

Microfluidic mixing utilizing electrokinetic instability stirred by electric potential perturbations in a glass microchannel

LUO WINJET^{*}, YARN KAOFENG^a, MING-HSYAN SHIH^b, KUANG CHENG YU^a

Department of Refrigeration, Air Conditioning and Energy Engineering, National Chin-Yi University of Technology, Taiping City, Taichung County 411, Taiwan

^a*Department of Electronic Engineering, Far East University, Tainan 744, Taiwan*

^b*Department of Mechanical Engineering, Chinese Military Academy, Fengshan, Taiwan*

The electrokinetic instability (EKI) phenomenon occurs when microfluidic flows with an electrical conductivity gradient are driven by a high-intensity external electrical field. Although EKI limits the robust performance of complex electrokinetic bioanalytical systems, it can be actively exploited to achieve the rapid mixing of micro- and nanoliter volume solutions in microscale devices. This paper investigates the EKI phenomenon in a T-type glass microchannel, in which two aqueous electrolyte solutions with a 3.5:1 conductivity ratio are driven electrokinetically into the mixing channel via the application of a DC electrical field. It is found that fluctuating, unstable waveforms are formed in the entrance region of the mixing channel and then propagate in the downstream direction when the intensity of the electrical field is increased beyond a certain critical threshold value. Electrical field intensity perturbations aligned in the direction of the conductivity gradient are then added to the DC electrical field at the upper inlet of the T-type glass microchannel. It is found that these perturbations stir the microfluidic instability and antedate the onset of flow instability. Significantly, the application of the electrical field intensity perturbations induces flow instability, and hence improves the mixing efficiency, even when the intensity of the DC electrical field is lower than the critical threshold value.

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1. Introduction

Over the past fifteen years, many integrated electrokinetic microsystems have been developed with a wide range of functionalities, including sample pretreatment, mixing, separation, and so forth. Such systems are a key component of so-called micro-total-analysis systems, which aim to integrate multiple chemical analysis functions on a single microfabricated chip. However, as the complexity of these systems increases, achieving a robust control of electrokinetic processes involving heterogeneous samples becomes increasingly important. A common concern in the microfluidics field is that of on-chip biochemical assays with high conductivity gradients. These gradients may occur intentionally, as is the case in sample stacking processes, or unintentionally, as in the case of multi-dimensional assays. Such conductivity gradients are known to result in flow instability when the fluids are driven by an external electrical field of high intensity. The presence of these electrokinetic instabilities, commonly referred to as EKI, can be regarded as a particular form of electrohydrodynamic instability, which is generally associated with electroosmotic flow. Melcher and Taylor [1] and Saville

[2] developed Ohmic models to describe the instability of electrohydrodynamic flows under the assumption of zero interfacial electrokinetic effects. In the proposed models, the fluid streams were considered to have both polarizability and a free charge, and the intensity of the internal electrical field generated by accumulated charges could be of the order of the externally imposed field. The model often uses a formulation for conservation of net charge and conductivity as scalar quantities. Hoburg and Melcher [3] performed a stability analysis of microfluidic flow in which the external electrical field was applied parallel to the liquid–liquid interface (i.e. perpendicular to the conductivity gradient) and the flow was initially at rest. Their analysis, which neglected the effects of molecular diffusion, showed that the interface between the two liquids remained stable for all electrical fields when the liquid–liquid interface was assumed to be infinitely sharp, but became unstable for all applied electrical fields when the interface was modeled with a finite width. Baygents and Baldessari [4] performed an analysis of electroosmotic flow taking the effects of conductivity diffusion into account and showed that the flow became unstable when the intensity of the applied electrical field exceeded a certain critical value. Lin *et al.* [5] considered the case

of electroosmotic flow in a long, rectangular-cross-section channel in which the conductivity gradient was orthogonal to the main flow direction and the electrical field was applied in the streamwise direction. The results showed that the flow system became highly unstable when the electrical field intensity exceeded a critical threshold value, and resulted in fluctuating flow velocities and a rapid stirring effect. Oddy and Santiago [6] presented a four-species electrokinetic instability model to investigate electroosmotic flow in a high aspect ratio geometry with a base state in which the conductivity gradient was orthogonal to the applied electrical field. The numerical simulation results were shown to be in good qualitative agreement with the experimental image data for electrolyte solutions with a conductivity ratio of 1.05:1. Chen *et al.* [7] studied the EKI phenomenon in a T-type glass microchannel. The authors performed a linear stability analysis on an electrokinetic instability model and applied Briggs-Bers criteria to identify physically unstable modes and to determine the nature of the instability. Based on a scaling analysis and the numerical results, the authors showed that the EKI phenomenon is governed by two main parameters, namely the ratio of the dynamic force to the dissipative forces, which determines the onset of EKI, and the ratio of the electroviscous velocity to the electroosmotic velocity, which governs the convective versus absolute nature of the instability.

