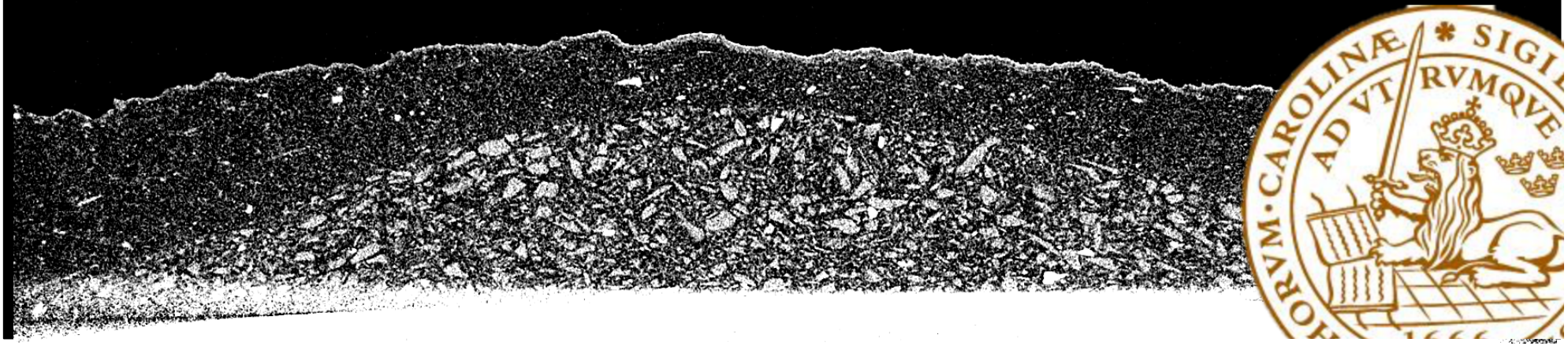
4D IMAGING LAB

LUND INSTITUTE OF ADVANCED NEUTRON AND X-RAY SCIENCE LINXS

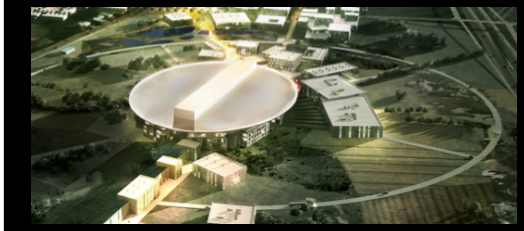
4D IMAGING OF CHARGING AND DISCHARGING PROCESSES IN LITHIUM-ION BATTERIES

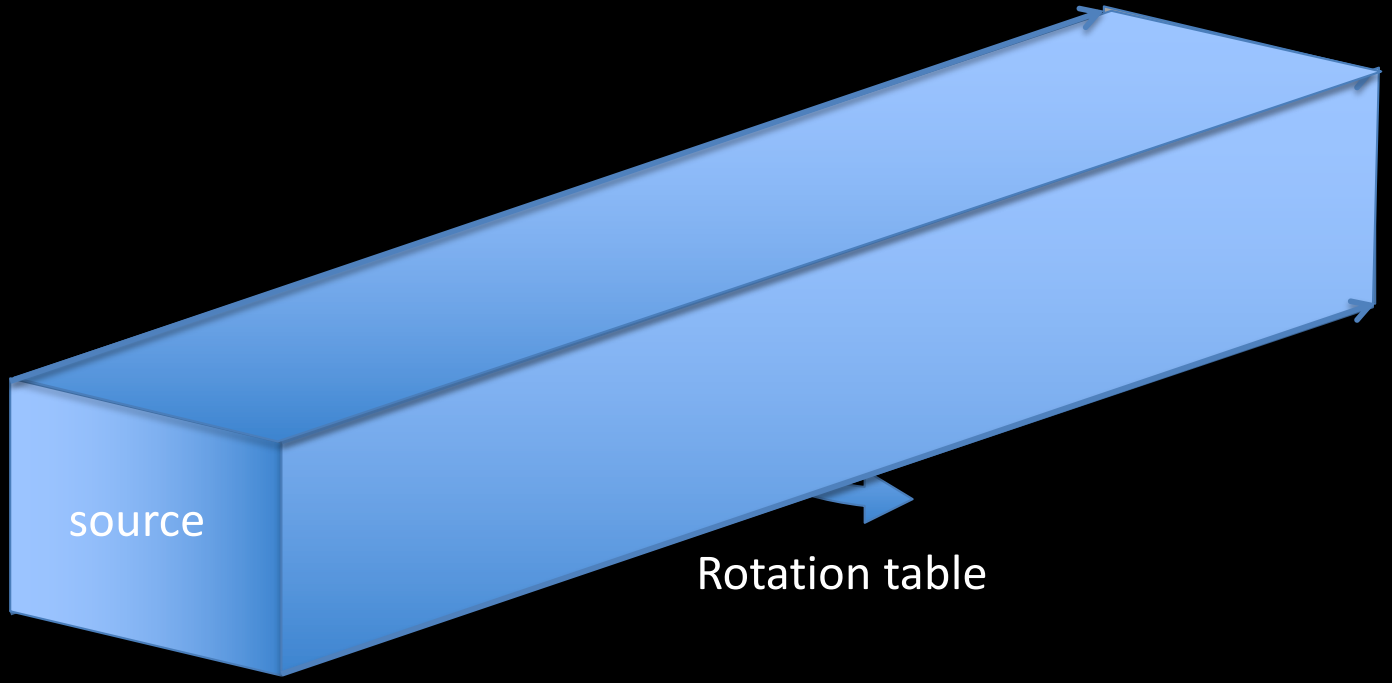
Stephen Hall

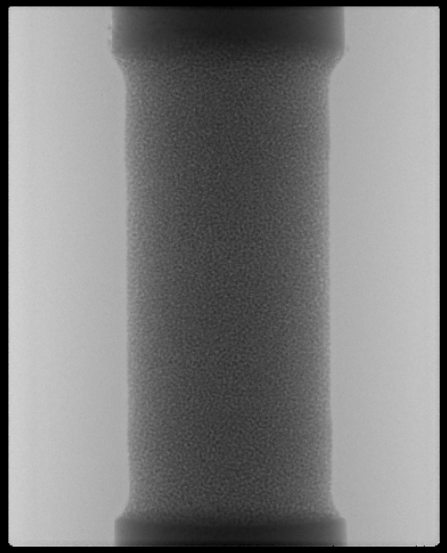
Division of Solid Mechanics, Lund University, Sweden
4D Imaging Lab, Lund University, Sweden
LINXS, Lund University, Sweden



