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Analysis of Structure Transport Interactions in Lithium Ion Batteries Supported by X Ray Imaging

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Overview

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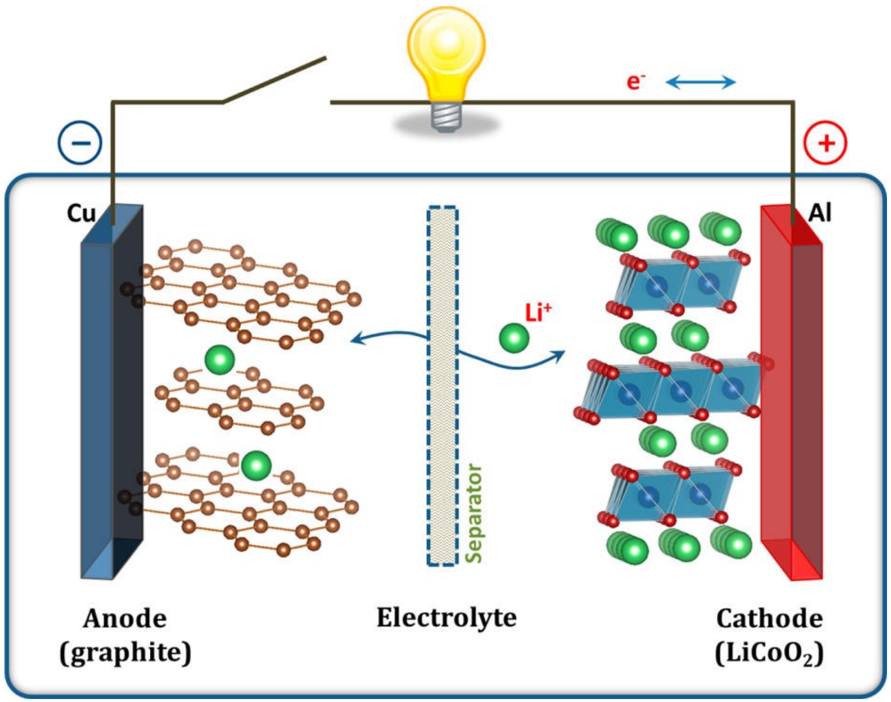
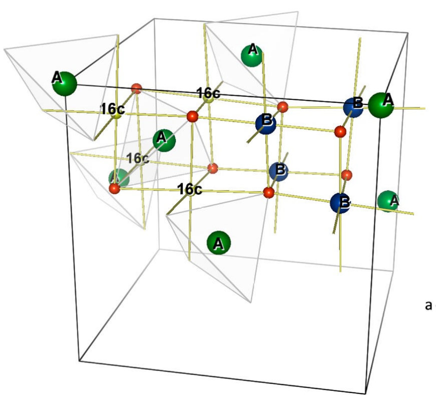
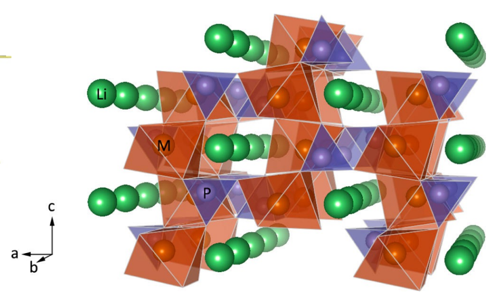
- The need for direct imaging of batteries
- Li-ion cathode characterization using X-ray nanotomography
- Particle scale intercalation studies
- Li-ion cathode characterization using X-ray microtomography (µCT)
- Analyzing electrode structure for fast charging of thicker electrodes
- Summary

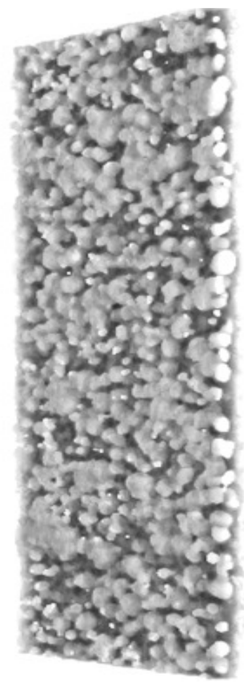
The Need for Direct Imaging

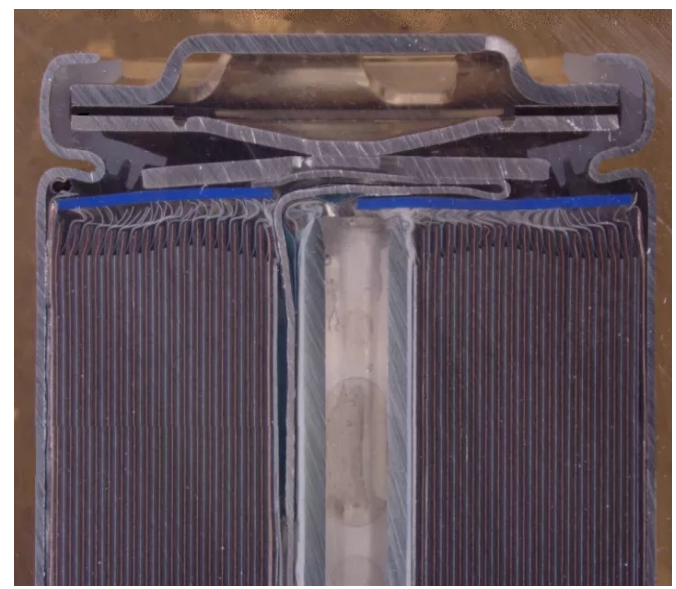


- Batteries are heterogeneous multiscale functional material systems.
- Multiscale design is a necessity.
- Direct imaging methods are a key tool for multiscale design.









https://www.electricbike.com/inside-18650-cell/

Goodenough and Park, *J. Am. Chem. Soc.*, 135, 1167–1176, 2013.

Nanotomography and µCT

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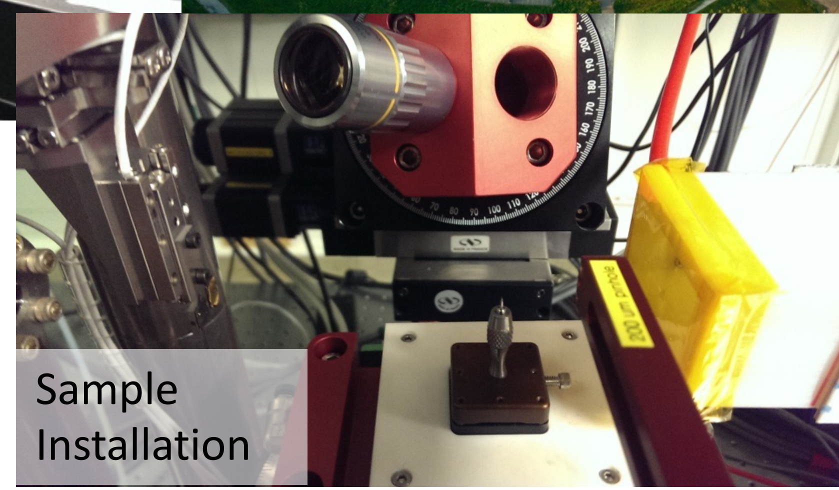
Argonne National Lab
Advanced Photon Source



Sample Installation

APS Beamline 32-ID-C TXM

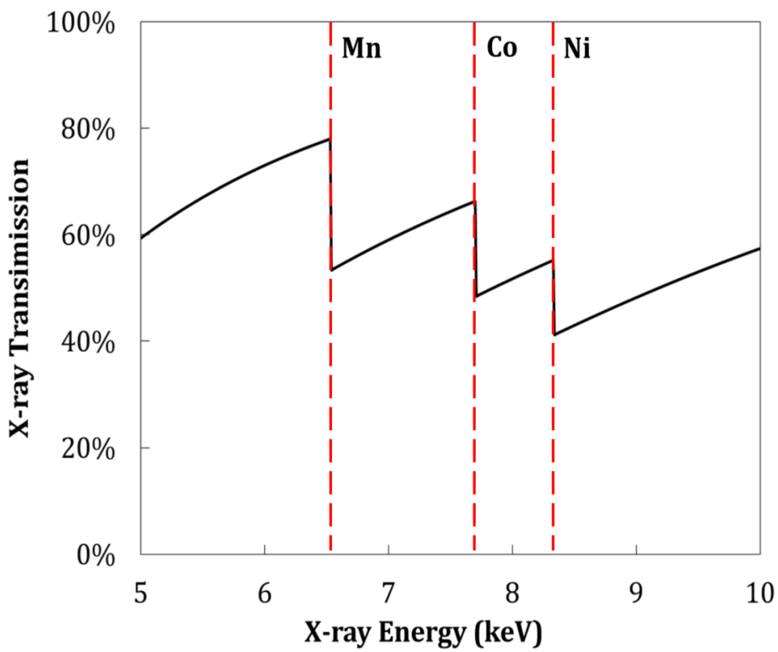
- X-ray energy: 7-40 keV
- 60 nm spatial resolution