This paper uses numerical simulations to investigate the EKI phenomenon induced by electrical field intensity perturbations in a T-type glass microchannel injection system. Two aqueous electrolytes with a conductivity ratio of 3.5:1 are driven electrokinetically into a common mixing channel via the application of a DC electrical field. Convectively unstable waves are observed within the mixing channel when the intensity of the electrical field exceeds a nominal threshold value. The unstable flow structure results in fluctuating flow velocities within the glass microchannel and a rapid stirring of the two electrolytes. Although both effects enhance species mixing, the relatively high value of the electrical field intensity at which flow instability is induced limits the practicability of the EKI phenomenon for microfluidic mixing applications. Accordingly, this study adds electrical field intensity perturbations in the direction of the conductivity gradient to the DC electrical field at the glass microchannel inlet in an attempt to antedate the onset of the EKI effect and improves the mixing efficiency, even when the intensity of the DC electrical field is lower than the critical threshold value.

2. Formula

The T-type glass microchannel considered in the present study has a uniform width and height of 60 μm .

The channel is filled with a monovalent binary electrolyte of uniform viscosity, μ and permittivity, ϵ . Since the characteristic height of the glass microchannel is in the order of magnitude of 10 μm , the interaction between the fluid and the wall is significant and must therefore be explicitly considered in the theoretical model. A review of the related literature suggests that a Ohmic model [7] for electrolytic solutions, and the Navier-Stokes equation with electric body force term provides a reasonable description of the electrokinetic instability flow in the present glass microchannel. In this model, the distributions of the electrical conductivity (σ) and the electrical field (ϕ), respectively, are both described by the electrolytic Ohmic model by assuming electroneutrality and negligible diffusive current [8-10]. For a monovalent binary electrolyte which is fully dissociated, charge density (ρ_f) and electric conductivity are related to concentration of cations (c_+) and anions (c_-) through

$$\rho_f = F(c_+ - c_-),$$

$$\sigma = F^2(c_+ m_+ + c_- m_-),$$

Where F is the Faraday constant, m is ionic mobility. Under electro-neutrality, $c_+ = c_- = c$, where c is the reduced ionic concentration, and the conductivity is proportional to this reduced ionic concentration by $\sigma = F^2(m_+ + m_-)c$. The conservation equations of charged species can be combined to yield,

$$\frac{\partial c}{\partial t} + (V \cdot \nabla)c = D_{eff} \nabla^2 c, \quad (1)$$

$$\nabla \cdot i = 0, \quad (2)$$

where V is fluid velocity and i is current density. The effective diffusivity D_{eff} is defined as

$$D_{eff} = \frac{2D_+ D_-}{D_+ + D_-}$$

where $D_{\pm}$ is ionic diffusivity, and is related to mobility by Einstein's relation $D_{\pm} = RTm_{\pm}$ where R is the universal gas constant, and T is temperature. By assuming negligible diffusive current, the current density can be

yield to $i \cong \sigma E$ where E is electric field intensity.

Applying the linear relationship between reduced concentration and conductivity, Eq. (1) and (2) reduce to

$$\frac{\partial \sigma}{\partial t} + (V \cdot \nabla) \sigma = D_{eff} \nabla^2 \sigma, \quad (3)$$

$$\nabla \cdot (\sigma E) = 0. \quad (4)$$

The electric field intensity is related to electric potential ϕ by $E = -\nabla \phi$. The conductivity distribution and electric field are governed by the electrolytic Ohmic model, Eqs (3) and (4), and the electroosmotic flow is described by the incompressible Navier-Stokes equation, i.e.

$$\frac{\partial \sigma}{\partial t} + (V \cdot \nabla) \sigma = D_{eff} \nabla^2 \sigma, \quad (5)$$

$$\nabla \cdot (\sigma \nabla \phi) = 0, \quad (6)$$

$$\nabla \cdot V = 0, \quad (7)$$

$$\rho \frac{\partial V}{\partial t} + \rho (V \cdot \nabla) V = -\nabla p + \mu \nabla^2 V + \varepsilon (\nabla^2 \phi) \nabla \phi, \quad (8)$$

where ρ is the mass density, p is the pressure and μ is the dynamic viscosity of the working fluid. Under electroneutrality, the electrical conductivity can be viewed as a material property which obeys a convective diffusion equation, Eq. (5). Equation (6) is simply Kirchhoff's Law valid for the special case where Ohmic current dominates. Through the electric body force in the momentum equation, Eq. (8), the electric field is coupled to the electroosmotic flow. In this model, electrical conductivity is not a passive scalar because a change in conductivity will alter the electric field and induce net charge, and the resulting electric body force will alter the velocity field.

