Numerical analysis of mixing by electrothermal induced flow in microfluidic systems

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An electrothermal flow induced chaotic mixing in microfluidic systems is studied analytically and numerically. The flow is induced due to the Coulombic and dielectric forces arising from the variation of the dielectric properties with respect to the temperature in the presence of an electric field. The numerical model is validated using an analytical solution derived for basic flow patterns in a simplified geometry. The computational model has been used to illustrate the mixing in microcavity and *T*-sensor constructs. The simulations predict the chaotic nature of the mixing process, where the material interface evolution shows exponential growth. © 2007 American Institute of Physics. [DOI: 10.1063/1.2734910]

I. INTRODUCTION

Microfluidic devices used in biological sample preparation and assays often require fast and thorough mixing of different analytes in a buffer medium. Although the typical characteristic length scale associated with microfluidic devices is small, mixing based on diffusion alone leads to inappropriate operational times or channel lengths to facilitate complete mixing. To overcome these difficulties, passive and active means of introducing forcing functions have been reported in the literature. For example, rapid changes in flow directions through use of tortuous flow paths¹ or grooved surfaces,² electro-osmosis in surface modified systems,³ use of conductivity gradients,⁴ magnetic fields,⁵ bubbles,⁶ gaseous slugs,⁷ acoustic stirring,⁸ and even motile bacteria⁹ have been studied. A comprehensive review of mixing in microfluidic devices can be found in Ref. 10.

When an AC electric field is applied to an electrolyte solution, several modes of AC electrokinetic phenomena can occur, all of which primarily depend on the applied voltage and frequency. When the applied voltage is low and the frequency is sufficiently high, electrothermal flow (ETF) dominates the electrolysis and AC electro-osmosis.¹¹ When an AC electric field is applied to an electrolyte solution, the solution is subjected to Coulombic and dielectric forces due to variations in the dielectric properties (electrical permittivity and conductivity) as a function of temperature.¹² Though the Joule effect is often the primary mechanism for heating the solution, other external sources, such as laser illumination, can induce thermal gradients, and hence, electrothermal flows. This flow depends on electrical conductivity and permittivity of the buffer, the voltage and frequency of the applied electric field, and electrode configuration and dimensions of the microfluidic device. The dielectric force due to the variation of dielectric fluid properties, induced by nonthermal means, has also been reported in previous literature¹⁹ as a means to facilitate mixing and is not in the scope of the present study.

In a recent publication, Gonzalez *et al.*¹⁸ investigated self-similar solutions of electrothermal induced flow. In addition to the temperature variation generated by internal sources, these authors also applied an external temperature gradient. In this article, we will first derive the analytical solutions for the flow velocity and the temperature field induced due to the electrothermal flow in

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FIG. 1. Coplanar electrode is used in the validation study. Solid lines illustrate the electric field. Dashed lines illustrate streamlines.

a simplified geometry. The analytical solution is obtained via dual integral/series and Green's function methods¹³ applied to solve the Stokes flow and heat conduction equations. Analytical solutions have been used to validate the computational model, where the full set of governing equations is solved using the finite volume approach. The model is used to demonstrate application of the electrothermal flow to facilitate mixing of various analytes in microcavity and T -sensor constructs. Moreover, it is shown that this flow induces chaotic mixing based on short-term evolution of a material segment.

II. ELECTROTHERMAL INDUCED FLOW

A. Mathematical formulation

Let us consider a microfluidic channel consisting of a pair of coplanar electrodes or electrode arrays kept at the bottom floor of the channel as shown in Fig. 1. The electrodes have a width of d_2 and a gap of $2d_1$ between adjacent electrodes. They are energized by an electric potential of $-V_0$ and V_0 , respectively. The force density due to application of the electric field is given by¹⁴

$$\mathbf{f} = \rho_e \mathbf{E} - \frac{1}{2} \mathbf{E}^2 \nabla \varepsilon + \frac{1}{2} \nabla \left[\rho_m \left(\frac{\partial \varepsilon}{\partial \rho_m} \right)_T \mathbf{E}^2 \right], \quad (1)$$