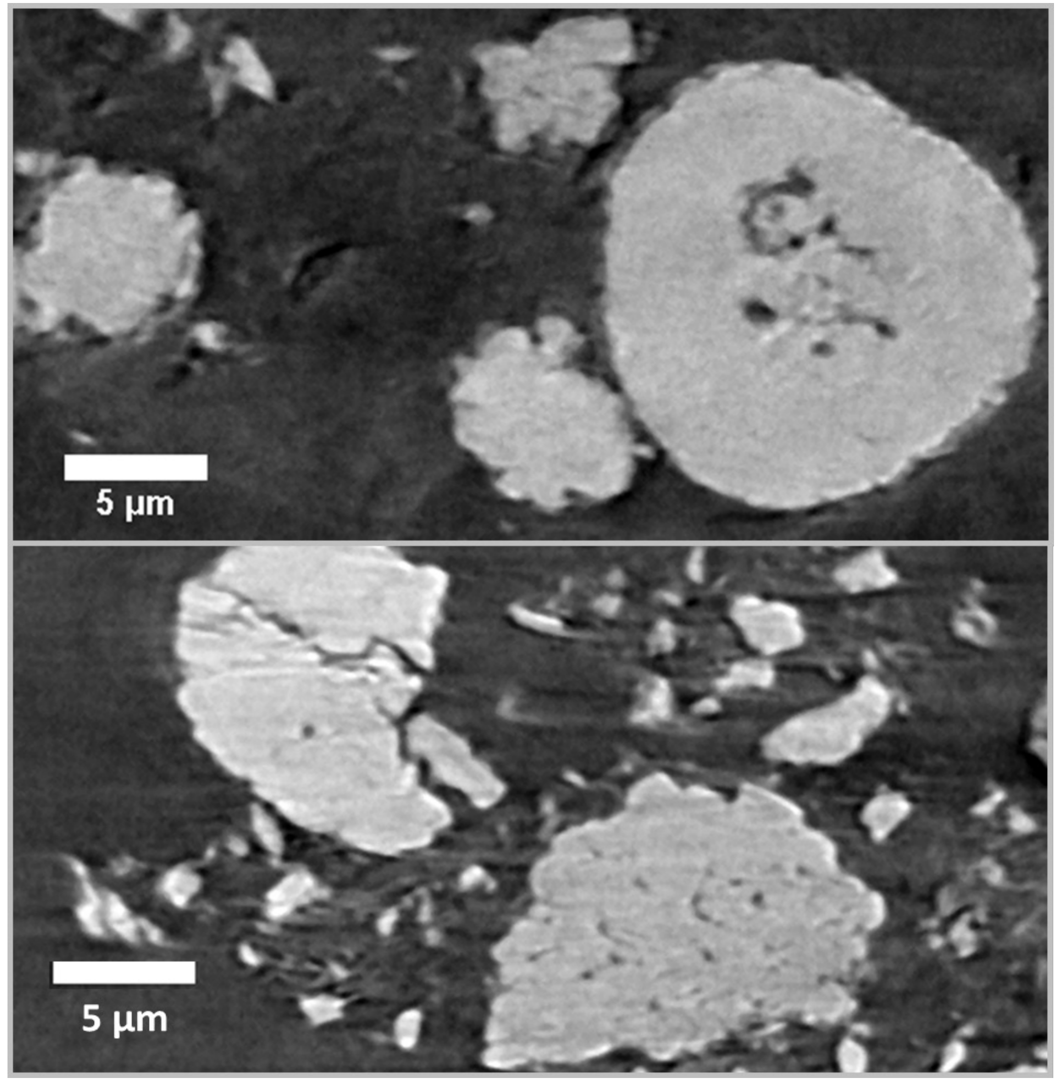
APS Beamline 2-BM-A

- X-ray energy: 11-35 keV
- 1.3 µm spatial resolution

NMC Cathode Nanotomography

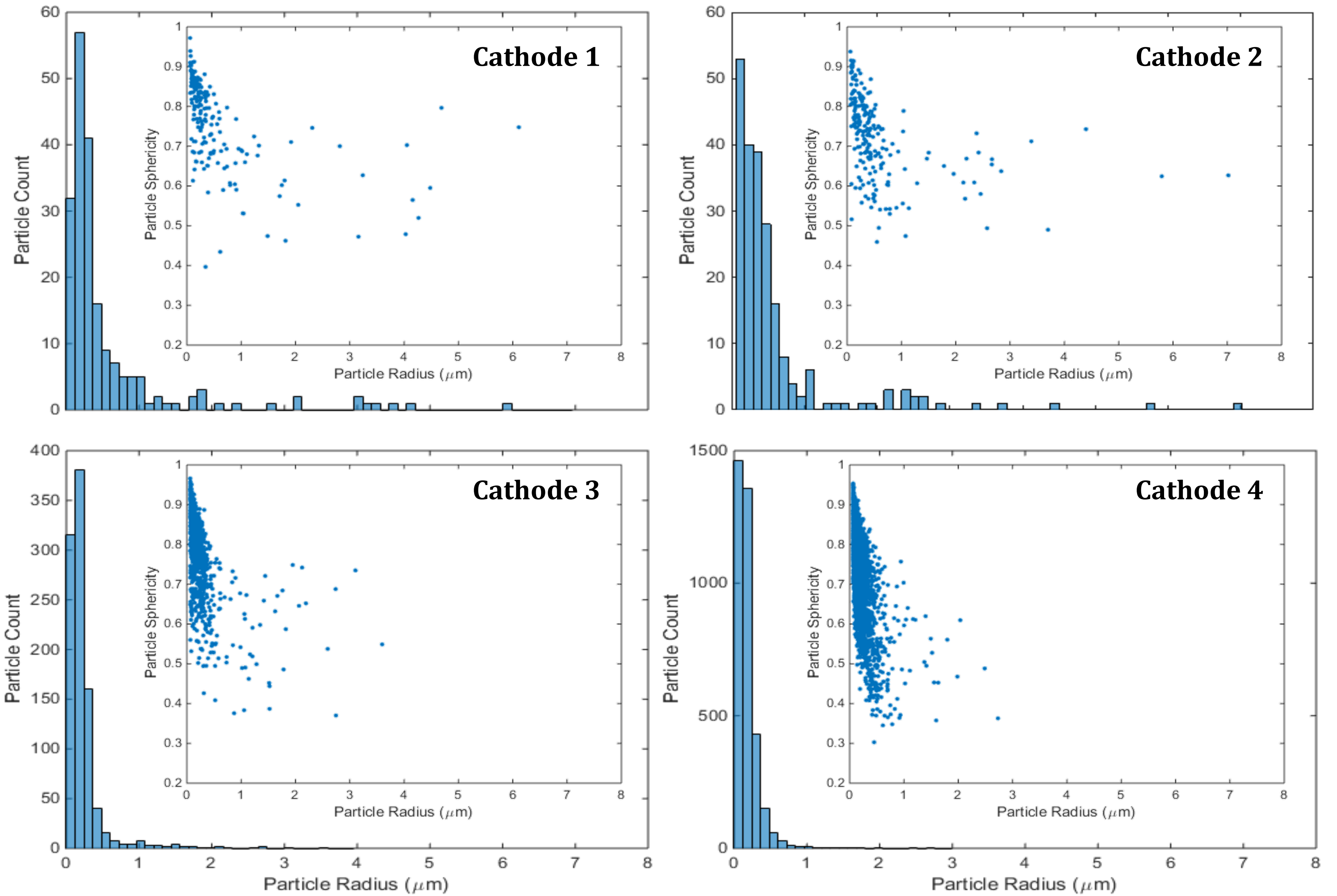
- Processing variants
  - Cathode 1: Gradual drying
  - Cathode 2: Rapid drying
  - Cathode 3: Rapid drying followed by calendering
  - Cathode 4: Ball milling followed by gradual drying





Nelson et al., *J. Electrochem. Soc.*, **164**(7) A1412-A1424, 2017.

Particle Size and Sphericity



Nelson et al., *J. Electrochem. Soc.*, **164**(7) A1412-A1424, 2017.

