ac electroosmotic pumping induced by noncontact external electrodes

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(Received 2 August 2007; accepted 22 August 2007; published online 21 September 2007)

Electroosmotic (EO) pumps based on dc electroosmosis is plagued by bubble generation and other electrochemical reactions at the electrodes at voltages beyond 1 V for electrolytes. These disadvantages limit their throughput and offset their portability advantage over mechanical syringe or pneumatic pumps. ac electroosmotic pumps at high frequency (>100 kHz) circumvent the bubble problem by inducing polarization and slip velocity on embedded electrodes,¹ but they require complex electrode designs to produce a net flow. We report a new high-throughput ac EO pump design based on induced-polarization on the entire channel surface instead of just on the electrodes. Like dc EO pumps, our pump electrodes are outside of the load section and form a cm-long pump unit consisting of three circular reservoirs (3 mm in diameter) connected by a 1×1 mm channel. The field-induced polarization can produce an effective Zeta potential exceeding 1 V and an ac slip velocity estimated as 1 mm/sec or higher, both one order of magnitude higher than earlier dc and ac pumps, giving rise to a maximum throughput of $1 \mu\text{l/sec}$. Polarization over the entire channel surface, quadratic scaling with respect to the field and high voltage at high frequency without electrode bubble generation are the reasons why the current pump is superior to earlier dc and ac EO pumps. © 2007 American Institute of Physics. [DOI: [10.1063/1.2784137](https://doi.org/10.1063/1.2784137)]

I. INTRODUCTION

Recently miniaturized total analysis system devices to implement chemical and biochemical assays have attracted significant attention because these devices are portable, consume considerably less amount of samples and reagents and reduce manual intervention. In the miniature devices, embedded mechanical micropumps with moving parts are extremely undesirable because micromechanical units are prone to frictional wear. The fabrication of miniaturized mechanical pumps also requires sophisticated and expensive processes. Larger external mechanical pumps such as peristaltic pumps and syringe pumps are, on the other hand, extremely bulky and hence defeat the purpose of miniaturization. In contrast, electrokinetic pumps without moving parts, which only require strategically placed electrodes, are inexpensive to produce and are straightforward to integrate into microdevices. Several types of electrokinetic pumps using dc voltages have been developed to drive fluids in miniaturized devices.^{2–4} Although some of these electrokinetic pumps are able to produce high pressures and are particularly useful for packed microchannels in HPLC,⁵ dc pumps tend to generate bubbles at the electrodes even at very low voltage (<1 V). This distinct disadvantage limits their throughput and offsets their portability and wear-free advantages over mechanical syringe or pneumatic pumps.

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Alternate current (ac) electrokinetic pumping with asymmetric embedded electrodes that operate at high frequency (>100 kHz) to circumvent the bubble problem has been demonstrated.¹ However, these microelectrodes within the microchannel require elaborate fabrication techniques to produce net flows and can contaminate the sample.

Using electrokinetics to pump fluid in microchannels requires substantial Smoluchowski slip velocity on the conduit walls, which is proportional to the thickness of the double layer with ion polarization, to generate electroosmotic flow. In a capillary wall with charged functionalities such as hydrolyzed fused silica, this double layer, with excess counter ions and depleted co-ions, has a thickness from 10 to 100 nm. The double layer screens the field of the charged functionality to produce a potential drop of about ~ 0.1 V, termed the Zeta potential. When a charged surface is on a conducting substrate such as ion-exchange resin, the depletion of co-ions is enhanced when the conducting material attracts field lines and the normal component of the external field drives the co-ion into the bulk. Under a moderate electric field (100 V/cm) this depletion zone creates an over-potential (effective Zeta potential) of 1 V or greater.⁶ The thickness of this layer with field-induced polarization is therefore extended to 1 μm to generate fast Smoluchowski slip velocity on the resin surfaces. This fast slip velocity is measured to be as high as several cm/s at a moderate field. This mechanism of electroosmotic flow around a granule is first proposed by Dukhin⁷ and has also been confirmed with visualization and theoretical studies.^{8–10} Our group has used fast electroosmotic flows on the ion exchange resin surfaces to generate microvortices to perform effective mixing in miniaturized reservoirs.¹¹

