Experimental verification of Faradaic charging in ac electrokinetics

Wee Yang Ng,^{1,2} Yee Cheong Lam,^{2,a)} and Isabel Rodríguez^{1,b)}

¹*Institute of Materials Research and Engineering, Agency for Science, Technology and Research (A*STAR), 3 Research Link, Singapore 117602, Singapore*

²*School of Mechanical and Aerospace Engineering, Nanyang Technological University, Nanyang Avenue 50, Singapore 639798, Singapore*

(Received 13 February 2009; accepted 21 March 2009; published online 23 April 2009)

This paper investigates the phenomenon of Faradaic charging in ac electrokinetics. Faradaic reactions were suggested as a key effect responsible for the reversal of pumping direction in ac micropumps. However, this hypothesis has yet to be proven convincingly and directly. Here we present an ion detection strategy to determine the production of ions through Faradaic hydrolytic reactions originating from direct application of voltage to electrolytic solutions during ac electrokinetics. Experiments were performed with symmetrical planar electrodes aligned along a microfluidic channel. Fluorescein, a *pH*-dependent dye, was employed as the *pH* indicator for the detection of ion production. Images were captured for analysis at various voltage levels. From analyzing the fluorescence intensity and its distribution, it can be concluded that the production of ions from hydrolytic reactions takes place and increases with the ac voltage. The coefficient of deviation indicates a significant enhancement at ac voltage above 11 V_{pp}. Lastly, we demonstrate a strategy using dc-biased ac electrokinetics to achieve controllability in direction and magnitude of the net fluid flow in pumping application. © 2009 American Institute of Physics. [DOI: 10.1063/1.3120273]

I. INTRODUCTION

The ac electrokinetics (ACEK) phenomenon has been utilized in a variety of microfluidic applications. One key advantage is that it offers localized flow control on microfluidic devices, and thus a viable platform to be integrated onto miniature diagnostic kits.^{1,2} Applications with ACEK include particle/cell transportation, trapping, sorting,^{3–6} and fluid pumping and mixing.^{7–19} Dielectrophoresis, ac electro-osmosis (ACEO), and ac electrothermal (ACET) are identified as the main categories in ACEK.²⁰ Conventional ACEO flows are generated via a pair of coplanar microelectrodes. With ac voltage applied, ions in the electric double layer respond to the electric field and create a flow movement. This mechanism is referred to as capacitive charging. It has been widely used as an actuation mechanism in ac electrokinetic micropumping^{7–16} and recently as a strategy in a micromixer to enhance mixing in a fluidic channel.^{17–19} One particular phenomenon in ACEO is Faradaic charging where hydrolytic reactions occur at the electrode surfaces which generate coions enhancing the generation of fluid movement. This effect is being debated and has yet to be conclusively proven directly.^{11–16} It is observed that the injected charges interact with the applied electric field producing a stronger fluid flow and in some instances the flow direction is opposite to that of capacitive type of charging.

Faradaic charging in ACEO was first discussed independently by Lastochkin *et al.*¹¹ and Studer *et al.*¹² They observed the reversal pumping effect experimentally in their micropump at higher ac voltages and higher frequencies. Lastochkin *et al.*¹¹ used an orthogonal *T* electrode array

^{a)}Electronic mail: myclam@ntu.edu.sg.

^{b)}Electronic mail: i-rodriquez@imre.a-star.edu.sg.

for their micropump. Sodium chloride (NaCl, 100 mS/m) electrolytes were used and reversal pumping was observed with voltage greater than $8 V_{\text{rms}}$ ($22.6 V_{\text{pp}}$, > 100 kHz). In the work of Studer *et al.*,¹² an asymmetrical electrode array was employed to achieve fluid pumping. Using potassium chloride (KCl, $10^{-4} M$, ~ 1 mS/m) electrolytes, reversal pumping was observed with voltage greater than $2.5 V_{\text{rms}}$ ($7.1 V_{\text{pp}}$, > 10 kHz). Using a symmetrical electrode array with potassium chloride (KCl, 2 mS/m) as the electrolyte, Ramos *et al.*¹³ reported pumping reversal in their traveling-wave electro-osmosis when the applied voltage was greater than $2 V_0$ ($4 V_{\text{pp}}$, 1 kHz). More recently, Urbanski *et al.*¹⁴ observed similar reversal pumping effects with a nonplanar three-dimensional (3D) electrode array with de-ionized water (~ 0.03 mS/m) as the working fluid. Pumping reversal occurred when the frequency exceeded 25 kHz at $3 V_{\text{rms}}$ ($8.5 V_{\text{pp}}$). Similar studies on reversal pumping were reported by Garcia-Sanchez *et al.*¹⁵ and by Yang and Wu.¹⁶ Theoretical models considering nonlinear analysis and the effect of charge injection (i.e., inclusion of reaction resistance R_s and Warburg impedance Z_w) had been reported, but a full understanding for the reversal pumping effect is yet to be established.^{20–22}