Particle Intercalation Studies

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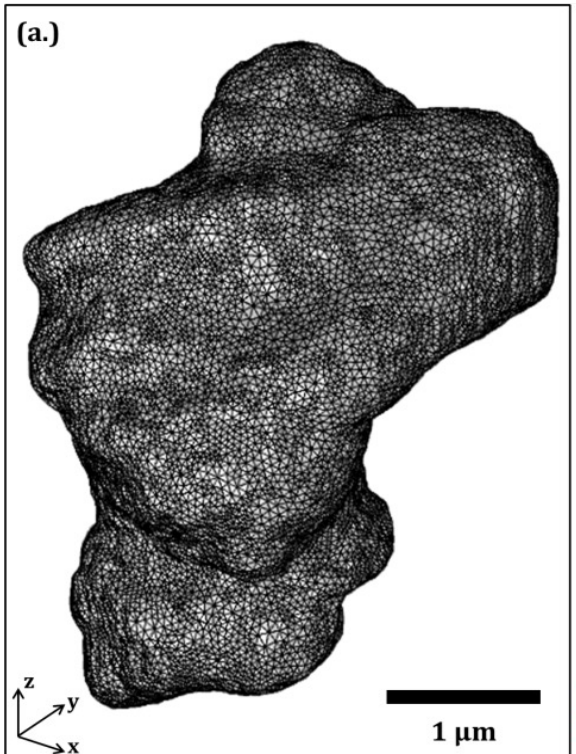
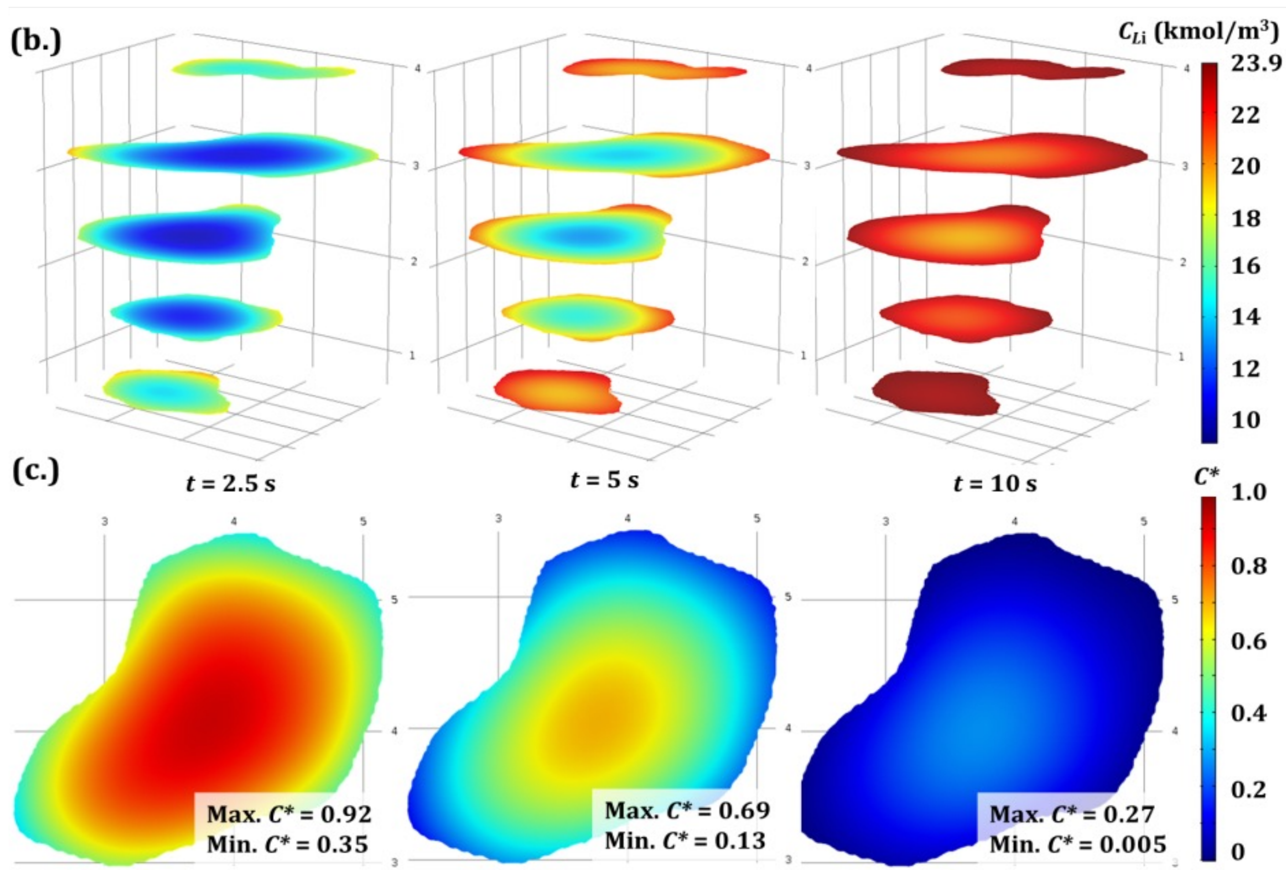
- Simulation of lithium diffusion, particle as sole domain (COMSOL Multiphysics).
- Butler-Volmer with fixed overpotential and galvanostatic operation.
- Intercalation characterized based on mass transfer Biot number and Fourier number.



Li Concentration (mol/m^3)



Particle Characteristics

- Radius: 1.3 μm
- Sphericity: 0.7

Nelson et al., *J. Electrochem. Soc.*, **164**(7) A1412-A1424, 2017.

Geometry Effect on Intercalation

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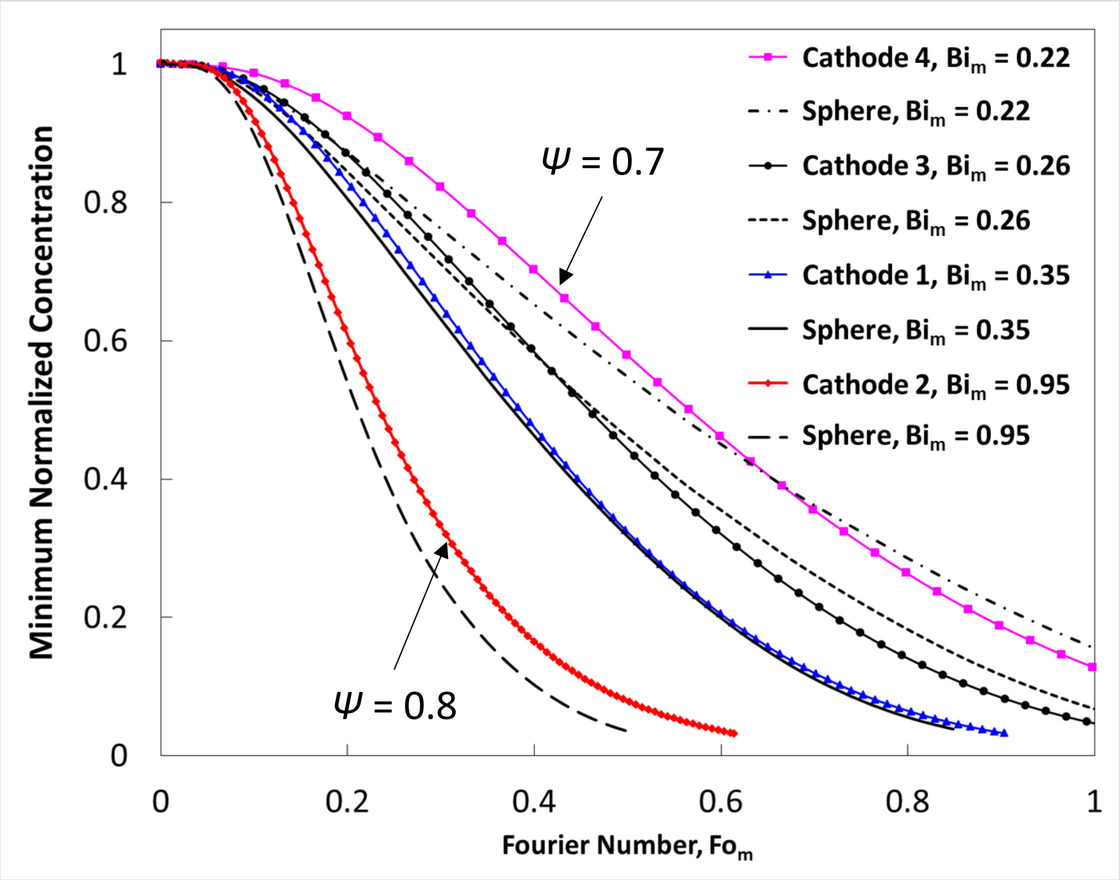
- Four particles extracted from cathode data sets.
- Compared to spheres of equivalent Bi_m

