Enhancement of micro-flow mixing using DC nonlinear electrokinetic vortices
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ABSTRACT

A nonlinear electrokinetics near a sharp corner was found to present in a dc electric field. Concentration polarization is the main mechanism responsible for the formation of a corner-vortex. Sharp corners (the angle of the corner is 30 degree) are designed in a microchannel to create vortices at the upstream near the corners. The rotational direction of the vortex near the corner is observed in experiments. This corner-vortex is used to enhance micromixing in this study. Furthermore, the fluorescent intensity is employed to observe the mixing index of fluid in the mixer. The degree of the mixing reaches to 78% when the fluid flows through an array of corners within 800 µm length.

Keywords: Concentration gradient / EOF / Nonlinear electrokinetics / Microfluidics

1 Introduction

Microfluidic devices, lab-on-a-chip systems, and the micro-TAS (Micro Total Analysis Systems) concept have attracted considerable attention over the past decade for bioanalytical and chemical processing applications. In recent years, electroosmotic flow (EOF) is a preferred mode of manipulating fluids in microdevices. Many applications of the EOF have been found in the literatures [1-4]. Generally, the characteristic velocity of EOF is considered by the Smoluchowski relation \( v_{slip} = \varepsilon \varepsilon_0 \nabla \phi / \mu \) (\( \varepsilon \) is the dielectric permittivity of the solvent, \( \varepsilon_0 \) is the vacuum permittivity, \( \mu \) is the viscosity of the fluid, and \( \nabla \phi \) is the external potential drop) [5]. This characteristic velocity is linear with respect to the external potential drop and the related phenomena are referred to classical linear EOF. According to the Smoluchowski relation, the velocity is independent on channel shape and dependent on the surface charges of the channel walls. Usually, the critical Reynolds number of classical linear EOF is under 10 and Cummings et al. [6] demonstrated that the electric field lines coincide with the streamlines in the linear EOF. Therefore, the linear EOF behaves like an inviscid potential flow due to the absence of wall shear. Interestingly, the potential flow is irrotational and it is a major challenge to generate vortices in linear EOF microfluidic systems.

Recently, Thamida et al.[7], Takhistov et al.[8] and Yossifon et al.[9] observed that, beyond a critical level of external-field intensity of dc electrical field, vortices appear within the flow around sharp corners of microchannel junctions. A large field penetration exists near sharp corners of the channels. The external field can penetrate the double layers and produce the nonequilibrium electrokinetics near the sharp corners. The nonequilibrium electrokinetics is termed dc nonlinear electrokinetics and this phenomenon possesses considerable applications of microfluidic systems. Thus, the generation of such vortices is potentially useful in certain applications as a means to enhance and control microfluidic mixing.

The dc nonlinear electrokinetics near sharp corners has not been utilized in applications of micro-systems. In this paper, we use the dc nonlinear electrokinetics to achieve a micro-mixer design for microfluidic systems. Using the symmetric corners, the vortices are induced near the sharp corners and compress the fluid in the channel. Two different kinds of samples are mixed when they flow through the symmetric corners. The mixing index is enhanced in a short distance across the symmetric corners.

2 Materials and methods

The geometry is represented in Fig. 1. The diameter of each reservoir is 4 mm. The length of main channel is 1 cm, the width of the channel is 200 µm and the depth of the channel is 95 µm. The angle of each corner is 30 degree. The material of the channel is polydimethylsiloxane (PDMS). Silicone elastomer and elastomer curing agent (Sil-More Industrial Ltd, USA Sylgard 184A and Sylgard 184B) were mixed in a ratio of 10:1 and then poured onto an SU-8 microstructure mold. The SU-8 microstructure mold is heated at 100°C for 1 hour to solidify the PDMS layer. The PDMS inverse structure was then mechanically peeled off the template and bonded with glass. The chip manufactured by PDMS process is performed by an oxygen plasma treatment, and it needs filling deionizer water in microchannel to maintain the hydrophilic property.
The width of the channel is $200 \mu m$; The length of the channel is $1 \text{cm}$; The diameter of each reservoir is $4 \text{mm}$; The symmetric corners are located at the middle of the channel and the interval of the corners is approximately $100 \mu m$; The angle of each corner is 30 degree.

Latex particles (diameter is $3 \mu m$) are used to observe the induced fluid field near the symmetric corners in the channel. The mixing experiments were performed using Rhodamine B ($10^{-3}$M) as the sample. The fluid field within the microchip was observed by mercury lamp induced fluorescence using a charge-coupled device camera (CCD, model: SSC-DC50A, Sony, Japan). The experimental images were captured by an optical microscope (model: Eclipse 50i, Nikon, Japan), filtered spectrally, and then measured by the CCD camera.

