view of our detection system. In this study, the gate voltage is modulated between 5 [V] to 45 [V] by keeping the drain voltage at a fixed value. The resultant value of the capacitance ratio becomes $C_{total} / C_{EDL2} = 0.0009 \sim 0.0004$ [-] in correspondence to $-19 < \Delta \zeta < 10$ [mV] and $-20 < \Delta V_{gs} < 20$ [V]. There are two reasons for not using higher gate voltages in this study. First, the EDL capacitance (C_{EDL2}) changes with the zeta potential, and, thus, saturation is expected to be seen at higher gate voltages. Second, at higher gate voltages, seeding particles used to track the flow become attracted to the control wall, creating nonuniform particle distribution within the microchannel followed by a flow obstacle in the vicinity of the channel

wall as well as inaccurate PIV vector calculations. Hence, the flow control at low gate voltage $(|V_g| < 40 \text{ [V]})$ is demonstrated in this chapter, and no reverse flow is observed due to the fact that the zeta potential in all cases never reaches positive value.

Figures 6-5 (a) and (b) demonstrate the field-effect electroosmotic flow results inside a straight microchannel for $\Delta V_g = 0$ [V] and 20 [V], respectively. As mentioned earlier, in order to see only electroosmotic flow phenomenon, the electrophoretic contribution of velocity is subtracted from the observed flow velocity obtained from the µPIV technique. In these experiments, the flow vectors in the EDL regions are not able to be captured because the spatial resolution of our μ PIV system is 5.36 [μ m] for a 20X objective lens. Therefore, the experimental results show velocity close to the wall, but not at the wall. Here, the drain and source voltages are kept constant at 50 [V] and 0 [V]. Equation (14) tells us that no change in zeta potential ($\Delta \zeta = 0$) will occur if there is zero current through the leakage resistance. For the above mentioned experimental setup, this can be achieved for a gate voltage of ~ 25 [V] simply because the side channel is located at the half length of the main channel. Hence, if the gate voltage is different from 25 [V], the flow control can be expected. In the case of Fig.6-5(b), the gate voltage for the control case is 45 [V], and the corresponding zeta potential at the controlled wall is -45.4 [mV]. Here, it is supposed that potential at the other side of wall is not significantly affected by the gate voltage due to thermal diffusion layer in EDL, so the zeta potential at the uncontrolled wall is assumed to be -35.2 [mV]. This zeta potential difference across the main channel contributes to lateral velocity in the microchannel from the bottom to the top direction (positive y direction as seen in Fig.7-6(b)). On the other hand, no such lateral velocity is observed in Fig.6-5(a) due to identical zeta potential at both side walls. The streamlines presented in Figs.6-5(a) and (b) indicate the mean flow direction in the straight microchannel.



Figure 6-5: The velocity distribution inside a straight channel for (a) no-control and (b) control cases. Here, drain voltage (V_d) is 50 [V], and the gate voltage (V_g) are adjusted at 25 [V] and 45 [V] for no-control and control cases, respectively. Streamlines show the net flow direction for both no-control and control cases.

Figure 6-6(a) shows the stream-wise velocity (u) profile across the main channel for different values of ΔV_g . Experimental evidences indicate that the electroosmotic velocity in the controlled case is significantly different from that of the no-control case. For straight

channel applications, experimental results are also compared with the leakage model prediction (in Table 2), and an excellent agreement is obtained between experimental values and theoretical predictions. As expected, for $\Delta V_g = 0$, stream-wise velocity is almost uniform throughout the channel. In this case the electroosmotic mobility $(M_{EOF} = 2.3890 \times 10^{-4} \text{ [cm}^2/(\text{V} \cdot \text{s})\text{]})$ is identical to that of a pure electroosmotic flow in a PDMS-glass microchannel (Horiuchi and Dutta, 2005). For the no-control case, the deviation from the uniform profile can be explained from the following facts. In the flow velocity estimation, the mean electrophoretic velocity is subtracted from the observed velocity under the assumption that the seeding particles are distributed uniformly and have identical charges and sizes. In practice, each particle experiences slightly different electrophoretic migration.

Table 6-2: Comparison of theoretical and experimental zeta potentials at the controlled surface in straight channel application. V_{con} is the velocity in the vicinity of the controlled region.

Gate		C_{EDL}/A (theory)	C_{Total}/A (theory)		Zeta potential (ζ) [mV]	
Voltage [V]	ΔV_{g}	$[F/m^2]$	[F/m ²]	V_{con}	experiment	theory
	[V]			$[\mu m / \text{sec}]$	1	5
5	-20	8.770E-01	8.101E-04	28.00	-16.5	-17.2
15	-10	1.186E+00	8.105E-04	47.00	-27.7	-28.8
25	0	1.524E+00	8.108E-04	60.00	-35.4	-35.4
35	10	1.875E+00	8.109E-04	70.00	-41.3	-39.9
45	20	2.178E+00	8.110E-04	77.00	-45.4	-43.4



Figure 6-6: (a) Stream wise (u) and (b) cross-stream (v) velocity distribution across the straight channel for different values of gate voltage at x= 175 [μ m]. Here, the reference gate voltage (V_{q0}) is 25 [V].

The cross-stream velocity (v) distribution across the channel is depicted in Fig.6-6(b) for different values of ΔV_g . For the planar channel considered in this study, the value of cross-stream velocity is almost uniform for a particular ΔV_g except at the wall region. In the vicinity of the wall, the velocity approaches zero due to no penetration at the side walls. Here, the little deviation from the exact uniform profile might be related to the procedure used in eliminating electrophoretic contributions. For positive values of ΔV_g , the cross-stream velocity is positive because the electrokinetic force acts from the bottom direction to the top direction and vice versa for negative values of ΔV_g . It is also clear that the magnitude of cross-stream velocity is proportional to the lateral electric field generated from the change in zeta potential at the side walls. These findings are consistent with existing electrokinetic theory where electroosmotic velocity is linearly proportional to the electric field.

6-4-5 : FLOW CONTROL AT A T-JUNCTION

T-junctions are widely used in microfluidic based "lab-on-a-chip" devices for sample extraction or loading. Although T-junction is an essential component of a microfluidic chip for automated sample handling, it is one of the major sources of dispersion for transporting samples such as protein and DNA. The primary reason for sample dispersion at a T-junction is the deformation (bending) of electric field lines as the current passes by the branch channel. Hence, in this chapter, we attempt to keep the streamlines straight by applying gate voltage from a field effect controlled transistor as one of the practical applications of the field-effect control device. The detailed experimental setup for the T-junction experiments is presented in Fig.6-7(a). The drain and gate reservoirs are connected to the positive voltages, and the source reservoir is connected to a common ground while the branch channel reservoir is

electrically floating. Initially, the pressure heads at each reservoir are maintained at the same value so that there is no movement of the fluid before applying the external electric field.

Figure 6-7(b) shows the fluid flow behavior under no-control scenario. Under this condition, the drain voltage is set as 48 [V] and the gate voltage is fixed at 25 [V]. This gate voltage corresponds to an equilibrium state in which the leakage current between the gate and source/drain is zero; we call this the reference gate voltage, V_{g0} . The streamlines (black solid lines), presented in Fig.6-7(b), show the direction of the flow in both main and branch channels under the above-mentioned electrical condition. It should be pointed out that the velocity distributions presented in Fig.6-7 are free from the electrophoretic component because the electrophoretic velocity is obviously nonuniform at the junction. The streamlines are expected to be distributed symmetrically about the axis of the branch channel (Patankar and Howard, 1998), yet the result revealed that some lines deviated from the main stream. This implies that the pressure is induced along the main channel due to electroosmotic pumping action, and the balance of the pressure among three reservoirs was broken. Moreover, the current meter which connects the gate and the source reservoirs may influence the electrokinetic potential distribution near the controlled region.