$$Bi_m = \frac{k_{eff} L_C}{D_{Li}} \qquad Fo_m = \frac{D_{Li} t}{r^2}$$

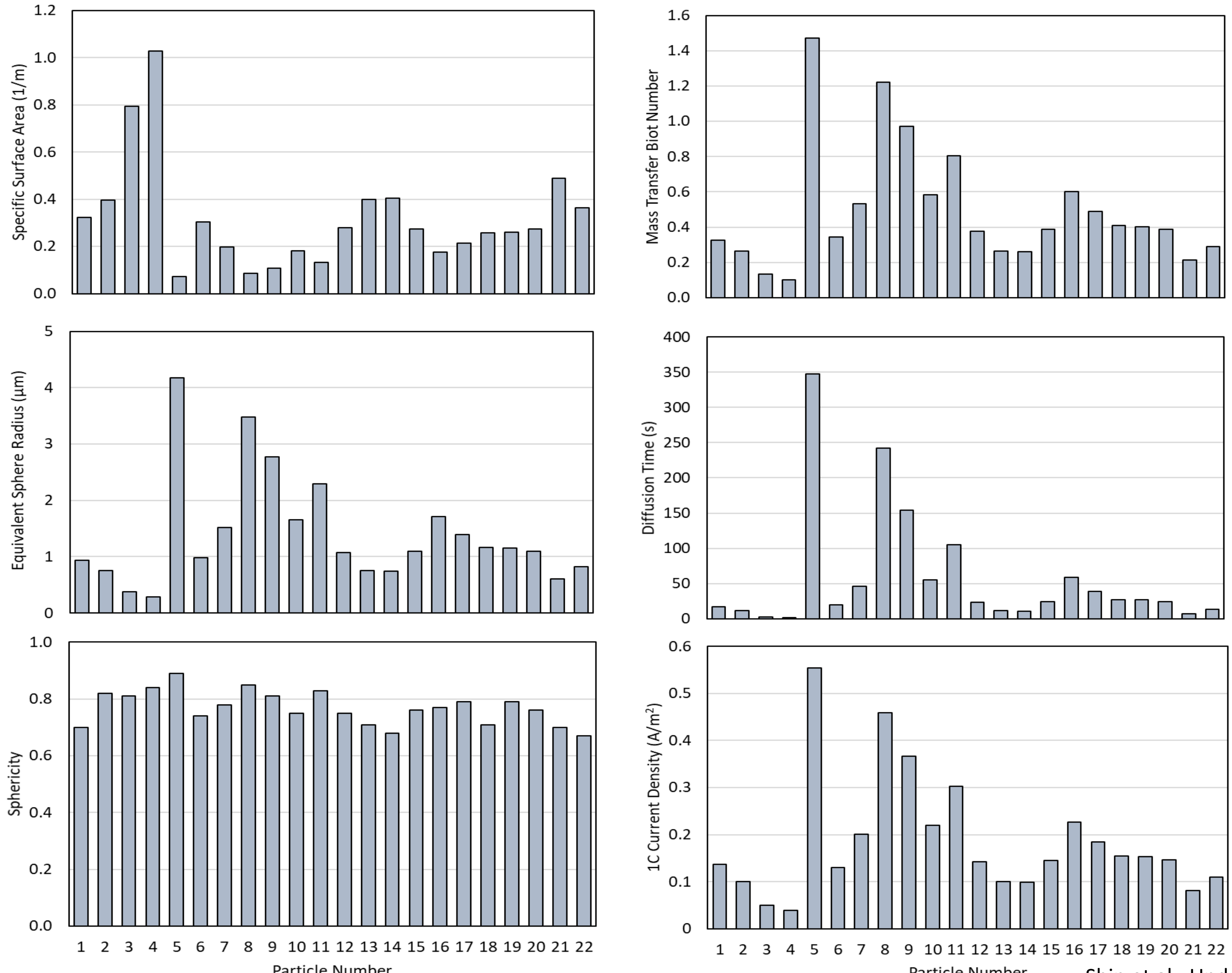
- Departure from spherical particle model depends on sphericity (ψ) and Bi_m.



Nelson et al., *J. Electrochem. Soc.*, **164**(7) A1412-A1424, 2017.

Metric	Cathode 1	Cathode 2	Cathode 3	Cathode 4
Characteristic Length (nm)	330	900	250	200
Mean CSD Length (nm)	370	910	270	230
Particle Biot Number	0.35	0.95	0.26	0.22

Assessing the Spherical Model



Shin et al., Under Review, 2019.

Galvanostatic Case Studies

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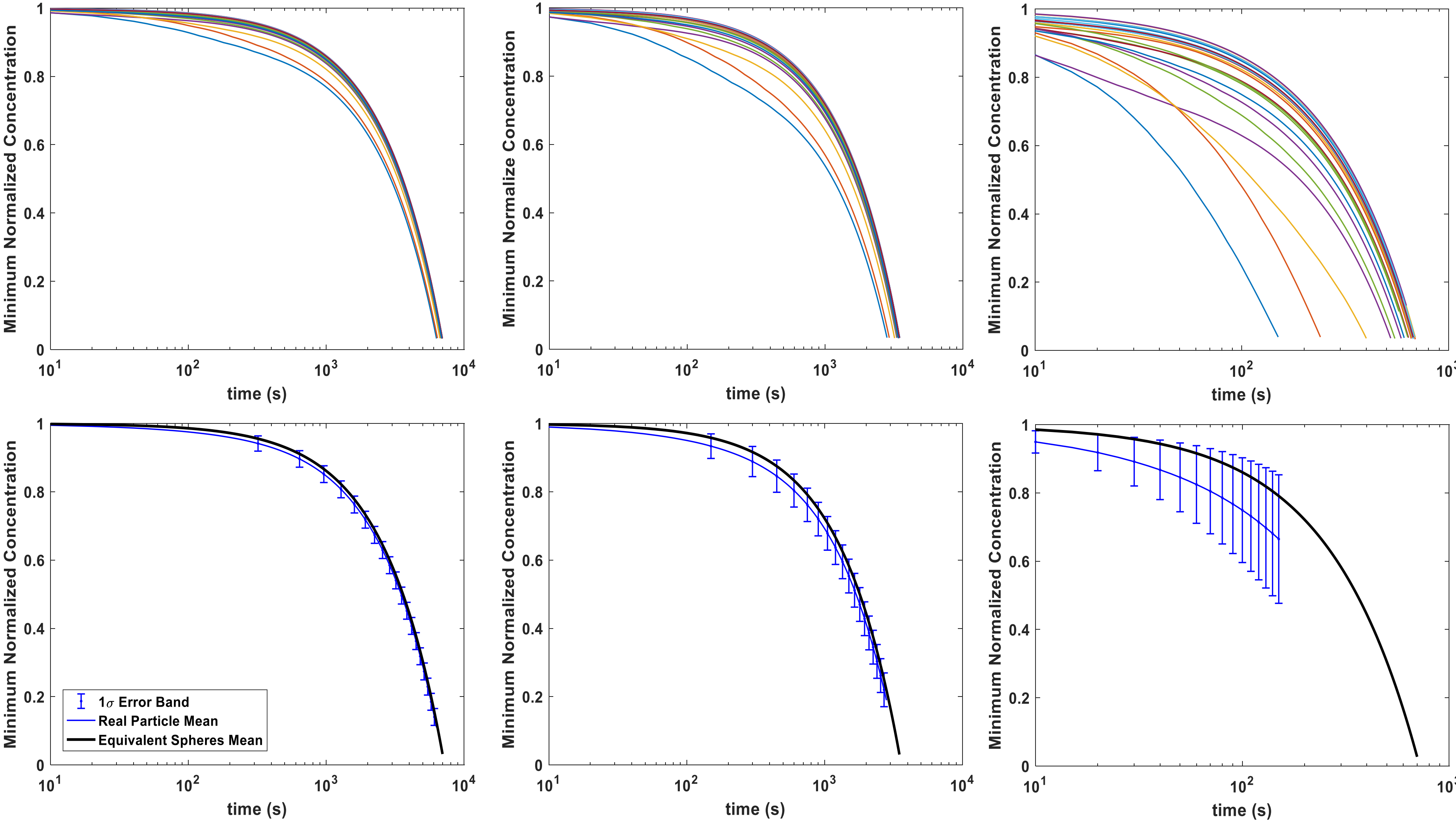

Shin et al., Under Review, 2019.

- Departure from spherical behavior seen at high C-rate.
- Increased discrepancy observed for lower sphericity.

