Mixing-dynamics of a passive scalar in a three-dimensional microchannel

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Abstract

The mixing of a diffusive passive-scalar driven by electro-osmotic fluid motion in a micro-channel is studied numerically. Secondary time-dependent periodic or random electric fields, orthogonal to the main stream, are applied to generate cross-sectional mixing. This investigation focuses on the mixing dynamics and its dependence on the frequency (period) of the driving mechanism. For periodic flows, the probability density function (PDF) of the scaled concentration settles into a self-similar curve showing spatially repeating patterns. In contrast, for random flows there is a lack of self-similarity in the PDF for the time interval considered. An exponential decay of the variance of the concentration, and associated moments, is found to exist for both periodic and random velocity fields. The numerical results also indicate that measures of chaoticity (in a deterministic chaotic system) decay exponentially in the frequency – at large frequencies – in agreement with the theory.

1. Introduction

The present work continues a series of studies on the mixing dynamics of an electrolyte solution flowing in a micro-channel [1–3]. These studies were motivated by the need of efficient and rapid mixing for biological analyses and rapid medical diagnosis [4,5]. A wide variety of passive and active methods have been used to generate mixing in micro-devices [6,7,5,8]. An active micro-mixer was proposed in [1], in which the mixing was obtained with time-dependent transverse electric fields to generate either periodic or random velocity fields. The numerical results showed a dependence of the degree of chaos in the system on the period of the advecting flow, as well as the strength of randomization. It was found that by decreasing the frequency and/or increasing the strength of the stochasticity (which would obviously generate better mixing), undesired Taylor dispersion effects were minimized. Chaotic flows occur when material lines and surfaces are stretched exponentially. The dependence of the degree of mixing on the frequency of the advecting flow described above (and also exhibited in other similar flows) is currently understood in a fairly qualitative sense.

The decay properties of the variance in concentration, without sources or fluxes at the boundaries, have been commonly used to quantify the mixing of a diffusive passive scalar quantity that is advected by a fluid. For example, studies of two-dimensional fully chaotic flows, have shown to exhibit an exponential decay of scalar variance in the long time limit [9–11]. The exponential divergence of nearby trajectories is characterized by the local finite-time Lyapunov exponent r, defined as the logarithm of stretching divided by time for this problem. The distribution of r, usually expressed by the probability density function, has also been used to explain the rate of decay in scalar variance [11,12]. It has been argued that if the domain scale is significantly larger than the flow scale, the description based on Lyapunov exponents alone can be inadequate for the prediction of decay rates during the final stage of mixing [13,14].

Accurate experimental measurements of [15,16] in chaotic two-dimensional time-periodic flows revealed an exponential decay in tracer variance, with evidence for persistent spatial patterns in the concentration field. These repeating patterns are also known as strange eigenmodes; at the late stages of mixing, the scalar represents a periodic eigenfunction of the linear advection–diffusion equation. The appearance of self-similar asymptotic probability density function (PDF) of the scalar field normalized by its variance, suggests that strange or statistical eigenmodes appear in flows with aperiodic time-dependence and has been examined in detail in [10,17–19]. Camassa et al. [20] studied the PDF for a...
decaying passive scalar advected by deterministic velocities. They found a lack of self-similarity in the PDFs with time-periodic flows and self-similar PDFs in steady flows. Liu and Haller [18] have shown among other things, that for this advection–diffusion problem, a finite-dimensional inertial manifold exist, in which the decay of the tracer at the late stages of mixing is governed by the structure of the slowest-decaying mode in the manifold.

Our study addresses two important issues that to the best of our knowledge have not been documented before. The first is the appearance of strange eigenmodes in three-dimensional flows. The second focuses on connecting the theory regarding measures of chaoticity and its exponential decay (in a deterministic chaotic system) with results from numerical simulations; such connection is conspicuously lacking [21–24].