Now in order to control the flow, the leakage current is generated by intentionally changing the value of the gate voltage from its equilibrium state. Like the aforementioned straight channel experiment, this leakage current modifies the zeta potential in the controlled region. The change in the zeta potential on one side of the wall will result in a transverse electric field, creating a cross-stream velocity component. Figure 6-7(c) shows velocity distribution in a control case for which the gate voltage is 18 [V] ($\Delta V_g = -7$ [V]). For this case, almost no sample leakage to the branch channel is evident, and this specific scenario is particularly useful if one desires to transport the sample plug through a T-junction with

minimum band distortion. Here, the field effect flow at the T-junction acts like an electroswitching valve using the benefit of this cross-stream velocity component. Thus, the ability of controlling the cross-stream velocity component at this particular surface plays an important role in obtaining the desired flow pattern. For the control-case, the deviation of the streamwise velocity component, u, is also minimized (not shown here).

It is worthwhile to mention that by changing the value of ΔV_g the amount of the fluid into the branch channel can be precisely controlled. For example, increasing the gate voltage from its reference value leads to acceleration of the fluid entering the branch channel. On the other hand, for $\Delta V_g = -15$ [V] (in Fig.6-7(d)), fluid is coming from the branch channel into the downstream direction due to the scenario of over-control. In this case, the controlled leakage current from the gate voltage is more than geometric racetrack effect near the first corner of the branch channel. Hence, this field-effect flow demonstrates that there many potential applications in using locally controlled zeta potential.



Figure 6-7 (a) : Schematic view of experimental setup to control flow at the T-channel junction. Here, separation distances between the main channel and the sub channel are $d_1=50[\mu m]$ and $d_2=125[\mu m]$. The length of the main channel (L_{main}) and the sub channel (L_{sub}), and controlled region ($L_{control}$), are 2 [cm],2.5 [mm], and 1[mm], respectively. As shown in the figure, the length of the branch channel is the half of the main channel ($L_{branch}=0.5L_{main}$). In this study, drain voltage is fixed at 50 [V], while the gate voltage is modulated between 10 [V] and 35 [V]. The onset figure shows the formation of EDL next to the channel surface.



Figure 6-7 (b) : Vector plots of flow velocity in the T-channel junction for $V_g\text{=}$ 25 [V]



Figure 6-7 (c) : Vector plots of flow velocity in the T-channel junction for V_g =18 [V]



Figure 6-7 (d) : Cross-stream (v) velocity distribution at the entry of the branch channel (along A-A' line) for different values of gate potential. Here the reference gate voltage (V_{g0}) is 18 [V].

6-5 : CONCLUSIONS

Electroosmotic flow controls are demonstrated in microchannel geometries using field effect transistors. The microchannel structures and the field effect transistors are formed on PDMS, while a glass slide is used to cover the channels. The flow control in straight channels and T-channel junctions are obtained both qualitatively and quantitatively using an in-house micro scale particle image velocimetry technique. Experiments show that there was current leakage from the side channel to the primary channel if a potential is applied in the gate reservoir. In our PDMS-glass microchip, it is possible to control the flow with low gate voltage (on the order of 10^1 [V]); current leakage was present but fluid leakage did not occur. Although the

resistivity of PDMS and glass is very high, small amounts of current leakage (up to 20 [nA]) took place most likely through the interface between PDMS and glass surfaces due to their finite roughness or improper bonding. To explain the flow control at low gate voltages, a leakage capacitance model is proposed in this study.

Experimental results in a straight microchannel ensured the existence of secondary electroosmotic flow in the lateral direction due to asymmetric zeta potential distribution. In straight channel experiments, the flow can be controlled with a relatively low gate voltage (in the order of 10^1 [V]). The flow velocities obtained from control experiments are in agreement with those calculated based on leakage capacitance models.

In this study, the field effect control in a T-channel junction is particularly selected as a practical application. In the T-channel, the field-effect transistor acts as an electro-switching valve. The amount of flow coming from or going into the branch can be controlled by simply adjusting the gate voltage, which is located along the main channel in the upstream direction on the branch channel side. When an optimum gate voltage is applied ($\Delta V_{gs} = -7[V]$), the net flow in the direction of the branch channel reaches zero.

CHAPTER 7

ELECTROOSMOSIS WITH STEP CHANGES IN ZETA POTENTIAL

7-1: INTRODUCTION

A very high pressure gradient is needed to drive fluid flows through capillaries and microchannels due to the friction caused by shear stress (Hau et al., 2003). Thus, electrokinetic (EK) phenomena such as electroosmosis, streaming potential, electrophoresis, electroacoustics, etc., have been receiving more attention as effective methods of fluid and chemical transportation in microchannels. Electroosmosis and electrophoresis are the most popular EK phenomena because they enjoy a number of practical applications in molecular biology and medical science, such as the separation of deoxyribonucleic acids (DNAs), nucleic acids, viruses, small organelles, and proteins. Over the last fifteen years, capillaries have mainly been used for electrophoretic methods due to their high separation efficiencies and peak capacities. More recently, microchannels have attracted the attention of researchers because they are less expensive, simpler to fabricate, and more amenable to integration than capillaries.

In 1879, Helmholtz developed an analytical model for electroosmotic flow (EOF); Smoluchowski expanded upon Helmholtz's attempts in seeking to derive a formula for the electroosmotic velocity in 1921. According to their analysis, a plug-like velocity profile is expected and its amplitude is linearly proportional to the zeta potential (ζ) and the electric field. This is true if and only if the externally-applied electric field is uniform and both the ζ potential and channel cross section are constant.

For electrophoresis, some researchers may attempt to uniformly suppress electroosmotic flow (Markstrom et al., 2002; Loughran et al., 2003) while others may decide to take advantage of the electroosmotic flow (Maeso et al., 2004; Simal-Gandara, 2004) in

trying to increase performance. In either case, the surface should be electrokinetically homogeneous in order to avoid convective sample dispersion, which lowers separation efficiency. However, in practice, it is not easy to maintain a homogeneous surface with a uniform ζ -potential. Therefore, a deeper and more comprehensive understanding of the effects of the surface inhomogeneity on peak broadening is required. Some experimental work has shown that polymeric materials, e.g., poly-di-metheyl-siloxane, develop inhomogeneous surface charges or ζ -potential (Jo et al., 2000; Ma et al., 2000). Also, variation of solution pH near the channel surfaces strongly affects the ζ -potential distribution (Lambert and Middleton, 1990; Towns and Regnier, 1991; Ocvirk et al., 2000) and, hence, the electroosmotic flow.

The first theoretical treatment of nonuniform ζ -potential on electroosmotic flow was performed within capillaries (Anderson and Idol, 1985). They observed that the velocity of electroosmotic flow depends primarily on the distribution of ζ -potential along the wall, which in turn can either cause flow separation or circulation. Extensions of this work to shallow rectangular channels with wavy surface potential distribution (Adjari, 1995; 1996) and spot defects (Long et al., 1999) were undertaken ten years later. Anaslytical studies allowed for the discovery of recirculating flows, which may prove to be useful in the design of microfluidic mixers. Experimental work by Stroock et al. (2000) verified the existence of patterning electroosmotic flow in both longitudinal and transverse directions. Polyethylene glycol was utilized reduce the local surface charge of organic polymers. On the other hand, a timedependent electroosmotic flow with nonuniform ζ -potential due to chaotic advection was demonstrated by Qian and Bau (2002). However, all of the theoretical models mentioned above are limited to spatially periodic boundary conditions. Lately, Herr et al. addressed step changes in ζ -potential (Herr et al., 2000) in a capillary. Analytical and experimental results indicated the presence of induced pressure gradients and the associated band broadening. On the other hand, Dutta et al. (2002) evaluated the performance of a micropump numerically by sandwiching one type of material between another type of material with different values of ζ -potential. Later on, Christopher and Davis (2004) showed that sudden changes in either ζ -potential or cross section would cause distortion of the plug-like velocity profile. In those cases, they solved for the velocity distribution only in the hydraulically fully-developed region, but did not attempt to calculate the velocity distribution occurring in or near the region of step change.

