Electronic Supplementary Material (ESI) for Analyst.

Optimal Voltage for Nanoparticle Detection with Thin Nanopores

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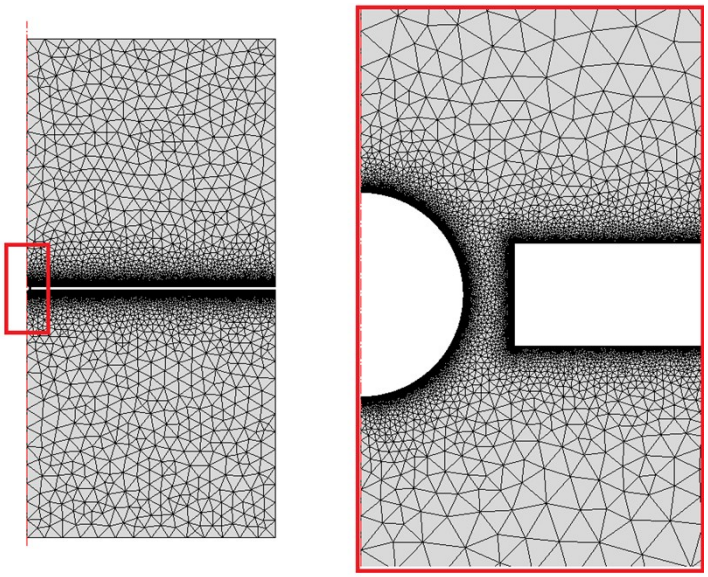


Figure S1 Plot of mesh for the cases with 100 nm particle in the pore. Pore is 150 nm in diameter and 50 nm in length. For the inner surface of the pore and particle surface, the mesh size of 0.1 nm was used to consider the effect of electrical double layers. For the charged boundaries of the reservoirs the mesh of 0.5 nm was chosen to lower the memory cost during calculation.

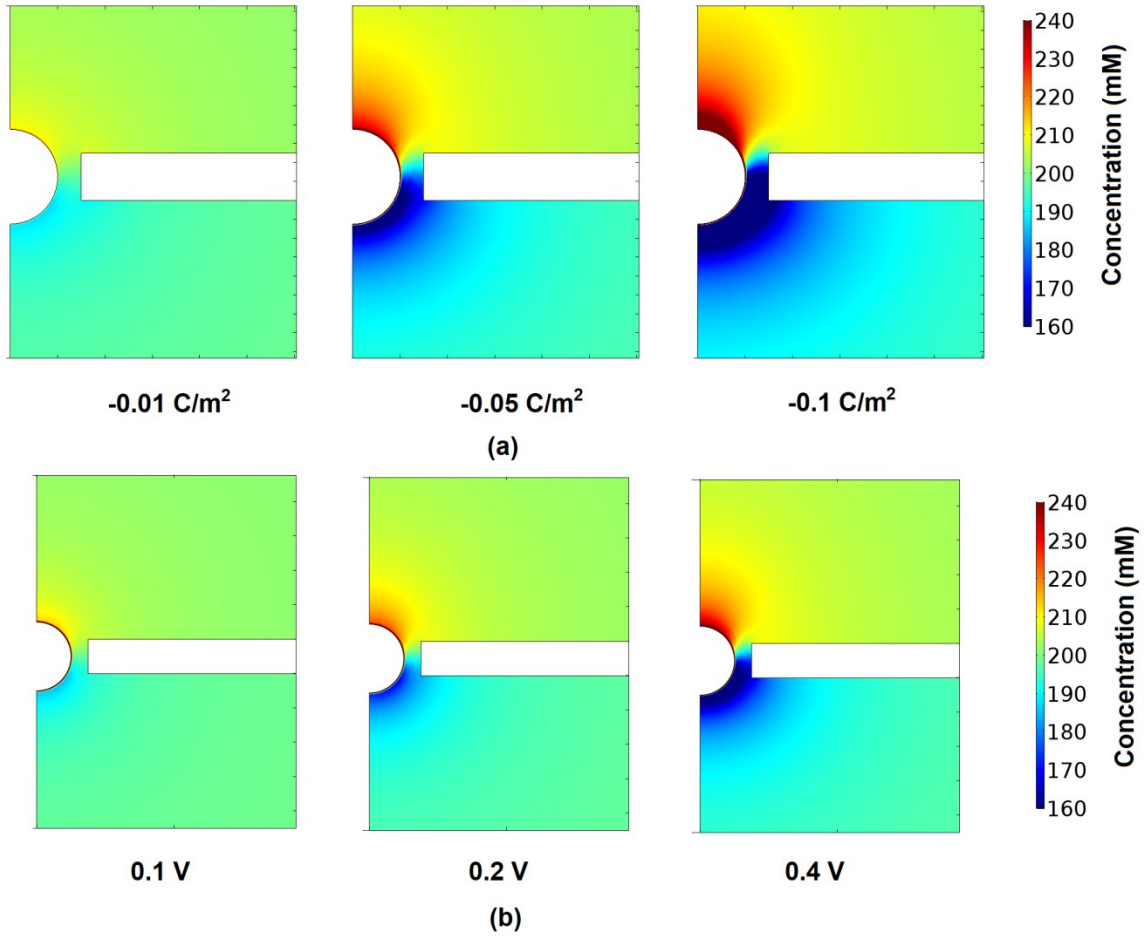


Figure S2 (a) Cocnetration distribution of totoal ions in the pore with particles under different surface charge densities at 0.4 V. (b) Cocnetration distribution of totoal ions in the pore with particles under -0.06 C/m^2 at different voltages. Particle size is 100 nm in diameter. Pore is 150 nm in diameter and 50 nm in length. 0.1 M KCl was selected as the solution.

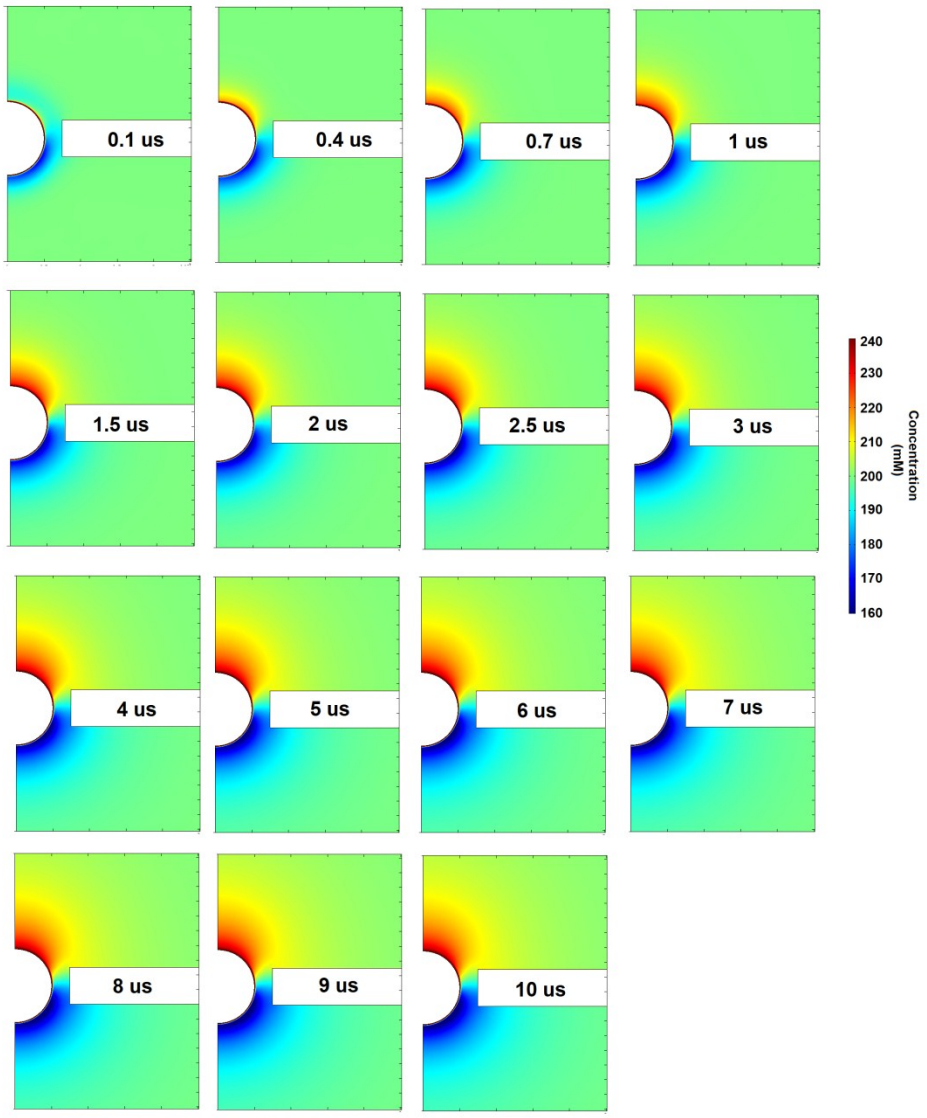


Figure S3 Dynamic total ionic distributions in the nanopore with the particle in the pore center. The case contains a nanoparticle with 100 nm in diameter and a nanopore with 150 nm in diameter and 50 nm in length. The surface charge densities of the particle and the pore were set as -0.08 and -0.005 C/m^2. Voltage was chosen as 0.25 V. In our simulations, the dynamic process of the first 10 μs was investigated with a time step as 0.1 μs. 0.1 M KCl was selected as the solution.