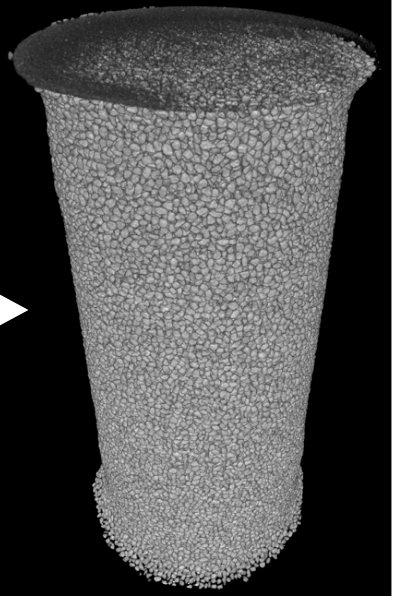
Full field Tomography imaging



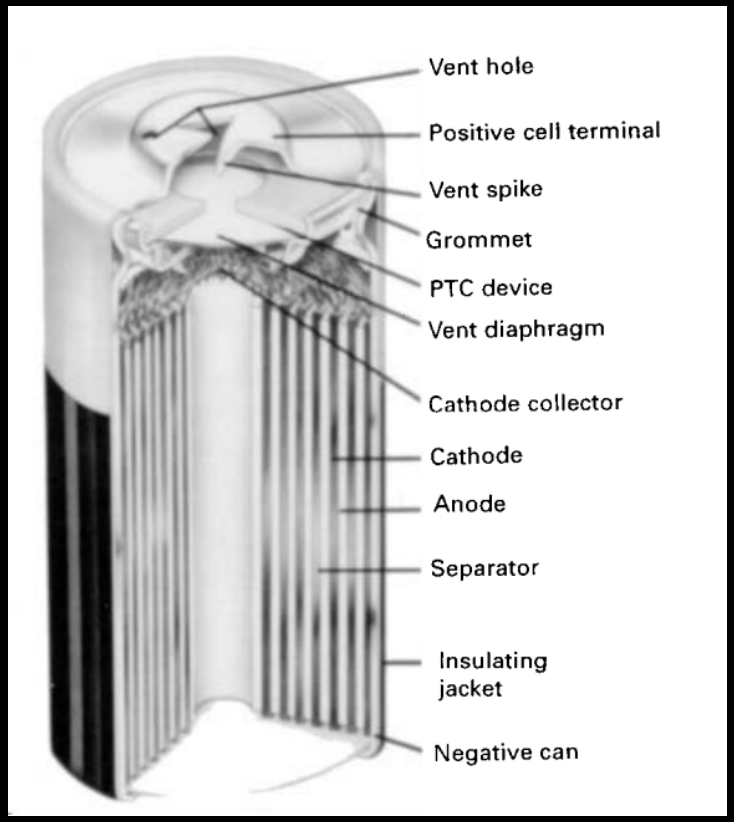