This chapter presents an analysis of the effects of a step change in ζ -potential on flow in a two-dimensional straight microchannel. Such a step change might occur when an oxidative surface modification is carried out inhomogeneously, for example, by masking parts of the surface (Stroock et al., 2000). A step change may also be caused by the adsorption of particles to the particular regions on the channel wall (Herr et al., 2000). The velocity and pressure distributions near the step change are determined from the Navier-Stokes equations by first solving for the fully-developed velocities far upstream and downstream from the step, and then subtracting the upstream velocity profile from the general expression for the velocity. Next, the continuity equation is eliminated by substituting the stream function, and the pressure is eliminated by taking the curl of the conservation equation. The resulting biharmonic partial differential equation (PDE) is reduced to a fourth-order ordinary differential equation (ODE) in the transverse coordinate by application of the two-sided Laplace transformation technique. The appropriate no-slip conditions are applied to the solution of this ODE, which in turn is inversely transformed using Heaviside expansion. This solution in terms of the stream function is then used to generate the velocity and pressure fields near the step change.

7-2 : MATHEMATICAL MODEL

Consider two parallel plates confining an electrolyte solution as a model of our microchannel. Figure 7-1 defines origin of our coordinate system as well as the channel half height, *H*, and illustrates the velocity profile in far-field, $V_{x\infty}$. The superscripts, "-" and "+", indicate whether the variables are upstream (x < 0) or downstream ($x \ge 0$), respectively. The axial step change in the ζ -potential occurs at the position, x = 0, and causes a perturbation in the otherwise straight streamlines as illustrated in Fig. 7-1. Our goal is to find both the velocity and the pressure distribution throughout the channel, focusing especially in the region situated near the step change in ζ -potential at x = 0.



Figure 7-1 : Schematic view of mixed electroosmotic and pressure driven flow with different values on ζ -potential between upstream (x < 0) and down stream ($x \ge 0$) regions.

7-2-1 : Governing Equation and Boundary Conditions

If a uniform electric field, \vec{E} , is applied in axial direction throughout the microchannel, the electrolyte in that channel experiences an electric body force per unit volume, due to the

Lorenz force, where ρ_e equals net charge density. However, this electrical body force is active only within the Debye layer. If the Debye layer is infinitesimal compared to the channel depth, then it is possible to approximate the steady-state Navier-Stokes equations for incompressible, creeping flow, i.e., Re«1, as

$$\nabla \cdot \vec{V} = 0 \tag{7-1a}$$

$$-\nabla P + \eta \nabla^2 \vec{V} = 0 \tag{7-1b}$$

with replacement of the no slip boundary conditions by implicit slip boundary conditions (Anderson and Idol, 1985),

$$\vec{V} = \mu_{eo}\vec{E} = -\frac{\varepsilon\varsigma}{\eta}\vec{E}$$
(7-2)

where μ_{eo} is the electroosmotic mobility, ε is the dielectric constant, ς is the zeta potential, and η is constant dynamic viscosity coefficient. Here the ζ -potential is a function of axial position. This implies that a step change in ζ -potential is identical to the sudden change in EOF velocity along the channel wall. Note that the thin electric double layer (EDL) approximation is valid if the electrolyte ionic concentration is high enough, e.g., if univalent electrolytes are concentration $> \sim 1 \text{ mol/m}^3$ so that the EDL is significantly smaller than the typical channel depth of $\sim 10 \,\mu\text{m}$. Taking curl and the divergence of the momentum equation (7-1b), we find that both vorticity and pressure satisfy the Laplace equation. In the case of planar flows, both the vorticity and the stream-function vectors have a single scalar component as

$$\vec{\Omega} = \begin{bmatrix} 0\\0\\\omega \end{bmatrix} \text{ and } \vec{\Psi} = \begin{bmatrix} 0\\0\\\psi \end{bmatrix}$$
(7-3)

respectively, and hence, the vorticity can be related to the stream-function vector (Panton, 1996) as

$$\vec{\Omega} = \nabla \times \vec{V} = \nabla \times \nabla \times \vec{\Psi} = -\nabla^2 \vec{\Psi} \tag{7-4}$$

This becomes the biharmonic equation in terms of the stream-function vector

$$\nabla^2 \vec{\Omega} = -\nabla^4 \vec{\Psi} = \frac{\partial^4 \psi}{\partial x^4} + 2 \frac{\partial^4 \psi}{\partial x^2 \partial y^2} + \frac{\partial^4 \psi}{\partial y^4} = L^4(\psi) = L^2(L^2\psi) = 0$$
(7-5)

where the linear operator, L^2 , is defined as $L^2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}$. Equation (7-5) is a fourth-order

PDE so it requires four boundary conditions in y and four x. In this study, the electric field is continuous and one-dimensional, i.e., $\vec{E} = E \hat{e}_x$ because the channel length and width are infinitely long relative to the channel height and because electrostatic coupling of the electrolyte ions in the bulk solution has been ignored. With this in mind, the boundary conditions for the stream function are:

$$V_{y}\Big|_{y=\pm H} = -\frac{\partial \psi}{\partial x}\Big|_{y=\pm H} = 0 \qquad \text{for all } x \qquad (7-6a)$$

$$V_x^+\Big|_{y=\pm H} = \frac{\partial \psi^+}{\partial y}\Big|_{y=\pm H} = -\frac{\varepsilon \zeta^+}{\eta} E \equiv u^+ \qquad \text{for } x \ge 0$$
(7-6b)

$$V_x^{-}\Big|_{y=\pm H} = \frac{\partial \psi^{-}}{\partial y}\Big|_{y=\pm H} = -\frac{\varepsilon \zeta^{-}}{\eta} E \equiv u^{-} \qquad \text{for } x < 0 \tag{7-6c}$$

where $u - \frac{\varepsilon \varsigma}{\eta}$ is the pure EOF velocity without pressure gradient - also known as Helmholtz-Smoluchowski velocity.

7-2-2 : ASSUMPTIONS

- (i) Influence of conduit inlet and outlet at $x \to \pm \infty$ on the flow is ignored because the channel length is infinite compared to the channel height.
- (ii) The far-field flow and the uniform electric field are both one-dimensional. This yields ζ -potential variation only in axial direction.

- (iii) The ionic concentration is high enough to have infinitesimal EDL thickness relative to the channel height.
- (iv) The ion convection is negligible, so the Poisson-Boltzmann equation is valid in this study.
- (v) The dielectric constant is independent from the electric field.

7-2-3 : DERIVATION

Our solution was carried out in four basic steps: First, we solved for the velocity and pressure distributions far away $(x \rightarrow \pm \infty)$ from the interface between the two different ζ -potentials defined at x=0; we will refer to these expressions as the far-field solutions. Second, by subtracting the downstream far-field solution, x<0, from the entire domain, we ensure that the remaining solution goes to zero as $x\rightarrow-\infty$. This remaining near-field problem is solved by utilizing the double-sided Laplace transformation technique (van der Pol and Bremmer, 1959). Lastly, the downstream far-field solution is added back to the resultant near-field solution in order to obtain solutions in original field.

