Electrolyte Motion in Cylindrical Cells: The Effects of Gravity, Material Selection, and Cell Geometry

2025 NASA Aerospace Battery Workshop

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Canadian Light Source

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Overview

Background

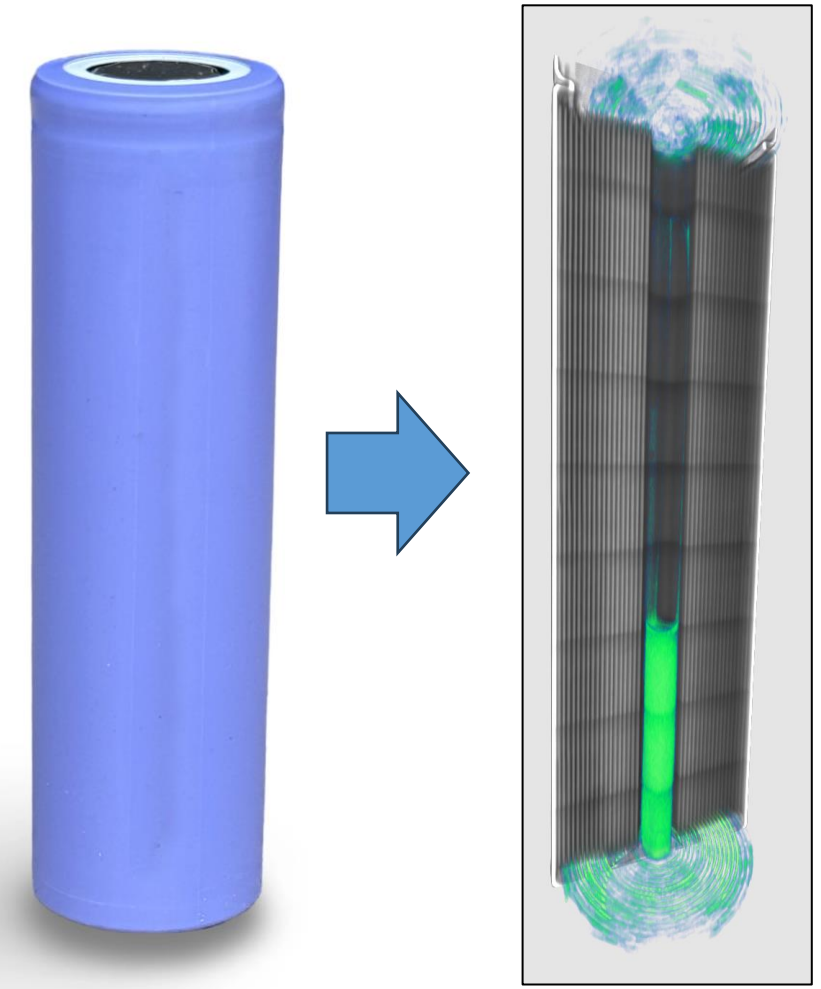
- Non-uniform degradation in large-format cells
- Electrolyte motion induced salt inhomogeneity (EMSI)

Synchrotron Computed Tomography (SR-CT)

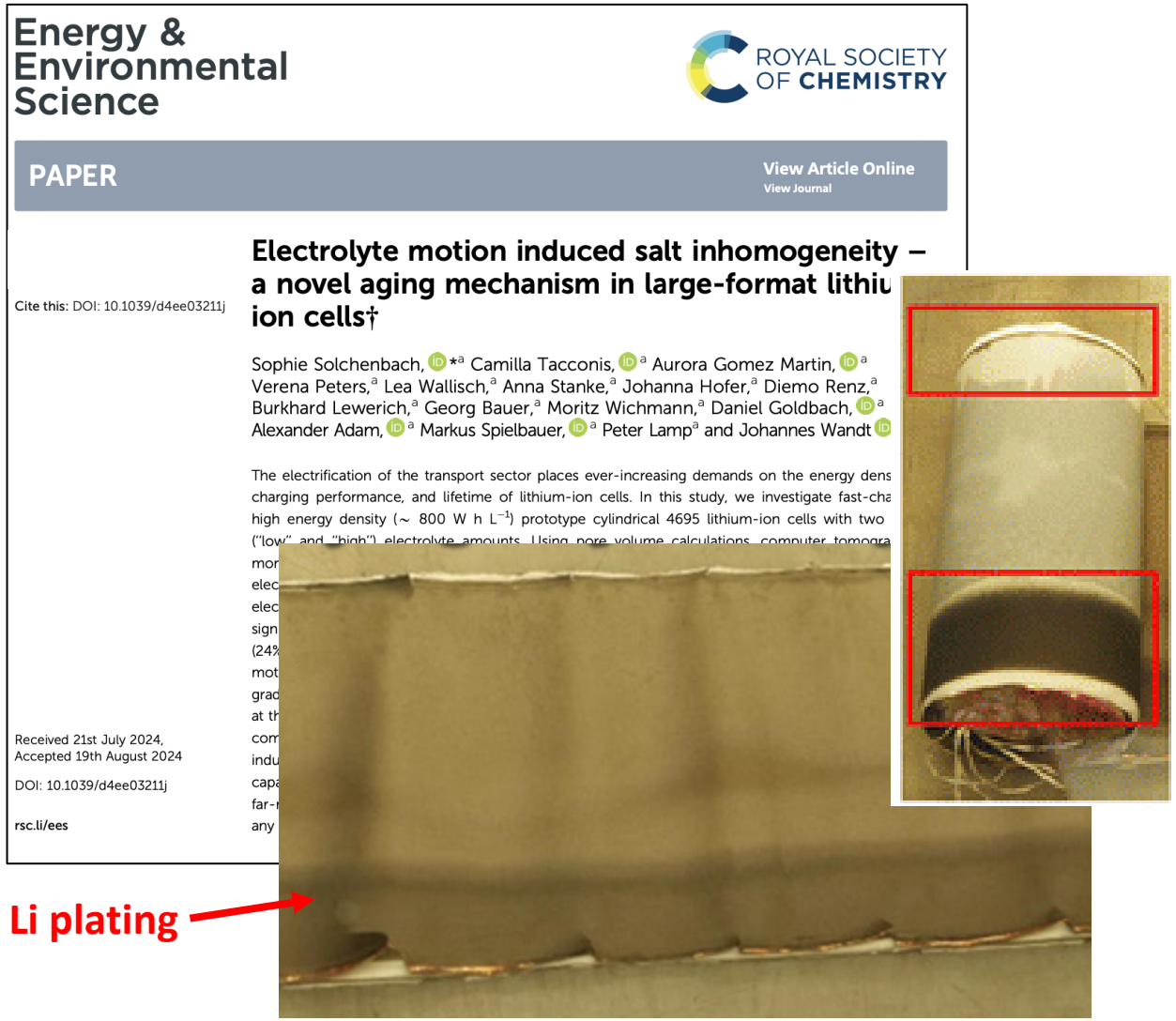
- What is it? why is it useful for battery research?
- Time-resolved and hi-res CT imaging

3D imaging of electrolyte motion in 18650 cells

- Electrolyte reservoirs and recirculation
- Effect of gravity on vertical vs horizontal cells
- Using electrolyte to measure pore volume changes



Li-plating linked to salt redistribution and electrolyte motion



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Electrolyte motion induced salt inhomogeneity – a novel aging mechanism in large-format lithiu ion cells†

Cite this: DOI: 10.1039/d4ee03211j

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The electrification of the transport sector places ever-increasing demands on the energy dens charging performance, and lifetime of lithium-ion cells. In this study, we investigate fast-cha high energy density (~ 800 W h L^{-1}) prototype cylindrical 4695 lithium-ion cells with two ("low" and "high") electrolyte amounts. Using pore volume calculations, computer tomogra

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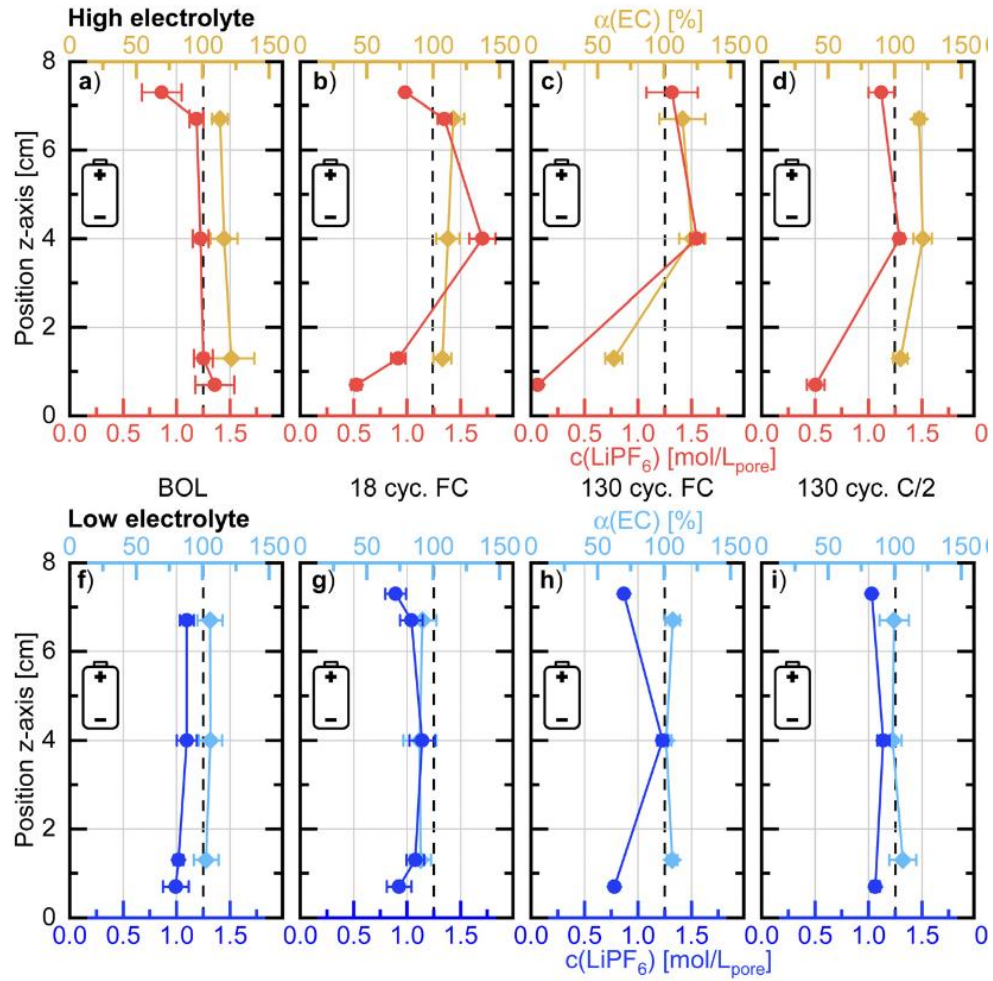

$$Pore\ filling\ ratio = \frac{V_{elyt}}{V_{pore}(SoC)}$$

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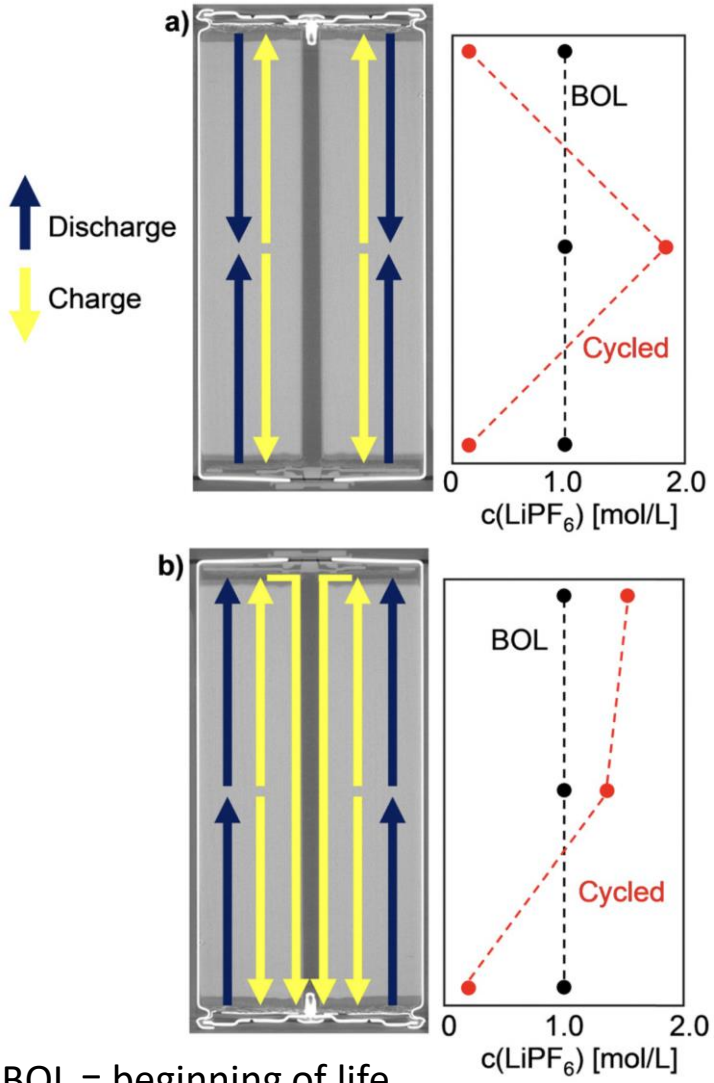
S. Solchenbach et al., Royal Soc. Chem., 19, 7294-7317 (2024)