Galvanostatic Case Studies

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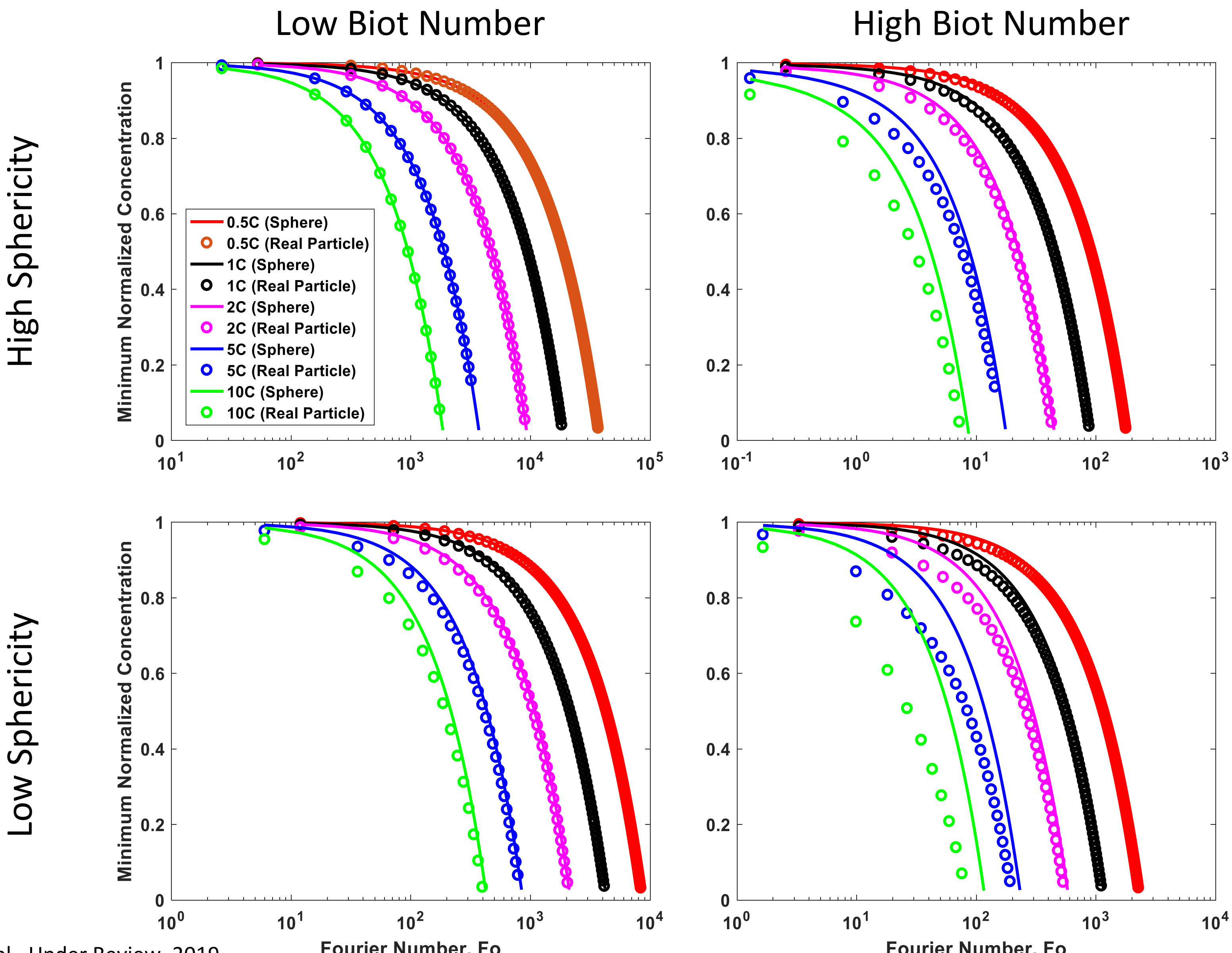
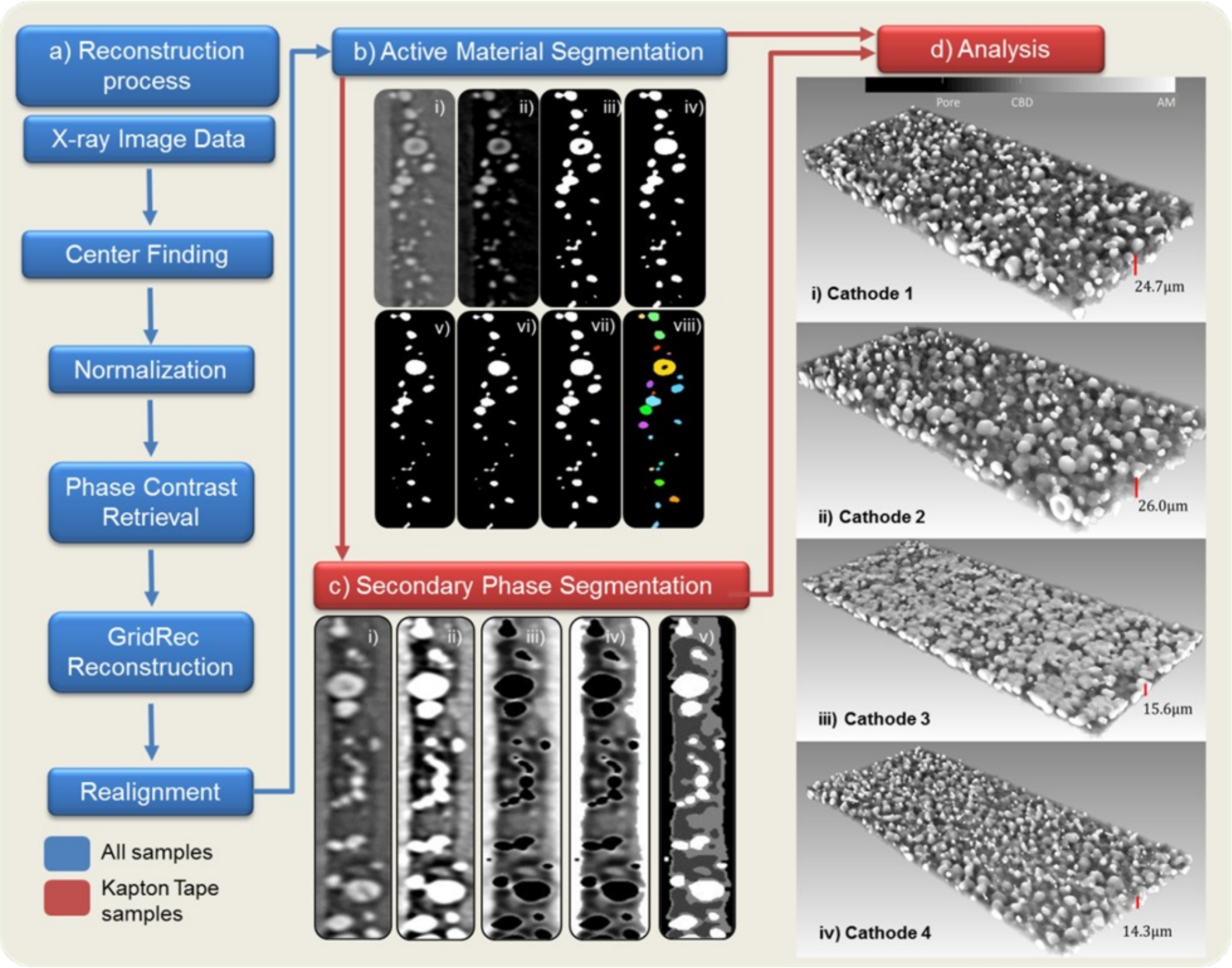


Shin et al., Under Review, 2019.

NMC Cathode Microtomography

- White beam µCT applied with phase contrast capability.
- Two sample encapsulation methods used: epoxy and Kapton tape.
- Phase contrast data preserved with Kapton tape samples.
- Watershed segmentation applied for AM.
- Secondary phases segmented using histogram data.



Rajendra et al., *ACS Appl. Mater. Interfaces*, 2019 (10.1021/acsami.8b22758).

Micron Scale Processing Effects

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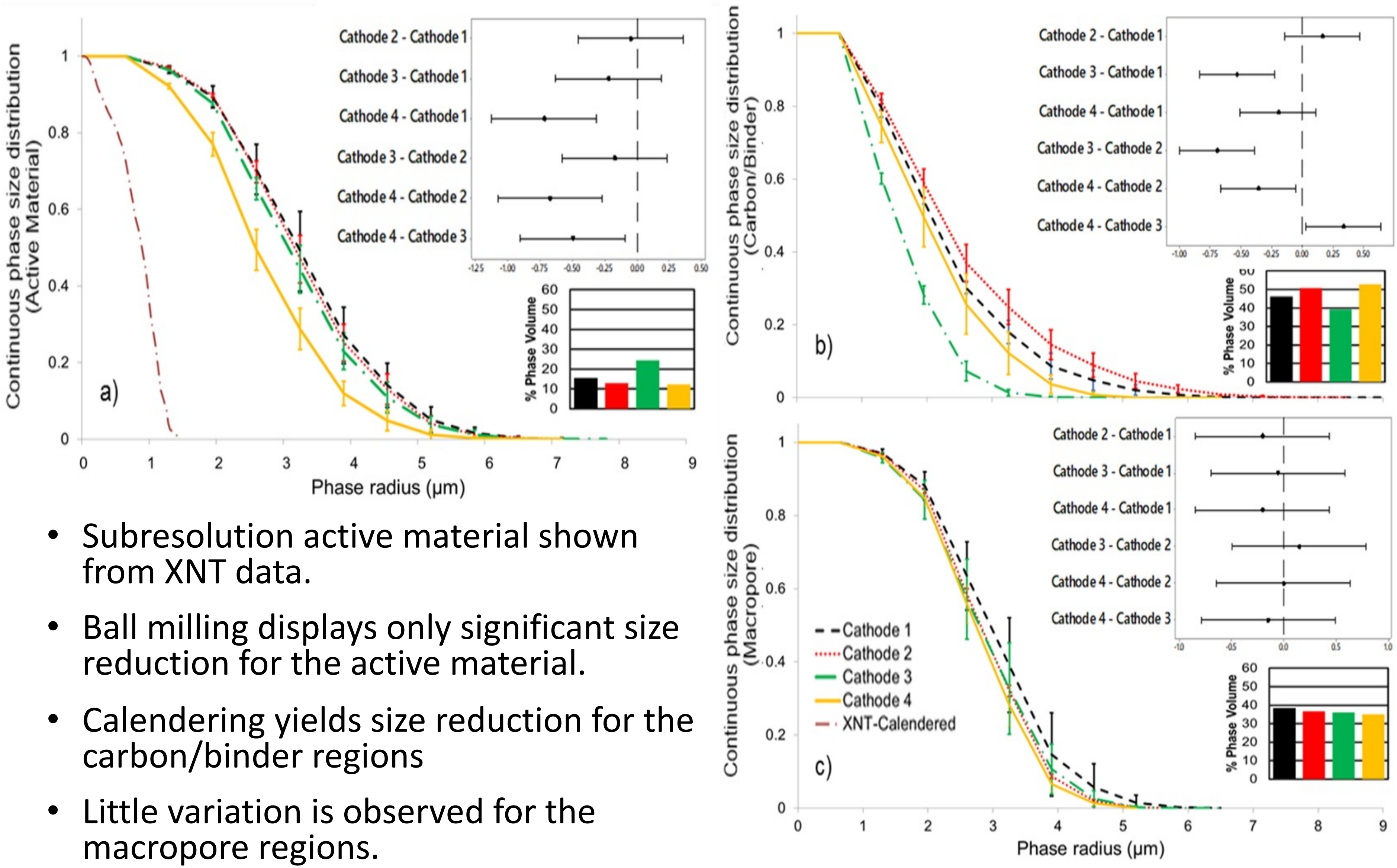

