Tutorial on Electrochemical Modeling

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“The head cannot take in more than the seat can endure.”

— Winston Churchill

Lesson Plan: 3 x 50-Minute Presentations

1. Introductions
2. Fundamentals of modeling
3. Particle-based models

Linear and nonlinear phenomena

Porous electrode battery models

15-minute breaks

1. Introductions
A little about you

Where are you living now?

- Europe
- Africa

Where do you work?

- University
- Industry
- Government

How many years studying electrochemistry (so far)

- 1 or less
- 2 to 4
- 5 to 10
- 11 or more

Your mathematical comfort level

- Algebra
- Basic calculus
- Partial differential equations

What do you want to get out of this tutorial?

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1. Introductions

A little about me

M

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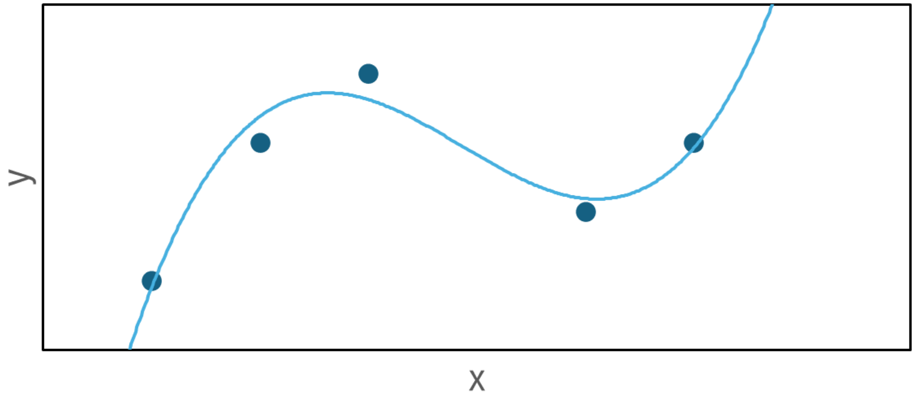
2. Fundamentals of Modeling

Why do we make mathematical models?

- Save time and money by avoiding experiments
- Get information not directly measurable $V = IR$
- Allow for computer control of system
- Demonstrate fundamental physical insight – do you really understand your system or not?
- Because it's fun!



A good model allows for accurate **extrapolation**, not just interpolation and smoothing

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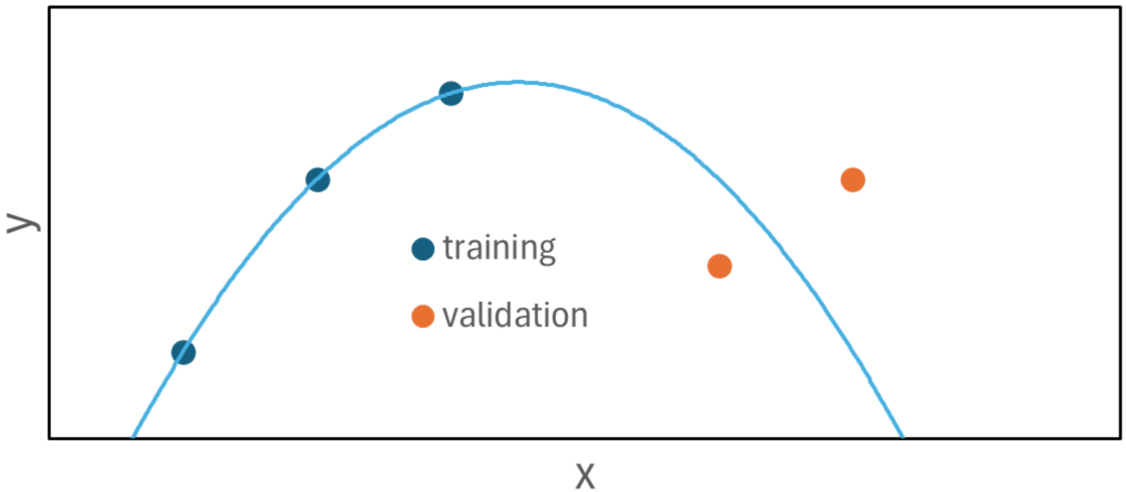
2. Fundamentals of modeling

Parameterization (i.e. fitting, regression)

- Models have fitting coefficients or parameters
- How many degrees of freedom to use?
  - The minimum needed! (Occam's razor)
  - Sensitivity analysis
  - Confounding in nonlinear systems
- Orthogonal experiments
- Training vs. validation data set



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2. Fundamentals of modeling

Complexity vs. Simplicity

- Complex system = more than the sum of its parts
- Paul Valéry's conundrum: "Everything simple is false. Everything complex is unusable."
- George Box's resolution: "All models are wrong, but some are useful."
- John Hedengren: "A model is not a set of equations; it is a set of assumptions."

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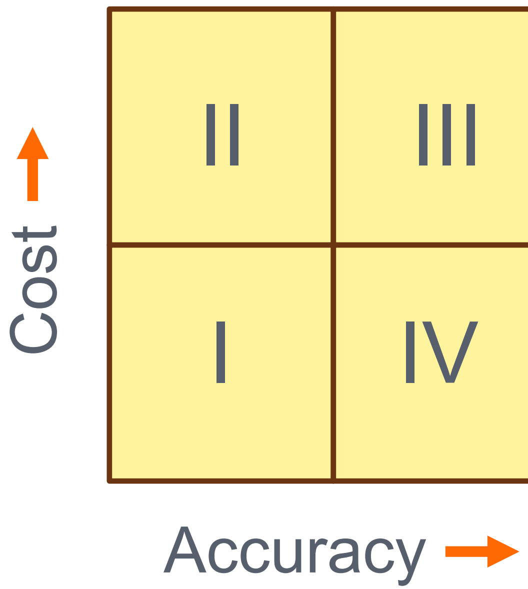
2. Fundamentals of Modeling

All Models Are Wrong…

- Is the wrong part important or not? PV=nRT
- Be a skeptic of your own model: what are its limits?
- Where do we want to be on this plot?



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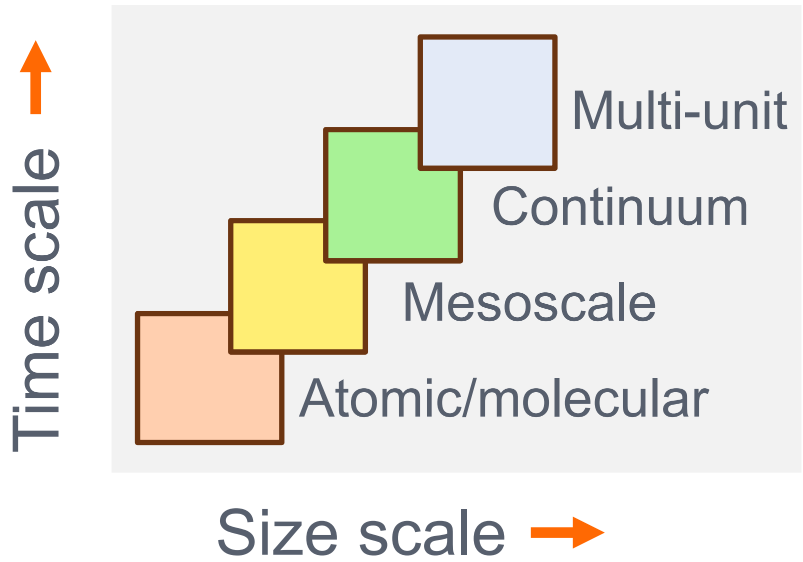
2. Fundamentals of modeling
Types of electrochemical models



Applications
- Sensors
- Corrosion
- Electroplating
- Batteries
- Fuel cells
- Chemical synthesis
- Biological systems
- Colloids

Which of these systems are of most interest to you?

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Lesson Plan

1. Introductions
2. Fundamentals of modeling
3. Particle-based models

Linear and nonlinear phenomena

Porous electrode battery models

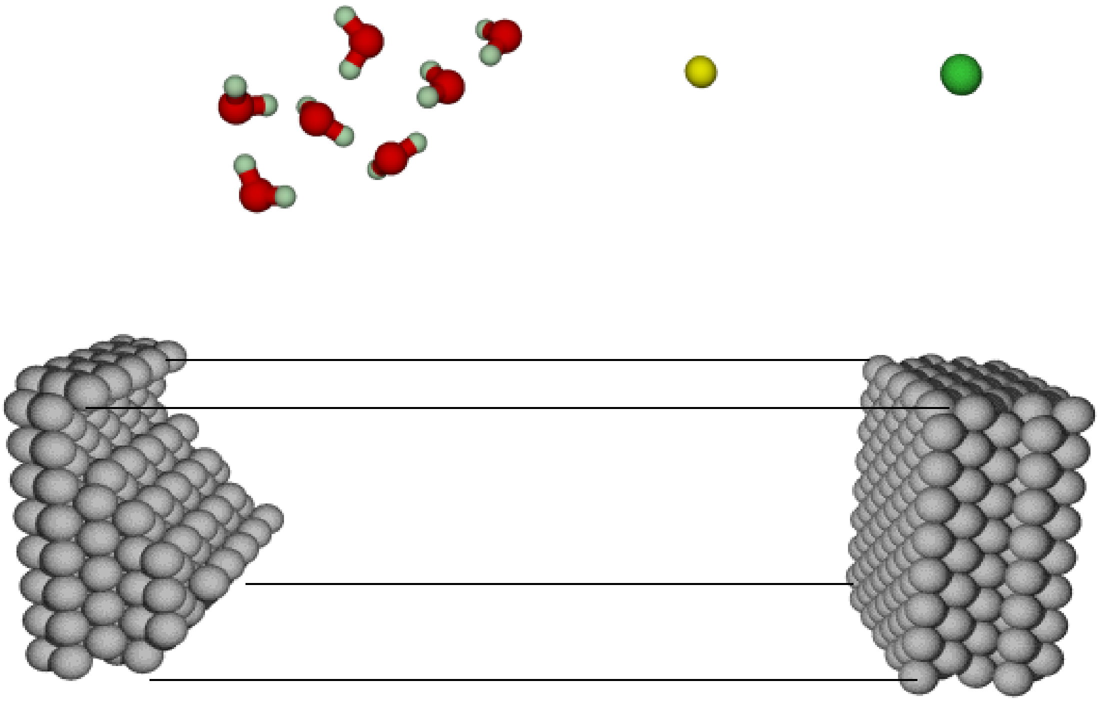
3. Particle-based models

A. Molecular dynamics simulations of the electrochemical double layer

Solve

$$\ddot{\mathbf{r}}_i = \frac{\mathbf{F}_i}{m_i}$$

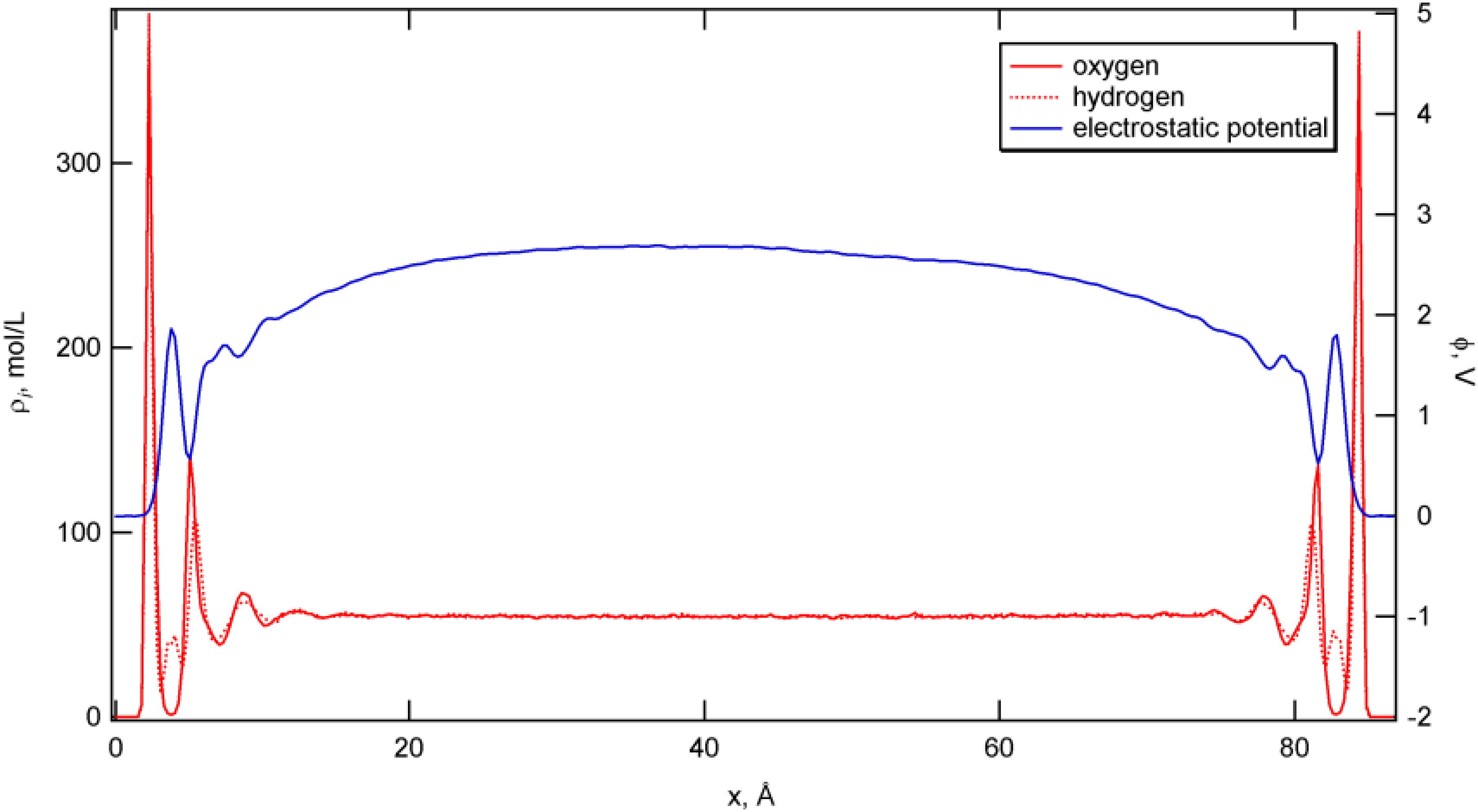
for 2500 H_2O 80 Na^+ 80 Cl^-



2700 Cu

Guymon et al., *Cond. Matt. Phys*. 8, 335-356 (2005).
Guymon et al. *J. Chem. Phys.* 128, 044717 (2008)

PURE WATER ON FLAT ELECTRODES: POTENTIAL AND DENSITY PROFILES



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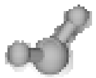
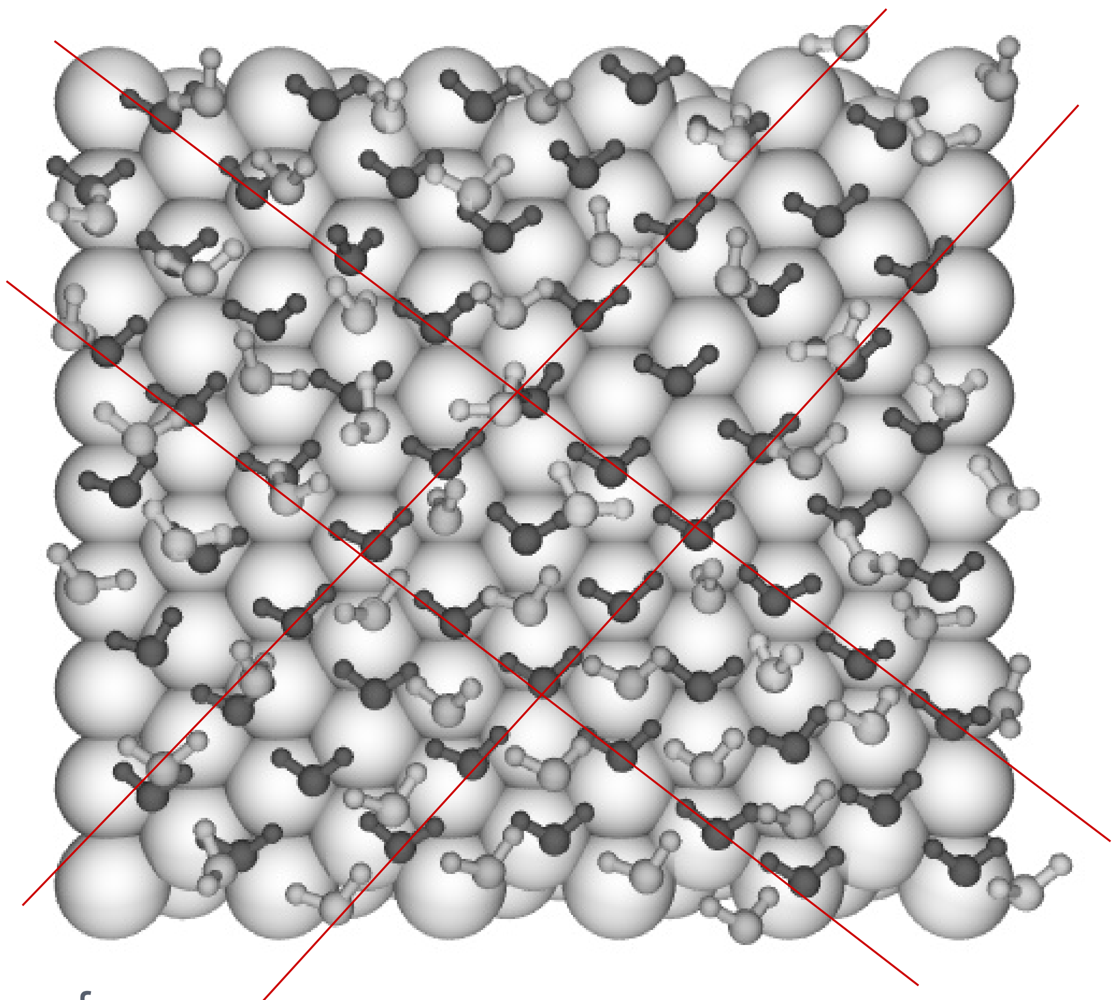
WATER LAYERS ON CU(111)

1st Layer

2nd Layer





neutral surface

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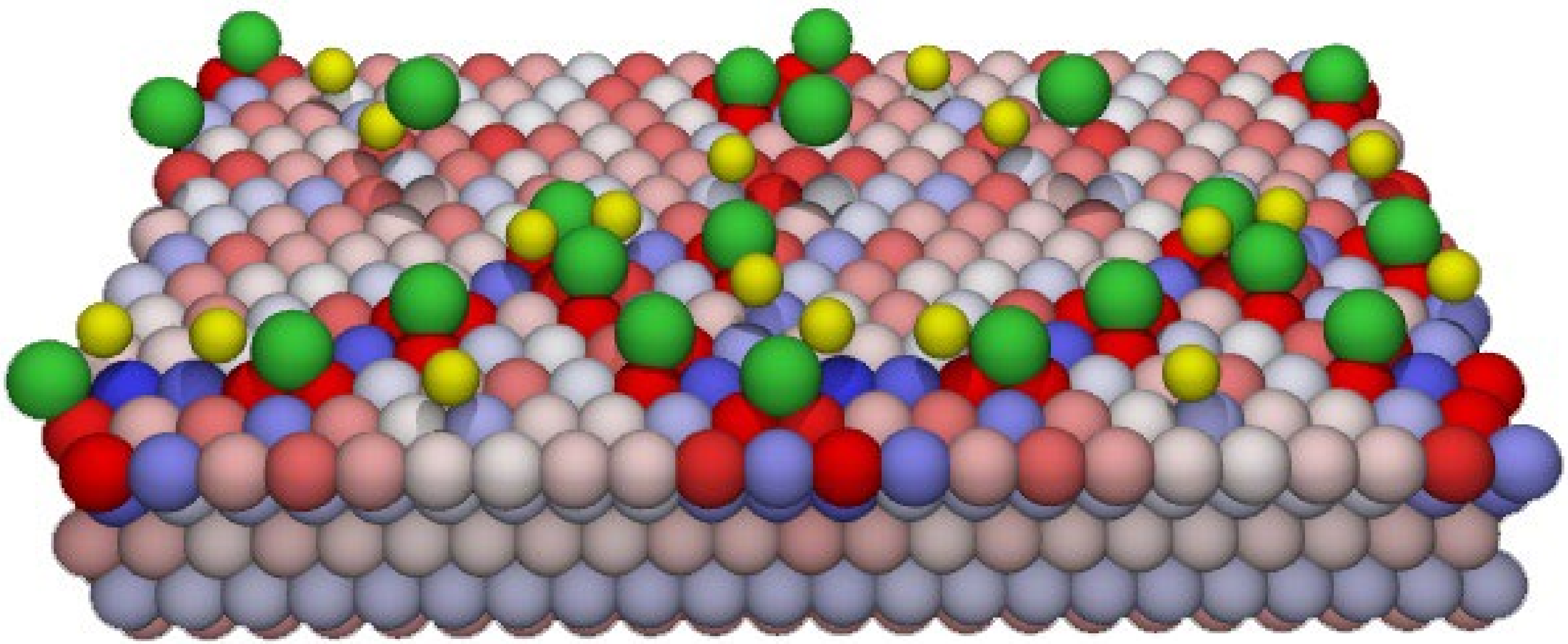
Flat electrode



Na^+ Cl^-

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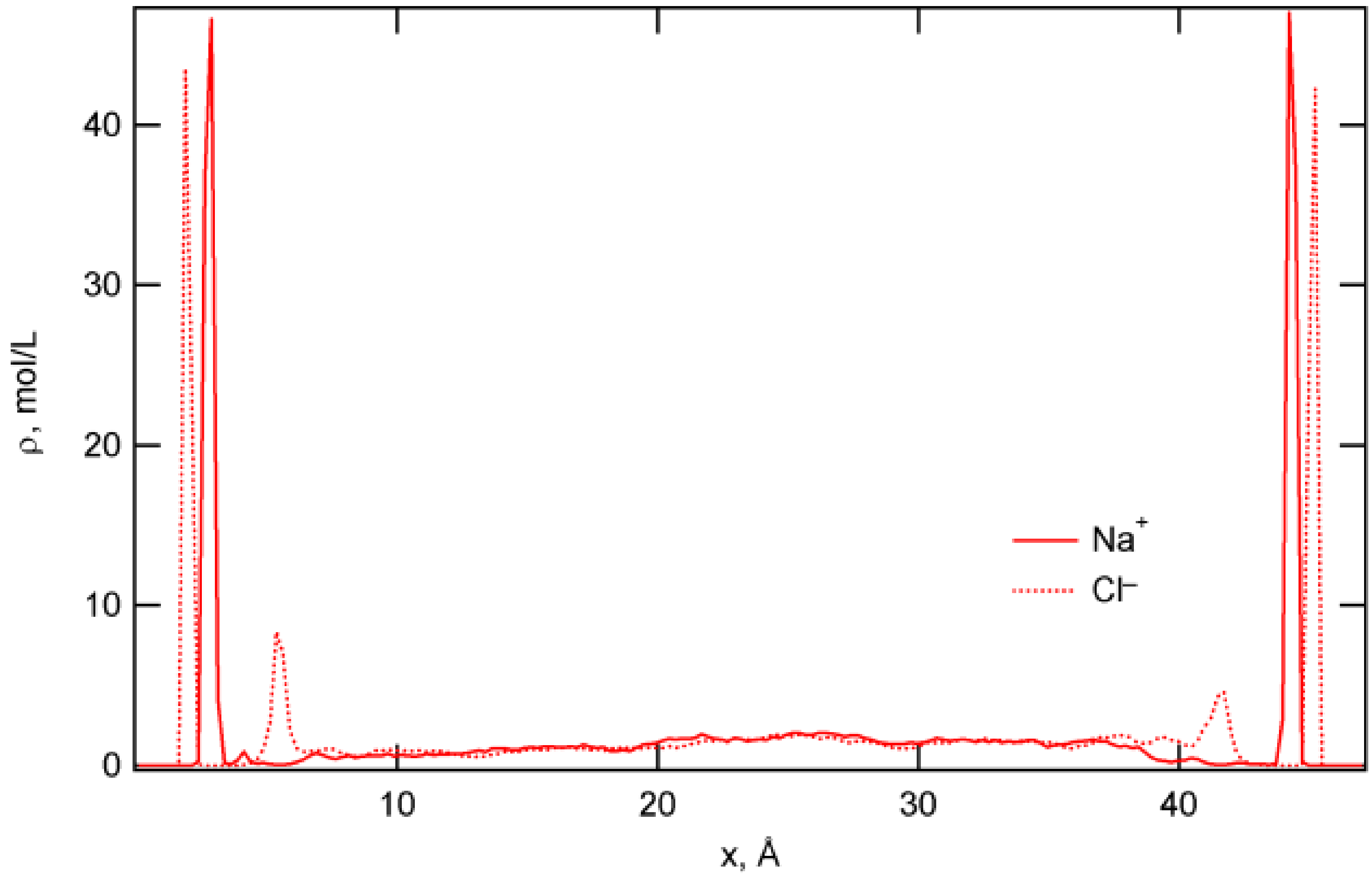
Ion density profiles for symmetric system



