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Effects of ionic concentration gradient on electroosmotic flow mixing in a microchannel

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ABSTRACT

Effects of ionic concentration gradient on electroosmotic flow (EOF) mixing of one stream of a high concentration electrolyte solution with a stream of a low concentration electrolyte solution in a microchannel are investigated numerically. The concentration field, flow field and electric field are strongly coupled via concentration dependent zeta potential, dielectric constant and electric conductivity. The results show that the electric field and the flow velocity are non-uniform when the concentration dependence of these parameters is taken into consideration. It is also found that when the ionic concentration of the electrolyte solution is higher than 1 M, the electrolyte solution essentially cannot enter the channel due to the extremely low electroosmotic flow mobility. The effects of the concentration dependence of zeta potential, dielectric constant and electric conductivity on electroosmotic flow mixing are studied. © 2014 Published by Elsevier Inc.

1. Introduction

Flow mixing in microchannels is an essential step for realizing biological and chemical reactions in lab-on-a-chip devices [1]. Electroosmotic flow (EOF), as an excellent transport method, has been widely used to pump chemical and biological reagents in microchannels because of its significant advantages over pressure-driving flow, i.e., easy to control, plug-like velocity profile and no mechanical moving parts. Electroosmotic flow based mixing is also widely used in microfluidic systems. Generally, electroosmotic flow based mixing can be categorized as laminar flow mixing and chaotic mixing [2]. In laminar flow mixing, flow velocity is low, no turbulence exists in the system, and the mixing of two liquid steams in the microchannel mainly depends on molecular diffusion. In the case of chaotic EOF mixing, the vortexes generated by the electrical field induced electroosmotic flow are used to enhance the mixing.

A variety of experimental and theoretical research has been done to study the mixing process in microfluidic systems. Most of them focused on enhancing the efficiency of mixing by passive or active methods. For passive mixers, complex channel geometries were used to increase the interaction area between the mixing

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mixing. Lin [11] studied EOF mixing by controlling a gradient distribution of zeta penitential by changing the frequency of electric power applied on the shielding electrodes along the channel walls. Induced-charge electrokinetic flow (ICEKF) is also a novel method to enhance the mixing efficiency and to control the flow rate by controlling the vortices of the induced charge electroosmotic flow [12–14]. It should be noted that all these works reported in the literature did not consider the ionic concentration effects on both the EOF and EOF mixing process. The ionic concentrations of most buffer solutions used in microfluidic systems are higher than 10 mM, giving rise to a thin electrical double layer (EDL) with a thickness on the order of 10 nm. For thin EDL, the velocity of the EOF can be calculated from the Helmholtz–Smoluchowski equation [15]:

liquids to achieve complete mixing within a short transport distance [3–6]. On the other hand, active mixing methods introduce external energy sources into the mixing process to enhance the

mixing efficiency [6,7]. Using variable zeta potentials, Erickson

[8] introduced oppositely charged surface patches into the micro-

channel, and obtained localized flow circulations to enhance the

mixing. Glasgow and Lin [9,10] designed T-form EOF mixers and

switched EOF alternatively by changing the electrical field period-

ically to control the flow rates of the two streams to enhance

$$\vec{V}_{EOF} = \varepsilon \zeta \vec{E} / \eta, \tag{1}$$

where ε is the local dielectric constant, ζ is the zeta potential of the solid boundary, η is the viscosity of the solution and \vec{E} is the





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Abbreviations: EOF, electroosmotic flow; ICEKF, induced-charge electrokinetic flow; EDL, electrical double layer.

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external electric field. All these parameters contribute to the EOF velocity and hence affect EOF mixing. It should be realized that the ionic concentration will directly affect the dielectric constant and the electrical conductivity of the electrolyte solution, as well as the zeta potential. However, the dependence of these parameters on the ionic concentration was not considered in previous studies of EOF mixing.

In this work, the ionic concentration effects on electroosmotic flow and the EOF mixing were investigated. A finite element numerical model was developed to study the ionic concentration effects on electroosmotic flow mixing in a straight microchannel. Concentration dependent zeta potential, dielectric constant and electrical conductivity were used in the model, which makes the electric field, the concentration field and the flow field fully coupled. The influences of ionic concentration on electroosmotic flow mixing are discussed especially for very high concentration difference between the two mixing streams.