- Subresolution active material shown from XNT data.
- Ball milling displays only significant size reduction for the active material.
- Calendering yields size reduction for the carbon/binder regions
- Little variation is observed for the macropore regions.

Rajendra et al., *ACS Appl. Mater. Interfaces*, 2019 (10.1021/acsami.8b22758).

Electrode Performance Effects

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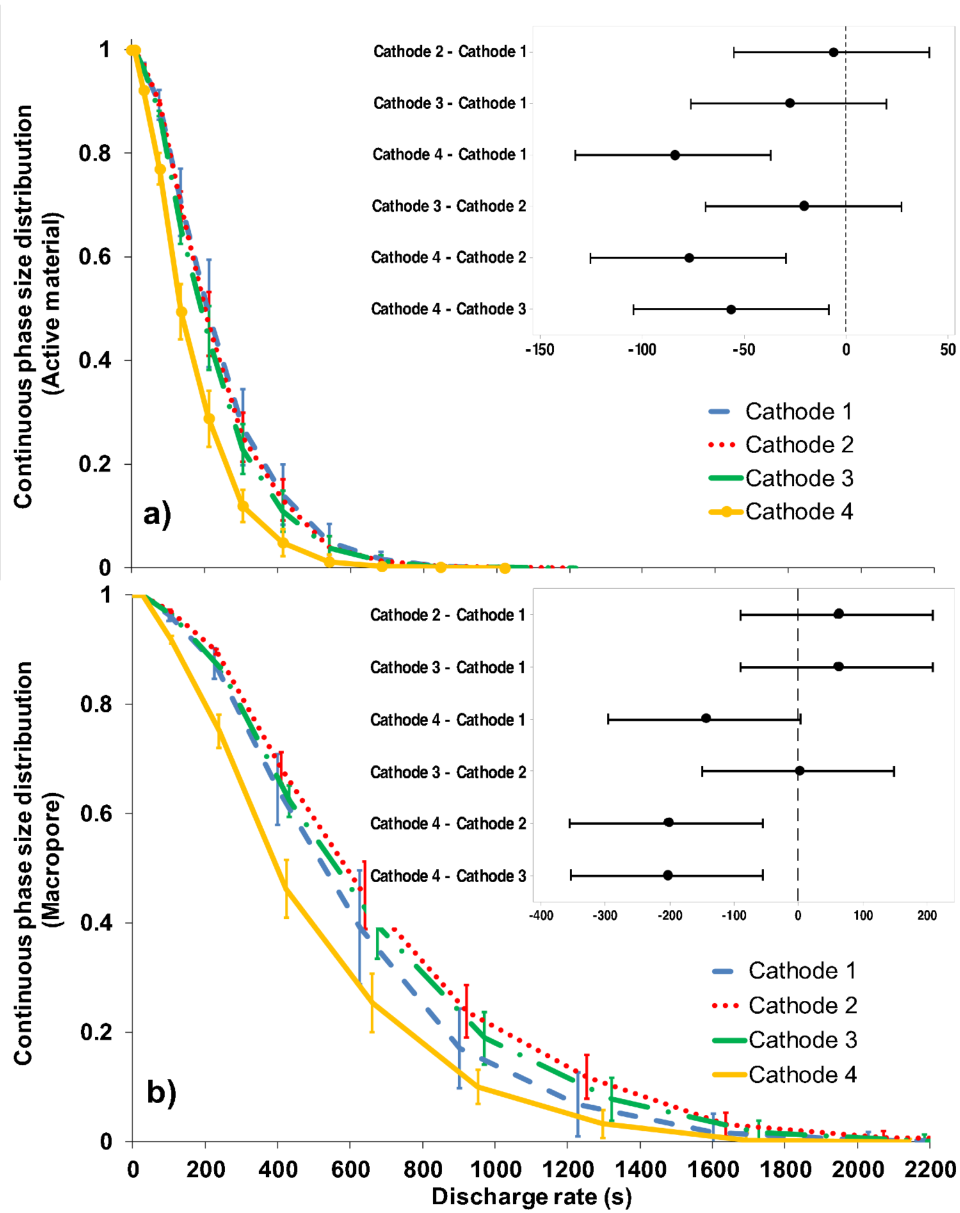
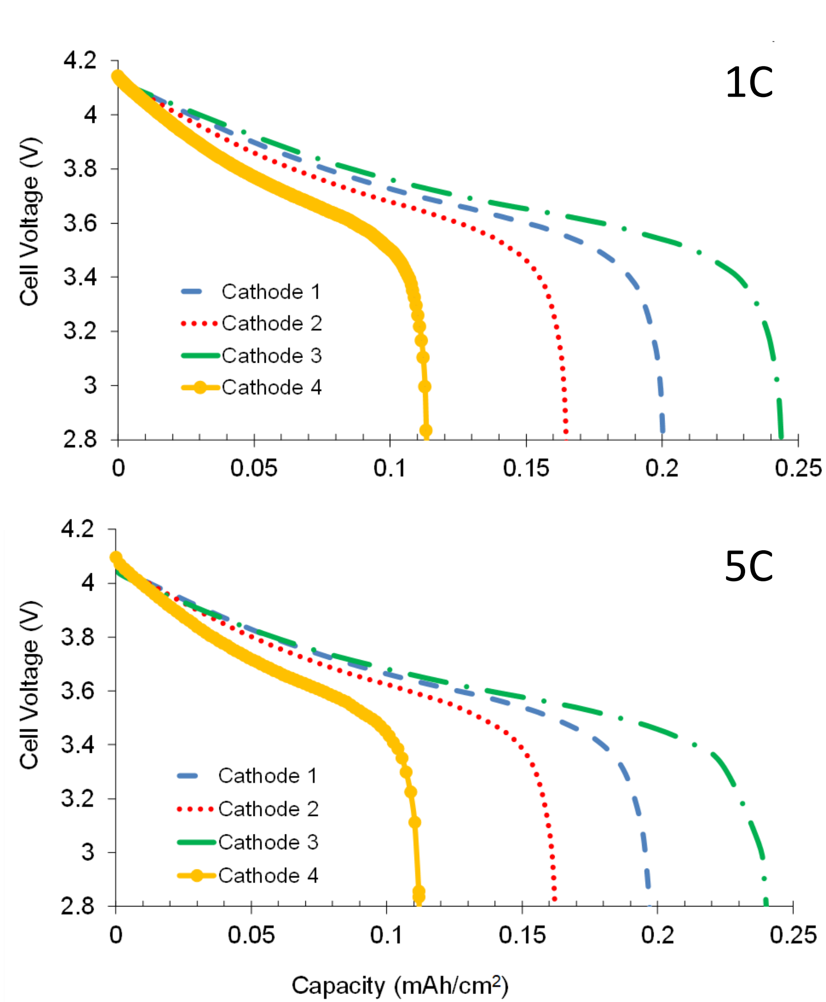
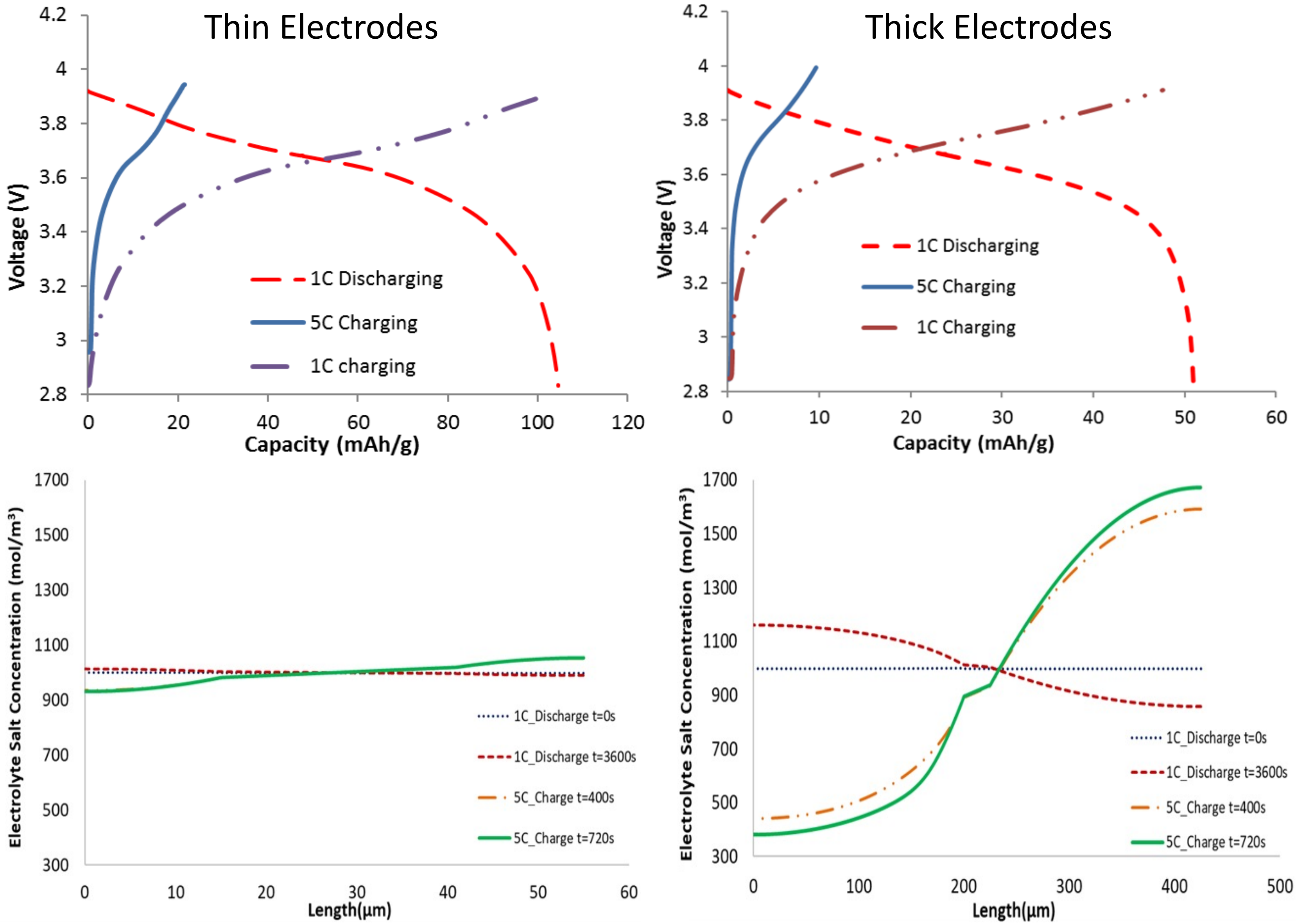
- Phase sizes yield aggregate diffusion time estimates based on $Fo_m = 1$.
- Simulated rate capability follows electrode diffusion time estimates.





