Nanofluidic Analyte Preconcentration Using Fluid Field-Effect Diodes

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Introduction

Preconcentration of charged analytes is often necessary when dealing with biological fluid samples due to low working fluid volumes and the dilute nature of many biomolecules of interest [1,2]. Electrokinetic preconcentration methods which exploit a local balance between ion electrophoresis & bulk electroosmosis are widely employed in micro & nanofluidics to address this issue [1-3]. These techniques provide a flexible platform for concentrating and separating molecules of different size and charge. Using numerical simulations, we show that nanofluidic channels with wall-embedded transverse electrodes (a configuration often utilized as transistors and/or diodes) can also be used to stack, focus, and/or separate charged molecules at distinct locations based on different measurable transport properties of the analyte such as valence, mobility, etc.

Nanofluidic Device

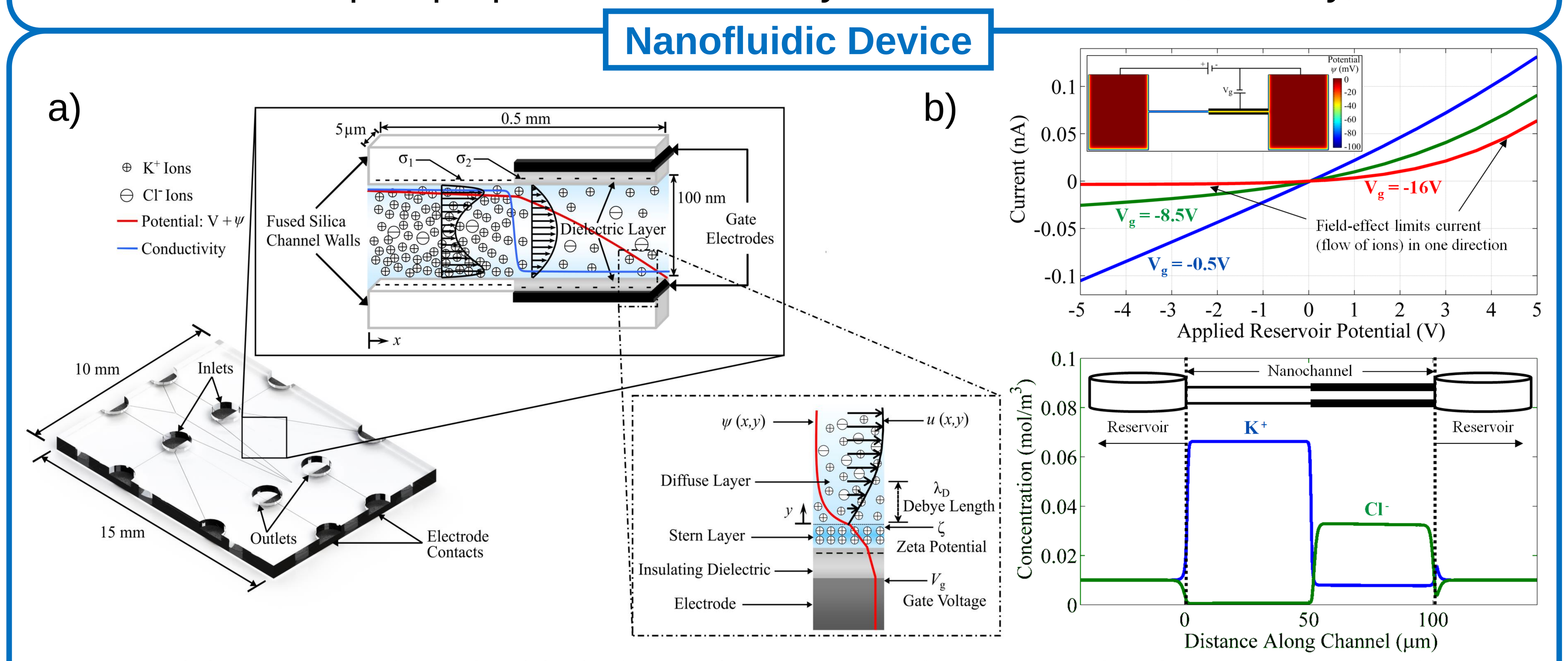

Figure 1. (a) Device with embedded electrodes -- insets show example electroosmotic velocity & ion distributions in the electric double layer (EDL), and (b) current-voltage characteristics of device displaying diode-like characteristics (top) along with axial electrolyte ion profiles (bottom).