Such a co-ion depletion layer can also be formed at a dielectric surface to create an enhanced overpotential when the induced polarization is induced with a high frequency ac field. When dielectric polarization is induced by the normal component of the external electric field, the induced surface field produces an electrical double layer. Because the double layer is essentially charged like a capacitor, this particular charging mechanism is typically referred to as capacitive charging. Such capacitive charging has been studied for double layers on metal electrodes where the charging field is applied by the electrode,^{12,13} but there exist few studies on capacitive charging of dielectric surfaces by an external field except for the dc study by Thamida and Chang.¹⁴

The advantage of ac charging of dielectric surfaces is that the electrodes can be housed in external reservoirs and, with a sufficiently high frequency, bubble generation can be minimized at these electrodes. More importantly, the electrodes are not in contact with the working sample and hence contamination is avoided. Although the polarity of ac field flips each half cycle, the Smoluchowski slip velocity on the surface still moves toward the same direction because the polarity of induced charge reverses as well. However, this induced charge polarization would cease at low frequencies when the induced charges in the dielectric double layer have time to diffuse back into the bulk. Therefore, a high frequency ac field is required to maintain the polarization of induced charge. On the other hand, the half cycle of ac field has to be long enough to pump counter ions to the polarized surface. One hence expects an optimum frequency for the fastest slip velocity that corresponds to the inverse RC time of the double layer, when the double layer capacitor is charged but not excessively by the distributed Ohmic resistors of the bulk fluid.

Like the flow field close to the conducting surface containing ion exchange functionalities, the electroosmotic flows are no longer aligned with electric field lines and are highly nonuniform. In particular, when the dielectric surface has a curvature, field leakage across the dielectric medium is enhanced. At the wedgelike corners of central reservoir, where the side channels are joined, the enhanced corner electric field lines allow deep penetration through the dielectric substrate. The overpotentials at the two walls of a corner are in opposite polarities. Therefore, the electroosmotic flows along both surfaces move toward the corner tip to produce a microjet.¹⁴ But the geometry at the corner is not symmetric. The curvature at the reservoir edge is larger than that at the channel edge. Since reservoir curvatures should hence promote the formation and efficiency of microjets, these streams will drive the liquid from these two channels either outward or inward the center reservoir, which works as a micropump.

In this paper, we demonstrate efficient pumping in a 1×1 mm conduit using ac electroosmosis due to field-induced polarization. Our pumping device, of that electrodes are outside of the

FIG. 1. Schematic illustration of microfluidic mixing device using induced ac electroosmotic vortex flows on the dielectric channel. A. Side reservoirs; B. Central reservoir; C. Connection channels; D. Electrodes.

pumping unit, consists of three circular reservoirs (3 mm in diameter) connected by a 1×1 mm channel. Close to the center reservoir corners, this device provides efficient pumping to drive solutes at a speed approximately 1 mm/sec. We also study the effects of ac field frequency and field strength on pumping efficiency.

II. EXPERIMENTAL

The fabrication procedures of our microfluidic reaction device are the same as those in our previous report.¹¹ Copolyester plastic sheets were obtained from DSM Engineering Plastic Products (Sheffield, MA, USA). The dielectric constant of copolyester plastics is about 2.4. In order to sufficiently release the bubbles generated at the electrodes when the applied ac field is of low frequency, the anode at the left and the cathode at the right were housed in two side-reservoirs as shown in Fig. 1. These side-reservoirs were connected by two straight channels to a central reservoir between them. All three reservoirs were 3 mm in diameter and were drilled on one thermoplastic slide (20×40 mm; 1 mm in thickness). The connecting channels were 1.0 mm in width and each segment between two reservoirs was 10 mm long. The holes and grooves were machined on one plastic slide with conventional drilling and milling tools. The substrate slide was then thermally bonded to the drilled one at 80 to 100 °C. Pressure was briefly applied using binder clips when the slides were glued together. Electrodes of platinum wires were placed in the two side-reservoirs with their tips bent to dip into the reservoirs to contact the liquids and the remaining part taped on the slides. The electrodes were connected to a high voltage output transformer (Industrial Test Equipment) and rf amplifier (Powertron 250 A, 10 Hz–1 MHz), capable of delivering up to 6000 V (peak to peak) and 250 W; ac frequencies were produced by a function/arbitrary waveform generator (Hewlett-Packard 33220A).

One drop of organic solvent glycerin stained with food-color dye was held in pipette tips to add in the reservoir when the tips were dipped. The images of pumping process were recorded with a video camera when the applied field was activated. The images were digitized and transferred into a personal computer to measure the advancement of dyes inside the channel.