Several factors have been discussed to address the observation of reversal pumping: (1) Nonlinear capacitance across the diffuse layer when the voltage drop over the layer is larger than 25–50 mV;^{12,14,22} (2) concentration polarization which sets up a conductivity gradient between the electrodes;^{12,13,15} (3) Faradaic charging where coions are injected, giving rise to a charge layer above the electrode.^{11–13,16,21–23} The charge increase in solution is the common effect highlighted in all these works despite the differences in electrode and microchannel configurations, solution conductivity, and voltage level. In this investigation, we aim to demonstrate directly the existence of Faradaic reactions in ACEO using symmetrical electrodes. Detection of ions from incipient hydrolytic reactions was carried out using the pH-dependent fluorescein molecule.^{24,25} We first demonstrate the feasibility of this strategy with dc voltage where the detection of H^+ and OH^- ions is reflected by the fluorescence intensity change of the fluorescein solution. Subsequently, we investigate the effect of ac voltage on the production of ions due to Faradaic reactions. Lastly, we illustrate the use of dc-biased ACEK for fluidic control in pumping applications. We had already reported previously a micromixer with a mixing efficiency of 90% over 180 ms using dc-biased ACEK,¹⁸ and we will expand here the applicability of this strategy for the control of flow rate and direction in pumping.

II. DEVICES AND MATERIALS

A. Device fabrication

All fabricated microdevices consist of a Pyrex glass patterned with gold electrodes enclosed by a polydimethylsiloxane (PDMS) microchannel. The electrode design and PDMS microchannel vary accordingly for ion detection or fluid pumping experiments. Standard photolithography processes were employed to pattern the electrodes on glass. The metal electrodes were sputtered onto a Pyrex wafer with an adhesion layer of chromium (~ 20 nm) followed by a layer of gold (~ 100 nm). A lift-off process was performed to remove the undesired photoresist (AZ7220). The microchannels were molded on PDMS (Sylgard® 184, Dow Corning Corporation).²⁶ PDMS was prepared by mixing the base and curing agent at a 10:1 weight ratio. The PDMS mixture was poured onto a silicon master and cured for 1 h at 80°C .

The microdevice for H^+/OH^- ion detection consists of a pair of symmetrical gold electrodes (width=160 μm , gap=80 μm , thickness=120 nm) on a Pyrex wafer enclosed by a PDMS microchannel (width=400 μm , depth=45 μm). The microdevice for fluid pumping experiments consists of an array of asymmetrical gold electrodes of 120 nm thickness on a Pyrex wafer. The widths of the wide and narrow electrodes are 15 and 5 μm , respectively. The alternate gaps between the wide and narrow electrodes are 15 and 5 μm , respectively. These electrodes are enclosed by a PDMS microchannel (width=1 mm, depth=160 μm).

FIG. 1. Experimental setup illustrating device layout and electrical connections: (A1) ac/ac+dc voltage and (A2) dc voltage arrangement.

B. Microbeads and dye preparation

Potassium chloride (Merck) prepared with a conductivity of 1 mS/m is the electrolyte used in all experiments. Fluorescein, a pH -dependent fluorescent dye, is employed in the pH tracing experiments. The solution of fluorescein is colorless at a pH below 4 and exhibits a strong fluorescence at pH above 6.²⁴ Fluorescein sodium salt (Sigma-Aldrich) is prepared in the potassium chloride solution to a concentration of $10^{-5}M$ (pH 6.3, 1.2 mS/m). The pH of the fluorescent dye was later adjusted by hydrogen chloride (0.1M) to pH 3.8 (9.8 mS/m) or by sodium hydroxide (0.1M) to pH 10 (2.6 mS/m). An acidic fluorescent solution was used for the detection of OH^- ion production and a basic fluorescent solution was used for the detection of H^+ ions. The fluorescence emission spectrum over a range of pH was measured using a spectrofluorophotometer (Shimadzu, RF-5301PC) with the excitation wavelength set at 490 nm. Red fluorescent microbeads of 1 μm (Invitrogen, Molecular Probes) in potassium chloride (1 mS/m) were employed as the flow tracer for the fluid pumping experiments. pH and conductivity were measured using a pH -conductivity meter (Radiometer Analytical, IONcheck65).

