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Rapid mixing by turbulent-like electrokinetic microflow

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HIGHLIGHTS

- The work characterizes novel rapid turbulent mixing in a microchannel at relatively low *Re* in the viewpoint of statistical turbulence.
- The work opens a new avenue by redefining the limits where turbulent mixing can be present.
- The work provides a new perspective view on turbulent mixing at microfluidic length scales.

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ABSTRACT

Rapid mixing has been achieved in a pressure-driven microflow which is forced under AC electric field. In our previous investigations, the mixing on large scale at the centerline of microchannel has been studied. Here, we show that the evaluation on mixing effect by flow visualization could be misleading, if its temporal resolution is not sufficiently high. By using single-point laser induced fluorescence (LIF) method, the mixing on both large and small scales can be investigated with high spatiotemporal resolution. It is found fast mixing is not only achieved at the centerline, but also in the majority of cross-sectional area, even near the bottom wall region. This is resulted from the large scale secondary flow due to unbalanced AC electroosmotic flow near the bottom wall. The temporal −5/3 spectrum of concentration measured with LIF in our previous investigations is also supported by the spatial spectrum of concentration. The physical process of mixing on small scales is further investigated by the flatness of concentration gradients.

1. Introduction

In chemical engineering, relatively slow mixing processes are often a bottleneck that restrict the reaction process, especially when the desired reaction rate is high. For this purpose, fast mixing is highly required to avoid the reactive process being delayed by the relatively slow mixing processes, and to reduce potential side products. Essentially say, the principle of mixing enhancement is to accelerate the cascade process of scalar structures in the entire cross section from large to small scales where molecule diffusion becomes dominant. In macroflows, this can be achieved by generating turbulent flows, through many ways, such as agitated tanks (Paul et al., 2003; Armenante and Huang, 1992; Ascanio et al., 2002; Tsouris and Tavlarides, 1994; Rudolph et al., 2007; Kresta, 1998; Ali et al., 1981; Alvarez et al., 2002; Chang et al., 1981), mixing layers (Fiedler et al., 1998; Ho and Huang, 1982; Ho and Huerre, 1984; Wang, 2003; Wang, 2006; Dimotakis and Brown, 1976; Koochesfahani and Dimotakis, 1985; Koochesfahani and Dimotakis, 1986; Koochesfahani and Mackinnon, 1991; Wygnanski and Fiedler, 1970; Dimotakis, 2005) or jets (Dimotakis, 2005; Catrakis and Dimotakis, 1996). However, in microreactors and other lab-on-a-chip applications, where Reynolds number (*Re*) is normally on the order of unity or below, it was believed that there was no turbulence in microfluidics at such a low Reynolds number (Brody et al., 1996).

Turbulence is commonly believed to be a feature of high *Re* flows. Fundamental theory of generating turbulence in low *Re* flows is not yet established. In microchannel, due to the strong viscosity effect, any initially generated velocity fluctuation will be dissipated immediately. Hence, to achieve turbulent flow, there should be a mechanism of providing high and long-lasting turbulent energy. Normally, the turbulent energy can be generated

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Nomenclature

x	streamwise direction and position
t	time
W	width of channel
f	frequency
η	dynamic viscosity
σ	electric conductivity
σ_1	initial σ in stream 1
ζ	zeta potential
$\vec{u}$	flow velocity vector
E_y	y-directional component of $\vec{E}$
C	dye concentration
$C = C - \overline{C}$	temporal concentration fluctuation around $\overline{C}$
$C''_{rms} = \sqrt{\overline{C}''^2}$	root-mean-square of C''
λ	degree of mixedness
$u_e = \sqrt{\frac{\varepsilon d E_0^2(\sigma_2-\sigma_1)}{\rho\sigma_1}}$	electric-inertial velocity
$Gr_e = \frac{\rho\varepsilon d^2 E_0^2(\sigma_2-\sigma_1)}{\sigma_1\eta^2}$	electric Grashof number
$Fn = \frac{\overline{\left(\frac{dC'}{dx}\right)^4}}{\left[\overline{\left(\frac{dC'}{dx}\right)^2}\right]^2}$	flatness of concentration gradient
y	spanwise direction and position
l	length scale
h	height of channel
f_f	forcing frequency of AC voltage
$\upsilon = \eta/\rho$	kinematic viscosity
ε	electric permittivity
σ_2	initial σ in stream 2
$E_0 = V/W$	nominal electric field
$\vec{E}$	electric field
$U_{HS} = -\varepsilon\zeta E_y/\eta$	Helmholtz-Smoluchowski velocity
C	spatial averaging of C
$C'' = C - C$	temporal concentration fluctuation around C
C_0	initial dye concentration in dye side
E_c	power spectrum of C by single-point LIF
$Pe = WU_b/D$	Peclet number
$l_d \sim 2\sqrt{Dt}$	diffusion length scale
$\overline{\cdot}$	temporal averaging
z	vertical direction and position
d	characteristic transverse length scale of channel
$k = 2\pi/l$	wavenumber
ρ	fluid density
D	diffusivity of dye in water solution
ρ_f	initial free charge density
V	applied voltage
p	pressure
$\vec{F}_e$	electrical body force
U_b	bulk flow velocity
$\overline{C}$	temporal averaging of C
$C'_{rms} = \sqrt{\overline{C'^2}}$	root-mean-square of C
$C_{ref} = \sqrt{C(C_0 - C)}$	a reference dye concentration of fully mixed
$E_{c,s}$	spatial spectrum of C by visualization
$Re_e = du_e/\upsilon$	electric Reynolds number
$l_{de} = d/Re_e$	nominal electric-force-dissipation scale
$\cdot$	spatial averaging

