Electrothermal blinking vortices for chaotic mixing

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ABSTRACT

At microscale, an efficient mixing is a key step which dramatically accelerates bio-reactions. For thirty years, Dynamical System theory has predicted that chaotic mixing must involve at least 3 dimensions (either time dependent 2D flows or 3D flows). However, in microfluidics, few works have yet presented efficient embedded micro mixers. In this regards, electrokinetic has emerged as an efficient way to generate time dependent flows with embedded electrodes, thus requiring a battery rather than external pumps. This paper presents the first experimental and theoretical study of 2D time dependent electrothermal chaotic mixing. To do so, asymmetric vortices are blinked to perform time dependent asymmetric flows. Successive horizontal plans of velocity are vortices are blinked to perform time dependent asymmetrical flows. To tackle this issue, more and more research has been devoted to chaotic advection to perform non diffusive mixing with time dependent flow. By folding and stretching flows, laminar chaotic motion has been achieved at microscale decreasing exponentially mixing time $t_m \approx ln(Pe)$ [1]. Today, a huge effort is made to provide portable lab-on-chip with embedded flow actuators. For this reason, electrokinetic has emerged over the last fifteen years as one of the major solution to drive flows with simple electrodes integrated in the microfluidic environment [2]. To that end, AC electro-osmosis has been extensively studied and applied to drive and mix low conductivity solutions [2]. Yet, most biological assays have a high conductivity ($1 mS.cm^{-1}$ (ELISA buffer) < $\sigma < 16 mS.cm^{-1}$ (PBS)). AC electrothermal flows, which have been scarcely studied, occur in high conductivity solutions allowing to drive physiological solutions. In this paper, we will study theoretically and experimentally 2D electrothermal blinking vortices to mix reagent in microwell plates. After justifying the choice of the ACET model based on our previous work [3] we will explain our $\mu$PIV experimental setup. We will then validate flow model with $\mu$PIV flow measurement and discuss the mixing efficiency at multiscale with the mix-variance method.

Keywords: micro-mixing, chaotic advection, electrokinetic, blinking vortices

1 INTRODUCTION

The subject of mixing is an old problem which has been extensively studied this last century from theoretical and technological perspectives. Today, economic revenues of product involving fluid mixing is measured in billions of dollars per year. One realm where major technical improvements in mixing are required is lab-on-chip also called ($\mu$TAS), where microfluidic flows are strongly laminar. Diffusion based passive mixers have been hindered by long time reaction occurring especially for large particles (high Peclet number) at micro and meso-scale (low Reynolds number). To tackle this issue, more and more research has been devoted to chaotic advection to perform non diffusive mixing with time dependent flow. By folding and stretching flows, laminar chaotic motion has been achieved at microscale decreasing exponentially mixing time $t_m \approx ln(Pe)$ [1]. Today, a huge effort is made to provide portable lab-on-chip with embedded flow actuators. For this reason, electrokinetic has emerged over the last fifteen years as one of the major solution to drive flows with simple electrodes integrated in the microfluidic environment [2]. To that end, AC electro-osmosis has been extensively studied and applied to drive and mix low conductivity solutions [2]. Yet, most biological assays have a high conductivity ($1 mS.cm^{-1}$ (ELISA buffer) < $\sigma < 16 mS.cm^{-1}$ (PBS)). AC electrothermal flows, which have been scarcely studied, occur in high conductivity solutions allowing to drive physiological solutions. In this paper, we will study theoretically and experimentally 2D electrothermal blinking vortices to mix reagent in microwell plates. After justifying the choice of the ACET model based on our previous work [3] we will explain our $\mu$PIV experimental setup. We will then validate flow model with $\mu$PIV flow measurement and discuss the mixing efficiency at multiscale with the mix-variance method.

2 THEORY

Electrothermal flows are due to the interaction of an electric field with non-uniform permittivity and conductivity generated by Joule heating. For fluid with large conductivities, the temperature raise in the fluid due to Joule heating is significant. We are using here the enhanced model of ACET presented in [3] developed for a general temperature raise above 5°C. This model uses an electrical thermal coupling and temperature dependent expression for the electrical conductivity and dynamic viscosity.

For aqueous solutions, a linear approximation of the temperature dependence of the electrical conductivity and permittivity is adequate, $\sigma_m(T) = \sigma_m(T_0)(1+c_\sigma(T-T_0))$ and $\varepsilon_m(T) = \varepsilon_m(T_0)(1+c_\varepsilon(T-T_0))$ where $c_\sigma = \frac{1}{\sigma_m(T_0)} \frac{\partial \sigma_m}{\partial T} |_{T_0} \approx 0.02 C^{-1}$ and $c_\varepsilon = \frac{1}{\varepsilon_m(T_0)} \frac{\partial \varepsilon_m}{\partial T} |_{T_0} \approx -0.0044 C^{-1}[4]$, with $T_0$ being a temperature of reference.