7-3-1: FAR-FIELD SOLUTION

Both the pressure gradient and the velocity lose their x-dependence far away from the origin, so the equations of motion are simplified to the ordinary differential equation

$$\frac{d^2 V_{x\infty}}{dy^2}^{\pm} = \frac{K^{\pm}}{\eta}$$
(7-7)

Applying boundary conditions given by Eqs.(7-6),

$$V_{x\infty}^{\pm} = u^{\pm} + \frac{3}{2} \left(\langle V_x \rangle - u^{\pm} \right) \left\{ 1 - \left(\frac{y}{H} \right)^2 \right\}$$
(7-8)

where $\langle V_x \rangle$ is the mean velocity which is defined as

$$\left\langle V_{x}\right\rangle \equiv \frac{1}{2H} \int_{-H}^{H} V_{x}(y) dy \tag{7-9}$$

and the far-field pressure gradient can be expressed as

$$K^{\pm} = \frac{3\eta}{H^2} \left(u^{\pm} - \left\langle V_x \right\rangle \right) \tag{7-10}$$

which is identical to the one presented by other researchers (Dutta and Beskok, 2001). The stream functions associated with the far-field solutions can be found by integrating Eq.(7-7) with the following boundary conditions:

$$\psi_{\infty}^{\pm} = \frac{y}{2} \langle V_x \rangle \left\{ 3 - \left(\frac{y}{H}\right)^2 \right\} - \frac{y}{2} u^{\pm} \left\{ 1 - \left(\frac{y}{H}\right)^2 \right\} - H \langle V_x \rangle \text{ for } \psi_{\infty}^{\pm} = 0 \text{ at } y = H$$
(7-11a)

where the difference between far-field stream functions is

$$\delta \psi_{\infty} = \psi_{\infty}^{+} - \psi_{\infty}^{-} = \frac{y}{2} (u^{-} - u^{+}) \left\{ 1 - \left(\frac{y}{H}\right)^{2} \right\}$$
(7-11b)

Similarly, the difference between the far-field pressures is

$$\delta P_{\infty} = (K^+ - K^-) x = \frac{3\eta}{H^2} (u^+ - u^-) x$$
(7-11c)

We will further incorporate Eqs. (11b) and (11c), later.

7-3-2: NEAR-FIELD SOLUTION

The near-field stream function with respect to downstream is defined as

$$\vec{\Psi}^* \equiv \vec{\Psi} - \vec{\Psi}_{\infty}^{-} = \begin{bmatrix} 0, \ 0, \ \psi - \psi_{\infty}^{-} \end{bmatrix}^T$$
(7-12)

$$\vec{V}^* = \left[V_x^*, V_y^*, 0\right]^T = \left[V_x - V_{x\infty}, V_y, 0\right]^T = \nabla \times \vec{\Psi}^* = \left[\frac{\partial}{\partial y}(\psi - \psi_{\infty}), -\frac{\partial \psi}{\partial x}, 0\right]^T$$
(7-13)

From Eqs.(7-6) and (7-7), the governing equation is

$$\Delta^{2}\psi^{*} = \nabla^{4}\psi^{*} = \frac{\partial^{4}\psi^{*}}{\partial x^{4}} + 2\frac{\partial^{4}\psi^{*}}{\partial x^{2}\partial y^{2}} + \frac{\partial^{4}\psi^{*}}{\partial y^{4}} = L^{4}(\psi^{*}) = L^{2}(L^{2}\psi^{*}) = 0$$
(7-14a)

and the boundary conditions are

$$V_{y}^{*}\Big|_{y=\pm H} = -\frac{\partial \psi^{*}}{\partial x}\Big|_{y=\pm H} = 0$$
(7-14b)

$$V_x^{*+}\Big|_{y=\pm H} = \frac{\partial \psi^{*+}}{\partial y}\Big|_{y=\pm H} = -\frac{\varepsilon(\zeta^+ - \zeta^-)}{\eta} E \equiv u^+ - u^-$$
(7-14c)

$$V_x^{*-}\Big|_{y=\pm H} = \frac{\partial \psi^{*-}}{\partial y}\Big|_{y=\pm H} = -\frac{\varepsilon(\varsigma^- - \varsigma^-)}{\eta} E \equiv u^- - u^- = 0$$
(7-14d)

It is convenient to use the unit (Heaviside) step function in order to combine Eqs.(7-14c) and (7-14d), resulting in

$$V_x^* \Big|_{y=\pm H} = \frac{\partial \psi^*}{\partial y} \Big|_{y=\pm H} = H_{(x)}(u^+ - u^-) \text{ where } H_{(x)} = \begin{cases} 0 & x < 0\\ 1 & x \ge 0 \end{cases}$$
(7-14e)

Two-sided Laplace transformation (van der Pol and Bremmer, 1959) is carried out by

$$\phi_{\psi}(s,y) \equiv \int_{-\infty}^{+\infty} \psi^*(x,y) e^{-sx} dx$$
(7-15)

which coverts the biharmonic equation Eq.(7-14a) into a fourth-order ordinary differential equation:

$$\frac{d^4\phi_{\psi}}{dy^4} + 2s^2 \frac{d^2\phi_{\psi}}{dy^2} + s^4\phi_{\psi} = \left(\frac{d^2}{dy^2} + s^2\right) \left(\frac{d^2}{dy^2} + s^2\right) \phi_{\psi} = 0$$
(7-16a)

The boundary conditions also need to be transformed into Laplace space, accordingly. From Eqs.(7-14b) and (7-14e), we arrive at

$$-s\phi_{\psi}\Big|_{y=\pm H} = 0 \tag{7-16b}$$

$$\left. \frac{d\phi_{\psi}}{dy} \right|_{y=\pm H} = \frac{u^+ - u^-}{s}$$
(7-16c)

The general solution of Eq.(7-16a) is

$$\phi_{\psi}(s, y) = C_1 \cos[sy] + C_2 y \cos[sy] + C_3 \sin[sy] + C_4 y \sin[sy]$$
(7-16d)

where the C_i are constants which can be calculated by using the four boundary conditions, Eqs.(7-16b,c). The solution to the problem in Laplace space is

$$\phi_{\psi}(s, y) = \frac{2(u^{+} - u^{-})(H\cos[Hs]\sin[sy] - y\sin[Hs]\cos[sy])}{s(2Hs - \sin[2Hs])} = \frac{N_{\psi}(s, y)}{D_{\psi}(s)}$$
(7-17)

where $N_{\psi}(s, y) \equiv 2(u^+ - u^-)(H \cos[Hs] \sin[sy] - y \sin[Hs] \cos[sy])/s$ and $D_{\psi}(s) \equiv 2Hs - \sin[2Hs]$ denote the numerator and the denominator of the solution. The Heaviside expansion technique (Churchill, 1958) is now used to invert the solution from Laplace space:

$$\psi^{*+} = \lim_{S \to 0} \left[s\phi_{\psi}(s, y) \right] + \sum_{\text{All } S_j < 0} \frac{N_{\psi}(s, y)}{\frac{\partial}{\partial s} D_{\psi}(s)} e^{S_x}$$
(7-18a)

$$\psi^{*-} = -\sum_{\text{All } S_j > 0} \frac{N_{\psi}(s, y)}{\frac{\partial}{\partial s} D_{\psi}(s)} e^{-S x}$$
(7-18b)

where s_j are the roots of the characteristic equation, $D_{\psi}(s_j) = 0$.