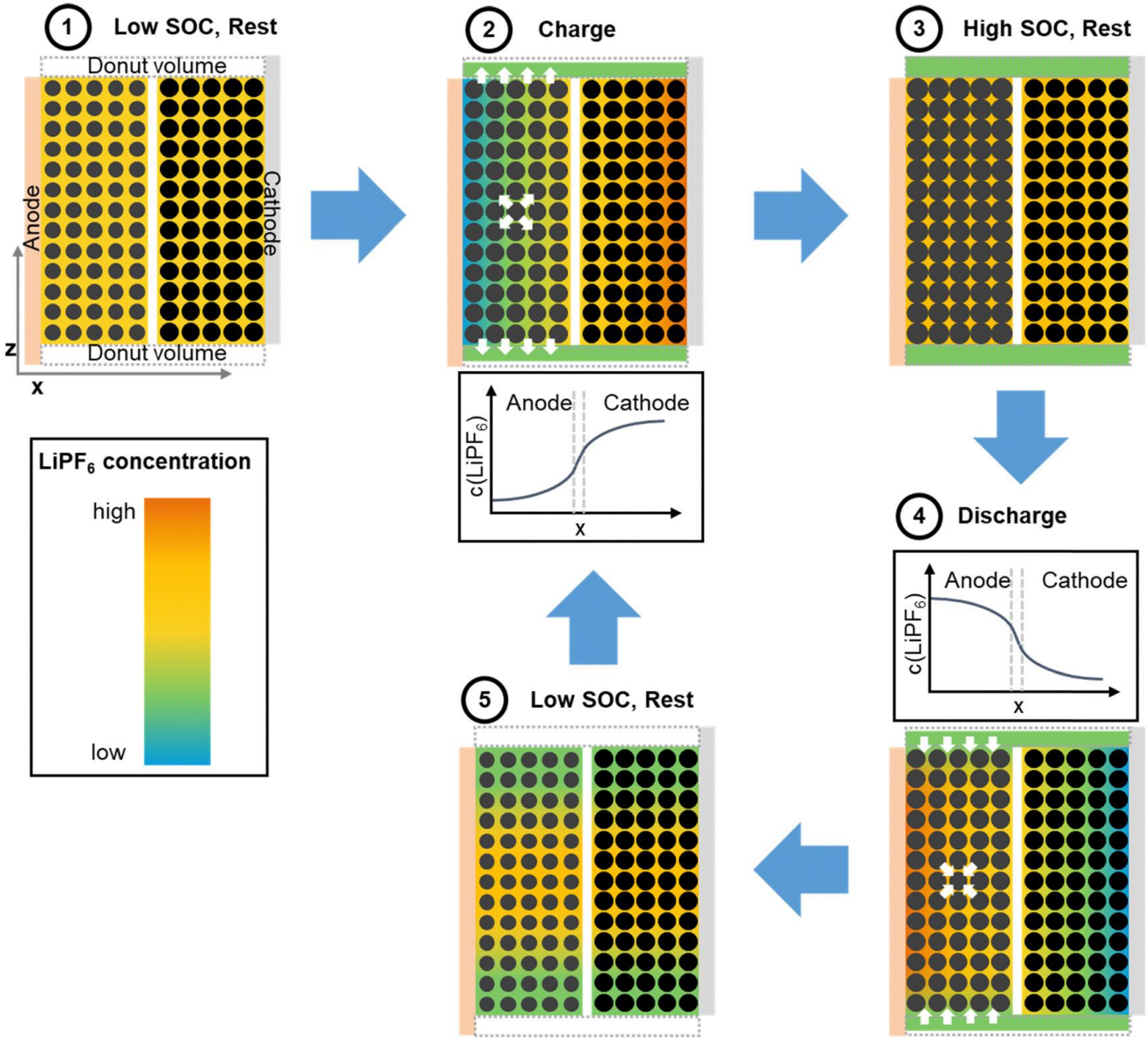
Electrolyte motion induced salt inhomogeneity (EMSI)



Excess electrolyte stored equally at both ends and is reabsorbed

Excess electrolyte pools at the bottom and is recirculated

BOL = beginning of life

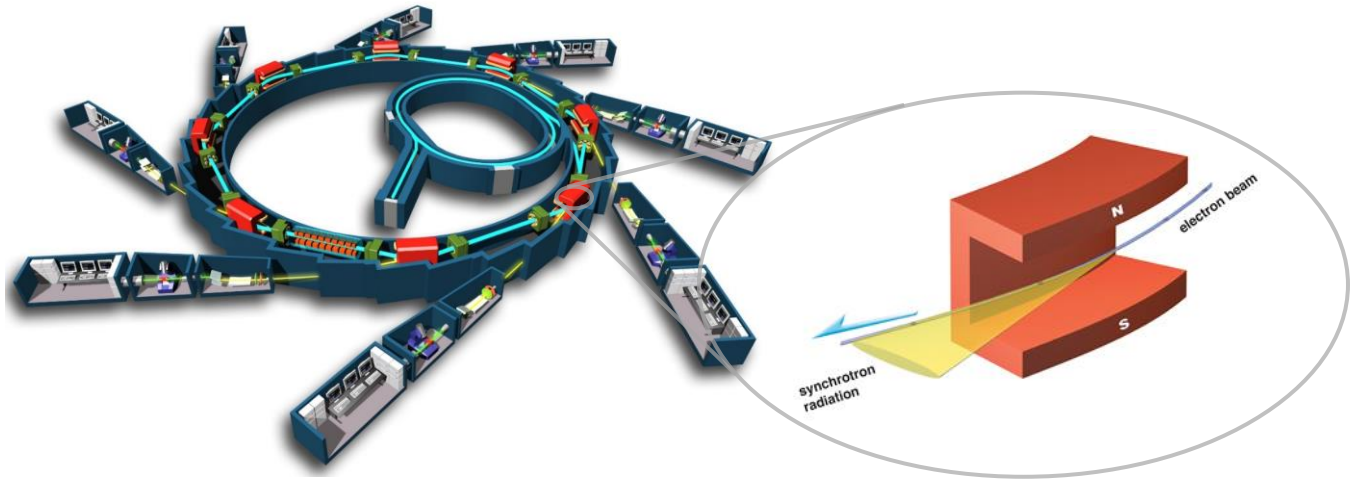


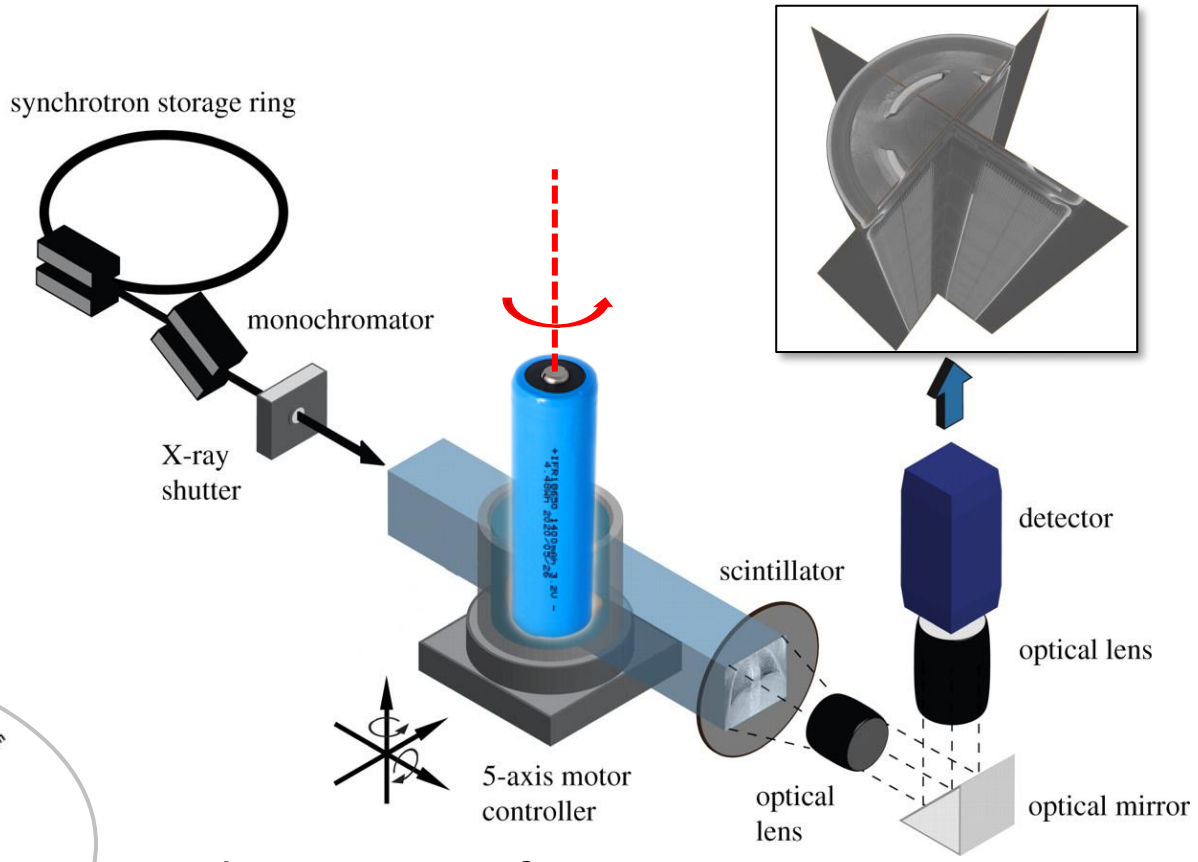
S. Solchenbach et al., Royal Soc. Chem., 19, 7294-7317 (2024)

Synchrotron computed tomography (SR-CT)

Advantages of Synchrotron X-rays:

- Very high x-ray beam intensity
- Tunable x-ray beam energy
- Highly collimated (parallel) beam geometry

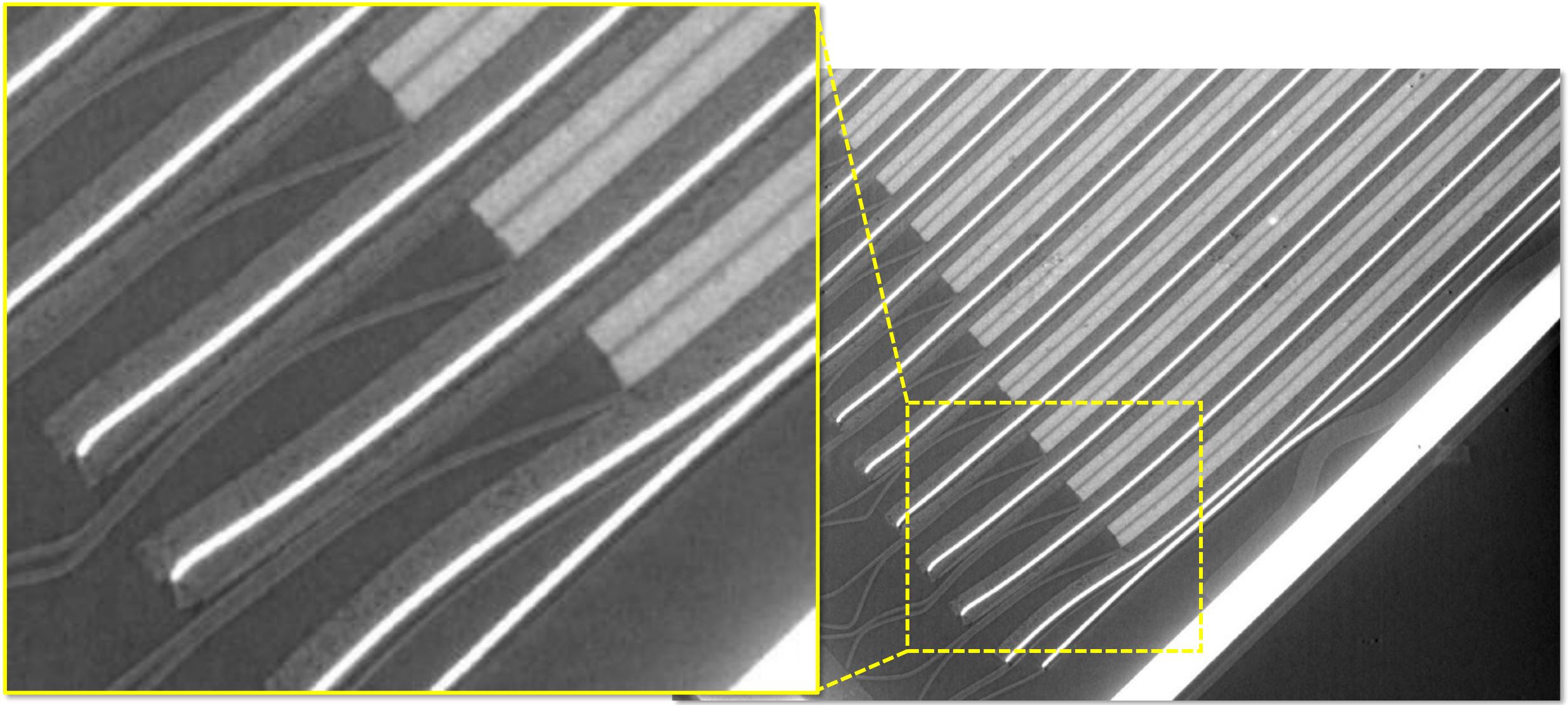




Advantages of SR-CT:

- Very fast scan times
- High-contrast for light elements
- Can scan parts of large objects at high magnification

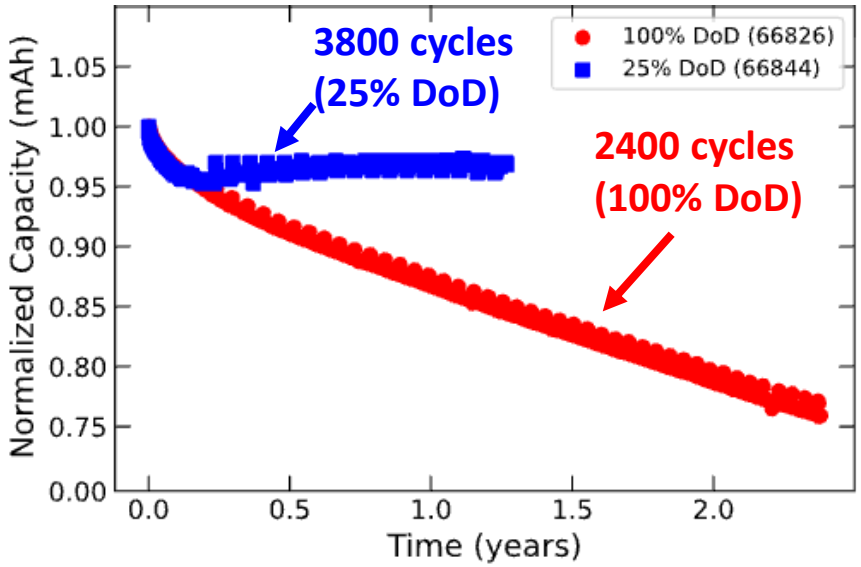
High-resolution imaging of cylindrical cells