C. Experimental setup and electrical connections

Figure 1 depicts the experimental setup for our experiments. The microdevice was mounted onto an upright microscope (Olympus, BX51) with a mercury lamp attachment for fluorescence imaging. Charge-coupled device cameras (Roper Scientific, Photometrics Cascade 512B and JVC, TK-C1481BEG) were used to capture the images and videos of the experiments for analysis. Experiments were conducted for micropumping (with asymmetrical electrode array) and ion detection (with a pair of symmetrical electrodes). For ion detection experiments, a syringe pump (Harvard Apparatus, PicoPlus) was used to inject the fluorescent solution.

For the electrical signals, a function generator (Instek, function generator GFG-8216A) was used to supply the ac voltage and dc-biased ac voltage signals. For experiments with dc voltage

FIG. 2. Superimposed image of the microbeads flow path in (a) forward pumping, 10 V_{pp} at 120 kHz and (b) reversal pumping, 20 V_{pp} at 120 kHz. Electrode configuration: widths=15 and 5 μm , gaps=15 and 5 μm .

alone, a dc power supply (GW Instek, PPE-3323) was used instead. An amplifier (Thurlby Thandar Instruments, WA301 wide band amplifier 30V pk-pk) was used to amplify the voltage signals if necessary and the signal was monitored by an oscilloscope (Hewlett Packard, 54600B oscilloscope 100 MHz). The electrical signals were then connected to the device electrodes through a relay (Hamlin, HE3621A0510) where a LABVIEW program was written to control the switching of the relay via a computer card (National Instruments, PCI 6723).

III. RESULTS AND DISCUSSION

A. Asymmetrical electrode array for pumping

Reversal pumping has been reported by several authors.^{11–16} Here, we demonstrate the effect of voltage and frequency on pumping direction with an asymmetrical electrode array. We fabricated and tested a device with the following electrode configuration: width=15 and 5 μm , gaps=15 and 5 μm . Figure 2 presents two typical dominant flow directions, i.e., forward and reversal pumpings. The flow paths of 1 μm microbeads at 120 kHz are shown to have a net forward pumping direction at 10 V_{pp} in Fig. 2(a) and a net reversal pumping direction at 20 V_{pp} in Fig. 2(b).

To better understand this phenomenon, knowledge of the origin of the vortices, the dominant flow direction, and the effect of the size of vortices at various voltage-frequency conditions is essential. In this paper we focus on addressing the effects of the ac voltage level and the verification of charge injection from Faradaic reactions and also the resulting effect on the flow direction in ACEK pumping. The injection of ions into the solution will alter the solution conductivity, thus setting up a conductivity gradient (if any). This could be a reason contributing to this phenomenon.

B. Fluorescein dye characterization and ion detection strategy

Faradaic reactions at high ac voltage have been hypothesized to be accountable for the flow reversal in ac micropumps, although they have yet to be observed directly. We aim here to provide a direct observation of these Faradaic reactions at the electrodes at high ac voltage. One direct approach for the observation of Faradaic reactions is to track the ion production from these hydrolytic reactions by observing the change in pH of the solution above the electrodes. This can be achieved with a pH-dependent fluorescent dye.

Chemical equations of electrolyte with different pH can be represented differently.²⁷ Here, we adopted the general electrolytic reactions for water that are written as²⁸

FIG. 3. Effect of electrolyte pH on fluorescence intensity at a concentration of $10^{-5}M$.

where H^+ ions are produced at the anode and OH^- ions are produced at the cathode. The pH -dependent fluorescein dye we employed could detect the pH changes associated with this ion production due to fluorescence intensity change. This dye exhibits low fluorescence intensity in an acidic environment and high fluorescence intensity in a basic environment as depicted in Fig. 3. The increase in H^+ ions at the anodic electrode protonates fluorescein molecules and causes a decrease in fluorescence intensity at the anodic side of the channel. In contrast, the increase in OH^- ions at the cathode deprotonates fluorescein, causing an increase in fluorescence intensity at the cathodic side of the channel.