7-3-3: VELOCITY AND PRESSURE DISTRIBUTION

Using Eqs.(7-12) and (7-13), the original stream function and local velocity components are

found as

$$\psi^{\pm} = \psi^{*\pm} + \psi_{\infty}^{-} \tag{7-19a}$$

$$V_x^{\pm} = \frac{\partial \psi^{\pm}}{\partial y} = \frac{\partial \psi^{\pm}}{\partial y} + \frac{\partial \psi_{\infty}}{\partial y} = \frac{\partial \psi^{\pm}}{\partial y} + V_{x\infty}$$
(7-19b)

$$V_{y}^{\pm} = -\frac{\partial \psi^{\pm}}{\partial x}^{\pm} = -\frac{\partial \psi^{\pm}}{\partial x}^{\pm} - \frac{\partial \psi_{\infty}^{-}}{\partial y}^{\pm} = -\frac{\partial \psi^{\pm}}{\partial x}^{\pm}$$
(7-19c)

implying that the solutions given by Eqs.(7-11a) and (7-18) provide explicit analytic solutions. Similarly, near-field solution of pressure in Laplace space has a general solution as

$$\phi_{P}(s, y) \equiv \int_{-\infty}^{+\infty} P^{*}(x, y) e^{-Sx} dx = C_{5} \cos[sy] + C_{6} \sin[sy]$$
(7-20)

where C_5 and C_6 are constant, and $P^* \equiv P - K^- x$. By relating pressure to the velocity with Eq.(7-2) in Laplace space, we can find these constants

$$C_5 = -2\eta \, s \, C_2 \tag{7-21d}$$

$$C_6 = -2\eta \, s \, C_4 \tag{7-21e}$$

as well as the near-field pressure distribution,

$$\phi_{P}(s, y) = \frac{4\eta (u^{+} - u^{-})(\sin[Hs]\cos[sy])}{2Hs - \sin[2Hs]} = \frac{N_{P}(s, y)}{D_{P}(s)}$$
(7-22)

where $N_P(s, y) \equiv 4\eta (u^+ - u^-) (\sin[Hs]\cos[sy])$ and $D_P(s) \equiv 2Hs - \sin[2Hs] = D_{\psi}(s)$. Note that the denominator of Eq.(7-22) is exactly the same as the one in Eq.(7-17), explaining that the roots for the velocity coincide with pressure due to the characteristics of the momentum equation. For the inversion, the Heaviside expansion technique is again used.

$$P^{*+} = x \lim_{s \to 0} \left[s^2 \phi_{P}(s, y) \right] + \sum_{\text{All } S_j < 0} \frac{N_P(s, y)}{\frac{\partial}{\partial s} D_P(s)} e^{S_x}$$
(7-23a)

$$P^{*-} = -\sum_{\text{All } S_j > 0} \frac{N_P(s, y)}{\frac{\partial}{\partial s} D_P(s)} e^{-Sx}$$
(7-23b)

Downstream far-field pressure is then added:

$$P^{\pm} = P^{*\pm} + K^{-}x \tag{7-23c}$$

We list first-10 nondimensional roots ($\hat{s} \equiv H s$) determined by the Secant method with 5000 iterations in Table 7-1.

Table 7-1 : First 10 roots for our analytical solutions based on characteristic equation, $\hat{D}_{\psi}(\hat{s}_j) = \hat{D}_P(\hat{s}_j) = 2\hat{s}_j - \sin[2\hat{s}_j] = 0$.

J	ŝ _j <0	ŝ _j >0
1	$-3.748838138888 \pm 1.384339141494 i$	$3.748838138888 \pm 1.384339141494 i$
2	-6.949979856988 \pm 1.676104942427 i	$6.949979856988 \pm 1.676104942427i$
3	$-10.119258853915 \pm 1.858383839876i$	$10.119258853915 \pm 1.858383839876i$
4	$-13.277273632746 \pm 1.991570820170i$	$13.277273632746 \pm 1.991570820170i$
5	$-16.429870502535 \pm 2.096625735216i$	$16.429870502535 \pm 2.096625735216i$
6	$-19.579408260032 \pm 2.183397558835 i$	$19.579408260032 \pm 2.183397558835 i$
7	$-22.727035732178 \pm 2.257320224741 i$	$22.727035732178 \pm 2.257320224741 i$
8	$-25.873384151411 \pm 2.321713978526i$	$25.873384151411 \pm 2.321713978526i$
9	$-29.018831029547 \pm 2.378757559041 i$	$29.018831029547 \pm 2.378757559041 i$
10	$-32.163616856643 \pm 2.429958323949 i$	32.163616856643 ± 2.429958323949 <i>i</i>

7-4: REMARKS

We wish to address three points of significance based upon Eqs.(7-18) and (7-23).

7-4-1 : FINAL VALUE THEOREM

In order to confirm our inverse Laplace transformation, we apply the final value theorem to find the difference between the far-field stream functions at both ends from the solution to the near-field stream function in Laplace space. The theorem states that

$$\lim_{x \to \infty} [\psi^*(x, y)] = \lim_{s \to 0} [s\phi(s, y)] = \frac{y}{2}(u^- - u^+) \left\{ 1 - \left(\frac{y}{H}\right)^2 \right\}$$
(7-24a)

$$\lim_{x \to \infty} [P^*(x, y)] = x \lim_{s \to 0} [s^2 \varphi(s, y)] = \frac{3\eta}{H^2} (u^+ - u^-) x$$
(7-24b)

are identical to Eqs.(7-11b) and (7-11c), respectively. Equations (7-24a) and (7-24b) represent and provide physical meaning of the first terms found in Eqs. (7-14a) and (7-23a). Note that inverse Laplace transformation of $1/s^2$ is x.

7-4-2 : FILTERING OUT GIBBS RINGING

Our analytical solutions are basically trigonometric series solutions. When we try to reconstruct discontinuous functions with our series solutions, many terms are needed to avoid truncation error, and the solution exhibits Gibbs ringing (Arfken, 1985) near x=0. This problem is mitigated by using one of several available filters (Gottlieb and Shu, 1997). In this case, we utilized a Lanczos Sigma factor (Hamming, 1986) to reduce the Gibbs phenomenon, but we also tested a first-order Hölder and a Cesàro mean (van Der Pol and Bremmer, 1959) as a filtering method with satisfactory results.

The x component of nondimensional velocities across the channel at $\hat{x} = 0$ is illustrated in Fig. 7-2 on step-change in ζ -potential along the wall. Here, dashed lines are the solution in upstream, $\hat{V}_x^- \equiv V_{x\infty}^-/(u^+ - u^-)$, while solid lines indicate the solution in downstream, $\hat{V}_x^+ \equiv V_{x\infty}^+/(u^+ - u^-)$. The figure shows a dramatic improvement of accuracy by utilization of the filtering technique and by increasing the number of eigenfunctions.



Figure 7-2 : The *x* component of nondimensional velocities, $\hat{V}_x^- \equiv V_x^-/(u^+ - u^-)$ and $\hat{V}_x^+ \equiv V_x^+/(u^+ - u^-)$ given by Eq.(7-19b), across the channel at the junction ($\hat{x} = 0$) by applying step change in ζ -potential along the wall ($\hat{y} = \pm 1$) for (a) 10 terms in the series solution without filter, (b) 10 terms in the solution with filter, (c) 200 terms in the solution with filter. Here $u^+ = 1.0 \text{ [mm/s]}$, $u^- = 0$, and $\langle V_x \rangle = u^+$.

