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Electroosmotic flow in micro/nanochannels with surface potential heterogeneity: An analysis through the Nernst–Planck model with convection effect

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ABSTRACT

A complete two-dimensional model is discussed to analyze the electroosmotic flow near a step jump in surface charge distribution of a micro- or nanochannel. The present model does not require the core neutrality of the fluid and it can handle electrolyte of arbitrary ionic valence as well. The governing equations consists of the flow field, mass transfer and electric potential equations. A pressure correction-based iterative algorithm (SIMPLE) is employed for computation. An analytic solution is obtained near the surface heterogeneity based on the Debye–Hückel approximation. The EOF is investigated both for weak and strong electrolytes at different channel heights. The form of the vortical flow near the potential patch and the dependence of the vortex strength on the patch overpotential is analyzed. A linear pressure drop is observed above the vortex. Average streamwise velocity overshoots the Helmholtz–Smoluchowski velocity. A comparison of the present model with the model based on the equilibrium Boltzmann distribution of ions is made. The results due to the two models differ significantly in the region where the convection effect is strong.

1. Introduction

Electroosmotic flow (EOF) is widely used to transport and mix fluids in micro- and nanofluidic systems based on the lab-on-a-chip concept. The electroosmotic flow in a microchannel is a laminar flow without any circulation. Mixing in microchannels is considered to be a non-trivial task. In microfluidic systems, it is difficult to incorporate moving components, and since the flow is laminar, there exists no turbulence to stir the fluid. Hence, mixing is a challenge.

In general, devices to enhance mixing in microchannels are classified into two categories: passive and active mixers [1]. Passive mixers typically use particular channel geometry configurations to increase the interfacial area between the liquids to be mixed [2]. The major difficulty in passive mixing is the fabrication complexity in microscale. The active mixers introduce moving parts inside microchannels or apply an AC-electric field. Though the above-mentioned techniques effectively reduce the mixing time, they may suffer from the difficulty in getting integrated into microfluidic systems since it requires external variable frequency pumps or internal mechanical moving parts [3]. In our previous work, we made a discussion on AC-electroosmotic flow in micro- and nanochannels [4].

The enhancement of mixing in microfluidics has been demonstrated by several authors through surface potential modulation. The generation of vortices in electroosmosis with non-uniform surface potential in microchannels was demonstrated by Ajdari [5]. The electroosmotic velocity strictly depends on the surface potential (ζ-potential). For a thin electric double layer, the electroosmotic velocity U_{EOF} is given by the Helmholtz–Smoluchowski equation as $U_0 = -\epsilon_e E_0 \zeta/\mu$. Thus the magnitude and the direction of the electroosmotic velocity can be controlled by the ζ-potential. The presence of the local circulation zone forces a portion of the mixed downstream fluid back to the unmixed upstream region.

The EOF in microchannel with non-uniform surface potential distribution has drawn the attention of several authors in recent years. Non-uniform ζ-potentials can be obtained by coating the channel walls with different materials or by using different buffer solutions [6]. Alternatively, when the solid's surface is photosensitive (such as a semiconducting TiO_2 film), the surface charge can be modified with light [7]. Erickson and Li [8] investigated the circulation region through surface charge heterogeneity in a T-shaped micromixer. They found that recirculation is strongest when the ζ-potential of the heterogeneous surface is of equal and opposite sign to that of the homogeneous surface. The potential and velocity distribution close to a step jump in potential has been investigated analytically by Yariv [9]. Fu et al. [10], based on the Nernst–Planck equation for ionic distribution, found that a step change in ζ-potential causes a significant variation in the velocity profile and in pressure distribution. However, their result could not predict any flow recirculation. Lee et al. [11] analyzed the EOF with

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non-uniform ζ-potential in a microchannel assuming that the electric body force is zero outside the electric double layer. Tian et al. [1] observed that the excellent mixing through surface non-uniform surface potential in microchannels may lead to a poor transport. In a recent article, Chang and Yang [12] provided a detailed account on the development of research related to electrokinetic mixing in microfluidic systems.

Qian and Bau [13] demonstrated that by the temporal modulation of the ζ-potential, it is possible to induce chaotic advection. They considered a spatially periodic mixing chamber, where the bottom and top surfaces each have two surface electrodes that are covered with a thin insulator. The ζ-potential on the insulated surface can be altered by applying electrostatic potential on these electrodes. It is possible to create various flow patterns in the mixing chamber by using different ζ-potentials under horizontal electric field. The time-dependent electroosmotic flow driven by an AC-electric field with patchwise surface heterogeneity distributed along the microchannel walls was considered by Luo [14,15]. He found that the circulation region grow and decay periodically in phases with the applied periodic AC-electric field intensity. Another method of effective mixing was proposed by Wu and Li [16,17] through an induced charged electroosmosis, in which triangle-shaped conducting objects are immersed in an aqueous solution.

It may be noted that all the earlier studies available in the literature reported the vortex formation near a patch when the patch potential is exactly opposite to the wall ζ-potential. A quantitative measure of the vortex strength, the form of the vortical flow and its dependence on the flow parameters has not been addressed in details. In the present paper we have varied the overpotential of the patch at a fixed value of the ζ-potential.