where ρ_e is the charge density, $\mathbf{E}$ is the electric field, ε is the permittivity of the buffer, and ρ_m is the buffer density. The first, second, and third terms on the right side of the above equation represent the Coulomb force, the dielectric force, and the electrostriction forces. The last term in Eq. (1) is the electrostriction force term; however, this term usually can be omitted for the incompressible fluids. If the applied electric field is an alternating one, then the averaged force density can be expressed as¹²

$$\langle \mathbf{f} \rangle = \frac{1}{2} \operatorname{Re} \left[\frac{\sigma \varepsilon (\alpha - \beta)}{\sigma + i \omega \varepsilon} (\nabla T \cdot \mathbf{E}_0) \mathbf{E}_0^* - \frac{1}{2} \varepsilon \alpha |\mathbf{E}_0|^2 \nabla T \right]. \quad (2)$$

Linear variations of the electric properties are assumed in deriving the above equation. In Eq. (2), σ is the electrical conductivity, T is the temperature, and α and β are the coefficients of variations of the electrical properties with respect to the temperature. The coefficients α and β are expressed as:

$$\alpha = \frac{1}{\varepsilon} \frac{\partial \varepsilon}{\partial T}, \quad \beta = \frac{1}{\sigma} \frac{\partial \sigma}{\partial T}. \quad (3)$$

Considering the Joule heating ($=\sigma \mathbf{E}^2$) alone as a mechanism to induce thermal gradients, the conservation equation for thermal field is expressed as

$$\rho_m c_p \frac{\partial T}{\partial t} + \rho_m c_p (\mathbf{u} \cdot \nabla) T = k \nabla^2 T + \sigma \mathbf{E}^2, \quad (4)$$

where c_p is the specific heat capacity, $\mathbf{u}$ is the velocity vector, and k is the thermal conductivity. The fluid flow is governed by Navier-Stokes and continuity equations

$$\rho_m \left[\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} \right] = -\nabla p + \mu \nabla^2 \mathbf{u} + \langle \mathbf{f} \rangle, \quad (5)$$

$$\nabla \cdot \mathbf{u} = 0,$$

where p is pressure and $\langle \mathbf{f} \rangle$ is the momentum source induced due to the electrothermal flow. The electric field satisfies the following relations:

$$\mathbf{E} = -\nabla V, \quad \frac{\partial \rho_e}{\partial t} + \nabla \cdot \mathbf{j} = 0, \quad (6)$$

$$\nabla \cdot \mathbf{D} = 0, \quad \mathbf{D} = \epsilon \mathbf{E}, \quad \mathbf{j} = \sigma \mathbf{E},$$

where V is the electric potential, $\mathbf{j}$ is the current density, $\mathbf{D}$ is the displacement vector, and σ is the electrical conductivity of the buffer. The solutions to the coupled Eqs. (4)–(6), subjected to appropriate boundary conditions, describe the thermal, electric, and flow fields. The mixing of the analytes is studied either by solving an advection-diffusion equation (when the analyte size is below a micron) or by solving a kinematic equation (when the analyte size is above a micron).¹⁷

B. Exact solution

We follow the procedure used by Ramos *et al.*,¹² and rewrite Eqs. (4)–(6) for a low Reynolds number ($\ll 1$) steady-state flow in simplified geometries, coplanar electrode structure shown in Fig. 1. The normalized electric potential ϕ (normalized by V_0) distribution is given by the Fourier integral

$$\phi(x, y) = \int_0^\infty A(\lambda) \lambda^{-1} e^{-\lambda y} \sin(\lambda x) d\lambda, \quad (7)$$

where

$$A(\lambda) = \frac{2}{K(ab)} \sum_{n=1}^{\infty} (-1)^{n-1} P_{n-1}(\cos v) J_{2n-1}(\lambda b), \quad (8)$$

and

$$v = 2 \cos^{-1} a/b, a = d_1, b = d_1 + d_2. \quad (9)$$

Here, P_n and J_n are the Legendre and Bessel functions of order n , K is the complete elliptic function of the second kind. The general solution to temperature is expressed as

$$T = -\frac{\sigma \phi^2}{2k} + Ax + By + C, \quad (10)$$

where A , B , and C are constants determined from thermal boundary conditions. By assuming an isothermal condition on the electrode surface, and solving heat conduction equation, we obtain