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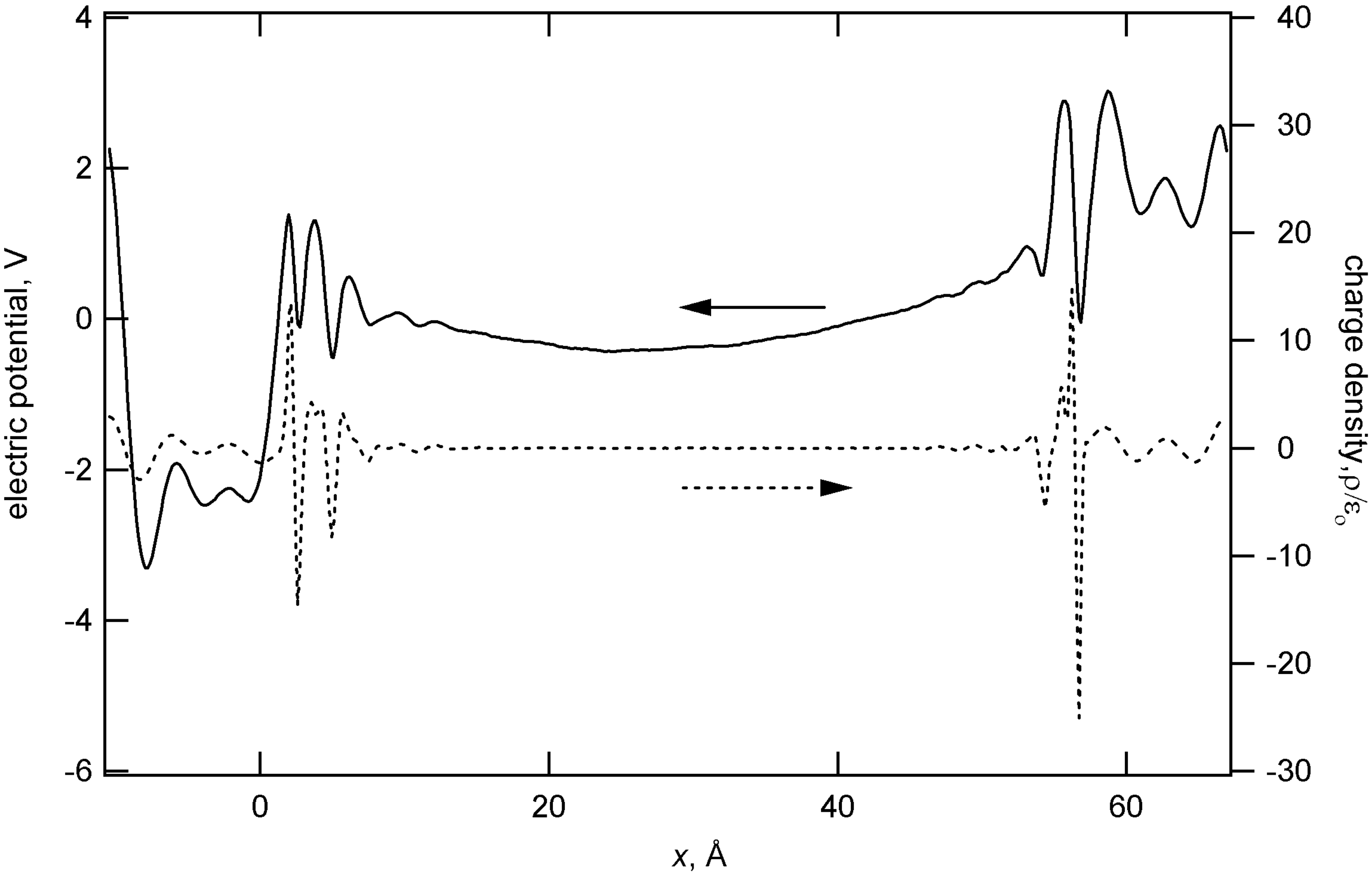
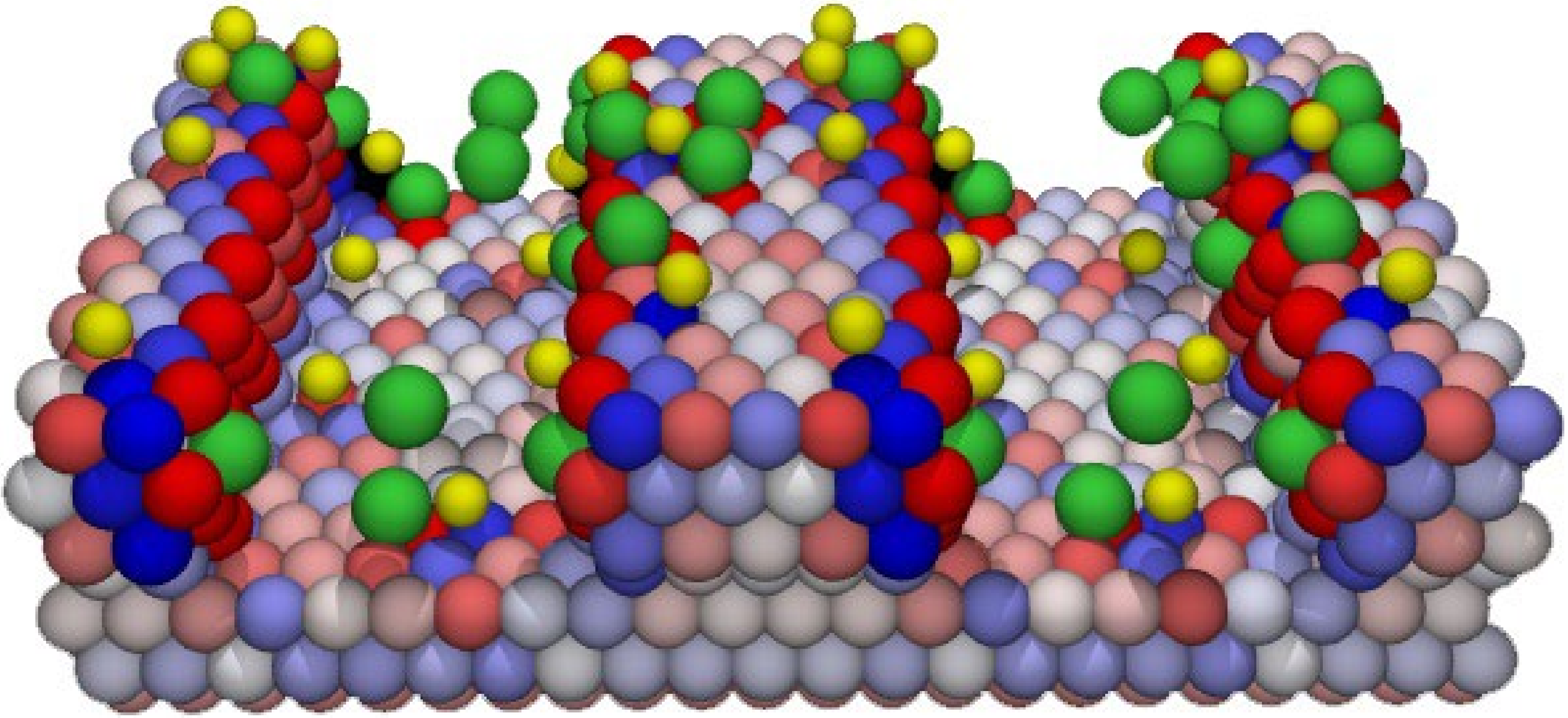
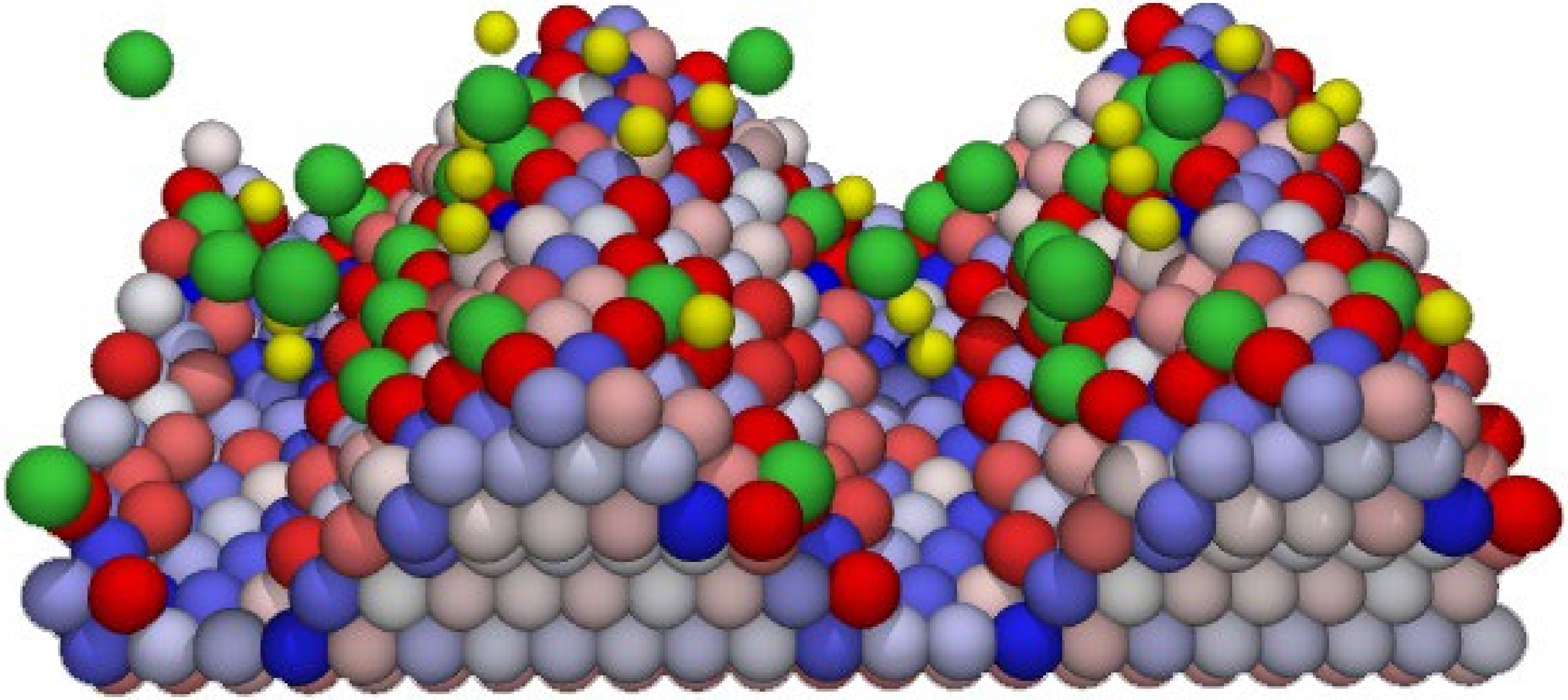
FLAT ELECTRODES, ΔV=3.6V



STEPPED ELECTRODE



SAWTOOTH ELECTRODE



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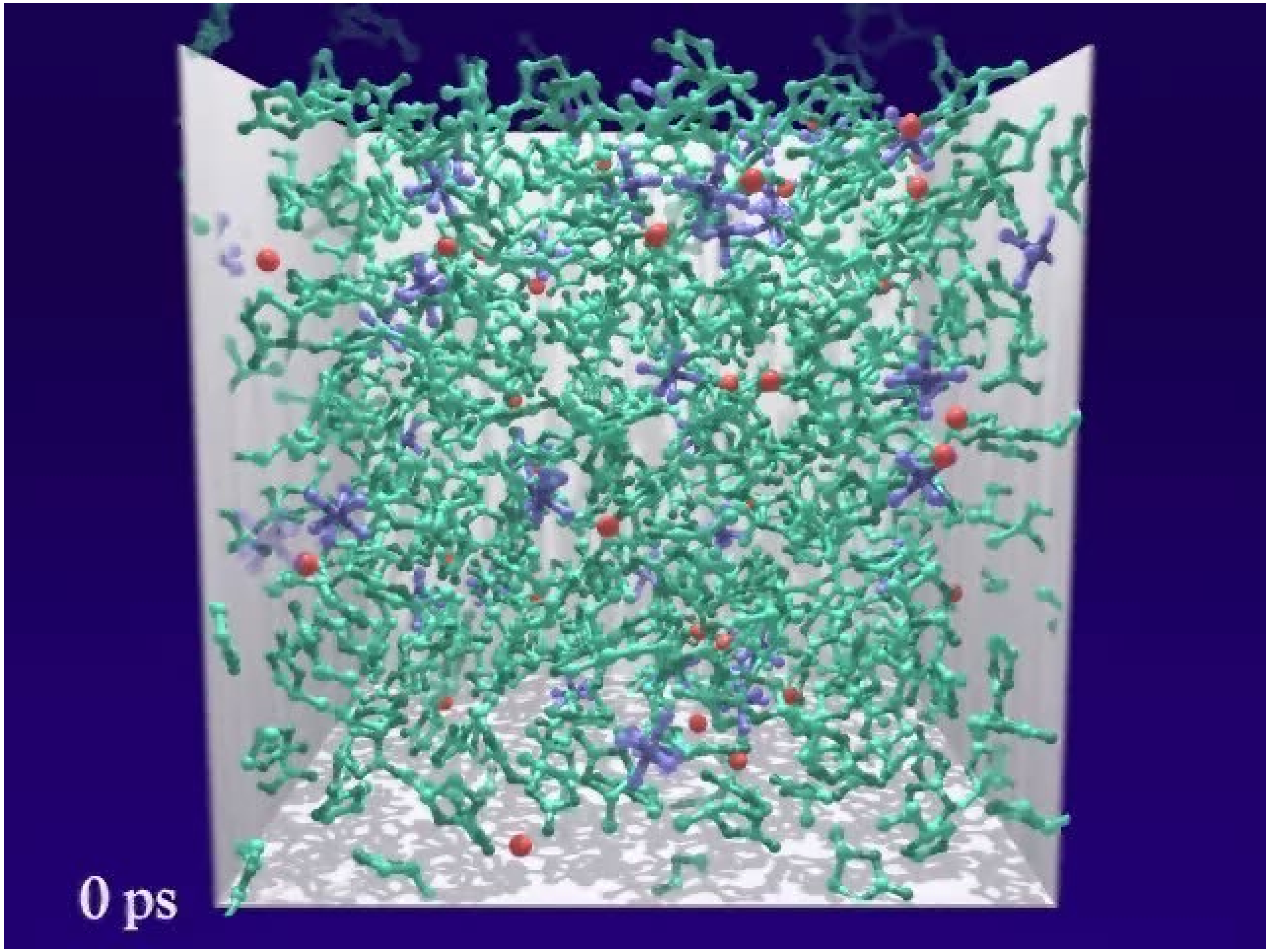
3. Particle-based models

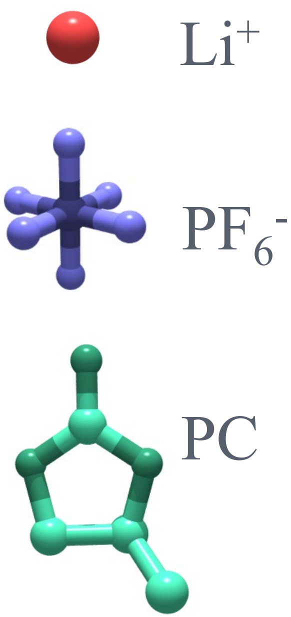
B. Molecular dynamics simulations of diffusion processes in electrolyte

- You compute all positions, velocities, and forces for a few thousand molecules/ions
- Can get multiple thermodynamic and transport properties from one simulation, if you have the right algorithms
- Forcefield must be accurate if want to match to experiment

Wheeler and Newman, J. Phys. Chem. B 108, 18362-18367 (2004).
Wheeler and Newman, J. Phys. Chem. B 108, 18353-18361 (2004).
Newman at al., J. Power Sources 119, 838-843 (2003).

1M $LiPF_6$ in PC





Watch the dynamics of the solvent and ions in this equilibrium simulation

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Macroscopic Framework: Concentrated Solution Theory

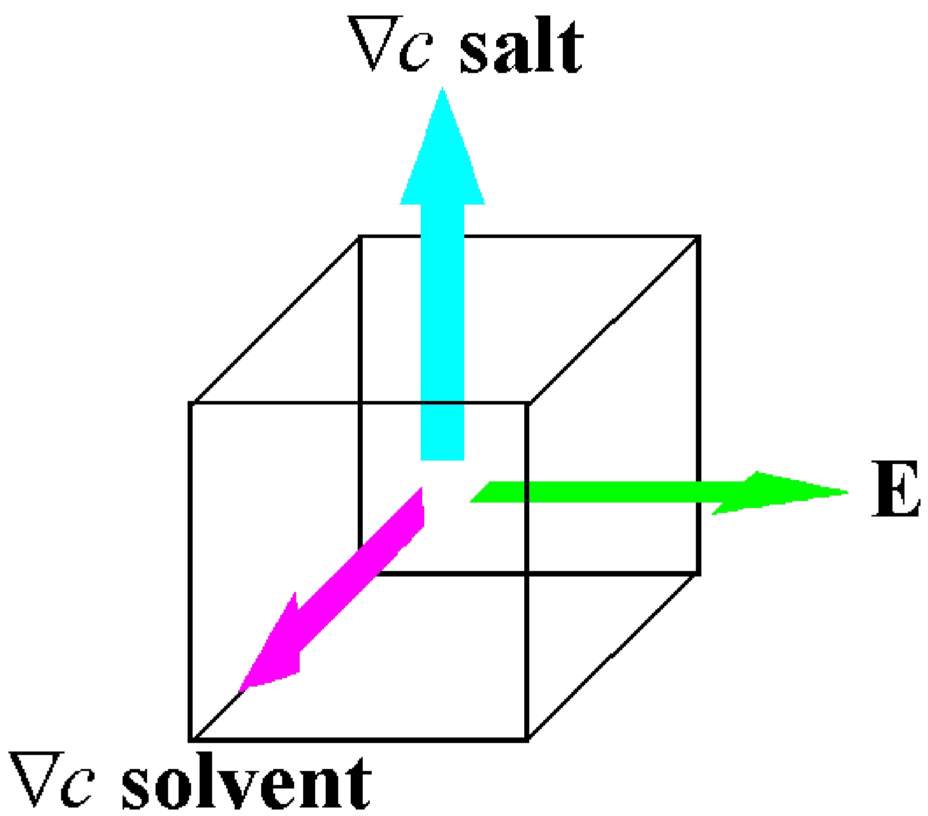
- *n*(*n*-1)/2 independent transport coefficients for an isothermal *n*-component system.
- Three equivalent ways to represent mass transport coefficients.

e.g. for a binary salt (**+** -) in solvent (0):

$$\boxed{\mathcal{D}_{0+} \quad \mathcal{D}_{0-} \quad \mathcal{D}_{+-}} \qquad \boxed{L^{0}_{++} \quad L^{0}_{--} \quad L^{0}_{+-}} \qquad \boxed{\mathcal{D}_{salt} \quad \kappa \quad t^{0}_{+}}$$

Nonequilibrium MD allows simultaneous Orthogonal ‘Experiments’

Body forces placed on particular speces



One simulation can resolve up to 6 pairwise diffusion coefficients (n = 4).

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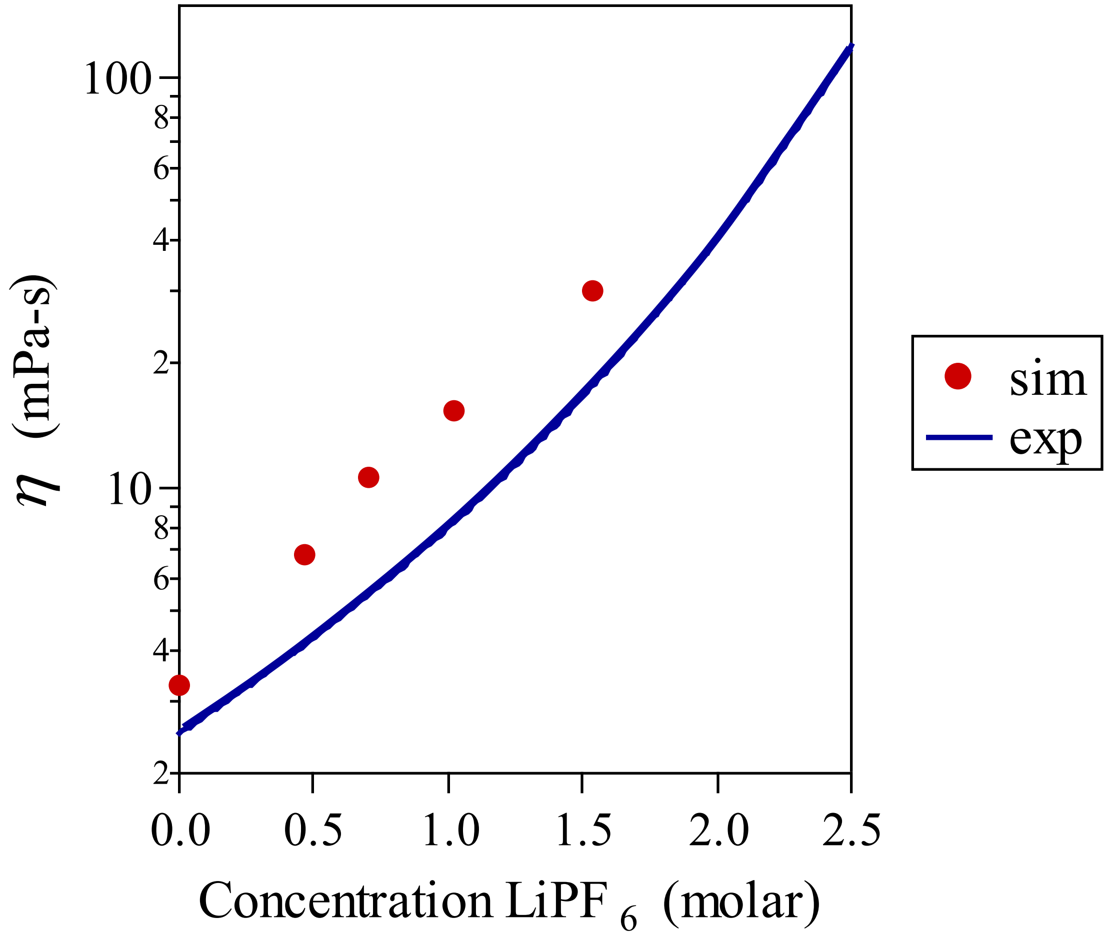
Viscosity
$LiPF_6$ in PC



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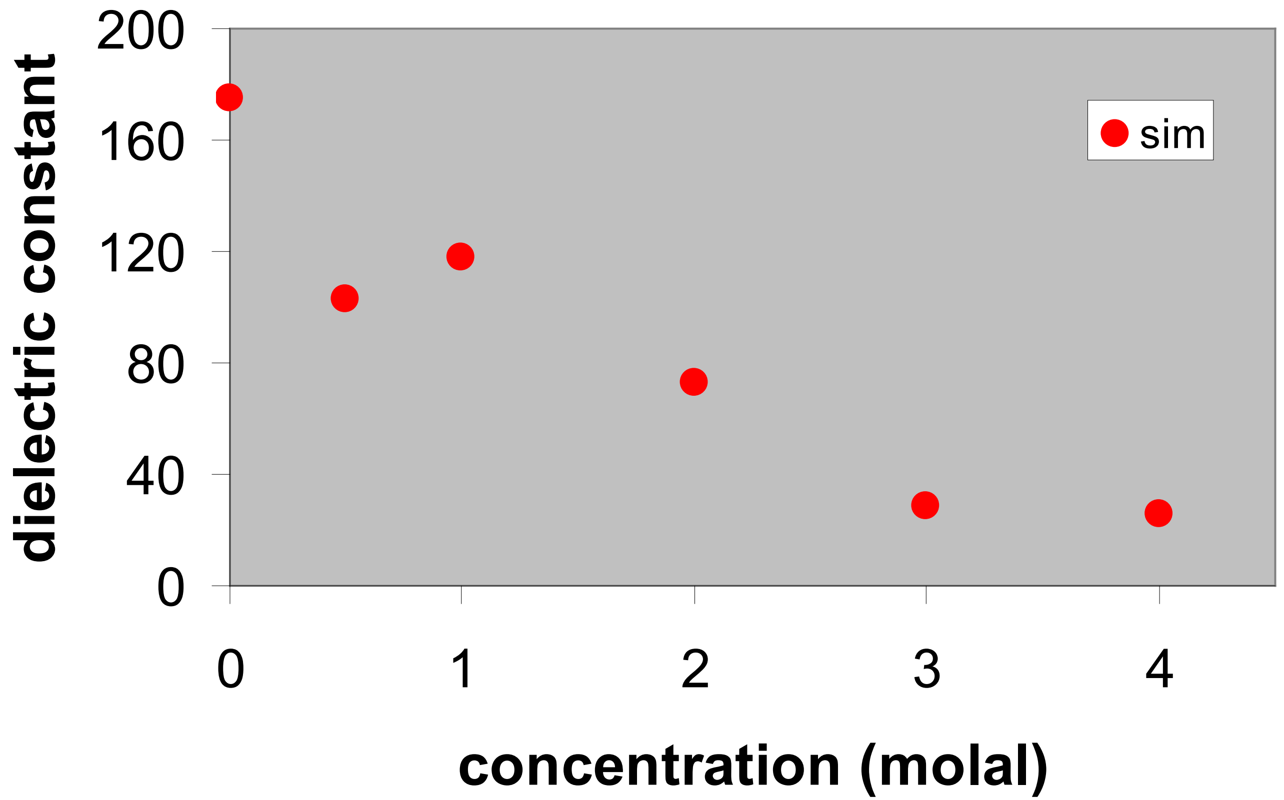
Dielectric Constant: $LiPF_6$ in EC



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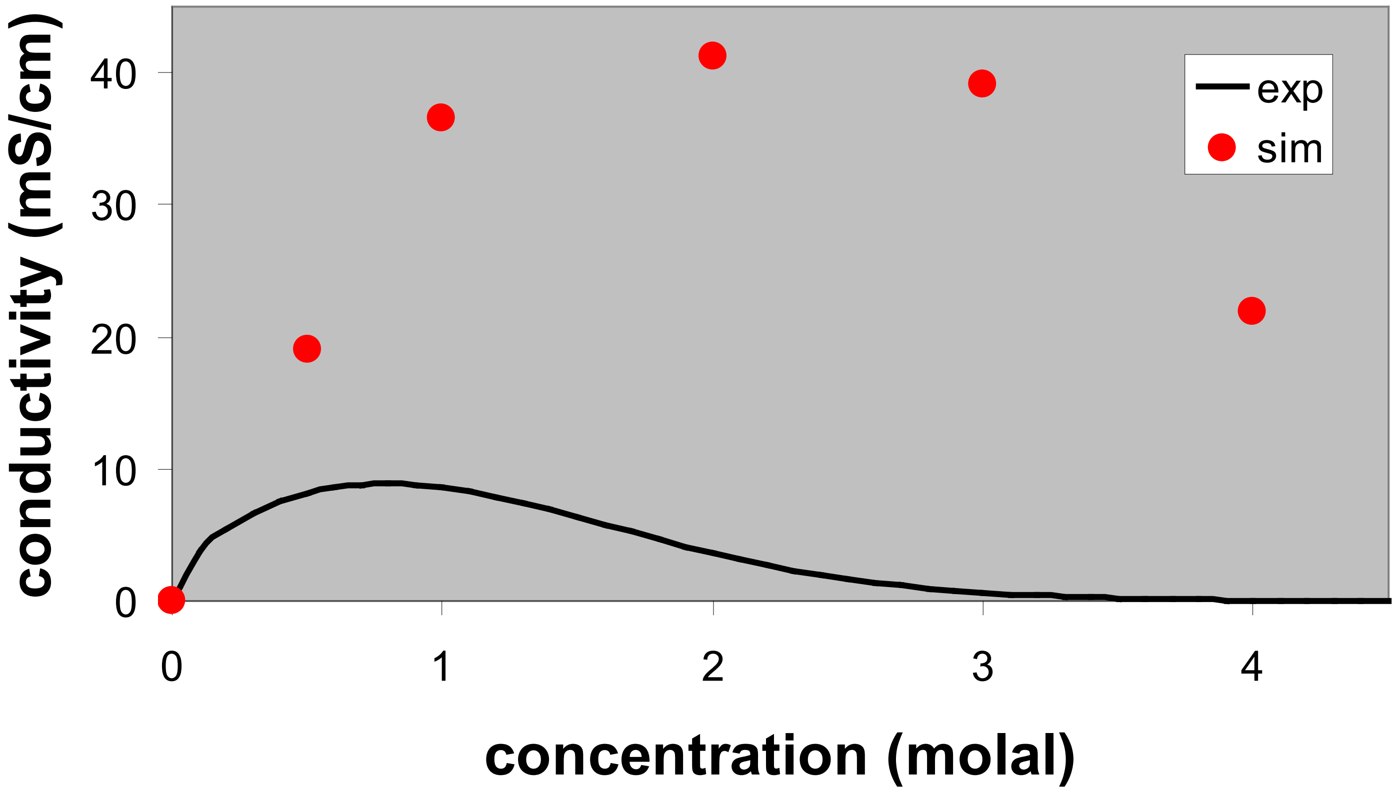
Electrical Conductivity
$LiPF_6$ in EC