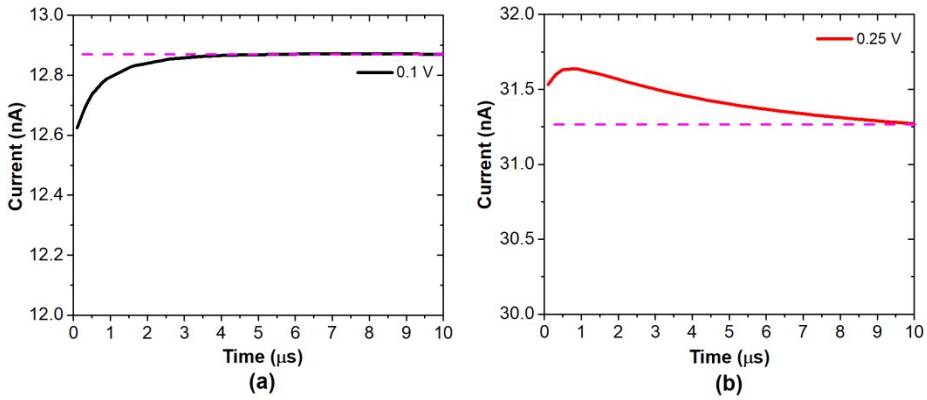


Figure S4 Dynamic ionic current through the nanopore with the particle in the pore center. The case contains a nanoparticle with 100 nm in diameter and a nanopore with 150 nm in diameter and 50 nm in length. The surface charge densities of the particle and the pore were set as -0.08 and -0.005 C/m^2. Voltage was chosen as 0.1 (a) and 0.25 V (b). In our simulations, the dynamic process of the first 10 μs was investigated with a time step as 0.1 μs. 0.1 M KCl was selected as the solution.

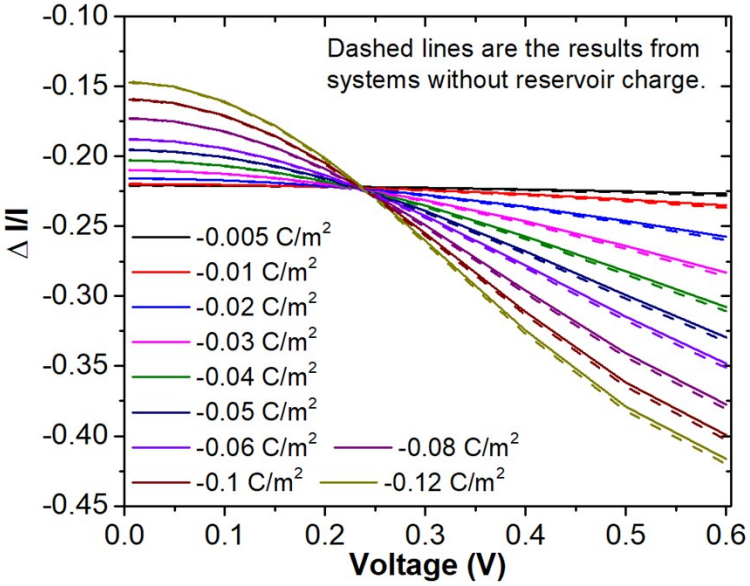


Figure S5. Current blockade ratio obtained from differently charged particles under different voltages. Dashed lines are the results from systems without surface charges of reservoirs. Particle size is 100 nm in diameter. Pore is 150 nm in diameter and 50 nm in length. 0.1 M KCl was selected as the solution.

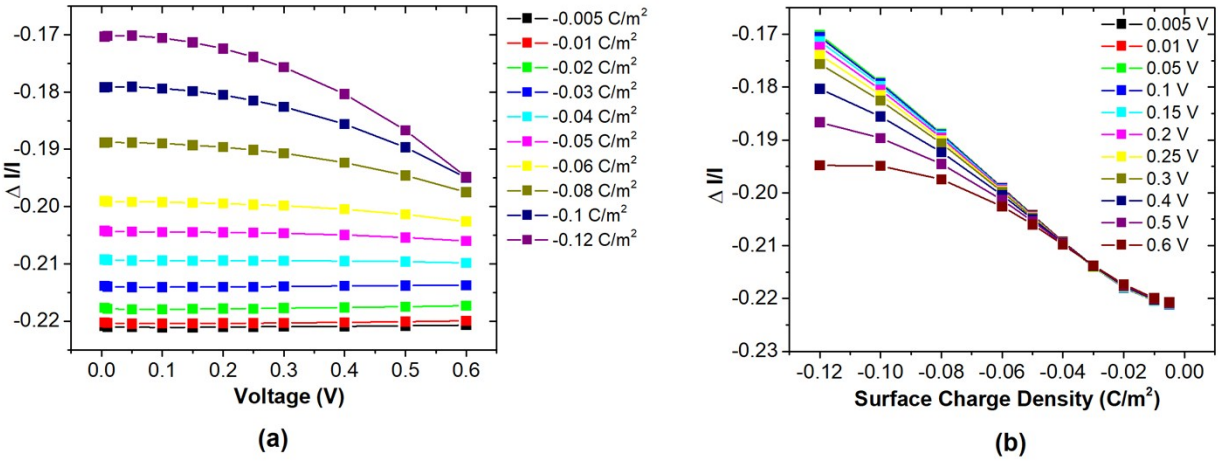


Figure S6. Current blockade ratios obtained from differently charged particles under different voltages. (a) Applied voltage as the variable, and (b) Surface charge density as the variable. Particle size is 100 nm in diameter. The pore is 150 nm in diameter and 50 nm in length. 0.1 M KCl was selected as the solution. There is no consideration of electroosmotic flow in this group of simulations.

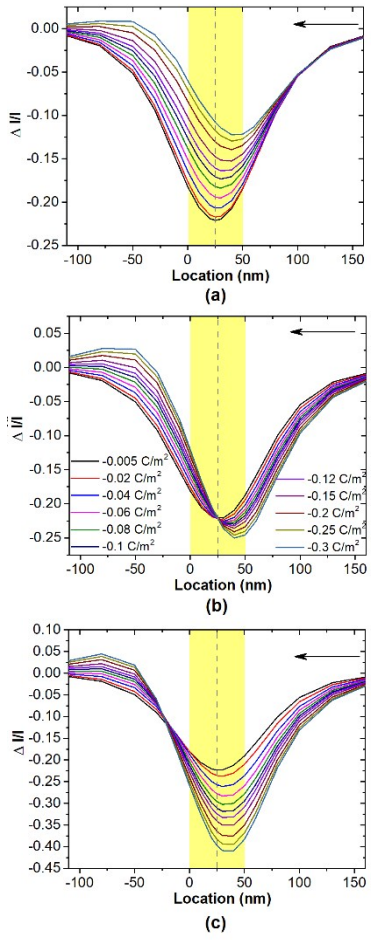


Figure S7 Current traces from 100 nm particles with different surface charge densities through a pore with 150 nm in diameter and 50 nm in length. (a) 0.1 V, (b) 0.237 V and (c) 0.4 V. The arrows show the electrophoresis direction of negatively charged particles. The pore region is shown as yellow. The dashed grey lines show the position of the pore center.

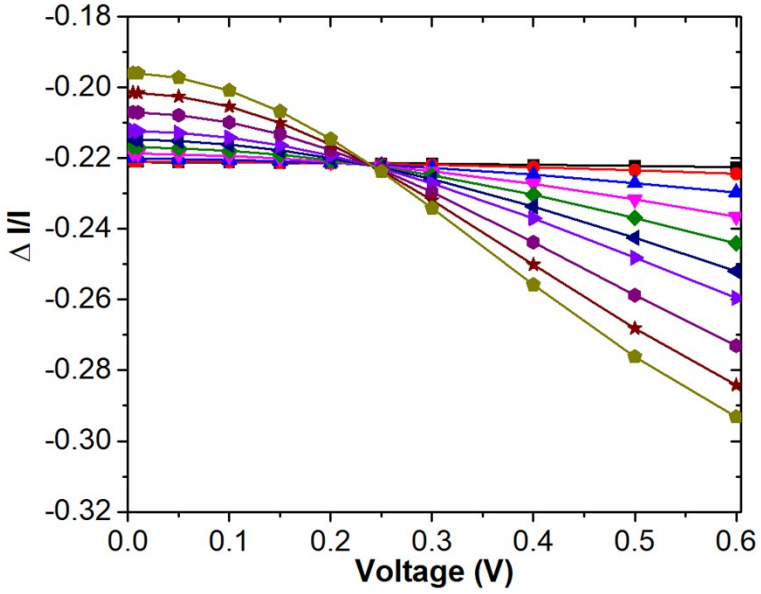


Figure S8. Current blockade ratios obtained from differently charged particles under different voltages. Particle size is 100 nm in diameter. Pore is 150 nm in diameter and 50 nm in length. 0.3 M KCl was selected as the solution.