7-6 : CONVERGENCE AND CONTINUITY AT THE JUNCTION

While the same cannot be said for the discontinuous change in ζ -potential along the wall boundary, the streamfunction, velocity, and pressure should all display continuity at the junction across the channel from a physical point of view. Such continuity is accomplished by increasing the number of eigenfunctions in our series solutions. If only a few terms are considered, the solutions in upstream region (x < 0) and downstream region ($x \ge 0$) do not match at the interface (x = 0). On the other hand, as mentioned in the previous section the infinite series converges very slowly near x = 0. We are now in a position to test continuity between the upstream and downstream variables at the junction by estimating the nondimensional stream function differences based on Eqs.(7-11a), (7-18), and (7-19a),

 $\delta \hat{\psi} = \hat{\psi}^+ - \hat{\psi}^- \equiv \psi_{\infty}^+ / \{H(u^+ - u^-)\} - \psi_{\infty}^- / \{H(u^+ - u^-)\}$. As expected from Fig.7-2, filtering flattens out the wavy errors as manifested in Fig.7-3. By contrast, both filters swell the error near the wall boundary. In general, oscillations on the Fourier series solutions curve can be reduced by multiplying (n)-th eigenfunction by the Lanczos sigma factor: $\sin(n\pi/(N+1))/(n\pi/(N+1))$ where N is the number of roots used to express finite series solutions. For comparison and contrast of filtering performance, the Cesàro mean technique is also considered by multiplying by (1-(n-1)/N) instead of the Lanczos sigma factor. Increasing the number of eigenfunctions in series solutions helps to lower the amount of discontinuous error. By trading off between artificial errors caused by filtering near the wall and convergence time, we decided to use 200 terms with filtering in this study. This allows for both fast convergence and good matching at the junction. The maximum absolute error in Fig.7-2(c) is about 0.1%, which can be quantified from Fig.7-3(c). Using the Lanczos sigma factor seems to yield better performance for the most part, but also introduces higher frequency waves. Cesàro mean has less of an effect on high pitch waves, yet it is not able to

diminish the lower order terms, which badly affects the bulk region. Nonetheless, difference between two filtering techniques becomes invisible in the cases of velocity and pressure.



Figure 7-3 : The nondimensional stream function differences, $\delta \hat{\psi} = \hat{\psi}^+ - \hat{\psi}^- \equiv \psi_{\infty}^+ / \{H(u^+ - u^-)\} - \psi_{\infty}^- / \{H(u^+ - u^-)\}$ from Eqs.(7-11a), (7-18), and (7-19a) across the channel at the junction ($\hat{x} = 0$) by applying step change in ζ -potential along the wall ($\hat{y} = \pm 1$) for (a) 10 terms in the series solution without filter, (b) 10 terms in the solution with filter, and (c) 200 terms in the solution with filter. Here $u^+ = 1.0 \text{ [mm/s]}, u^- = 0$, and $\langle V_x \rangle = u^+$.

-0.001

(c)

7-5 : DISCUSSIONS

This section consists of three parts. One illustrates the properties of the roots corresponding to the eigenfunctions in the series solutions. Another one verifies the accuracy of our analytical solutions. The third section focuses mainly on recirculation and separation of the flow, Taylor dispersion, band broadening effects, and resulting band shapes in order to connect with practical implications in electrophoresis. All the results displayed in this writing were arrived at under the consideration of using deionized (DI) water within a microchannel 5µm in height (i.e. $\eta = 1.25 \times 10^{-3} \text{ kg/(m s)}$ and $H = 2.5 \times 10^{-6} \text{ m}$), although the solution to the biharmonic equation will apply even with the use of a wide variety of fluids and microchannels of different geometrical proportions.

7-5-1 : BASE SOLUTION

From Table 7-1, it is obvious that all of the roots are complex and that the roots appear in conjugate sets of four. In other words, if \hat{s}_i is an eigenvalue, so are $-\hat{s}_i$ as well as complex conjugates of both \hat{s}_i and $-\hat{s}_i$. Care must be taken to ensure that $\hat{s} = 0$ is not a root and that there are no repeated eigenvalue. The real parts of the roots increase approximately by $(n+1/4)\pi$ for large *n*. The corresponding positive imaginary parts increase at a much slower rate. More importantly, the near-field solutions given by Eqs.(7-18) and (7-23) decay exponentially to zero in both axial directions where the real part of roots is the spatial decay parameter. The real part in the leading eigenvalue ($\operatorname{Re}[\hat{s}_1] \cong \pm 3.745$ as shown in Table 7-1) is the most significant number among all roots since the first decay parameter (i.e. principal eigenvalue) primarily localizes where the near-field solutions vanish. The prediction of complex roots is typically executed by using numerical techniques such as Newton's method and the Secant method in order to maximize accuracy. Although the eigenvalues are rounded
to 12 digits to the right of the decimal point in Table 7-1, all the calculations are actually done with higher precision numbers (at least 20 digits).

7-5-2 : PRACTICAL IMPLICATIONS

In case of undulated surface charges by Ajdari (1995), $\sigma = \sigma_{max} \cos(qx + \gamma)$, the axial EOF velocity component is taken to be $u = u_{max} \cos(qx + \gamma)$ where q and γ are constant. The flow recirculation shown by their work leads to band broadening, which reduces electrophoretic efficiency. In reality, patterning surface charge may not be able to be expressed by cosine function at all times. It is more natural to regard the modified surface as a discontinuous boundary (Abraham et al, 2000). Additionally, different materials also offer the step change in ζ -potential (Dutta et al, 2002). If there is a sudden change in ζ -potential at a particular location of the channel, the flow pattern is illustrated in Fig.7-4(a) by computing streamlines given by Eqs.(7-18) and (7-19a). Here symmetric EOF velocity ($u^+ = -u^-$) with no flow rate, $\langle V_x \rangle = 0$, is considered. The fluid approaches very close to the wall to satisfy discontinuous boundary conditions. Figure 7-4(b) describes anticipatable distorted band shape at each location. For instance, if a sample is located in far-field region, its shape is skewed like the shape of a "crescent" because the flow around the center of the channel has opposite direction from the region close to the wall as shown in Fig.8-4(a). It seems that step changes in ζ potential may result in less effect from recirculation in contrast to Ajdari's work. Nevertheless, skews of the sample band are actually inevitable.



Figure 7-4 : Applying step change in ζ -potential along the wall ($\hat{y} = \pm 1$) for $u^+ = 1.0 \text{ [mm/s]}$, $u^- = -u^+$, and $\langle V_x \rangle = 0$, (a) the nondimensional streamlines given by Eqs. (7-18) and (7-19a), and (b) possibilities of sample band deformation due to this flow pattern are presented. All bands are skewed at each location.

Figure 7-5 explains dependences of the nonzero mean velocity on streamlines. the symmetrical step change condition, $u^+ = -u^-$, is maintained to observe how the bulk velocity affects flow patterns. In Fig.7-5(a), $\langle V_x \rangle = 0.1u^+$ is applied; some of the fluid partially travels toward the down stream direction, especially at the center of the channel upstream. Thus, intersection is shifted downstream. The more the flow rate is increased, the more fluid is transported to the downstream direction. The interaction will eventually vanish as the flow rate is increased, and most of the fluid will now head downstream as shown in Fig.7-5(b). Consequently, the higher mean velocity moderates effects of discontinuity. Due to the fact that u^- is negative, reverse flow happens near the wall boundary at the junction even if relatively high flow rate is applied. In Fig.7-5(c), almost all streamlines are situated parallel to each other in the far-field region of the downstream side. There should not be any distortion of the sample in the region where longitudinal location, x, is equal to or greater than one characteristic length, H, namely $\hat{x} \ge 1$ according to Fig.7-5(c).