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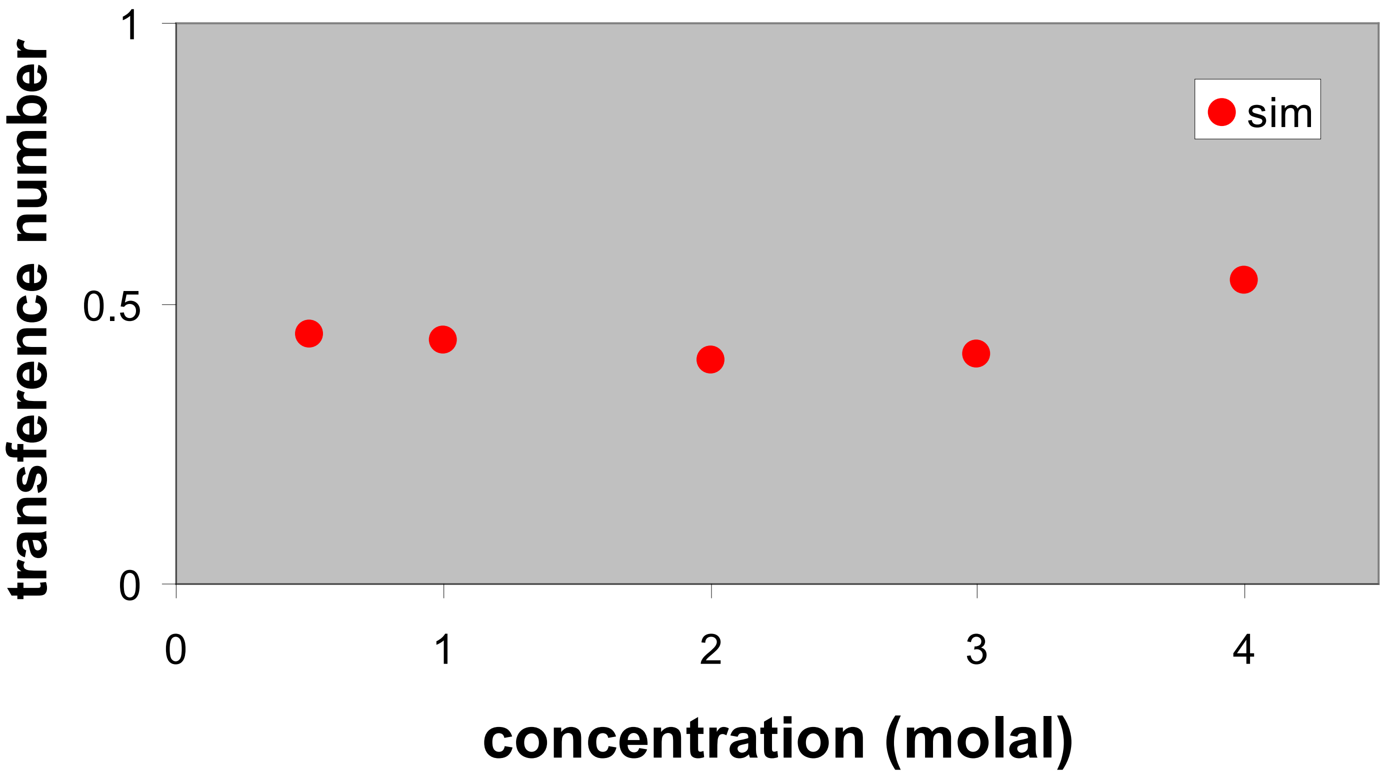
Cation Transference Number
$LiPF_6$ in EC



experimental $t_+^0 \approx 0.41$

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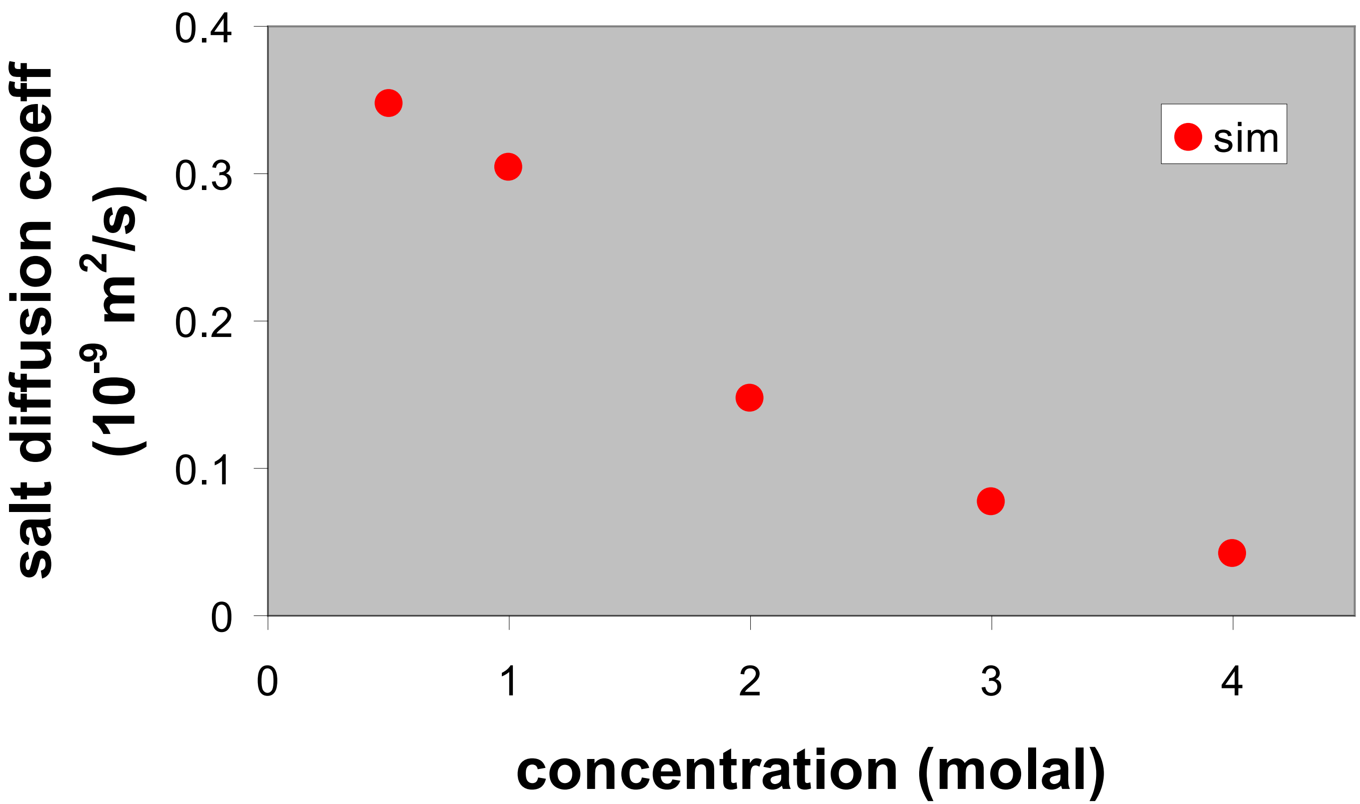
Salt Diffusion Coefficient: $LiPF_6$ in EC



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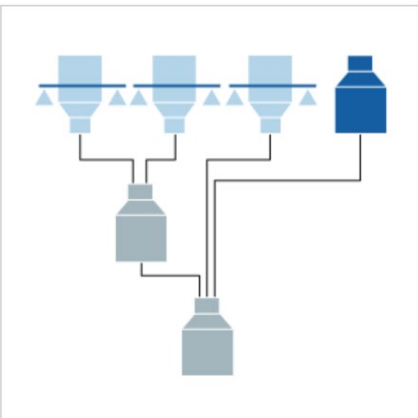
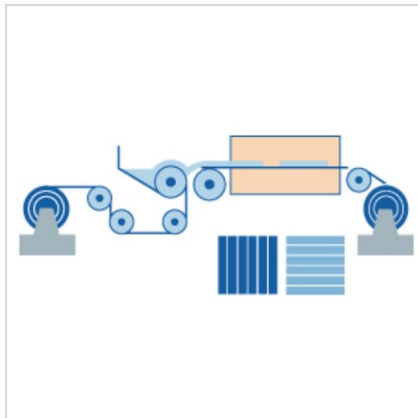
3. Particle-based Models

C. Simulating Microstructure During Electrode Manufacturing

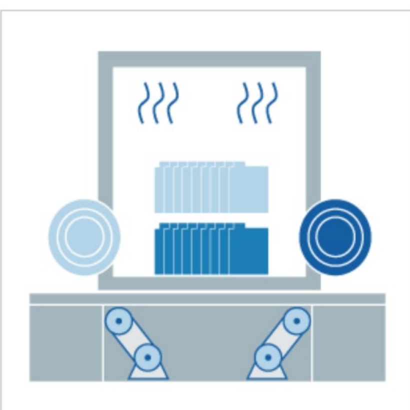
Mixing



Coating



Drying



Calendering

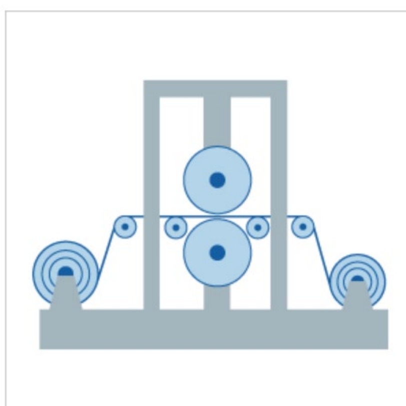


Diagram from w3.siemens.com

- Forouzan et al., J Power Sources 312, 172 (2016)
- Nikpour et al. J Electrochem Soc, 168, 060547 (2021)
- Nikpour et al. J Electrochem Soc, 168, 120518 (2021)
- Nikpour et al., Batteries, 8, 107 (2022)

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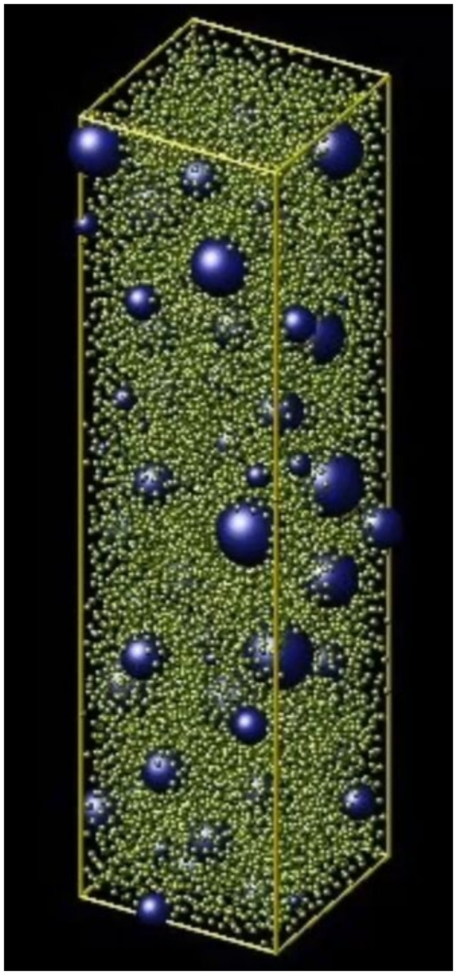
Discrete element method (DEM) model of drying process

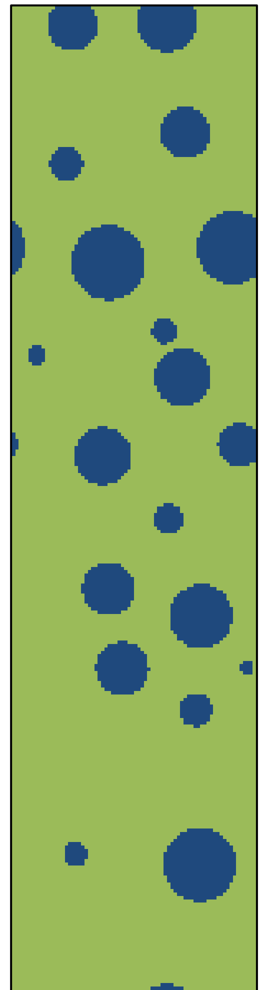


Blue = active matl

Green = CBD, solvent

White = macropore

Liquid simulation

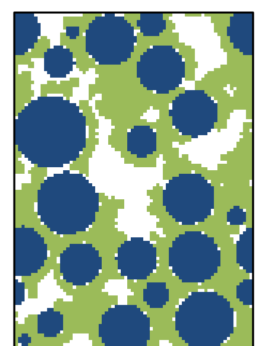


Toda NMC523 cathode

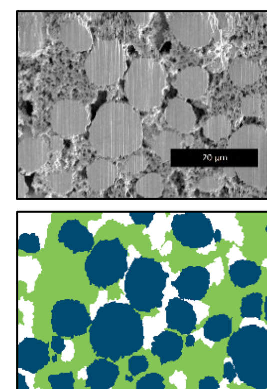
Volume Fractions of Active Film

Domain	SEM/FIB	Sim
Active	0.443	0.402
CBD	0.367	0.376
Macropore	0.190	0.222

Dried film simulation



Experimental dried film



Forouzan et al., J Power Sources 312, 172 (2016)

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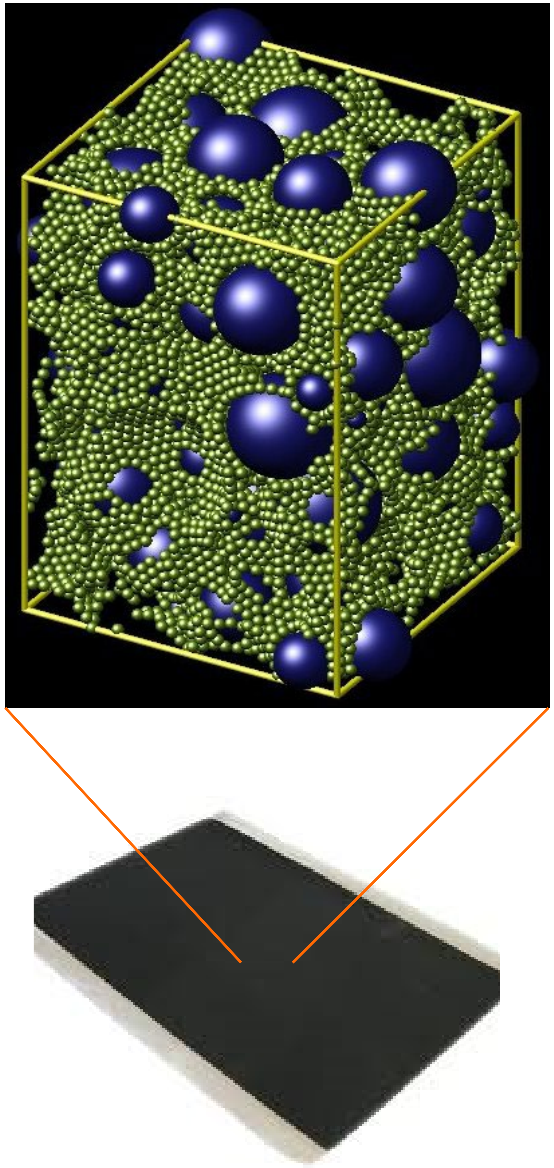
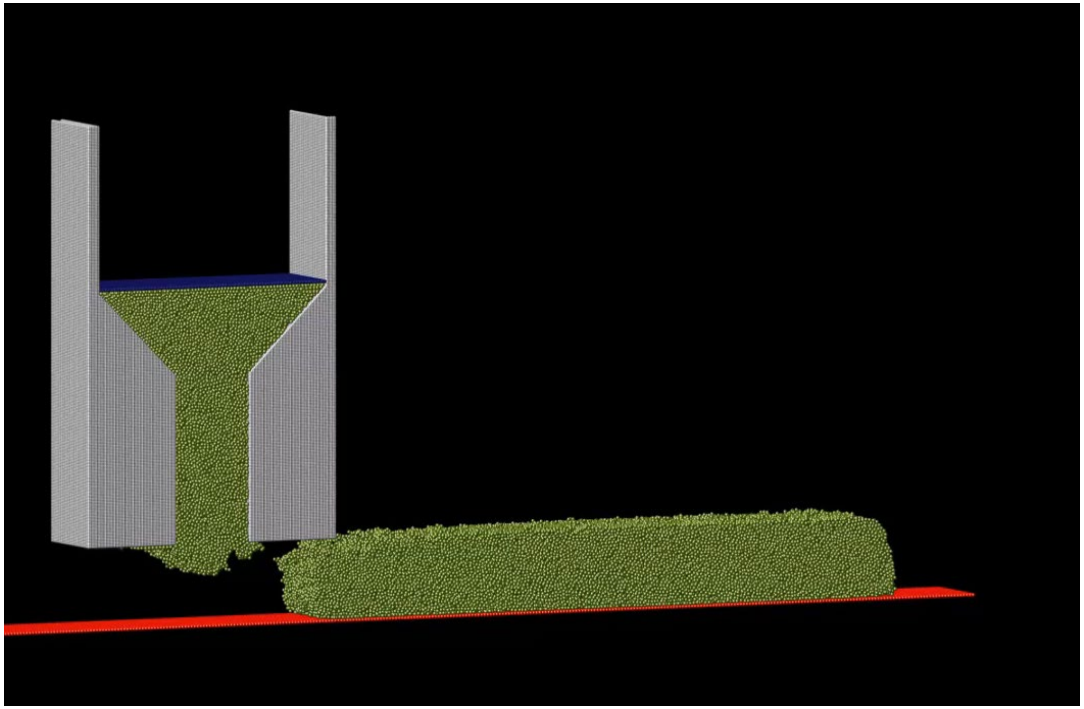
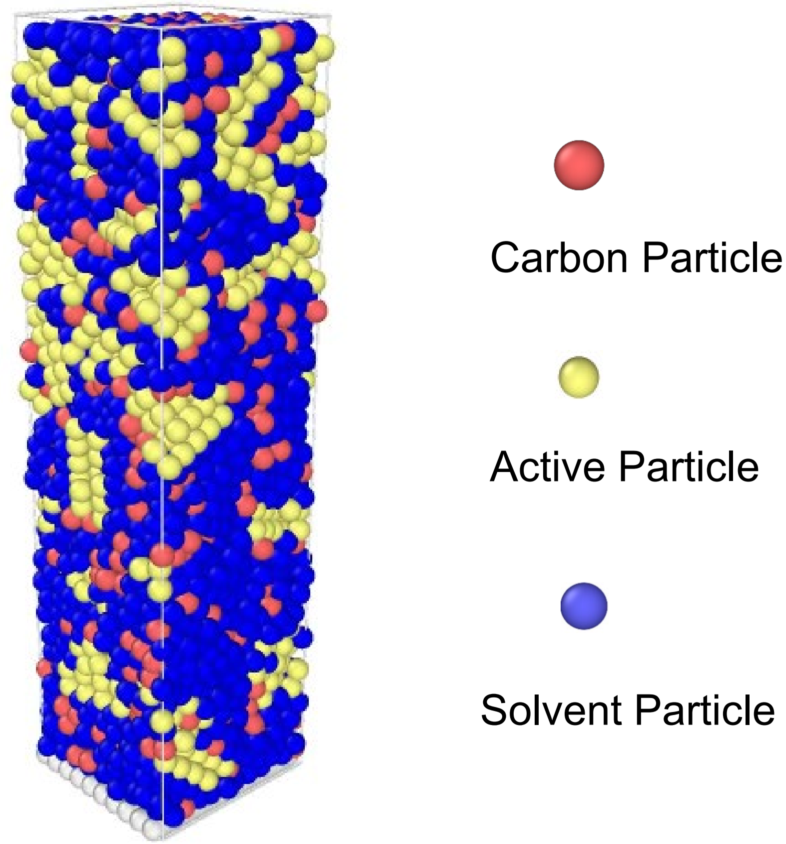
FILM COATING SIMULATIONS





FILM DRYING SIMULATIONS WITH 3 PARTICLE TYPES



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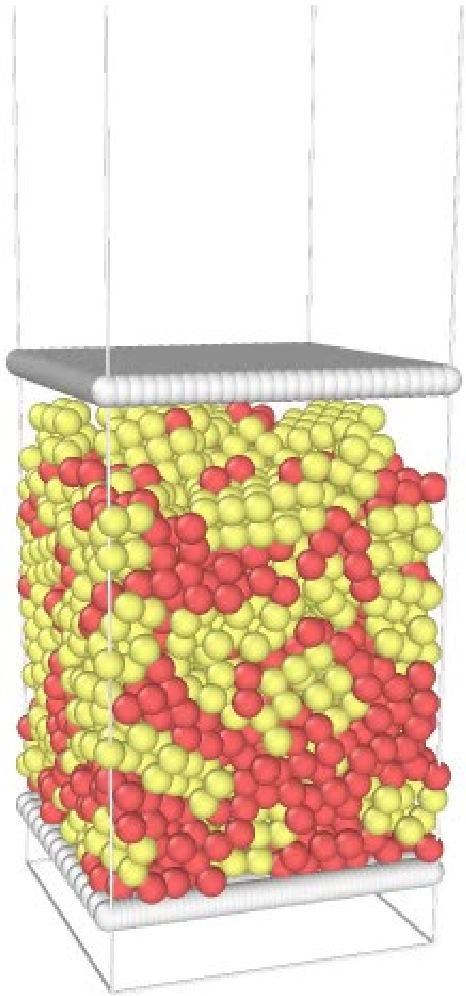
Film Calendering Simulations




Carbon Particle


Active Particle

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Break Time

We will start again at 14:35

LESSON PLAN

1. Introductions
2. Fundamentals of modeling
3. Particle-based models

Linear and nonlinear phenomena

4. Linear equivalent-circuit models
5. Nonlinear phenomena
6. Battery background

Porous electrode battery models

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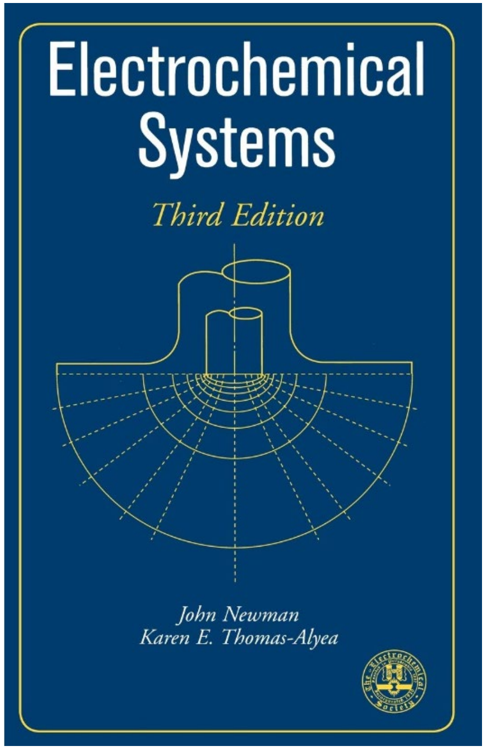
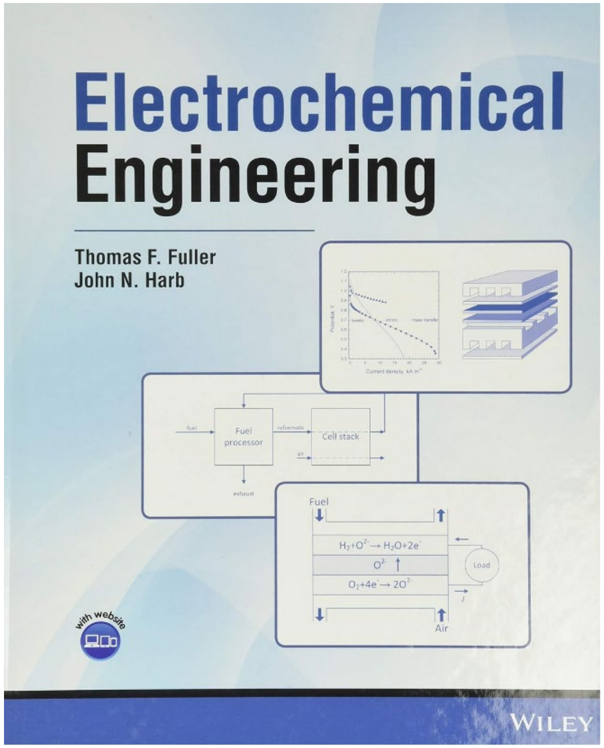
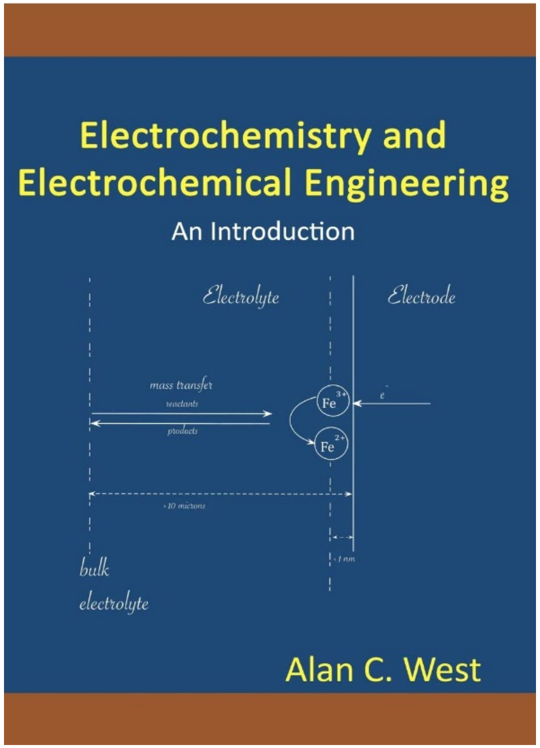
Some Resources

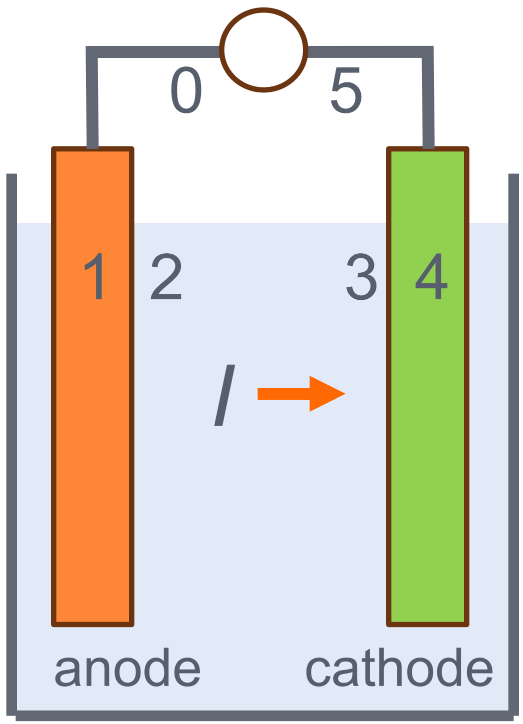






4. EQUIVALENT-CIRCUIT MODELS

- Electrochemical cell = electrical circuit
- Ions act like electrons, but with differing charge



Numbers indicate positions in electrode or electrolyte

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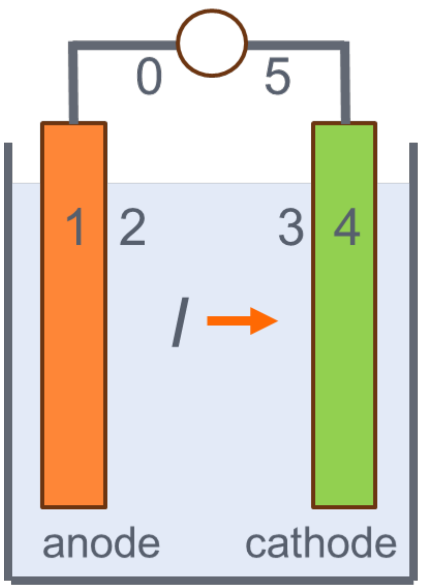
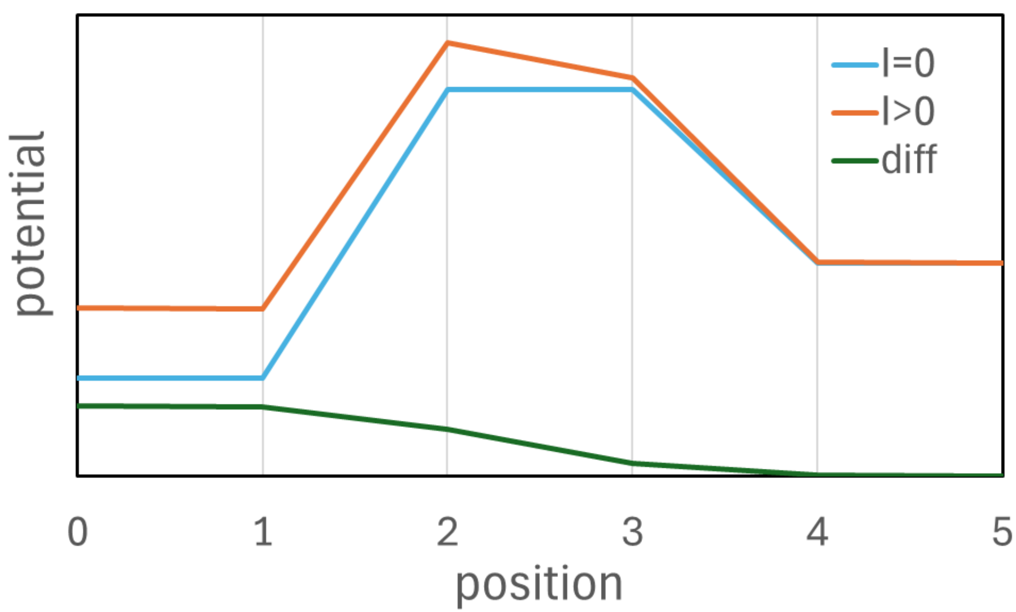
4. EQUIVALENT-CIRCUIT MODELS

- See if you can rationalize the relative potentials at each position
- Is this an electrolytic (needs power) or galvanic (battery) cell?