To avoid diffusion and neutralization of ions over time from the cathodic side to the anodic side or vice versa and to have delimited areas of ions, the fluorescein solution was continuously pumped through the microchannel at a relatively low flow rate of $1 \mu l/min$. With a flowing stream, there will be a negligible effect caused by diffusion and the intensity profile downstream of the electrodes will reach steady state and could be more distinguishably detected.

As the ion production could be small (depending on the voltage condition) and near the electrode surface, the ion production may not cause an intensity change throughout the entire depth of the microchannel for a deep channel. For ease of detection, we employed a microchannel of $45 \mu m$ depth. Similarly, for ease of detection, a sufficiently large electrode gap with operation conducted at sufficiently high ac voltage and low frequency was required to generate sufficient intensity change for detection. However, the ac voltage employed could not be too high in order to avoid electrolysis with gas formation caused by a high electric field. As such, we employed a larger electrode gap of $80 \mu m$, as compared to the asymmetrical micropump with electrode gaps of 15 and $5 \mu m$. This strategy on ion detection by pH tracing was briefly reported without arriving at a definite conclusion on Faradaic charging in our previous work.^{18,19}

FIG. 4. Experimental images and 3D intensity plot of fluorescein dye (pH 6.3) flowing in the microchannel without and with dc voltage applied: (a) no voltage, (b) +2 V, and (c) -2 V. (d) Fluorescence intensity across the microchannel width. (e) Distribution of fluorescence intensities in the area of evaluation.

C. Detection of H^+/OH^- production under dc voltage

To prove our proposed strategy of ion detection by observing pH changes, we first conducted experiments with only dc voltage. Fluorescein solution at pH 6.3 was continuously pumped through the microchannel at a flow rate of $1 \mu l/min$, the results are depicted in Fig. 4. Without voltage applied to the electrodes, a homogeneous fluorescence intensity was observed in the microchannel as shown in Fig. 4(a). The 3D intensity plot exhibits higher intensity in the electrode region due to the amplification of the fluorescence intensity by ~ 1.6 -fold by the gold electrodes. Although amplification of the observed intensity by the electrodes could aid detection at low ion production, the amplification was only over the electrodes and not right across the channel. As such, all the analyses of the intensity and its distribution were performed outside the electrode region at $5 \mu m$ downstream of the electrodes with an evaluation area of $400 \mu m$ (width) by $420 \mu m$ (length).

With +2 V voltage as shown in Fig. 4(b), the right electrode (anode) exhibited a decrease in fluorescence intensity. This indicates a protonation of the fluorescein by H^+ ions which were produced in the hydrolytic reaction ($2H_2O \rightarrow 4H^+ + O_2 + 4e^-$). The fluorescence intensity plot in-

FIG. 5. Experimental images and 3D intensity plot of fluorescein dye ($pH\ 3.8$, $pH\ 10$) flowing in the microchannel without and with dc voltage applied: (a) no voltage, (b) +2 V, and (c) -2 V. (d) Fluorescence intensity across the microchannel width. (e) Distribution of fluorescence intensities in the area of evaluation.

indicates a drop from an arbitrary value of ~ 85 to ~ 30 on the right electrode (anode) as shown in Fig. 4(d). As for the left electrode (cathode), OH^- ions were produced ($4\text{H}_2\text{O} + 4e^- \rightarrow 4\text{OH}^- + 2\text{H}_2$). However, its production would not result in a higher intensity due to the pH of the fluorescein solution employed (i.e., $pH\ 6.3$). Figure 4(e) indicates that the fluorescence intensity distribution shifted from a single tall peak at high intensities (80–95) when no voltage was applied to a flatter peak (60–90) and a tall peak at lower intensities (25–35) when +2 V was applied. This tall peak at lower intensities verifies the production of H^+ ions. Similarly, as we switched the electrode polarity as shown in Fig. 4(c) by applying -2 V, H^+ ions were similarly detected at the left electrode (anode) and had similar intensity and distribution plots as expected.