The practical relevance of electrokinetic transport in channels where the electric double layer formed on the wall occupies a substantial part of the channel interior is established in a recent articles by Yuan et al. [18] and Baldessari [19]. The continuum hypothesis is still valid for channel height greater than the size of the solvent molecules dissolved ions [20]. In most of the previous studies (i.e., [8,14] and the references therein) the channel height is considered to be large so that the EOF is modeled by the coupled Poisson–Boltzmann (PB) equation and the Navier–Stokes equations. The Boltzmann distribution for ions is proper when the bulk fluid is assumed to be electrically neutral. Thus the PB model, based on the Poisson–Boltzmann equation for electric potential, is valid for large values of ζ-potential, which corresponds to a thin electric double layer. However, it does not take care of the convective transport and the influence of external electric field on the ionic distribution. Comparison of the model where the charge distribution is governed by the Nernst–Planck equation (NP model) and the model based on Poisson–Boltzmann equation to describe the EOF in microchannel with a step change in surface ζ-potential has been made by Park et al. [21]. They observed a significant discrepancy between the results predicted by the two models near the region of inhomogeneous ζ-potential, when the electric double layer is thicker and/ or the applied electric field is stronger. Thus the difference in results between the two models in a heterogeneous microchannel is observed only when the EDLs are close. Recently, Wang et al. [22] examined the validity of the PB model in micro- and nanoscale EOF. The previous comparisons between NP and PB models were limited to microchannels where the EDLs are far apart for a strong electrolyte. One of the objectives of the present study is to compare the solutions between the NP and PB models at low value of wall potential and low channel heights with heterogeneous ζ-potential. The EOF conditions such as, external electric field, EDL thickness and overpotential of the patch, which influence the difference between the two models is also addressed in the present analysis.

The present work deals with the EOF in a long rectangular channel with a modulation of ζ-potential on the lower wall of the channel. The height of the channel is varied from 20 nm to 60 nm. The present analysis is based on the Nernst–Planck model. We have analyzed the two-dimensional flow field by computing the Navier–Stokes equations in its full form along with the mass transfer equations and electric potential equation. The separation of the flow field from the flat wall is observed through a step jump in ζ-potential. The flow separation is followed by a vortex formation above the patch. We have analyzed the form of the vortex and its dependence on the channel height and the patch overpotential. The flow field and the ionic concentration are computed for various values of the flow parameter. An asymptotic solution based on the Debye–Hückel approximation is derived for the present EOF near a surface potential heterogeneity.

2. Physical model and numerical method

Consider a long rectangular channel whose height h is nanoconstrained and is filled with an incompressible Newtonian electrolyte of uniform permittivity ϵ_e and viscosity μ, and is subjected to an uniform external electric field directed along the length of the channel, say x-axis. Fig. 1 shows the co-ordinate system for the present problem. A potential patch of length $l \sim O(h)$ which has a different surface potential than the channel walls is embedded in the lower wall of the channel. The channel width is considered to be of the order of its length. Thus a two-dimensional flow is considered.

The dimensional electric field $\vec{E}^* = (E_x, E_y, E_z)$ satisfying the Maxwell's equations, $\vec{E}^* = -\vec{\nabla}\Phi^*$, is governed by the following relation:

$$\nabla \cdot (\epsilon_e \vec{E}^*) = \rho_e = F\sum_i z_i c_i = Fc\sum_i z_i X_i. \tag{1}$$

where ρ_e is the charge density per unit volume, Φ^* the electric potential, c_i is the molarity of ionic species i and c is the total molar concentration, which is assumed to be constant. z_i is the valence and $X_i = c_i/c$ is the mole fraction of species i.

The electric potential Φ^* can be written as $\Phi^*(x, y, z) = -E_0 x + \phi^*(x, y, z)$. Here E_0 represents the externally applied electric field, which is considered to be constant, and ϕ^* is the induced electric potential. Using Eq. (1), the non-dimensional form of the equation for the potential can be written as

$$\frac{\partial^2\phi}{\partial y^2} + \epsilon_1^2\frac{\partial^2\phi}{\partial x^2} + \epsilon_2^2\frac{\partial^2\phi}{\partial z^2} = -\frac{\beta}{\epsilon^2}\sum_i z_i X_i. \tag{2}$$

The potential is scaled by $\phi_0 = RT/F$. For non-dimensionalization we have used (l, h, W) for coordinates (x, y, z) for example, $x = (x^*/l)$. We define $\epsilon_1 = h/l$, $\epsilon_2 = h/W$, $\epsilon = \lambda/h$ and $\beta = c/I$. Here λ, the EDL thickness, is $\sqrt{\epsilon_e RT}/FI^{1/2}$ and the ionic strength $I =$

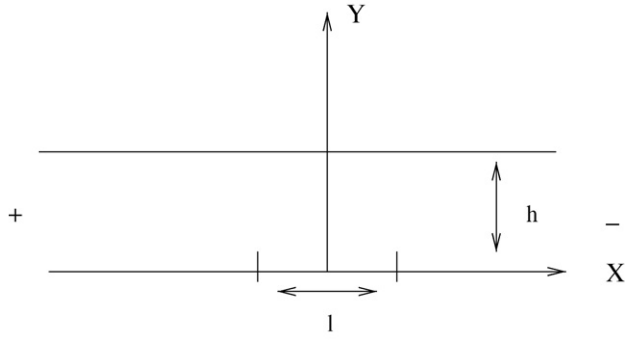

Fig. 1. Schematic diagram of the nanochannel and computational domain.