Microchannel electrokinetics of charged analytes in buffered solutions near floating electrodes

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Microchannel electrokinetics of charged analytes in buffered solutions near floating electrodes

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We present both experimental and numerical studies of nonlinear electrokinetic flow of buffered solutions seeded with dilute analytes in a straight microchannel (0.6 µm high, 250 µm wide, and 9000 µm long) with a 0.15 µm high 60 µm wide electrode situated at the bottom center of the channel. Such studies will enable a fundamental understanding of nonlinear transport effects of ions in electrolyte systems with a significant Debye screening layer. Initial experimental studies have shown an order of magnitude increase of concentration near the electrodes, but numerical studies have so far failed to accurately predict such behavior in these flow regimes. Experimentally, using conventional fluorescence microscopy, we investigated the concentration gradient (as well as the associated electroosmosis, induced-charge electro-osmosis, and electrophoresis) of the charged analyte near the floating electrode as a function of analyte (1 to 10 µM fluorescein and bodipy) and buffer (1 to 10 mM borate and phosphate) concentrations and an externally applied voltage drop (50 to 100 V) along the channel. We have implemented a nonlinear continuum kinetics model of the system involving the electric potential, the buffer flow velocity, the pressure, and the four ionic concentration fields and compared the resulting numerical simulations with experiments.
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Induced-charge electro-osmosis: UCSB experimental setup I

**Extreme aspect ratios – Debye layer overlap**

- \( L = 9000.00 \ \mu m \)
- \( w = 250.00 \ \mu m \)
- \( l = 60.00 \ \mu m \)
- \( H = 0.50 \ \mu m \)
- \( \lambda_D = 0.03 \ \mu m \)
Induced-charge electro-osmosis: UCSB experimental setup II

- Mercury lamp
- Inverted fluorescence microscope
- Microchip stage
- Data analysis
- Power supply
- Optical filter cubes
- CCD camera/eyepiece
- Microchip
- Fluorescence dye
- Objective
- Filter cube
- Dichromatic mirror
- Emission filter
- Ocular
- Hg lamp
- CCD
The continuum description of the electric screening in electrolytes: the double layer

- **Governing equations**
  - **Navier-Stokes equation** \( \mathbf{u} \): \[ \rho \left[ \partial_t \mathbf{u} + (\mathbf{u} \cdot \nabla) \mathbf{u} \right] = -\nabla p + \eta \nabla^2 \mathbf{u} - Z e (c_+ - c_-) \nabla \phi \]
  - **Incompressibility** \( \rho \): \[ \nabla \cdot \mathbf{u} = 0 \]
  - **Electrostatic Maxwell eq.** \( \phi \): \[ \nabla \cdot (\epsilon \nabla \phi) = -Z e (c_+ - c_-) \]
  - **Nernst-Planck equation** \( \mathbf{J} \): \[ \mathbf{J}_i = -\frac{D_i}{k_B T} c_i \mathbf{\nabla} \mu_i + \mathbf{u} c_i \]
  - **Continuity equation** \( c \): \[ \nabla \cdot \mathbf{J}_i = -\partial_t c_i \]

![Diagram of the double layer model](image)

- Bulk electrolyte
- Debye layer: \( \lambda_D \sim 30 \text{ nm} \)
- Stern layer
- Charged wall

\( \lambda_D \sim 30 \text{ nm} \)
Induced-charge electro-osmosis: Basic principle

- An external potential difference is applied (+$V_0$ and $-V_0$) to electrolyte
- Induced polarization charge appears in the metallic electrode
- Ions in the electrolyte screens out the $E$-field normal to the electrode
- The tangential $E$-field drives an ionic current along the dielectric
- The moving ions drags the liquid along the dielectric
- Flow-rods are consequently induced in the electrolyte

![Diagram](image-url)
Induced-charge electro-osmosis: UCSB experimental setup III
Experimental parameters and calibration of fluorescent detection

### Experimental Parameters

<table>
<thead>
<tr>
<th>Electric Fields</th>
<th>Ion/Analyte Densities</th>
</tr>
</thead>
<tbody>
<tr>
<td>± 13.3 kV/m</td>
<td>1 mM / 1 mM</td>
</tr>
<tr>
<td>± 26.6 kV/m</td>
<td>1 mM / 100 mM</td>
</tr>
<tr>
<td>± 39.9 kV/m</td>
<td>1 mM / 10 mM</td>
</tr>
<tr>
<td>± 53.2 kV/m</td>
<td>1 mM / 1 mM</td>
</tr>
<tr>
<td>± 66.5 kV/m</td>
<td>10 mM / 1 mM</td>
</tr>
<tr>
<td>± 79.8 kV/m</td>
<td>10 mM / 100 mM</td>
</tr>
<tr>
<td>± 93.1 kV/m</td>
<td>10 mM / 10 mM</td>
</tr>
<tr>
<td>± 106.4 kV/m</td>
<td>10 mM / 1 mM</td>
</tr>
<tr>
<td>± 119.7 kV/m</td>
<td>10 mM / 1 mM</td>
</tr>
</tbody>
</table>

### Electrode Geometries

- Ellipsoid
- Circle
- Square

### Analytes

- Rhodamine 6G (positive)
- Bodipy DiSulfonate
- Fluorescein (negative)
Real-time CCD recordings of one experiment

Transverse average of the fluorescence signal

Flow direction

Top view

Electrode

-\( V_0 \)

+\( V_0 \)

\( x \)
Theoretical model: the 2D computational domain

Extreme aspect ratios
- \( L = 9000.00 \ \mu m \)
- \( l = 60.00 \ \mu m \)
- \( H = 0.50 \ \mu m \)
- \( \lambda_D = 0.03 \ \mu m \)
Theoretical model: Induced-charge electro-osmosis in microchannels

The symmetry is broken by the EO-flow from the surrounding walls
Theoretical model: potential, pressure and velocity profiles

The potential $\phi$

$t = 0$

$t \rightarrow \infty$

The pressure $p$

$\Delta V$

$\Delta p$

$Q_{eo}$ $Q_p$

$Q$

$V_0$

$-4500 \mu m$

$4500 \mu m$

$0 \mu m$
Dye concentration: comparing simulation with experiment

(a) $c_{(d)}(x)$ [m$^{-3}$]

- $c_{(b,-)} = 10$ μM
- $c_{(b,+)} = 11$ μM
- $c_0^{(d)} = 1$ μM

(b) $I$ [AU]

- $c_0^{(ph)} = 1$ mM
- $c_0^{(BDP)} = 1$ μM

$V_0$, flow

5V, 10V, 15V, 20V, 25V, 30V

50V, 75V, 80V, 90V
Concluding remarks

- Experimental setup for observing induced-charge electro-osmosis (ICEO) in micro/nanochannels
- Transient development of dye concentration has been measured
- Theoretical model combining EO flow with ICEO has been established for extreme aspect ratios
- Qualitative agreement between experiments and numerical simulation has been achieved
- A good basis for further ICEO studies have been developed and successfully tested
Theoretical model

$c^{(c)}(x)$ [m$^{-3}$]

100 μM buffer
1 μM dye

15V, 10V, 5V, 20V, 25V
Theoretical model

![Graph showing a theoretical model with various voltage levels and concentrations.](image-url)