Due to the choice of the initial pH of the fluorescein solution the production of OH^- ions at the cathode was not convincingly proven as the low intensity changes could not be detected. To avoid this difficulty, two streams of fluorescein solution with different pH , namely, $pH\ 3.8$ and 10 for the right and left streams, respectively, were injected into the microchannel as shown in Fig. 5(a). With an applied dc voltage of +2 V shown in Fig. 5(b), the acidic stream ($pH\ 3.8$) on the right electrode (anode) exhibited even lower intensity, indicating the production of H^+ ions which

protonated the fluorescein dye further. However, the production of OH^- ions on the left electrode could not be convincingly detected due to the insensitivity of the $p\text{H}$ 10 stream to the production of OH^- ions. As we switched the electrode polarity to -2 V as shown in Fig. 5(c), the acidic stream ($p\text{H}$ 3.8) on the right electrode (cathode) exhibited higher intensity, indicating clearly the production of OH^- ions which deprotonated the fluorescein at the cathodic side of the channel. In addition, the $p\text{H}$ 10 stream on the left electrode (anode) had a significant reduction in intensity, indicating the production of H^+ ions which protonated the fluorescein dye. This indicates the production of H^+ and OH^- ions simultaneously at the anode and cathode, respectively. Thus, the proposed strategy is inclined toward detecting the production of H^+ ions at mid- and high values of $p\text{H}$ and the production of OH^- ions at low $p\text{H}$. The results obtained were summarized in Figs. 5(d) and 5(e).

D. Detection of H^+/OH^- production under ac voltage

Here we speculate that with a sufficiently high ac voltage level due to Faradaic reactions both H^+ and OH^- ions will be produced at the same electrodes (if any) at different parts of the cycle. These cyclic alternations of coion production are difficult to detect as H^+ and OH^- ions will tend to neutralize each other, and also the high frequency of the alternating voltage allows only a small quantity to be produced.

For ease of detection, we exploit the fact that with a properly chosen $p\text{H}$ ($p\text{H}$ 6.3) for the fluorescein solution, a fluorescence intensity change due to the small production of H^+ ions is detectable but not the small production of OH^- ions. As such, with any intensity change detected, it can be concluded that H^+ ions are produced. With the aperture of the camera opened (exposure time = 50 ms) over many cycles of the alternating voltage, the cumulative intensity change of the fluorescein solution over these cycles would be detected, thus avoiding the complications caused by the high frequency of the applied ac voltage.

Figure 6 shows the experimental images and 3D intensity plot from low to high ac voltages at 100 Hz with the fluorescein solution at $p\text{H}$ 6.3 continuously pumped through the microchannel at a flow rate of $1 \mu\text{l}/\text{min}$. With no voltage applied to the electrodes, a homogeneous fluorescence intensity was observed in the microchannel. With a low ac voltage of 8 V_{pp} , the production of H^+ ions was not obvious. As the ac voltage increased to 12 V_{pp} , the center of the microchannel exhibited a drop in intensity, indicating the production of H^+ ions. The cyclic nature of ac voltage resulted in the production of H^+ ions from both electrodes. The electrical field was stronger at the edges and thus the intensity changes were more dominantly observed at the center of the microchannel. The fluorescence intensity plot indicates a drop from a high arbitrary value of ~ 85 (no voltage) to lower values (~ 65 at 12 V_{pp} , ~ 38 at 18 V_{pp}) at the center of the microchannel, as shown in Fig. 6(f). In addition, the fluorescence intensity distribution shifted from a tall single peak at high intensities (80–95) to a flatter distribution toward the lower intensities, as shown in Fig. 6(g). Both plots indicate the detection of H^+ ion production and this production increased with an increase in ac voltage.

Although the experiments did not explicitly detect the production of OH^- ions, it is reasonable to expect that the production of OH^- ions would follow the same trend as the production of H^+ ions.

For quantification of the effect of ac voltage on the production of ions at the electrodes, we measured the deviation of the fluorescence intensities at different ac voltage levels. A coefficient of deviation is defined as

$$\varepsilon_{\text{dev}} = \frac{1}{N} \frac{\sum \sum |I_{ij} - \bar{I}|}{\bar{I}}, \quad (1)$$

where I_{ij} is the observed intensity, $\bar{I}$ is the mean intensity from a reference image with no voltage, and N is the number of pixels in the area of evaluation (outside the electrode region). With this definition, a high value of ε_{dev} indicates greater deviation and thus greater ion production due to

FIG. 6. Experimental images and 3D intensity plot of fluorescein dye (pH 6.3) flowing in the microchannel without and with ac voltage applied at 100 Hz: (a) no voltage, (b) 8 V_{pp}, (c) 12 V_{pp}, (d) 16 V_{pp}, and (e) 18 V_{pp}. (f) Fluorescence intensity across the microchannel width. (g) Distribution of fluorescence intensities in the area of evaluation.