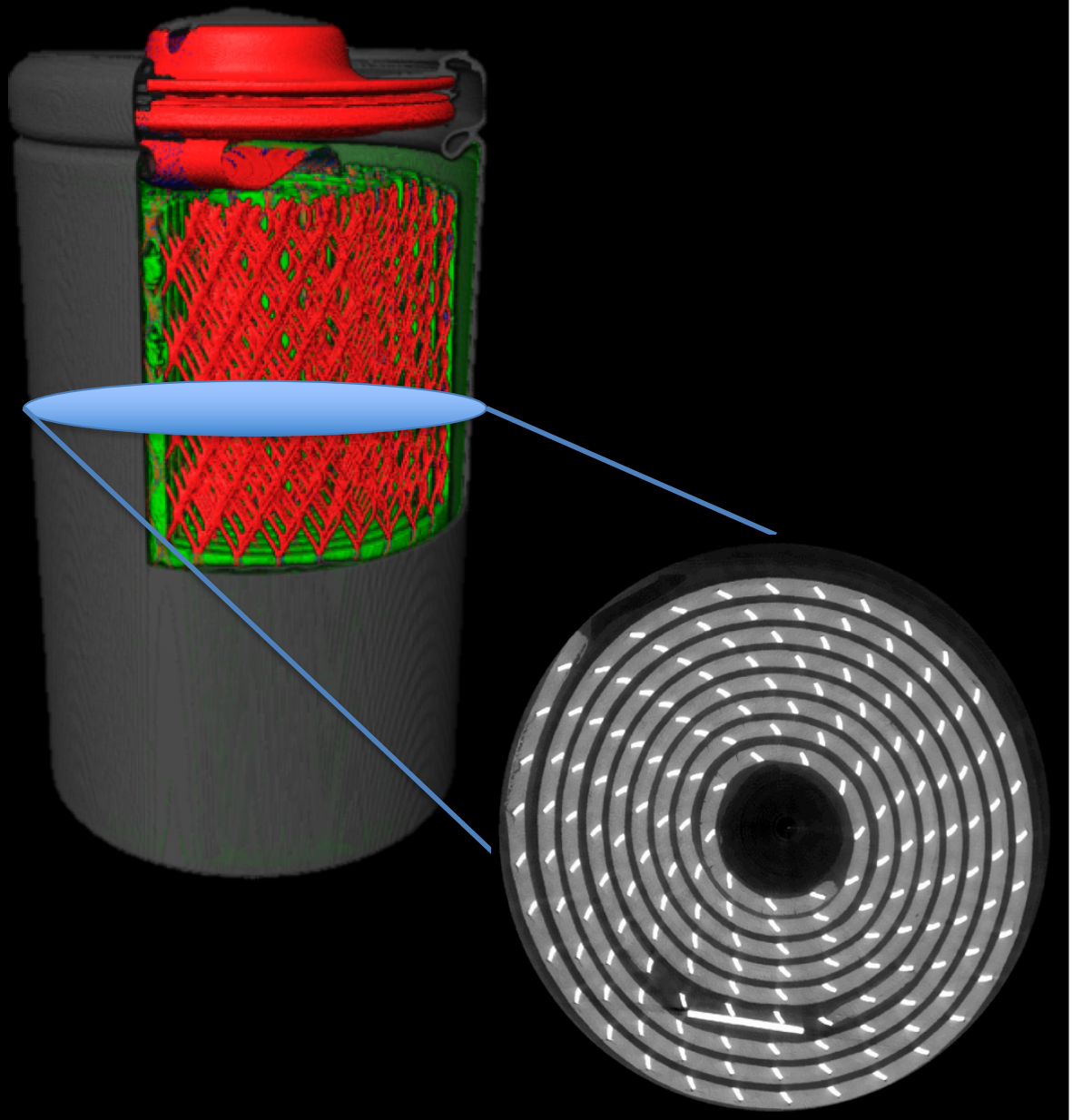


Reconstruction



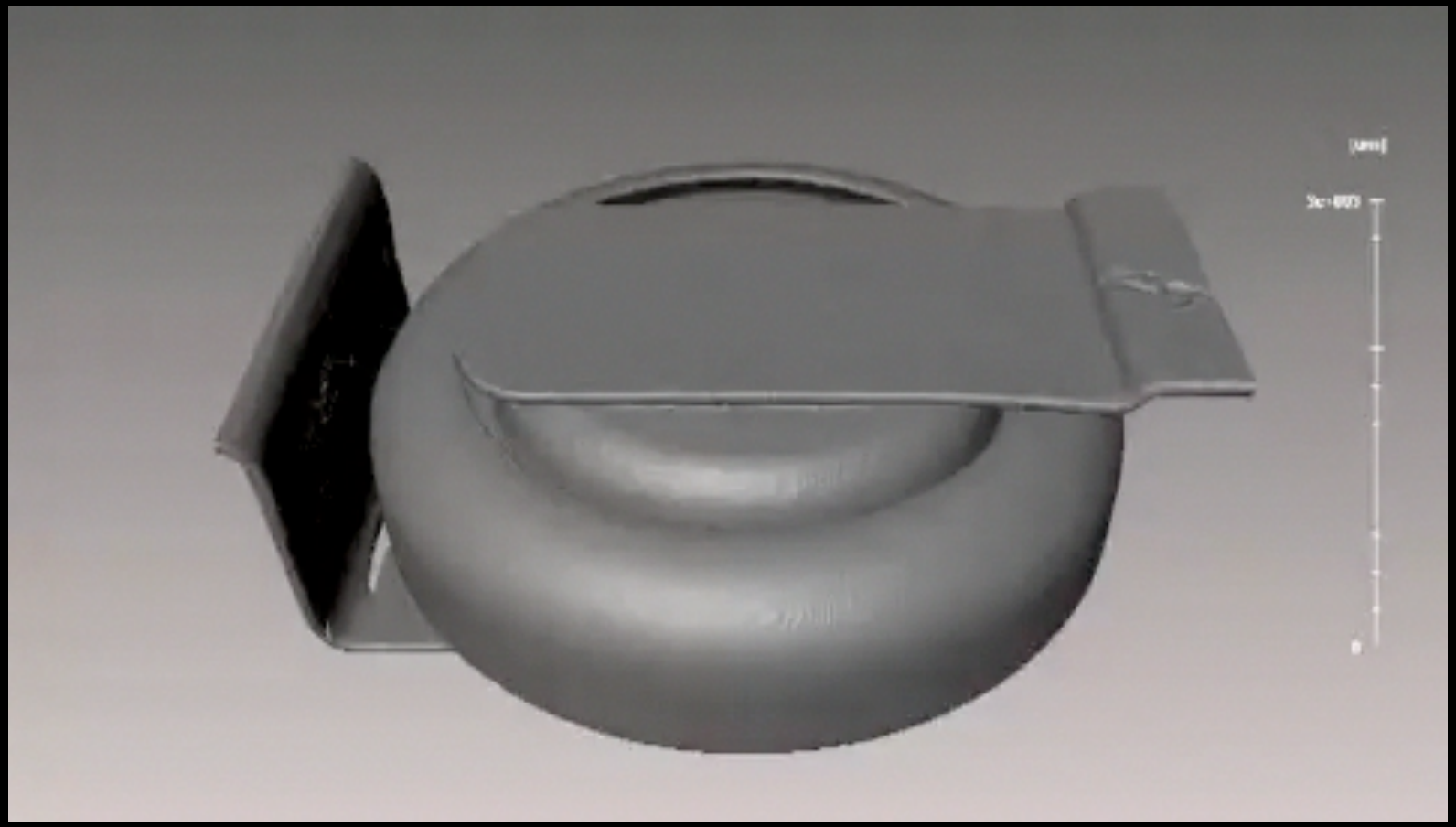
Full field Tomography imaging of batteries





