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Force criterion of different electrolytes in microchannel*

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The control and handling of fluids is central to many applications of the lab-on-chip. This paper analyzes the basic theory of manipulating different electrolytes and finds the two-dimensional model. Coulomb force and dielectric force belonging to the body force of different electrolytes in the microchannel were analyzed. The force criterion at the interface was concluded, and testified by the specific example. Three basic equations were analyzed and applied to simulate the phenomenon. The force criterion was proved to be correct based on the simulation results.

Keywords: force criterion, basic theory, specific example, numerical simulation
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1. Introduction

Dynamic control of fluids is central in the development of lab-on-a-chip (LOC) technology.[1−3] Although microelectromechanical devices are often used for this purpose, their moving parts make the fabrication and operation delicate. The use of electric fields can fulfill the requirement of particle and fluid manipulation without moving parts. The control of liquids using DC electric fields has been widely used in microsystems, particularly with capillary electroosmosis.[4] However, the required high voltages produce electrolysis and Joule heat. In this context, the use of AC electric fields presents several advantages, including lower power requirements, little or no electrolysis and simple integration. Both the induction pump[5] which exerts electrical forces in the liquid bulk, and the AC electroosmotic pump[6] which exerts forces in the diffuse part of the electrical double layer, are examples of micropumps that employ AC electric fields.

Recently, the use of AC electric fields to deflect and manipulate co-flowing streams of two different electrolytes within a microfluidic channel was demonstrated experimentally.[7] The experimental arrangement consisted of a microchannel with microelectrodes fabricated on the channel bottom. Two electrolytes with different conductivities were pumped along the channel side by side. When the voltage was applied to the electrodes, the boundary between the two fluids moved to the region of lower conductivity.

We had analyzed a much simpler two-dimensional (2D) physical system in order to understand the mechanisms that lead fluid flow and quantify the voltage required to have significant flow in a microdevice.[8] We expect that the 2D problem captures some essential features of the flow in the cross-section of the channel in Ref.[7]. The simulations proved that our analysis is correct.

However, we only analyzed the same solute with the same permittivity.[9] In fact the most significant conditions we need to face are the controls of different electrolytes, and the central problem is how to assure the flow direction. In this context the 2D physical system with different electrolytes was analyzed, the force at the interface of the two kinds of electrolytes with conductivity and permittivity gradient was calculated and the force criterion was concluded. At last the experimental phenomenon was simulated to testify the theoretical calculation.

2. Force criterion

The physical system modelled by us is shown in Fig.1. An array of electrodes occupies the width of the bottom of the channel. Two water saline solutions with different conductivities σ_1 and σ_2, different

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permittivities ε_1 and ε_2, fill the channel with the interface at the centre of the channel. An AC potential difference of amplitude $2V_0$ is applied between consecutive electrodes, i.e. each electrode is subjected to a voltage $V_i(t) = (-1)^i V_0 \cos(\omega t)$, where $i = 1, 2, 3 \ldots$ and ω is the angular frequency of the applied AC signal. We assume that there is a continuous supply of electrolyte at left and right walls so that the conductivity can be considered fixed at these boundaries. This is required in order to have conductivity gradients and fluid flow in steady conditions, otherwise, diffusion would homogenize the saline concentration in the domain and there would be no fluid flow in steady state. Our physical system could be realized experimentally with porous side walls in contact with reservoirs of saline solutions.

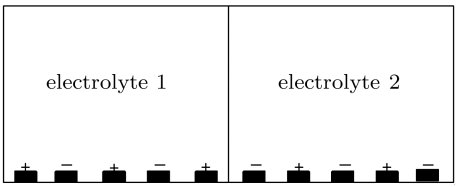


Fig.1. The analytical model.

The experiments in Ref.[7] were explained, considering that the electrical forces are actuated in the liquid bulk, because the frequency of the applied signal was high enough to neglect double-layer effects.[8] The electrical body force on a fluid is[10]

$$\boldsymbol{f}_{\mathrm{e}} = \rho_q \boldsymbol{E} - \frac{1}{2}\boldsymbol{E}^2 \nabla \varepsilon + \frac{1}{2}\nabla \left[\rho_m \left(\frac{\partial \varepsilon}{\partial \rho_m} \right)_T \boldsymbol{E}^2 \right], \quad (1)$$

where ρ_q is the charge density, ρ_m the mass density, ε the permittivity, T the temperature, and $\boldsymbol{E}$ is the the electric field.