Here is the galvanic cell





The *diff* curve subtracts the open circuit potential (*I*=0) from the other curve (*I*>0)

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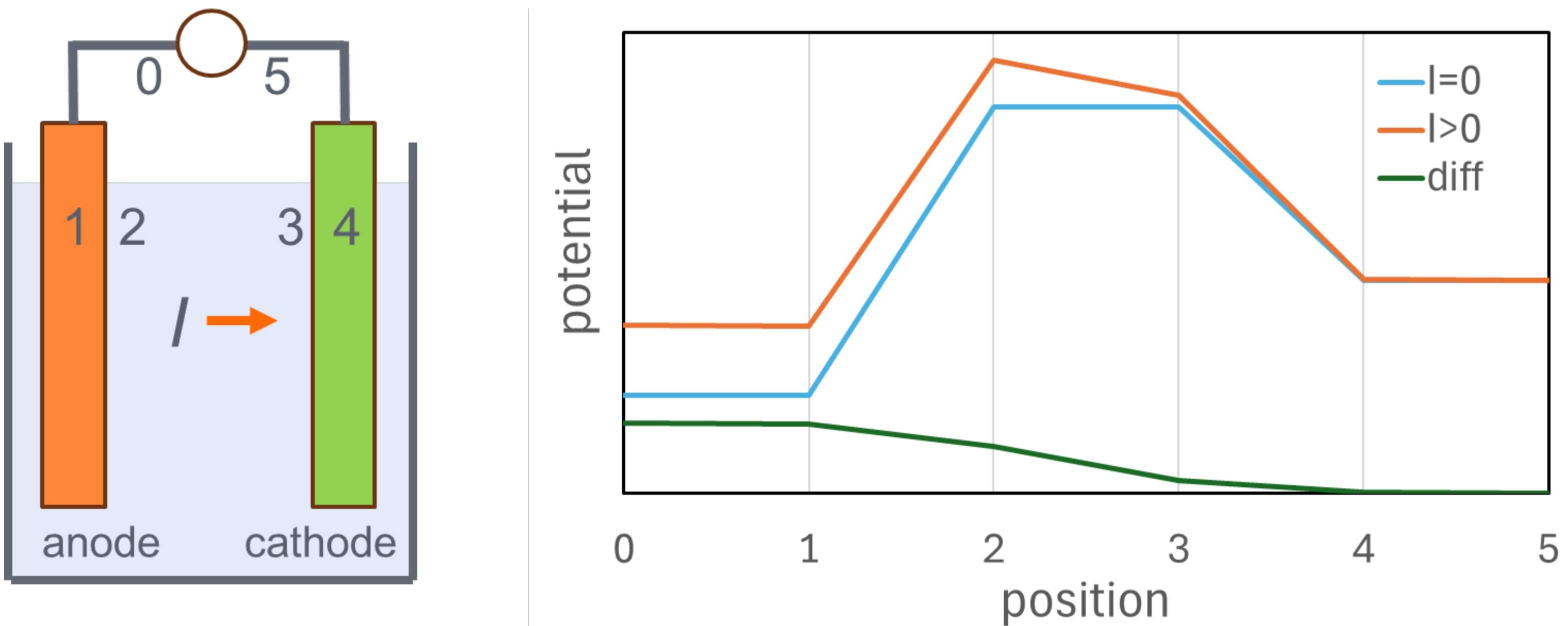
4. EQUIVALENT-CIRCUIT MODELS

- The system can be described as a voltage source and a series of resistors *
- Which resistors are least/most significant?



*with parallel capacitors

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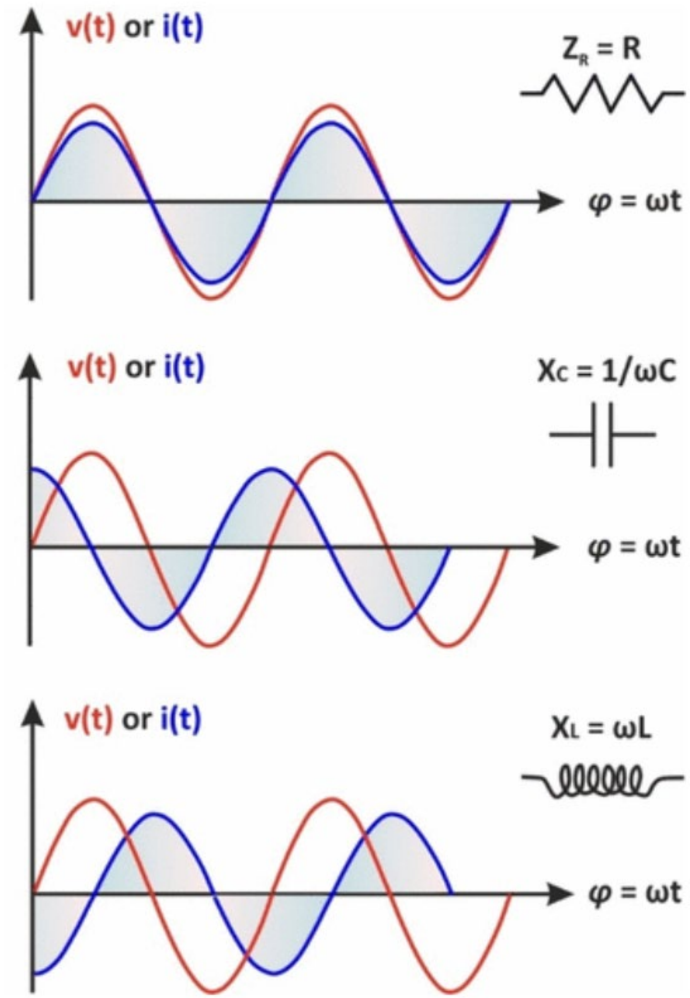
4. EQUIVALENT-CIRCUIT MODELS

EIS

- Electrochemical Impedance Spectroscopy (EIS) = AC signal across cell
- Randles cell model



- $Z = R_1 + \frac{1}{\frac{1}{R_2} + j\omega C}$ is impedance, a complex resistance

Lazanas, ACS Meas Sci Au 2023 3, 162.

doi.org/10.5796/electrochemistry.22-66071

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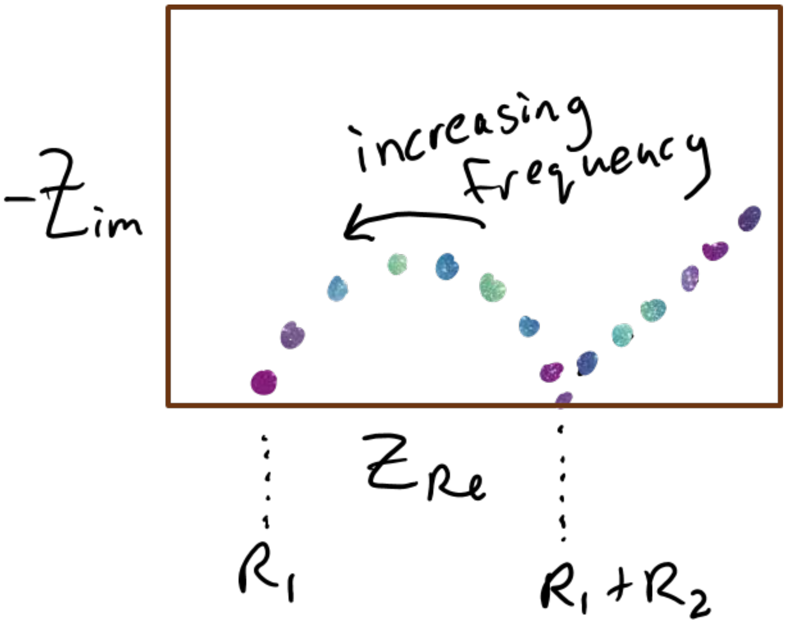
4. EQUIVALENT-CIRCUIT MODELS

EIS

- Nyquist plot is often used to describe response of the system to an EIS test with multiple frequencies (around 1 mHz to 1MHz)



- Equivalent circuits can get a lot more complicated to describe more complicated conditions or results

EQUIVALENT CIRCUIT EXAMPLE: BLOCKING ELECTROLYTE METHOD FOR MEASURING ELECTRODE TORTUOSITY

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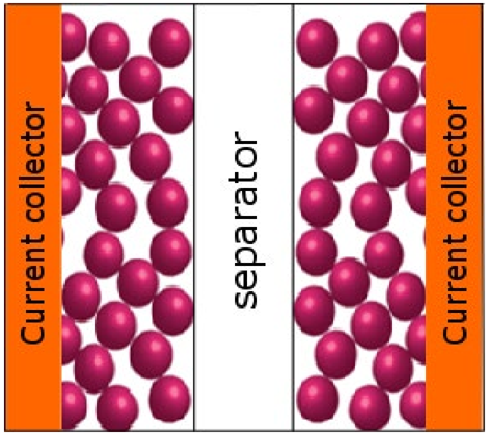
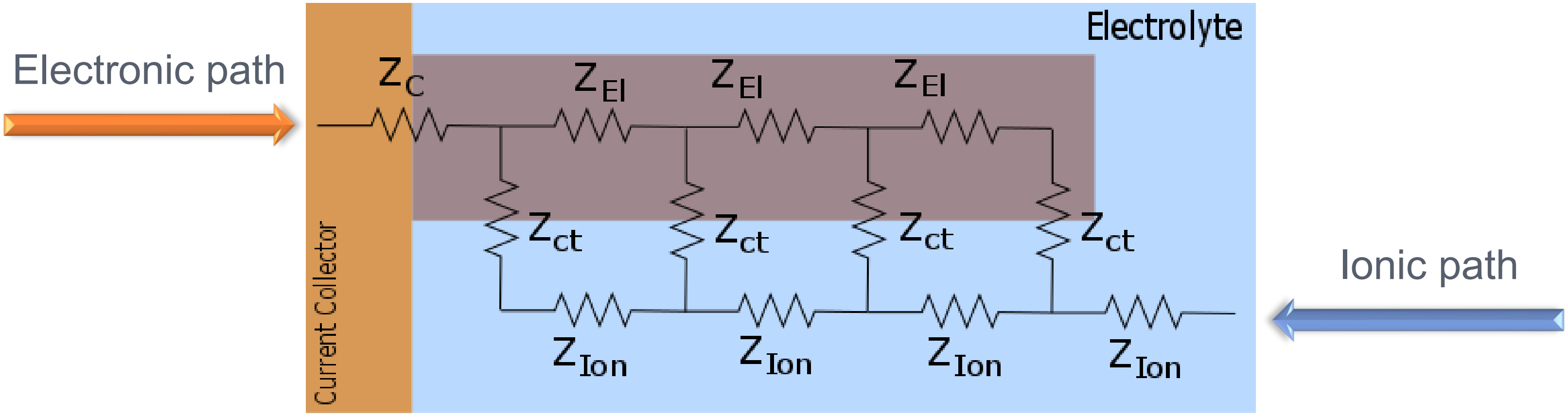
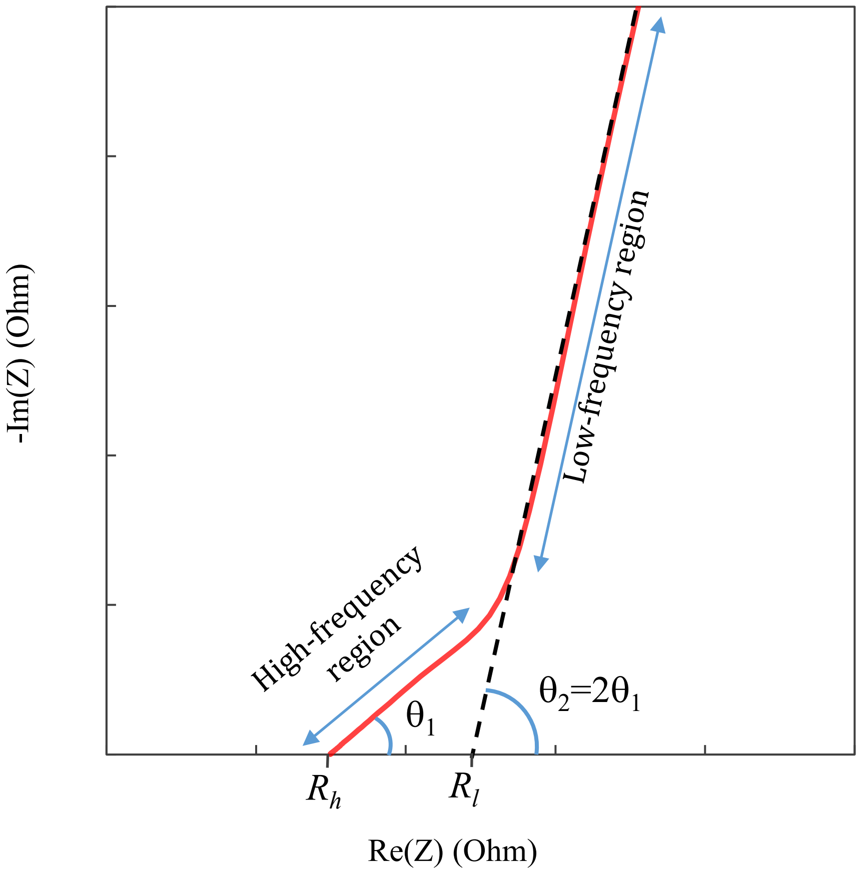


Pouraghajan et al., J Electrochem Soc 165, A2644 (2018).
Pouraghajan et al., J Power Sources 492, 229636 (2021)
Liu et al., J Electrochem Soc 169, 010517 (2022)

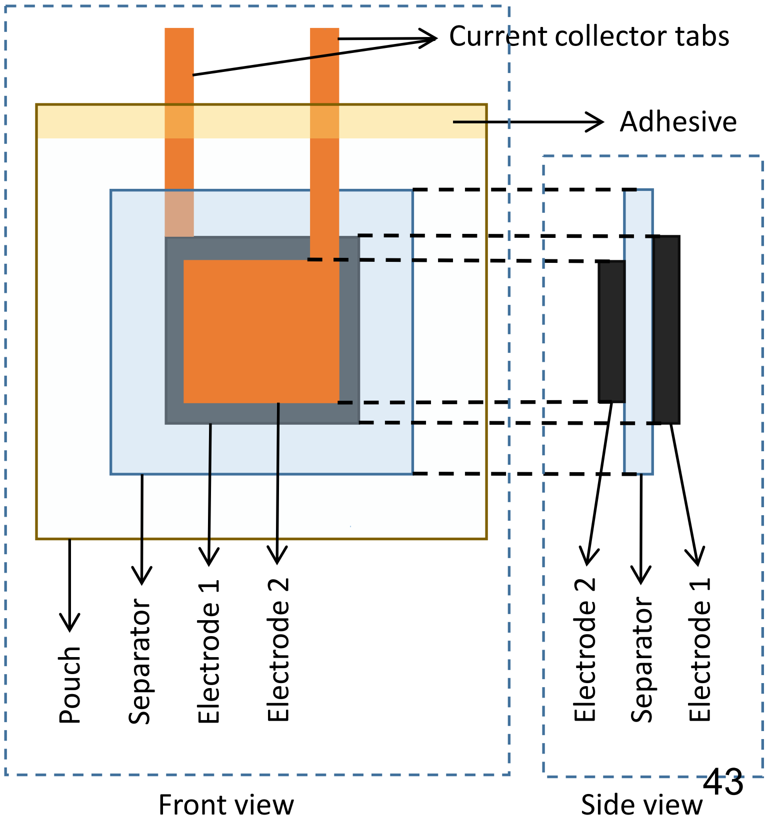
- A blocking electrolyte (no Li+) stops the faradaic reaction
- For porous electrodes, the EC has two or more "rails" like this and is called a transmission line model (TLM)



BLOCKING ELECTROLYTE METHOD



The Nyquist plot is analyzed to obtain Ionic resistance, a value proportional to tortuosity.



$$R_h - R_l \sim \frac{R_{\text{ion}}}{3}$$

$$R_{\text{ion}} = \frac{L}{k_{eff} \cdot A}$$

$$\frac{k}{k_{eff}} = \frac{\tau}{\epsilon} = \frac{R_{\text{ion}} \cdot A \cdot k}{L}$$

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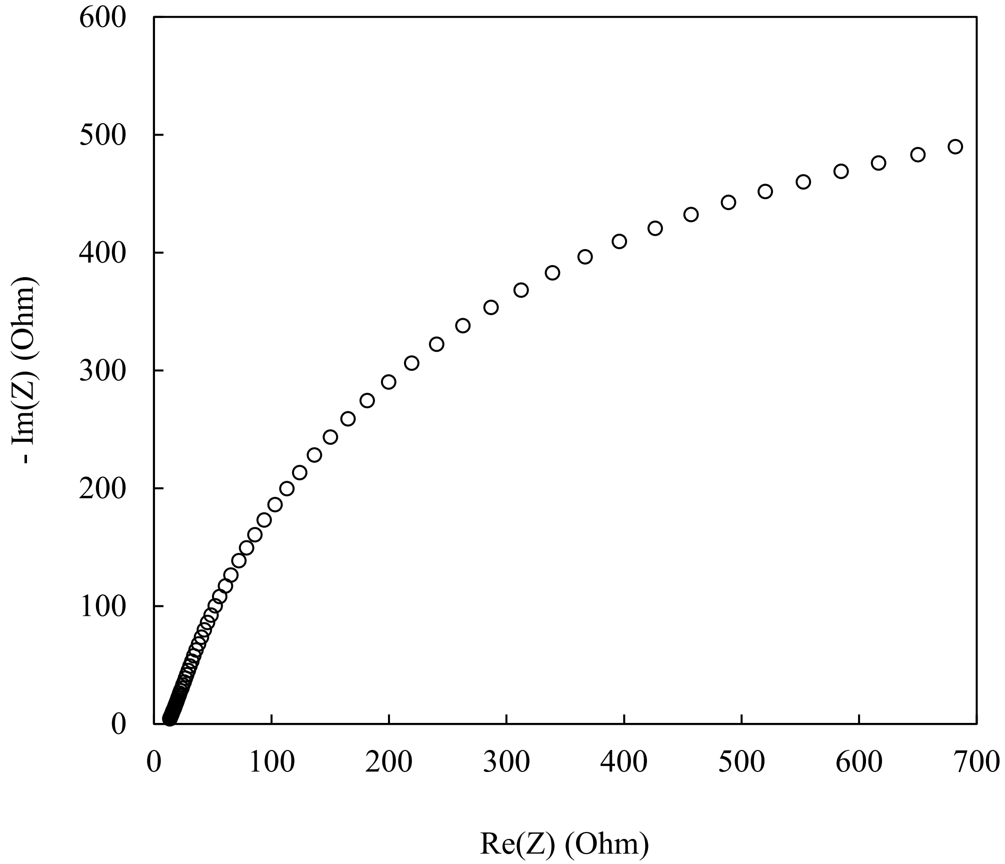
BLOCKING-ELECTROLYTE CHALLENGES



Nyquist impedance plot of blocking electrolyte method for cycled anode.

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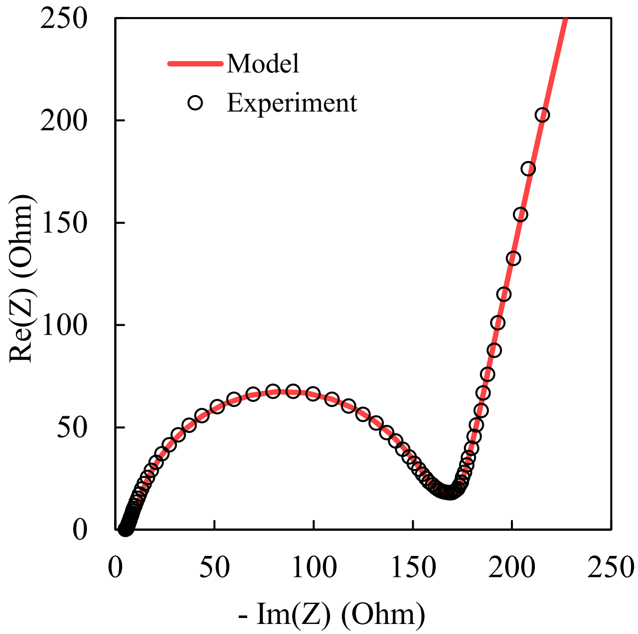
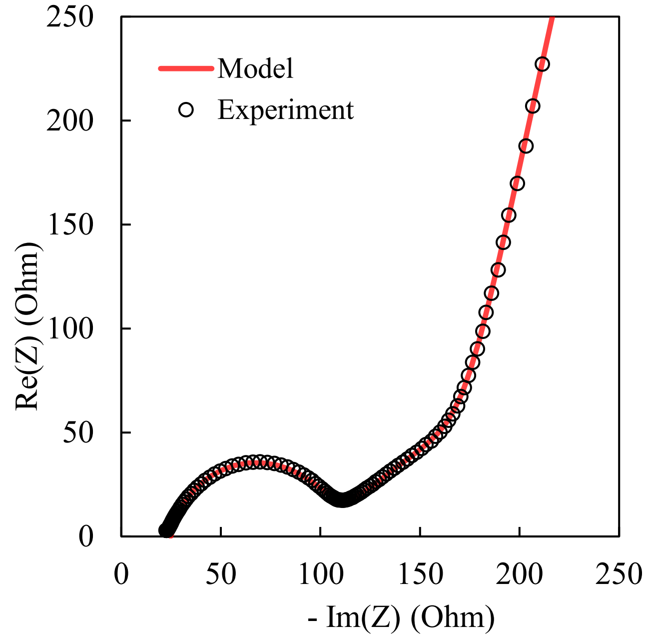
Blocking-Electrolyte Challenges

Use of orthogonal experiments to get more info



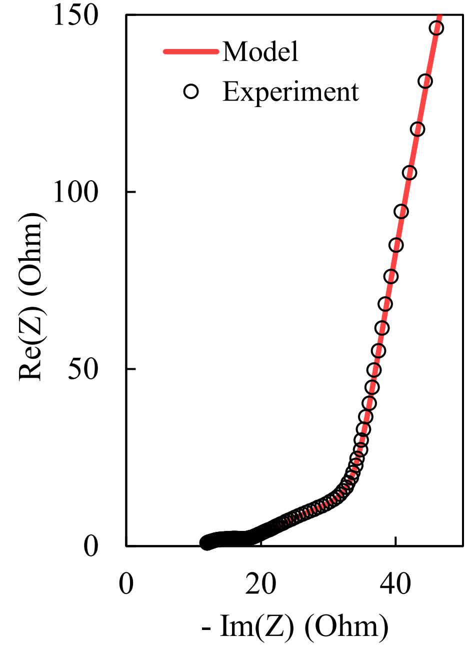
τ = 3.5

Intact cathode



τ = 3.05

Intact cathode
Low conductivity electrolyte



τ = 2.8

Delaminated cathode with copper as a current collector

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4. EQUIVALENT-CIRCUIT MODELS

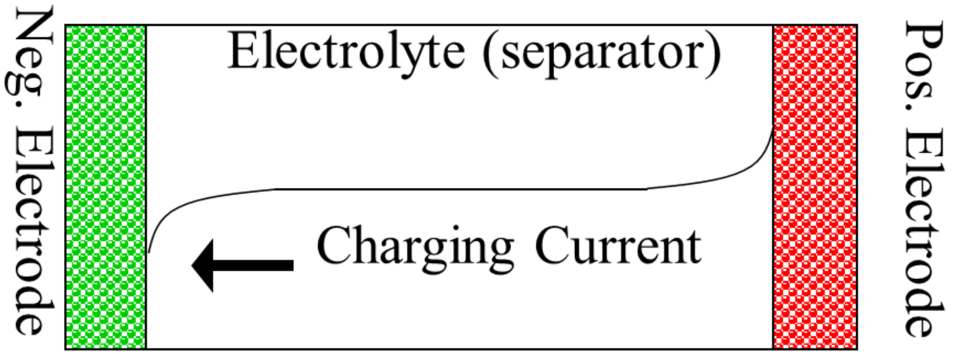
SUMMARY

- ECs and TLMs are intuitive and ***linear*** models
- They work for *stationary* situations (either DC or AC after transients have died down)
- They work when cell currents/overpotentials are low
- EIS spectra are easy to measure; *interpretation* can be quite difficult in real-world cases (e.g. porous electrodes) with many circuit elements

5. Nonlinear Phenomena in electrochemical systems

What are some phenomena in electrochemistry that are nonlinear (and would be included in a model)?

- Nernst Equation (effect of concentrations on OCP)
- Butler-Volmer Kinetics
- Concentration overpotential and transient diffusion of ions



- Others?