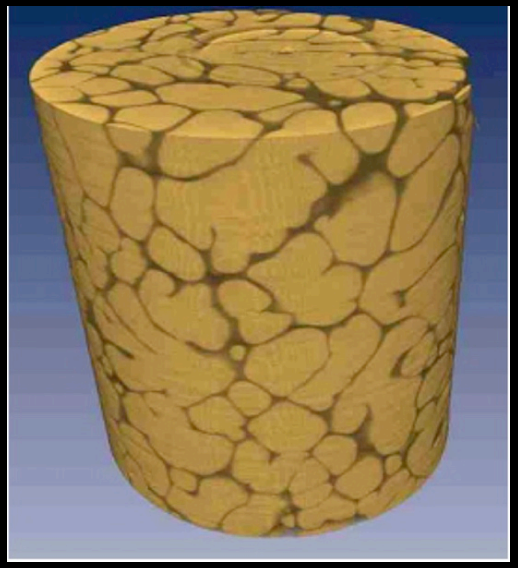
Collaboration Donal Finegan and Paul Shearing, UCL
Finegan et al., 2016, Advanced Science

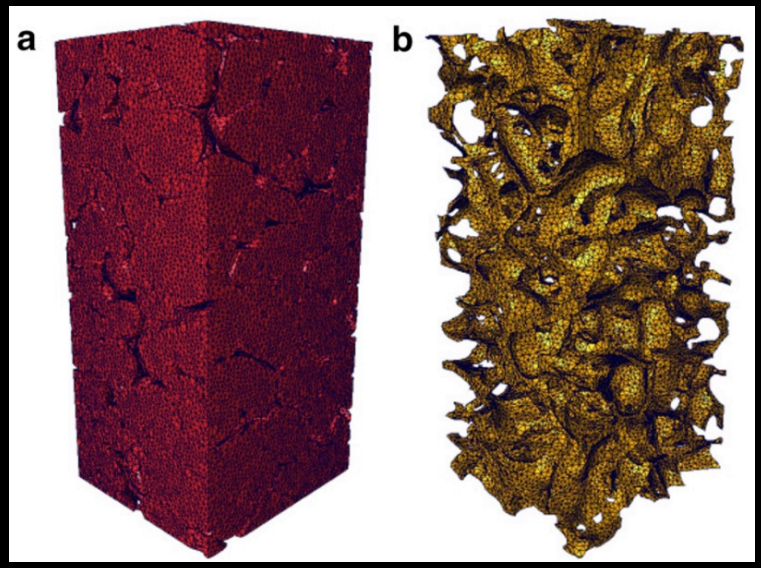
Full field Tomography imaging of batteries



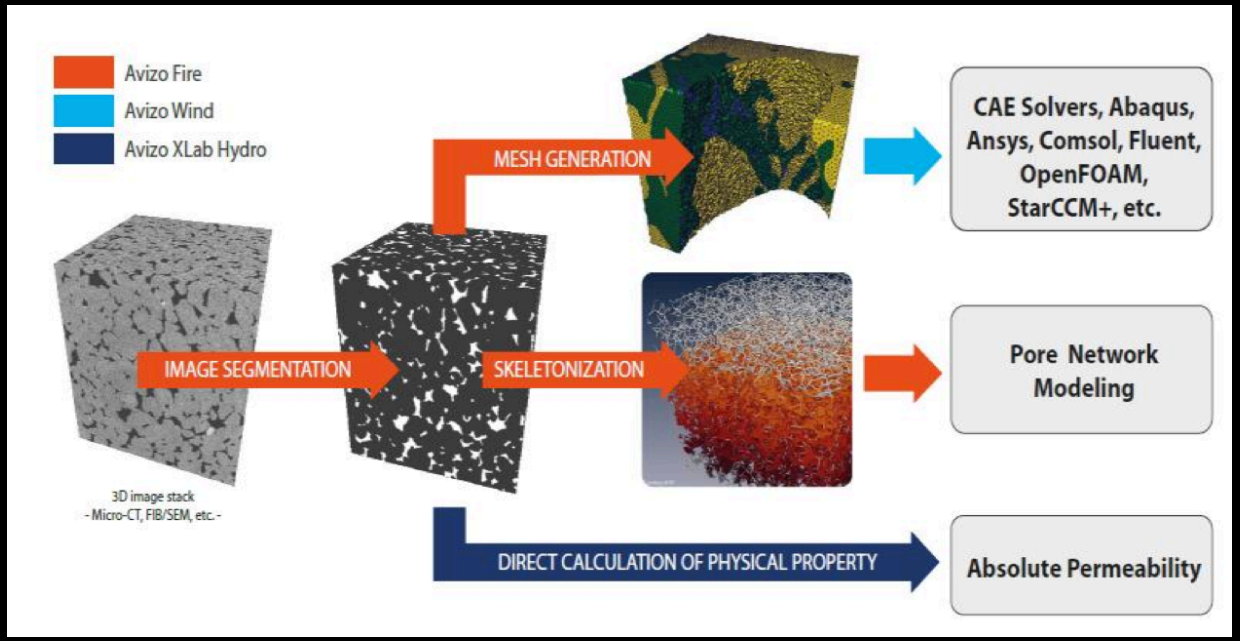
P. Shearing, UCL

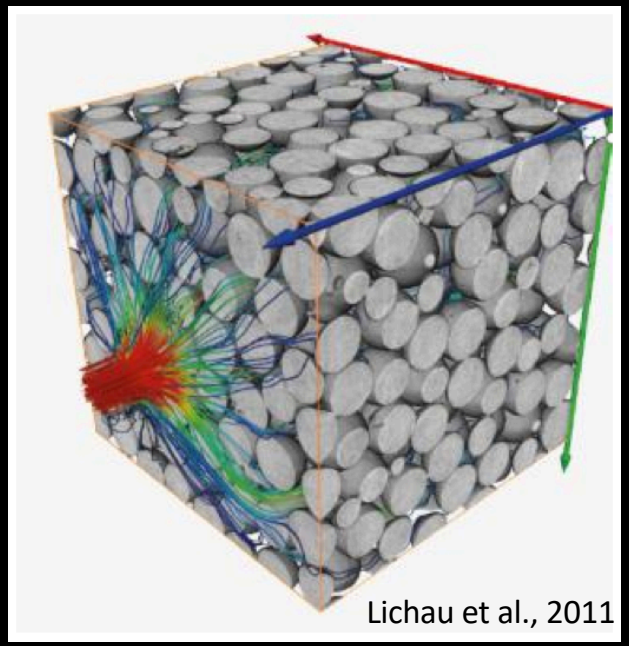
Structural imaging and characterisation → models





