Experiments on pumping of liquids using arrays of microelectrodes subjected to travelling wave potentials

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Abstract. Net fluid flow of electrolytes driven on an array of microelectrodes subjected to a travelling-wave potential is presented. Two sizes of platinum microelectrodes have been studied. In both arrays, at low voltages the liquid flows according to the prediction given by ac electroosmotic theory. At voltages above a threshold the fluid flow is reversed. Measurements of the electrical current when the microelectrode array is pumping the liquid are also reported. Transient behaviours in both electrical current and fluid velocity have been observed.

1. Introduction
The development of ‘lab-on-a-chip’ devices or micro total analysis systems (μTAS) demands the precise control of fluids in small channels (a process called microfluidics) [1]. A great variety of strategies have been developed to pump liquids in microsystems [2]. Electric forces applied directly on the liquid allow for the pumping of small amounts of liquids without moving parts. Electroosmosis [3], electro-wetting [4], ion-drag pumping [5], electrohydrodynamic induction pumping [6] and ac electroosmosis [7] are examples of electric-field-driven micropumping.

In this work, we study experimentally the pumping of electrolytic solutions using ac electroosmosis (ACEO) [8, 9]. The ACEO pumping of electrolytic solutions employs arrays of micro-electrodes subjected to a low potential ac signal (less than 10 V). The arrays of planar microelectrodes are mounted in a microfluidic channel and net fluid flow is observed after application of the electric potential. The liquid is driven at the surface of the electrodes by the lateral force that the electric field exerts on the induced charge in the diffuse double layer. Unidirectional fluid flow is obtained either using asymmetric arrays of electrodes subjected to a single ac signal [7, 10, 11, 12] or using arrays of symmetric electrodes subjected to a travelling wave signal [13, 14]. The theoretical predictions at low voltage explain reasonably the experimental observations [13, 15]. Nevertheless, the ACEO theory [16] is at present insufficient in order to explain all the experimental observations [11, 14].

In this paper we study the pumping of electrolytes using travelling-wave arrays made of platinum, see Fig. 1. A sketch of the fluid velocity profile produced over the electrodes is shown in Fig. 2. The electrodes are of equal width and gap and energized with a 4-phase ac signal. We
have previously reported experiments where electrolytic solutions (KCl in water) were pumped with an array of titanium electrodes of 20 µm width [17]. We now present experimental results with platinum electrodes of two different widths (10 and 20 µm). The different travelling-wave arrays show the same qualitative behaviour. At low voltages the liquid flows according to the prediction given by the ACEO electroosmosis, i.e. in the direction of the travelling wave. At voltages above a threshold the fluid flow is reversed. The threshold voltage is dependent on the array characteristics.

We also report measurements of the electrical current when the microelectrode array is pumping the liquid. Transient behaviours in both electrical current and fluid velocity are reported.

2. Experimental details

The microelectrode array for the travelling-wave (TW) experiment is shown in figure 1. The device consists of an array of 55 interdigitated electrodes fabricated on a planar glass wafer using photolithography. Two arrays has been studied: array A consists of 20µm wide electrodes separated by 20µm gaps, and array B consists of 10µm wide electrodes and 10µm gaps. Each electrode is driven by an ac voltage $V_i(t) = V_0 \cos(\omega t + \phi_i)$. The difference in phase between consecutive electrodes is $\phi_{i+1} - \phi_i = 90^\circ$, as shown in fig. 1. This produces a travelling-wave potential with a wavelength $\lambda = 160\mu m$ for array A and $\lambda = 80\mu m$ for array B.

Microscope coverslips were used to fabricate a long fluidic channel on the microelectrode array with 700µm × 180µm cross-section, where 180µm is the height of the channel. A 10$^{-4}$ M electrolytic solution of KCl (conductivity is 1.5 mS/m) was used as working fluid. The end points of the channel were connected to PTFE tubings and these to a syringe. The electrolyte was renewed after every measurement.

Fluorescent latex particles (500 nm diameter) were suspended in the electrolyte and used as flow tracers. An Epifluorescence microscope (Nikon Optiphot-100) is used for observing the flow tracers. A digital camera attached to the microscope is used and videos are captured by a computer. The fluid velocity measurements were carried out by particle tracking velocimetry (PTV).

Electrical current measurements were also carried out. The TW array is energised by a four-output-signal generator. We measure the current flowing in the device with four electrical resistors (of low resistance) connected to the device, see Fig. 3. The voltage drop across a resistor is measured using a lock-in amplifier, where the components in-phase $V_x$ and out-of-phase $V_y$ with respect to the driving signal are measured.
3. Results and discussion

3.1. Steady velocity measurements

When an ac voltage is applied to the electrode arrays, fluid flow is driven at the electrode surface. Because the channel is closed with valves at both ends (input and output), the fluid recirculates, and the tracer particles at the top move in the opposite direction to those above the electrodes (see Fig. 2). The pump behaviour was characterized as a function of voltage and frequency by measuring the fluid velocity at a height of 120 µm above the electrodes, which is 2/3 of the channel total height. At this point, the fluid is moving in the opposite direction to that at the electrode surface. Figure 2 shows the theoretical velocity profile obtained as a consequence of slip boundary condition at the electrodes arrays and non-slip at the top of the channel and it was observed experimentally in a previous work [17]. Velocity measurements at the height of 120 µm were considered to be representative of the global behaviour of the pump. The average slip velocity at the electrode level can be seen to be three times the measured velocity.