based on intrinsic flow instability under either high *Re*, or from external forcing. However, the former one is difficult to realize in microfluidics. While the latter one can be achieved by both active methods, e.g. electrokinetic (EK) flow (Baygents and Baldessari, 1998; Chen et al., 2005; Chang and Yang, 2007; Huang et al., 2006; Park et al., 2005; Posner and Santiago, 2006; Ramos et al., 1998), and passive ways, e.g. specially designed geometry of channels (Stroock et al., 2002; Hessel et al., 2005). So far, based on recent review (Lee et al., 2011), all the disturbed flows generated by external (or active) forcing are chaotic flows, not turbulence.

Wang et al. (2014) first observed turbulent-like flows with fast mixing in microfluidics with *Re* on the order of unity without forcing. Then, the scalar turbulence generated by the AC EK mechanism was investigated in details (Wang et al., 2016). However, their investigations on mixing effect are related to the large scale scalar structures only. And there are little data about the transverse evolution of mixing process.

In this manuscript, the misleading of mixing effect caused by flow visualization is shown first. Then, the accurate mixing effect is evaluated by a single-point laser induced fluorescence (LIF) measurement with high spatial and temporal resolution. The mixing process is further investigated by both spatial and temporal spectra of concentration at different positions. Here, the uniformity of mixing on multiscale are evaluated and the effectivity of temporal spectrum is qualitatively supported by the spatial spectrum from visualization. Finally, the transport mechanism of scalar on small scale is also analyzed based on the LIF measurement.

2. Mechanism of generating EK turbulence

Previously, many investigations have been conducted to generate disordered and irregular flows in microchannel by EK methods, such as periodic electroosmotic flow (Lim et al., 2010), dielectrophoretic flow (Lee et al., 2011; Campisi et al., 2009; Choi et al., 2009; Zhao and Yang, 2011; Lee et al., 2001; Deval et al., 2002), and electrothermal flow (Ng et al., 2009). EK based micromixer is very common and can be simply induced by arrange an external electric field either parallel (El Moctar et al., 2003) or perpendicular (Chen et al., 2005) to solution conductivity gradient. Disordered flow can be generated due to EK instability when the electric Rayleigh number is beyond its critical value ($Ra_{e,c}$) (Posner and Santiago, 2006).

By increasing the external electric field, high electric-inertial velocity (u_e) can be generated. This leads to larger electric Reynolds number ($Re_e = du_e/\upsilon$, where d is a reference length scale of transverse direction, υ is the kinematic viscosity) and cause the flow to be unstable 28 compared with unforced flows. If the electric-inertial velocity is sufficiently high, it is possible to generate a turbulent flow region where viscous effect can be overcome. Hence, the key issue becomes how to generate a higher u_e. u_e can be estimated by dimensional analysis from the Navier-Stokes equation with electric body force, as below:

$$\rho\left(\frac{\partial\vec{u}}{\partial t} + \vec{u}\cdot\nabla\vec{u}\right) = \nabla p + \eta\nabla^2\vec{u} + \vec{F}_e \tag{1}$$

where ρ, $\vec{u}$, p, η and $\vec{F}_e$ are the fluid density, flow velocity, pressure, dynamic viscosity and electrical body force, respectively. Since both the electrical forces related to the gradient of electric permittivity and fluid compressibility are negligible (Wang et al., 2014, 2016), we approximately have $\vec{F}_e = \rho_f\vec{E}$, where $\vec{E}$ is the electric field and $\rho_f = -\varepsilon\vec{E}\cdot\nabla\sigma/\sigma$ denotes the free charge density (Chen et al., 2005). ε is the permittivity of the electrolyte, σ is the electric conductivity of medium and $\nabla\sigma$ is the conductivity gradient. From Eq. (1), we can easily conclude u_e is of the order $\sqrt{|d\varepsilon(\vec{E}\cdot\nabla\sigma)\vec{E}/\rho\sigma|}$, when $\vec{F}_e$ is on the same order of the convection term. Obviously, u_e can be increased by (1) increasing the conductivity ratio between the two streams; (2) applying higher electric