Faradaic reactions. A MATLAB program was written to analyze the intensities from the image pixels and compute the coefficient of deviation.

Figure 7 shows the coefficient of deviation as a function of ac voltage at 100 Hz for fluorescein solution (pH 6.3) flowing at 1 $\mu\text{l}/\text{min}$ in the microchannel. With ac voltage below 10 V_{pp}, the coefficient of deviation was less than 0.052. At an ac voltage of 11 V_{pp}, the coefficient of deviation increased significantly to a value of 0.091. This indicates that above this voltage for the given frequency and electrode configuration, the production of ions was evident. As the ac voltage increased, the coefficient of deviation increased, with a value of more than 0.25 for an ac voltage above 18 V_{pp}. These results verify the production of H^+/OH^- and Faradaic charging effects can be expected during high voltage ACEK experiments. Note that the voltage for the onset of Faradaic reactions for different experimental conditions, such as different electrode widths/gaps, microchannel depths, and solution conductivities, may differ from the values reported here.

E. dc-biased ac electrokinetics in pumping application

We exemplify here how charge injection from incipient hydrolytic reactions can be employed to control the magnitude and direction of a net ACEK flow. Figure 8(a) shows the schematic of a typical ACEK micropump consisting of pairs of asymmetrical electrodes. As shown in Figs. 8(b) and 8(c), microbeads were used as tracers of flow direction and for the estimation of the flow velocity. Typically, this type of geometry has been used in capacitive pumping conditions (10 V_{pp}, 10 kHz) in asymmetric electrodes to obtain a net flow from the big electrode to the small electrode (estimated average net velocity of 450 $\mu\text{m}/\text{s}$) as shown in Fig. 8(b). Reversal pumping in similar electrode geometry has been reported in literature at high ac voltage and a high frequency (17 V_{pp}, 50 kHz).¹² However, we show in Fig. 8(c) that reversal pumping can be achieved by

FIG. 7. Coefficient of deviation of fluorescent intensity signal as a function of ac voltage at 100 Hz for fluorescein solution (pH 6.3) flowing at 1 $\mu\text{l}/\text{min}$.

superimposing a dc bias to the ac signal (i.e., 10 V_{pp}, 10 kHz, −1 V, estimated average net velocity of −1220 $\mu\text{m}/\text{s}$). Thus, a reversal net flow which is ~ 2.7 times higher than in the original flow direction was achieved by the application of a small negative dc voltage of −1 V. This effect is attributed to the injection of additional charges and the generation of charge gradients. The dc bias here constantly injects charges into the solution resulting in a stronger net flow and change in flow direction. This observation illustrates the potential of charge injection from Faradaic reactions in the control of flow direction and flow velocity. The small dc-biased voltage required is significant as it is a desirable attribute for the implementation of a controllable pumping strategy in magnitude and directional controls of fluid flow in a fully integrated microfluidic device.

IV. CONCLUSIONS

In conclusion, we have demonstrated the production of H^+/OH^- from incipient hydrolytic reactions and thus a direct observation of Faradaic charging in ACEK. In experiments where a symmetrical pair of electrodes (width=160 μm , gap=40 μm) and a microchannel (width=400 μm , depth=45 μm) were employed, the change in fluorescence intensity of a fluorescein solution with pH was used as an indication of ion production in dc and ac conditions. The results indicate that H^+/OH^- production from hydrolytic reactions was evident for an ac voltage level above 11 V_{pp}. Our findings directly validated the existence of Faradaic reactions at high ac voltage in ACEK.

Lastly, we demonstrated that charge injection arising from the application of a small dc bias voltage can be employed to control the magnitude and direction of ACEK flow in micropumping.

FIG. 8. (a) 3D schematic of dc-biased ac electrokinetic micropump. A superimposed image of the microbead flow path in (b) capacitive pumping, 10 V_{pp} at 10 kHz and (c) dc Faradaic pumping, 10 V_{pp} at 10 kHz, -1 V. Electrode configuration: widths=15 and 5 μm , gaps=15 and 5 μm .

ACKNOWLEDGMENTS

This research work was performed at the Institute of Materials Research and Engineering, Agency for Science, Technology and Research (A*STAR). This work is also supported by A*STAR SERC Grant No. 052 101 0013.