In the current simulations, it is assumed that the physics of the electrical double layer (EDL) influence the instability dynamics only in that the EDL determines the electroosmotic velocity in the immediate vicinity of the glass microchannel wall. This assumption is supported by the fact that the EDLs adjacent to the current glass microchannel walls have a characteristic Debye length (λ_D) of less than 10 nm, which is considerably less than the characteristic channel width and height. The zeta potential on the homogeneous surface of the glass microchannel is assigned a value of -75 mV in this study. Thus, the boundary conditions at the walls, inlets, and outlet of the glass microchannel are given, respectively, by:

1. At the walls, with non-penetrable condition:

$$n \cdot \nabla \sigma = 0,$$

$$n \cdot \nabla \phi = 0.$$

For horizontal wall, with slip conditions:

$$u = -\frac{\varepsilon \zeta}{\mu} \frac{\partial \phi}{\partial x}, \quad v = 0,$$

$$\frac{\partial p}{\partial x} = \mu \frac{\partial^2 u}{\partial x^2} + \varepsilon \left(\frac{\partial^2 \phi}{\partial x^2} + \frac{\partial^2 \phi}{\partial y^2} \right) \frac{\partial \phi}{\partial x}.$$

For vertical wall, with slip conditions:

$$u = 0, \quad v = -\frac{\varepsilon \zeta}{\mu} \frac{\partial \phi}{\partial y},$$

$$\frac{\partial p}{\partial y} = \mu \frac{\partial^2 v}{\partial y^2} + \varepsilon \left(\frac{\partial^2 \phi}{\partial x^2} + \frac{\partial^2 \phi}{\partial y^2} \right) \frac{\partial \phi}{\partial y}.$$

where n denotes the wall-normal direction and ζ is the zeta potential of the EDL. The boundary conditions are consequences of the non-penetrable nature of the glass microchannel walls. (u, v) are the flow velocity

in the X - direction and Y - direction respectively. The electroosmotic velocity at the wall is a function of the local ionic concentration and the electrical field strength, and is given by the Smoluchowski equation. The value of the zeta potential is related to the ionic concentration, which is proportional to the ionic conductivity for dilute solutions under electro-neutrality conditions. In modeling the zeta potential, the following correlation is assumed: $\zeta/\zeta_r = (\sigma/\sigma_r)^{-j}$, where j is an empirical

constant and ζ_r is the reference zeta potential at the reference conductivity of σ_r . Note that in this study, j and reference zeta potential are assigned of zero and -75mv, respectively. Thus, zeta potential, ζ , on the homogeneous surface of the glass microchannel is -75mv.

2. At the inlets:

$$\phi = C_{EP} \text{ (lower inlet) and}$$

$$\phi = C_{EP} + \phi_m \sin(\omega t) \text{ (upper inlet),}$$

$$\frac{\partial u}{\partial y} = 0, \quad \frac{\partial v}{\partial y} = 0, \quad p = 0, \quad \frac{\partial \sigma}{\partial y} = 0,$$

where C_{EP} is the value of constant electric potentials

applied at the inlets, ϕ_m and ω denote the maximum electric potential and angular velocity generated by the AC electric power supply.

3. At the outlet:

$$\phi = 0, \quad \frac{\partial u}{\partial x} = 0, \quad \frac{\partial v}{\partial x} = 0, \quad p = 0, \quad \frac{\partial \sigma}{\partial x} = 0.$$

3. Numerical method

The numerical method used in this study employs the backwards-Euler time-stepping method to identify the evolution of the flow when driven by an AC electric field. The computational domain is discretized into 1001×701 non-equally spaced grid points in the X - and Y -directions. The calculated solutions are carefully proven to be independent of both the computational grid points and the time step. The governing equations, Eqs. (5)-(8), are discretized by of second-order central differences to form the following system of nonlinear algebraic equations:

$$H(Q, \Delta t) = 0, \quad (9)$$

where Q is the solution vector and Δt is the dimensionless marching time step.

The solution vector at various times can be obtained via a sequence of iterations, $[Q^{(m)}(t)]$, defined by

$$Q^{(0)}(0) \equiv \text{initial state}, \quad (10a)$$

$$H_Q(Q^{(m)}, \Delta t) [Q^{(m+1)}(t + \Delta t) - Q^{(m)}(t + \Delta t)] = -H(Q^{(m)}, \Delta t), \quad (10b)$$

where $m=0, 1, 2, 3, \dots$

In Eq. (10b), H_Q is the Jacobian matrix of Eq. (9) and t is the non-dimensional time.

