



Long-range electrothermal fluid motion in microfluidic systems



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ABSTRACT

AC electrothermal flow (ACEF) is the fluid motion created as a result of Joule heating induced temperature gradients. ACEF is capable of performing major microfluidic operations, such as pumping, mixing, concentration, separation and assay enhancement, and is effective in biological samples with a wide range of electrical conductivity. Here, we report long-range fluid motion induced by ACEF, which creates centimeter-scale vortices. The long-range fluid motion displays a strong voltage dependence and is suppressed in microchannels with a characteristic length below $\sim 300 \mu\text{m}$. An extended computational model of ACEF, which considers the effects of the density gradient and temperature-dependent parameters, is developed and compared experimentally by particle image velocimetry. The model captures the essence of ACEF in a wide range of channel dimensions and operating conditions. The combined experimental and computational study reveals the essential roles of buoyancy, temperature rise, and associated changes in material properties in the formation of the long-range fluid motion. Our results provide critical information for the design and modeling of ACEF based microfluidic systems toward various bioanalytical applications.

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1. Introduction

AC electrokinetics is a promising technology for the development of integrated point-of-care diagnostic systems [1,2]. AC electrokinetic phenomena, such as AC electrothermal flow (ACEF), enables fluid manipulation for automating bioanalytical procedures. ACEF arises from Joule heating induced temperature gradients developed within a fluid when an external electric field is applied [3]. The temperature gradients create local electrical conductivity, permittivity, viscosity, and density gradients in the solution. These gradients and their interactions with the electric field, in turn, create bulk fluid force and motion. ACEF is effective for fluids with a large range of electrical conductivity and is particularly useful for manipulating conductive physiological samples and biological buffers. By properly designing the electrokinetic devices,

ACEF has been demonstrated for pumping, mixing, concentration, separation, signal enhancement and noise reduction [4–18].

A theoretical model of ACEF was developed for parallel electrodes with a small gap [3]. In this model, the equilibrium temperature distribution was determined by considering the energy balance equation. The effect of fluid motion on the temperature distribution was ignored due to the low velocity. The gradients in permittivity and electrical conductivity, which interact with the electric field to generate the bulk fluid force, were approximated based on their linear temperature sensitivity. The buoyancy effect was not considered due to the small length scale of microfluidics. This classical model forms the basis of numerous ACEF studies and facilitates the design of various bioanalytical devices. With the rapidly expanding applications of ACEF, the classical model, however, does not fully describe the diverse behaviors of ACEF observed in microfluidic systems. Discrepancies between the fluid motion and the classical model were reported at high voltages and conductivities [9,19,20]. Several modifications of the classical model, including buoyancy effects, temperature-dependent parameters and convective heat transfer, have been proposed.

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Nomenclature

c	specific heat capacity of the fluid	\mathbf{u}	fluid velocity
E	magnitude of the electric field	V_{pp}	peak to peak voltage
\mathbf{E}	electric field	V_{rms}	root mean square voltage
E_{rms}	root mean square electric field	<i>Greek symbols</i>	
f	frequency of the AC voltage	α	parameter defined as $\alpha = (\partial\varepsilon/\partial T)/\varepsilon$
f_c	crossover frequency	β	parameter defined as $\beta = (\partial\sigma/\partial T)/\sigma$
\mathbf{F}_e	time-averaged electrothermal force	ε	electrical permittivity
\mathbf{g}	gravitational acceleration	μ	viscosity of the solution
\mathbf{I}	identity matrix	ρ_m	medium density
\mathbf{J}	the current density	ρ_q	charge density
k	thermal conductivity	σ	electrical conductivity
L	characteristic length	τ	ratio of $\varepsilon(T)/\sigma(T)$
\hat{n}_θ	unit vector in tangential direction	ϕ	electrical potential
p	pressure	ω	angular frequency of the AC voltage, $\omega = 2\pi f$
Pe	Péclet number		
Ra	Rayleigh number		
T	temperature		

Nevertheless, the effects of these modifications and the diverse behaviors of ACEF are only partially understood. A systematic experimental investigation along with modeling and simulation is required to decipher the diverse behaviors of ACEF and provide guidelines on the design of ACEF based bioanalytical devices.