As in [3], we assume that the zeta potential at the lateral electrode walls is zero. Due to the capacitive charging at the electrode, there is a field-induced electro-osmosis – termed as AC electro-osmosis – and the zeta potential is non-zero [25,26]. Since the velocity profiles for AC electro-osmosis in perfectly polarizable electrodes is zero at high and low frequencies, and maximum at some intermediate characteristic frequency [27], in order to quantify the sole effect of the period modulation of the electric field in the mixing process, the effects of the field-induced electro-osmosis are not considered in our model. As it will be shown in the following sections, this simplified model reveals novel features not present in previous mixing mathematical formulations. Specifically, it is demonstrated that spatially repeating patterns develop for periodic flows, and that an exponential decay of variance in concentration exists. Additionally, quantitative measures of chaoticity for both periodic and random flows are provided.

The article is organized as follows: the governing equations and numerical integration scheme are briefly presented in Section 2. The results and discussion from the numerical simulations are presented in Section 3. A summary is presented in Section 4, which concludes the paper.

2. Governing equations and numerical method

We consider the electro-osmotic flow (EOF) inside a long rectangular micro-channel of height $2H$, and width $2W$ as shown in Fig. 1. This is the EOF micromixer studied by Pacheco and co-workers [1–3], where the typical dimensions for the device are $10 \text{ cm} \times 100 \mu \text{m} \times 200 \mu \text{m}$. The primary steady flow along the channel is driven by a steady electric field, generated by zeta-potentials on the top and bottom surfaces. An unsteady transverse motion of the fluid is driven by secondary electric fields generated with four micro-electrodes placed on the lateral walls.

In Cartesian coordinates, $(x,y,z)$ with their corresponding unit vectors $(\hat{i},\hat{j},\hat{k})$, the non-dimensional velocity vector and pressure are denoted by $\vec{u}$ and $p$, respectively. The non-dimensional electric potentials due to an external electric field is denoted by $\phi$ and the electric potential due to the electric charge at the walls by $\psi$. This

![Fig. 1. Schematic of the flow apparatus to generate the velocity fields. The inset shows the streamlines of the transverse velocity field $\vec{u}_T$ for $(\phi_1,\phi_2,\phi_3,\phi_4) = (0,0,1,0)$. On the right, the primary flow is driven by the velocity generated by the electric potential $\phi$, at $y = \pm W$.](image1)

![Fig. 2. Three-dimensional view of $\vec{u}_L = \vec{u}(y,z,t)$ along the channel. (a) The maximum velocity occurs near the center of the channel and the slip-velocity model for this EOF is not valid; (b) the highest velocity occurs near the central portion of the upper and lower plates and the slip-velocity model may be valid.](image2)
system is governed by the Navier–Stokes equations in the limit of small Reynolds number and by the advection–diffusion equation

\[ \mathbf{V} \cdot \mathbf{u} = 0, \]

\[ \text{ReSt} \frac{\partial \mathbf{u}}{\partial t} = -\nabla p + \nabla \cdot (\kappa \nabla \mathbf{u}) - \kappa^2 \psi (C_1 \mathbf{j} - \mathbf{C}_0 \nabla \cdot \mathbf{u}), \]  

\[ \text{St} \frac{\partial c}{\partial t} + \mathbf{V} \cdot (c \mathbf{u}) = \text{Pe}^{-1} \Delta c, \]  

where \( t \) is the time and \( c \) is the concentration of the reagent.

The governing equations are scaled using \( L_c = H \) for the space variables, \( U_c = c \varepsilon \mathbf{E}_0 \) for the velocity, \( \kappa = 2\pi/\alpha_c \) for time, \( L_c \mathbf{E}_0 \) for the transverse electric potential \( \phi \), and \( \zeta \) for the induced electric potential \( \psi \). The electrical permittivity of the solution is \( \varepsilon \), \( \mathbf{E}_0 \) is the electric field along the channel, \( \mu \) is the fluid viscosity, and \( \alpha_c \) is a characteristic frequency of the transverse electric field \( \mathbf{E}_0 \). The Reynolds number is \( Re = \rho U_c L_c / \mu \), the Strouhal number is \( St = L_c \alpha_c / 2\pi U_c \), and the Péclet number is \( \text{Pe} = U_c L_c / D_m \), where \( \rho \) is the fluid density, \( D_m \) is the mass diffusivity of the reagent. The dimensionless Debye length is \( K^{-1} = (L_c^2 \kappa)^{-1} \) and \( \kappa^{-1} \) is the Debye length used to describe the thickness of the electric double layer. The constant \( C_1 = \mathbf{E}_0 / \varepsilon \) gives information on the resident time of the reagent, and \( C_0 = \zeta_0 / L_c \mathbf{E}_0 \).