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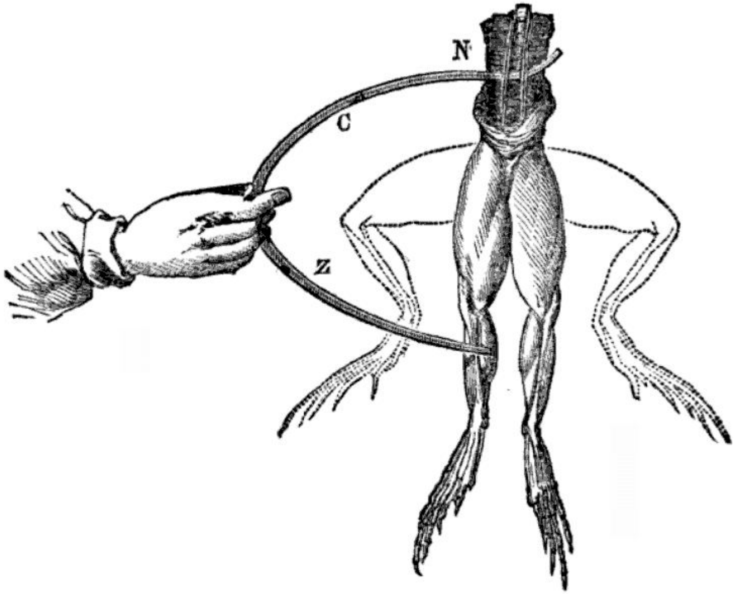
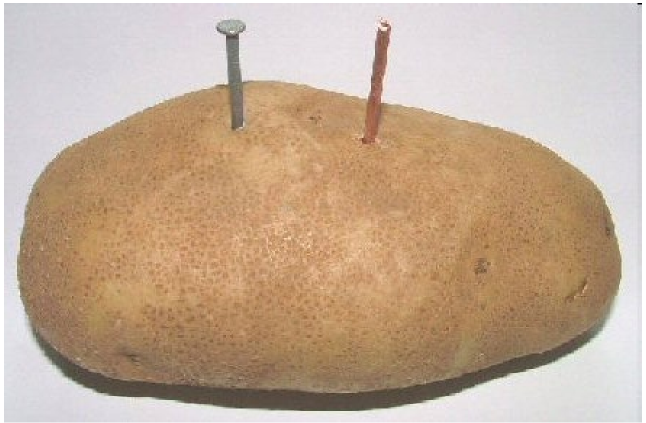
6. Battery Background

1786 Luigi Galvani



1799
Alessandro
Volta

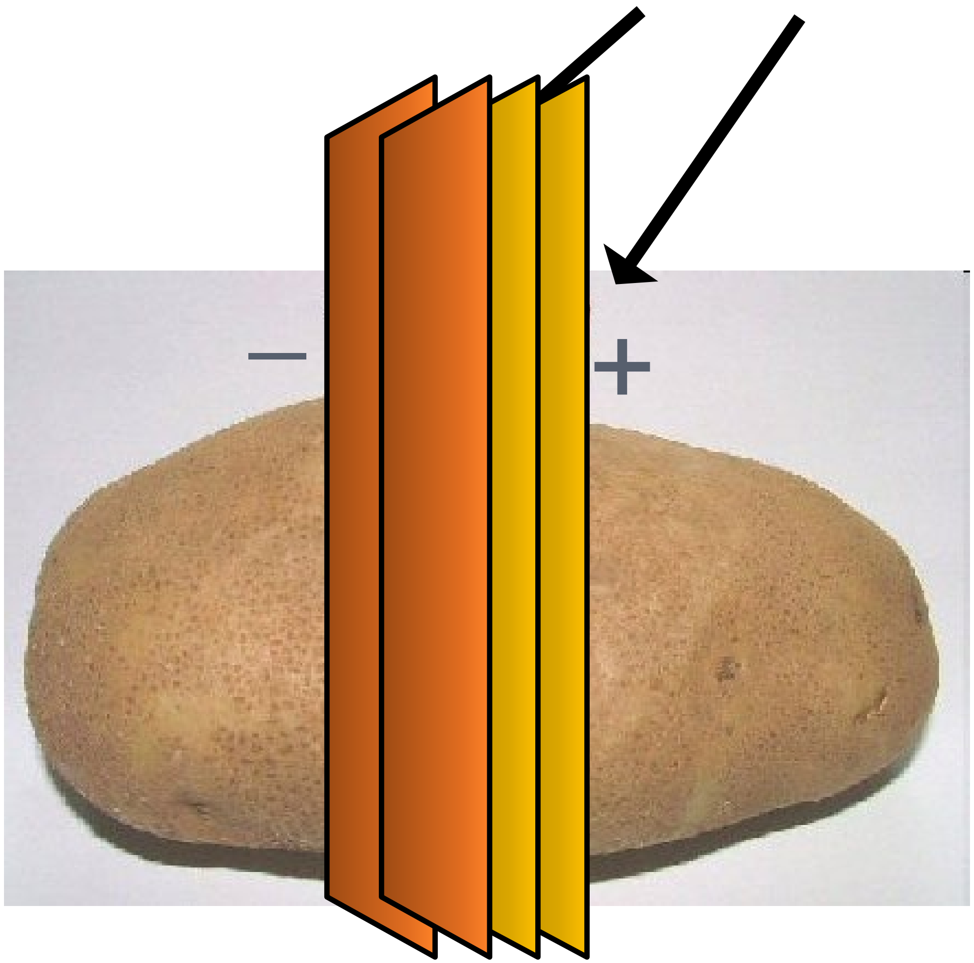


2024 science fair
battery

Image sources: David Ames Wells, GuidoB/Wikipedia

How to Make Batteries Work Better?

- Change the electrode chemistry
- Change the electrode geometry



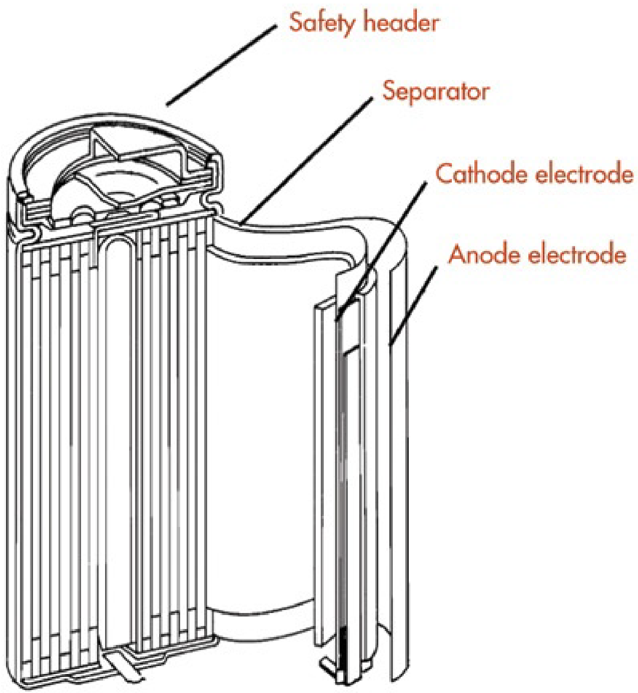


Image source:
electronicdesign.com

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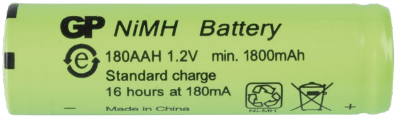
Kinds of Rechargeable Batteries

- Lead acid (invented 1859) – $100/kWh

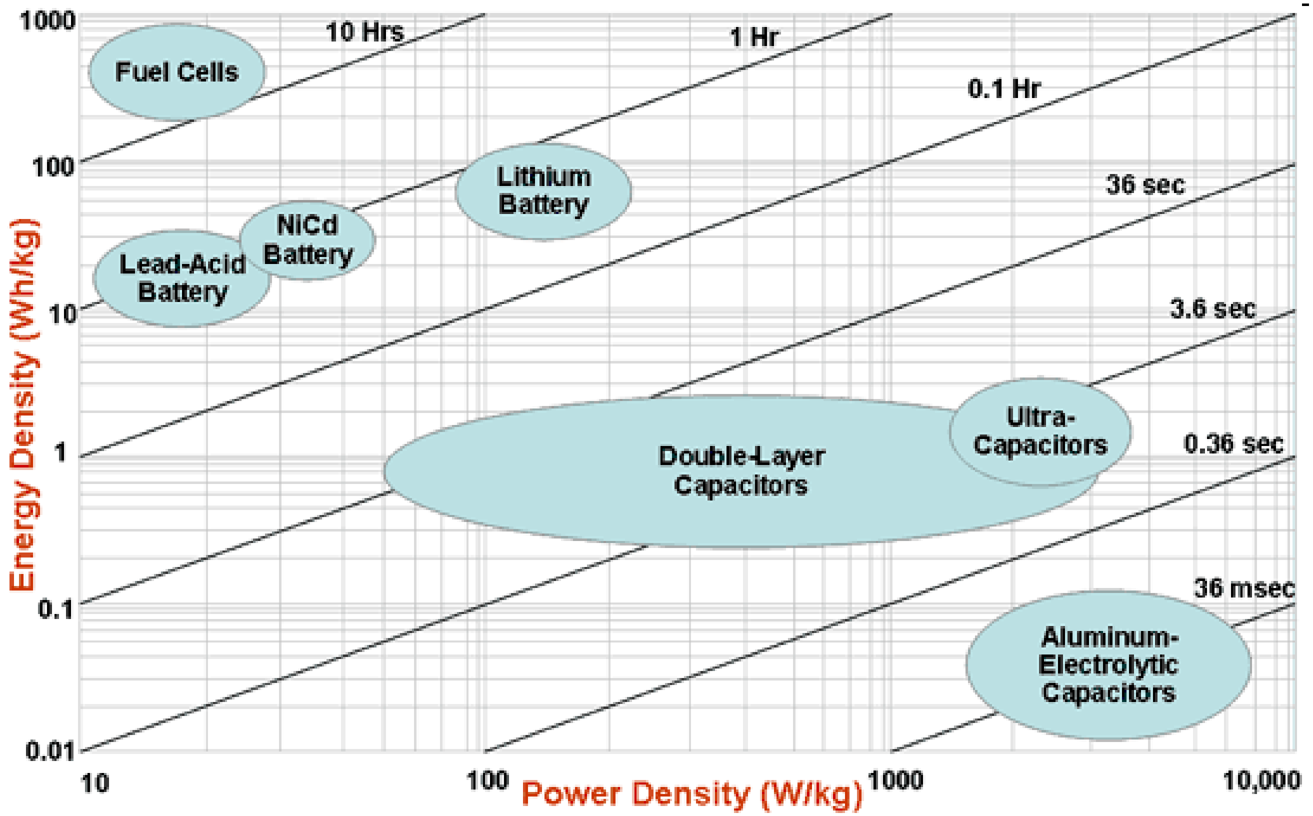
- Nickel cadmium, Ni metal hydride – $300/kWh
- Lithium-ion (commercialized 1990, light, long-lasting) – $100/kWh



- Other chemistries in development (5-10 years away)
- Warranty is big factor in commercialization

Image sources: voltaicsystems.com, megabatteries.com, amazon.com

ENERGY AND POWER CONNECTED BY DISCHARGE TIME (RAGONE PLOE)



Source US Defence Logistics Agency

One of the key parameters the battery engineer must know is charge/discharge time

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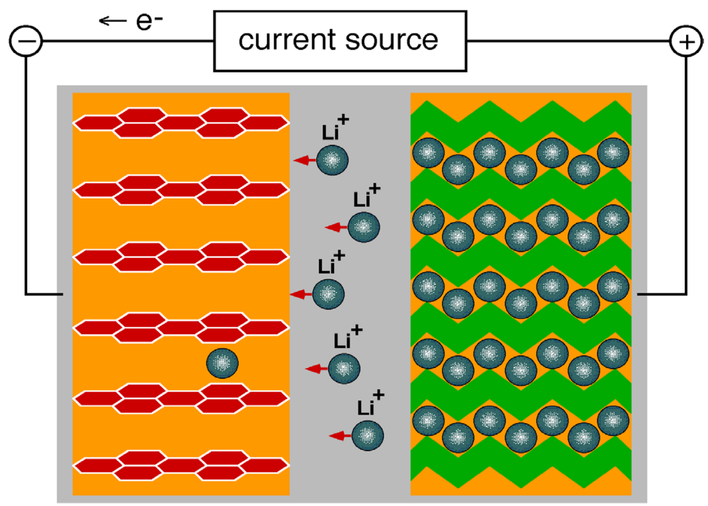
What is a Li-ion battery?

Lithium ions intercalate in interstitial lattice sites or alloy with other materials



1990:	Li_xC_6 (0<x<1)	Li_yCoO_2 (0.4<y<1)
1997:	Li_xC_6 (0<x<1)	Li_yFePO_4 (0<y<1)
newer:	Li_xSi (0<x<3.75)	$Li_yNi_aMn_bCo_cO_2$ (0.4<y<1)

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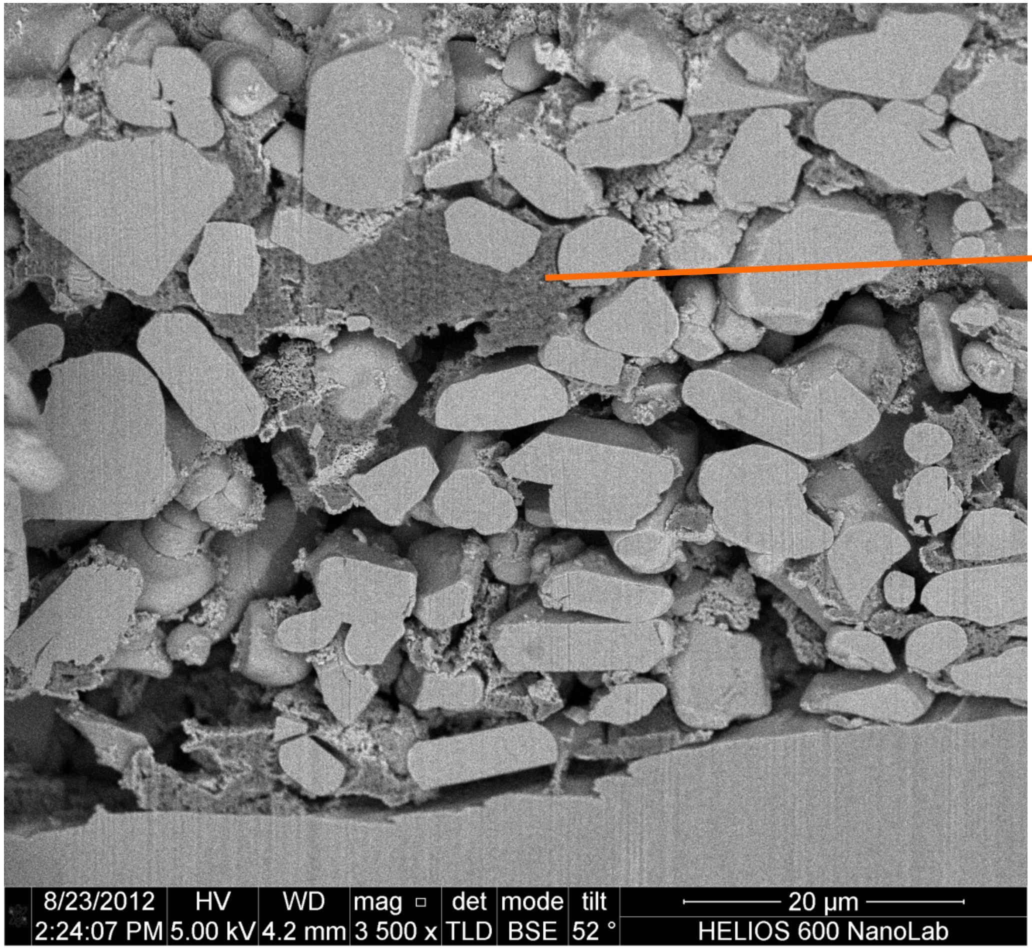
Porous Electrode Microstructure

SEM/FIB cross section of LCO cathode

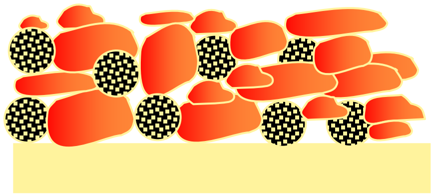


Concept of carbon binder domain (CBD)

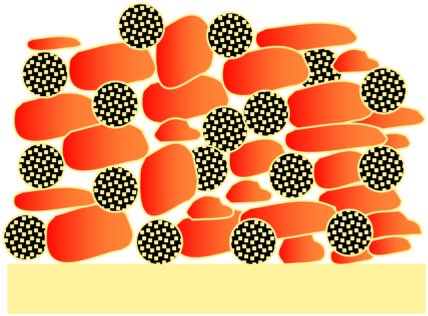
Stephenson et al., J. Electrochem. Soc. 158, A781 (2011)

OPTIMIZATION: TRADEOFFS IN ELECTRODE DESIGN

Thin



Thick



- ➢ Lower transport resistances
- ➢ Higher mass burden of inert materials (current collector + separator)

- ➢ Higher transport resistances
- ➢ Lower mass burden of inert materials (current collector + separator)

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Safety Is an Issue

- Batteries store lots of energy in a small space
- Fire can result because of “thermal runaway”
- Need to reduce heat transfer from one cell to another

Image from slate.com

Image from blog.tmcnet.com

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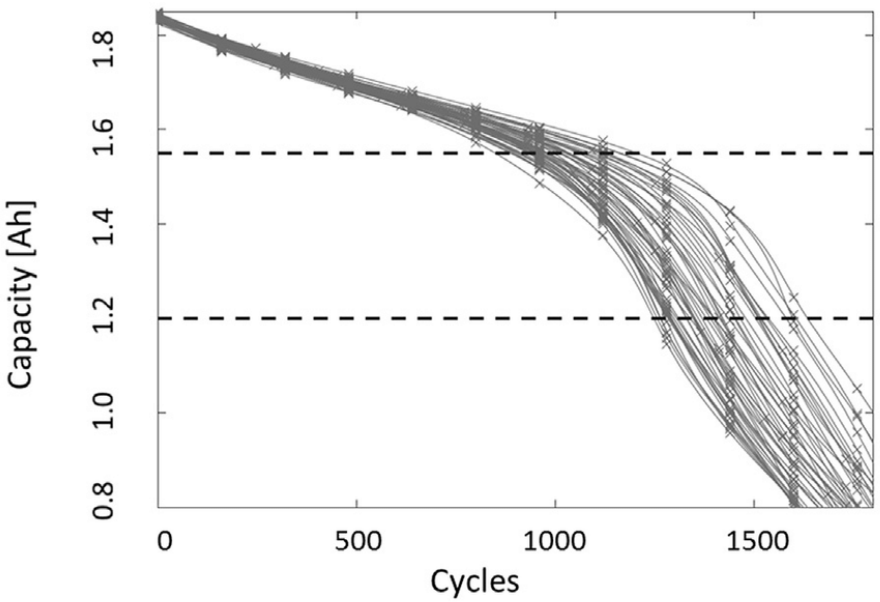
BATTERY LIFE IS AN ISSUE

"[device] failure is generally a statistical process, where life is determined by "hidden" variables over which we typically exercise little control. In batteries these variables could be microscale, such as heterogeneities within particles; or mesoscale, such as variation in local porosity or state of charge; or macroscale, such as the location in a pouch or cell-to-cell variation in the time-temperature history in a pack."

Harris et al., J. Power Sources 342 , 589 (2017)



Durability data of from 48 nominally identical Panasonic 18650 batteries. Baumhöfer et al., J. Power Sources 247 (2014) 332-338

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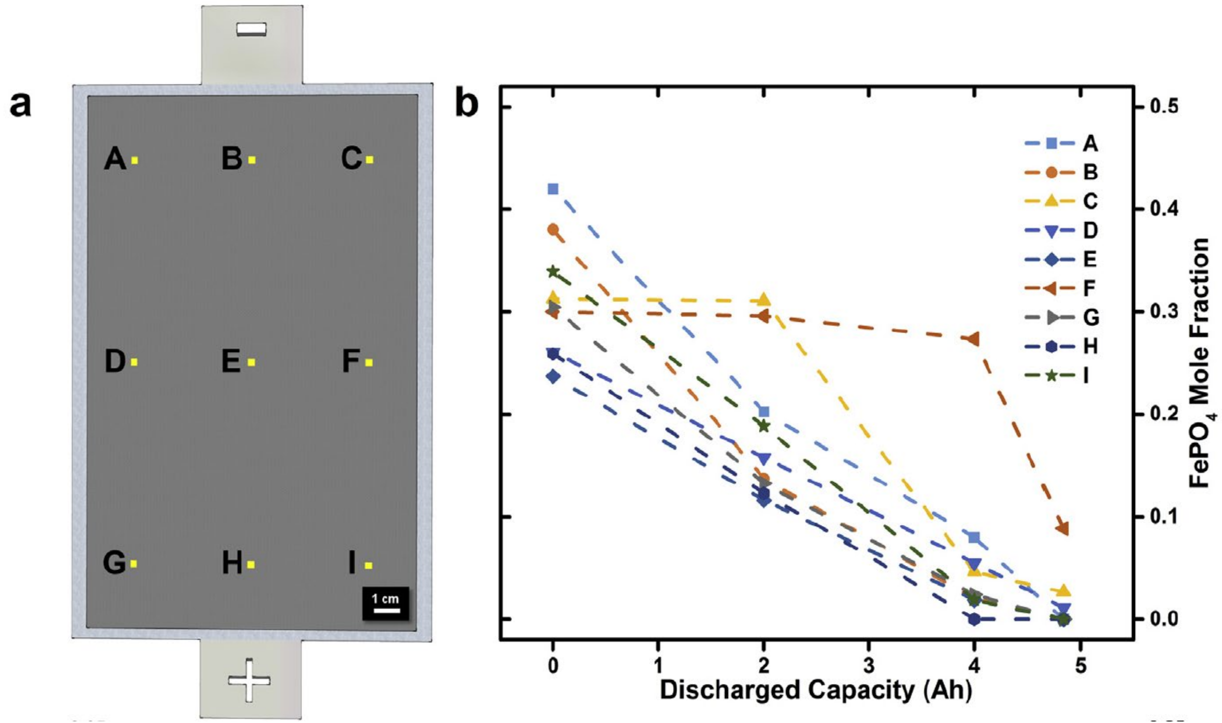
EXPERIMENTAL EVIDENCE OF HETEROGENEITY

EDXRD on LFP pouch cell to determine local state of charge



Paxton et al., J. Power Sources 275, 429 (2015)

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Delamination of films from current collector reveals growing mechanical heterogeneity during cycling

Pristine Formed Cycled

Pouraghajan et al., J Power Sources 492, 229636 (2021)

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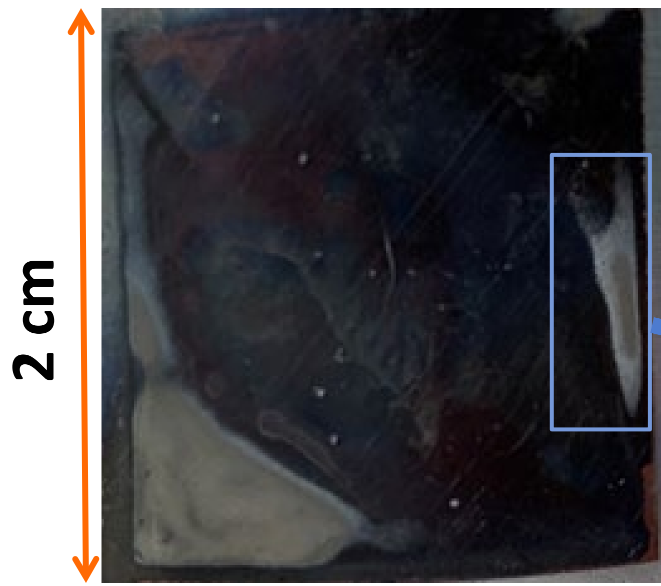
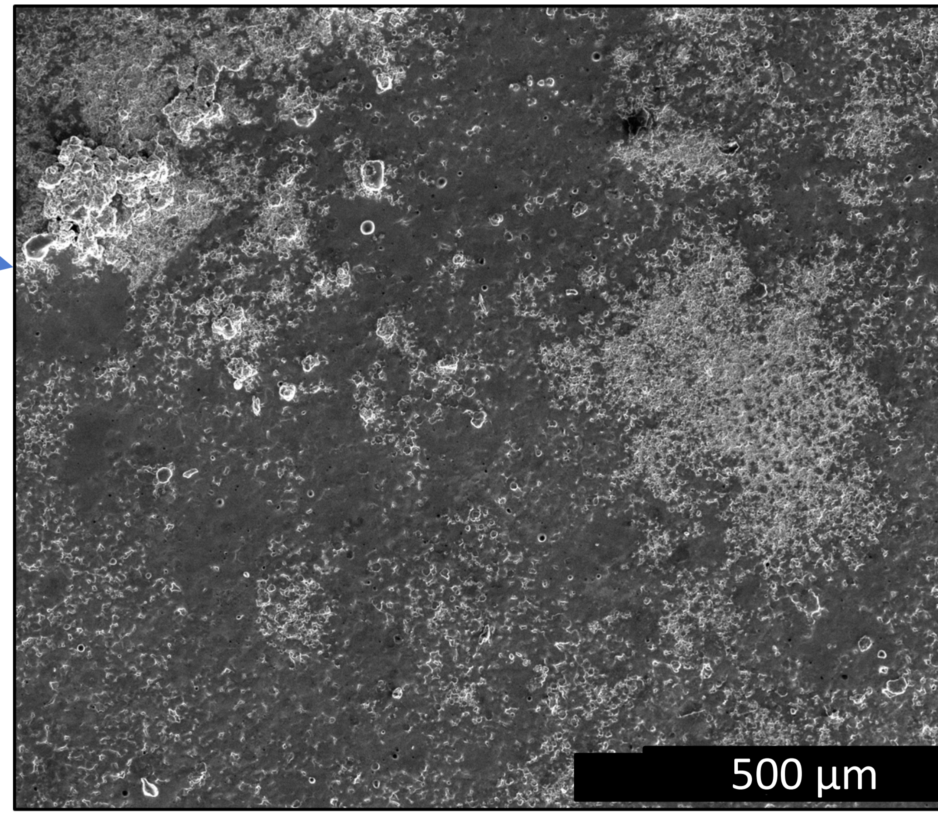
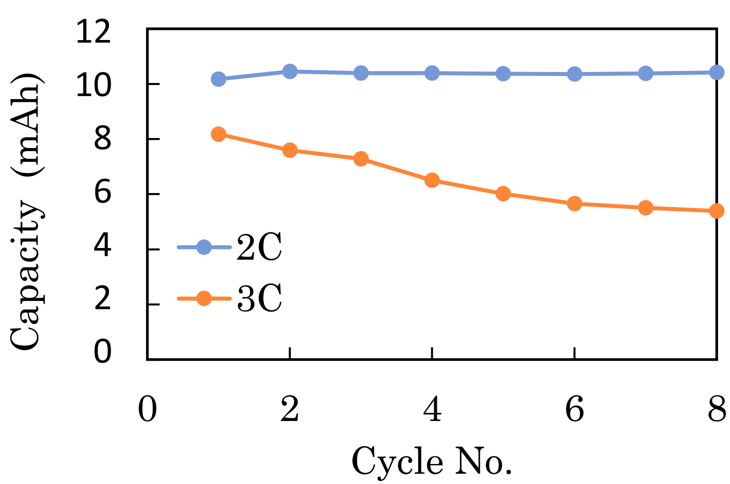
HETEROGENEITY IN LI-PLATING FOR HIGH-RATE CHARGING







Hamedi et al. J Electrochem Soc 169, 020551 (2022)
Liu et al., J Electrochem Soc 169, 010517 (2022)

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Break Time

We will start again at 15:40

Lesson Plan

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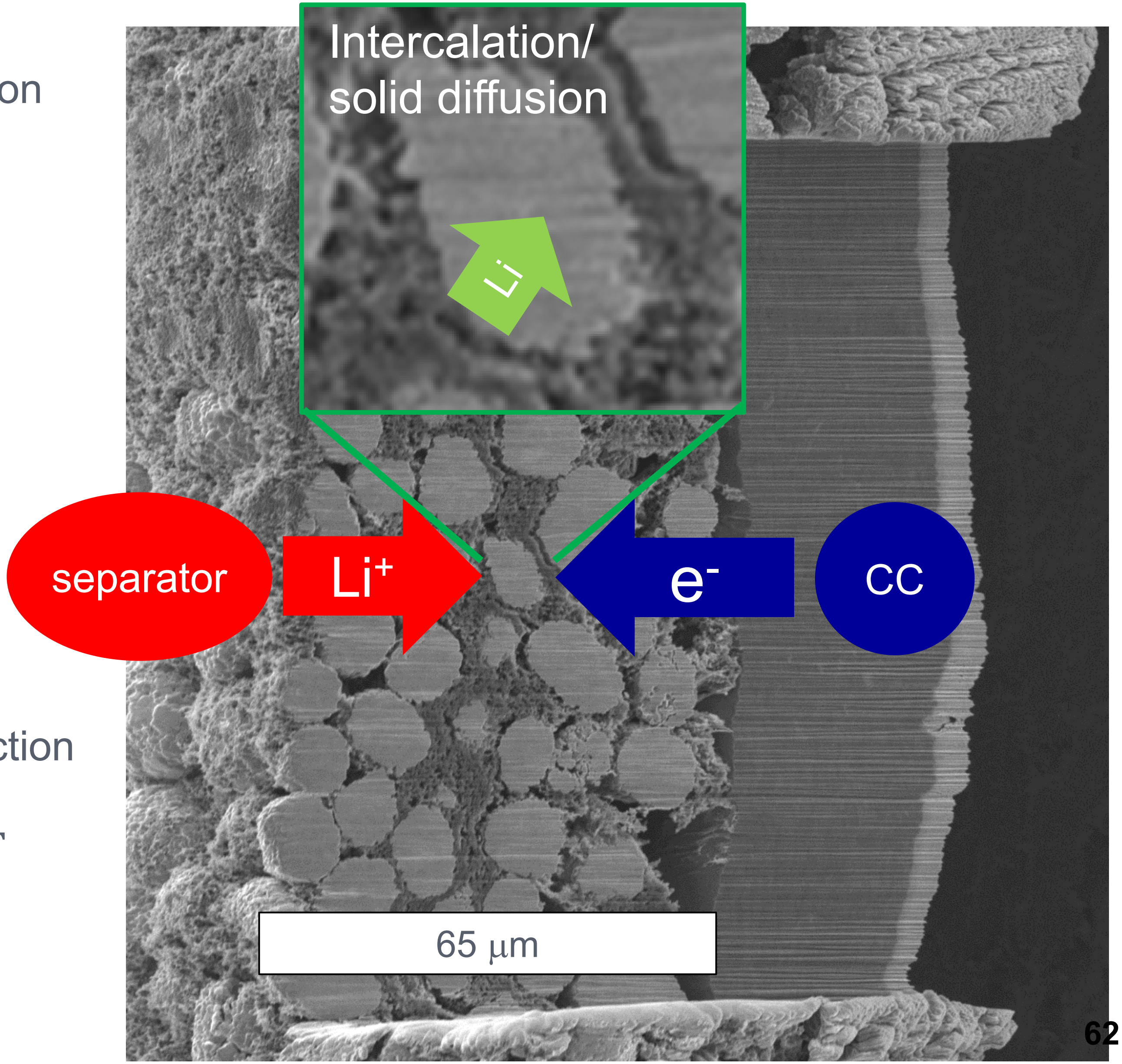
Porous electrode microstructure

FIB/SEM cross section

Materials are not uniformly distributed

- Active material
- Carbon
- Binder
- Pores



Electrochemical reaction requires 3 reactants

$\mathrm{Li}^{+} + e^{-} + \Gamma \longleftrightarrow \mathrm{Li}\,\Gamma$

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7. Pseudo-2D Model

P2D =
Newman-Type model =
DFN model =
Dual Foil model =
Macrohomogeneous model =
Porous Electrode Theory + Concentrated Solution Theory

Initially developed by John Newman and coworkers:

- Newman and Tiedemann, *AIChE J.* 21, 25-41 (1975).
- Doyle, Fuller, and Newman, *J. Electrochem. Soc.*, 140,1526 (1993).