Figure 1. The geometry of symmetric corners. The depth of the channel is $95 \mu m$; The width of the channel is $200 \mu m$; The length of the channel is $1 \text{cm}$; The diameter of each reservoir is $4 \text{mm}$; The symmetric corners are located at the middle of the channel and the interval of the corners is approximately $100 \mu m$; The angle of each corner is 30 degree. 

The induced zeta potential is given as

$$\zeta_i = \left( e / \delta A \right) = \mp \lambda \left( \cot \alpha \right) \phi_{\zeta}$$

(1)

$\zeta_i$ is the dimensionless induced zeta potential; $\zeta_i$ is the induced zeta potential near the corner; $e = e_f / e_w$ is the ratio of permittivity of fluid and wall; $L$ is the width of the channel; $A$ is the constant of the solution of Laplace equation (it is represented in Eq. (10a) in the reference 7); $\delta$ is the Debye length; $\lambda$ is the eigenvalue of Eq. (10a) in the reference 7; $r$ is the dimensionless radius of the corner; $\alpha$ is the half angle of the corner. The dimensionless zeta potential is $\zeta_i \sim 1.545 r^{-1.3}$ and the similar value is obtained by from Yossifon et al.[9]. It implies that $\zeta_i$ is very large when the radius of the corner, $r$, is infinite small. However, the radius of the corner is approximately $10 \mu m$ in our device. Hence, the induced zeta potential is very small with its magnitude of the order (i.e. $\zeta_i \sim 1mV$) and this value can be neglected in our case.

According to the reference 8, we use the concept of the conservation of current to explain the phenomena near the sharp corners in our experimental results. The symmetric electrolyte is considered i.e. $c_i = c_e = c$, $z_i = z_e = z$ ($z = 1$).

The current density, $i_{\text{Ohm}}$, is described by Ohm’s law at the location far from the corner.

$$i_{\text{Ohm}} = \sigma \left( - \nabla \phi_{\text{Ohm}} \right)$$

(2)

$\phi_{\text{Ohm}}$ is the potential governed by the Ohm’s law. $\sigma$ is the conductivity of the electrolyte and can be translated i.e. $\sigma = (F \sum z_i^2 \nu_i)$. $F$ is the Faraday constant; $z_i$ is the valence of $i$ -th kind of ion; $\nu_i$ is the mobility of $i$ -th kind of ion; $c_i$ is the concentration of $i$ -th kind of ion. $\nu_i$ can be represented as $\nu_i = D_i / RT$ i.e. Nernst-Einstein equation [5]. $D_i$ is the diffusivity of $i$-th kind of ion; $R$ is the gas constant and $T$ is the temperature. Finally, we can get $i_{\text{Ohm}}$

$$i_{\text{Ohm}} = \left( F \sum z_i^2 \frac{D_i}{RT} c_i \right) \left( - \nabla \phi_{\text{Ohm}} \right)$$

(3)

If we use the Ohm’s law to describe the current density near the corner, the assumption of the similitude between the electric field lines and streamlines are satisfied as described by Cummings et al. [6]. There will be no vortex formed near the corner. But, our experimental results showed the existence of the corner vortices which violate the similitude near the corner. Some mechanisms must be considered here.

According to the Nernst-Plank equation, the current density is described by

$$i_{\text{sc}} = \sigma \left( - \nabla \phi_{\text{sc}} \right) + \left( - F \sum z_i D_i \nabla c_i \right) + \left( F \nu_i \sum z_i c_i \right)$$

(4)

$i_{\text{sc}}$ is the current density near the sharp corner. $u$ is the velocity of the fluid in the channel. For current conservation, $i_{\text{Ohm}} = i_{\text{sc}}$, we obtained

$$\left( - F \sum z_i \frac{D_i + D_e}{D_i - D_e} \nabla \phi_{\text{sc}} \right) \left( - \nabla \phi_{\text{Ohm}} \right) = \frac{\nabla c}{c}$$

(5)

The current density near the corner is composed of electromigration and diffusion in Eq. 4. According to the current conservation i.e. Eq. 4, we infer that the concentration gradient is generated near the corner. The relation between potential drop and concentration gradient near the corner is given in Eq. 5. Takhistov et al. [8]
providing that the potential difference, $\phi_c - \phi_{\text{lim}}$, is approximately $16 \, mV$ when the vortex is formed in their case. The angle of the corner we designed is sharper than Takhistov et al. Therefore, we have potential difference larger than $16 \, mV$ in our design.