Numerical Model

- Nanochannel and reservoir electrokinetics modeled using COMSOL v5.2a
- Customized mesh accounts for disparate length scales ranging from ~ 0.1 nm to ~ 1mm (7 orders of magnitude)
- Transverse EDL potential governed by Poisson's Equation, modeled with "Coefficient Form PDE" module
- Potential within channel from applied electric field determined by current conservation using "Electric Currents" module
- Electroosmotic velocity field modeled by Stokes' equation & flow continuity with "Creeping Flow" module
- Electrolyte and analyte species modeled by species conservation (Nernst-Planck) equation using the "Transport of Diluted Species" module

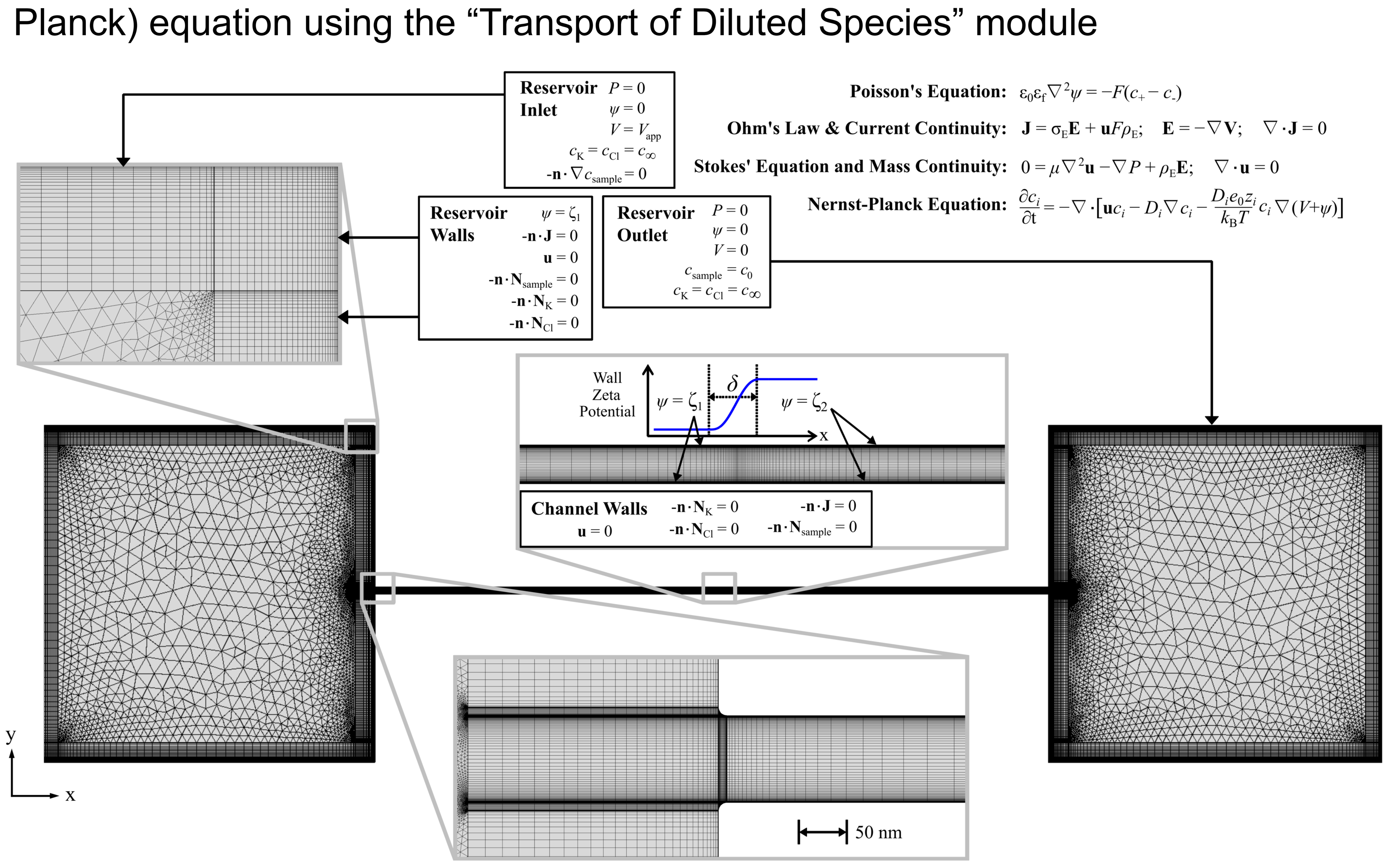

Figure 2. 2D COMSOL Multiphysics model, with governing equations and boundary conditions used in our numerical simulations.