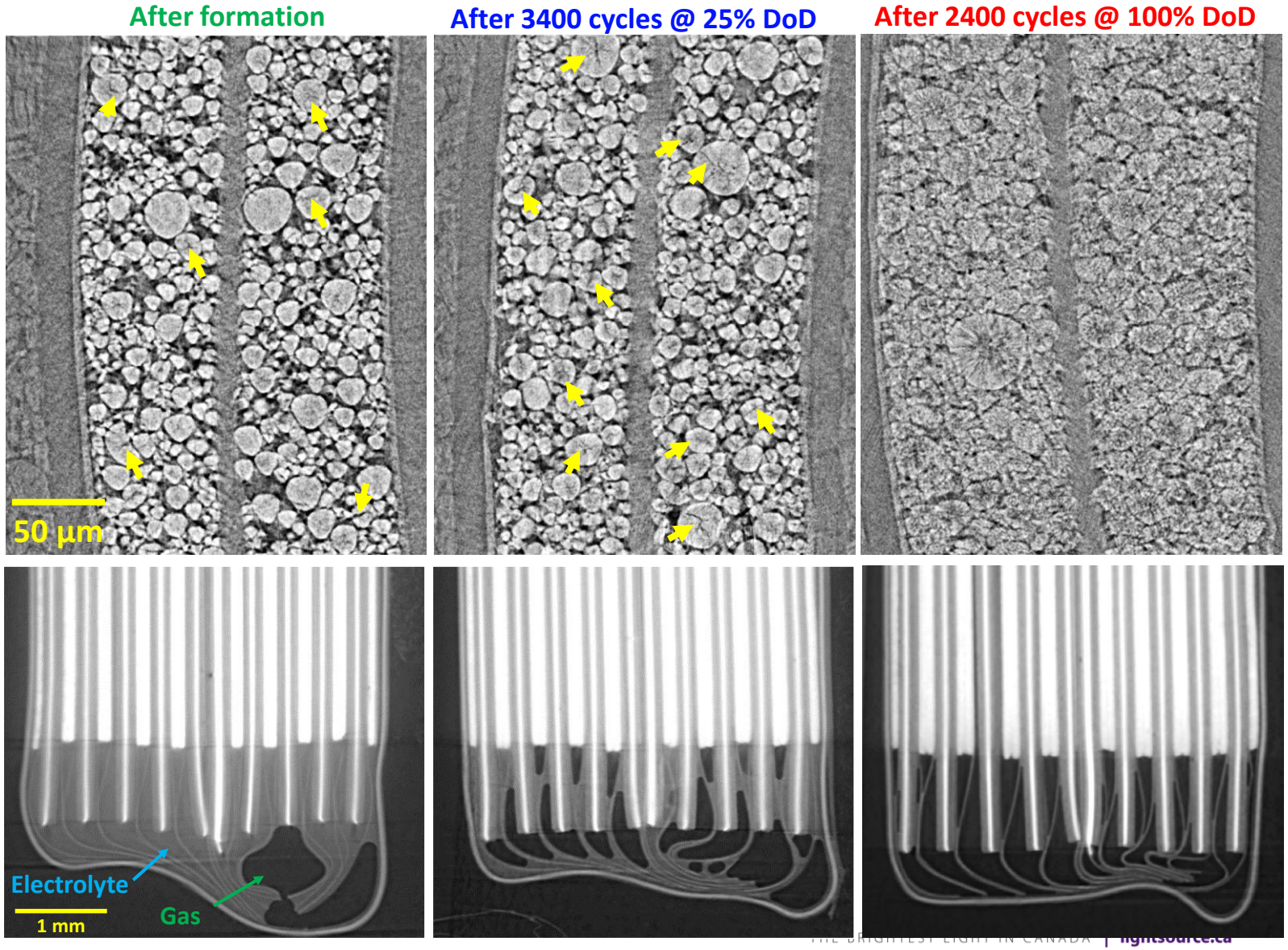


Electrode-level CT of 18650 cell (1.5 µm)

SR-CT can capture microstructural changes in large cells

- **Polycrystalline NMC622 / natural graphite cells with natural graphite** were cycled from 3.0 to 4.1 V at 40° C





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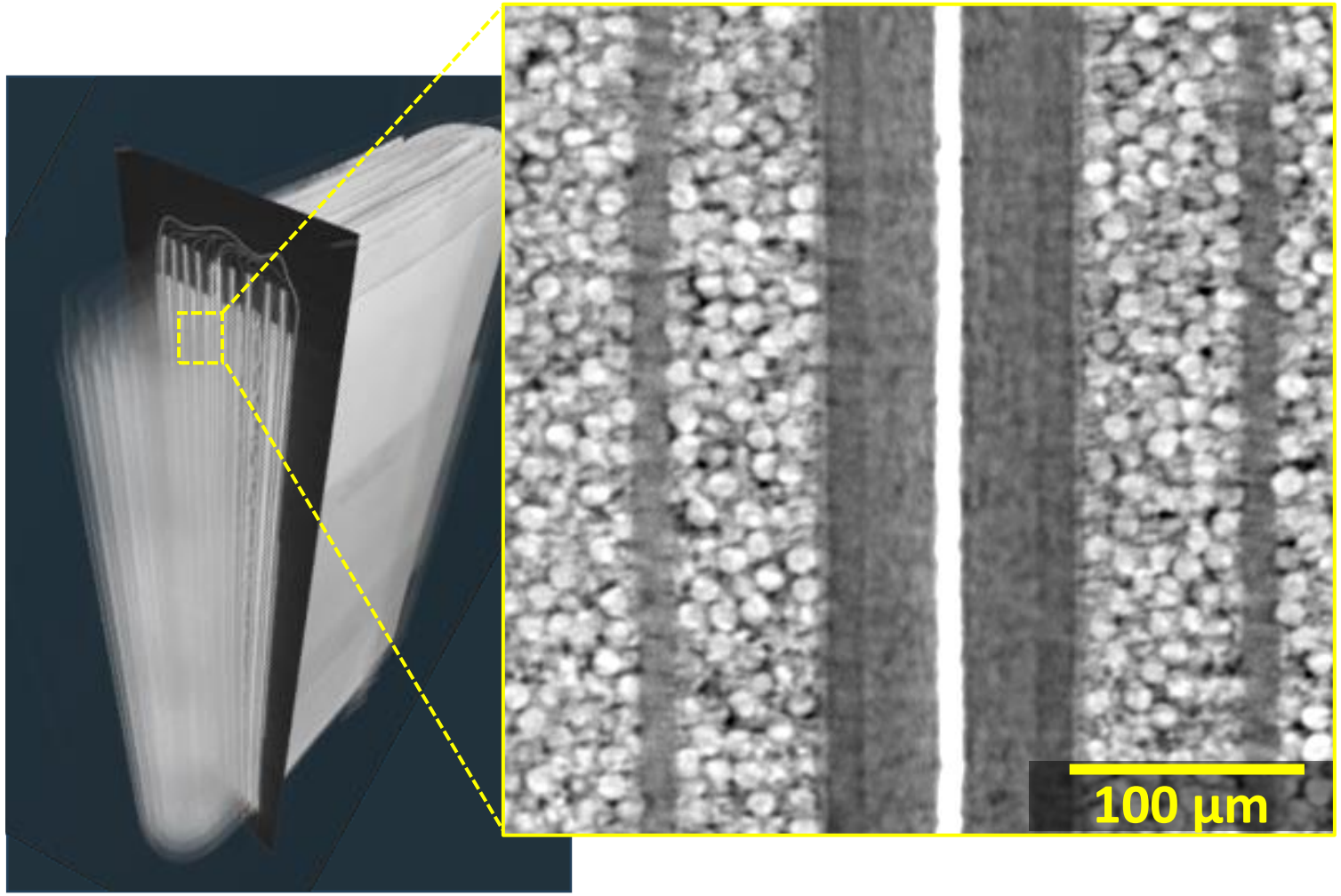
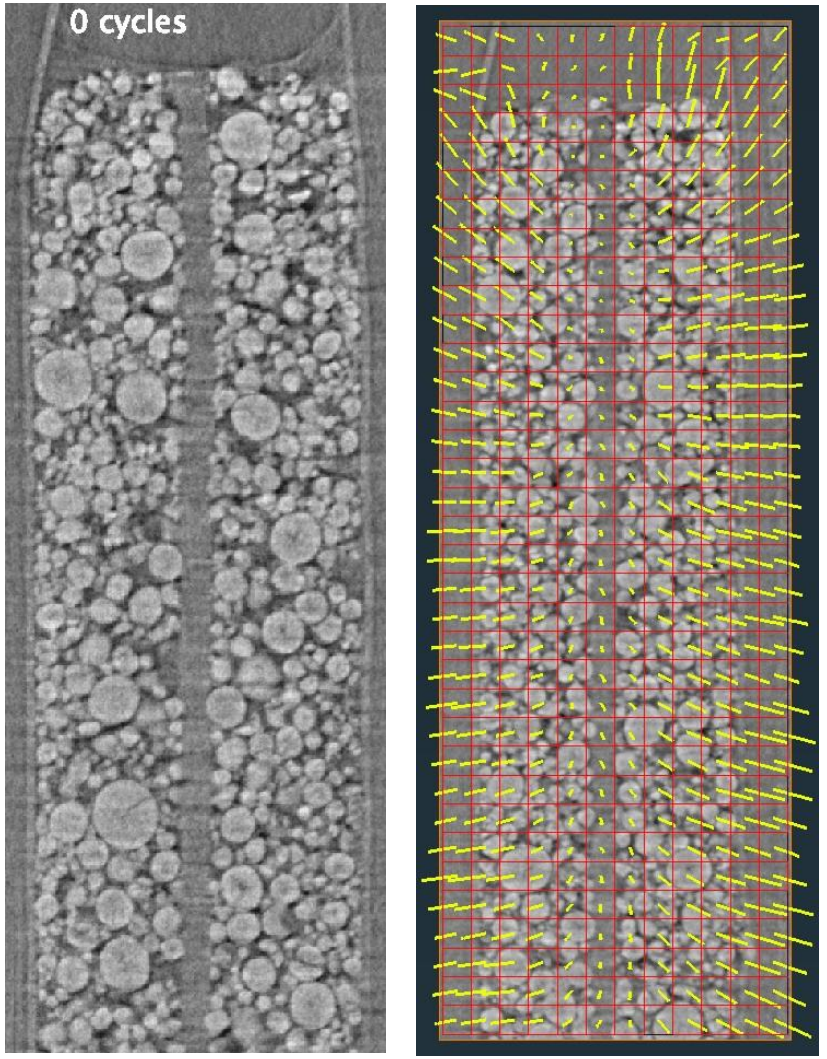
SR-CT can capture microstructural changes in large cells

Operando CT of Formation cycle (20% µSi/graphite pouch cell)



Time-lapse CT of microcracking over 1200 cycles



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Overview

Background

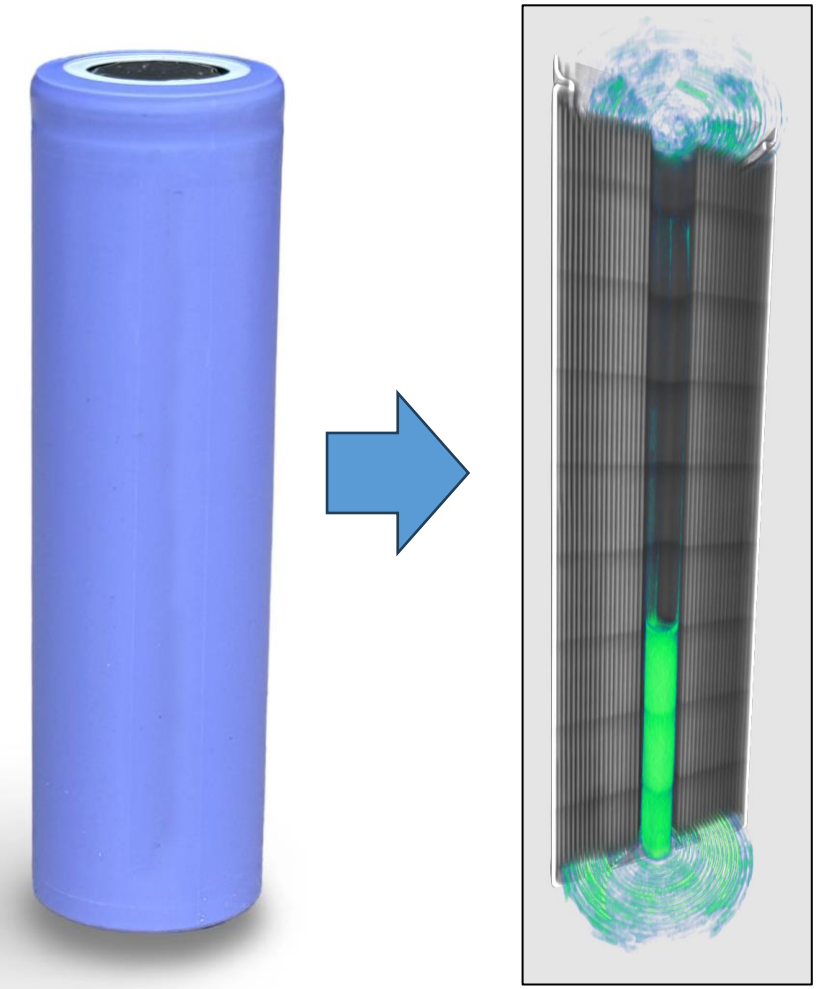
- Non-uniform degradation in large-format cells
- Electrolyte motion induced salt inhomogeneity (EMSI)
- Rotational inertia measurements (RIM)

Synchrotron Computed Tomography (SR-CT)

- What is it? why is it useful for battery research?
- Imaging microcracking in commercial pouch cells