The last three terms in the right-hand-side of the momentum equation correspond, respectively, to the normalized axial electric field \( \mathbf{E}_0 \), the transverse field \( \mathbf{E}_t \), and the induced electric field \( \mathbf{C} \cdot \mathbf{j} / \zeta \), and the induced electric field \( -\nabla \cdot \mathbf{u} \). The governing Eqs. (1)–(3) are solved subject to no-slip boundary conditions for the velocity on all walls \( \mathbf{u}(\pm 1, \pm 1, \cdot) = 0 \) and no mass flux normal to the wall \( \nabla \cdot \mathbf{n} = 0 \), where \( \mathbf{n} \) is the unit normal vector directed into the fluid and \( \Gamma = H/W \) is the aspect ratio. The electric potential \( \psi \) is the solution to the Helmholtz equation \( \nabla^2 \psi = K^2 \psi \) with \( \psi(y, \pm 1, \cdot) = 1 \) and \( \psi(\pm 1, z, \cdot) = 0 \). The electric potential \( \phi \) is obtained by solving the Laplace equation \( \nabla^2 \phi = 0 \) with linear boundary conditions, thus allowing us to write \( \phi(y, z, \cdot) = \sum \phi_i(t) \phi_i(y, z) \). The functions \( \phi_i(y, z) \) are solutions of the Laplace equation with Neumann boundary conditions on the top and bottom walls, and Dirichlet boundary conditions on the lateral walls, i.e., \( \phi_i(\pm 1, \cdot) = 0 \). The functions \( \phi_i(t) \) are solutions of the Laplace equation with periodic boundary conditions in space for \( t \), with \( \phi_i(0) = \phi_i(T) \) for \( i \) odd and \( \phi_i(0) = \phi_i(T) + \pi/2 \) for \( i \) even.

The dimensions of the channel are set to \( L_c = 5, L_r = 1, \) and \( L_r = 2 \) which fixes the aspect ratio at \( \Gamma = 0.5 \). Periodic boundary conditions are imposed on \( x = 0 \) and \( L_r \). The selection of a variable-length time-like interval protocol has proven to be most effective in generating mixing for this configuration [3]. Throughout most of this study, we shall keep \( K = 100, C_1 = 10^{-2}, \text{Re} = 0.04 \) and \( \text{Pe} = 10^4 \) and vary \( T \).

Fig. 3. Evolution of the concentration of the periodic flow (\( c = 0 \)) at multiple times showing the convergence to a strange eigenmode.
To solve (1)–(3) a semi-implicit second-order projection scheme is used on a staggered mesh, where all spatially-varying terms are treated implicitly [28]. A \( \frac{800}{C_2} \) \( \times \frac{200}{C_2} \) \( \times \frac{240}{C_2} \) grid mesh is used in the \( x \)-, \( y \)-, and \( z \)-directions respectively. Non-uniform grids, which are stretched away from the vicinity of the walls using a hyperbolic tangent function, are used in cross-section, whereas uniform grids are employed along the channel. The convergence of the code has already been described for the same flow in [3,2] and successfully tested against solutions of the velocity field obtained with a collocated arrangement of the variables on the grid [29,30].

3. Results and discussion

A comparison of the cross-section and lateral view between Figs. 2(a) and (b) demonstrate the effect of the Debye length on \( \vec{u}_i \). When the Debye length is comparable to the wall length scales, \((K^{-1} = 10^{-1})\) the velocity field shown in Fig. 2(a) resembles that of a pressure-driven flow along a channel. As the Debye length is reduced to \( K^{-1} = 10^{-2} \), the velocity field can be approximated as a slip-driven flow in a channel. Note, however, the sharp decrease of the velocity close to the upper and lower walls, as shown in Fig. 2(b). Only in the regions close to the top and the bottom walls, and only near the central portion of the cross-section \( z < |f| \), the primary velocity field \( \vec{u}_i \sim 1 \). Note that gas bubbles induced by electrochemical reactions, when embedded electrodes are used – as in the present case – may occur when the voltage applied is high enough. The formation of bubbles is detrimental to any and all electric-based microfluidic system; however, using AC fields, lower ionic strength buffers, short run times, and high frequencies, bubble creation can be largely mitigated [31–33].