Results

Our simulations show enhancement factors of up to 10^5 for cases with thick electric double layers and large variations in surface charge along the channel. In such cases, electric field gradients from ion concentration polarization effects & thick EDLs can cause mid-channel sample focusing or stacking. For certain conditions, the focusing location can be shifted from the edge of the electrode to some arbitrary position depending on the analyte properties, enabling simultaneous focusing & separation.

Enhancement Mechanisms

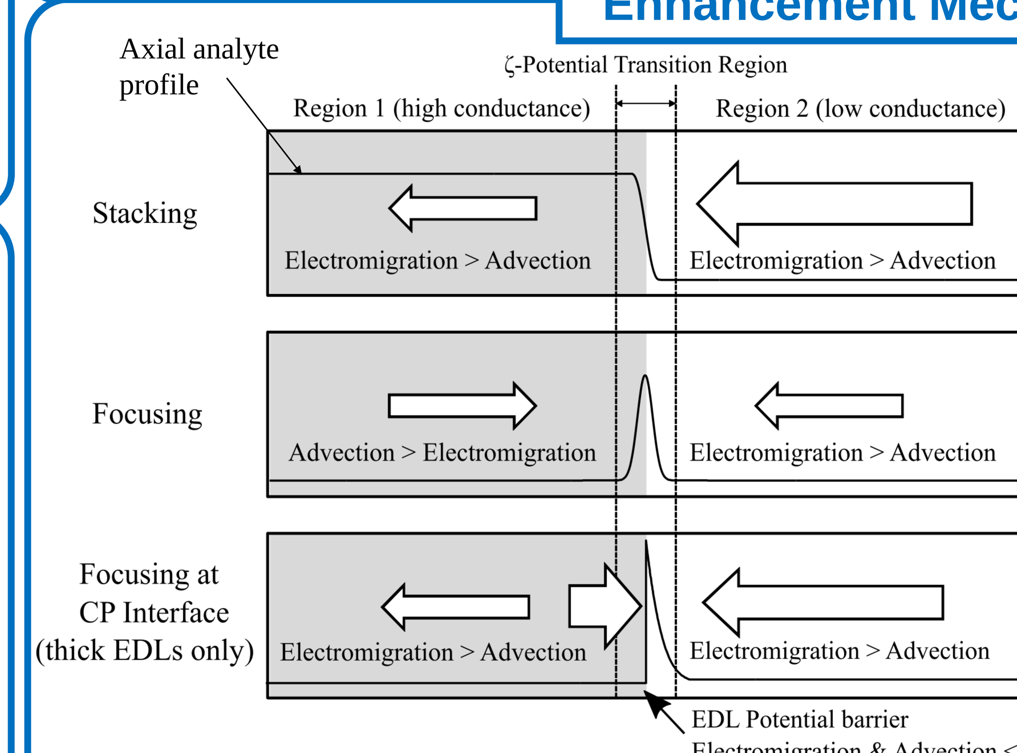

Figure 3. Diagram of the possible concentration enhancement mechanisms for our configuration.

- Stacking increases the analyte concentration in one region (analogous to a traffic jam)
- Focusing drives analyte ions to concentrate at a single location
- For thick electric double layers, ions can be focused at or near the concentration polarization (CP) interface between the two regions of different EDL potential