In this context, two different electrolytes are analyzed. The first and second terms are the Coulomb and dielectric forces, respectively, and the last term is electrostriction force, which can be ignored for incompressible fluids.[11] For the system described here, the main forces acting on the free charge at the interface between the two liquids are Coulomb force and the dielectric force.

In order to predigest the model, a pair of electrodes is analyzed, which are shown in Fig.2. For a non-uniform electric field, a small region '*abdc*' is chosen, where '*oo*′' is the centre line and also the interface of the two fluids. Define the right as the positive direction. The small region is nearly uniform.

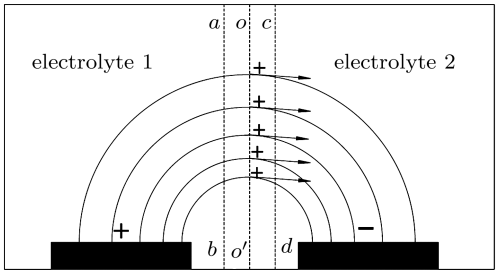


Fig.2. The model with two electrodes.

We assume that the left current density is $\boldsymbol{J}_1$ and right is $\boldsymbol{J}_2$, so

$$\boldsymbol{n} \cdot \boldsymbol{J}_1 = \boldsymbol{n} \cdot \boldsymbol{J}_2, \quad (2)$$

where $\boldsymbol{n}$ is the normal vector to the interface. The left electric field intensity is $\boldsymbol{E}_1$ and right is $\boldsymbol{E}_2$, the equations based on the Ohm's law can be expressed as

$$\boldsymbol{J}_1 = \sigma_1 \boldsymbol{E}_1 \quad (3)$$

and

$$\boldsymbol{J}_2 = \sigma_2 \boldsymbol{E}_2 \quad (4)$$

respectively.

Substituting Eqs.(3) and (4) into Eq.(2), we can obtain

$$\sigma_1 \boldsymbol{E}_1 \cdot \boldsymbol{n} = \sigma_2 \boldsymbol{E}_2 \cdot \boldsymbol{n}. \quad (5)$$

The net charge per unit area at the interface based on the Gauss's law can be described as[11]

$$Q_{\mathrm{surface}} = (\varepsilon_2 \boldsymbol{E}_2 - \varepsilon_1 \boldsymbol{E}_1) \cdot \boldsymbol{n}. \quad (6)$$

So the Coulomb force is

$$Q_{\mathrm{surface}} = (\varepsilon_2 \boldsymbol{E}_2 - \varepsilon_1 \boldsymbol{E}_1) \cdot \boldsymbol{n}. \quad (7)$$

In this context, the solutes are not the same, so the permittivities are different and the dielectric force must be considered. The permittivity gradient at the interface can be recognized as $\varepsilon_2/\varepsilon_1$ instantaneously. The dielectric force is expressed as

$$\boldsymbol{f}_{\mathrm{D}} = \frac{1}{2}\frac{\varepsilon_2}{\varepsilon_1}(\boldsymbol{E}_2^2 - \boldsymbol{E}_1^2) \cdot \boldsymbol{n}. \quad (8)$$

Substituting Eqs.(7) and (8) into Eq.(1), we can obtain

$$\boldsymbol{f}_{\mathrm{e}} = \boldsymbol{f}_{\mathrm{C}} - \boldsymbol{f}_{\mathrm{D}} = (\sigma_r - 1)[\sigma_r \varepsilon_a - \varepsilon_b] \cdot E_1^2 \cdot \boldsymbol{n}, \quad (9)$$

where σ_r is σ_1/σ_2, ε_a is $(\varepsilon_2 - \varepsilon_2/2\varepsilon_1)$ and the ε_b is $(\varepsilon_1 + \varepsilon_2/2\varepsilon_1)$, respectively.