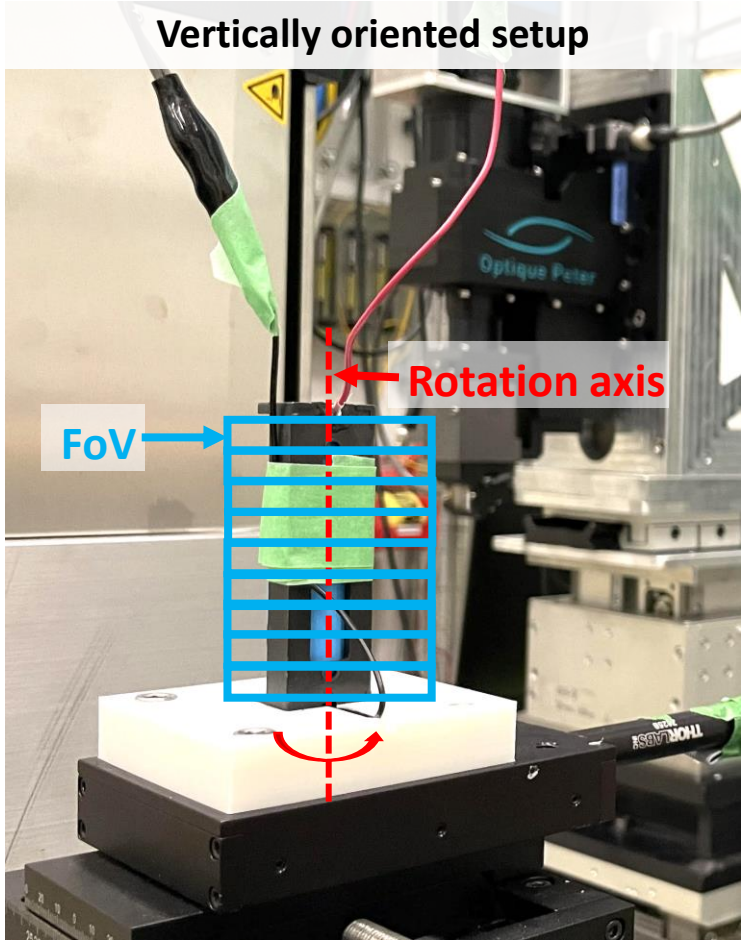
3D imaging of electrolyte motion in 18650 cells

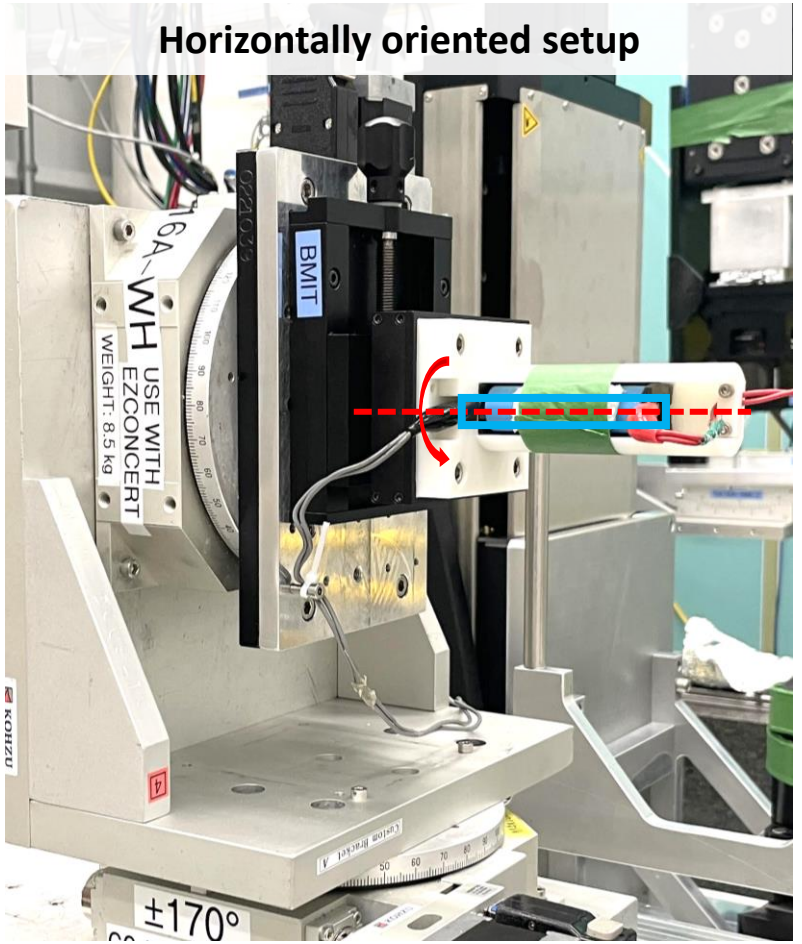
- Long-term cycling and salt redistribution
- Electrolyte reservoirs and recirculation
- Effect of gravity on vertical vs horizontal cells
- Using electrolyte to measure pore volume over time



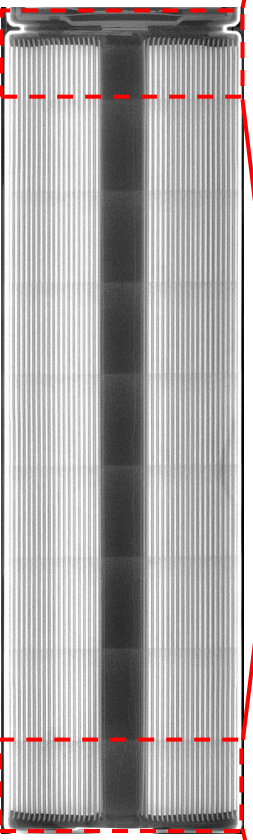
Imaging electrolyte motion in 18650 cells

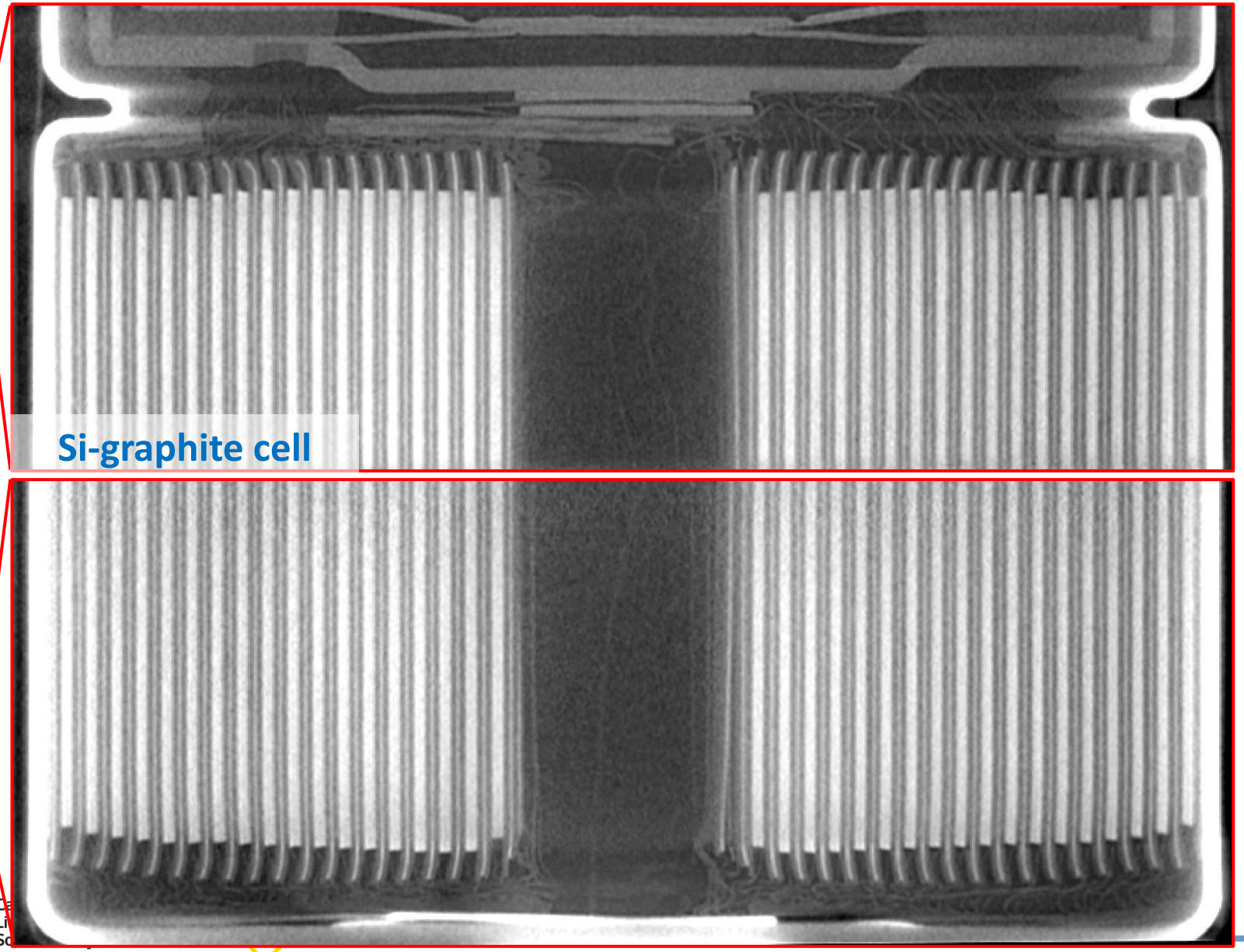
- Two cells were imaged:
  - **NCA / Si-graphite anode (5%)**
  - **NCA / graphite-only anode**
- cells were scanned in both vertical and horizontal orientation
- Cells were imaged at
  - 11 µm pixel size for vertical
  - 22 µm pixel size for horizontal
- Vertical scan time: 6 min
- Horizontal scan time: 50 sec
- Cells were discharged to 2.5 V before imaging
- During imaging, cells were charged at C/3 to 4.2 V and discharged at 1C to 2.5 V