$$T = T_0 - \frac{\sigma V_0^2 \phi^2}{2k}. \quad (11)$$

The solution for the electrothermal force is calculated from Eq. (2) by the Green's function method

$$u_i(x, y) = \int \int_{S_c} G_{ij}(x_1, x_2, x_{0,1}, y_{0,2}) f_j(x_{0,1}, y_{0,2}) dS_0, \quad (12)$$

where G_{ij} is Green's function or Stokeslet in the presence of a planar boundary as given in Ref. 13. We can extend the above methodology to a periodic array of electrodes and calculate the electric potential in terms of a Fourier series

$$\phi(x, y) = \sum_{n=1}^{\infty} \frac{A_n}{\lambda_n} \cos \lambda_n e^{-\lambda_n y}, \lambda_n = n - 1/2, \quad (13)$$

where

$$A_n = \frac{P_{n-1}(\cos c)}{K(\cos c/2)}. \quad (14)$$

Similarly, the flow field is calculated by Eq. (12), in which the two-dimensional singly periodic Stokeslet in the presence of a planar boundary is used.¹³ The above analytical solutions are used to validate the numerical method.

C. Numerical method

Microfluidic devices often have complex electrode patterns, which require solving the full set of governing equations using numerical methods. We use a finite-volume method-based solver available in the commercial software CFD-ACE+ (Ref. 15) to solve Eqs. (4)–(6) in a coupled manner. Analyte mixing is studied using an advection-diffusion equation or kinematic equations. The basic flow, electric, and thermal solvers are available in CFD-ACE+. We have added the source term to the momentum equations corresponding to the electrothermal flow. During simulations, we apply constant voltage on electrodes. Due to the high thermal conductivity, the temperature on the electrode is assumed constant and the same as the ambient temperature. The channel walls are assumed electrically and thermally insulated, which is a good approximation based on typical materials used in microfabrication. The no-slip condition is applied to all channel surfaces, where fluid makes contact.

III. RESULTS AND DISCUSSION

A. Model validation

The analytical results for the coplanar electrode array as shown in Fig. 1 is used to validate the full-scale numerical simulation. The electrode width and the gap used in the study are 50 μm . A DC signal of ± 2 V is applied to the adjacent electrodes. The ambient temperature is assumed to be 300 K. In all simulations, the KCl solution is used as the buffer. It has a density of 1000 kg/m^3 , viscosity of 10^{-6} m^2/s , thermal conductivity of 0.6 $\text{W}/\text{m}\cdot\text{K}$, heat capacity of 4100 $\text{J}/\text{kg}\cdot\text{K}$, electrical conductivity of 0.056/ Ωm , relative permittivity of 78, α value of $-0.004/\text{K}$ and β value of 0.02/K. Comparison between numerical and analytical solution in terms of velocity and temperature along a plane at an elevation of 5 μm and 25 μm from electrode surface is shown in Fig. 2. In numerical simulation, the grid used is 400×200 , and further increase of grid numbers does not improve the predictions. The numerical and analytical results agree with the error less than 0.1%. The maximum velocity in this region is ~ 4 $\mu\text{m}/\text{s}$. The maximum temperature difference is very small (0.2 K).

FIG. 2. Comparison of the numerical results (lines) with analytical data (circles). (a) Temperature field. (b) Velocity field.