Figure 4 shows a 2-D map of the velocity measurements for travelling-wave array A (20 µm wide electrodes). These measurements are plotted in the voltage versus frequency domain. A negative value of the velocity indicates that the fluid is driven on the electrodes in the direction of the travelling electric field. At low voltages, the fluid is driven in this direction, as expected by the low voltage theory of ac electroosmosis [13, 14]. We refer to this mode of behavior as Normal Pumping. At higher voltages, the fluid is driven in the opposite direction, a mode of behavior we designate as Reverse Pumping. The physical mechanism responsible for this behavior is not clear at present.

The figure shows four distinguishable regions corresponding to: normal pumping, reverse pumping, no net pumping and non-uniform motion. There is a region of voltage where the pumping mechanism changes from normal to reverse pumping. No net flow was measured in this region, although fluid rolls occurred over the electrodes. We designate this region as No Net Pumping. At low frequencies and for voltages where the reverse pumping occurs, there appears a region where nonuniform fluid motion is observed. The fluid pattern is fully 3D and the profile of figure 2 is not observed. This region is designated as Non-uniform motion. It should be noted that this is a qualitative difference with respect to the observations for the titanium arrays where electrolysis was found when the frequency was decreased before the fluid flow became nonuniform [17].

Figure 5 shows the fluid velocity as a function of frequency for array A at different voltages. The fluid flow is strongly dependent on frequency. In the reversal region we find the nonuniform
motion before any peak in velocity and in the normal region a maximum absolute velocity is not clearly observed.

The same measurements were carried out with the travelling wave array B (10 μm wide electrodes). The qualitative behaviour is similar as for array A. Reverse pumping is observed at voltages at and above 2.5 Vpp. Non-uniform flow is observed in the reverse pumping region when the frequency is low. Figure 6 shows the fluid velocity as a function of frequency for array B at several voltages. A quantitative difference between array A and B is that velocities observed in array B were much greater than those observed in array A at the same voltages. This indicates that the velocity is greater for smaller electrodes. In the normal pumping region a maximum velocity appears at a frequency around 100 Hz. This is different for the reverse pumping where, apparently, a maximum is found at higher frequencies. However, this maximum is very close to the frequency where nonuniform flow is observed and this apparent maximum may not be significant.

**Figure 4.** Recirculation velocities for the 20 micron array

**Figure 5.** Velocity versus frequency for the 20 micron array.

**Figure 6.** Velocity versus frequency for the 10 micron array.

The reproducibility of the velocity measurements is influenced by effects such as aging of...
the electrodes. Nevertheless, the threshold voltage for the appearance of flow reversal is very reproducible. The threshold voltage was observed to be more or less independent on frequency for frequencies below 1 kHz, and increased with frequency above. The threshold voltage, defined as the lowest value at which flow reversal is clearly observed, is shown in the following table for three cases: the titanium 20-µm-wide electrode array (previously investigated in [17]) and the two platinum electrode arrays studied here. The values shown are for frequencies below 1 kHz.

|          | Ti 20 µm | Pt 20 µm | Pt 10 µm |
|----------|----------|----------|----------|
| Threshold Voltage | 4 V<sub>pp</sub> | 3.5 V<sub>pp</sub> | 2.5 V<sub>pp</sub> |

3.2. Transient measurements

Electrical currents were measured as indicated in the experimental details section. Linear behaviour in the current-voltage characteristics was found at signals below 2 V<sub>pp</sub>. At higher voltages, the system was clearly nonlinear. More remarkably, we measured a time-dependent electrical current amplitude, i.e. the current was oscillating at the driving frequency \( I(t) = I_0 \cos(\omega t) \) but the amplitude \( I_0 \) changed slowly in time. The characteristic time for this slow variation was of the order of minutes, very much greater than the period of the ac signal. In array B, we observed the current amplitude changing with time at and above 2 V<sub>pp</sub>. In array A, current amplitudes are dependent on time at voltages above 2.5 V<sub>pp</sub>.

For array A at a voltage of 3 V<sub>pp</sub> and at 400 Hz, we observed that the fluid started to move in the normal pumping mode and, after around three minutes, the direction was reversed. These values of voltage and frequency correspond to a point in the map of fig. 4 inside the no net pumping region. This transient behaviour is shown in figure 7, where the measurements of the voltage drop through the resistor (the current amplitude) are also plotted. For this case, both velocity and current amplitude are strongly correlated.

Figure 7. Velocity and electrical current versus time at 3 V<sub>pp</sub> and 400 Hz
4. Conclusions

The two platinum microelectrode arrays showed similar flow behaviours in the voltage versus frequency domain. Both showed four distinct regions: normal pumping, reversal pumping, no net pumping and nonuniform motion. In both arrays, at low voltages the liquid flowed according to the prediction given by ac electroosmotic theory. At voltages above a threshold the fluid flow was reversed. Reversal flow occurred at lower voltages for the array with smaller electrodes (array B). Typical frequencies for fluid motion were lower for array A.

Comparison with the titanium microelectrode array shows that the threshold voltage was greater for these electrodes. Typical frequencies for fluid motion were lower for the platinum electrode array than for the titanium electrode array.

The measurements of the electrical current when the microelectrode array was pumping the liquid showed transient behaviours. Simultaneous measurements of velocity and current showed a strong correlation.

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5. References

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