Rajendra et al., *ACS Appl. Mater. Interfaces*, 2019 (10.1021/acsami.8b22758).

Response to Fast Charging



P. Patel and G. J. Nelson, J. Energy Resour. Technol., *Accepted*, 2019.

Microstructural Influence

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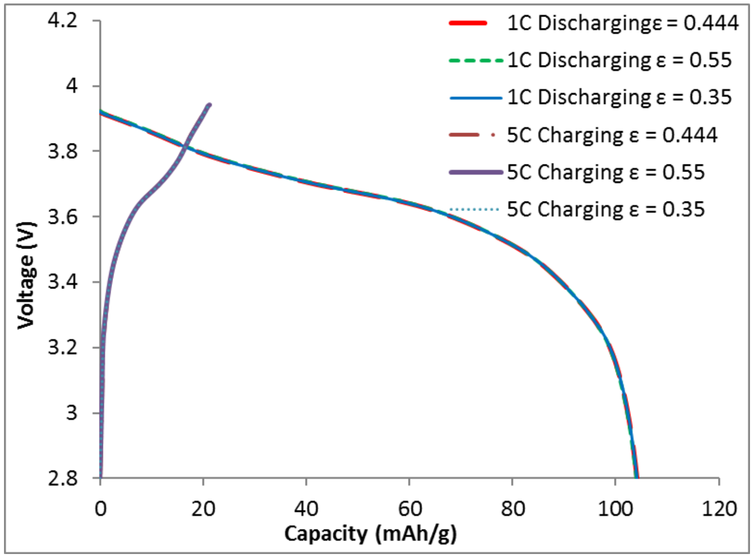
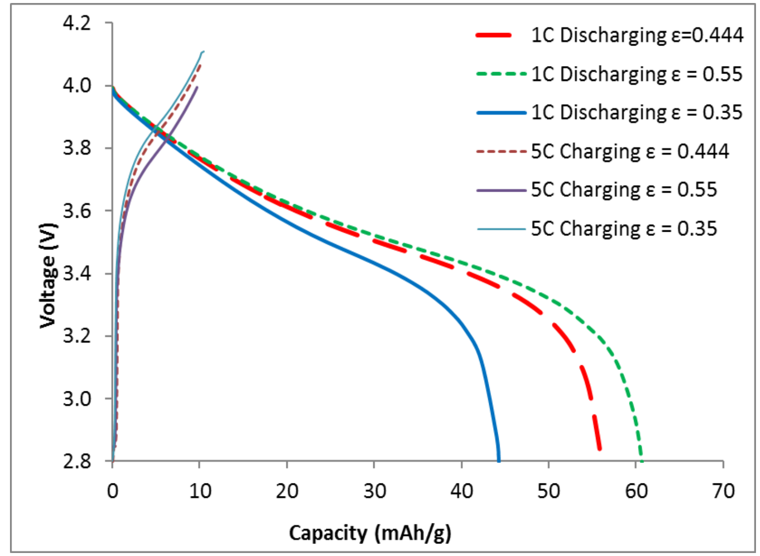
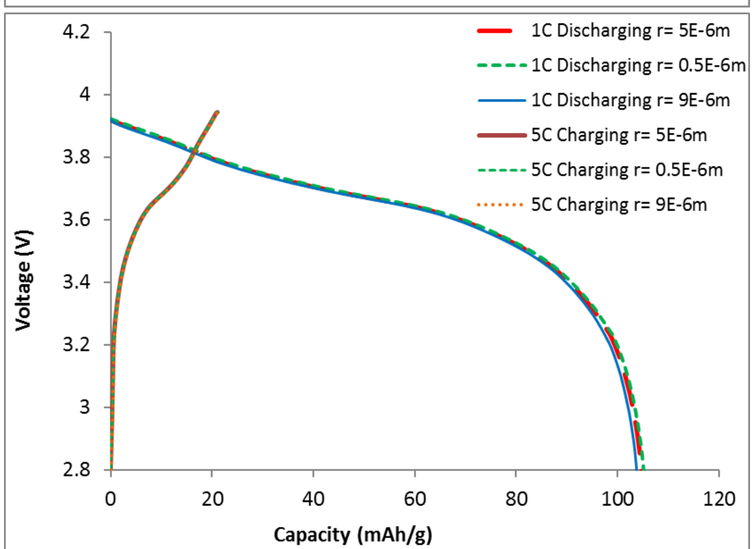
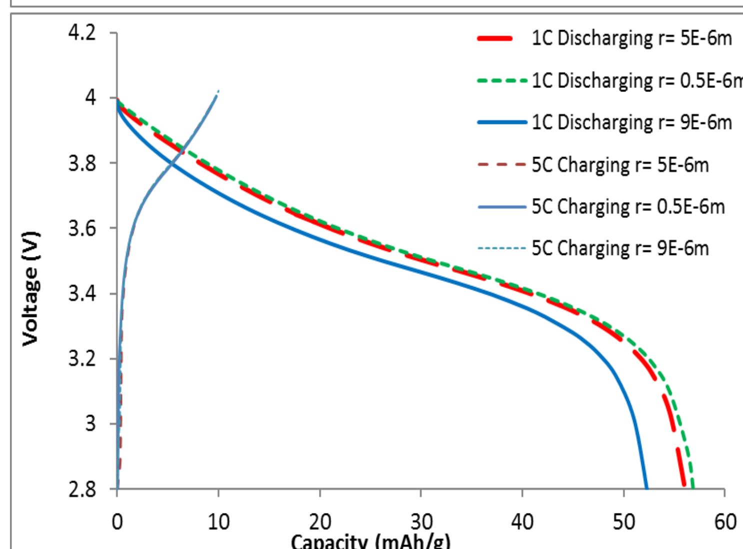
Thin Electrode



Thick Electrode







P. Patel and G. J. Nelson, J. Energy Resour. Technol., *Accepted*, 2019.

Summary

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- Multiscale nature of batteries predicates multiscale direct imaging methods.
- X-ray nanotomography
  - Processing alters particle geometry.
  - Particle geometry influences charge/discharge capabilities.
- X-ray µCT
  - Processing alters active material and secondary phase geometry.
  - Phase geometry influences charge/discharge capabilities.
- The role of geometry can be assessed with appropriate dimensionless metrics.
- Geometry at the microscale and macroscale may be altered to enhance performance, reliability, and safety.