2. Physical and mathematical models

Fig. 1 shows the top-view of the simplified flow-mixing system to be modeled in this work, two streams of different electrolyte solutions entering a straight microchannel under an applied electrical potential difference, V_1 at the entrance of the channel and V_2 at the exit. For simplicity, the electrolyte solutions are considered as aqueous NaCl solutions. The two streams of NaCl solutions have different concentrations (C_1 and C_2 , respectively). The microchannel is 200 µm in width, 50 µm in depth and 2 mm in length. The channel walls are made of glass. Because there is no concentration gradient in the channel depth direction, and the identical top and bottom channel walls will not affect the EOF in the width and length directions, this system can be simplified as a 2-D model as shown in Fig. 1 and this treatment will not affect the conclusions on EOF mixing of this paper.

2.1. Governing equations and boundary conditions

2.1.1. Electric field

The walls of the glass microchannel are electrically non-conducting. The electric current conversation in the channel must be satisfied, that is

$$\nabla \hat{\imath} = 0, \tag{2}$$

where \hat{i} is the current in the microchannel and can be written as

$$\hat{\imath} = \lambda(c) \vec{E} \,. \tag{3}$$

In the above equation, $\lambda(c)$ is the electric conductivity and a function of ionic concentration; the electric field, \vec{E} , can be calculated by the electric potential φ gradient:

$$\vec{E} = -\nabla \phi \tag{4}$$

Combining Eqs. 2–4, the electric field in the channel can be described by

$$\nabla(-\lambda(c)\nabla\varphi) = \mathbf{0}.$$
(5)

The boundary conditions of the electric field are given by the following:

$$\hat{n}\nabla \varphi = 0$$
 at the channel walls; (6a)

$$\varphi = V_1$$
 at the inlet; (6b)

 $\varphi = V_2 = 0$ at the exit. (6c)

2.1.2. Flow field

As the electroosmotic flow of the aqueous electrolyte solutions is an incompressible laminar flow, the flow field can be calculated by Navier–Stokes equation and the continuity equation as follows [16]:

$$\rho \left[\frac{\partial \vec{u}}{\partial t} + \vec{u} \nabla \vec{u} \right] = -\nabla P + \mu \nabla^2 \vec{u} + \vec{E} \rho_e - \frac{1}{2} \vec{E}^2 \nabla \varepsilon, \tag{7a}$$

$$\nabla \cdot \vec{u} = 0 \tag{7b}$$

where ρ is the density of the solution, *t* is the time, \vec{u} is the velocity vector, μ is the viscosity, and ∇P is the pressure gradient. ρ_e is the net charge density and $\vec{E} = -\nabla \varphi$ is the applied electrical field. The third term on the right hand side of Eq. (7a) is the Coulomb force term due to the net charge, and the last term on the right hand side presents the dielectric force term due to the existence of dielectric permittivity gradient. Net charge can be generated in the bulk solution due to electric field and conductivity gradient, leading to electrohydrodynamic flow instabilities in microchannels [17–19]. Dielectric force can also contribute to the instabilities. However, one essential condition for the instabilities is that the electric field should be very high [18]. In this work, only low electric field situation is considered and we assume that instabilities will not occur. Consequently, the Coulomb force term and the dielectric force term in the bulk liquid phase are considered negligible.

Because the local net charge density is not zero only in EDL and the thickness of the EDL is considered sufficiently thin, the driving force term $\vec{E} \rho_e$ in Navier–Stokes equation can be neglected and the Helmholtz–Smoluchowski slip flow boundary is applied to account for the electroosmotic flow:

$$\vec{u} = -\frac{\varepsilon_0 \varepsilon_r(c) \zeta(c)}{\mu} \overrightarrow{E}$$
 at channel walls, (8)

where ε_0 is the permittivity in vacuum, $\varepsilon_r(c)$ is the relative dielectric constant, and $\zeta(c)$ is the zeta potential. Both $\varepsilon_r(c)$ and $\zeta(c)$ are functions of ionic concentration.

Considering a steady state electroosmotic flow without pressure difference along the channel, it is given as follows:

$$P = 0$$
 at the inlets and the exit. (9a)

Also, the pressure gradient is set to be zero at the channel walls:

$$\hat{n}\nabla P = 0$$
 at channel walls. (9b)

2.1.3. Concentration field

According to the ionic transport theory [20], the concentration distribution can be described by

$$\frac{\partial C_i}{\partial t} = -\vec{u} \cdot \nabla C_i + D_i \nabla^2 C_i + \frac{z_i F D_i}{RT} (\nabla \cdot C_i \nabla \varphi) + R_i,$$
(10a)

 C_i is the concentration of *i*th species, D_i is the diffusion coefficient of *i*th ion, z_i is the valence of *i*th ion, F is the Faraday constant, $F = eN_A$, where N_A is the Avogadro constant and e is the electronic charge;



Fig. 1. Schematic of the model flow-mixing system of two streams in a microchannel.

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