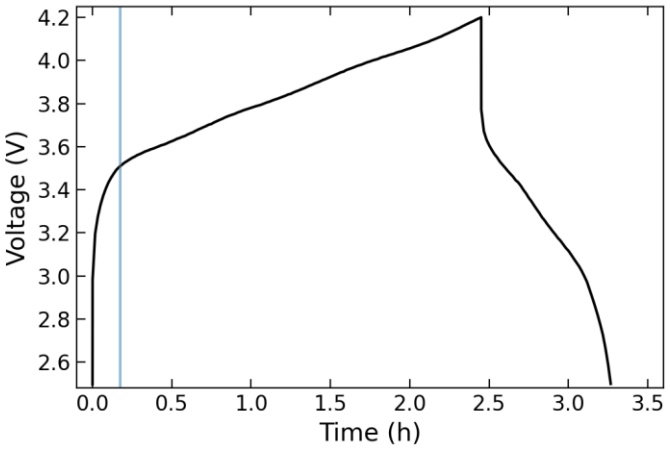




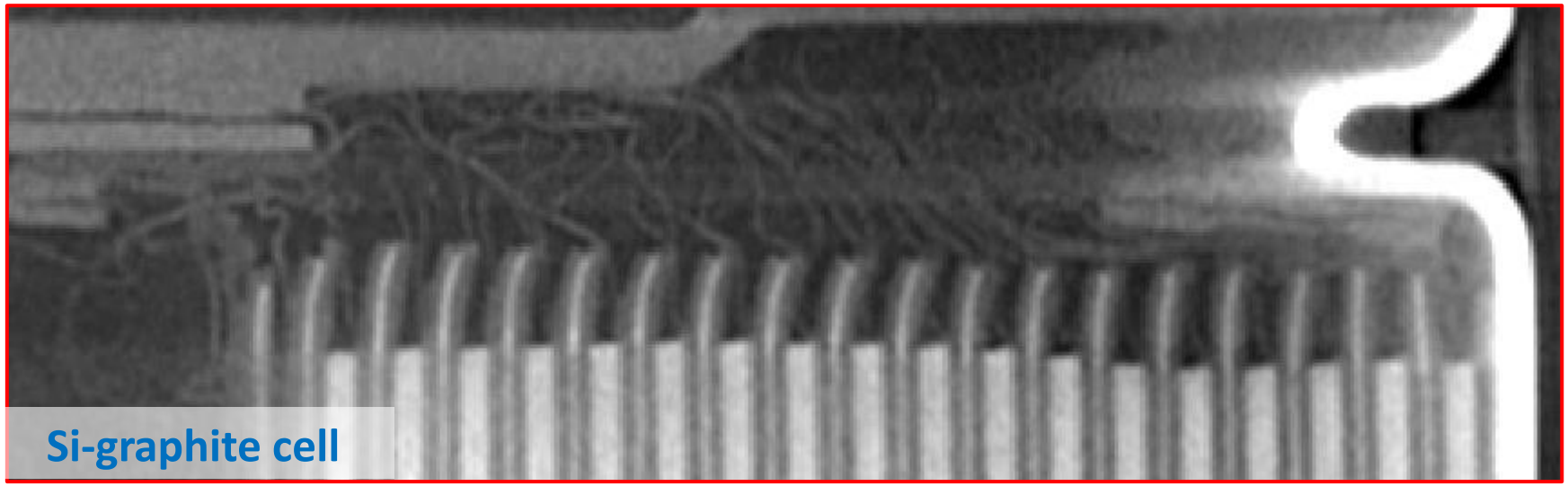
Vertically oriented cells

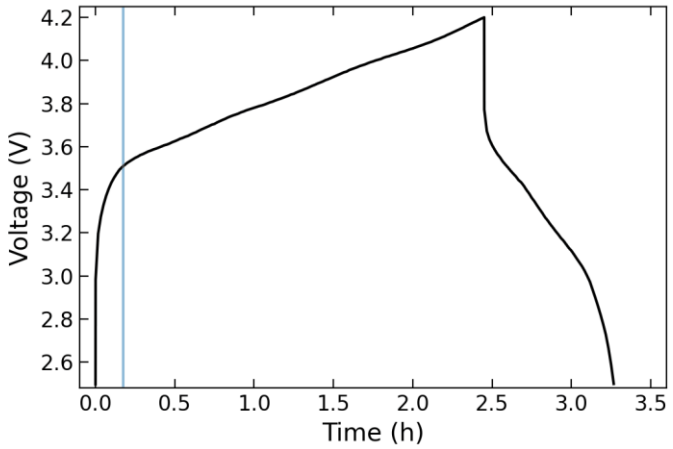


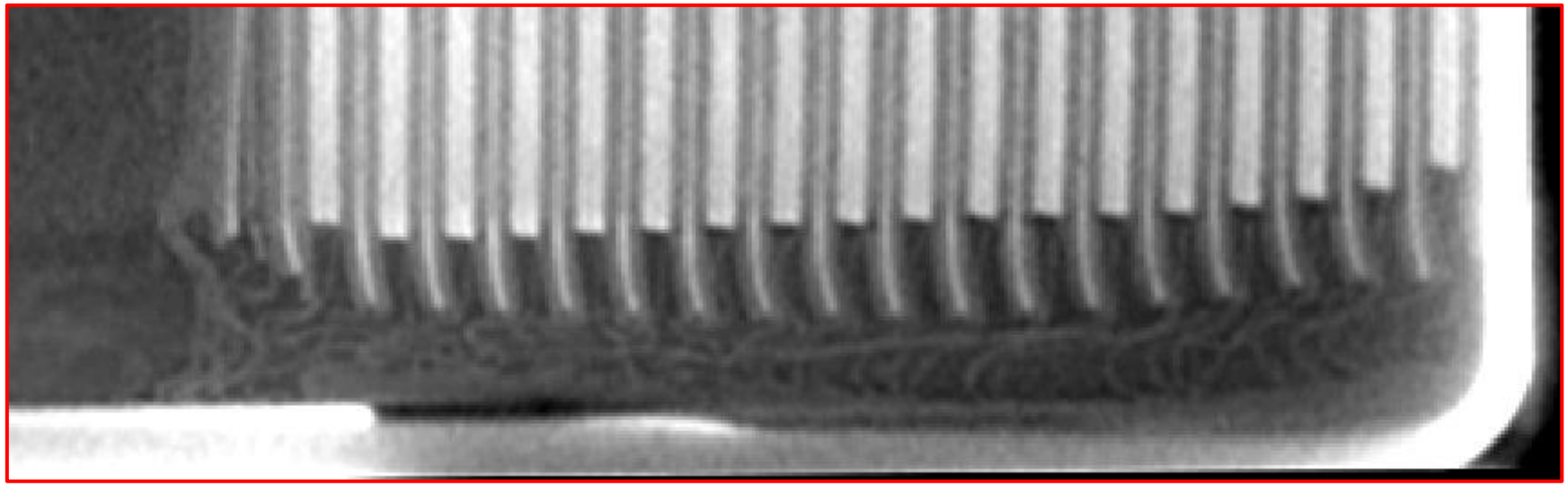




In-plane flow of electrolyte







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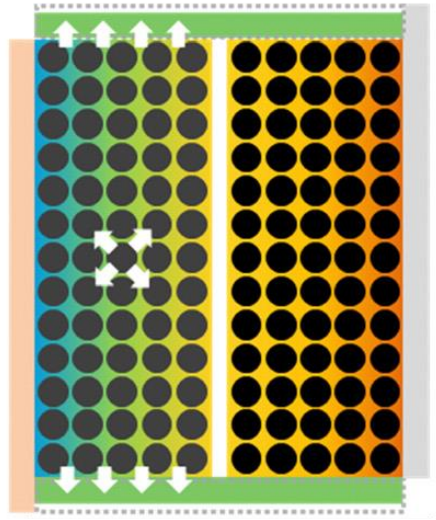
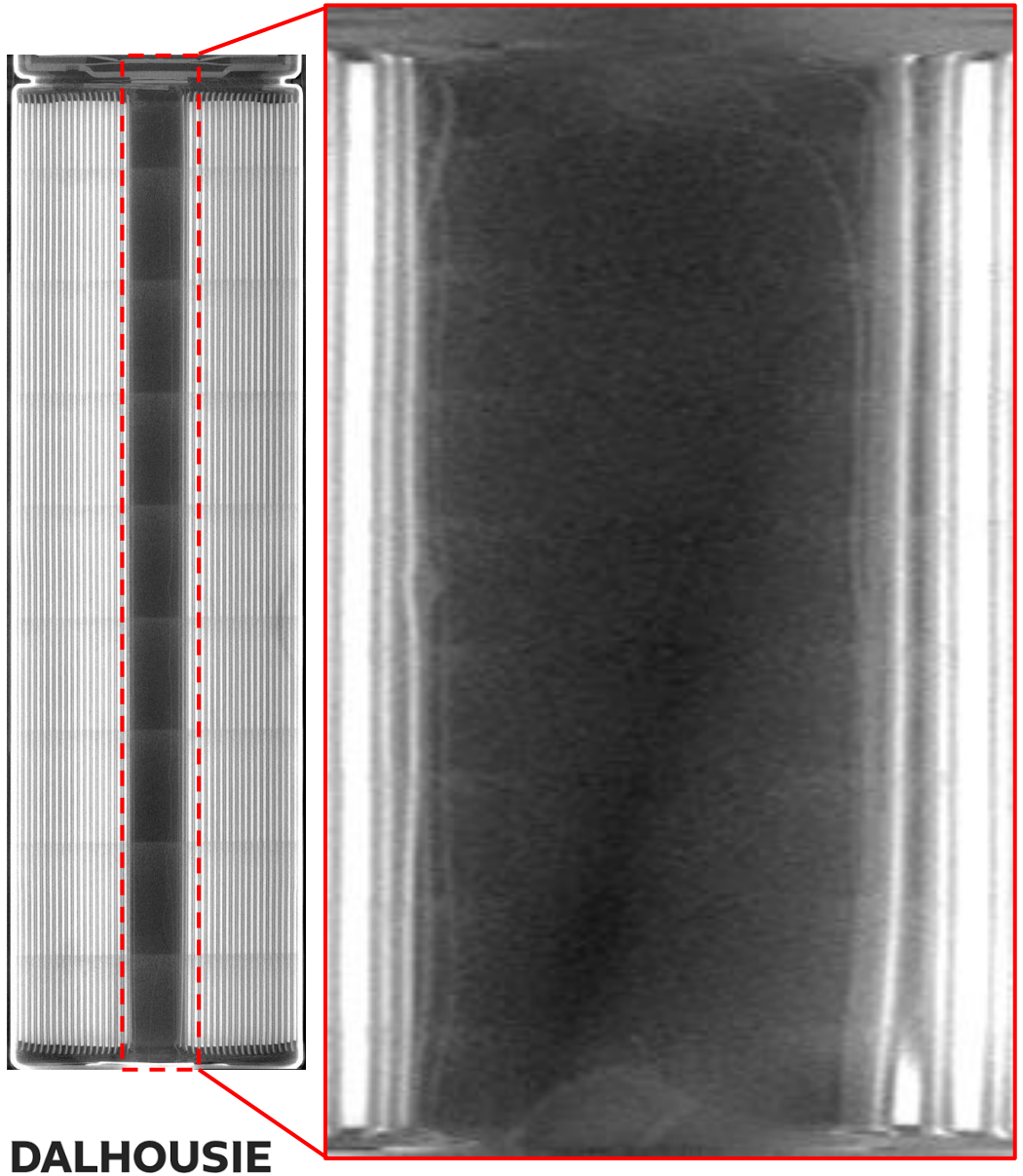
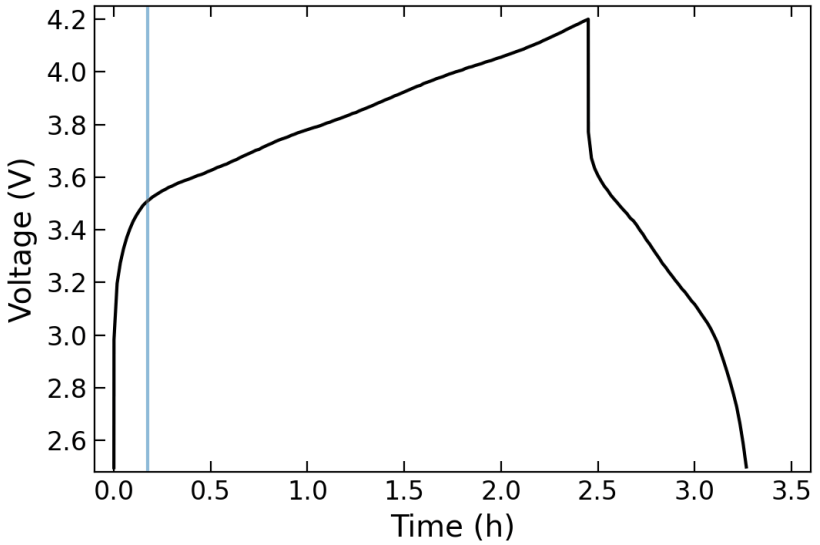


Imaging electrolyte motion in 18650 cells

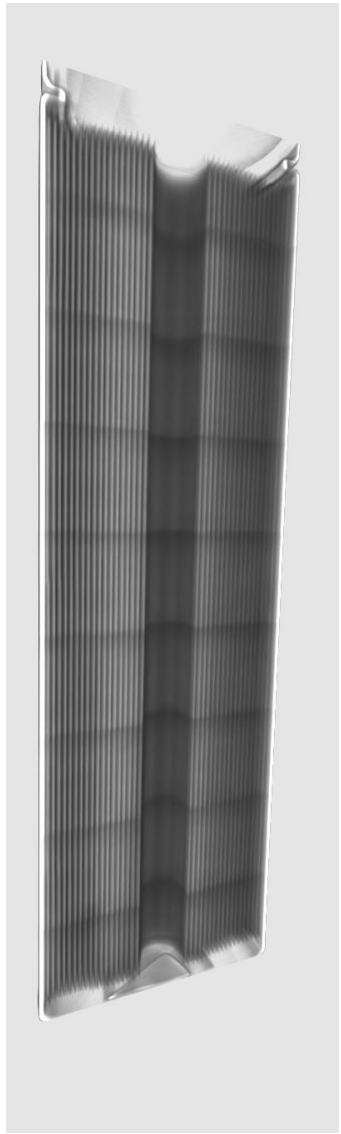
- Excess electrolyte drains down the core along channels formed by separator
- You can see this if you compress the core vertically (pixels binned by a factor of 8)