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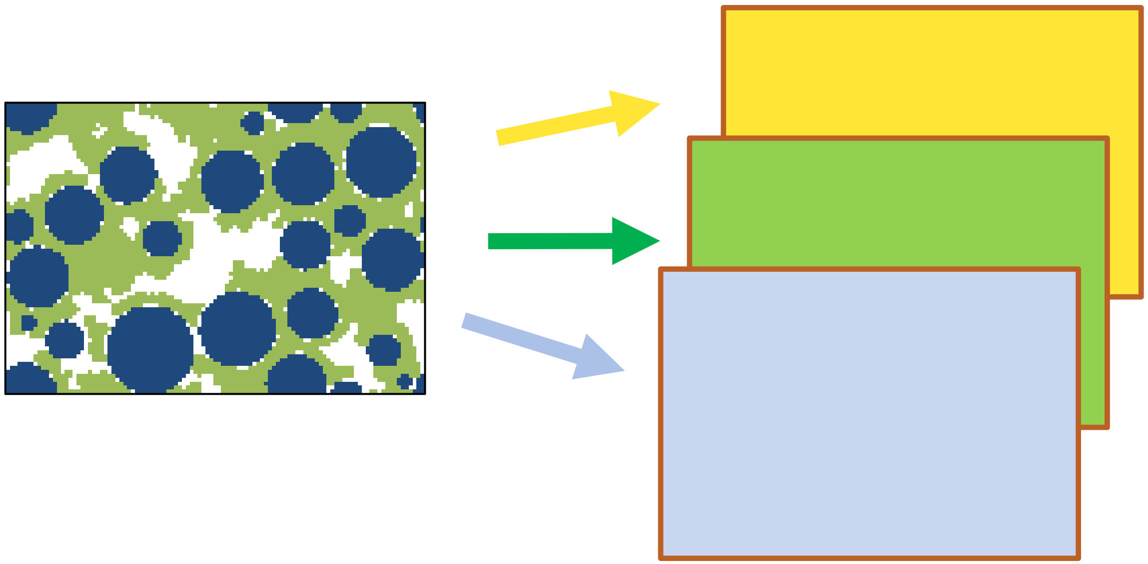
P2D MODEL CONCEPTS

- Homogenize conductivities
- Homogenize surface reaction
- Superposition of pathways



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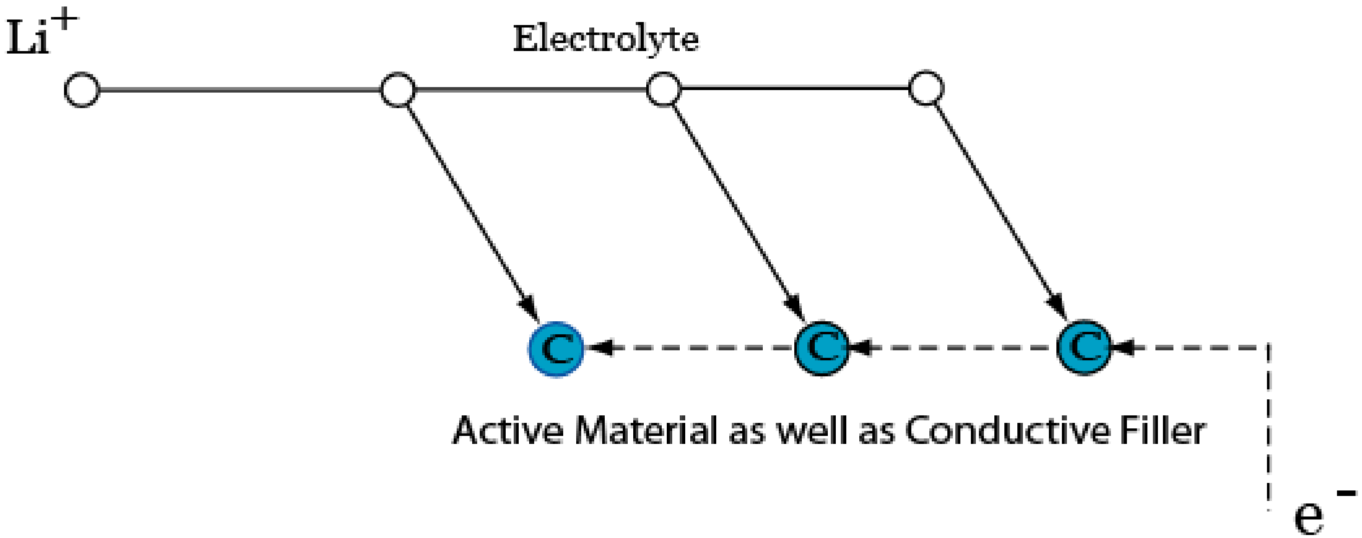
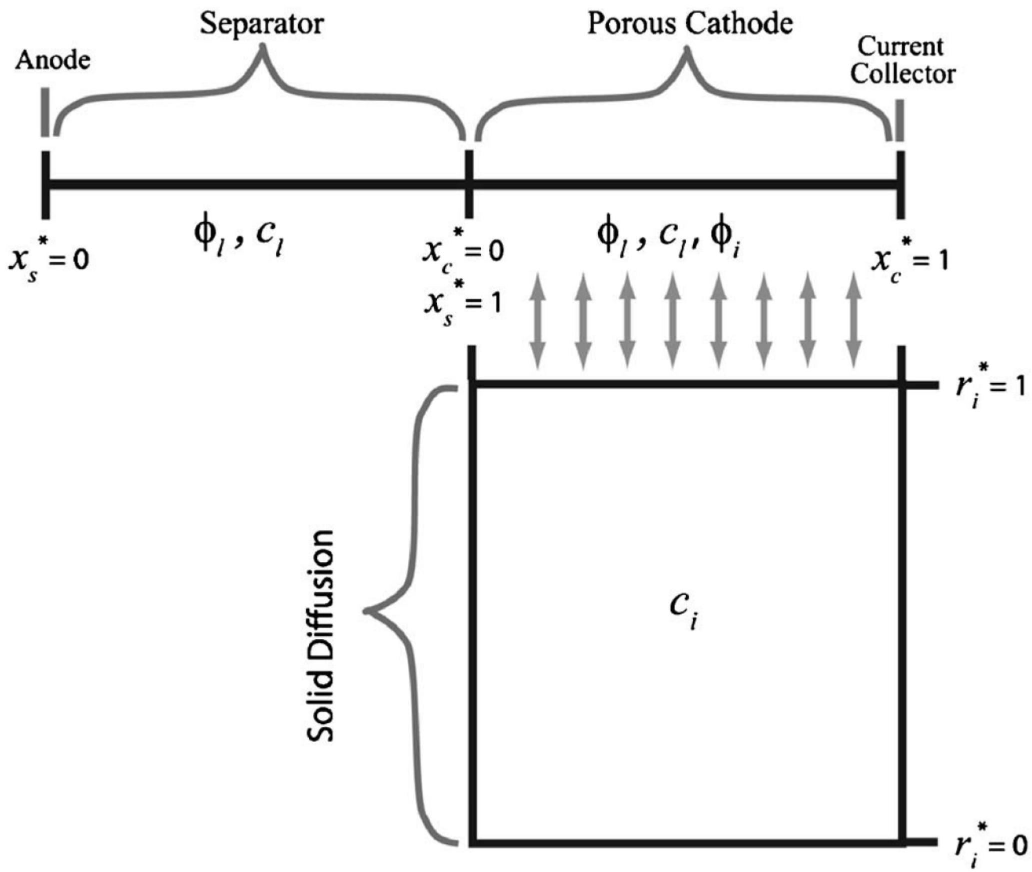
P2D MODEL CONCEPTS

- TLM representation for one electrode



P2D MODEL CONCEPTS

- Solid diffusion in radial direction coupled to ionic/electronic transport in *x* direction (cathode half cell shown here)



Stephenson et al., J Electrochem Soc 154, A1146 (2007)

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THE FOUNDATIONAL P2D EQUATIONS

- Assume binary electrolyte, no advection
- 4 dependent variables: solid potential, electrolyte potential, electrolyte concentration, solid concentration

$$\nabla \cdot \mathbf{i}_1 = -Faj_+ \qquad \mathbf{i}_1 = -\sigma_{\text{eff}} \nabla \phi_1$$

$$\nabla \cdot \mathbf{i}_2 = Faj_+ \qquad \mathbf{i}_2 = -\kappa_{\text{eff}} \nabla \phi_2 + \frac{2\kappa_{\text{eff}} RT}{F} (1 - t_+^0) \frac{1}{c} \nabla c$$

$$\epsilon \frac{\partial c}{\partial t} = D_{\text{eff}} \nabla^2 c + \frac{1 - t_+^0}{F} Faj_+$$

$$\frac{\partial c_s}{\partial t} = D_s \nabla^2 c_s$$

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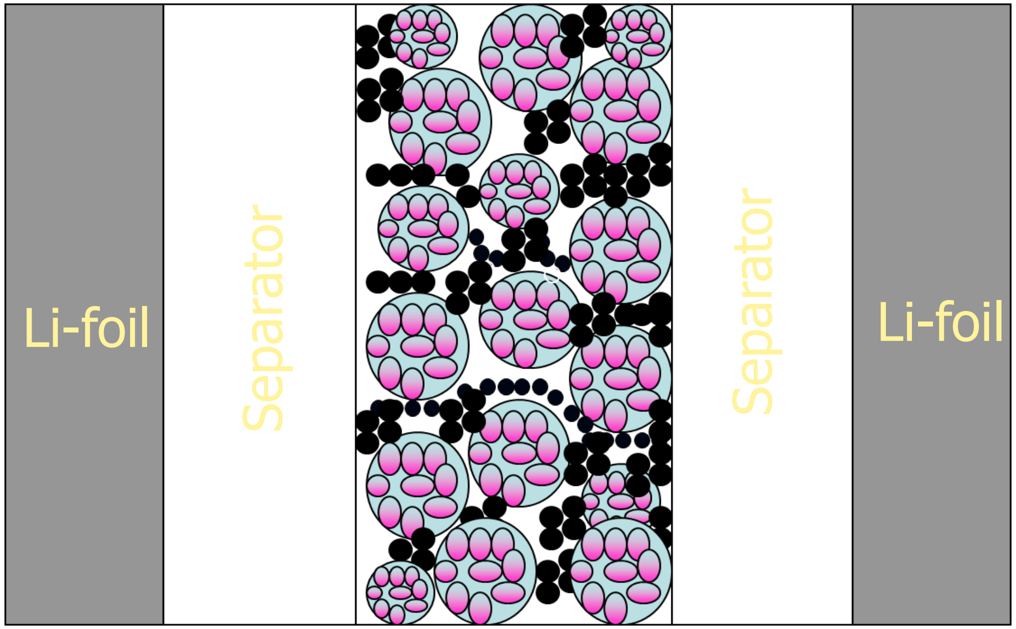
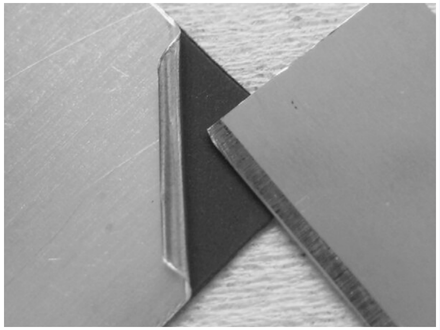
Summary of properties needed for simplest P2D model

- Open circuit potential as function of state of charge $U(y)$
- Ion transport properties κ_{eff}, D_{eff}, t_+
- Electron transport property σ_{eff}
- Solid-state diffusion D_s
- Surface kinetic rate constant ai_0
- Thicknesses and porosities of layers

IONIC TRANSPORT EXPERIMENTS





- Delaminate electrode film from current collector
- Restricted-diffusion experiment on free-standing film to determine effective ionic conductivity and salt diffusivity

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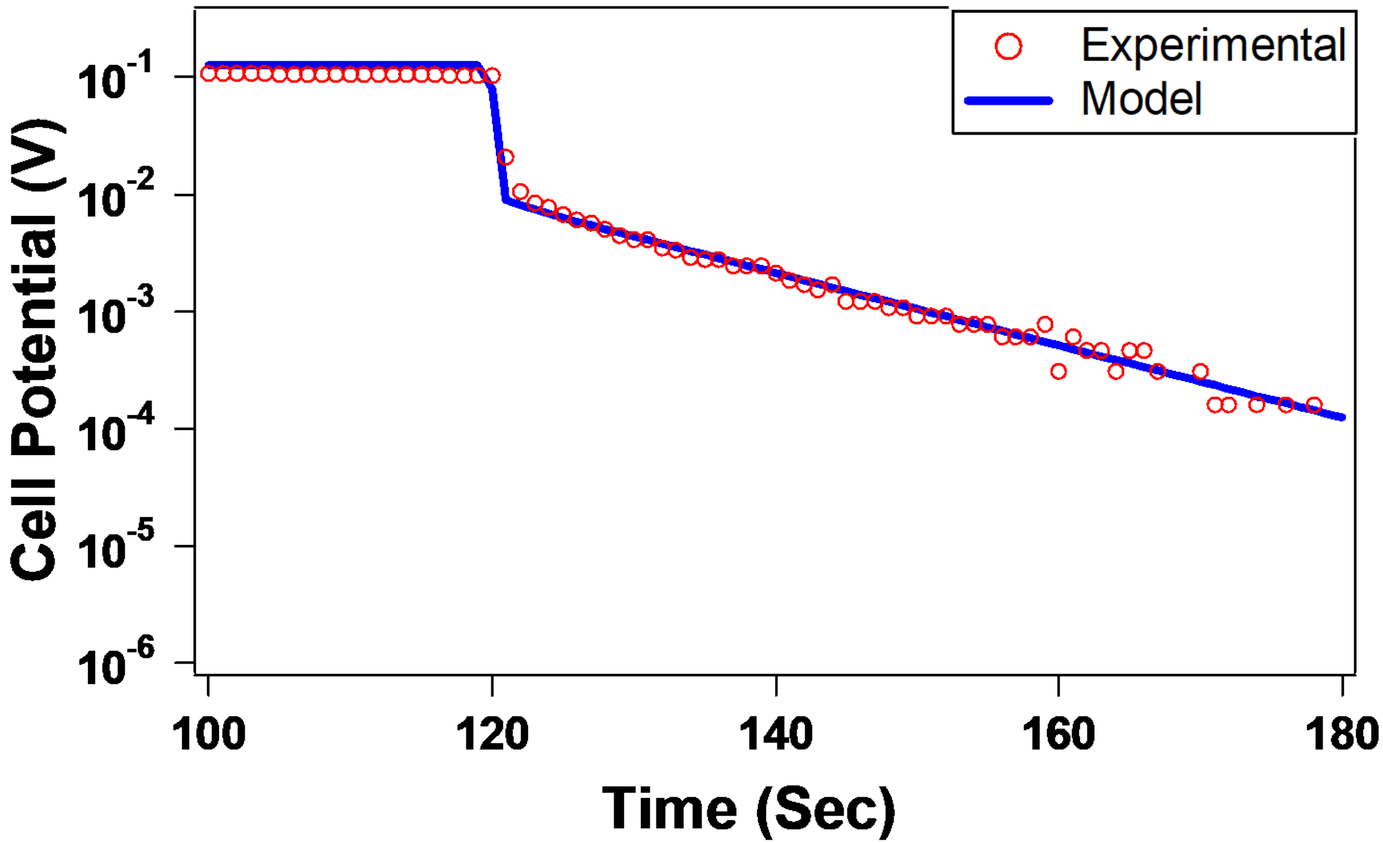
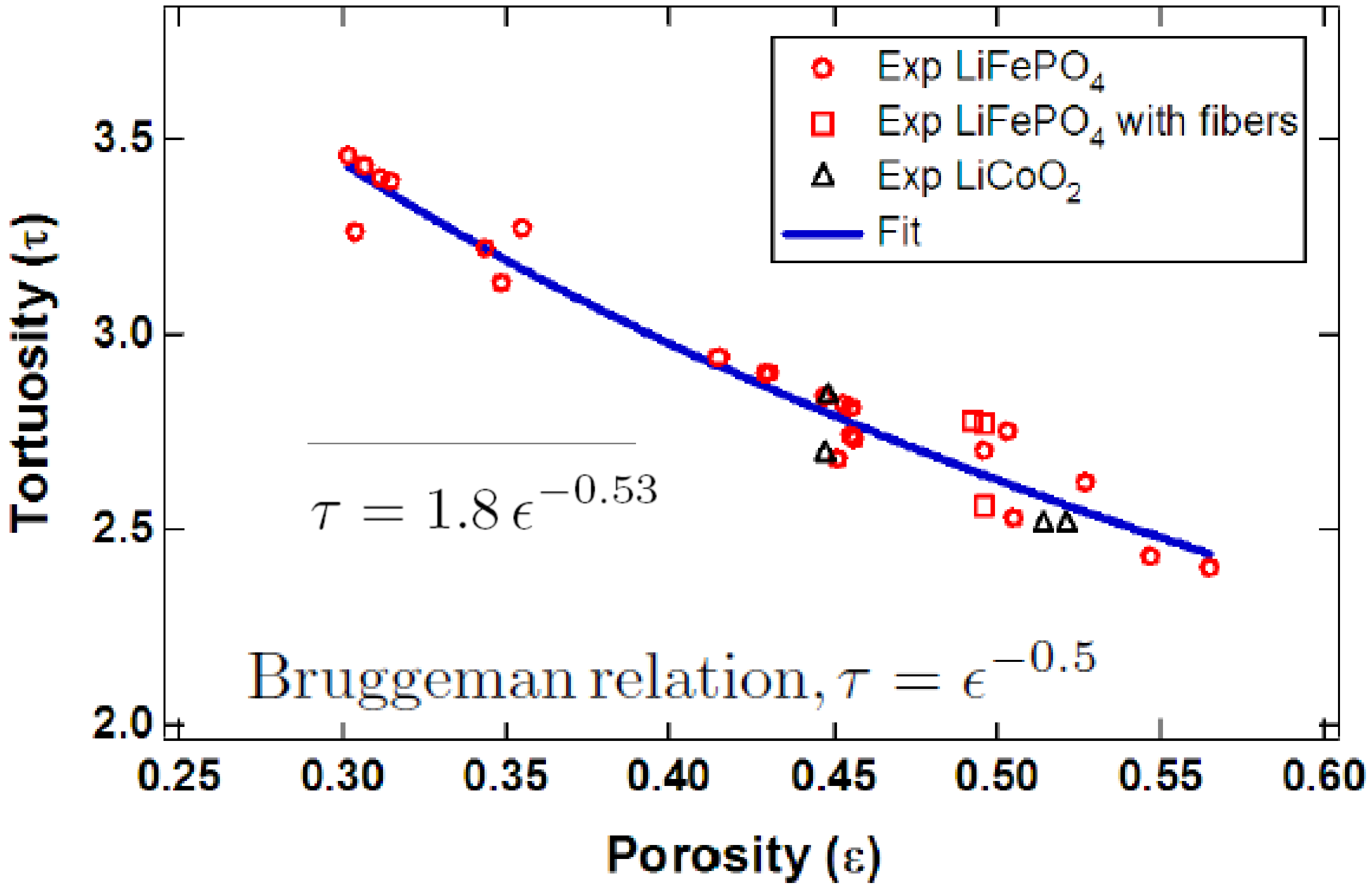
Polarization-interrupt experiment: P2D model used to interpret experiment



Thorat et al., *J. Power Sources* 188, 592 (2009).

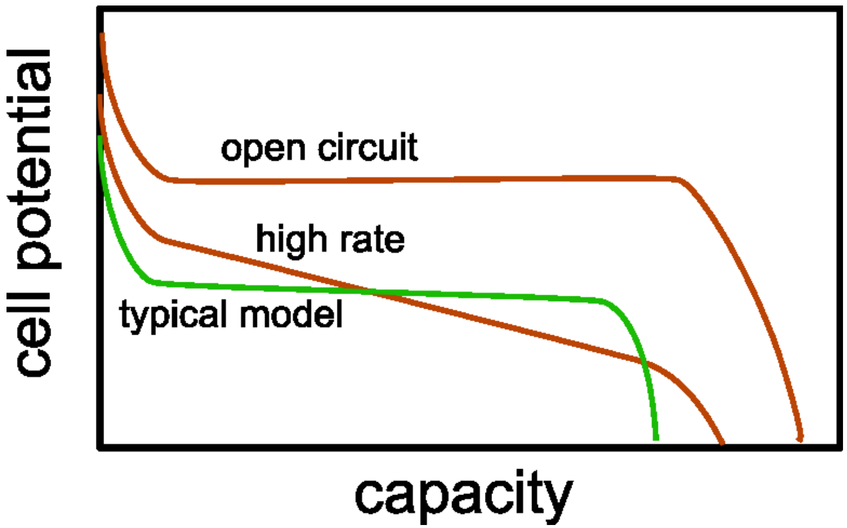
CATHODE TORTUOSITY VS. POROSITY



Thorat et al., *J. Power Sources* 188, 592 (2009).

Zacharias et al., *J. Electrochem. Soc*. 160, A306 (2013).

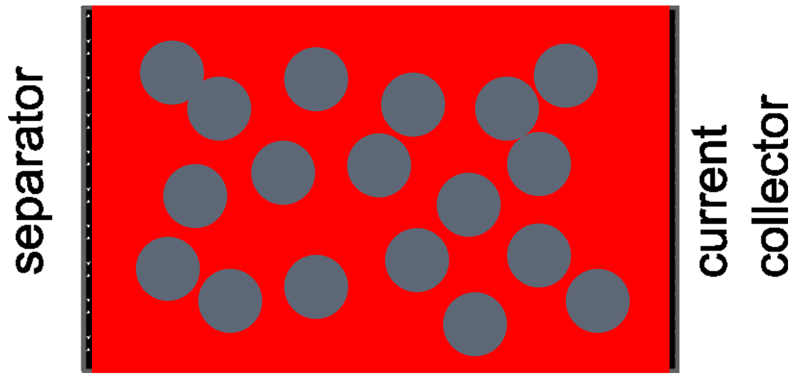
Observation of "Distribution of connectivity"



Apparent time-increasing resistance at high rate: What is happening?