- ¹ H.-C. Chang, *Can. J. Chem. Eng.* **84**, 146 (2006).
- ² H.-C. Chang and G. Yossifon, *Biomicrofluidics* **3**, 012001 (2009).
- ³ J. Wu, Y. Ben, and H.-C. Chang, *Microfluid. Nanofluid.* **1**, 161 (2005).
- ⁴ J.-R. Du, Y.-J. Juang, J.-T. Wu, and H.-H. Wei, *Biomicrofluidics* **2**, 044103 (2008).
- ⁵ N. Lewpiriyawong, C. Yang, and Y. C. Lam, *Biomicrofluidics* **2**, 034105 (2008).
- ⁶ C. H. Kua, Y. C. Lam, I. Rodríguez, C. Yang, and K. Youcef-Toumi, *Anal. Chem.* **79**, 6975 (2007).
- ⁷ A. B. D. Brown, C. G. Smith, and A. R. Rennie, *Phys. Rev. E* **63**, 016305 (2000).
- ⁸ M. Mpholo, C. G. Smith, and A. B. D. Brown, *Sens. Actuators B* **92**, 262 (2003).
- ⁹ S. Debesset, C. J. Hayden, C. Dalton, J. C. T. Eijkel, and A. Manz, *Lab Chip* **4**, 396 (2004).
- ¹⁰ J. Wu, *J. Appl. Phys.* **103**, 024907 (2008).
- ¹¹ D. Lastochkin, R. Zhou, P. Wang, Y. Ben, and H.-C. Chang, *J. Appl. Phys.* **96**, 1730 (2004).
- ¹² V. Studer, A. Pepin, Y. Chen, and A. Ajdari, *Analyst (Lond.)* **129**, 944 (2004).
- ¹³ A. Ramos, H. Morgan, N. G. Green, A. Gonzalez, and A. Castellanos, *J. Appl. Phys.* **97**, 084906 (2005).
- ¹⁴ J. P. Urbanski, T. Thorsen, J. A. Levitan, and M. Z. Bazant, *Appl. Phys. Lett.* **89**, 143508 (2006).
- ¹⁵ P. Garcia-Sanchez, A. Ramos, N. G. Green, and H. Morgan, *Langmuir* **24**, 9361 (2008).
- ¹⁶ K. Yang and J. Wu, *Biomicrofluidics* **2**, 024101 (2008).
- ¹⁷ N. Sasaki, T. Kitamori, and H. B. Kim, *Lab Chip* **6**, 550 (2006).
- ¹⁸ W. Y. Ng, S. Goh, Y. C. Lam, C. Yang, and I. Rodríguez, *Lab Chip* **9**, 802 (2009).
- ¹⁹ W. Y. Ng, Y. C. Lam, and I. Rodríguez, Proceedings of the Conference on Advances in Microfluidics and Nanofluidics, Hong Kong, 2009 (unpublished), Paper No. E7-1.
- ²⁰ A. Ramos, H. Morgan, N. G. Green, and A. Castellanos, *J. Phys. D* **31**, 2338 (1998).
- ²¹ A. Ajdari, *Phys. Rev. E* **61**, R45 (2000).
- ²² L. H. Olesen, H. Bruus, and A. Ajdari, *Phys. Rev. E* **73**, 056313 (2006).
- ²³ A. Ramos, A. Gonzalez, P. Garcia-Sanchez, and A. Castellanos, *J. Colloid Interface Sci.* **309**, 323 (2007).
- ²⁴ D. R. Lide, *CRC Handbook of Chemistry and Physics*, 88th ed. (CRC, Boca Raton/Taylor & Francis, New York, 2008), pp. 8–18.
- ²⁵ S. Fiedler, R. Hagedorn, T. Schnelle, E. Richter, B. Wagner, and G. Fuhr, *Anal. Chem.* **67**, 820 (1995).
- ²⁶ J. C. McDonald and G. M. Whitesides, *Acc. Chem. Res.* **35**, 491 (2002).
- ²⁷ M. Trau, D. A. Saville, and I. A. Aksay, *Langmuir* **13**, 6375 (1997).
- ²⁸ R. G. H. Lammertink, S. Schlautmann, G. A. J. Besselink, and R. B. M. Schasfoort, *Anal. Chem.* **76**, 3018 (2004).