

FREQUENCY DEPENDENT AC ELECTROOSMOTIC FLOW IN NANOCHANNELS

Wesley T.E. van den Beld¹, Wouter Sparreboom¹, Albert van den Berg¹, and Jan C.T. Eijkel¹

¹BIOS Lab-on-a-Chip group, MESA+ Institute for Nanotechnology, Twente University, the Netherlands

ABSTRACT

We report frequency-dependent bidirectional AC electroosmotic flow (AC-EOF) in a nanochannel with double layer overlap. This work follows our report in μ Tas 2008 of unidirectional AC-EOF in nanochannels [1]. Observed is a bidirectional pumping behavior; simulations of the low frequency pumping confirm a direction opposite to that of AC-EOF in microchannels. By this frequency-dependent bidirectional pumping, nanochannel AC-EOF behaves in fundamentally different way than microchannel AC-EOF. Generally, the results are of importance for the understanding of ion and liquid transport in nanoconfinement.

INTRODUCTION

The pumping mechanism of AC-EOF in microchannels has been investigated in several studies; main advantages of using AC-EOF compared to traditional DC-EOF are the use of low voltages ($\sim 1V$) and no need for external electrodes. The operational behavior in microchannels has been modeled theoretically and confirmed experimentally in other studies [4,5]. At present, AC-EOF has not been investigated in nanochannels, neither in experiment or simulation apart from our previous report. Here we present novel frequency dependent measurements in combination with simulation results.

EXPERIMENTAL

Glass chips with 50 nm high nanochannels were fabricated using cleanroom technology and sacrificial layer etching of chromium [2,3]. An asymmetric gold electrode array of 100 electrode pairs was integrated along the channel axis with dimensions as shown in Figure 1.

The fabricated device is displayed in Figure 3. Channels were filled with a 100 μ M KNO₃ solution, leading to a Debye length of 30 nm. This concentration is chosen to have overlapping double layers in the nanochannel. A PDMS layer with microchannels is attached to the chip to allow solution exchange. The measurement setup is schematically shown in Figure 2. Measurements were performed by first filling the nanochannels with 100 μ M solution and subsequently filling the connecting microchannels in the PDMS layer on one side with 50 μ M and on the other side with 200 μ M solution. Then a sinusoidal signal with an amplitude of 400 mV is applied to the electrode pairs using an impedance analyzer, with the large and small electrodes 180 degrees out of phase. The time-dependent increase or decrease of impedance then indicates pumping speed and direction.

SIMULATION

Simulations were performed in COMSOL Multiphysics 4.3a. The simulated 2D-geometry consists of the nanochannel with two pairs of electrodes with dimensions of geometry A as made clear in Figure 1. For the oxide surfaces a capacitance of 1 F/m² and for the metal surfaces 0.2 F/m² is modeled. The boundary condition for the oxide is a constant potential assumption of -0.09 V. At the two ends of this channel two microreservoirs are added to take into account the entrance effects. The behavior is mathematically described by three sets of equations:

- The Nernst-Planck equation is used to describe ionic transport.
- The Poisson equation is included to calculate the electric potential.
- Stokes flow is describing the fluid flow.

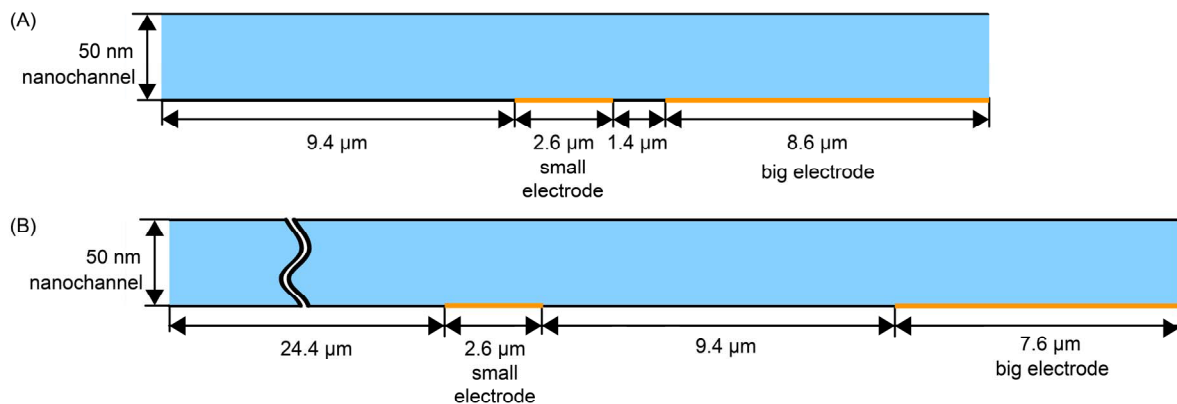


Figure 1: A cross-sectional schematic drawing with both tested geometries showing one repetitive electrode pair of the array.