A convergence criterion of

$$[Q^{(m+1)}(t + \Delta t) - Q^{(m)}(t + \Delta t)]^2 / Q(t)^2 < 10^{-16} \text{ is}$$

used to identify the convergence of the iteration process. The convergence efficiency of the iterations is determined by the accuracy of the initial estimate. An

effective means of obtaining a good initial estimate is to employ a Taylor expansion of a calculated convergent solution with respect to the parameter Δt , i.e.,

$$Q^{(0)}(t + \Delta t) = Q(t) + \Delta t Q_t(t). \quad (11)$$

Equation (11) is used to obtain Q_t , which satisfies

$$H_Q(Q, \Delta t) Q_t = -H_{\Delta t}(Q, \Delta t). \quad (12)$$

$H_{\Delta t}$ denotes the differential of the system of nonlinear algebraic equations with respect to Δt . The method described in Eqs. (10)-(12) is known as the backwards-Euler time-stepping method. Since this method employs second-order accuracy in time, it is necessary to provide two initial solutions at the beginning of the time-stepping calculation. One of these solutions is obtained from the initial state, while the other is obtained by the same numerical method, but with a first-order finite difference in time. Before the iteration algorithm is executed to obtain the convergence solution at the next time step, the predictor step in Eqs. (11) and (12) is applied to generate accurate estimates of the solution. The iteration algorithm is extremely effective and generally converges quadratically. Note that a detailed description of the iteration algorithm is reported in Yang [8].

3. Results and discussion

Fig. 1 presents a schematic illustration of the T-type glass microchannel considered in the current simulations. As shown, the T-type glass microchannel has a uniform width and height of $60 \mu\text{m}$, and the lengths are $708 \mu\text{m}$ and $1005 \mu\text{m}$ in Y - direction and X - direction, respectively. A driving force is developed within the T-type device by grounding the outlet of the mixing channel and applying a DC voltage across the two inlet channels. The parameters and properties of the glass microchannel considered in the present simulations are summarized in Table 1. During the injection procedure, two electrolyte solutions, one with a low conductivity and one with a high conductivity, are introduced into the microfluidic device through the lower and upper inlet channels, respectively. The conductivity ratio of the two fluids is assumed to be 3.5:1. The electrical field intensity conditions applied in the present simulations are summarized in Fig. 1(b).

Table 1. Parameters for the numerical analysis.

Height of the glass microchannel A (μm)	60
Viscosity of fluid μ ($\text{N}\cdot\text{s}\cdot\text{m}^{-2}$)	0.90×10^{-3}
Density of fluid ρ ($\text{kg}\cdot\text{m}^{-3}$)	10^3
Concentration of ions ζ (M)	10^{-6}
Dielectric constant ϵ	78.3
Permittivity of vacuum ϵ_0 (F/m)	8.854×10^{-12}
Charge of an electron e (c)	1.6021×10^{-19}
Valence Z	1
Bulk electrolyte concentration n_0 m^{-3}	6.022×10^{20}
Boltzmann constant k_b (J/K)	1.38×10^{-23}
Absolute temperature T (K)	298.16
Double layer potential ψ (mV)	-75

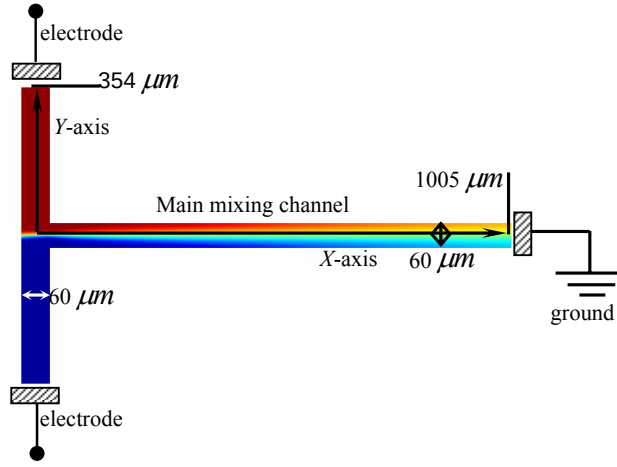


Fig. 1(a). Geometry and setup of T-type glass microchannel.

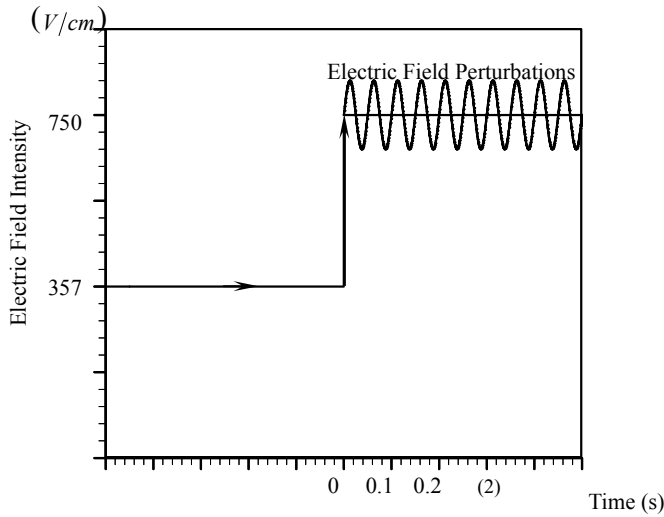


Fig. 1(b). Voltage scheme applied in current simulations showing both DC electrical field intensity and intensity perturbations.