Fig. 2(a)–(b) demonstrates the evolution of the deformation of the scalar field for the late stages of mixing, the advection–diffusion equation (3), with initial conditions \( c(x, t_0) = 0.5\sin(2\pi y)\sin(2\pi z) \), is considered next. Since the current study focuses on the decaying scalar of the field with \( \langle c(x, t) \rangle = 0 \), then the scaled concentration is defined as \( \chi = c/C_1 \), where the spatial average \( \langle \cdot \rangle \) is carried out over the entire domain.

Fig. 3. The extracted values of \( \nu_n \) vs \( n \) for \( \varepsilon = 0 \). The symbols correspond to \( \nu_n \) and the line to \( \nu_n = (n - 1)\nu_2 \).

Fig. 4. Decay of the various moments for \( \varepsilon = 0 \). \( \nu_n \) for \( n = 2 \); \(-\Delta-\), \( n = 3 \); \(-\cdots-\), \( n = 4 \).

Fig. 5. The extracted values of \( \nu_n \) vs \( n \) for \( \varepsilon = 0 \). The symbols correspond to \( \nu_n \) and the line to \( \nu_n = (n - 1)\nu_2 \).
(\langle c(\mathbf{x}, t)^n \rangle \sim e^{-\chi^2} [34] \) with \( \nu_n \) growing linearly with \( n \). This theoretical prediction is also valid when \( \nu_n \) is a nonlinear function of \( n \), in this case, the moments are not stationary. It is easy to verify, by extracting the slope from the values of \( \nu_n \) using the portion of the curves showing linear behavior in the semi-log plot of Fig. 4, that the moments decay as predicted by the theory. The equations of the lines are plotted in Fig. 5 along with the values of \( \nu_n \) showing that decay-rates grow linearly with \( n \). The long-time behavior of the re-scaled moments \( \langle \chi^n \rangle \) for \( n = 2, 3, 4 \) is depicted in Fig. 6. Note that the moments become stationary which also implies that \( \nu_n \) is a linear function of \( n \), and the PDF of the re-scaled variable \( H(\chi) \) must be self-similar. Note also that for higher values of \( T \) the time at which the moments become stationary decreases.

The evolution towards the self-similar stage for \( H(\chi) \) is shown in Fig. 7 for \( T = 10 \) and \( 20 \), with \( \varepsilon = 0 \). The regions that remain relatively isolated are captured by the bi-modal characteristic of \( H(\chi) \) shown in Fig. 7(a). The bi-modal nature of \( H(\chi) \) tends to disappear due to the decrease in size of regions of poor mixing, forming a long core and tailed PDF, as shown in Fig. 7(b). Self similar behavior also appears for values of \( Pe \) number different from the one illustrated here, except that for higher values of \( Pe \), the time at which the evolution becomes self-similar increases.

The long-time evolution of \( H(\chi) \) and the moments are shown in Fig. 8 for \( T = 20 \) and \( \varepsilon = 1 \). Since the flow is now fully chaotic, \( H(\chi) \) is unimodal with a Gaussian structure. However, it can be seen that never settles down into a self-similar eigenmode as demonstrated by the oscillatory nature of the higher-order moments of Fig. 8.

Sukhatme [19] has argued that in the strange eigenmode regime the maximum scale of variation of the scalar field \( (l_c) \) is of the same order as the scale of variation of the velocity field \( (l_v) \), and that the exponential decay of moments is also valid when \( l_c < l_v \) but in this case, \( \nu_n \) is a nonlinear function of the order of the moment \( n \). For small values of \( Pe \), large-amplitude fluctuations of the rescaled moments were also reported.