In this study, we investigate an uncharacterized long-range fluid motion induced by ACEF, which creates centimeter scale vortices. The long-range fluid motion is studied under various channel dimensions, sample properties, and operating parameters. Microfluidic channels with characteristic lengths from 100 μm to 1 mm are designed to investigate the temperature and velocity profiles. Fig. 1a and b shows the cross-sectional and top views of the microchannel. The fluid motion and the temperature profile in the microchannel are experimentally characterized by particle image velocity and infrared thermometry. The effects of sample electrical conductivity, voltage, and frequency, are investigated systematically. An extended computational model of ACEF, which describes the effects of the density gradient and temperature-dependent parameters, is also developed to compare with the experimental results. Using the combined experiment and computation approach, we elucidate the effects of buoyancy, convective heat transfer, boundary conditions, and temperature-dependent parameters on the long-range electrothermal fluid motion.

2. Materials and methods

2.1. Microfabrication

The microfluidic device consisted of a pair of parallel electrodes in a polydimethylsiloxane (PDMS) microchannel (Supplementary Fig. S1). The length, width and distance of the electrodes were 7 mm, 100 μm , and 50 μm respectively. The electrodes were fabricated by evaporation and lift-off on glass microscope slides (Thermo scientific, Portsmouth, NH). Briefly, the glass slides were cleaned by acetone and isoamyl alcohol, dried with nitrogen, and baked for 15 min at 125 $^\circ\text{C}$. The electrode pattern was defined by photolithography with AZ-3312 photoresist (AZ Electronic Material, Branchburg, NJ). The electrodes were deposited by evaporating 50 nm of titanium, 150 nm of gold, and 50 nm of titanium onto the glass slides followed by the lift-off process with acetone. The microchannels were fabricated by PDMS molding. The mold was created by a rapid prototyping process based on a computer numerical control system with micrometer precision (HAAS automation Inc., Oxnard, CA). The length and width of the

microchannel were 5 cm and 5 mm respectively. The depth of the microchannel varied from 100 μm to 1000 μm for the theoretical and experimental study of ACEF. PDMS was mixed at 1:10 curing agent to elastomer base ratio and incubated at 65 $^\circ\text{C}$ for 5 h. Inlets and outlets were punched into the PDMS chamber by a 0.7 mm Harris Uni-Core punch tool (Ted Pella, Inc., Redding, CA). The sample chamber and electrode glass substrate were sterilized and bonded by air plasma treatment (Harrick Plasma, Ithaca, NY). An assembled device is shown in Supplementary Fig. S1.

2.2. Experimental setup

The microfluidic chip was loaded on a Leica DMI-4000B inverted epi-fluorescence microscope (Leica Microsystems, Wetzlar, Germany). A Nikon Eclipse E800 upright epi-fluorescence microscope (Nikon Instruments INC., Melville, NY) was also used to observe the fluid motion on the electrode. Images and videos were captured using a Cooke SensiCAM QE CCD camera (PCO-TECH Inc., Romulus, MI). A 5 mm prism (PS609, Thorlabs, Newton, NJ) was setup on the inverted microscope to monitor the fluid motion from the side (Supplementary Fig. S2). A function generator (33220A, Agilent, Englewood, CO) was used for generating the AC potential. The AC potential was monitored by a digital oscilloscope (DSOX2012A, Agilent, Englewood, CO) during the experiment.

2.3. Image processing and particle image velocimetry

Fluorescent microspheres (F8810; Invitrogen, Carlsbad, CA) with a diameter of 200 nm were applied for particle image velocimetry. The particles were diluted using Mueller Hinton (MH) broth with an electrical conductivity ~ 1.1 S/m (at 21 $^\circ\text{C}$). The images and videos were taken by a 10x objective (Leica HC PL FL, NA = 0.3) and processed using the NIH ImageJ software. The velocity field was calculated using the open source JPIV software package. The depth of focus for PIV measurement was approximately 8 μm . The CCD pixel size was 6.4 μm . The measurement uncertainty can be calculated applying the following formula $\delta_x \approx \frac{d_e}{10M} = 20$ nm, where d_e is the effective particle diameter [21,22]. In the experiment, the particle density was 6–8 particles in each 128×128 pixel interrogation window. All velocity measurements were performed 100 μm above the bottom of the channel surface and 500 μm away from the electrode (Fig. 1a and b). The setting was optimized for electrohydrodynamic measurement with minimal influence of dielectrophoresis [23,24].

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