When the intensities of constant electrical fields applied at the two inlets of the glass microchannel keep lower, two flows from the inlets have a stratified form in the entrance region of the glass microchannel and the interface between two streams from the inlets is well-defined. However, the bandwidth of the interface gradually expands along the downstream direction as a result of diffusive mixing, as shown in Fig. 1. The stratified flow conditions can be maintained when the applied electric fields is below a threshold value. For the flow conditions in this study the threshold value is found to be 550 V/cm. However, for a case a relatively low DC voltage corresponding to an electrical field intensity of 357 V/cm was applied to the inlet channels to inject the electrolyte solutions into the mixing channel and to establish an initial interface between them. Subsequently, at time $t=0$ s, a DC voltage corresponding to an electrical field intensity of 750 V/cm, i.e. higher than the critical voltage, was applied to produce instability waves in the two fluid streams. Fig. 2 presents a series of numerical images showing the temporal evolution of the electrical conductivities of the two fluid streams under a constant DC electrical field intensity of 750 V/cm. Note that in these images, the blue regions correspond to the low conductivity stream, while the red regions correspond to the high conductivity stream. The images show that at $t=0.03$ s, the interface between the two streams is only slightly perturbed, i.e. only slight flow perturbations are apparent. However, over the interval of $t=0.05\sim 0.07$ s, the wave-like form of the interface increases rapidly both in wavelength and in amplitude. At $t=0.11$ s, it can be seen that the low conductivity regions of the flow extend into the high conductivity regions in the form of a series of finger-like structures. At $t=0.14$ s, the interface and finger-like structures break down into a more complex flow pattern and the concentration fluctuations occupy the full width of the channel. The fluctuating flow velocities in the transverse and longitudinal directions associated with this unstable flow pattern result in a rapid mixing of the two streams. Overall, the results presented in Fig. 2 show that the EKI phenomenon is induced when the value of the applied electrical field intensity exceeds a critical threshold value.