Theoretical prediction of duration times for particle translocation through a pore with 50 nm in length and 150 nm in diameter.

With the classical theory, we evaluated the rough translocation time of the particle through a 50-nm-in-length pore.

$$v = -\frac{\varepsilon_0 \varepsilon_r \left(\zeta_{particle} - \zeta_{pore}\right) E}{\eta}$$

in which $v, \varepsilon_0, \varepsilon_r, \zeta_{particle}, \zeta_{pore}, E$ and η are the particle speed, dielectric constant of vacuum, dielectric constant of water, zeta potential of particle, zeta potential of inner wall surface, electric filed and viscosity of water, respectively.

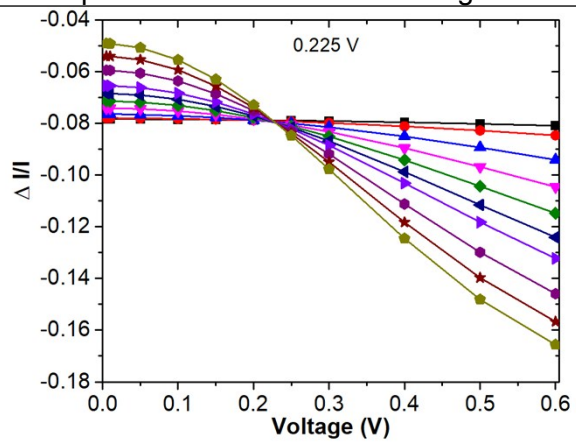
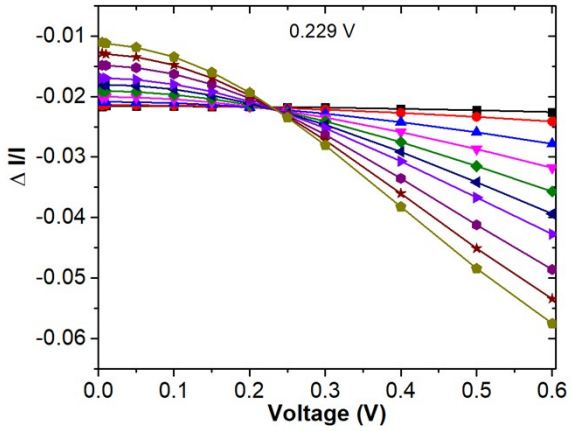
Here, we set $\varepsilon_0 = 8.85 \times 10^{-12} F/m$, $\varepsilon_r = 80$, $\zeta_{particle} = -75mV$, $\zeta_{pore} = -5mV$, E=1.49×10^6 V/m and $\eta = 8.9 \times 10^{-4} Pa \cdot s$. The electric filed strength was calculated with the method in Ref 1. The translocation time for a particle passing through a pore with 50 nm in length is around 0.6 μs.

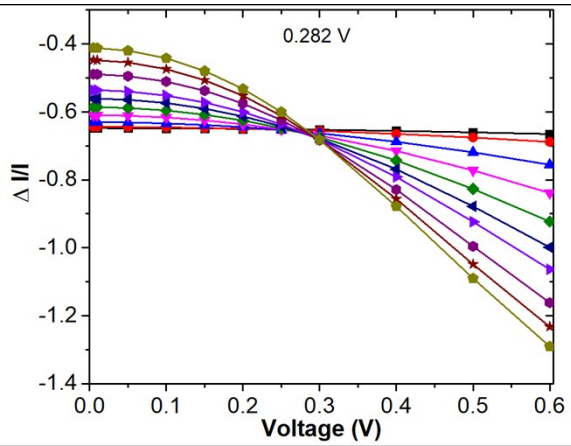
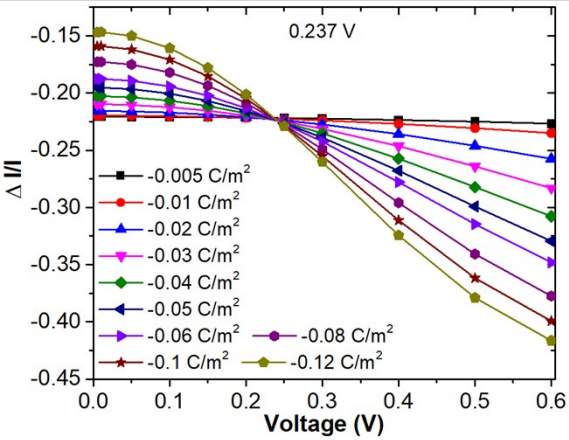
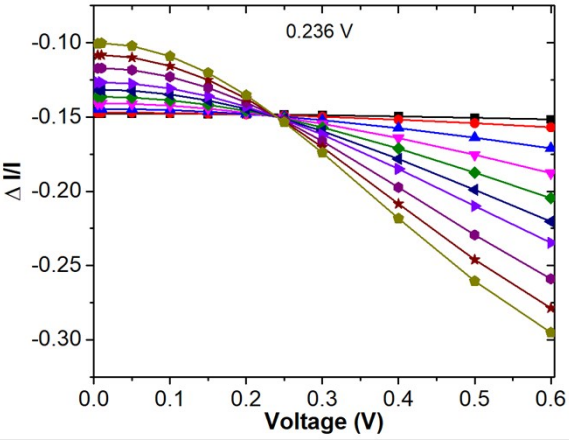
References:

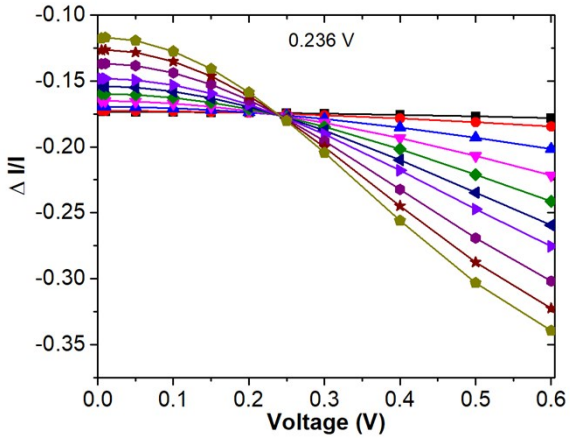
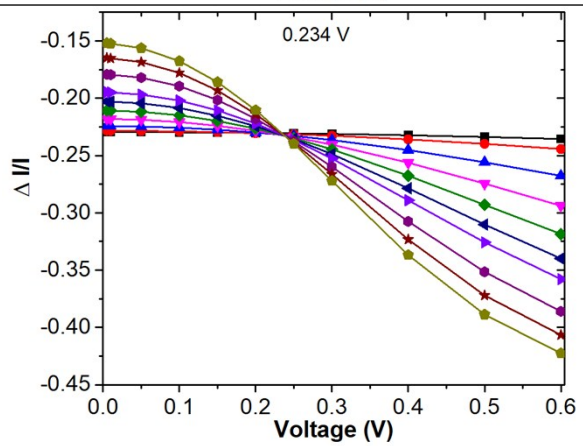
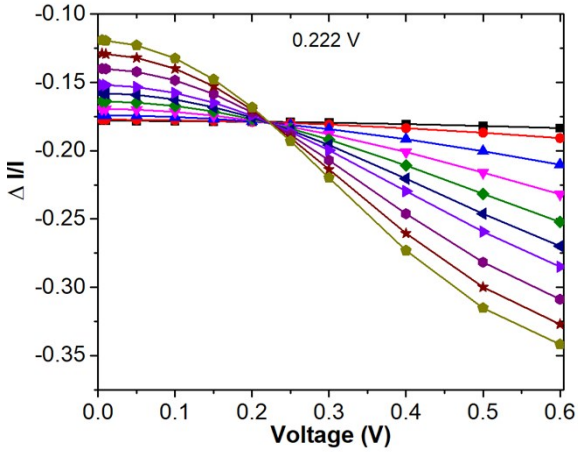
1. M. Davenport, K. Healy, M. Pevarnik, N. Teslich, S. Cabrini, A. P. Morrison, Z. S. Siwy and S. E. Létant, *ACS Nano*, 2012, **6**, 8366-8380.