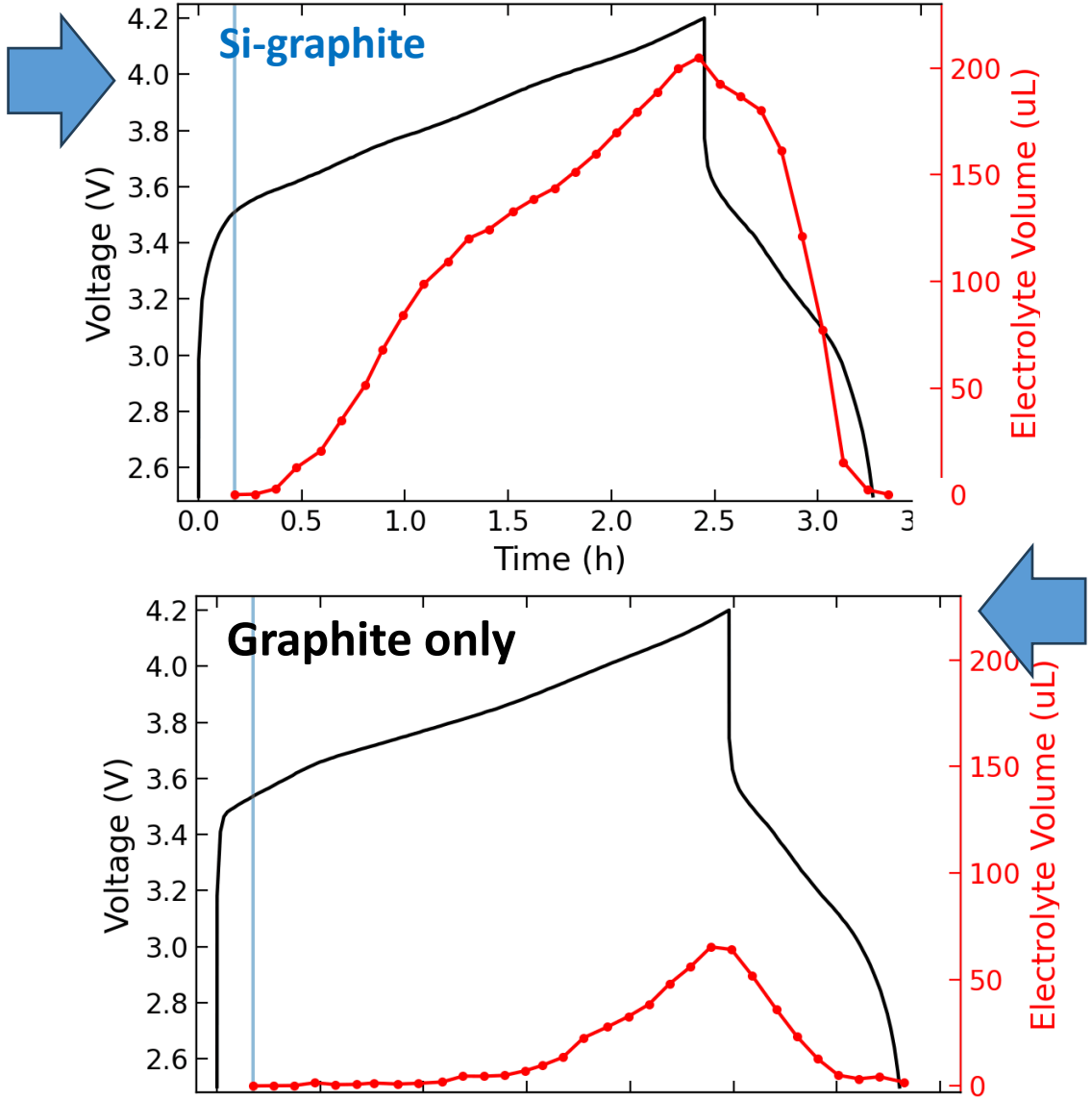


Vertically oriented cells

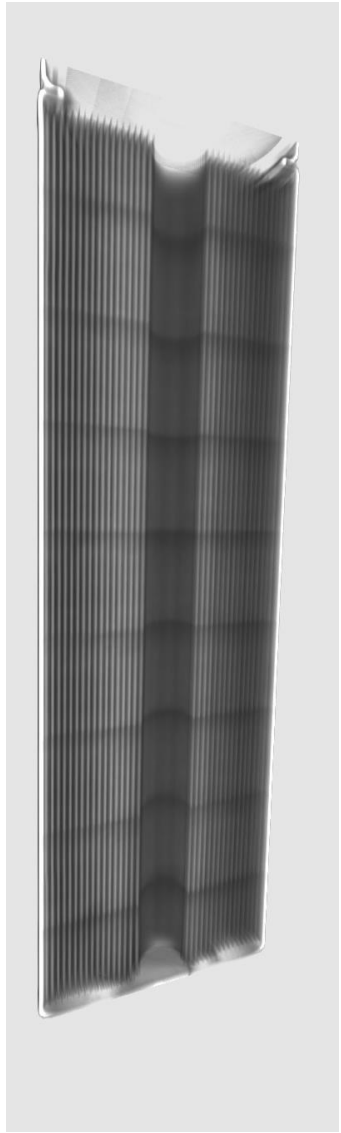
Si-graphite



Gravity



Graphite-only



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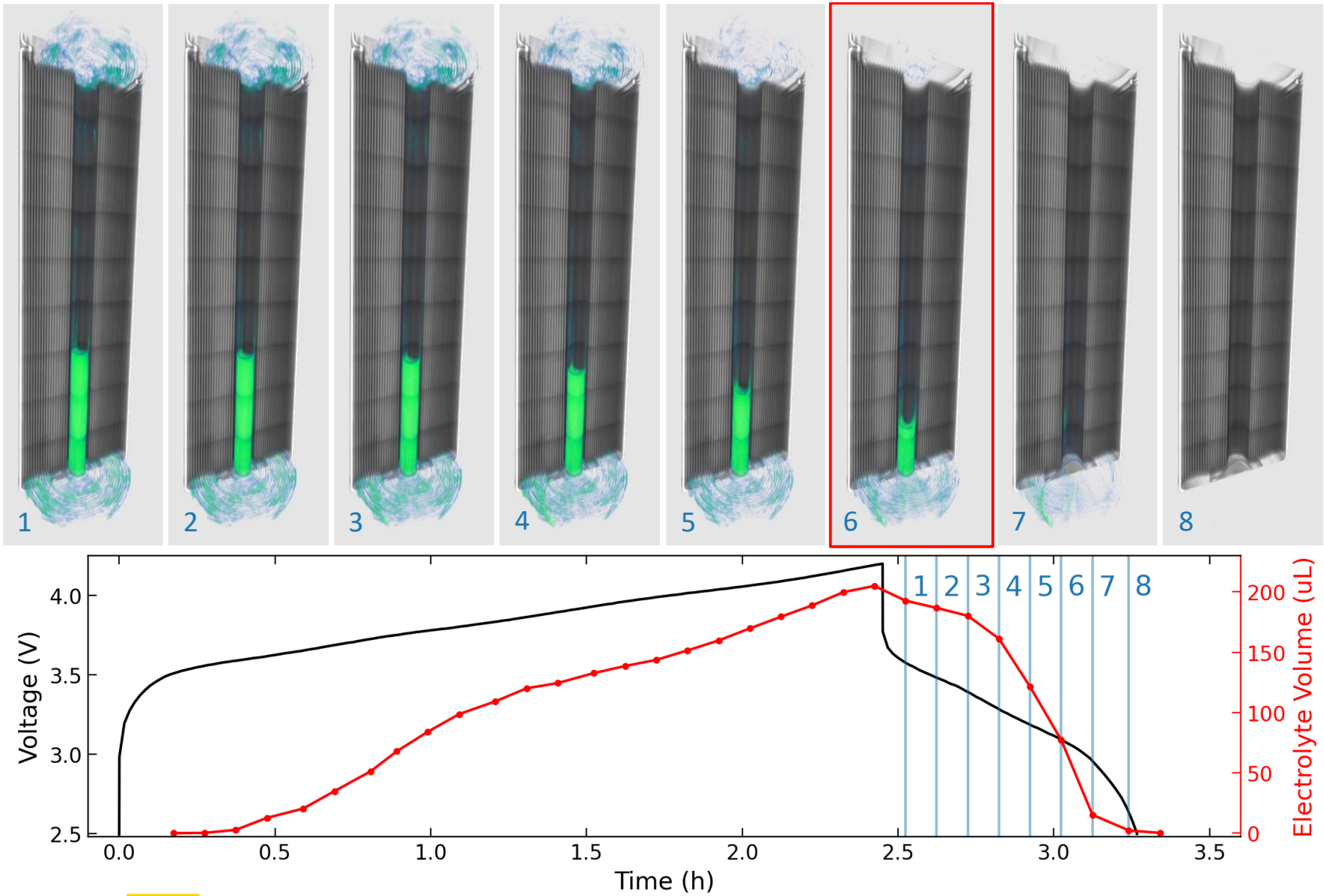
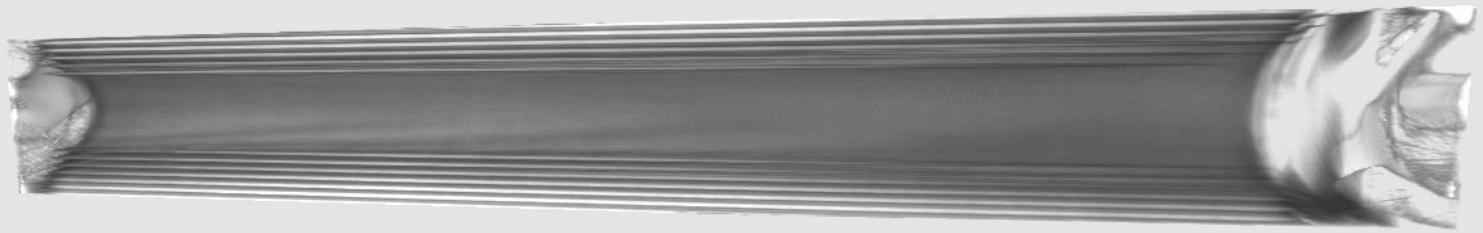
Excess electrolyte depletes at top before bottom

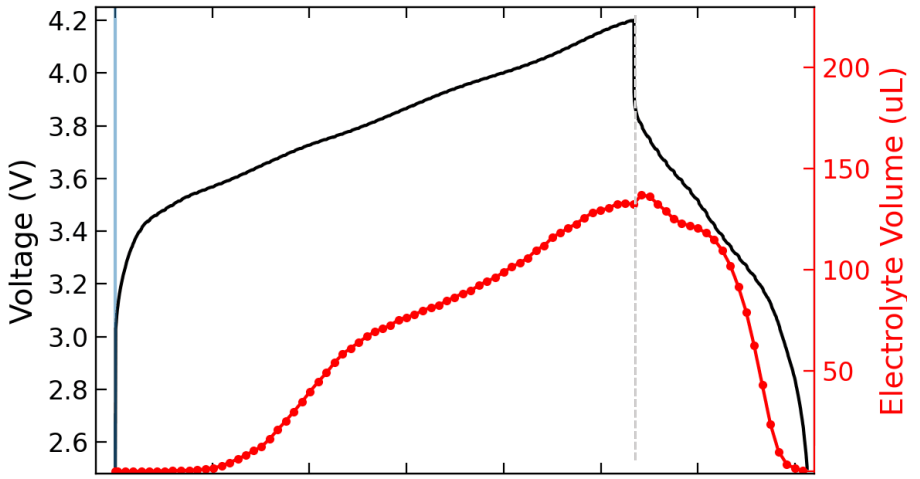


Horizontally oriented cells

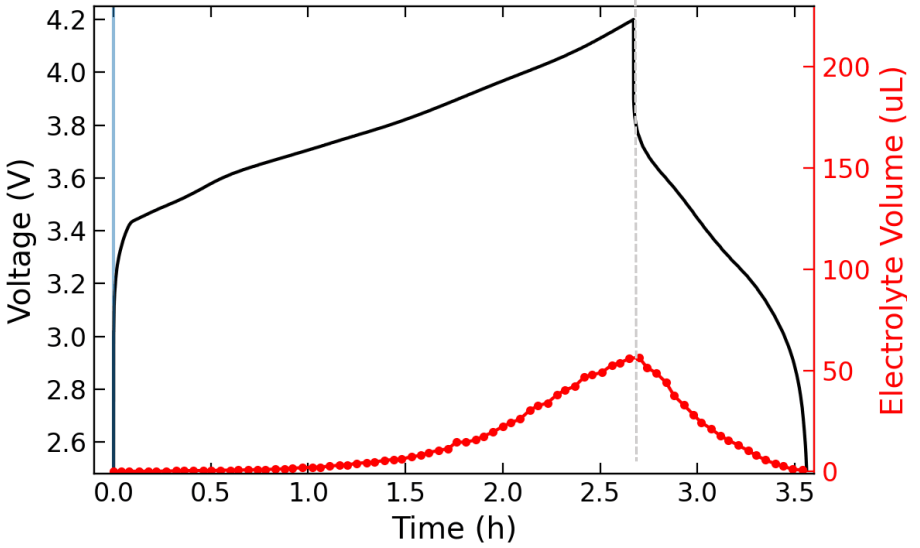
Si-graphite cell



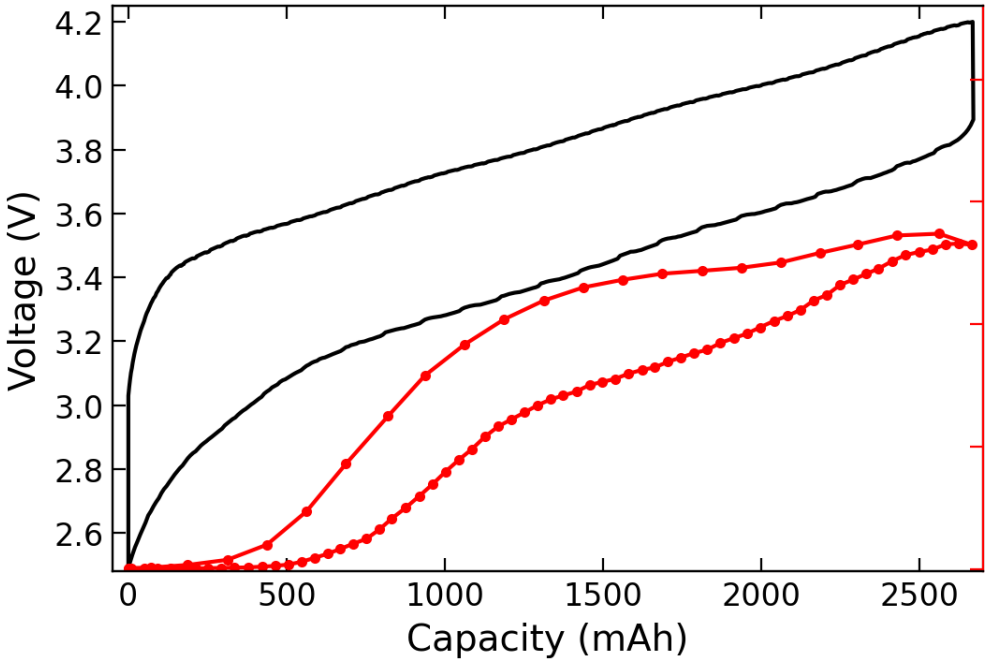
Gravity

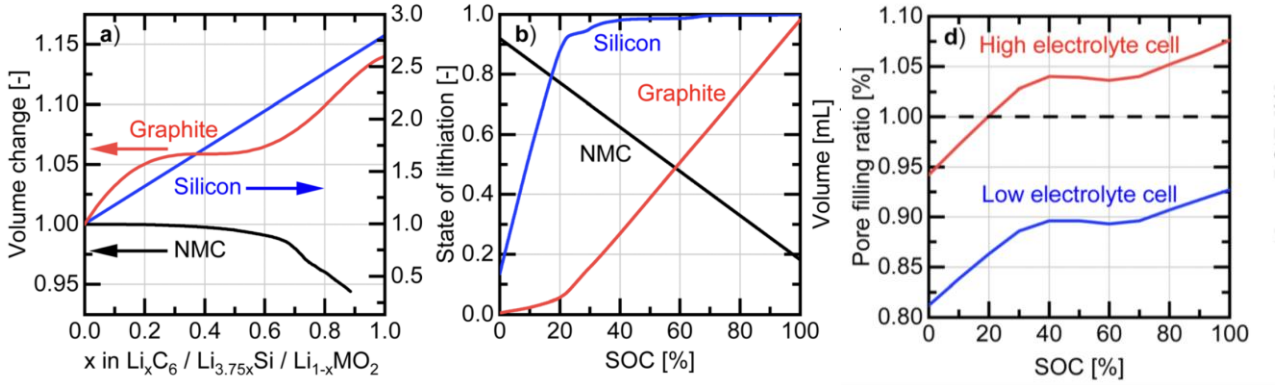


Graphite-only cell



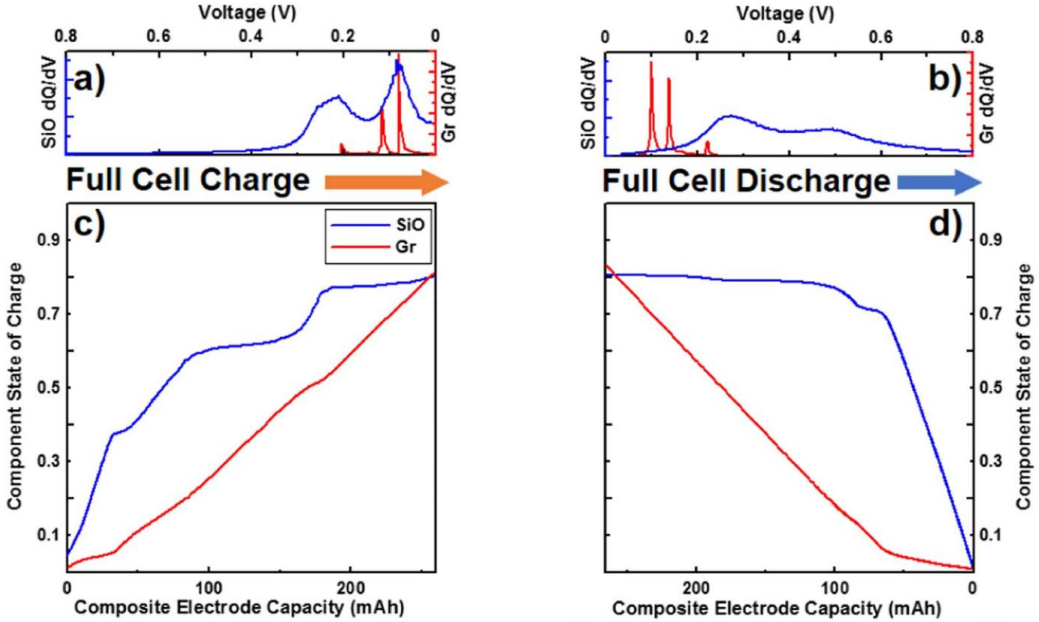
Shape of excess electrolyte curve





S. Solchenbach et al., Royal Soc. Chem., 19, 7294-7317 (2024)

Inherent hysteresis in Si lithiation state



A.J. Louli et al., J. Electrochem. Soc., 164 (12) A2689-A2696 (2017)