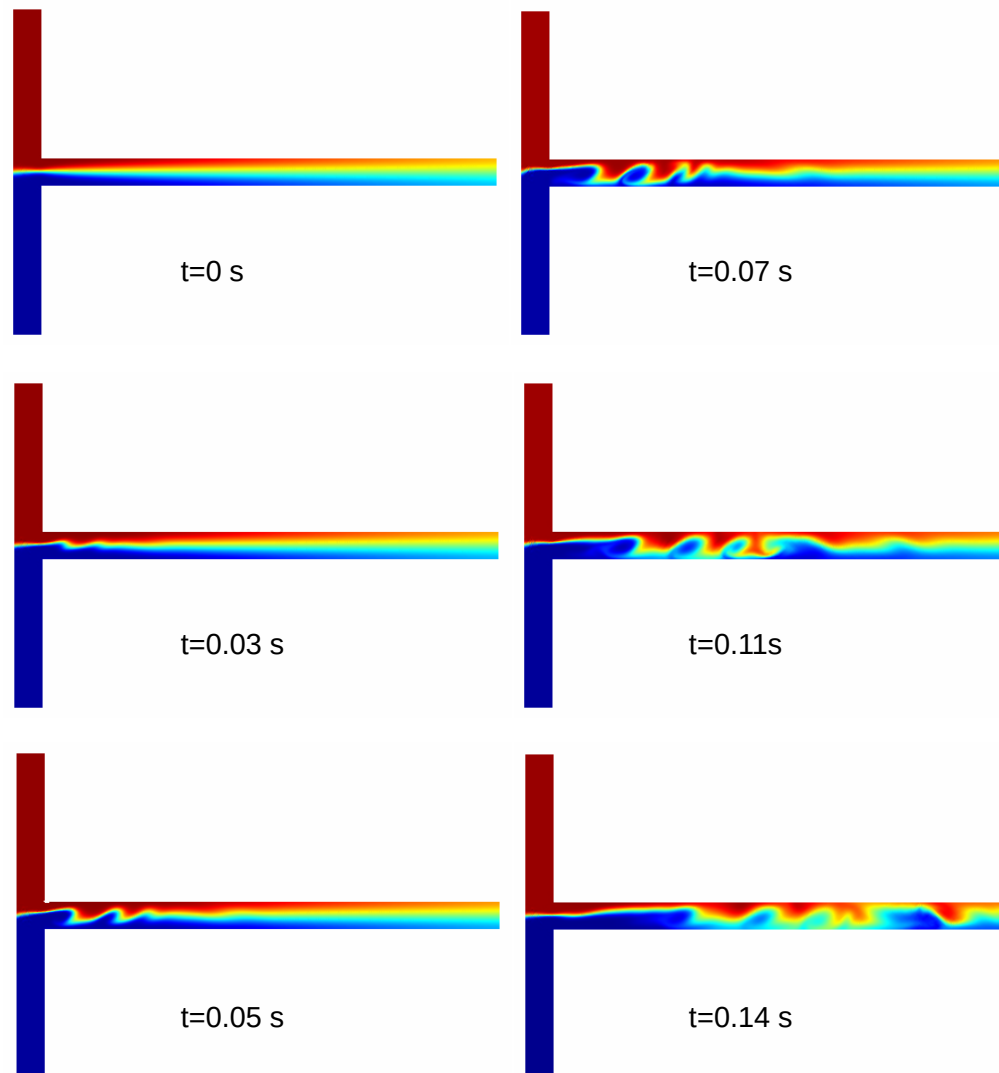


Fig. 2. Temporal evolution of electrical conductivity of two fluid streams under constant DC electrical field intensity of 750 V/cm.

In the second simulation, a sinusoidal electrical field with an amplitude of 20 V and a frequency of 20 Hz was added to the DC electrical field (intensity of 750 V/cm) at the upper inlet of the T-type glass microchannel while a DC electrical field intensity of 750 V/cm was maintained at the lower inlet. The distribution of the applied perturbation electrical field applied at the upper inlet is shown in Fig. 1(b). The resulting temporal evolution of the electrical conductivities of the two electrolyte streams is shown in Fig. 3. In contrast to the electrical field with no perturbations (shown in Fig. 2), it is observed that three weak waves are generated at the interface of the two streams in the junction region of the T-type glass microchannel and rapidly grow wave pattern at $t=0.03$ s. During the interval $t=0.03\sim0.07$ s, the presence of the electrical field perturbations at the upper inlet causes these wave patterns to twist and break down into a more complex pattern and to propagate downstream at a rate equal to the electroosmotic velocity. At $t=0.11$ s, several very mild fluctuations are formed at the interface of the two streams in the upstream region of the mixing channel. These fluctuations gradually grow in size until they occupy the full width of the channel, resulting in a second chaotic state, as shown in the final image of the sequence in Fig. 3, corresponding to $t=0.14$ s. Comparing Figs. 2 and 3, it is observed that a complex flow instability state exists at $t=0.11$ s in Fig. 2, but at $t=0.07$ s in Fig. 3. Therefore, the results indicate that the application of electrical field intensity perturbations at one inlet of the glass microchannel antedates the onset of chaotic instability in the mixing channel.