B. Mixing of two analytes in a microcavity

The ETF induced mixing of two analytes is studied in a closed microcavity construct. The cavity has a dimension of $100 \times 200 \mu\text{m}$ cross section as shown in Fig. 3(a). Four electrode pairs are kept; one on the top, bottom, and each sidewall. The electrodes at the sidewall are $25 \mu\text{m}$ wide, while those at bottom walls are $50 \mu\text{m}$ wide. In the top wall, three electrodes of sizes 25 and $50 \mu\text{m}$ wide, are kept while complementing those at the bottom wall. Initially, analyte A occupied the top half of the cavity, while analyte B occupied the bottom half. The electrodes are energized with a field of $\pm 5 \text{ V}$ and 10 KHz frequency. The bottom, side, and top electrode pairs are energized for 2 s in succession (total cycle time of 6 s). To activate the electrodes on the top wall, the $50 \mu\text{m}$ electrode at the center is grounded and the two $25 \mu\text{m}$ electrodes are energized. The concentration distribution is computed using an advection-diffusion equation for each analyte separately. The results are summarized in Fig. 3(b). At 12 s, the concentration is uniformly distributed and the system is well mixed. Even after 100 s, the analyte has hardly diffused in a static situation. Based on this simulation, we conclude that it will take at least several minutes for the diffusion process to achieve the well-mixed state if the analytes' diffusion coefficients are in the order of $10^{-10} \text{ m}^2/\text{s}$. The concentration profile of A and B with and without electrothermal flow is shown in Fig. 3(b) at different time levels. As time progresses, the concentrations evolve to a more uniform profile within a short span of time when the electrothermal flow is activated. Circulation zones near the electrode edge [Fig. 3(a)], which are created by the electrothermal effect, appear to be similar to a microfluidic system subjected to AC electro-osmotic flow.¹⁶ The flow direction is determined primarily by the sign of the electrical properties (α and β).¹² The rotational flow usually causes stretching of the material lines in the fluid. If the stretched material lines are folded back and the process repeated, a chaotic mixing can be induced.¹⁷ The flow exhibits a similarity to those observed previously by Green *et al.*,¹² in the context of electro-osmotic flow.

A similar mixing concept is demonstrated in a continuous flow system in Fig. 4. Here, electrodes are embedded on the sidewalls of a T-sensor and activated in succession for a period of

FIG. 3. (a) Microcavity is used in two analytes mixing study. (b) Comparison of concentration along the vertical axis of the cavity with (solid lines) and without (symbols) the electrothermal flow at various time levels.

2 s. The channel height is $20 \mu\text{m}$ and the main channel width is $40 \mu\text{m}$. Two streams consisting of analytes A and B are introduced into the main channel at a velocity of $100 \mu\text{m/s}$ from the top and flow downward to the exit at the bottom of the T channel. Based on the scaling arguments, we estimate that a construct with a main channel that is about a meter long will be necessary to obtain

FIG. 4. Mixing of two streams consisting of two different analytes in a T junction under the action of the electrothermal flow is shown at time $T=5$ and 10 s. Results from the pure diffusion mixing is shown on the left at time $T=40$ s.

FIG. 5. Evolution of a $10 \times 10 \mu\text{m}$ material blob in a cavity simulated via (a) particle tracking model and (b) advection-diffusion model.

a well-mixed system at the exit. To further investigate the nature of the mixing process, the evolution of a tracer blob subjected to the electrothermal flow in a cavity system shown in Fig. 5 was simulated. A blob of $10 \times 10 \mu\text{m}$ was placed at $30 \mu\text{m}$ from the mid point of the bottom wall. The analyte concentration was tracked through both the advection-diffusion model and the particle dynamics model. Results from both models are shown in Fig. 5. The electric field used was the same as the one used in Fig. 3(a). The position of the blob was tracked for two periods of 12 s. The particle lines [Fig. 5(a)] and the concentration fields [Fig. 5(b)] clearly illustrate the chaotic mixing. The material interface repeatedly stretches and folds, facilitating the mixing. The short-term growth of the length of the analyte-fluid interface (normalized by the initial interface length) is shown for two blobs of different sizes in Fig. 6. Exponential growth of this measure confirms the chaotic behavior. In particular, the interfacial growth was faster for the smaller blob ($L_0 = 10 \mu\text{m}$) compared to the larger blobs ($L_0 = 100 \mu\text{m}$), indicating more thorough mixing at small length scale.

IV. CONCLUSION

The electrothermal flow in microfluidic channels and cavities has been analyzed using both analytical and full-scale numerical simulations. The simulations show that mixing can be accomplished in a very short period of time using electrothermal flow rather than by the diffusion mechanism alone. This is applicable in both open flow and closed cavity systems. The similarity lies in the appropriate arrangement of electrodes and activation patterns to generate circulatory flow to facilitate mixing of fluids. Moreover, this mixing exhibits a chaotic behavior. Based on

FIG. 6. Short-term evolution of material interface length.

these observations, a combination of pressure-driven and electrothermal flow can be exploited to quickly and efficiently rid microsystems of trapped contaminants, or use of electrothermal flow alone, to facilitate transport/pumping of analytes.

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