Consider an applied AC voltage with angular frequency $\omega$. Complex notations can be used for the electric field, $E$, and electrical potential, $V$, ($E = -\nabla V$):
\[ E = Re \left[ \tilde{E} e^{i \omega t} \right] , \quad V = Re \left[ \tilde{V} e^{i \omega t} \right] . \] The AC electrothermal flow is modelled by solving for the voltage \( \tilde{V} \), the temperature \( T \) and the fluid velocity \( \mathbf{u} \) using the following set of equations:

\[
\begin{align*}
\nabla^2 \tilde{V} &= c_e \nabla T \cdot \nabla \tilde{V}, \quad \tilde{E} = -\nabla \tilde{V} \\
\n\nabla \cdot (k_m \nabla T) + \frac{\sigma_m(T)}{2} |\tilde{E}|^2 &= 0 \\
\n\nabla \cdot (\mu_m(T) (\nabla \mathbf{u} + (\nabla \mathbf{u}^T))) + \mathbf{F} &= \nabla P, \quad \nabla \cdot \mathbf{u} = 0
\end{align*}
\]

(1)

where the fluid heat conductivity, \( k_m \), is considered constant, the fluid dynamic viscosity \( \mu_m(T) \) is described by a polynomial equation in \( T \) [4] and \( P \) is the pressure.

The force \( \mathbf{F} \) is expressed as the sum of ACET Force and Buoyancy Force: \( \mathbf{F} = < \mathbf{F}_{ACET} > + \mathbf{F}_b \).

The time-averaged electrostatic body force (ACET force) is given by:

\[
\mathbf{F}_{ACET} = \frac{\varepsilon_m(T_0)}{2} \left[ (c_e - c_o) (\nabla T) \cdot \tilde{E} - \frac{1}{2} c_e \nabla T |\tilde{E}|^2 \right]
\]

(2)

The buoyancy force is given by:

\[
\mathbf{F}_b = \rho_m(T_0) \beta(T - T_0) \mathbf{g}
\]

(3)

where \( \beta = 10^{-3} \text{C}^{-1} \) is the coefficient of thermal expansion, \( \mathbf{g} = 9.8 \text{m} \cdot \text{s}^{-2} \mathbf{z} \) is the acceleration of gravity, and \( T_0 \) is the temperature of reference.

The assumptions used here are: small Reynolds number, \( Re = \frac{\rho_m u_0 L}{\mu_m} \ll 1 \) (\( \rho_m \) is the fluid density, \( u_0 \) is the characteristic velocity, \( L \) is the characteristic length), small electric Reynolds number, \( Re_{el} = \frac{\varepsilon_m u_0}{\sigma_m L} \ll 1 \), small thermal Peclet number, \( Pe_T = \frac{\rho_m c_p u_0 L}{k_m} \ll 1 \) (\( c_p \) is the heat capacitance of the fluid), large AC voltage frequency \( \omega \tau \ll 1 \) \( (\tau = \frac{\tau_{rel}}{\varepsilon_m}) \) is the charge relaxation time, and the ratio of charge relaxation time and thermal diffusion time is small, \( \tau_{rel} = \frac{\varepsilon_m k_m}{\sigma_m \rho_m c_p L^2} \ll 1 \).

For a characteristic length, \( L = 320 \mu m \), the thermal-diffusion time in water is \( \tau_{diff} = \frac{\rho_m c_p L^2}{k_m} \approx 0.8 \text{s} \) and the viscous-diffusion time in water is \( \tau_{fl} = \frac{\rho_m L^2}{\mu_m} \approx 0.1 \text{s} \) thus the transient time for the system to reach the stationary solution given by the system of equations 1 is of the order of 0.8s. In this work we will ignore the transient behaviour of the system as the period of switching between configurations of applied voltage on the electrodes is much greater than 1s.

### 3 MATERIAL AND METHODS

Experiments are carried out in a 320 \( \mu m \) thick and 8 \( \text{mm}^2 \) wide PDMS well. Beneath this micro-well, a sinusoidal electric signal is applied between 3 interdigitated gold electrodes, separated by a 100 \( \mu m \) gap. The phase signal \( V_{pp} = 11 V \) and the ground are switched between the two side electrodes at a frequency 0.01Hz \( < F_s < 0.2 \text{Hz} \), whereas the opposite phase signal \( -V_{pp} \) is steadily applied to the center electrode (Figure 1). The temperature of the PCB is set by a thermo-electric device at 20°C so as to control the temperature rise.