In the current work, the periodic flow \( (\varepsilon = 0) \) is in the self-similar regime, with \( l_c \sim l_v \) and the rescaled moments decay exponentially. For the random flow \( (\varepsilon = 1) \), the exponential decay of the moments is also exponential, but it does not exhibit self-similarity for the PDFs, at least for the times considered in this investigation. Since the introduction of randomization in the process is equivalent to increasing the diffusion, and the results of self-similarity are valid for large \( Pe \), it is expected that a strong modulation of the flow would produce fluctuations with large amplitude in \( \langle c(\mathbf{x}, t)^n \rangle \) in the late stages of mixing, preventing the PDFs from becoming stationary. The argument here is that the lack of self-similarity of the PDFs is due to the large number of eigenmodes of the advection–diffusion operator that are present throughout the randomization process [18]. Nevertheless, the asymptotic
self-similarity of the tracer PDFs remains to be established for our velocity fields with aperiodic time-dependence. The location of 10,000 passive non-diffusive particles for \( e = 0 \), 0.5 and 1 and \( T = 3, 10 \) and 20 is shown in Fig. 9. The particles are initially located within the circular disk of radius of 0.1 centered at \((x, y, z) = (1, 0, 0)\). When the flow is periodic \((e = 0)\) there are two regions of poor mixing whose size shrinks as \( T \) increases from 3 to 20 as shown in the left-hand-side of Fig. 9(a)–(c). As the strength of the randomization \( e \) increases from 0 to 1, the particles spread to a wider region covering nearly the entire \( yz \)-plane for all the values of \( T \) considered. The work of [21,23,24,22] indicate that measures of chaoticity in a deterministic chaotic system decay exponentially in the frequency at large frequencies (or small period \( T \)).

This decrease with the frequency is exhibited in Fig. 9, an apparently valid conclusion for \( e = 0 \) and \( e \neq 0 \). This is one issue that this study investigates in more detail. To this end, we consider the measure of chaoticity \( \lambda \), as the ratio between the measure of chaotic domain in the Poincaré section to the area of the whole domain. The chaotic domain is determined by extracting the sign of the largest Lyapunov exponent of 400\( \times \)200 particles placed on the center of uniform grid cells on the plane \( yz \) at \( x = 1 \).

Fig. 10 shows the measure of chaoticity \( \lambda \) \((e = 0 \) and \( e = 0.5)\) with the analytical expression \( e^{\alpha/T} \) as function of the period \( T \). Theoretically, \( \lambda \sim e^{\alpha/T} \) for \( e = 0 \), where \( \alpha = 0.2 \) is a constant whose value was obtained by regression analysis [35]. The shape of the curve \( \lambda \) for the periodic flow remains relatively unchanged for \( t > 1 \) (plotted here at \( r = 1,000 \)), but it gradually degenerates due to errors associated with the numerical integration of particle trajectories. On the contrary, the measure of chaoticity \( \lambda \) for \( e \neq 0 \) is time-dependent, and global chaos is achieved rather quickly for \( T > 0 \). Fig. 10 depicts \( \lambda \) for \( e = 0.5 \) at \( t = 200 \). The transient nature of this process is an interesting problem that requires further investigation.

4. Summary

This manuscript has discussed the mixing enhancement induced by periodic and random velocities in an electro-osmotic flow of an electrolyte solution flowing inside a three-dimensional micro-channel. It was found that the PDF of the scaled concentration settles into a self-similar curve for the periodic flows in which the scalar field shows spatially repeating patterns. When stochasticity is introduced, the random flow does not enter into the strange eigenmode regime for the interval of time considered in this investigation. It was confirmed an exponential decay of the variance of concentration for both the periodic and random flows. Also found was the fact that the measures of chaoticity decay exponentially with the frequency at large frequencies of the advecting flow in agreement with the theory. There are many aspects of mixing in electro-osmotic flows which require more exploration, and this study provides the framework for further investigations. One of them is the quantification of the AC electro-osmosis in parameter space \((e, Pe, T)\) for fixed \( K \), which will be the subject of forthcoming papers; another is the use of Eulerian predictor for optimal mixing appropriate for steady, three-dimensional channel flows [36].

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Fig. 9. Influence of $\varepsilon$ on the location of 10,000 passive non-diffusive particles projected onto the yz-plane at $t = 500$. The particles are initially located within a circular disk of radius 0.1 with center at $(x, y, z) = (1, 0, 0)$.

Fig. 10. Measures of chaoticity and theoretical prediction vs $T$: $(\Diamond)$, $\varepsilon = 0$ at $t = 1,000$; $(\bigodot)$, $\varepsilon = 0.5$ at $t = 200$; $(\square)$, $e^{-\lambda T}$ with $\lambda = 0.2$.

References