III. RESULTS AND DISCUSSION

Figure 2 shows the lateral dye motion toward both side reservoirs via the induced electroosmotic pumping action with an ac sine wave at 120 kHz. (The field across the two electrodes is 133 Vrms/cm.) The speed of the two advancing fronts of dyes is approximately 1.0 mm/sec, one order of magnitude higher than earlier dc and ac pumps of comparable field strength, giving rise to a maximum throughput of 1 μ l/sec. We estimate the effective overpotential to be as large as 1 V at these conditions.

FIG. 2. Liquid motion using induced ac electroosmotic flow produced by applying a sine waveform ac field ($133 \text{ V}_{\text{rms}}/\text{cm}$; 120 kHz) along a $1 \times 1 \text{ mm}$ channel with a 10 mm length. Glycerin solution stained with food-color dyes dipped in the center reservoir is driven toward the side reservoir at the speed of approximately 1 mm/sec .

FIG. 3. (a) Measured pumping speed as a function of ac frequency for three different applied fields (133, 100, and $50 \text{ V}_{\text{rms}}/\text{cm}$). The upper and lower curve in Fig. 3(a) depict the trends of pumping speed as a function of ac frequency for three different applied 133 and $100 \text{ V}_{\text{rms}}/\text{cm}$ respectively. The maximum pumping speed appears at 120 kHz . (b) the insert frame showing a linear log-log relation of pumping speed dependence on field strength at 120 kHz ac field. The slope of this linear curve is 2.00. The R^2 value of regressed curve is 0.927.

The expected optimum frequency for the slip velocity, roughly corresponding to the inverse RC time of the double layer on the dielectric, is also confirmed by the measured dye velocity. Figure 3 shows the pumping speed dependence on the applied field frequency. Curves in Fig. 3 all have a maximum pumping speed at around 120 kHz.

This optimum frequency for slip velocity is consistent with that measured on electrodes for dilute electrolyte solutions.¹⁵ The co-ion depletion layer of induced ac electroosmosis is created when the surface dielectrics is polarized by an external field. Therefore, like the polarization on the surface of a conducting granule, the induced polarization for ac electroosmosis on dielectric also scales linearly with the applied field strength. Since the Smolouchowski slip velocity is due to the Maxwell force that is the product of the polarization and the applied field, the effective velocity of induced ac electroosmosis should scale quadratically to the applied field strength. Using the data in Fig. 3 to perform regression, as shown in the insert frame, the log-log relation of maximum pumping speed at 120 kHz versus the applied field strength approximately yields a straight line of power of two. This quadratic scaling relation confirms the quadratic nature of induced electroosmotic flow.

Theoretically, the pumping rate can be much higher with larger voltage. However, the plastic substrates cannot resist an applied voltage beyond 1 kV.

IV. CONCLUSIONS

Pumping speed one order of magnitude higher than that by previous dc or ac electroosmotic pumps has been achieved in small channels with and induced ac electroosmotic flow at the corners of dielectric substrates. These flows resulted from fluid slips driven by nonuniform polarization of dielectric wedges inserted in a high frequency ac electric field (50–133 Vrms/cm; 50–140 kHz) to avoid generating bubbles on the electrodes. The applicability of this unique pumping mechanism giving rise to a throughput of 1 μ l/sec was demonstrated under moderate electric fields and without internal electrodes, flow pumping by any mechanical compressor with moving parts.

In addition, since the maximum pumping efficiency of this device is achieved at very high ac frequency 120 kHz, bubble generation at the electrodes is no longer problematic when this device is in enclosed format. Such a pumping speed of 1 mm/sec is still not able to create enough pressure to push liquid out off the channel end when the device is modified to a closed format. But since the pumping speed increases quadratically as the field strength increases, high pressure pumping is expected around 2 kV/cm. Although this device made of plastic substrates cannot resist high voltages, other dielectric materials such as glass or quartz are alternative substrates to solve this problem. In addition, machining the same reservoir and channel geometries on these materials should be straightforward fabrication effort. Electrodes can be fabricated by depositing conductive materials on the ends of the channel when the device is modified to enclosed format. Unlike the previous ac pumping device using polarization on electrodes, this novel device employs induced electroosmosis without the presence of internal electrodes. Therefore, contaminating reactions do not occur inside the pumping device.

ACKNOWLEDGMENTS

The financial support by National Science Council, Taiwan (NSC 94-2113-M-194-015) and National Chung Cheng University is acknowledged.

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