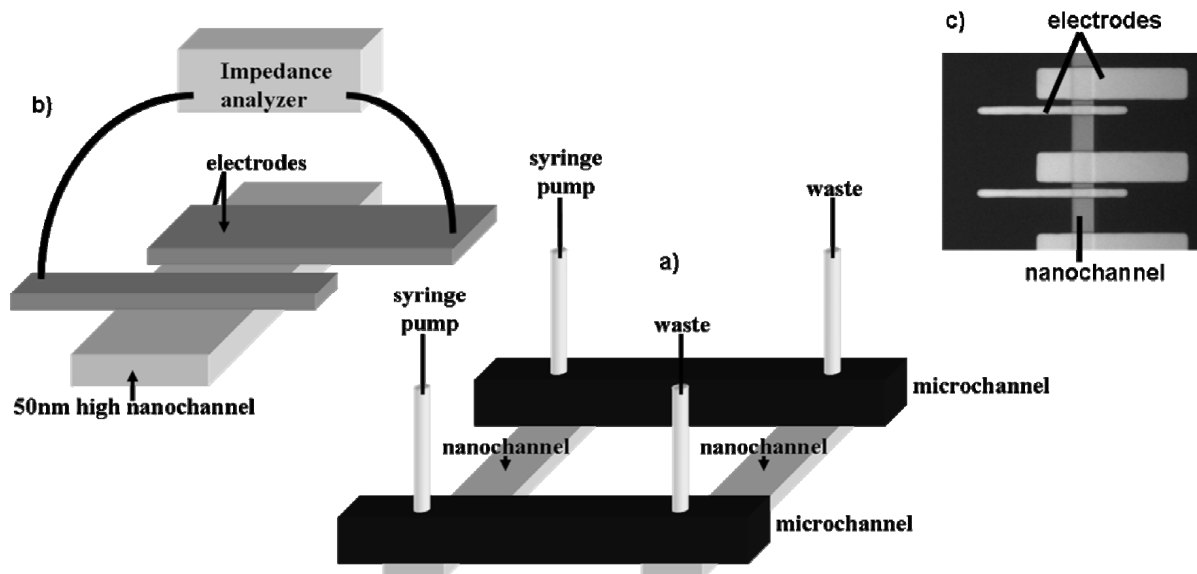


Figure 2: Schematic overview of the measurement setup. The nanochannels are interfaced by two microchannels, each connected to a syringe pump injecting liquids with different conductivities (a). The electrodes are connected to an impedance analyzer for actuation and measuring of the pumping velocity in the nanochannel (b). Photomicrograph of the asymmetric electrodes array situated in the nanochannel (c). [2]

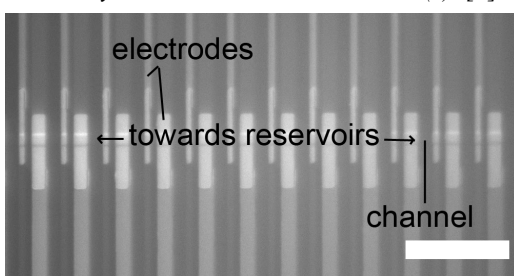


Figure 3: Top-view micrograph of the fabricated device with nanochannel and asymmetric electrode array, the scale bar is $50\ \mu\text{m}$ [1].

RESULTS - EXPERIMENTS

Bidirectional flow is observed in the measurements when changing the applied electrode frequency; results of simulations and measurements are shown in Figure 4. Error bars in the measurements are shown, when multiple experiments were performed at these frequencies. Experiments and simulations show identical flow directions for low frequency actuation, though the magnitude of the simulated flow for geometry A is about threefold lower than the measured flow. In the experiments the flow changes direction at 200 Hz for geometry A and around 30 Hz for geometry B. Lastly, the observed maximum flow speed of $8\ \mu\text{m/s}$ is comparable to the flow speed observed in microchannels at the same applied voltage [4].

Temperature influence on the system can be safely neglected: calculations indicate that the generated power ($0.5\ \mu\text{W}$) after a typical experiment duration of 1000 s only causes a temperature increase of 4 mK in the chip (lumped capacitance $1.2\ \text{J/K}$).