Could have

1. High bulk resistance (ionic or electronic) to get moving reaction front
2. Distribution of particle types with varying local resistance



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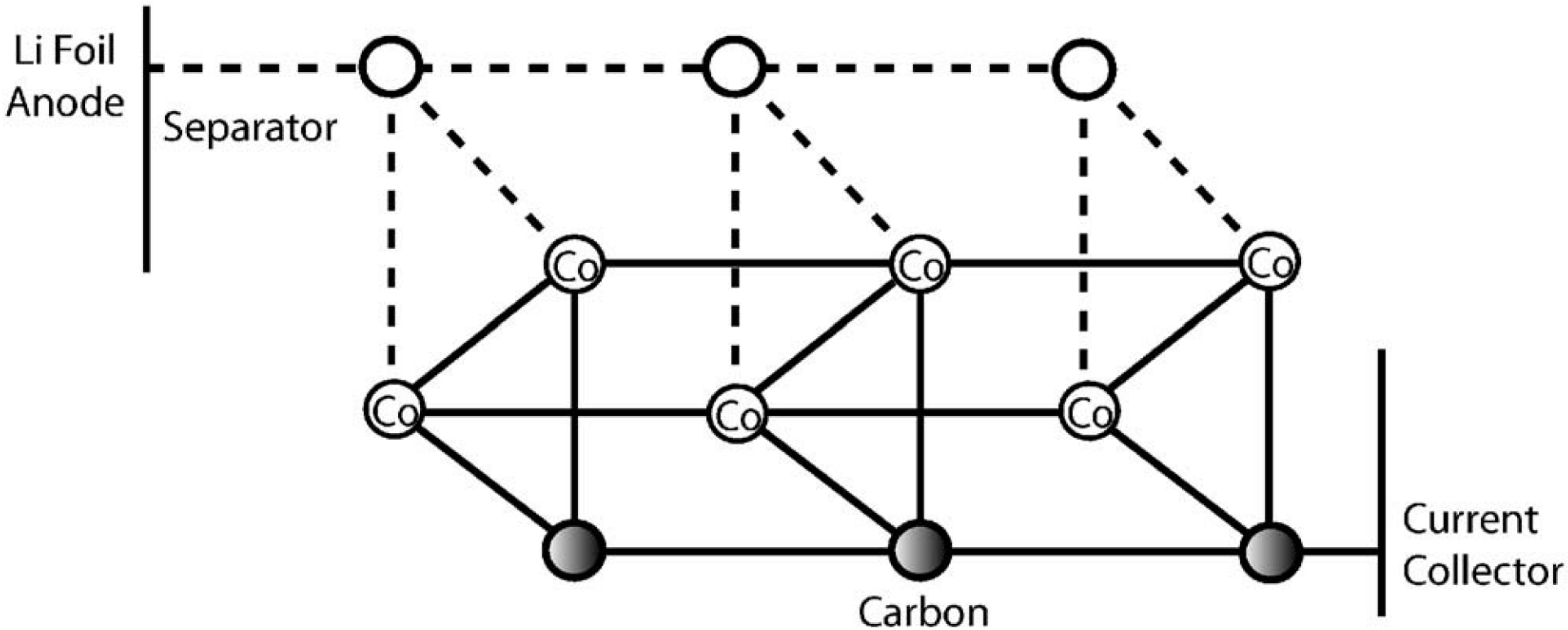
8. Extensions to Newman P2D model

A. Multiple particle types/sizes to provide "distribution of connectivity"

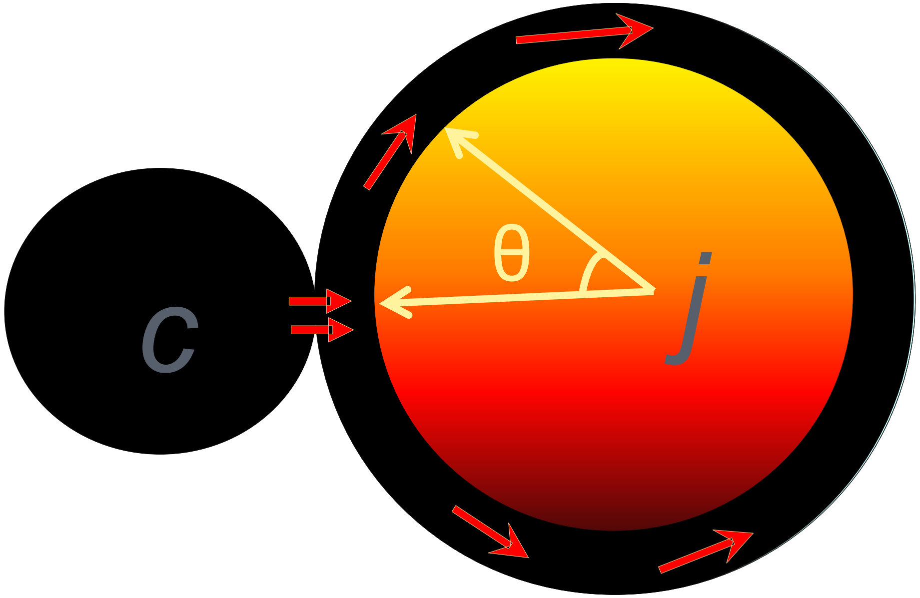


Stephenson et al., J. Electrochem. Soc. 154, A1146 (2007)
Thorat et al., J. Electrochem. Soc. 158, A1185 (2011)

Thomas-Alyea, ECS Trans. 16, 155 (2008)
Safari & Delacourt, J. Electrochem. Soc. 158, A63 (2011)

MODELING INTERPARTICLE CONTACT BETWEEN ACTIVE MATERIAL AND CARBON ADDITIVE

$$R''_{cj} = R''_{\text{contact}} + R''_{\text{spread}}$$



$$R''_{\text{spread}} = \frac{r_{\text{con}}^2}{2\, L_{cc}\, \sigma_{cc}} \ln\left(\frac{\sin\theta_c}{1-\cos\theta_c}\right)$$

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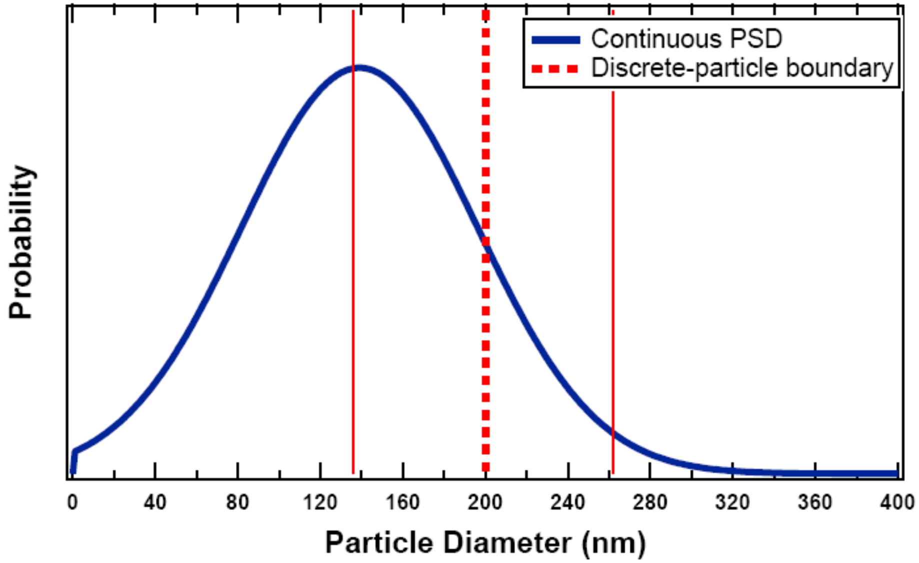
PARTICLE SIZES/TYPES IN LFP MODEL



Index (j)	R_j(nm)	m_j	R''_{cj} (Ω cm^2)
1	68	0.1615	2.6
2	68	0.3135	9.7
3	131	0.3500	12.0
4	131	0.1750	15.0

Smoothed curve measured from TEM images on Phostech material

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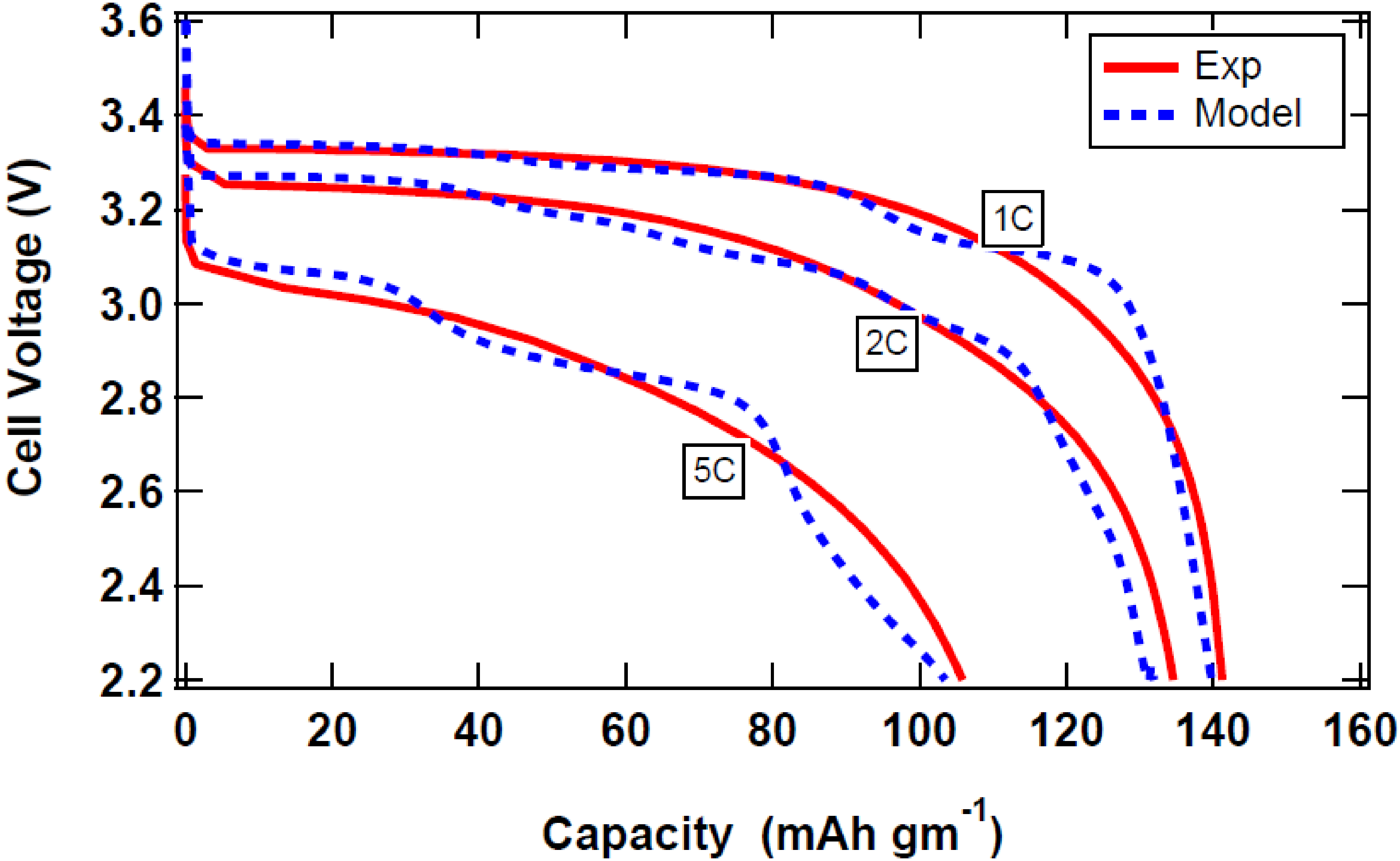
LFP Model validation: thin cell

34 um thick



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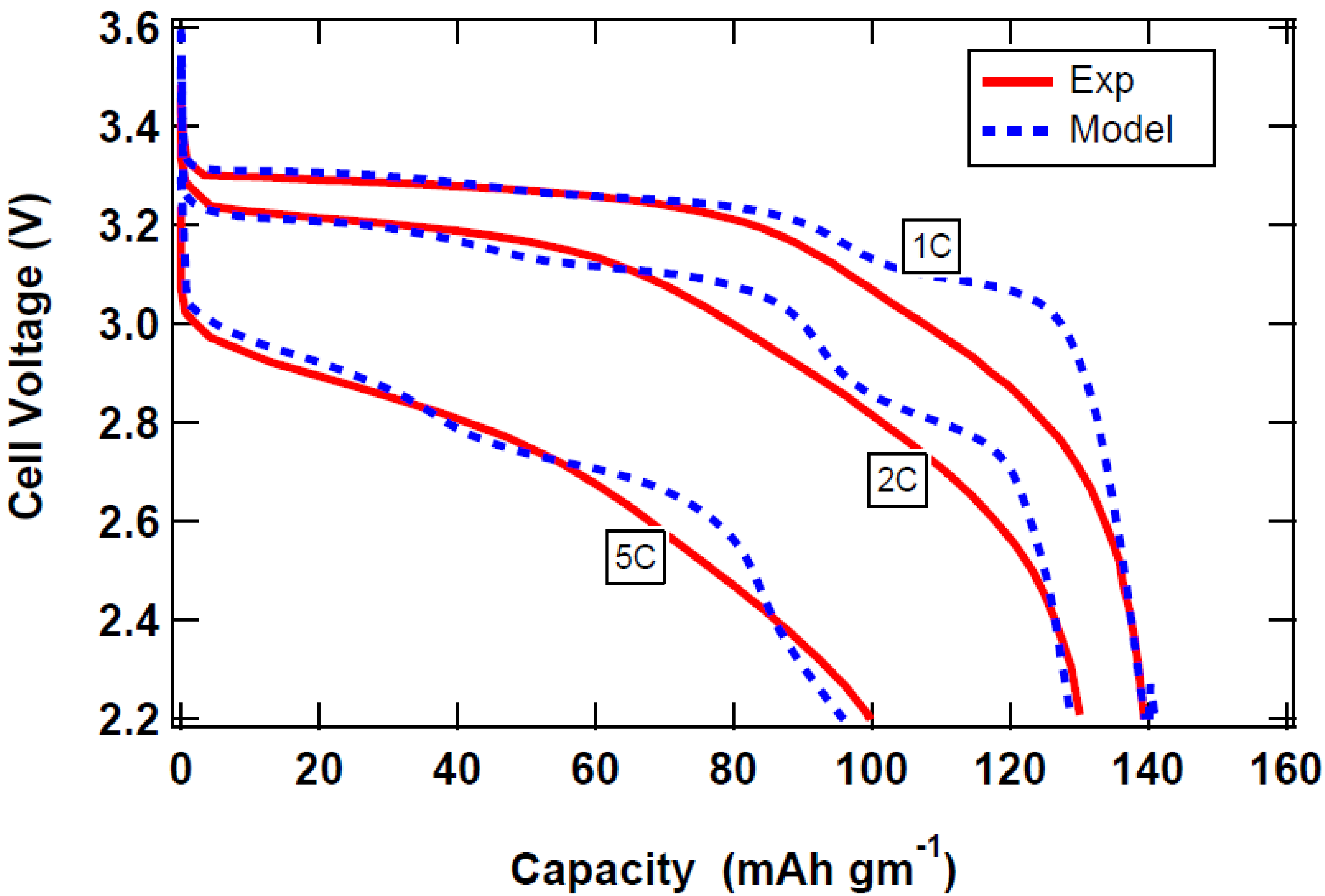
LFP Model validation: medium cell

57 um thick



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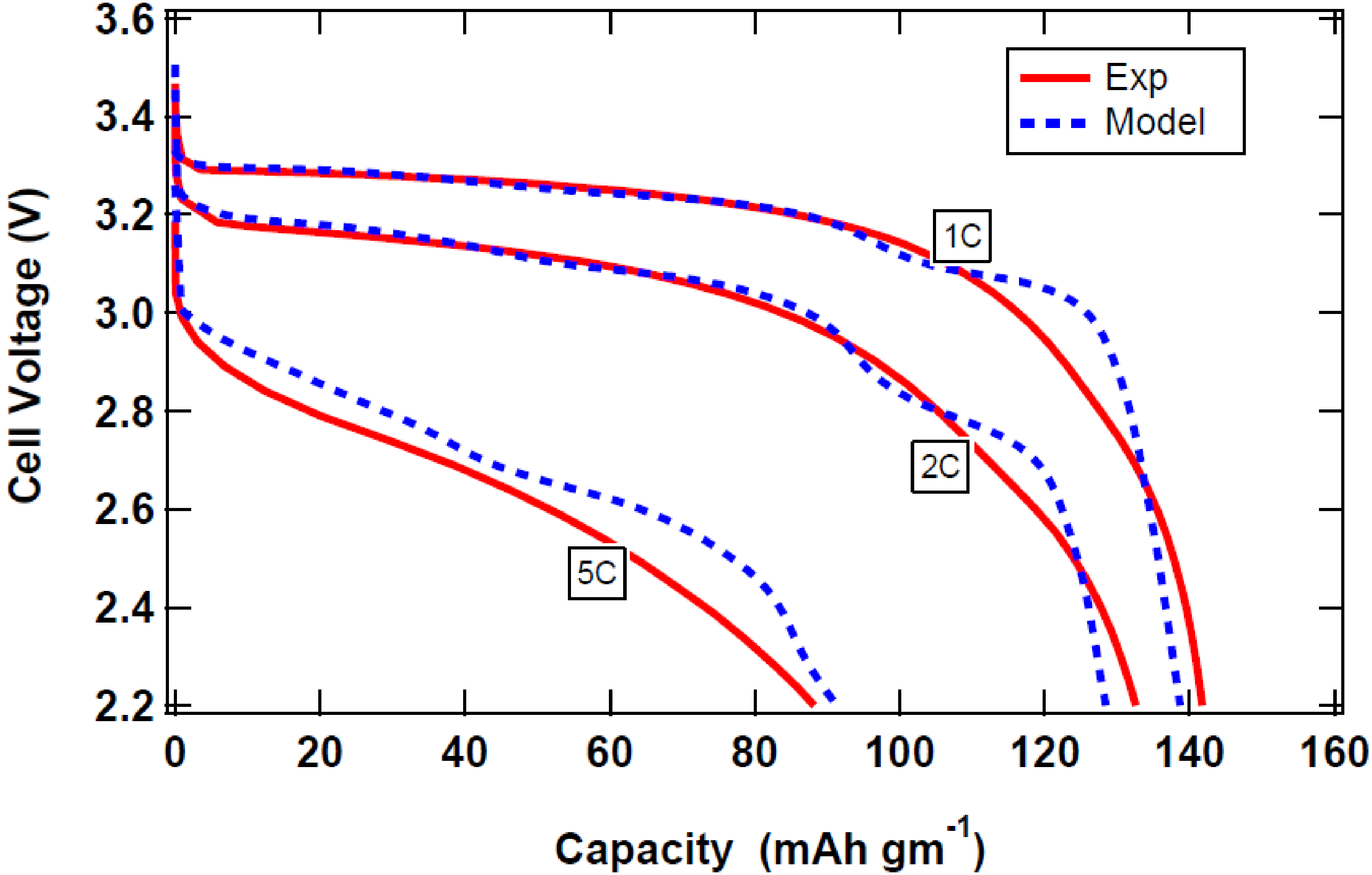
LFP MODEL VALIDATION: THICK CELL

66 um thick



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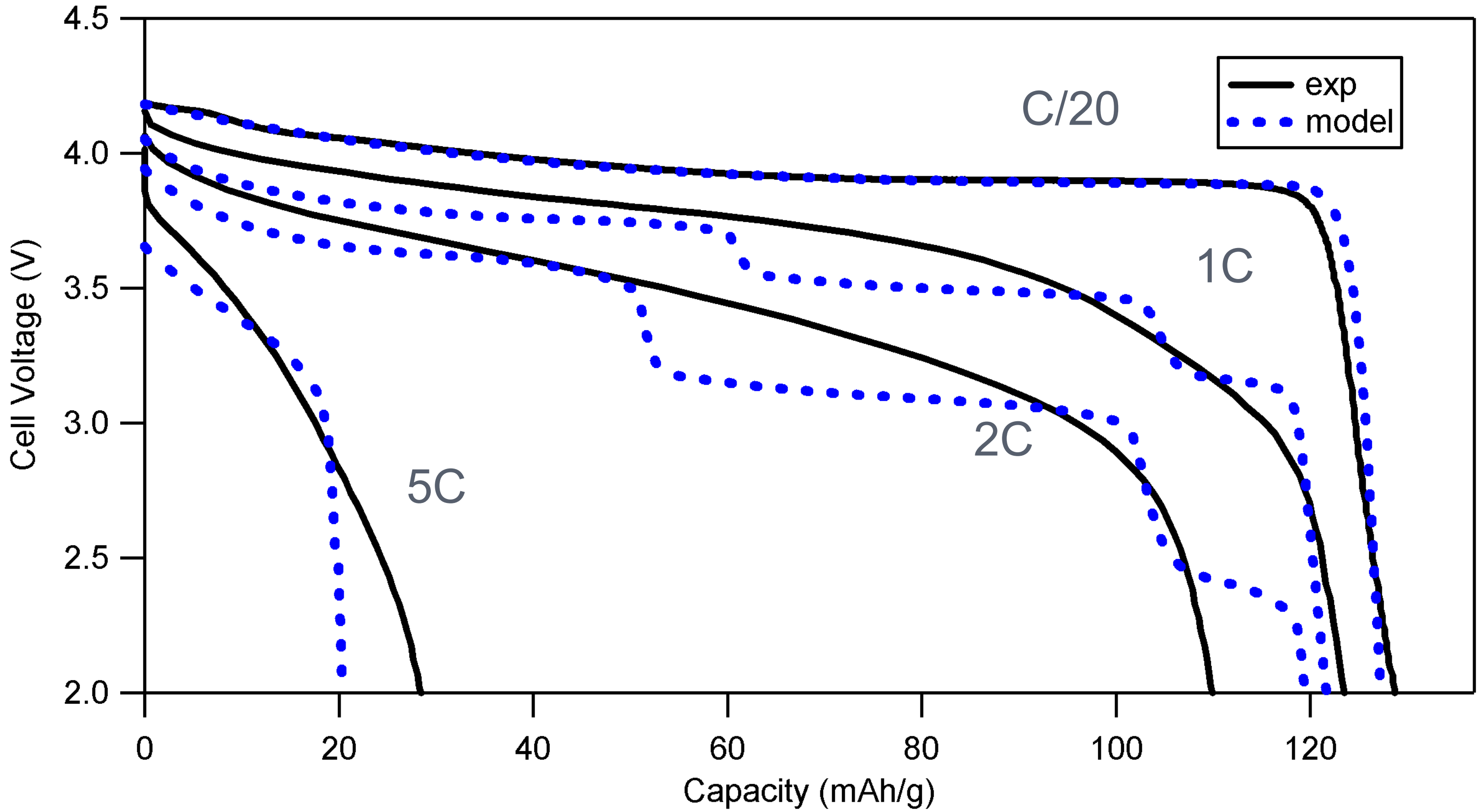
MODEL VALIDATION FOR $LiCoO_2$ ELECTRODE



Thick $LiCoO_2$ electrode

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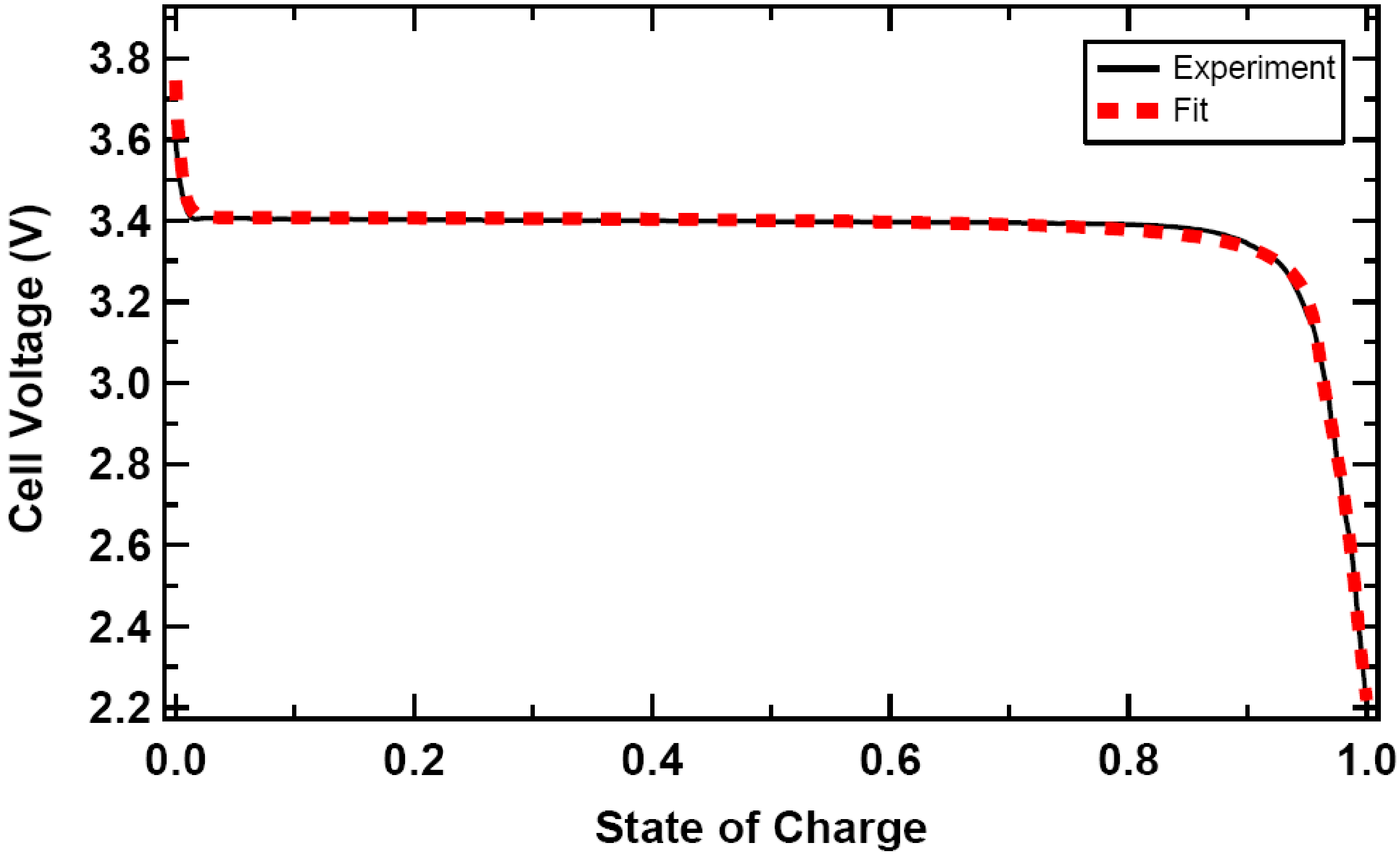
MANY ELECTRODE MATERIALS INVOLVE PHASE CHANGE