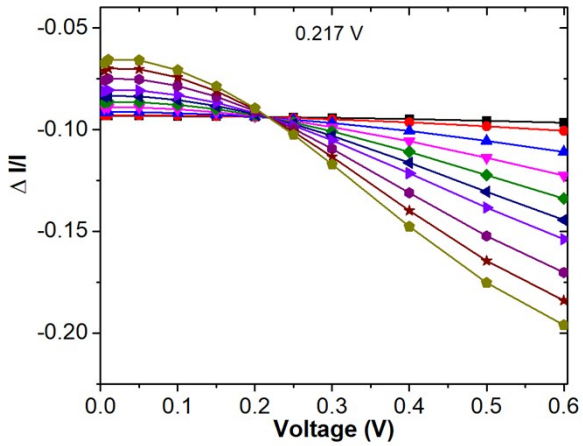
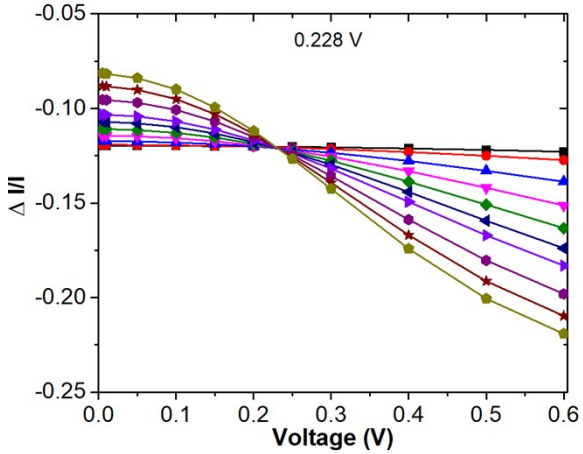
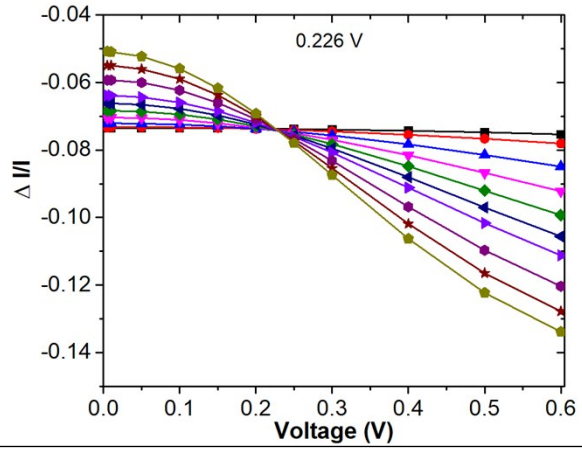
Appendix

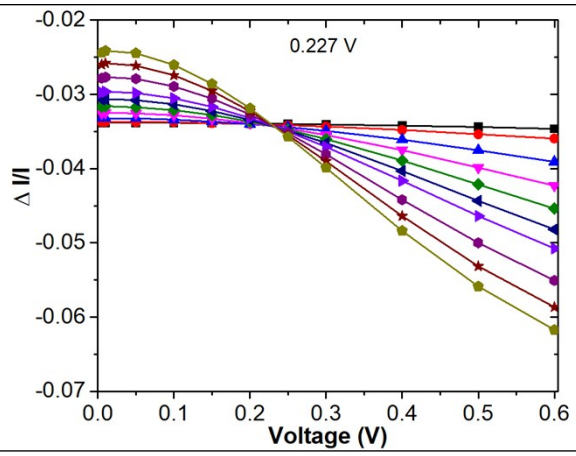
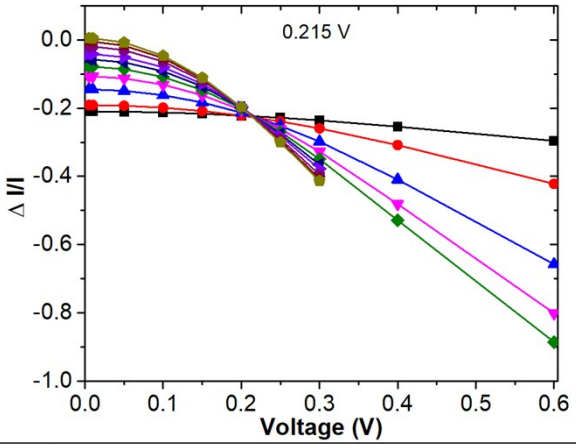
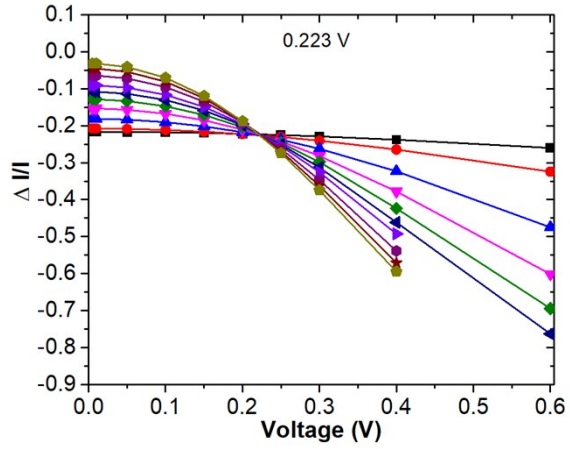
The following shows current blockade raito obtained from differently charged particles under different voltages from all the cases used in this paper.

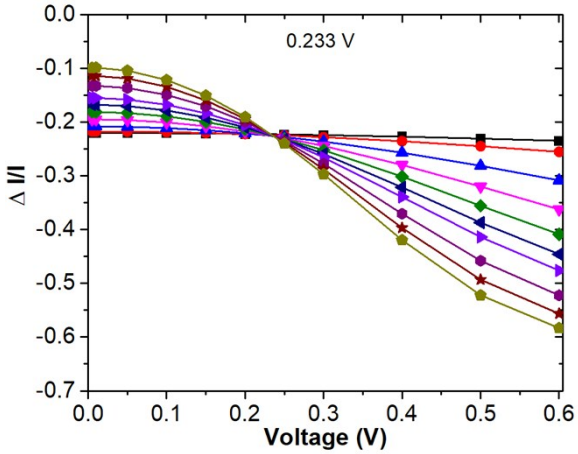
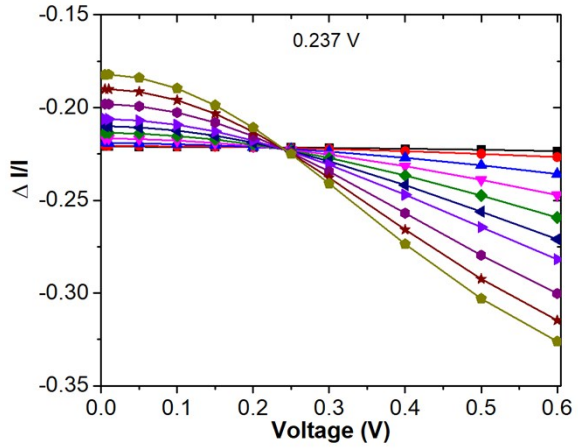
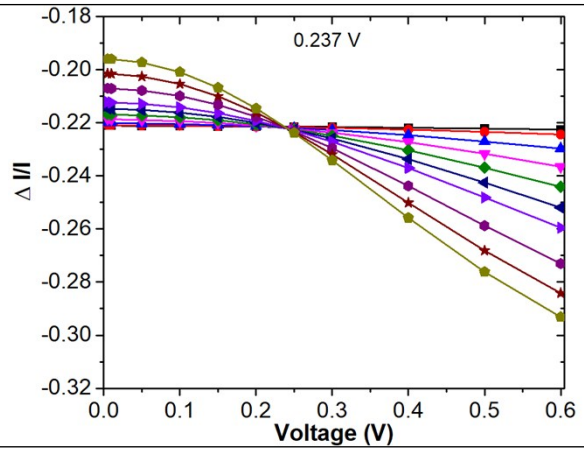
Cases	Current blockade ratio obtained from differently charged particles under different voltages
Particle size is 75 nm in diameter. Pore is 150 nm in diameter and 50 nm in length. 0.1 M KCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	
Particle size is 50 nm in diameter. Pore is 150 nm in diameter and 50 nm in length. 0.1 M KCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	