Figure 7-5 : Influences of the mean velocity on the nondimensional streamlines given by Eqs.(7-18) and (7-19a) for (a) $\langle V_x \rangle = 0.1 u^+$, (b) $\langle V_x \rangle = 0.5 u^+$, and (c) $\langle V_x \rangle = 1.0 u^+$. Here symmetrical step change in ζ -potential is maintained (e.g. $u^+ = 1.0 \text{ [mm/s]}$ and $u^- = -u^+$).

We must point out that changing the mean velocity requires consideration of band broadening effect and Taylor dispersion. In the van Deemter equation (van Deemter et al., 1956)

$$HETP = A + B \frac{1}{\langle V_x \rangle} + C \langle V_x \rangle$$
(7-25)

HETP is the theoretical plate height, and a lower value of *HETP* stands for a higher sample separation efficiency. Here *A*, *B*, and *C* are constants for a particular system. The first term indicates eddy diffusion, which is simply a constant. The second term represents the longitudinal diffusion, whereas the third term represents the resistance to mass transfer. Both the second and third terms greatly depend on the mean velocity, $\langle V_x \rangle$. Originally, Eq.(7-25) was introduced for gas chromatography (van Deemter et al., 1956), but the equation applies for liquid chromatography as well. In the case of microchannel electrophoresis, eddy diffusion can be eliminated since carrier liquid is a single phase and uniformly filled. Nevertheless, as long as the second and third terms exist, attention needs to be paid for the process to finite values of $\langle V_x \rangle$.

It is well documented that Taylor dispersion also causes band broadening because the velocity profile across the channel is not plug-like in mixed electroosmotic and pressure driven flows. The effective dispersion coefficient for Poisseuille flow in a tube is $D + \langle V_x \rangle^2 r^2 / (48D)$ (Guell, 1987) where *D* is the molecular diffusivity and *r* is the tube radius, while in the case of two parallel plates the coefficient is $D + \langle V_x \rangle^2 H^2 / (210D)$ (Beard, 2001). Beard's work draws attention to the treatment of mean velocity in our study. Indeed, dispersion in the longitudinal direction is higher in Fig.7-5(c).

We now readjusted the back the mean velocity back to zero again, and vary the EOF velocities (or ζ -potentials) in upstream, u^- , and in downstream, u^+ , separately. We no longer have a constraint, $u^+ = -u^-$, so that unsymmetrical step change in ζ -potential is carried out. Figure 7-5 depicts streamlines when the relationship between u^- and u^+ is arbitrary. The band probably forms a "crescent" shape on the downstream side in Fig.7-6(a). Since there is almost no velocity component in the upstream region, the bandwidth is not affected by the flow until it reaches the downstream area. Figures 7-6(b) and (c) have the similar trend in the far-field region of the downstream side, yet they show different flow profiles before approaching the interface between upstream and downstream. The sample may be shaped like a "planar concave" near the junction in Fig.7-6(b) for $u^- = -0.5 u^+$. If the same direction but different magnitude of the EOF velocity is applied in both upstream and downstream, a shape between a "crescent" and a "trapezoid" will be obtained as shown in Fig.7-6(c). This kind of parametric study allows for a vriety of different resulting shapes including the bi-convex, biconcave, trapezoid, rectangle, half moon, and crescent facing either left or right. Unless the sample forms a perfect rectangular shape, the band is skewed somehow due to the existence of flow variation.



Figure 7-6 : Influences of the relationship between upstream EOF velocity (u^{-}) and downstream EOF velocity (u^{+}) on the nondimensional streamlines given by Eqs. (7-18) and (7-19a) for (a) $u^{-} = 0$, (b) $u^{-} = -0.5u^{+}$, and (c) $u^{-} = +0.5u^{+}$. Here, no volume flow rate (i.e. $\langle V_x \rangle = 0$) and $u^{+} = 1.0$ [mm/s] are maintained. Slight changes of the

relationship between u^- and u^+ significantly affects flow pattern as well as the shapes of the sample.

The pressure gradient is also important to predict dispersion of samples. We present nondimensional pressure distribution $(\hat{P}^{\dagger} \equiv P^{\pm} / \{u^{+} - u^{-})\eta / H\})$ in Fig.7-7. By recalling the results in Fig.7-6, the corresponding pressure distribution is manifested in Fig.7-7(a). Spikes shown in all cases at the wall on the interface between upstream and downstream indicate that there are flows toward these holes. This suggests that the velocity in cross-stream direction cannot be ignored in contrast to the hypothesis made by Ren and Li. The lateral component of velocity is experimentally observed in the previous chapter (Chap 6). Band distortion happens especially near the wall where discontinuous ζ -potential is applied. Figure 7-7(b) represents the pressure distribution under the same conditions as those in Fig.7-7(a). In spite of no flow rate (i.e. $\langle V_{x} \rangle = 0$), samples in the downstream region seem to move toward the upstream direction by induced pressure gradient. Strictly speaking, flat pressure profile in upstream space does not allow too much intrusion of the samples in order to maintain mass conservation. The flow rate is controlled to achieve the same mean velocity as u^+ preserving $u^- = 0$. As expected, the flow of the entire region travels in the downstream direction as shown in Fig.7-7(c).



Figure 7-7 : The nondimensional pressure distribution given by Eq.(7-23) by applying step change in ζ -potential along the wall ($\hat{y} = \pm 1$) for (a) $u^- = -u^+ \& \langle V_x \rangle = 0$, (b) $u^- = 0 \& \langle V_x \rangle = 0$, (c) $u^- = 0 \& \langle V_x \rangle = 1.0 u^+$. Here $u^+ = 1.0 \text{ [mm/s]}$. Spikes near the wall on the interface ($\hat{x} = 0$, $\hat{y} \approx \pm 1$) indicate that there are flows toward these holes.

Finally, we demonstrate velocity field in this section. First of all, normalized streamwise velocity component, \hat{V}_x , is presented in Fig.7-8 for the identical flow parameters used in Fig.7-7. The exact same process is repeated for nondimensional cross-stream component, \hat{V}_y , in Fig.7-9. We can see how the velocity develops under the step ζ -potential boundary conditions. Parabolic velocity profiles in Figs.7-8(a) and (c) promises the existence of pressure gradient, which corresponds to pressure distribution in Fig.7-7. In other words, there is no axial velocity in upstream region in the case of Fig.7-8(b) as expected from Fig.7-7(b). Moreover, maximum velocity coincides with the magnitude of the mean velocity. On the other hand, it is evidently proved that velocity in y-direction occurs surrounding the junction for all cases as shown in Fig.7-9. This is also observed in Fig.7-7. From the figure, a flow travels from center of the channel to the wall boundary near $\hat{x} = 0$.



Figure 7-8 : The nondimensional streamwise velocity distribution given by Eqs.(7-19b) by applying step change in ζ -potential along the wall ($\hat{y} = \pm 1$) for (a) $u^- = -u^+$ & $\langle V_x \rangle = 0$, (b) $u^- = 0$ & $\langle V_x \rangle = 0$, (c) $u^- = 0$ & $\langle V_x \rangle = 1.0 u^+$. Here $u^+ = 1.0 \text{ [mm/s]}$, there is no axial velocity in the upstream region in the case of (b). The other cases show parabolic velocity profile.



