Nanofluidic Analyte Preconcentration Using Fluid Field-Effect Diodes

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Abstract

Introduction

Throughout the field of micro- and nanoscale electrokinetics, floating bipolar electrodes (BPEs) have proven to be useful in manipulating charged ionic species and biomolecules for electroanalytical studies. The growing use of integrated electronics in working towards fully functional lab-on-chip devices has led to the development of BPE applications ranging from analyte separation and preconcentration, electrochemical voltammetry, and many other electrokinetic transport phenomena. BPEs are of particular interest due to the simplicity of fabrication and ability to control electrodes without directly addressing them with an electrical signal, while the choice of buffer solution properties and the magnitude of the external potential difference applied across the fluid provide the additional flexibility to tailor the operating conditions to the precise needs of the desired application.

Use of COMSOL Multiphysics

In this work, we use COMSOL Multiphysics® v5.2a to simulate the 2D and 3D transient electrokinetic response of a floating BPE within a nanofluidic channel filled with an electrolyte buffer solution. We solve the highly coupled, nonlinear Poisson-Nernst-Planck system of equations with convection in the fluid domain coupled with charge conservation within the solid electrode using a combination of the Coefficient Form PDE, Transport of Diluted Species, Creeping Flow, and Electrostatics physics interfaces. The current model represents situations in which the applied potentials are sufficiently low to prevent Faradaic reactions from occurring, and serves as a framework upon which the effects of these surface reactions will be added and explored.

Results and Conclusions

Our numerical results match the observed trend from fluorescence experiments, in which an applied DC electric field causes accumulation and depletion of a charged tracer species on either side of the BPE due to electrostatic attraction and repulsion from the induced surface charge. We show that after the applied field is removed, a transient focusing mechanism causes the accumulation and depletion sides of the BPE to briefly invert before the tracer species ultimately diffuses away. This focusing arises as a result of the competing time scales between charge relaxation within the fluid and solid electrodes, and creates local electric field gradients within the fluid due to the accumulated charge distribution of background electrolyte and tracer species ions near the BPE. The nonzero free charge density from these accumulated ions causes a local fluid potential distribution which decays more slowly than the applied potential near the channel inlet and outlet, leading to an electric field which persists for a short time after turning off the applied field and which reverses direction everywhere except above the electrode. By including Faradaic reactions and surface adsorption of ions on the BPE, this model will ultimately enable us to investigate more complicated electrochemical processes in which other types of localized analyte enrichment and transient discharging have been experimentally observed but heretofore unexplained fully.

Figures used in the abstract

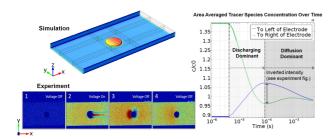


Figure 1: Numerical and experimental tracer species concentration profiles, as well as a plot showing transient focusing above one side of the electrode after the applied electric field is turned off.