Madi et al., 2006

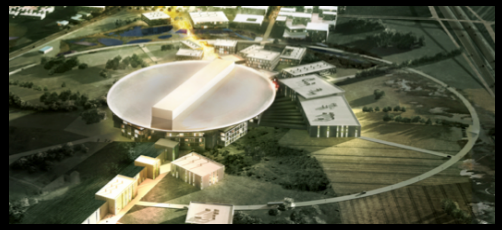




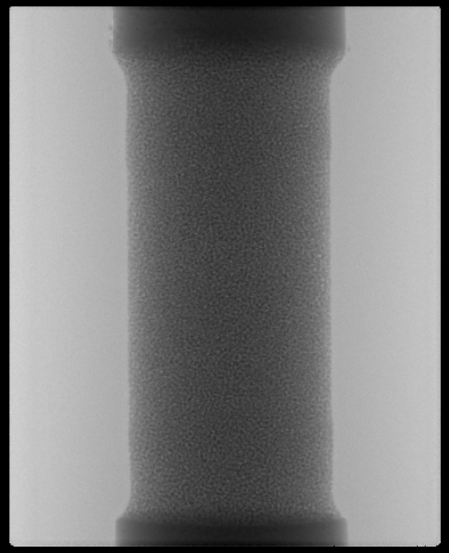
Lichau et al., 2011

4D imaging…

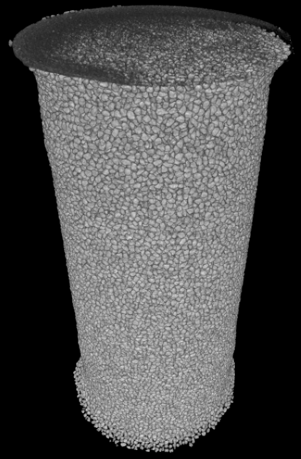
Imaging data



Radiographies

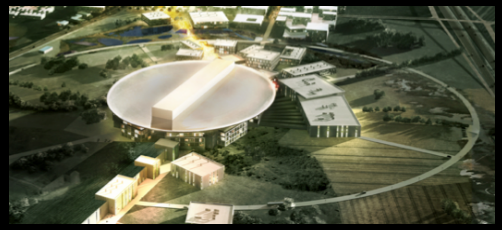