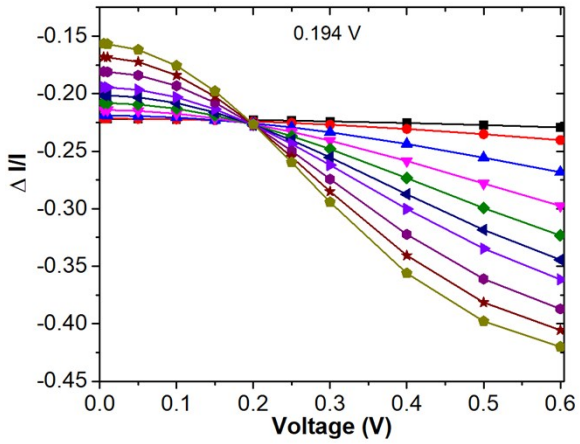
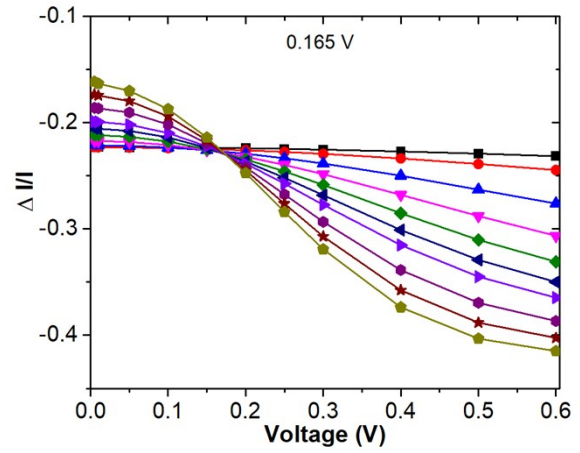
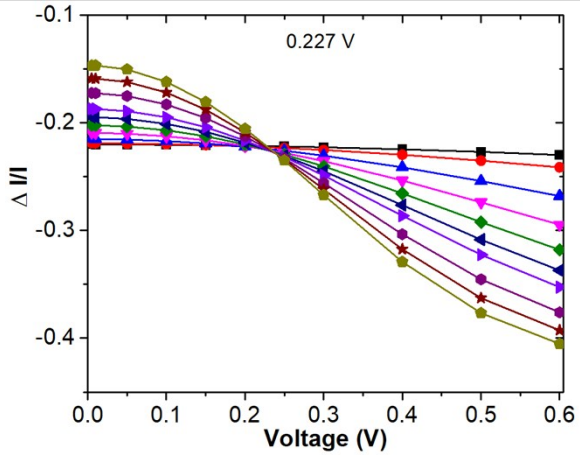
Particle size is 125 nm in diameter. Pore is 150 nm in diameter and 50 nm in length. 0.1 M KCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	
Particle size is 100 nm in diameter. Pore is 150 nm in diameter and 50 nm in length. 0.1 M KCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	
Particle size is 100 nm in diameter. Pore is 150 nm in diameter and 10 nm in length. 0.1 M KCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	

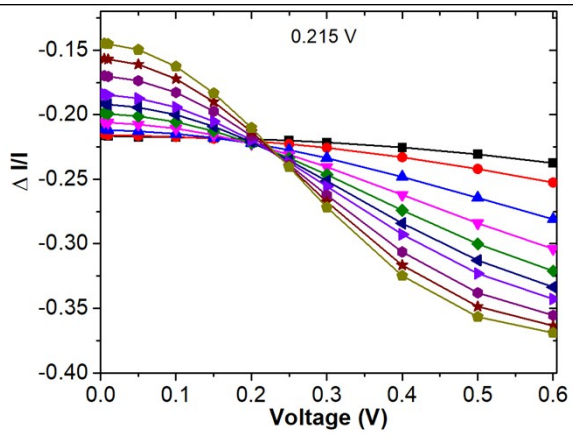
Particle size is 100 nm in diameter. Pore is 150 nm in diameter and 20 nm in length. 0.1 M KCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	
Particle size is 100 nm in diameter. Pore is 150 nm in diameter and 100 nm in length. 0.1 M KCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	
Particle size is 100 nm in diameter. Pore is 150 nm in diameter and 200 nm in length. 0.1 M KCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	

Particle size is 100 nm in diameter. Pore is 150 nm in diameter and 500 nm in length. 0.1 M KCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	
Particle size is 100 nm in diameter. Pore is 175 nm in diameter and 50 nm in length. 0.1 M KCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	
Particle size is 100 nm in diameter. Pore is 200 nm in diameter and 50 nm in length. 0.1 M KCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	

Particle size is 100 nm in diameter. Pore is 250 nm in diameter and 50 nm in length. 0.1 M KCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	
Particle size is 100 nm in diameter. Pore is 150 nm in diameter and 50 nm in length. 0.01 M KCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	
Particle size is 100 nm in diameter. Pore is 150 nm in diameter and 50 nm in length. 0.02 M KCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	

Particle size is 100 nm in diameter. Pore is 150 nm in diameter and 50 nm in length. 0.05 M KCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	
Particle size is 100 nm in diameter. Pore is 150 nm in diameter and 50 nm in length. 0.2 M KCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	
Particle size is 100 nm in diameter. Pore is 150 nm in diameter and 50 nm in length. 0.3 M KCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	

Particle size is 100 nm in diameter. Pore is 150 nm in diameter and 50 nm in length. 0.1 M NaCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	
Particle size is 100 nm in diameter. Pore is 150 nm in diameter and 50 nm in length. 0.1 M LiCl was selected as the solution. Surface charge density of the pore was -0.005 C/m^2.	
Particle size is 100 nm in diameter. Pore is 150 nm in diameter and 50 nm in length. 0.1 M KCl was selected as the solution. Surface charge density of the pore was -0.01 C/m^2.	

Particle size is 100 nm in diameter. Pore is 150 nm in diameter and 50 nm in length. 0.1 M KCl was selected as the solution. Surface charge density of the pore was -0.02 C/m^2.	

COMSOL Report Example

COMSOL MULTIPHYSICS®

D150nml50nm-0.1M-100nm

Report date

Jul 31, 2018 9:30:55 AM

1 Global Definitions

Date	Jul 30, 2018 3:24:15 PM

Global settings

Name	D150nml50nm-0.1M-100nm-charge-voltage KCl-Figure S1.mph
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COMSOL version	COMSOL 5.2 (Build: 220)

Used products

COMSOL Multiphysics
Batteries & Fuel Cells Module

1.1 Parameters 1

Parameters

Name	Expression	Value	Description
Res_l	5e-6	5E−6	
Tip_r	75e-9[m]	7.5E−8 m	
Pore_l	50e-9[m]	5E−8 m	
Volt	400[mV]	0.4 V	
Diffcl	2.03e-9[m^2/s]	2.03E−9 m²/s	
T	298[K]	298 K	
Bulk	100[mmol/L]	100 mol/m³	
F	96485.3415[A*s/mol]	96485 C/mol	
charge	-0.005[C/m^2]	−0.005 C/m²	
Parti_r	50e-9[m]	5E−8 m	
pos	Pore_l/2	2.5E−8 m	
Diffk	1.92e-9[m^2/s]	1.92E−9 m²/s	
chargep	-0.08[C/m^2]	−0.08 C/m²	