Li_xFePO_4 half cell

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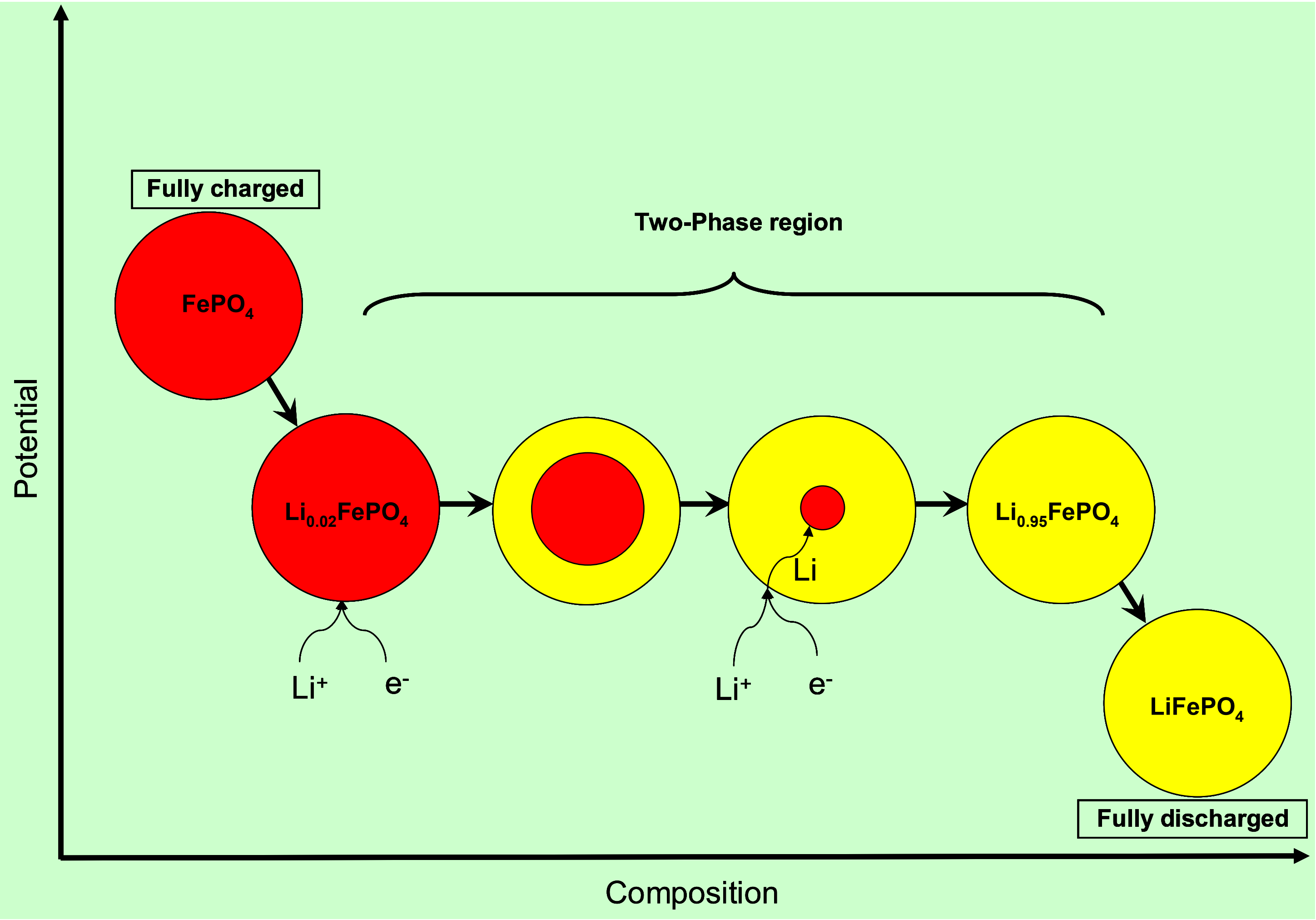
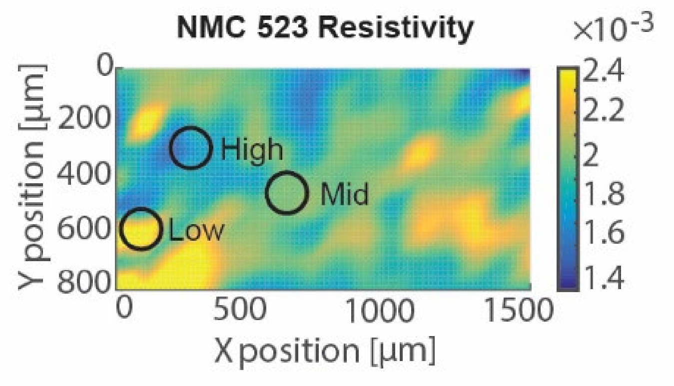
8. P2D MODEL EXTENSIONS
B. TWO-PHASE MODEL

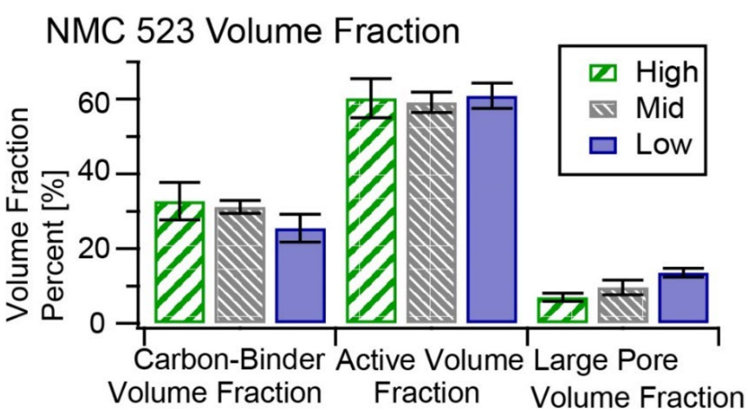


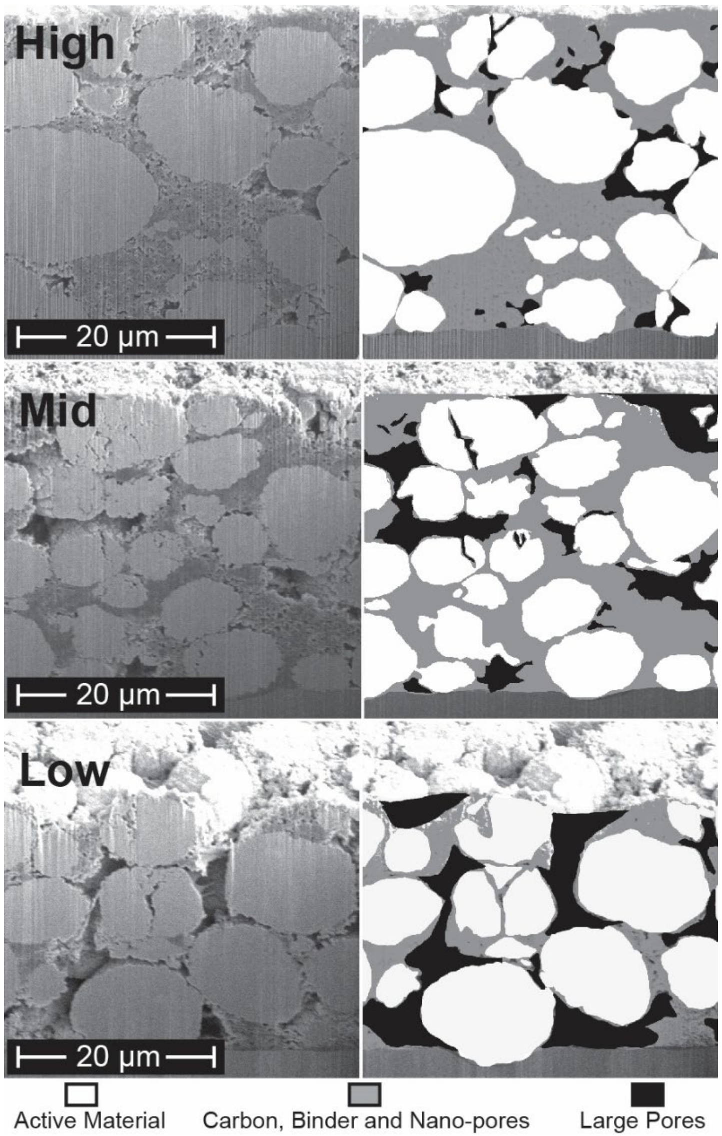
Srinivasan and Newman, J. Electrochem. Soc. 151, A1517 (2004)
Thorat et al., J. Electrochem. Soc., 158, A1185 (2011)

Microstructures Correlate with Electronic Impedance Heterogeneity in Cathode Films

NMC cathodes were tested electronically and subsequently imaged using scanning electron microscopy (SEM) with focused ion beam milling (FIB). Areas with measured **high** and **low** conductivity show a significant difference in microstructure.







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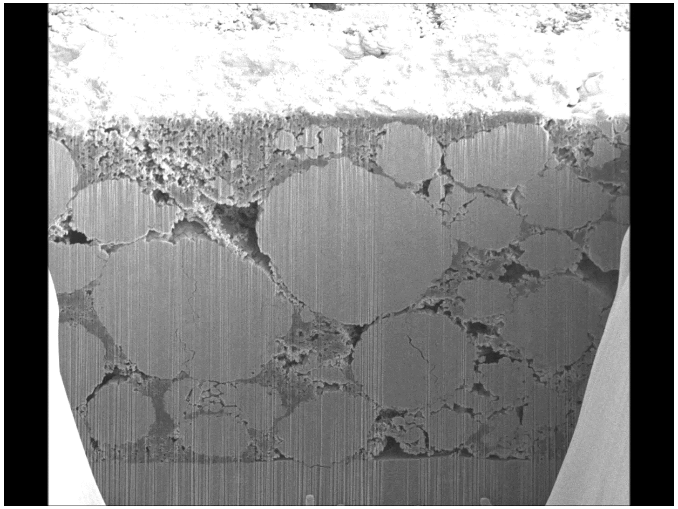
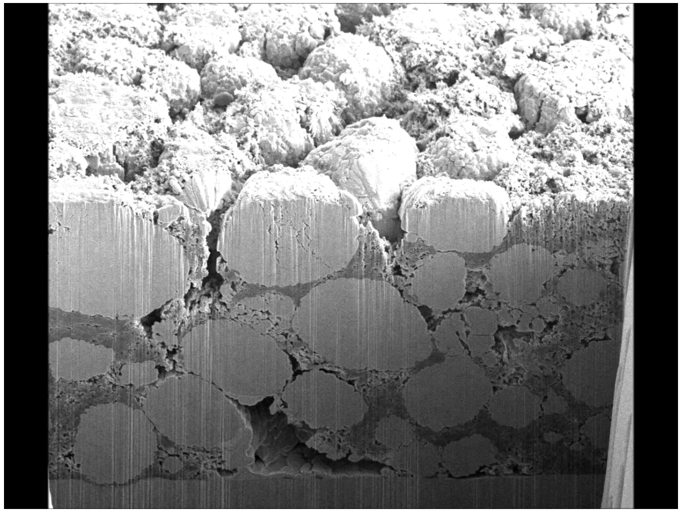
Video Comparison of High- and Low-Impedance Regions – NMC 523



Low Impedance



High Impedance

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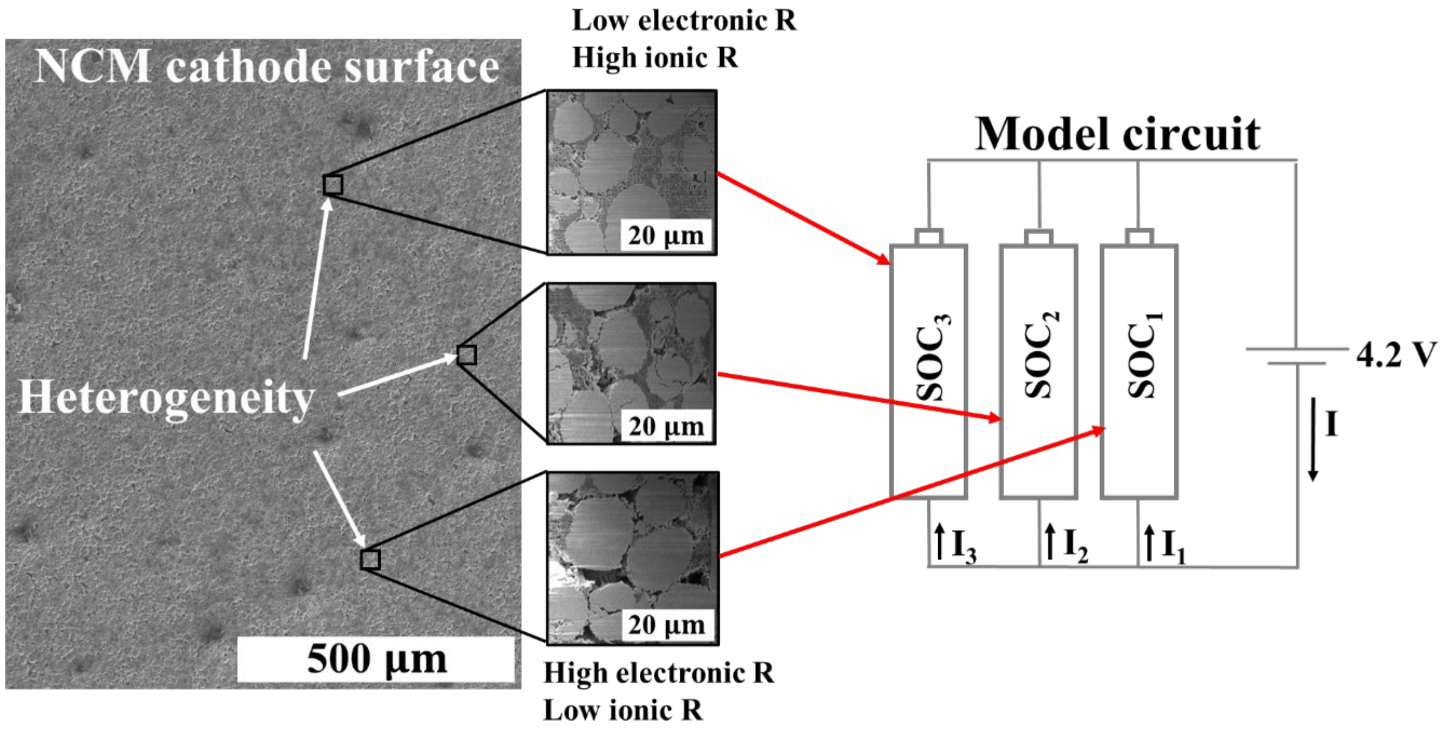
8. P2D MODEL EXTENSIONS
C. PARALLEL P2D MODELS TO DESCRIBE MISMATCH BETWEEN ELECTRODE REGIONS DUE TO HETEROGENEITIES

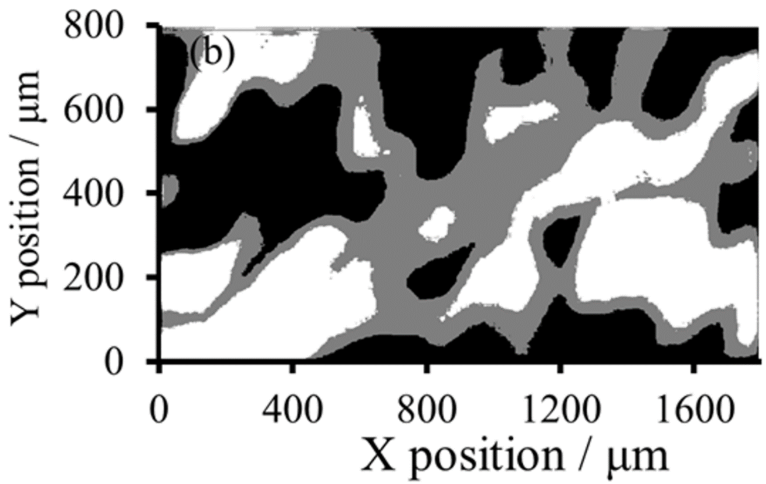


Forouzan et al., J Electrochem Soc 165, A2127 (2018).
Hamedi et al., J Electrochem Soc 169, 020551 (2022).

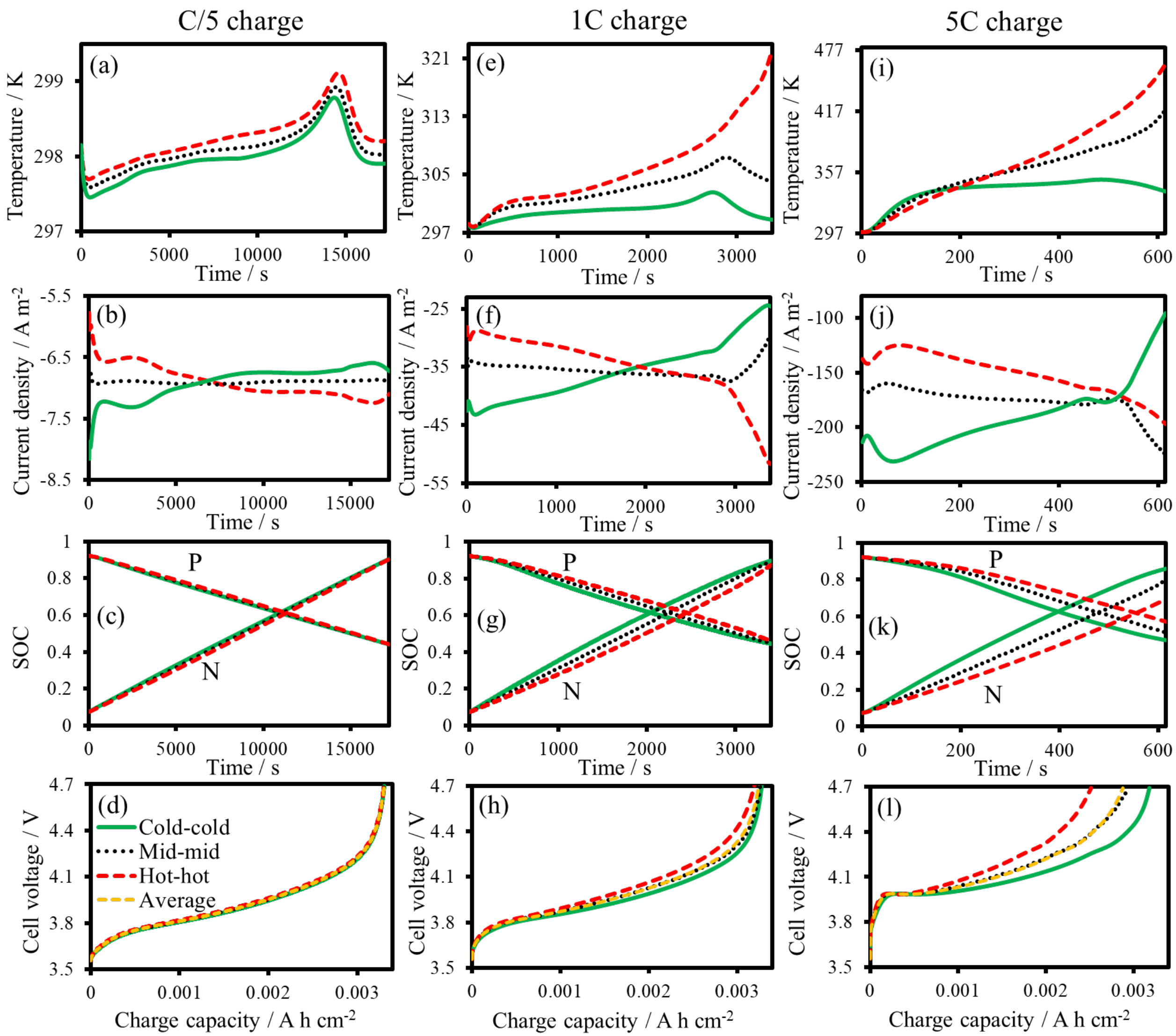
Divide Electrodes into Separate Hot/Middle/Cold Regions

N_M stands for MacMullin Number, and ε_f is volume fraction of the filler (carbon additives and binder).

		Cold spot	middle spot	Hot spot
Negative electrode [43, 111]	ε_N	0.37	0.35	0.33
	ε_f	0.01	0.03	0.05
	τ_N	2	5	8
	N_M	5.4	14.3	24.2
Positive electrode [59, 111]	ε_P	0.37	0.35	0.33
	ε_f	0.01	0.03	0.05
	τ_P	1.5	3	4.5
	N_M	4.1	8.6	13.6



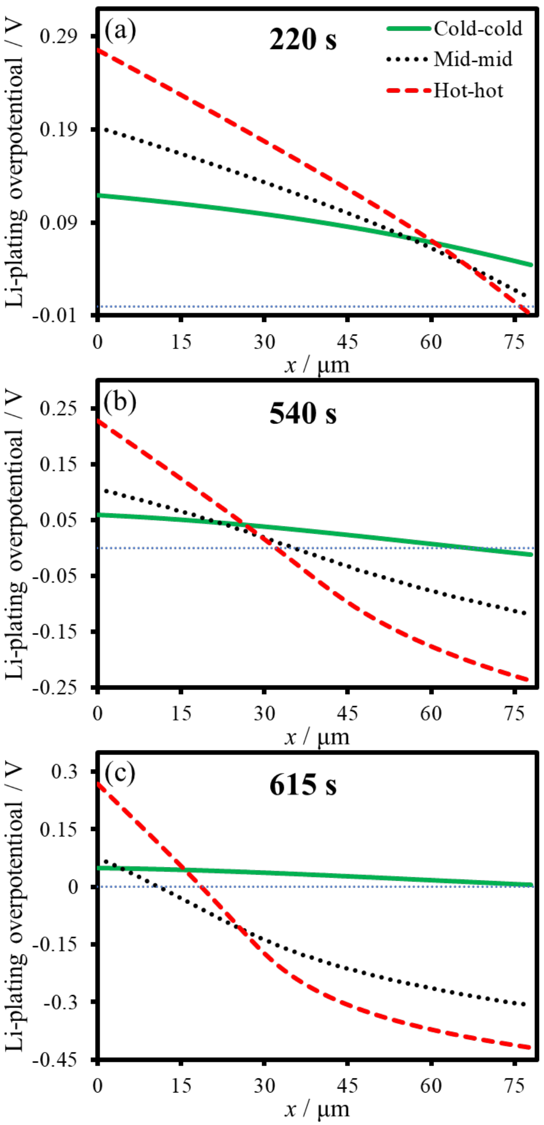
Model Results for Different Regions: Charging NMC-GR Heterogeneous Electrodes



Tendency for Lithium Plating During Fast Charge (5C)

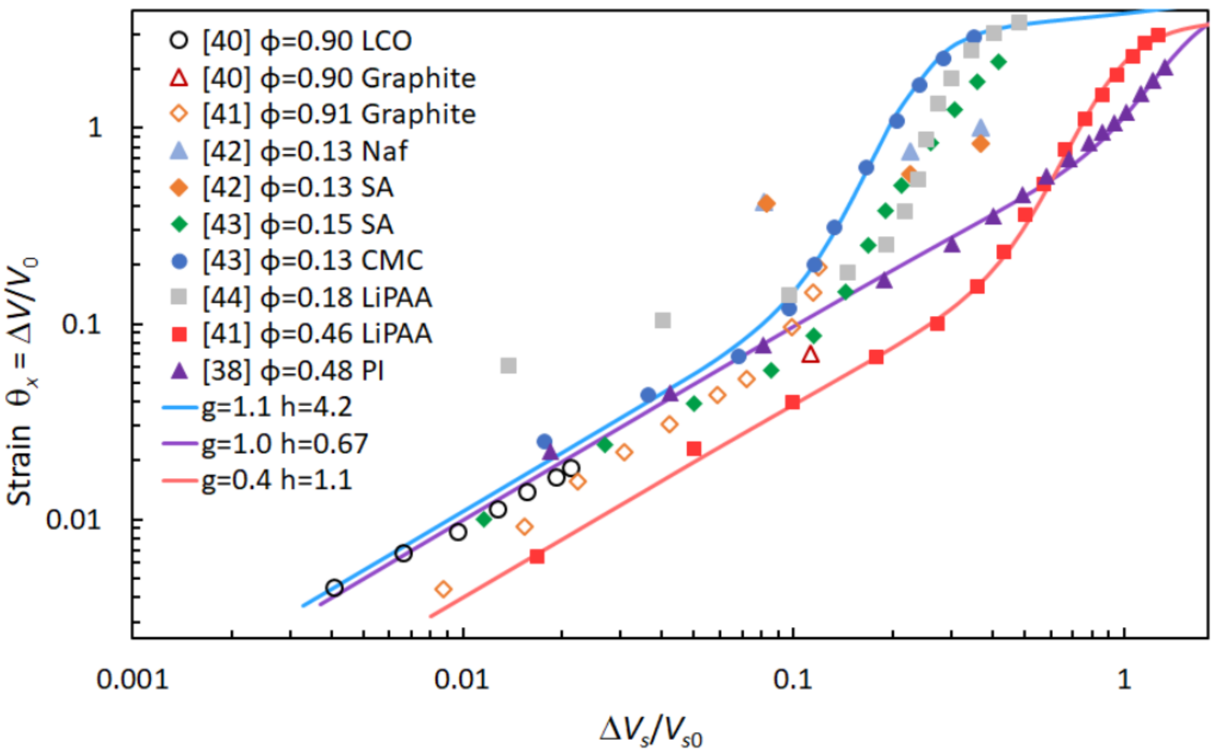
Li can plate when overpotential negative

Plating occurs for growing regions in anode, beginning next to separator



SILICON ELECTRODES UNDERGO TREMENDOUS STRAIN

On full lithiation, active material has 275% volume change (2% for LCO ad 12% for graphite)



This plot shows large measured strains also of film

Hamedi et al, manuscript in preparation

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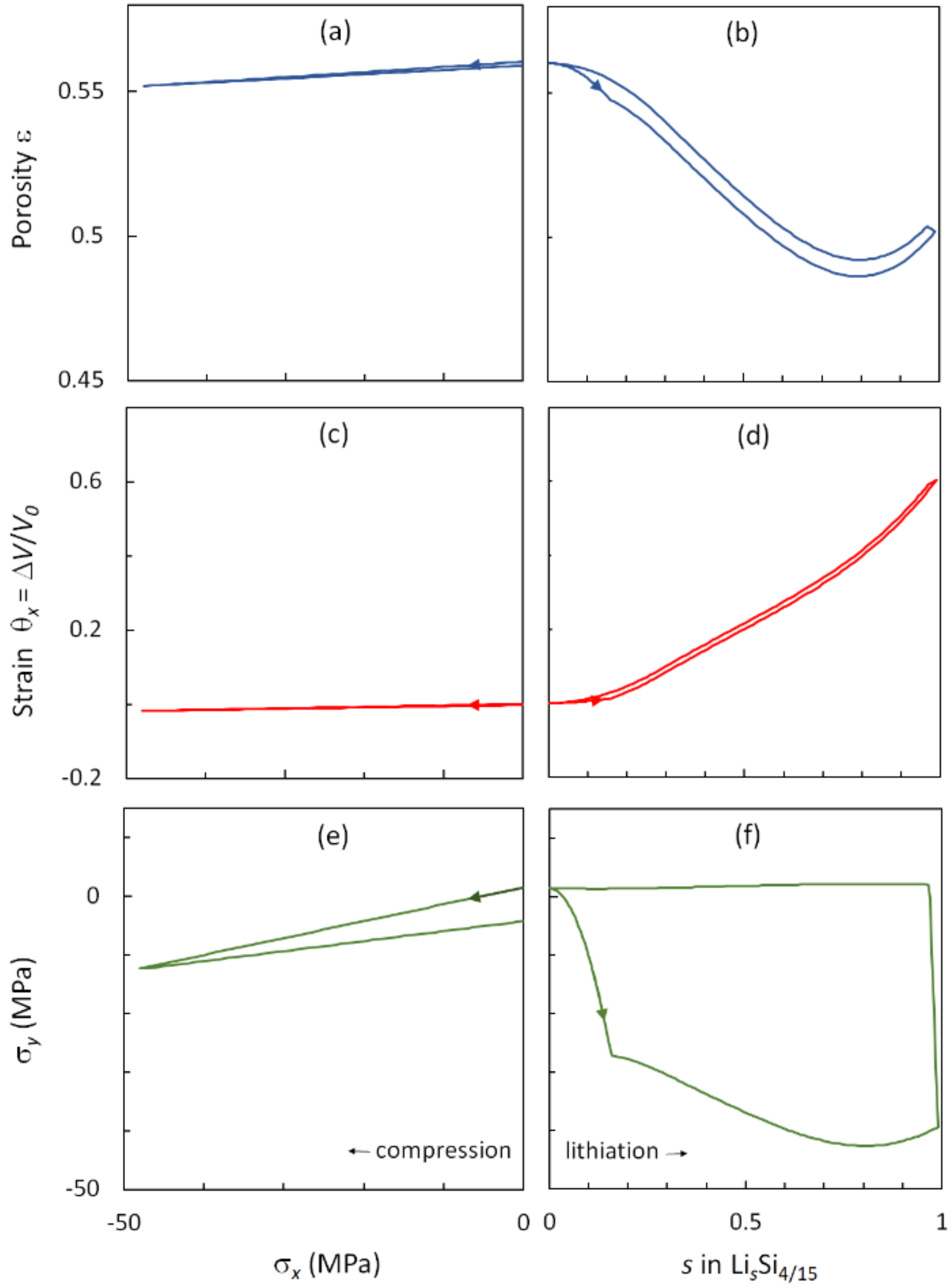
8. P2D MODEL EXTENSIONS

D. STRESS AND STRAIN FOR SILICON-BASED ELECTRODES



Hamedi et al, manuscript in preparation

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