Reconstruction

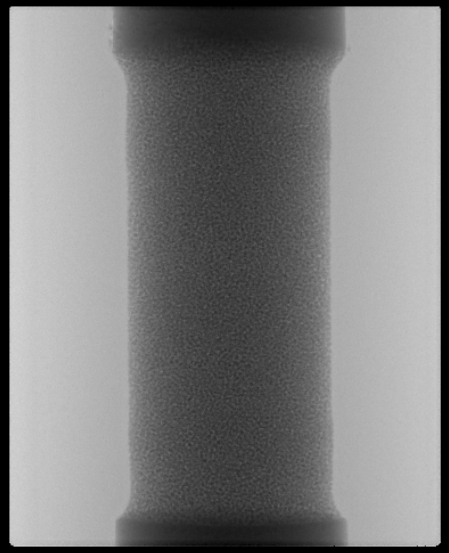


4D imaging…

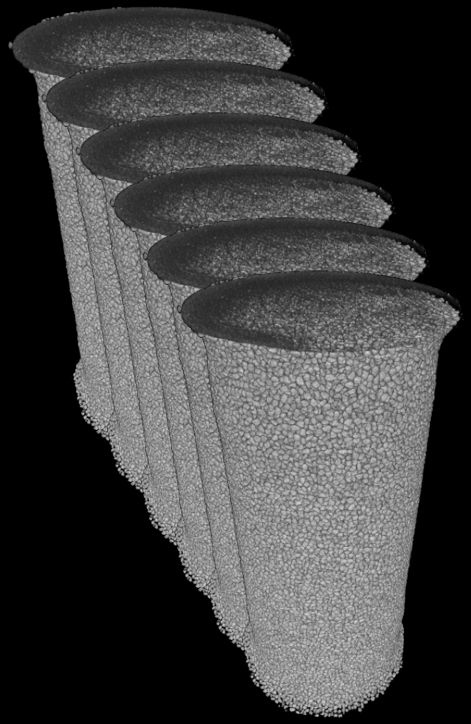
Imaging data



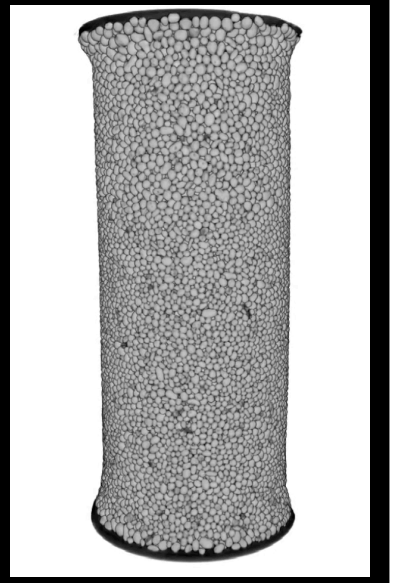
Radiographies



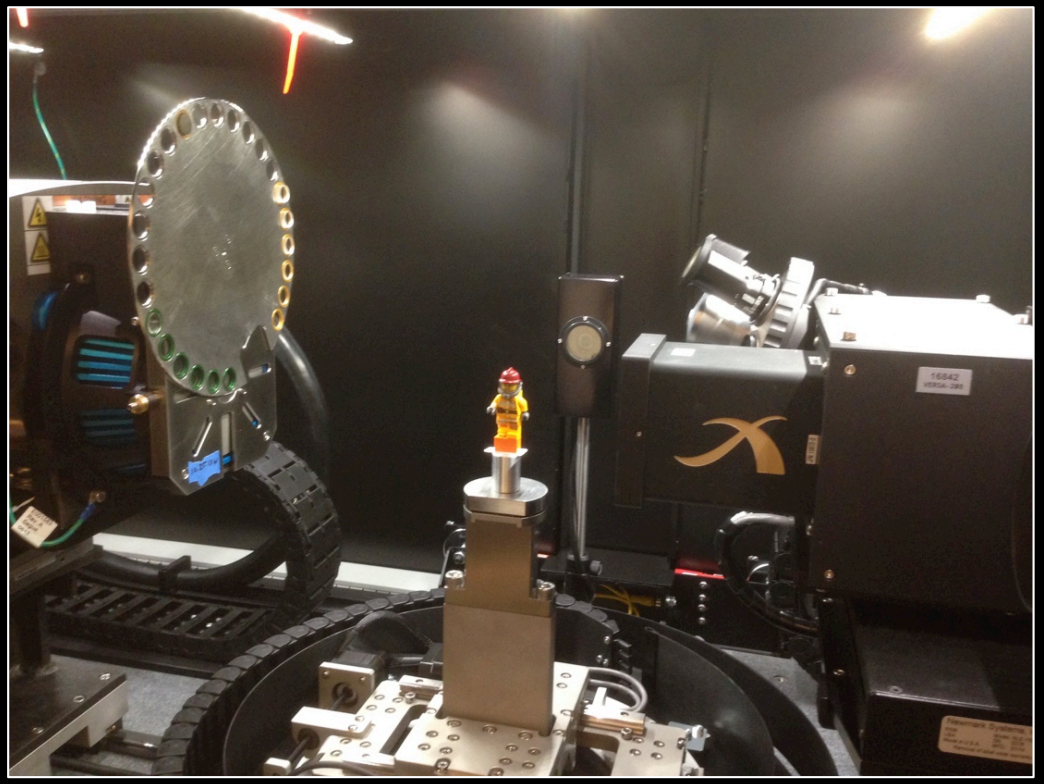
Reconstruction(s)