The force at the interface can be calculated easily by Eq.(9), which is the force criterion. So if $\boldsymbol{f}_{\mathrm{e}} > 0$,

the interface will be driven towards right, contrarily, left. Equation (9) proves that when σ_1 is equal to σ_2, $\boldsymbol{f}_\mathrm{e}$ is zero and the interface will be static no matter what the electrolytes are. Using more electrodes, we shall get the same prediction. Because the line 'CD' can be chosen freely, so in all the microchannel the phenomenon is constant.

3. Calculative example and simulation

3.1. Calculative example

In order to test and verify the force criterion with example, the sodium chloride (Nacl) and potassium chloride (Kcl) are chosen as the electrolyte 1 and electrolyte 2, respectively. The specific parameters of the two electrolytes are as follows: σ_1 is 0.01 mS/m, σ_2 is 0.02 mS/m; ε_1 is 80 F/m and ε_2 is 86 F/m (and the corresponding relative permittivities); the temperature is 20 °C.

Substituting the specific parameters into the force criterion Eq.(9), we can conclude that $\boldsymbol{f}_\mathrm{e} < 0$ with the negative direction, so we can estimate that the interface will be driven towards left.

However, the theoretic calculation still needs to be validated by the simulation. The force criterion only provides us a qualitative method to estimate the deflexion of the different electrolytes interface.

3.2. Electrical equations

The electrical current in the bulk of a dilute electrolyte is given by

$$\boldsymbol{J} = \sum_i q_i(n_i\mu_i\boldsymbol{E} - D_i\nabla n_i + n_i\boldsymbol{u}), \tag{10}$$

where q_i, n_i, μ_i, D_i are, respectively, the charge, ion number density, mobility and diffusivity of ionic species i, and $\boldsymbol{u}$ is the electrolyte velocity. For electrolytes, the convective current $q_i n_i \boldsymbol{u}$ is usually much smaller than the conduction current $q_i n_i \boldsymbol{u}\boldsymbol{E}$.[8] Also, in many situations in microsystems, the ratio between diffusion and electro-migration $|D_i\nabla n_i| \,/\, |n_i\mu_i\boldsymbol{E}|$ is much smaller than unity.[8] In effect, it is of the order of $D_i/\mu_i\boldsymbol{E}d$, with d a typical distance of the system. From Einstein's relation for monovalent ions, $D_i/\mu_i = k_\mathrm{B}T/e = 0.025$ V at room temperature (where k_B is the Boltzmann's constant, T is the absolute temperature and e is the charge of a proton) is much smaller than the typical voltage drop in the liquid bulk $\boldsymbol{E}d$ (of the order of 10 V). With these approximations and applied signals with periods much shorter than the typical time of variation of ionic concentration, the charge conservation equation leads to[8]

$$\nabla\cdot((\sigma + \mathrm{i}\varepsilon\omega)\nabla\phi) = 0, \tag{11}$$

where $\sigma = \sum_i q_i n_i \mu_i$ is the electrolyte conductivity and ϕ is the phase of the electrical potential. The boundary conditions for this equation are: fixed potential at the electrodes and normal current equal to zero at the remaining boundaries. On the micrometer length scale, electrolytes are quasi-electroneutral, i.e. $\sum_i z_i n_i / \sum_i n_i \ll 1$ where z_i is the ion valence.[8] For a binary electrolyte, under the electro-neutrality assumption, the conductivity satisfies a convection-diffusion equation[12,13]

$$D\nabla^2\sigma = \frac{\partial\sigma}{\partial t} + (\boldsymbol{u}\cdot\nabla)\sigma, \tag{12}$$

where $D = 2D_+D_-/(D_+ + D_-)$ is the ambipolar diffusion coefficient, and $D_\pm$ the diffusion coefficients of positive and negative species.