2 Component 1

2.1 Definitions

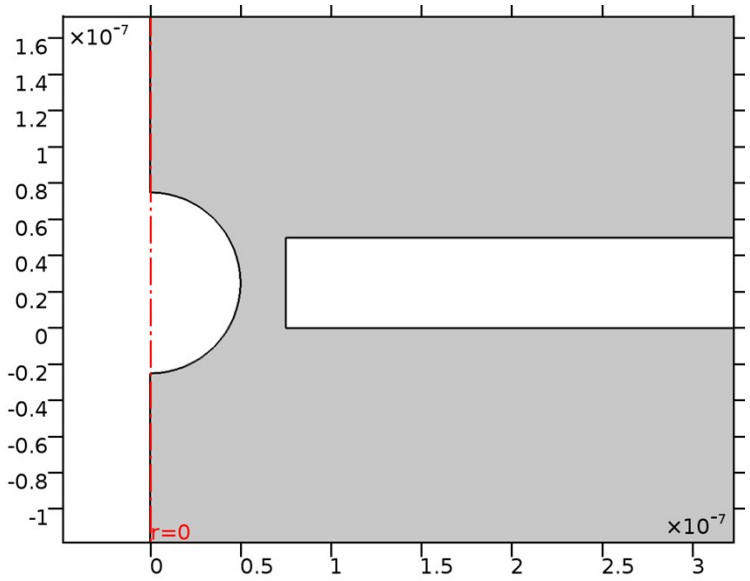
2.1.1 Component Couplings

2.1.2 Coordinate Systems

Boundary System 1

Coordinate system type	Boundary system
Tag	sys1

2.2 Geometry 1



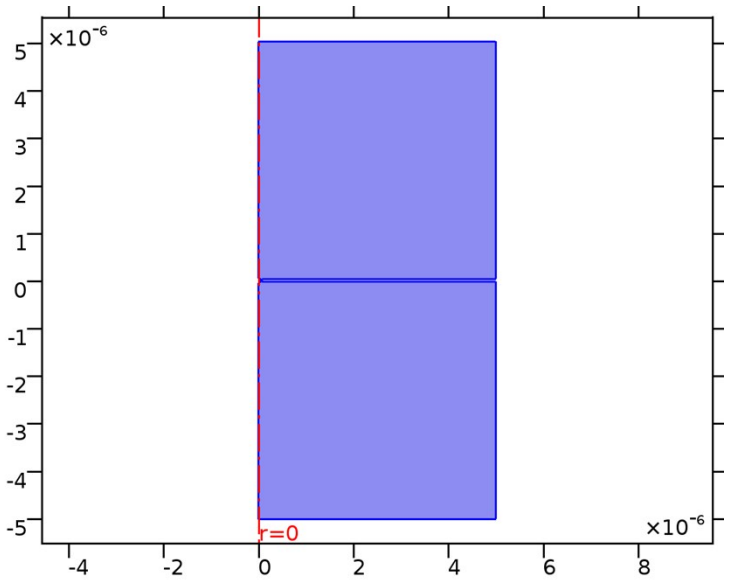
Geometry 1

Units

Length unit	m
Angular unit	deg

2.3 Materials

2.3.1 Water

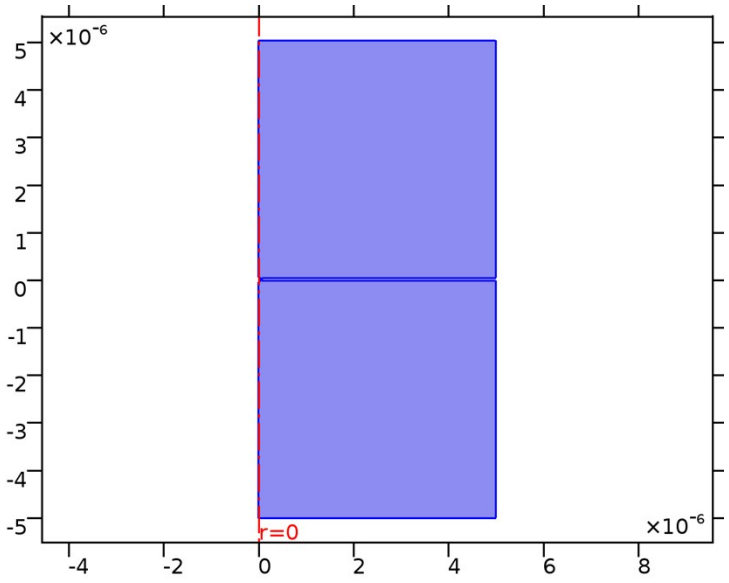


Water

Selection

Geometric entity level	Domain
Selection	Domain 1

2.4 Electrostatics



Electrostatics

Equations

$$\nabla \cdot \mathbf{D} = \rho_v$$

$$\mathbf{E} = -\nabla V$$

Features

Charge Conservation 1
Axial Symmetry 1
Zero Charge 1
Initial Values 1
Space Charge Density 1
Ground 1
Electric Potential 1
Surface Charge Density 1
Surface Charge Density 2

2.4.1 Charge Conservation 1

Equations

$$\mathbf{E} = -\nabla V$$

$$\nabla \cdot (\epsilon_0 \epsilon_r \mathbf{E}) = \rho_v$$

2.4.2 Zero Charge 1

Equations

$$\mathbf{n} \cdot \mathbf{D} = 0$$

2.4.3 Space Charge Density 1

Equations

$$\nabla \cdot \mathbf{D} = \rho_v$$

2.4.4 Ground 1

Equations

$$V = 0$$

2.4.5 Electric Potential 1

Equations

$$V = V_0$$

2.4.6 Surface Charge Density 1

Equations

$$\mathbf{n} \cdot (\mathbf{D}_1 - \mathbf{D}_2) = \rho_s$$

2.4.7 Surface Charge Density 2

Equations

$$\mathbf{n} \cdot (\mathbf{D}_1 - \mathbf{D}_2) = \rho_s$$

2.5 Transport of Diluted Species

Transport of Diluted Species

Equations

$$\nabla \cdot (-D_i \nabla c_i - z_i u_{m,i} F c_i \nabla V + \mathbf{u} c_i) = R_i$$
$$\mathbf{N}_i = -D_i \nabla c_i - z_i u_{m,i} F c_i \nabla V + \mathbf{u} c_i$$

Features

Convection, Diffusion, and Migration
Axial Symmetry 1
No Flux 1
Initial Values 1
Concentration 1

2.5.1 Convection, Diffusion, and Migration

Equations

$$\nabla \cdot (-D_i \nabla c_i - z_i u_{m,i} F c_i \nabla V + \mathbf{u} c_i) = R_i$$
$$\mathbf{N}_i = -D_i \nabla c_i - z_i u_{m,i} F c_i \nabla V + \mathbf{u} c_i$$

2.5.2 No Flux 1

Equations

$$-\mathbf{n} \cdot \mathbf{N}_i = 0$$

2.5.3 Concentration 1

Equations

$$c_i = c_{0,i}$$

2.6 Laminar Flow

Laminar Flow

Equations

$$\rho(\mathbf{u} \cdot \nabla)\mathbf{u} =$$
$$\nabla \cdot \left[-p\mathbf{I} + \mu\left(\nabla\mathbf{u} + (\nabla\mathbf{u})^T\right)\right] + \mathsf{F}$$
$$\rho\nabla \cdot (\mathbf{u}) = 0$$

Features

Fluid Properties 1
Axial Symmetry 1
Wall 1
Initial Values 1
Volume Force 1
Inlet 1

2.6.1 Fluid Properties 1

Equations

$$\rho(\mathbf{u} \cdot \nabla)\mathbf{u} =$$
$$\nabla \cdot \left[-p\mathbf{I} + \mu\left(\nabla\mathbf{u} + (\nabla\mathbf{u})^T\right)\right] + \mathsf{F}$$

$$\rho\nabla\cdot(\mathbf{u}) = 0$$

2.6.2 Wall 1

Equations

$$\mathbf{u} = \mathbf{0}$$

2.6.3 Volume Force 1

Equations

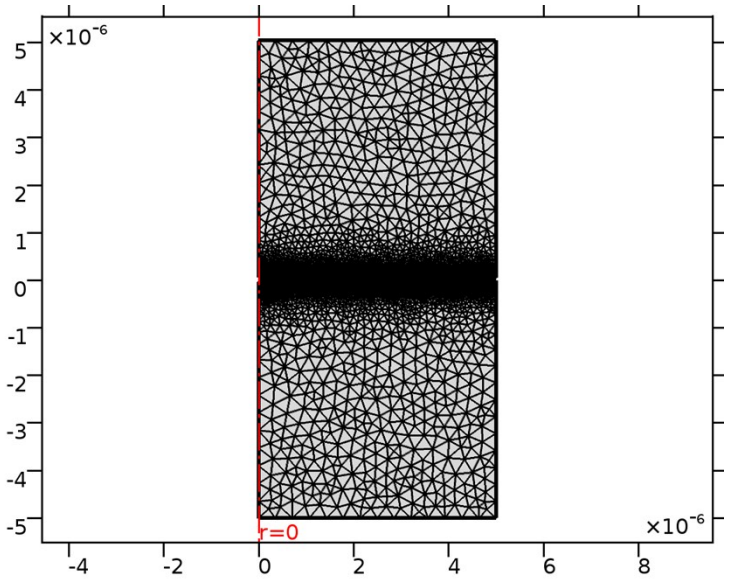
$$\rho(\mathbf{u}\cdot\nabla)\mathbf{u} =$$
$$\nabla\cdot\left[-p\mathbf{I} + \mu\left(\nabla\mathbf{u} + (\nabla\mathbf{u})^T\right)\right] + \mathbf{F}$$

2.6.4 Inlet 1

Equations

$$\mathbf{n}^T\left[-p\mathbf{I} + \mu\left(\nabla\mathbf{u} + (\nabla\mathbf{u})^T\right)\right]\mathbf{n} = -\hat{p}_0$$
$$\hat{p}_0 \geq p_0\,,\quad \mathbf{u}\cdot\mathbf{t} = 0$$

2.7 Mesh 1



Mesh 1

3 Study 1

Computation information

Computation time	29 min 13 s
CPU	Intel(R) Core(TM) i5-2400 CPU @ 3.10GHz, 4 cores
Operating system	Windows 8

3.1 Parametric Sweep

Parameter name	Parameter value list
Bulk	150[mmol/L]

3.2 Parametric Sweep 2

Parameter name	Parameter value list
chargep	-0.01,-0.05,-0.1

3.3 Parametric Sweep 3

Parameter name	Parameter value list
Volt	0.005,0.01,0.05,0.1,0.15,range(0.2,0.2,0.6)

3.4 Stationary

Study settings

Description	Value
Include geometric nonlinearity	Off

Physics and variables selection

Physics interface	Discretization
Electrostatics (es)	physics
Transport of Diluted Species (chds)	physics
Laminar Flow (spf)	physics