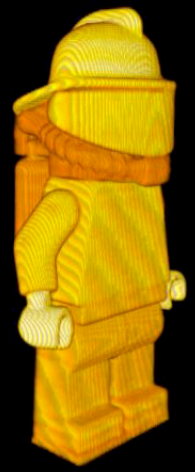
4D Visualisation

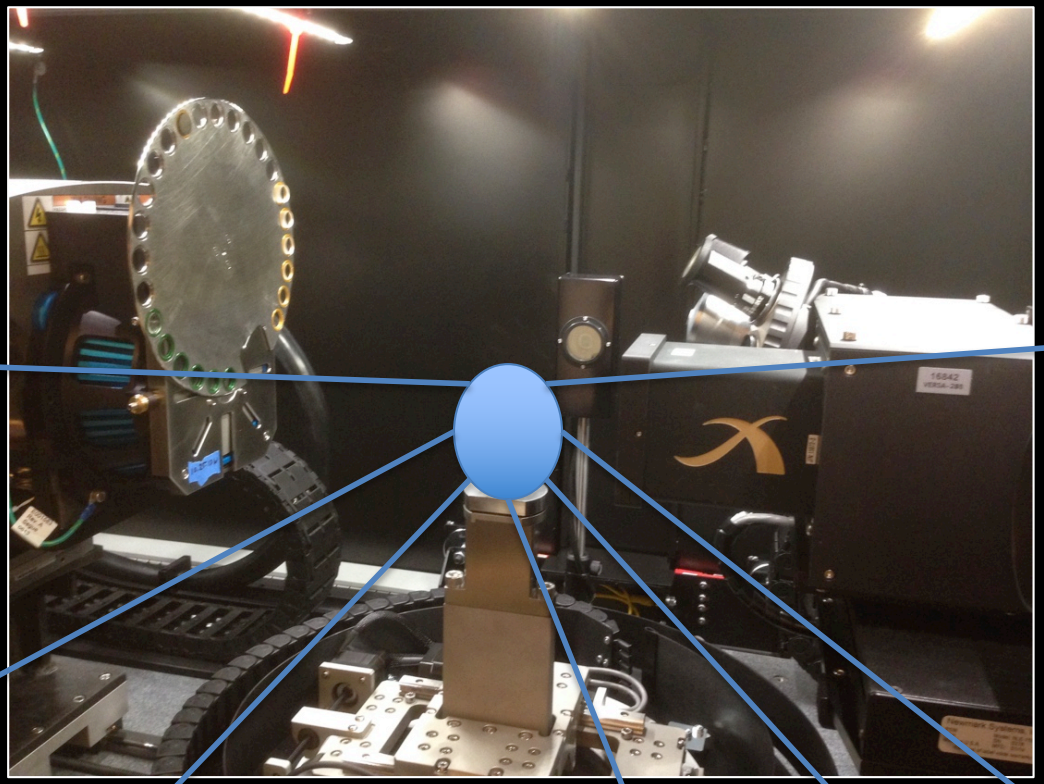


In-situ devices @ 4D Imaging Lab

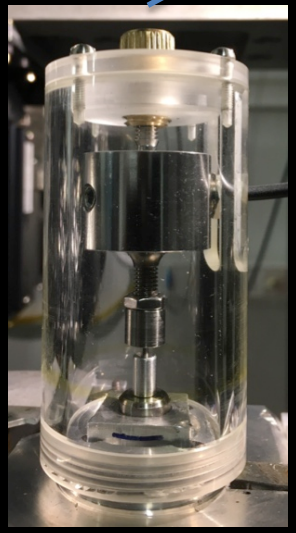


In-situ devices @ 4D Imaging Lab

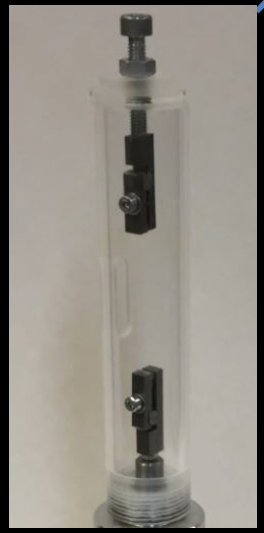


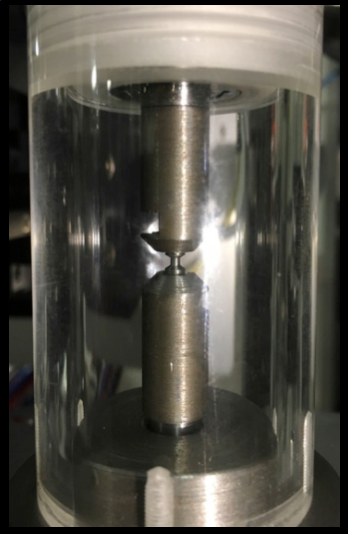


Electrochemical



Compression





Tension

Peeling

Vacuum/pressure

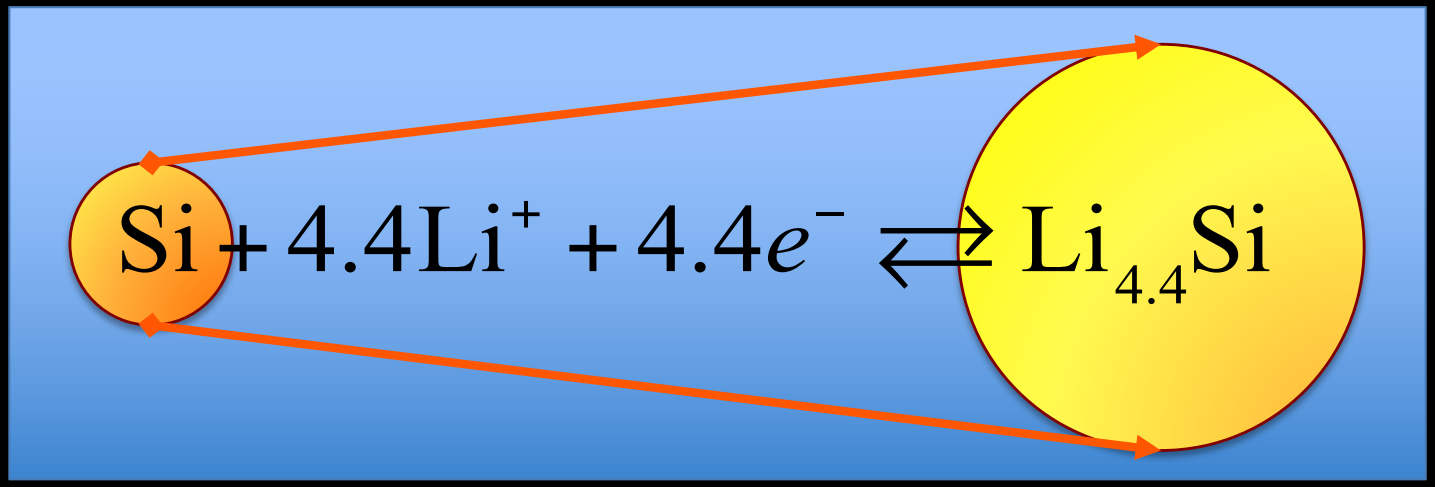
Humidity

Others and Future:

- Fluid flow
- Chemical reactions
- Heating
- Cooling (freezing)
- …

Si–Anode LIB

- Lithiation/delithiation of silicon during charge/discharge of LI-ion battery cells causes **dramatic volume changes (up to 330%)**.