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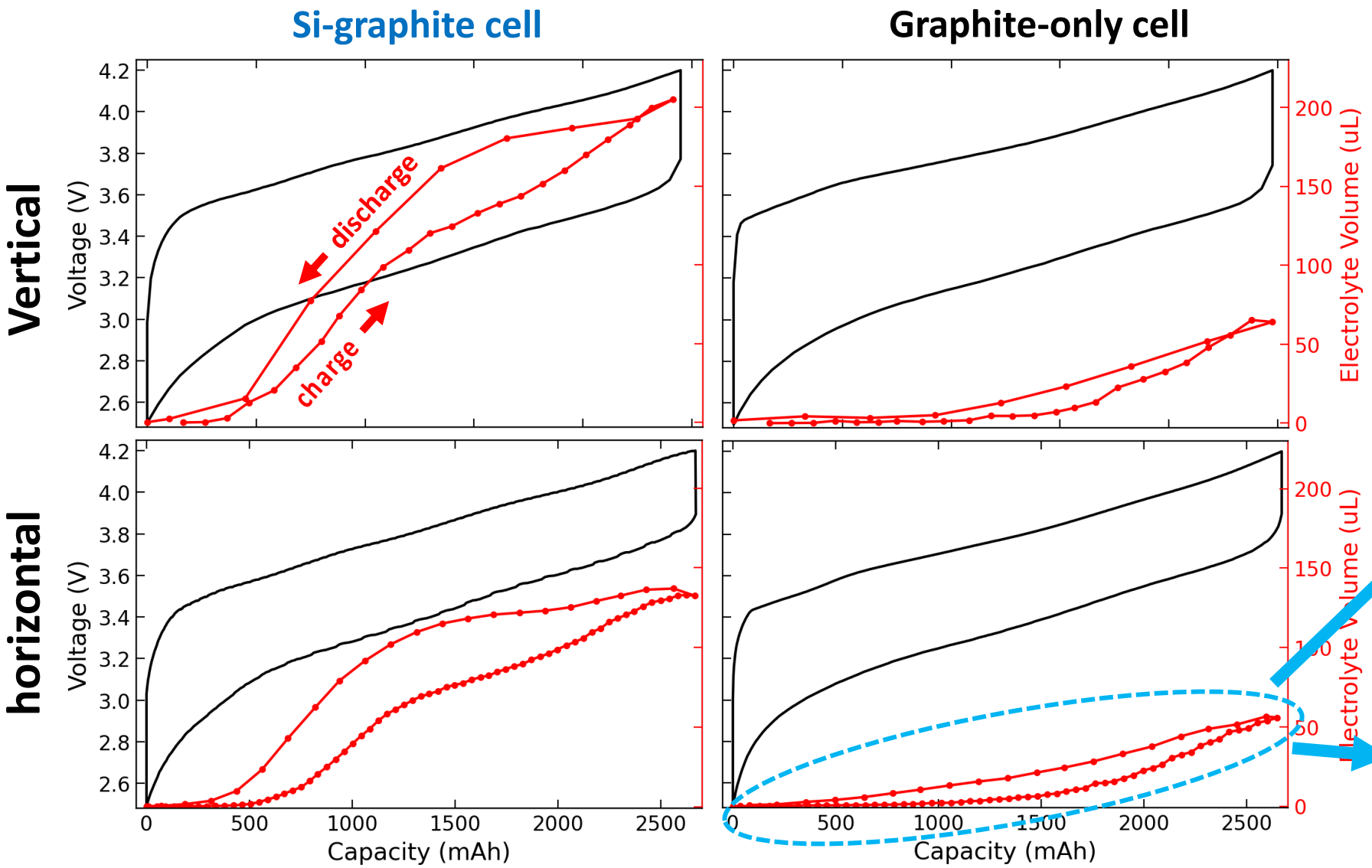
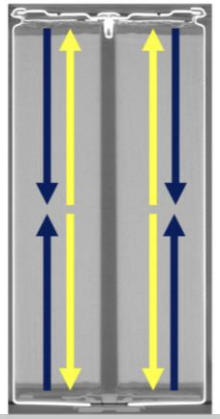
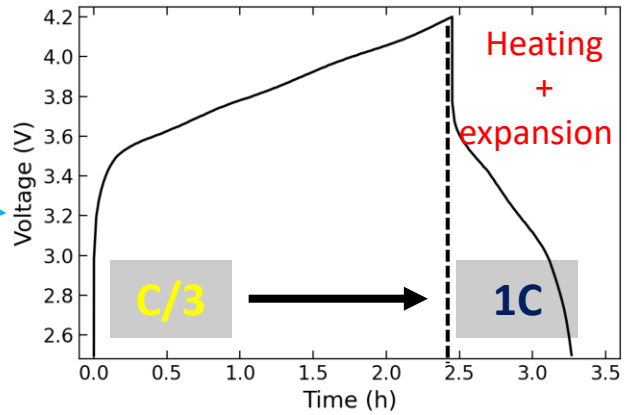
Comparison of electrolyte volume curves



High pressure differential (charge)



Low pressure differential (discharge)



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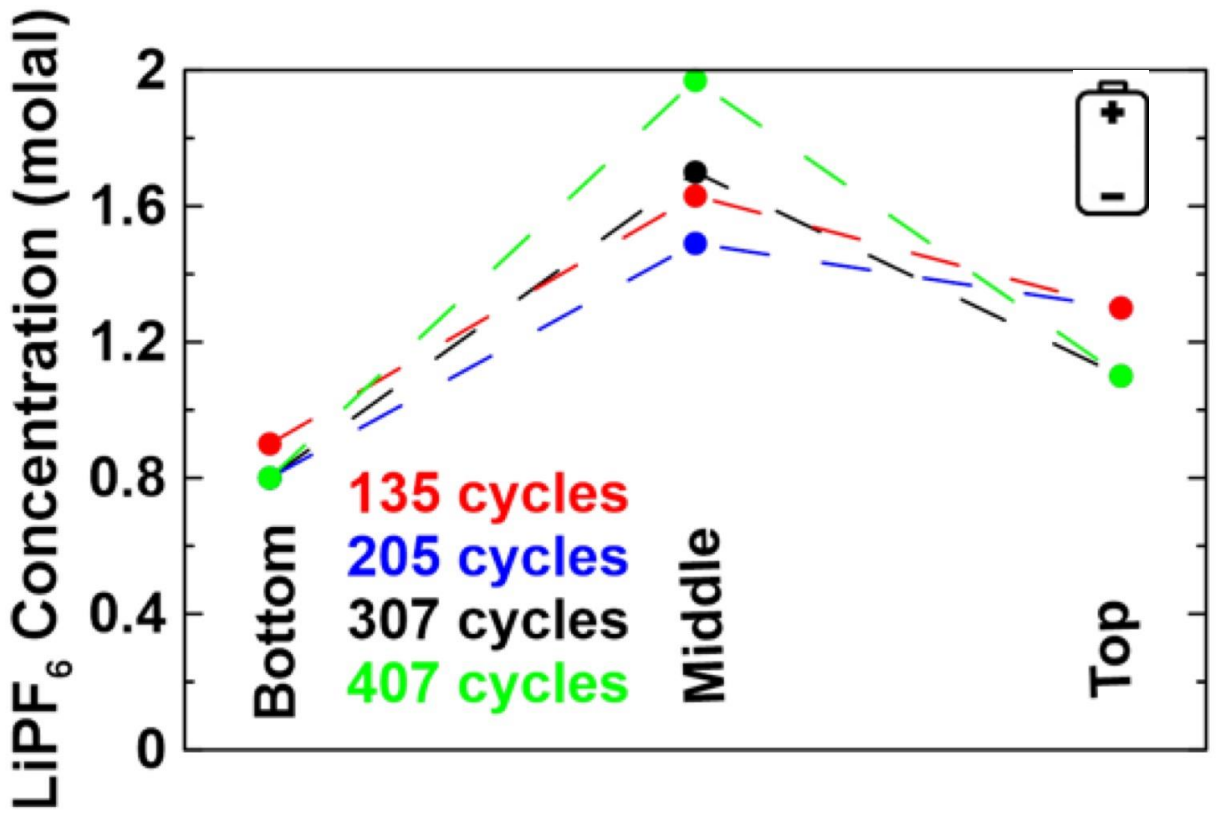
Salt concentration measurements after long-term cycling

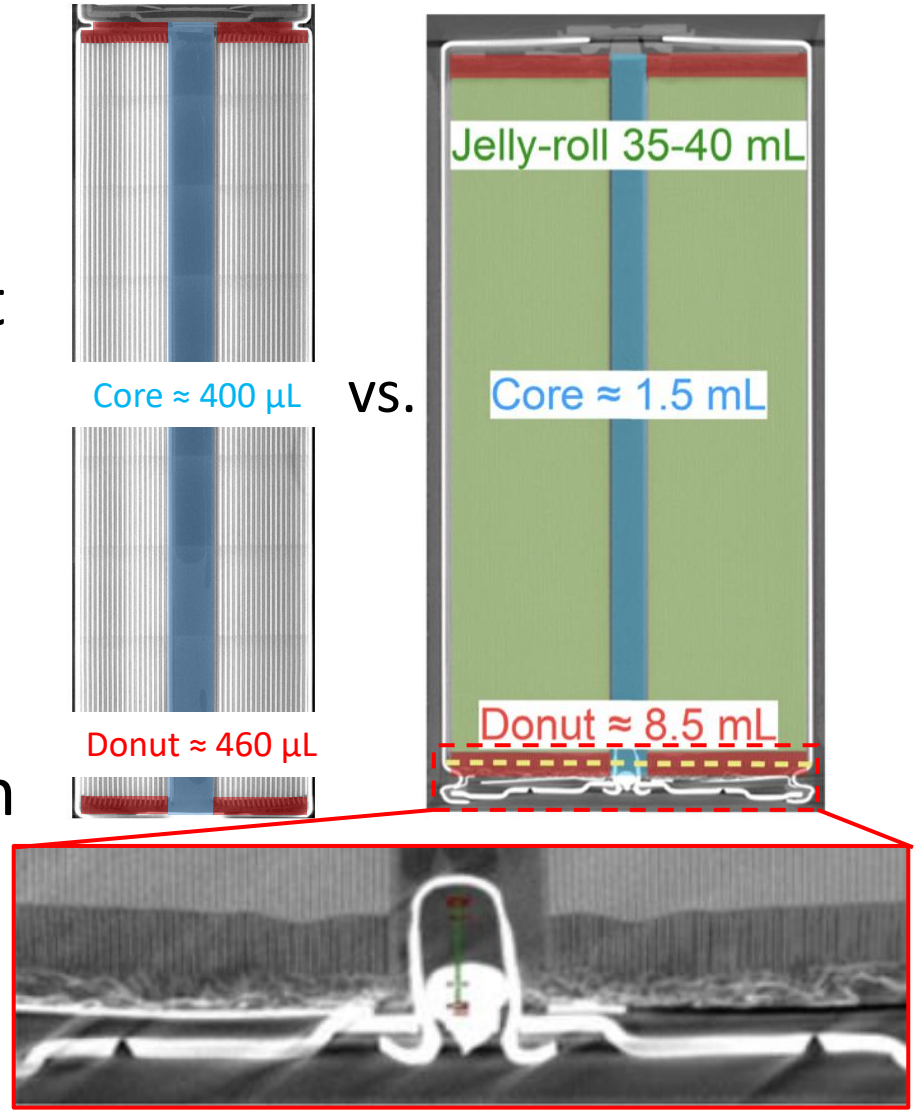
- four Si-containing cells were cycled offline
- Cycling conditions:
  - Vertical orientation (cap up)
  - Charged at C/4 to 4.2 V
  - Discharged at C/3 to 2.85 V
  - Temperature = 20° C
- After cycling different amounts, cells were discharged to 0 V, sawed into 3 segments for solvent extraction and ICP-MS



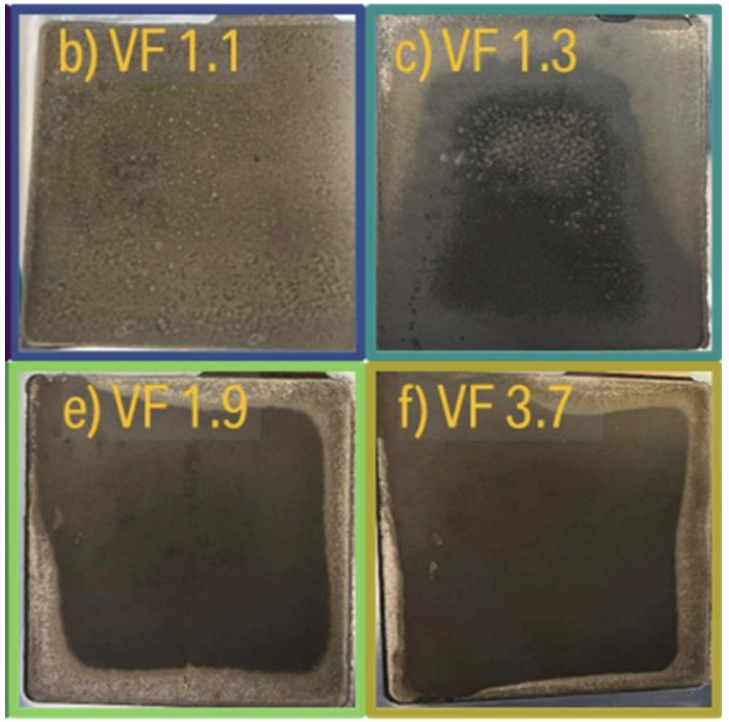
EMSI can occur at modest C-rates

A note on cell geometry

- Solchenbach et al. used 4695 cells with different features:
  - Core volume is small relative to “donuts”
  - Tabless design
- EMSI also happens in prismatic cells, and even single-layer pouch cells