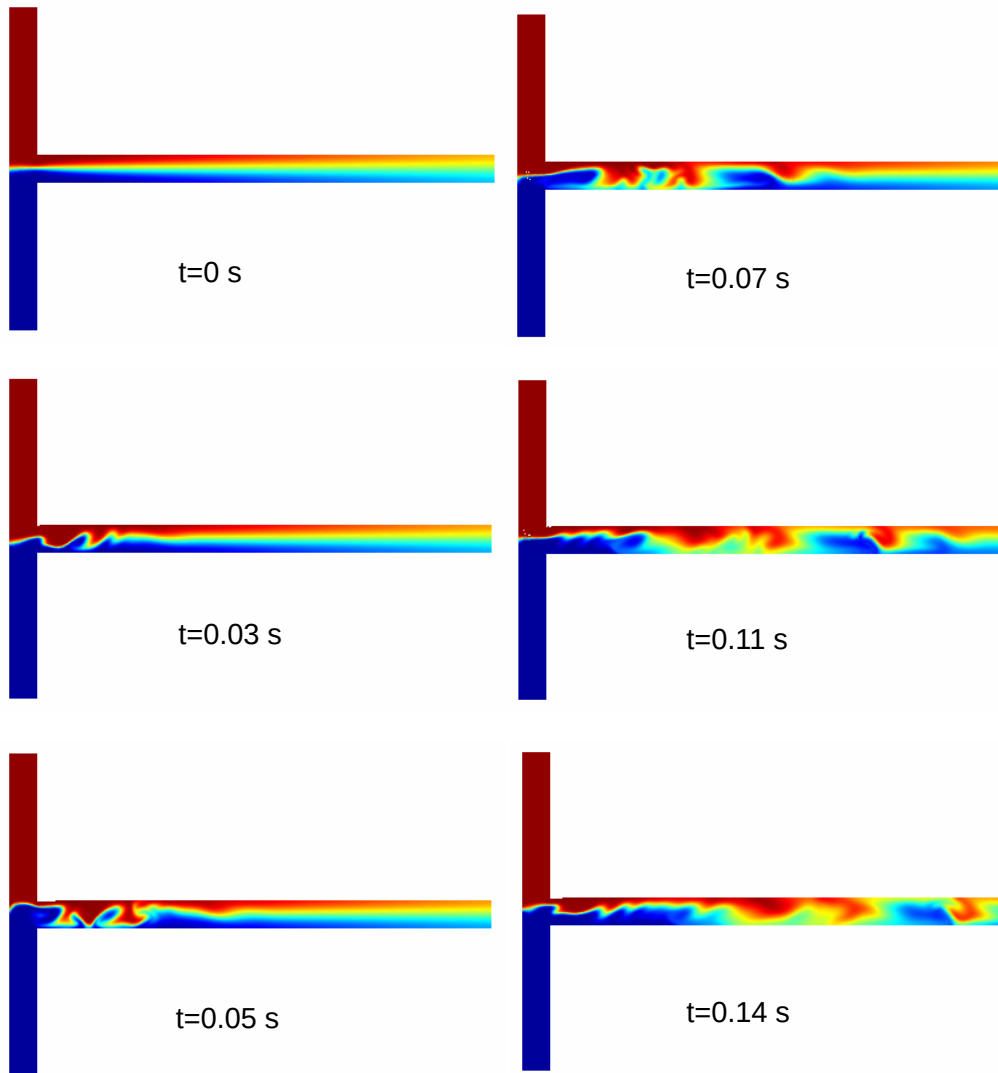


Fig. 3. Temporal evolution of electrical conductivity of two fluid streams under DC electrical field intensity of 750 V/cm with electrical field intensity perturbations.

Fig. 4 shows the distribution of the mixing efficiency along the X -axis direction of the glass microchannel at a time of $t=0.1$ s. Note that the solid line denotes the case where a constant electrical field intensity of 357 V/cm is applied, while the dotted line corresponds to the case of a constant electrical field intensity of 750 V/cm and, finally, the solid line with square symbols indicates the case where sinusoidal electrical field perturbations are applied at the upper inlet in addition to a constant electrical field intensity of 750 V/cm. The results clearly show that the flow instability conditions created by the higher electrical field intensity and by the application of electrical field perturbations, respectively, enhance the mixing efficiency. Furthermore, it can be seen that the electrical field perturbations not only increase the flow instability, but also antedate the onset of flow instability.

The images presented in Fig. 3 have shown that the

application of electrical field intensity perturbations to the original DC electrical field prompts the formation of flow instabilities when the intensity of the steady electrical field exceeds the critical threshold value of 550 V/cm. However, in practical microfluidic applications, it is desirable to induce this EKI phenomenon at lower electrical field intensity. Accordingly, a third simulation was performed to investigate the effect of adding sinusoidal electrical field perturbations to a steady DC electrical field with an intensity lower than the threshold value. Fig. 5 illustrates the temporal evolution of the electrical conductivities of the two electrolyte solutions when a sinusoidal electrical field with an amplitude of 20 V and a frequency of 20 Hz is added to a constant electrical field of 357 V/cm at the upper inlet of the glass microchannel while a constant electrical field of 357 V/cm is applied at the lower inlet. The electrical field perturbations cause the

fluid stream interface in the junction region of the glass microchannel to reciprocate periodically between the two walls of the main mixing channel, which results in the formation of a series of weak waves in the upstream region of the glass microchannel at $t=0.03$ s. These

waves gradually grow in size and propagate in the downstream direction with elapsed time, and eventually occupy the full width of the channel, as shown in the image corresponding to $t=0.3$ s.

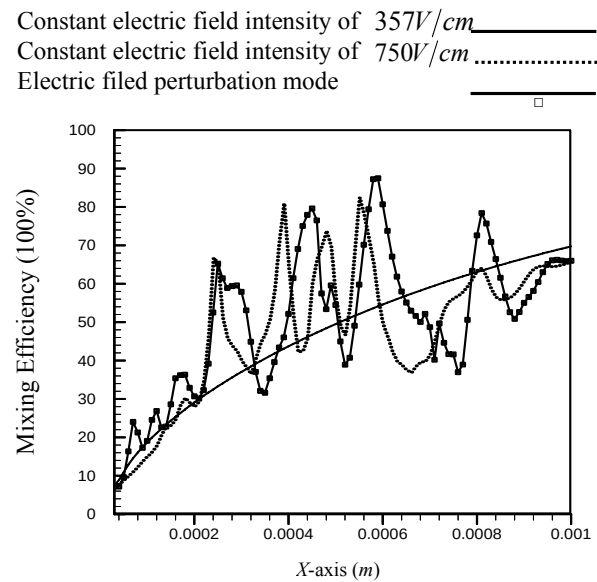


Fig. 4. Distribution of mixing efficiency along X -axis direction at $t=0.1$ s.