Mesh selection

Geometry	Mesh
Geometry 1 (geom1)	mesh1

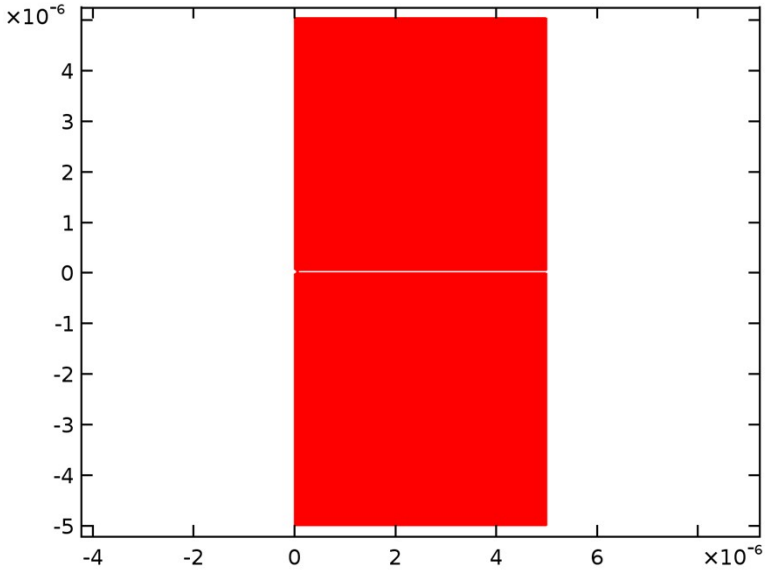
4 Results

4.1 Data Sets

4.1.1 Study 1/Solution 1

Solution

Description	Value
Solution	Solution 1
Component	Save Point Geometry 1



Data set: Study 1/Solution 1

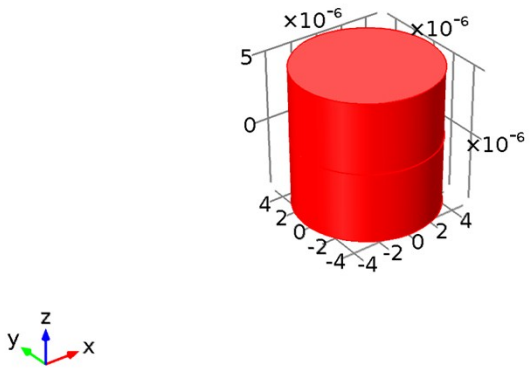
4.1.2 Revolution 2D 1

Data

Description	Value
Data set	Study 1/Solution 1

Axis data

Description	Value
Axis entry method	Two points
Points	{{0, 0}, {0, 1}}



Data set: Revolution 2D 1

4.1.3 Revolution 2D 2

Data

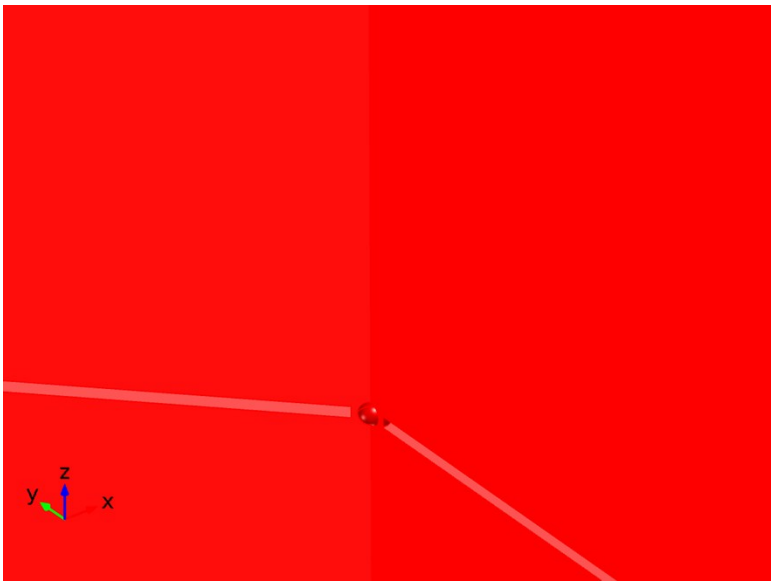
Description	Value
Data set	Study 1/Solution 1

Axis data

Description	Value
Axis entry method	Two points
Points	{{0, 0}, {0, 1}}

Revolution layers

Description	Value
Start angle	-90
Revolution angle	225



Data set: Revolution 2D 2

4.2 Tables

4.2.1 Table 1

Line Integration 1 (F*(H_lm-OH_lm))

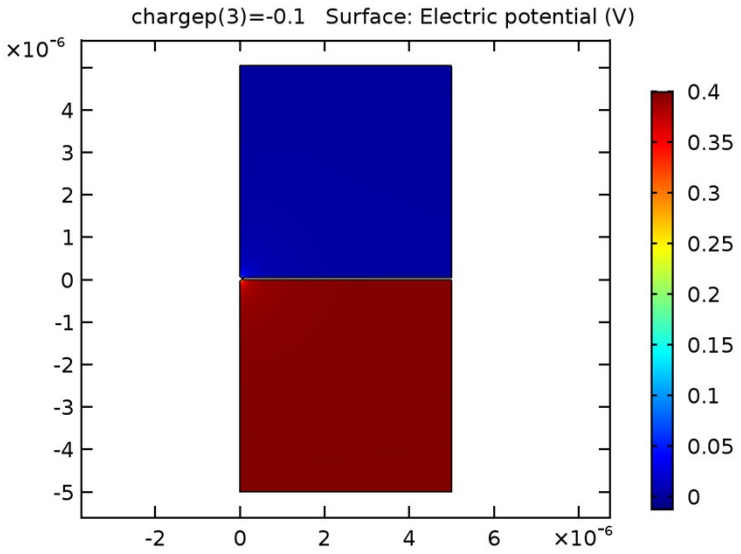
Table 1

Bulk (mol/m^3)	chargep	Volt	F*(H_lm-OH_lm) (m*s*A/mol)
150.00	-0.0050000	0.0050000	1.1129E-9
150.00	-0.0050000	0.010000	2.2258E-9
150.00	-0.0050000	0.050000	1.1129E-8
150.00	-0.0050000	0.10000	2.2255E-8
150.00	-0.0050000	0.15000	3.3377E-8
150.00	-0.0050000	0.20000	4.4491E-8
150.00	-0.0050000	0.40000	8.8839E-8
150.00	-0.0050000	0.60000	1.3295E-7
150.00	-0.010000	0.0050000	1.1129E-9
150.00	-0.010000	0.010000	2.2259E-9
150.00	-0.010000	0.050000	1.1129E-8
150.00	-0.010000	0.10000	2.2253E-8
150.00	-0.010000	0.15000	3.3369E-8
150.00	-0.010000	0.20000	4.4472E-8
150.00	-0.010000	0.40000	8.8686E-8
150.00	-0.010000	0.60000	1.3248E-7
150.00	-0.020000	0.0050000	1.1138E-9
150.00	-0.020000	0.010000	2.2276E-9
150.00	-0.020000	0.050000	1.1136E-8
150.00	-0.020000	0.10000	2.2262E-8
150.00	-0.020000	0.15000	3.3368E-8

Bulk (mol/m^3)	chargep	Volt	F*(H_lm-OH_lm) (m*s*A/mol)
150.00	-0.020000	0.20000	4.4444E-8
150.00	-0.020000	0.40000	8.8301E-8
150.00	-0.020000	0.60000	1.3124E-7
150.00	-0.030000	0.0050000	1.1155E-9
150.00	-0.030000	0.010000	2.2309E-9
150.00	-0.030000	0.050000	1.1152E-8
150.00	-0.030000	0.10000	2.2285E-8
150.00	-0.030000	0.15000	3.3383E-8
150.00	-0.030000	0.20000	4.4427E-8
150.00	-0.030000	0.40000	8.7849E-8
150.00	-0.030000	0.60000	1.2981E-7
150.00	-0.040000	0.0050000	1.1177E-9
150.00	-0.040000	0.010000	2.2355E-9
150.00	-0.040000	0.050000	1.1173E-8
150.00	-0.040000	0.10000	2.2318E-8
150.00	-0.040000	0.15000	3.3408E-8
150.00	-0.040000	0.20000	4.4418E-8
150.00	-0.040000	0.40000	8.7370E-8
150.00	-0.040000	0.60000	1.2842E-7
150.00	-0.050000	0.0050000	1.1204E-9
150.00	-0.050000	0.010000	2.2409E-9
150.00	-0.050000	0.050000	1.1198E-8
150.00	-0.050000	0.10000	2.2357E-8
150.00	-0.050000	0.15000	3.3439E-8
150.00	-0.050000	0.20000	4.4414E-8
150.00	-0.050000	0.40000	8.6894E-8
150.00	-0.050000	0.60000	1.2717E-7
150.00	-0.060000	0.0050000	1.1233E-9
150.00	-0.060000	0.010000	2.2467E-9
150.00	-0.060000	0.050000	1.1225E-8
150.00	-0.060000	0.10000	2.2399E-8
150.00	-0.060000	0.15000	3.3475E-8
150.00	-0.060000	0.20000	4.4413E-8
150.00	-0.060000	0.40000	8.6437E-8
150.00	-0.060000	0.60000	1.2608E-7