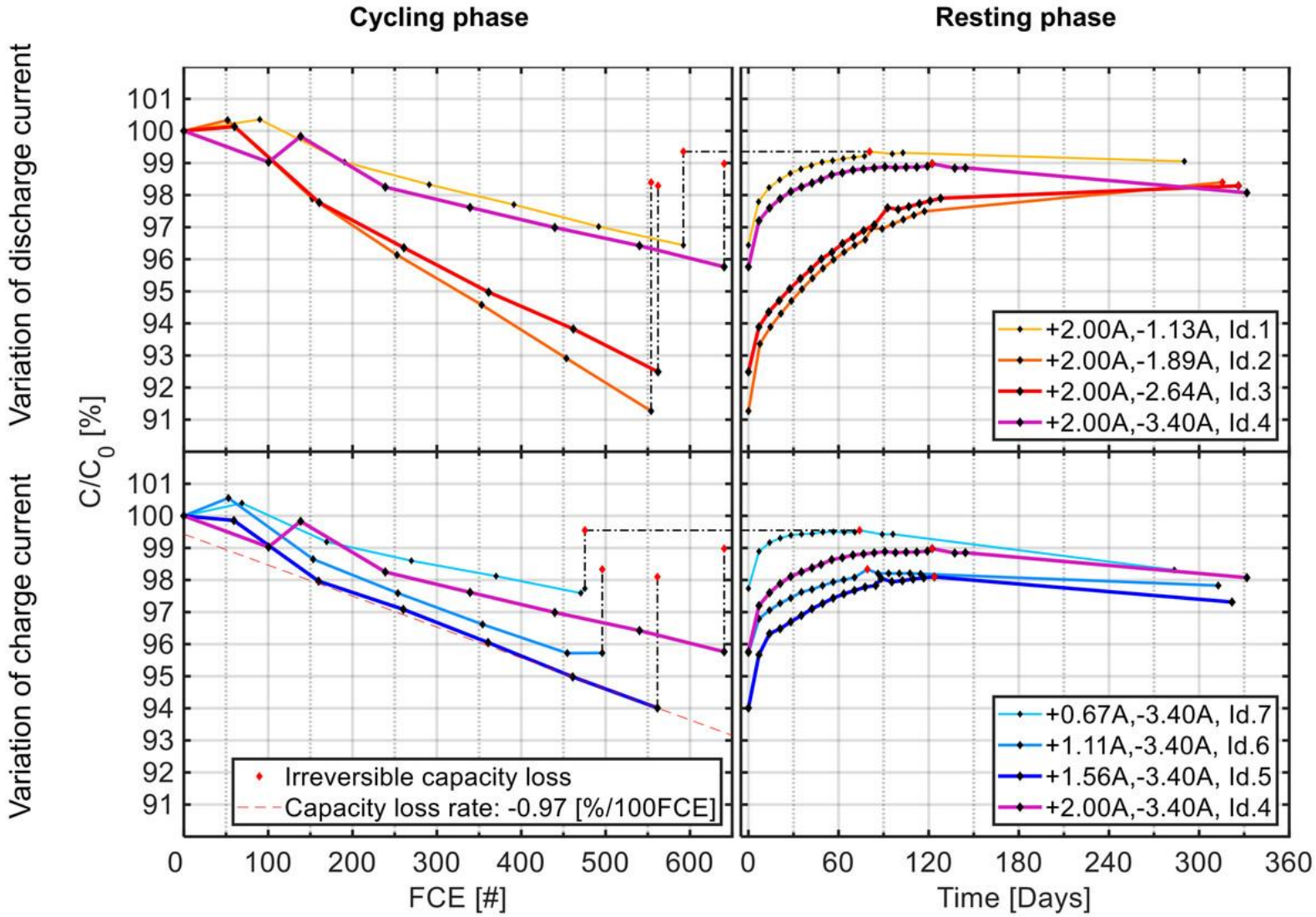
S. Solchenbach et al., Royal Soc. Chem., 19, 7294-7317 (2024)



H. C. Hsiao, J. of The Electrochem. Soc., 173 010503 (2026)

Recovery after repeated fast charging

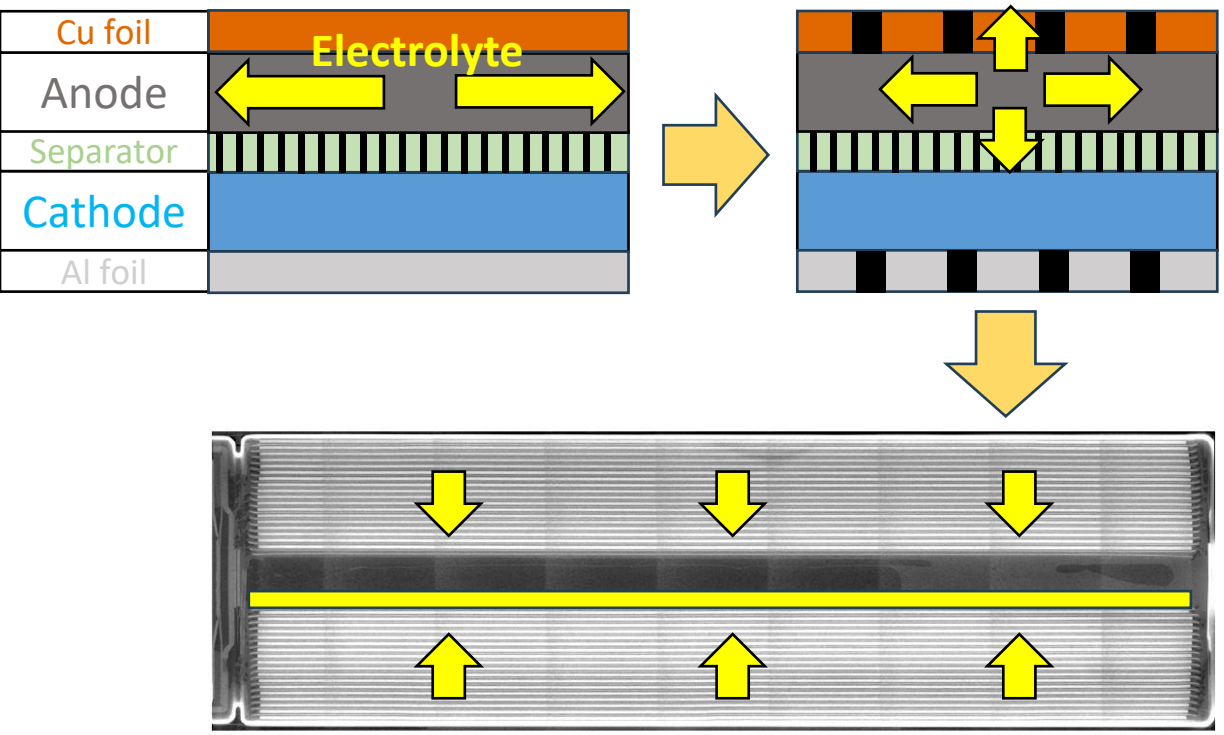
- The capacity fade and resistance increase due to EMSI is reversible to an extent
- Rate and extent of recovery vary widely with different cells and cycling conditions



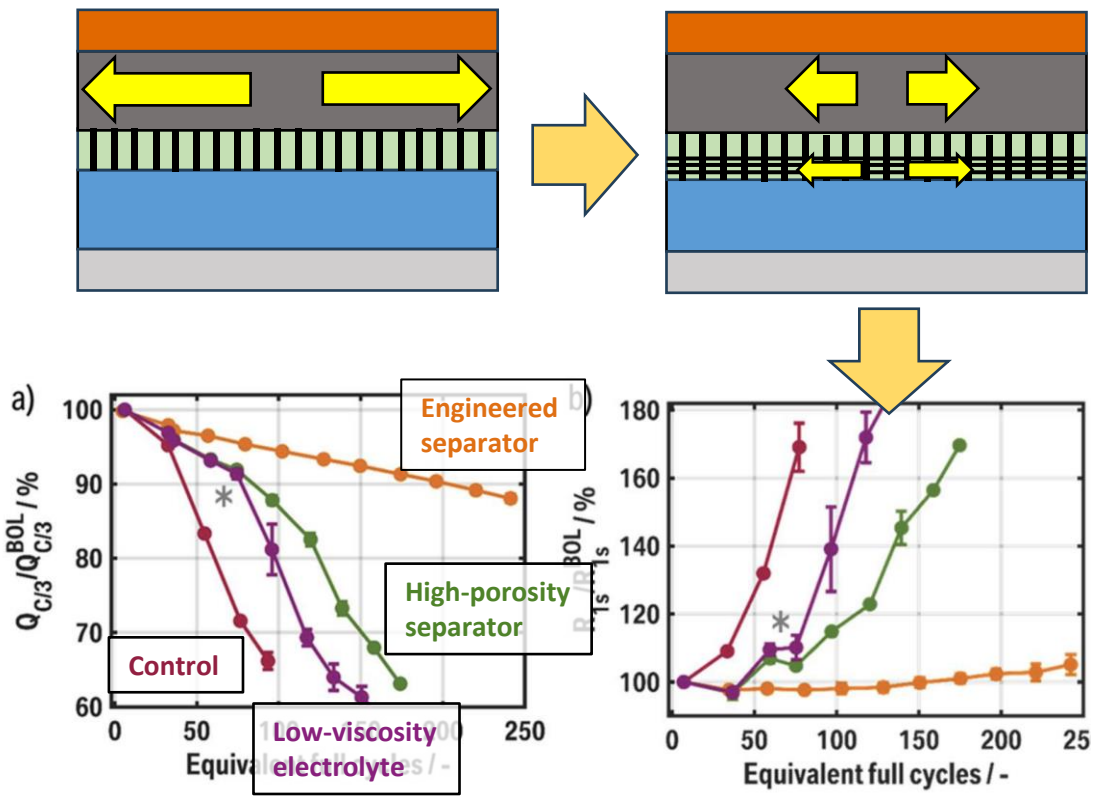
P. Torricos et al., Batteries & Supercaps, 00, e202500559 (2025)

Mitigating EMSI by redirecting electrolyte flow

Perforated current collectors



“Engineered” separators



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Summary

- SR-CT is an excellent tool for time-resolved electrolyte imaging
- Electrolyte flow is affected by:
  - Cell geometry (even small details)
  - Gravity and cell orientation
  - Active materials
  - Separator and foil engineering
- EMSI can occur at modest C-rates
- This work validates key assumptions of EMSI

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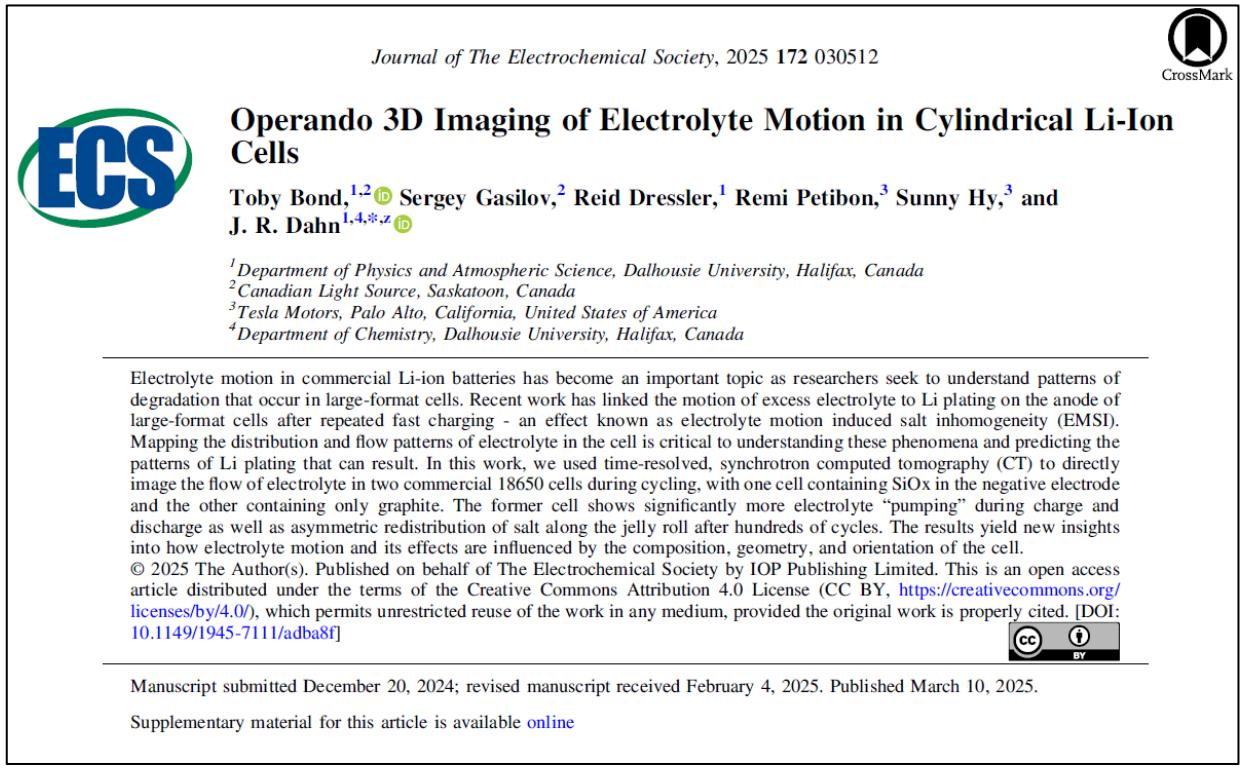

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Operando 3D Imaging of Electrolyte Motion in Cylindrical Li-Ion Cells

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[3]*Tesla Motors, Palo Alto, California, United States of America*
[4]*Department of Chemistry, Dalhousie University, Halifax, Canada*

Electrolyte motion in commercial Li-ion batteries has become an important topic as researchers seek to understand patterns of degradation that occur in large-format cells. Recent work has linked the motion of excess electrolyte to Li plating on the anode of large-format cells after repeated fast charging - an effect known as electrolyte motion induced salt inhomogeneity (EMSI). Mapping the distribution and flow patterns of electrolyte in the cell is critical to understanding these phenomena and predicting the patterns of Li plating that can result. In this work, we used time-resolved, synchrotron computed tomography (CT) to directly image the flow of electrolyte in two commercial 18650 cells during cycling, with one cell containing SiOx in the negative electrode and the other containing only graphite. The former cell shows significantly more electrolyte "pumping" during charge and discharge as well as asymmetric redistribution of salt along the jelly roll after hundreds of cycles. The results yield new insights into how electrolyte motion and its effects are influenced by the composition, geometry, and orientation of the cell.

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