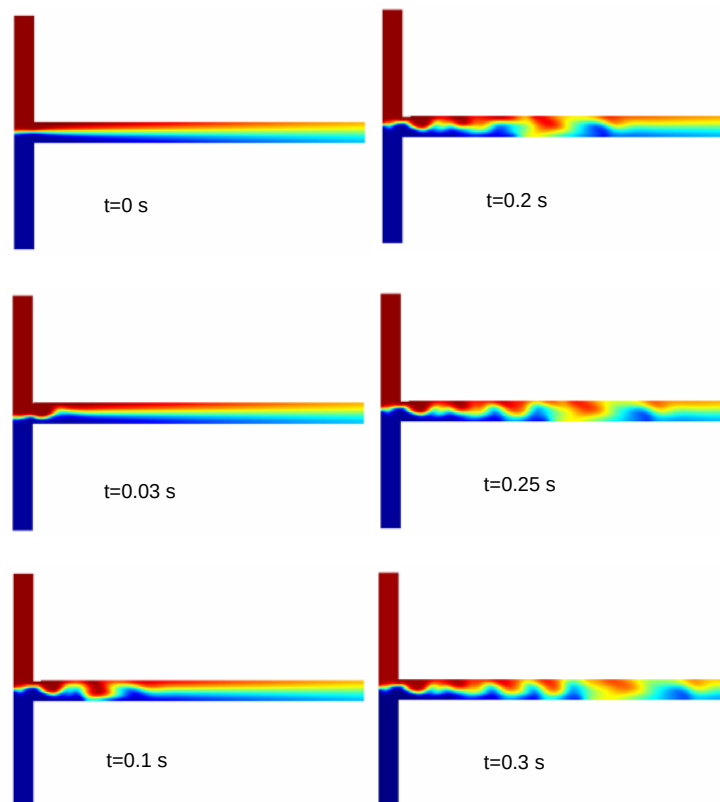


Fig. 5. Temporal evolution of electrical conductivity of two fluid streams under constant DC electrical field intensity of $357 V/cm$ with electrical field intensity perturbations.

Fig. 6 shows the distribution of the mixing efficiency along the X -axis direction of the glass microchannel at $t=0.3$ s. Note that the solid line corresponds to the application of a constant electrical field intensity of 357 V/cm, while the solid line with square symbols denotes the case where sinusoidal electrical field perturbations are applied to the constant electrical intensity of 357 V/cm at the upper inlet. The results show that the electrical field perturbations prompt flow instability conditions, and hence enhance the mixing efficiency, even when the intensity of the constant electrical field is lower than the critical threshold value.

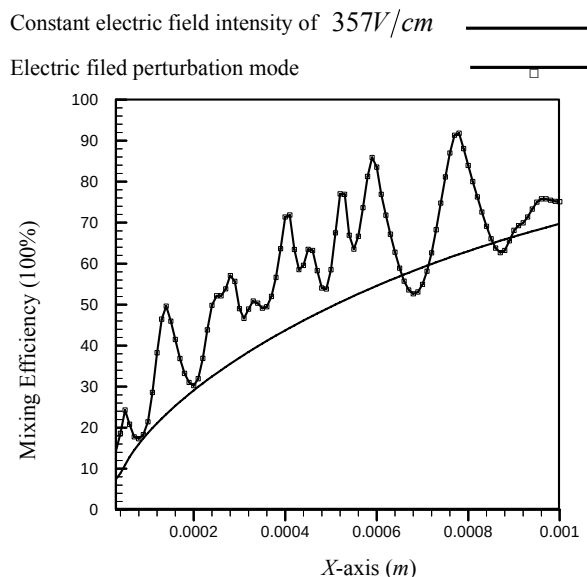


Fig. 6. Distribution of mixing efficiency along X -axis direction at $t=0.3$ s.

4. Conclusions

As the complexity of electrokinetic microsystems increases, achieving a robust control of electrokinetic processes involving heterogeneous samples becomes increasingly important. A common concern in the microfluidics field is that of on-chip biochemical assays with high conductivity gradients. Under high electrical fields in the presence of electrical conductivity gradients, electrokinetic instabilities can be induced. Such instabilities limit the robust performance of complex electrokinetic bio-analytical systems, but can be exploited for rapid mixing and flow control in microscale devices.

This paper has investigated the EKI phenomenon in a T-type microfluidic injection system in which two aqueous electrolyte solutions with a 3.5:1 conductivity ratio are driven electrokinetically into a mixing channel through the application of a DC electrical field to the two inlet channels of the device. The results have shown that when the intensity of the applied electrical field exceeds a nominal threshold value (550 V/cm in the current device), fluctuating unstable waves are formed in the inlet region of the mixing channel and then propagate in the downstream direction resulting in a mixing of the two species. The results have also shown that the application of electrical field intensity perturbations in the direction of the conductivity gradient at the upper inlet of the glass microchannel not only enhances the EKI effect, and therefore improves the mixing performance, but also antedates the onset of flow instability. Finally, the simulations have revealed that the application of electrical field intensity perturbations induces flow instability conditions even when the intensity of the DC electrical field is lower than the critical threshold value.

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*Corresponding author: wjluo@ncut.edu.tw