Preconcentration Capabilities

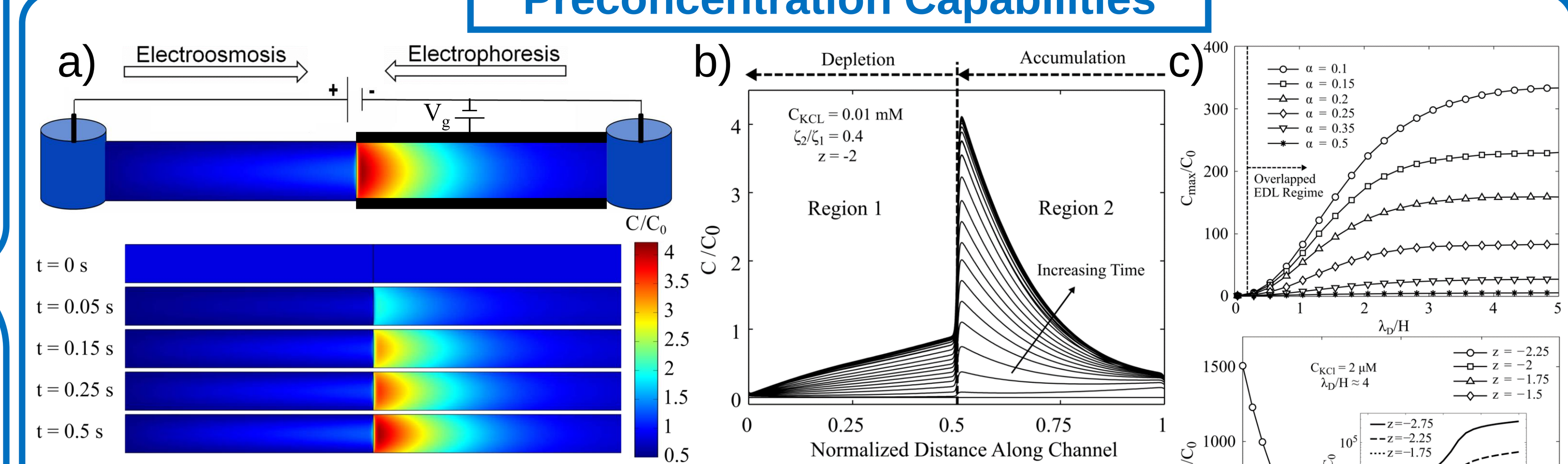

Figure 4. (a) Transient 2D sample concentration profiles, (b) time evolution of centerline axial sample profile, (c) enhancement for varying parameters c_{KCl}, $E_{applied}$, and ζ.

Stationary Focusing & Separation

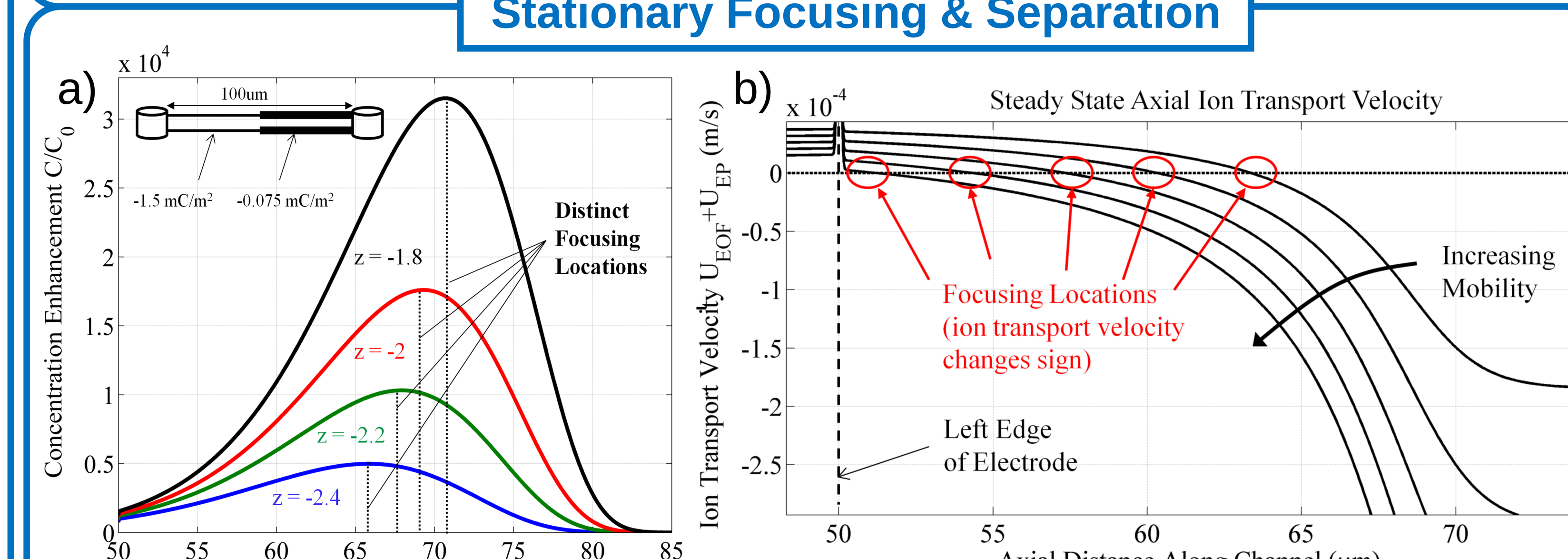

Figure 5. (a) Axial concentration profiles for several analytes demonstrate that ions of different valence can be focused 10,000-fold at unique locations, and (b) net ion transport velocities show predicted focusing locations from the model.

Conclusions

- Nonuniform axial electric fields induced by field-effect surface charge modulation in nanochannels can be leveraged for stacking, focusing, & separation of analytes
- Method allows for tunable, stationary sample preconcentration by varying the potential applied to an embedded gate electrode
- Can potentially achieve up to hundred-thousandfold concentration enhancement for certain analytes
- Less dispersion and greater enhancement than microchannel-based methods [1,3] without the need for multiple electrolyte solutions

References

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