4 Results and discussion

In Fig. 2, we apply an electric field, $500 \, V/cm$, to the channel filled with deionized water. When the dc electric field is activated, the vortices are generated immediately. Some latex particles are aggregated near the upstream of the tip of the corners. Once the induced vortex is formed, it catches particles in itself. We observe that the vortex forms a close shape in a constant dc electric field. The closed vortices produce a nozzle structure in the middle of the symmetric corners. The dash-line is used to depict the nozzle structure near the symmetric corners. The fluid is accelerated in the nozzle structure. In the video of our experiments we found that the vortices have three dimensional structures. The ratio of the depth and width in the channel is approximately 0.5 (i.e. depth/width = 95/200 = 0.475). The EOF of the upper and lower walls is considered. The fluid on upper and lower walls is continuously moved downward but the middle fluid is retarded by the vortices due to the concentration polarization near the corners.

In Fig. 3 the transient state of the particle trajectories is shown. In Fig. 3a the dc electric field (i.e. $500 \, V/cm$) is applied and immediately the vortex is formed near the corner. The latex particles are trapped inside the vortex and the red arrow shows the rotational direction of the vortex clearly. We estimated that the zeta potential on the PDMS is $30-40 \, mV$ (the electrolyte is deionized water). We estimated that the vortex velocity of the particle near the corner is approximately $6.59 \, mm/s$. In the same dc electric field, the velocity of the classical EOF is approximately $1.06 \, mm/s$. According to the rotational direction of the vortex, the mechanism similar to the reference 8 can be proven in this experiment. The zeta potential is changed near the corner due to the concentration gradient of the electrolyte. In Fig. 3b the red circles are used to mark the locations of the vortices. Each vortex is located at the upstream near the corner and the two vortices are symmetric.

We use the symmetric corners to function as a mixer. The array of symmetric corners is designed and the geometry is shown in Fig. 4a. The latex particles are used to trace the flow field in the mixer. The latex particles are filled in the channel first and then we apply a dc electric field (i.e. $E = 210 V/cm$). When the electric field is applied, the vortices are formed near the corners immediately. When the
dc electric field is increased \( i.e. E \sim 380 \text{ V/cm} \), the vortices near corners become larger and the particle trajectories are more complicated. According to the result of the particle trajectories, we infer that the array of symmetric corners is valid to enhance mixing. To investigate the mixing performance of the array of symmetric corners, experiments were performed using fluorescent dye, Rhodamine B \( (10^{-4}\text{M}) \), as the sample and deionized water as the buffer liquid. In Fig. 4c, the mixing index is expressed by the intensity of Rhodamine B. The formula of the mixing index is referred to the reference 10. The mixing index is approximately 3% before the fluids flow into the array and the mixing index is approximately 78% when the fluids flow passed the array and the length of the array is approximately \( 800 \ \mu\text{m} \). By the three dimensional vortices, the fluids are compressed and the mixing length between two samples is reduced so that the mixing index is enhanced.

![Figure 4a](image1.png)  
**Figure 4a** The geometry of the array of symmetric corners. The part of the array is approximately \( 800 \ \mu\text{m} \); The depth of the channel is \( 95 \ \mu\text{m} \); The width of the channel is \( 400 \ \mu\text{m} \). The interval of symmetric corners is \( 200 \ \mu\text{m} \); The interval between corners in a symmetric corner is \( 100 \ \mu\text{m} \); The angle of each corner is 30 degree. The intensity of the dc electric field is approximately \( 210 \ V/\text{cm} \). b) The intensity of the dc electric field is approximately \( 210 \ V/\text{cm} \). The fluorescent dye, Rhodamine B \( (10^{-4}\text{M}) \), is used to observe the mixing index in the device. The mixing indices are 3% and 78% before fluid flow into the array and after fluid flow out of the array, respectively.

5 Conclusion

A nonlinear electrokinetic phenomenon is observed near a sharp corner and vortices are observed clearly in our experiments. The mechanism to form small symmetric vortices due to field effect in the reference 7 is not the major cause to form the vortices near the corners in our study. Furthermore, we considered that the main mechanism to form the vortices near the corners is due to concentration polarization. The figure 3 proves the concentration polarization is the main mechanism. Due to the result of the corner-vortex, the three dimensional structure is found near the symmetric corners. The ratio of the depth and the width is approximately 0.5 and the three dimensional effect must be considered. We inferred that the EOF on the upper and lower walls affects the vortex near the symmetric corners. Further, we use the three dimensional corner-vortex to create a novel mixer in our experiment. The mixing index is enhanced to 78% when the fluid flows through the array of symmetric corners in \( 800 \ \mu\text{m} \). The length between corners must be considered to obtain the optimum mixing index. The corner-vortex is dependent on the concentration of electrolyte and its relation will be studied in our future work.

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