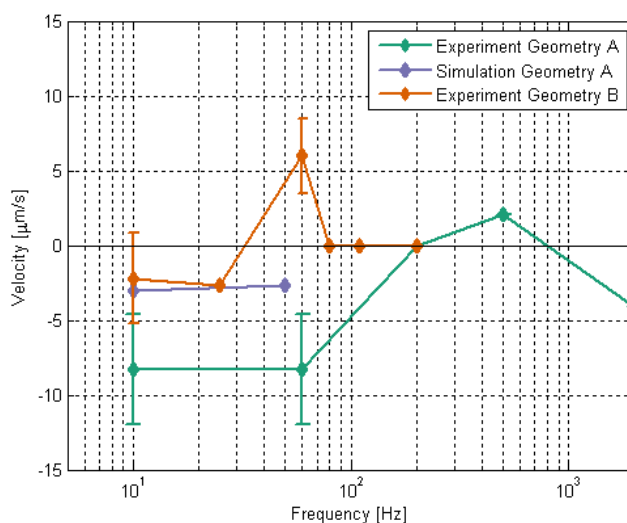


Figure 4: Frequency response of pumping velocity, simulation and measurement [2]. Bidirectional pumping behavior as function of frequency was clearly experimentally observed.

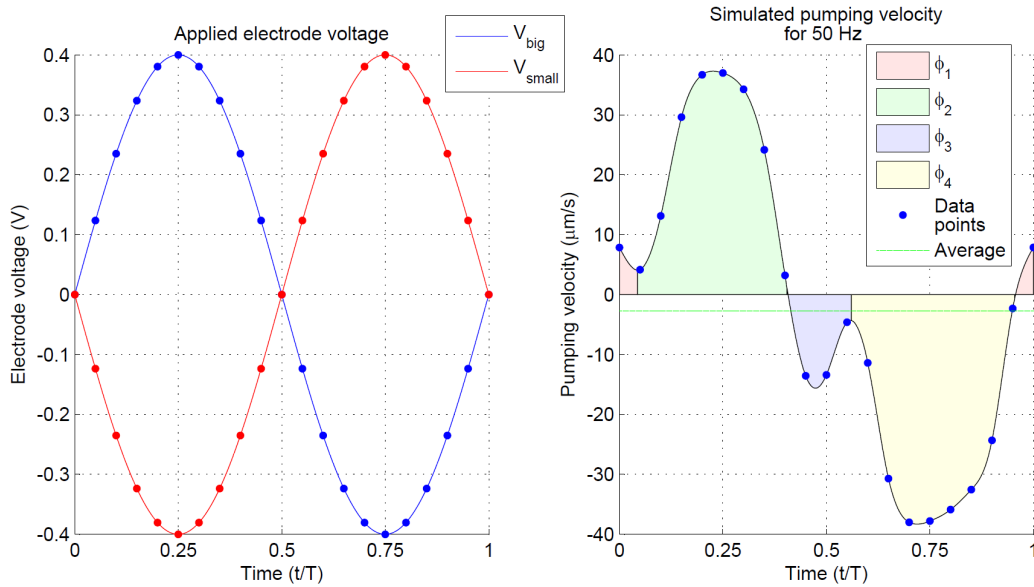


Figure 5. On the left the voltage applied on the big and small electrodes in the array on the bottom of the nanochannel, the simulated pumping velocity on the right for an actuation frequency of 50 Hz. In the pumping behavior four phases ϕ_1 - ϕ_4 can be discriminated.

RESULTS - SIMULATIONS

In nanochannels, the operational principle is significantly different from microchannels. As a result of the large surface-to-volume ratio in nanoconfinement, the surface charge on the walls is dominating the flow behavior. Oxide surfaces deprotonate in contact with an aqueous solution as a function of the acidity. When the salt concentration is chosen such that the negative oxide surface charge cannot be screened, double layers will overlap in the nanochannel. Therefore the nanochannel is mostly filled with positive ions and negative ions are mostly excluded.

In a symmetric electrode configuration no net directional flow can be present in the nanochannel due to the balance in forces. However, in an asymmetric configuration a net directional liquid flow can be generated. When a sinusoidal voltage is applied on the electrodes, they attract or repel ions, involving RC-times related to the geometry and the ion concentrations in the channel. Since ions are accumulated or repelled, charge gradients are induced, leading to electric fields and thus generating coulomb forces. When these forces are aligned a directional flow is obtained.

At low frequencies, simulations indicate the flow direction is largely oscillatory and in-phase with the applied electrode potential. The electrodes are screened for the majority of time for these low frequencies, flow is generated by a type of electroosmotic flow driven by the field in the interelectrode spaces. Simulation results of the pumping velocity versus time actuated with 50 Hz are displayed in Figure 5. Four pumping phases can be distinguished, two small (phase 1 and 3) and two large flow contributions (phase 2 and 4). The overall direction of the flow is determined by the dominant flow, which is caused by the

large charge gradient in the large interelectrode space. Minor differences in this largely oscillatory flow driven by conventional electroosmotic forces lead to a net flow, as will be treated now.

The abundant positive ions are dominating the pumping behavior, negative ions have a negligible influence. Figure 6 reveals the positive ion distribution in the four different phases. The origin of the minor differences is as follows. In the first phase, the positive ions are repelled from the big electrode and injected in the large interelectrode space (LIES) and the small interelectrode space (SIES), generating charge gradients. The largest charge gradient is present in the LIES, however it is counteracted by the smaller gradient in the SIES, leading to a small net flow in this phase.