The PIV image pairs are taken using an infinity corrected objective lens (Nikon, \( M = 20, NA = 0.45 \)) mounted with a 0.5× magnification relay lens to provide the view of the whole vortices. Relatively large, \( d_p = 1 \mu m \), diameter particles were chosen to decrease bias and un-bias error and to dampen beads diffusion without exhibiting significant DEP forces. The depth of the PIV measurement is therefore estimated to be \( \delta_z = \pm 7.5 \mu m \) [5]. The successive horizontal plans of velocity are measured snapping 2 images at 100ms with two pulsed lasers (MiniLase II-30 (New Wave research, Fremont, CA, US) synchronized with a camera PIVCAM 13.8 (TSI Shoreview, MN, US)). Bead velocities are then extracted after ensemble averaging 30 correlation functions of bead positions using a custom PIV interrogation software [6] (Figure 2.a,b). Then 25 transversal velocity vectors \( \mathbf{u} \) are spatially averaged above each gap. The measured results are then compared to the modeled velocity vertically averaged over \( h \pm \delta_z \) (Figure 2.c).

Since electrodes are parallel, the flow is 2D. Electrothermal flows are thus modelled with 2D finite element method using Comsol Multiphysics v4.2a (COM-
SOL Inc., Stockholm, Se) (Figure 2). The three equations, (1), are solved taking into account electrothermal coupling. Thermal diffusion time, $\tau_{diff}$, and viscous diffusion time, $\tau_{vis}$, being both negligible compared to the switching period, blinking vortices are assumed to follow a square shape signal. To theoretically evaluate mixing efficiency, particles trajectory is modelled using COMSOL particle tracking module. We use the mix-variance coefficient (MVC) [7] to evaluate mixing at different scales (Figure 4). To do so, the domain is successively divided in boxes along the x and y direction up to $n_x$ and $n_y$ boxes, respectively. For each box configuration, average bead concentration is computed. The variance of these concentrations is then evaluated:

$$MVCS = \frac{1}{n_x n_y} \sum_{i=0}^{n_x} \sum_{j=0}^{n_y} (\rho_{ij} - 0.5)^2 \quad (4)$$

Figure 2: Experimental Measurement of fluid velocity for the configuration $11V_{pp} - 11V_{pp} 0$. $\mu$PIV results at height $z = 30\mu m$ (a) and $z = 195\mu m$ (b). (c) Experimental and simulation flow velocity averaged above left and right gap for different height $z$.

4 RESULT AND DISCUSSION

Breaking and shifting the symmetry increase the chaotic advection efficiency by enhancing stretching and folding motion [8]. For this reason, we switched an assymetric electric field over time. Figure 1 displays the side-view of both states of the blinking vortices. For the first configuration (i), the applied potential on the left, central and right electrodes are $V_L = 11V_{pp}$, $V_c = -11V_{pp}$, $V_r = 0V_{pp}$. A faster vortex occurs with its center above the edge of the left electrode. Conversely, a right slower vortex whose center is shifted above the right gap slips under the left vortex, thereby the separatrix between the two vortices is leaning at $\approx 20^\circ$ deg (Figure 1b). Switching the voltage of both side electrodes (i.e configuration (ii)) leads to a shift of the vortices and a rotation of the separatrix. As a result, there is an overlap between the two different flow configurations leading to a transfer of particles from one side to the other.

Figures 2a,b display the in-plane velocity field measured by $\mu$PIV corresponding to the configuration (i).

Two different heights (30$\mu$m and 195$\mu$m) behind and above the vortex center are displayed, respectively. As predicted by the theory, in both configurations the flow goes down above the central electrode. The separatrix is shifted to the left by $\approx 60\mu m$ over a height of 165$\mu m$, thereby confirming a tilt of $\approx 20^\circ$ deg and thus the overlap of the vortex configurations.

Figure 3 compares the vertical profile of both modelled and measured horizontal velocities spatially averaged above each gap. Both velocities match with slight differences, certainly coming from uncertainties in the geometrical and thermal characterisation of the experimental device. At the vicinity of the electrode, the gradient of velocity being very high, the velocity is not null, since it is integrated on $\pm \delta z$. With 10 percent of difference, this confirms the relevance of our model described in Eqs.1.

Figure 3 shows snapshots of the simulated mixing with particle tracking. Two kind of particles are confined on two different side of the device. The slow left vortex drag down blue particles along the 20$^\circ$ diagonal, thus stretching the separatrix. After 4s green particles are encircled by blue ones in the left vortex. After 10 seconds, vortices blink, folding the trapped green particles in the left side vortex and stretching the separatrix to encircle blue particle in the right side vortex. During folding, both particles are layering 3 times around the center of the vortex. Hence, in that case every half period $T/2 = 10s$, vortices switch, consecutively stretching and folding flows forming particle layers of smaller and
smaller thickness which is characteristic from chaotic advection.

Using the concentration of beads at different scales, the efficiency of mixing has been evaluated using the mix-variance norm (MVC), Eq. 4, for different period of blinking vortices (Figure 4). When MVC is 0.25 there is no mixing, i.e., particles are surrounded by neighbours of the same kind. When MVC decreases, mixing is enhanced with a perfect mixing reached at MVC=0. Confirming the earlier described stretching and folding, which are achieved between 4s and 10s, the optimal mixing is reached for a period between 10 and 20s. For longer period, the mixing is slower, whereas for faster period less stretching and folding are achieved.

REFERENCES