Figure 7-9 : The nondimensional cross-stream velocity distribution given by Eqs.(7-19c) by applying step change in ζ -potential along the wall ($\hat{y} = \pm 1$) for (a) $u^- = -u^+ \& \langle V_x \rangle = 0$, (b) $u^- = 0 \& \langle V_x \rangle = 0$, (c) $u^- = 0 \& \langle V_x \rangle = 1.0 u^+$. Here $u^+ = 1.0 \text{ [mm/s]}$, locations of spikes near the wall on the interface ($\hat{x} = 0$, $\hat{y} \approx \pm 1$) coincide with those in Fig.7-7.

7-6 : CONCLUSIONS

One of the aims of our work is to solve the biharmonic equation with double-sided Laplace transformation. Many problems including potential flow, wave propagation theory, and elasticity in engineering and physics often lead to the mathematical formulation of biharmonic equations. There are a tremendous number of numerical and analytical techniques performed in order to solve this biharmonic equation, but to the best of the authors' knowledge, no explicit analytical solution with discontinuous ζ -potential has been found in infinite domain so far. Consequently, our solutions may impact not only microfluidics or electrophoretics but also other fields of science. For uniqueness and existence of the solutions to the biharmonic equation in Cartesion coordinates, see Benedikt's paper (Benedikt 2002).

The explicit mathematical expressions are found for the streamfunction, the velocity, and the induced-pressure inside a straight microchannel with nonuniform (either continuously or discontinuously) ζ -potential variation. In this paper, step change in ζ -potential is emphasized. This happens when partial surface modification or two different materials are considered. The validation check is executed by comparing our solutions with other studies.

Practically, our analytic solutions can be used to optimize band forming processes such as IEF. Here, the flow pattern in terms of the channel depth, ζ -potentials, and the mass flow rate can be controlled. To be specific, input parameters are either the pressure at two points in the microchannel or the flow rate. In practice, the flow rate is the preferable input parameter although pressure can be interchangeable if known. The theoretical treatment (double-sided Laplace transformation) of a biharmonic equation is straightforward, so our analytic solutions may impact not only microfluidics or electrophoretics but also other fields in science such as elasticity. Filtering techniques (particularly Lanczos sigma factor and Cesàro mean) dramatically reduce the Gibbs ringing, so 200 terms in the series solution represent good results with reasonable convergence speed. Indeed, continuity at the junction indicates the accuracy of our results.

In this chapter, it is observed how the variation of flows contributes hydrodynamic dispersion, band broadening, flow separation, and recirculation. If the ζ -potential is discontinuously changed, the distortion of a sample band due to the flow pattern takes place. A sample shape of bi-convex, bi-concave, trapezoid, rectangle, half moon, and a crescent facing left and right are examined. Increasing flow rate moderates mathematical discontinuity at the junction, but care should be taken that the mean velocity influences band broadening and Taylor dispersion. On the other hand, the analytical pressure distribution suggests that there are flows toward the wall at the interface between upstream and downstream.

CHAPTER 8

SUMMARY AND SUGGESTIONS FOR FUTURE WORK

8-1: SUMMARY

8-1-1 : MIXED ELECTROOSMOTIC & PRESSURE DRIVEN FLOWS IN A TRAPEZOIDAL MICROCHANNEL

The experimental results of mixed electroosmotic and pressure driven flows in a trapezoidal shape microchannel was presented. Microchannels were formed on poly di-methyl siloxane using soft lithography technique, while micro particle image velocimetry (µPIV) was utilized to acquire velocity profile across the channel for pressure, electroosmotic and mixed electroosmotic-pressure driven cases. In mixed flow studies, both favorable and adverse pressure gradient flows were considered. Flow results obtained from µPIV were compared with three dimensional numerical predictions, and an excellent agreement was obtained between them. In numerical techniques, electric double layer was not resolved to avoid expensive computation, rather a slip velocity was assigned at the channel surface based on the electric field and electroosmotic mobility. This study showed that trapezoidal microchannel provides tapered-cosine velocity profile if there is any pressure gradient in the flow direction. This result was significantly different from that observed in rectangular microchannel. Our experimental results verify that velocity distribution in mixed flow can be decomposed into pressure and electroosmotic driven components.

8-1-2 : ELECTROOSMOTIC FLOW CONTROL IN MICROGEOMETRIES USING FIELD EFFECT

A field-effect transistor was developed to control flow in microfluidic chips by modifying the surface charge condition. By applying a gate voltage to one side of the microchannel wall,

zeta potential at that side was altered, while the zeta potential at the other side was maintained at the original value. This non-uniform zeta potential resulted in a secondary electroosmotic flow in lateral direction, which was used for flow control in microchannel geometries. In this study, microchannel structure and field effect transistors were formed on PDMS using soft lithography techniques, and a μPIV technique was used to obtain high resolution velocity distribution in the controlled regions. The flow control was observed both quantitatively and qualitatively at relatively low voltage (less than 50 [V]), and this local flow control was primarily due to the leakage current through the interface between PDMS and glass layers. A leakage capacitance model was introduced to estimate the modified zeta potential for straight channel case, and an excellent agreement was obtained between predicted and experimental zeta potential. This leakage-current based field-effect was then applied to T-channel junction to control flow in the branch channel. Experiments showed that the amount of discharge in the branch channel can be controlled by modulating gate voltage.

8-1-3 : ELECTROOSMOSIS WITH STEP CHANGES IN ZETA POTENTIAL

An analytical solution for two-dimensional fluid flow in a rectangular microchannel in the vicinity of a step change in the surface zeta potential was presented. The stream function was determined from the creeping flow approximation to the Navier-Stokes equations assuming a fixed volumetric flow, a constant electric field and thin, symmetric double layers. The resulting biharmonic equation was solved using a double-sided Laplace transformation which was then inverted by a Heaviside expansion. The resulting series solution provided closed-form expressions for the velocity and pressure fields which demonstrated how the recirculating flows generated by an abrupt change in the zeta potential may contribute both locally and globally to the hydrodynamic dispersion observed in straight microchannels.

8-2 : SUGGESTIONS FOR FUTURE WORK

For future work, there are a couple of things that can be done using the same facilities with minimal modifications. First of all, temperature measurement is possible by measuring Brownian motion of seeding particles (Park et al., 2005; Chamarthy et al., 2005). The resulting temperature field can then be compared with some existing analytical solutions (Horiuchi and Dutta, 2004; Horiuchi et al, 2005).

Local flow control using leakage current field-effect can be applied to not only Tchannels but also many other micro geometries. For example, the racetrack effect along a serpentine channel can be minimized when the gate voltage is applied along the outer curved line (Lee et al., 2004b). The local dispersion causes deformation of sample species, so straightened velocity profiles due to the modification of zeta potential on one side helps to avoid distortion of band shape. The same idea should be applicable to other geometries in which cross-sectional area suddenly expands. Moreover, complicated flows at the crossroads are expected to be able to control by applying the similar gate voltages to the T-channel results.

Analytical solution for the continuous zeta potential can be shown by simply changing the boundary condition on the wall, and it should be compared with numerical work performed by Ren and Li (2001), who were able to derive the solution numerically. This continuous boundary condition is an example of practical application in that the zeta potential is varied in correspondence with the pH gradient continuously for IEF. The experimental work by Cui et al. (2005) should be referred and we should check and compare our results with theirs to look for agreement. Additionally, end effects (effects of reservoirs) should be addressed by investigating a new approach within finite domain unlike our current problem definition. Because we possess the technologies to fabricate, detect, and analyze mixed electroosmotic and pressure driven flow in two dimensional microchannels with variable zeta potential, the analytical solution including end effects can be compared with experimental results in the future.

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