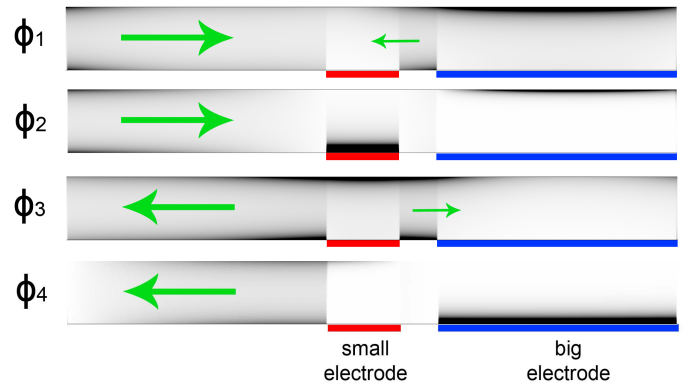


Figure 6. Simulated distribution of the positive ions in the nanochannel in the four phases (dark indicates higher concentration) as well as the flow direction (green arrow). In phase 1 and 3 there is a large gradient, counteracted by a smaller gradient, while in phase 2 and 4 there is a large gradient without a significantly counteracting gradient.

This smaller gradient in the SIES disappears when the small electrode starts to attract the positive ions. Then the pumping velocity starts to increase (phase two). Further screening of the small electrode is causing a stronger gradient in the LIES while the gradient in the SIES disappears, generating a strong flow. This flow will continue until the small electrode starts to repel the previously accumulated positive ions, injecting them in the LIES and the SIES. The consequence is that the charge gradient in the LIES switches direction, and therefore the pumping velocity switches sign.

In the third phase, a large gradient is present in the LIES, counteracted by a gradient in the SIES, similar to phase one. However, in phase three the charge is removed faster from the small electrode, hence generating a rapid gradient, having a larger accumulated influence on the net flow than phase one.

This description holds to the point that the big electrode starts to accumulate the positive ions, inducing a large charge gradient in the LIES and removing the gradient from the SIES (phase four). The mechanism is similar to that of phase two. The difference is that the gradient in the LIES is stronger, since the big electrode attracts more positive ions for screening. When the applied voltage on the big electrode approaches the oxide voltage, it repels the positive ions and injects them into the SIES and LIES. The pumping velocity switches and phase one starts again.

Summarizing, in simulation a largely oscillatory flow has been observed with the same direction as in the experiment. In phases one and three the positive ions are repelled from previously charged electrodes, causing opposing gradients and causing a small net force. In phases two and four, the positive ions are attracted, generating a large gradient and thus a large net force. Due to these subtle differences in charge gradients through the several phases a net flow results. The simulations thus provide an explanation for the pumping behavior of AC-EOF in nanochannels. At present we are adjusting our code to also simulate the high frequency behavior.

CONCLUSIONS

The AC-EOF pumping direction at low frequencies in nanochannels is opposite from microchannels[4]. In microchannels, hydrodynamic coupling causes the bulk liquid to move, generated by the induced electroosmotic forces above the electrodes. In nanochannels the force is mainly generated in interelectrode spaces, similar to traditional DC-EOF. Since an alternating signal is applied, an oscillatory flow results. Because of minor differences in the involved charging processes above small and big electrodes, different charge gradients are induced leading to a net flow.

REFERENCES

- [1] Sparreboom, W. and Cucu, C.F. and Eijkel, J.C.T. and van den Berg, A. (2008) *Ion pumping in nanochannels using an asymmetric electrode array*. In: 12th International Conference on Miniaturized Systems for Chemistry and Life Sciences, 12-16 Oct 2008, San Diego, California.
- [2] Sparreboom, W. (2009). *AC electro-osmosis in nanochannels*. University of Twente. thesis.
- [3] Sparreboom, W., Eijkel, J. C., Bomer, J., & van den Berg, A. (2008). Rapid sacrificial layer etching for the fabrication of nanochannels with integrated metal electrodes. *Lab on a Chip*, 8(3), 402-407.
- [4] Brown, A. B. D., Smith, C. G., & Rennie, A. R. (2000). Pumping of water with ac electric fields applied to asymmetric pairs of microelectrodes. *Physical review E*, 63(1), 016305.
- [5] Ramos, A., Morgan, H., Green, N. G., & Castellanos, A. (1999). AC electric-field-induced fluid flow in microelectrodes. *Journal of colloid and interface science*, 217(2), 420-422.

CONTACT

W.T.E. van den Beld, tel: +31-53-4892154;
w.t.e.vandenbeld@utwente.nl