3.3. Mechanical equations

In microsystems, the Reynolds number ($\mathrm{Re} = \rho_m u d/\eta$) is usually much smaller than one. In our system ρ_m and η are, respectively, mass density and viscosity of water, u is the typical velocity (∼1 mm/s), d is the typical length (∼10 μm), and we obtain $\mathrm{Re}\sim 10^{-3}$. Therefore, the liquid motion is governed by Stokes equations

$$\rho\frac{\partial\boldsymbol{u}}{\partial t} = \eta\nabla^2\boldsymbol{u} - \nabla p + \boldsymbol{f}_e, \tag{13}$$

$$\nabla\cdot\boldsymbol{u} = 0, \tag{14}$$

where p is the pressure and $\boldsymbol{f}_e$ is the electrical body force given by both the time-average Coulomb force and dielectric force

$$\begin{aligned}\boldsymbol{f}_\mathrm{e} &= \rho\boldsymbol{E} - \frac{1}{2}\boldsymbol{E}^2\nabla\varepsilon \\ &= -\varepsilon\frac{\nabla\sigma\cdot\nabla\phi}{\sigma}\nabla\phi - \frac{1}{2}(\nabla\phi)^2\cdot\nabla\varepsilon.\end{aligned} \tag{15}$$

The equations for potential, conductivity and velocity are fully coupled. This system of equations is solved here by using the commercial software COMSOL Multiphysics.

For the simulations, the rectangular domain was 250 μm wide and 120 μm high. There were ten equal electrodes at the bottom boundary, as in Fig.1. They were 15 μm wide, with negligible height, separated by 10 μm gaps. Each electrode is subjected to an AC potential of amplitude $\pm V_0$ with a voltage drop between consecutive electrodes equal to $2V_0$. We specified $\sigma_1 = 0.01$ S/m and $\sigma_2 = 0.02$ S/m, i.e. the right region is twice as conductive. For simplicity, in the simulations we have used angular frequency $\omega \ll \sigma/\varepsilon$ ($f \ll 10^6$ Hz) The voltage amplitude applied to each electrode was ± 1 V.

Setting the specific parameters of the example and coupling the three equations, we can obtain the time-dependent ($t = 0.01$ s and $t = 3$ s) simulation results as shown in Fig.3.

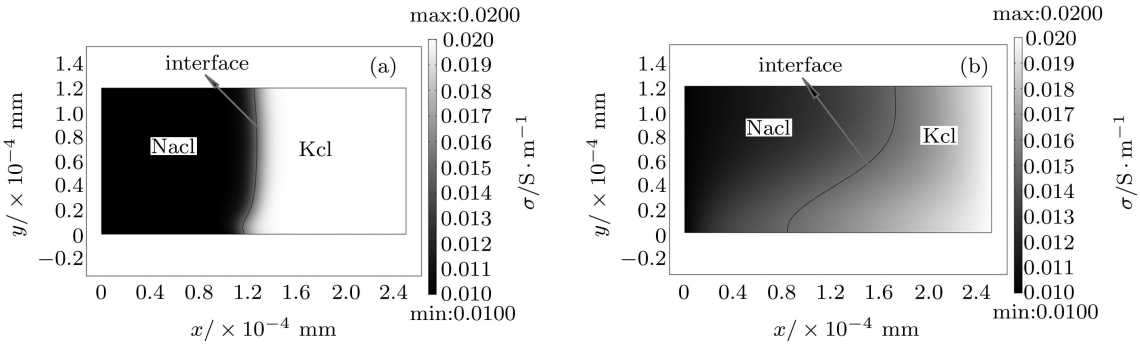


Fig.3. The simulation results at (a) $t = 0.01$ s and (b) $t = 3$ s.

In the figures, the interface is driven towards left, which is the same as the force criterion calculated above. The simuation proves that the force criterion is correct and believable.

4. Conclusion

Fluid manipulation is an important requirement in LOC technology. In this paper we have analysed numerically the electrical manipulation of different electrolytes with a steep gradient in saline concentration and permittivity by an array of microelectrodes subjected to AC voltages. In this situation, fluid flow is generated by the Coulomb force acted upon the charge and the dielectric force induced in the region with conductivity and permittivity gradients, respectively. The force criterion at the interface of the two kinds of electrolytes was calculated. The specific example with Nacl and Kcl was chosen to testify the force criterion. The three basic equations were analyzed specifically. At last, the force criterion was proved to be correct by the simulation in the software COMSOL. Using the force criterion, we can analyze the force of different electrolytes in microchannel conveniently and easily.

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