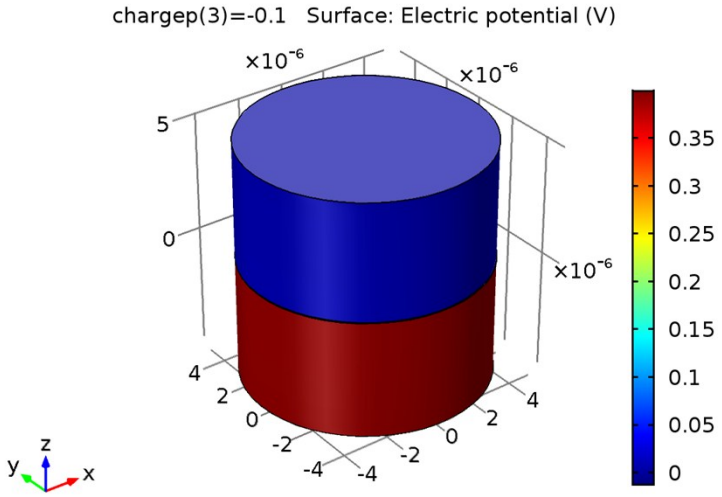
4.3 Plot Groups

4.3.1 Electric Potential (es)



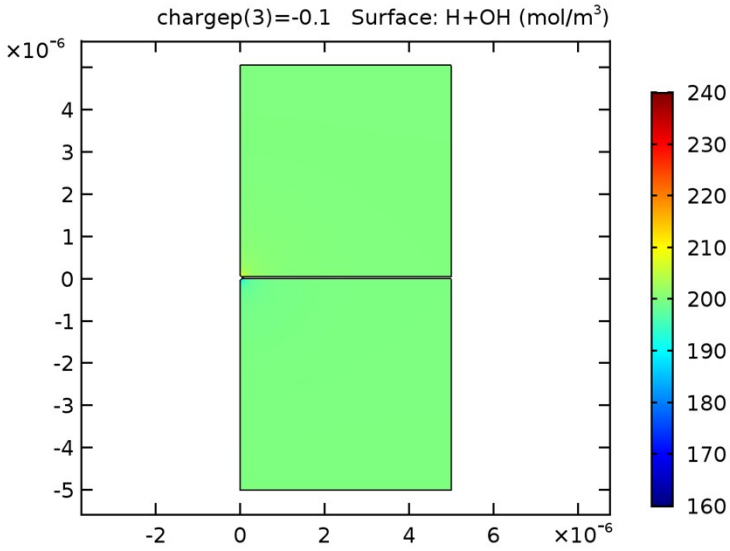
chargep(3)=-0.1 Surface: Electric potential (V)

4.3.2 Electric Potential, Revolved Geometry (es)



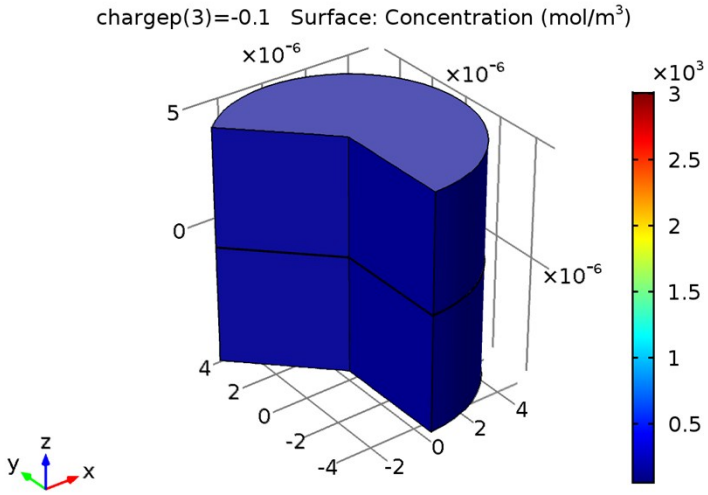
chargep(3)=-0.1 Surface: Electric potential (V)

4.3.3 Concentration (chds)



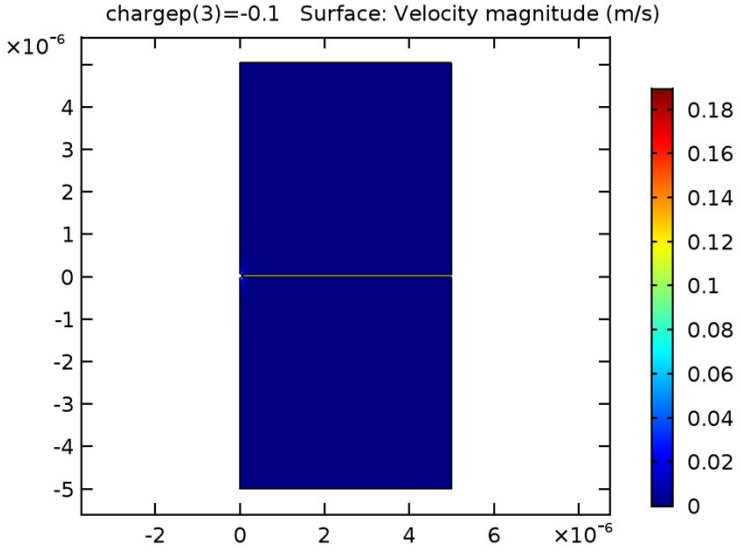
chargep(3)=-0.1 Surface: H+OH (mol/m³)

4.3.4 Concentration (chds) 1



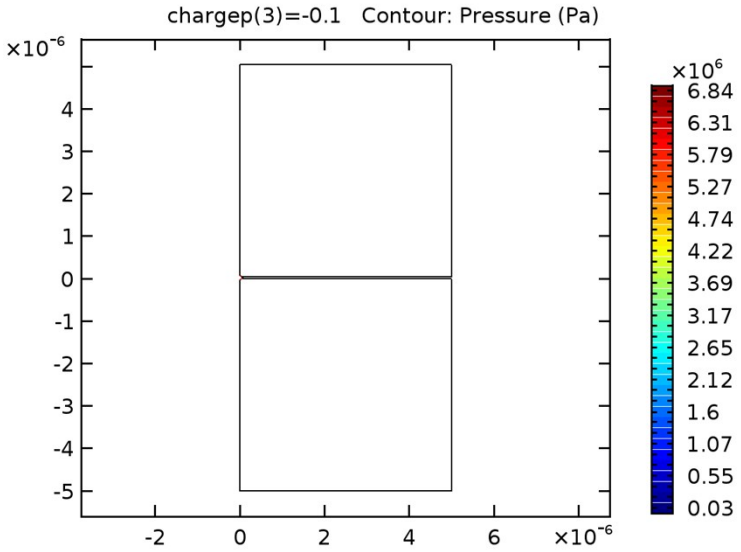
chargep(3)=-0.1 Surface: Concentration (mol/m³)

4.3.5 Velocity (spf)



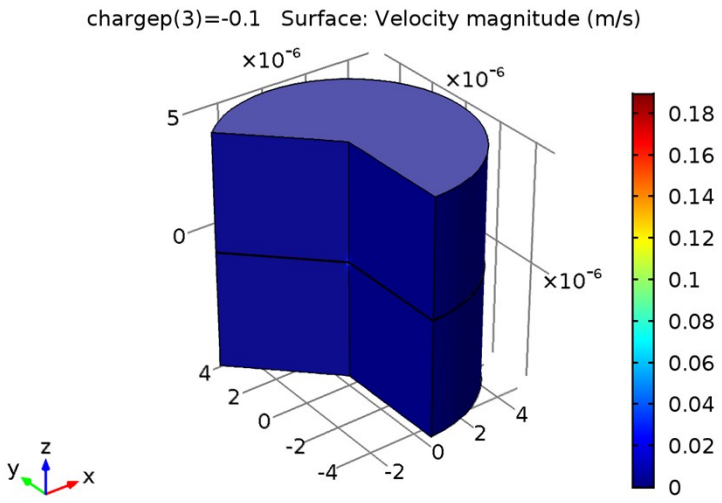
chargep(3)=-0.1 Surface: Velocity magnitude (m/s)

4.3.6 Pressure (spf)



chargep(3)=-0.1 Contour: Pressure (Pa)

4.3.7 Velocity (spf) 1



chargep(3)=-0.1 Surface: Velocity magnitude (m/s)