Acknowledgments

- Support
  - NSF Collaborative Research Project (CBET-1438683)
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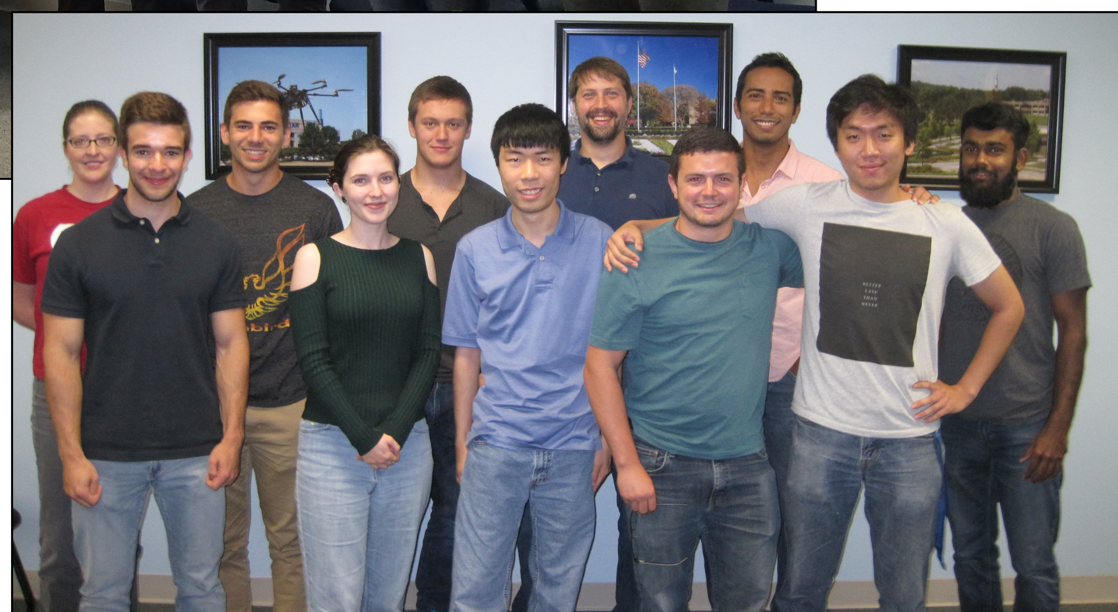
- Collaborators
  - Partha Mukherjee, Aashutosh Mistry, Daniel Juarez-Robles (Purdue)
  - Vincent De Andrade, Xianghui Xiao (Argonne National Lab)

Thank you for your time



Multiscale Transport and Energy Conversion

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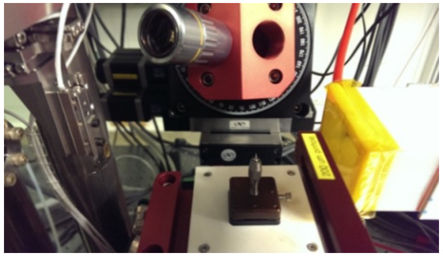
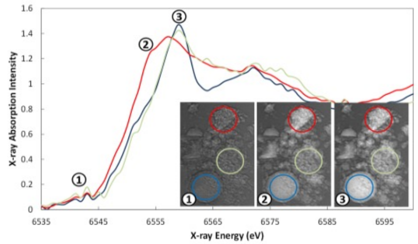
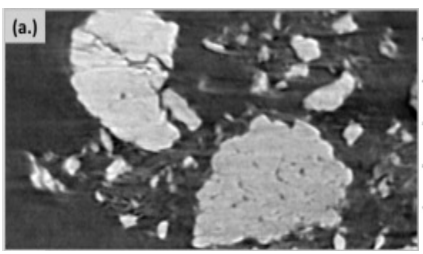
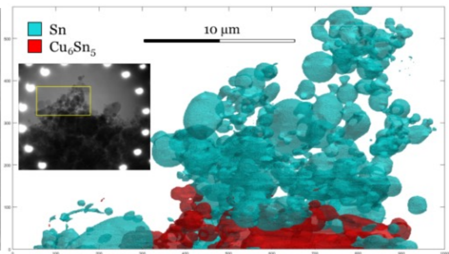
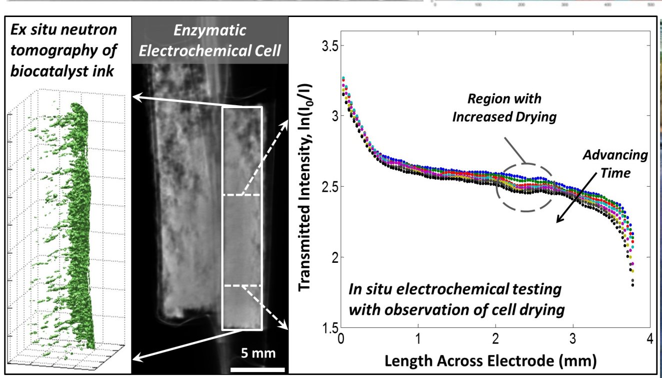
X-ray and Neutron Imaging



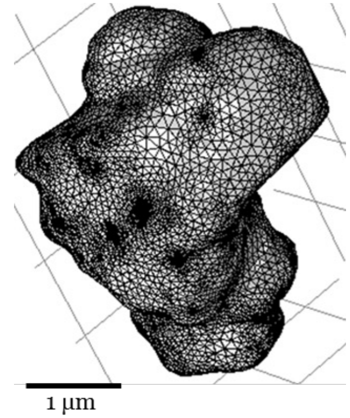




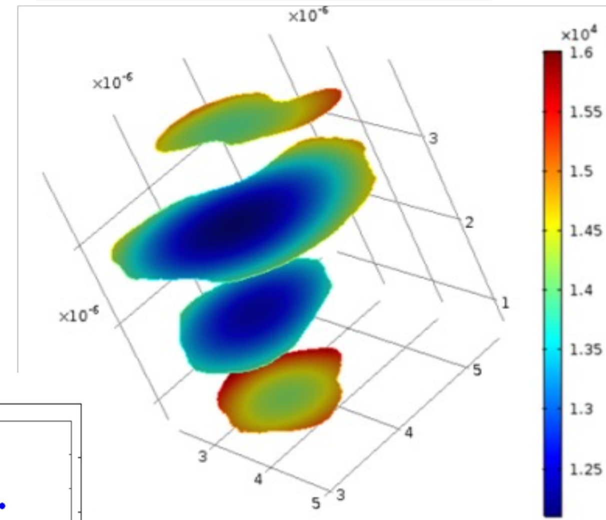


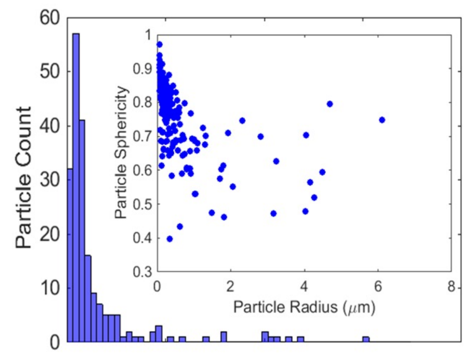


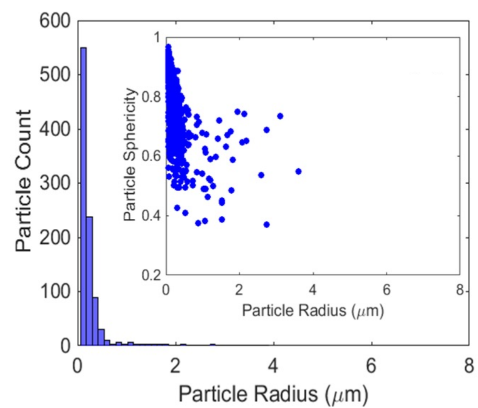
3D Data Analysis & Multiphysics Modeling



Li Concentration (mol/m³)







Projects include:

- X-ray and neutron imaging (3D and *in operando*)
- Microstructural analysis
- Multiphysics FEA
- Device testing
- Materials synthesis

Further details:

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