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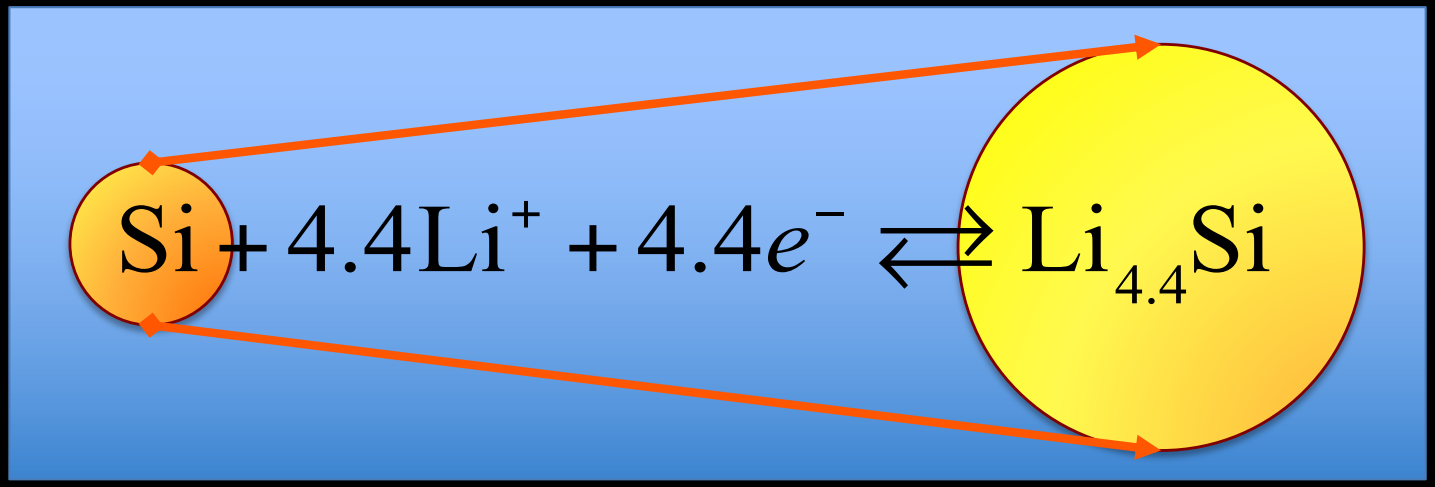
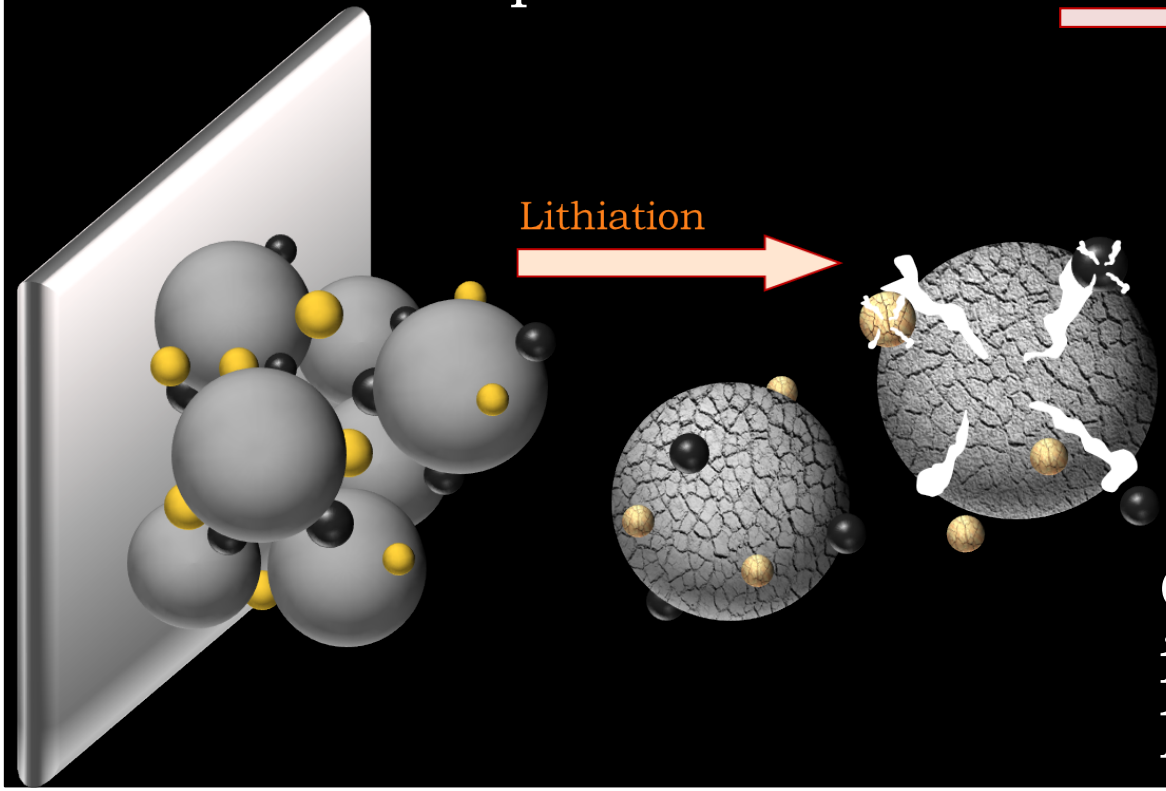


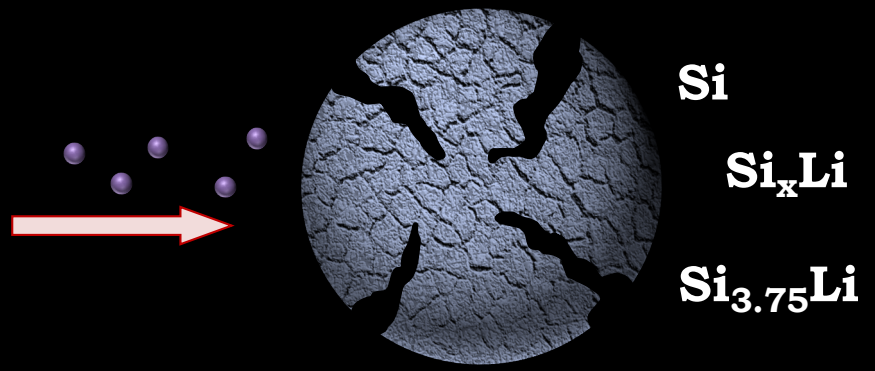
$$\frac{4.4\ \text{mol}_{\text{Li}}}{\text{mol}_{\text{Si}}}\ \frac{\text{mol}_{\text{Si}}}{28\ \text{g}_{\text{Si}}}\ \frac{96485\ \text{C}}{\text{mol}_{\text{Li}}}\ \frac{\text{mAh}}{3.6\ \text{C}} \approx 4200\ \text{mAh g}^{-1}$$

The energy density of Si-based electrodes is higher than graphite electrodes (~ 4200 mAh/g of Si against ~ 375 mAh/g of C).

Si–Anode LIB

- Lithiation/delithiation of silicon during charge/discharge of LI-ion battery cells causes **dramatic volume changes (up to 330%)**.
- Repeated expansion and contraction places huge strain on the silicon particles

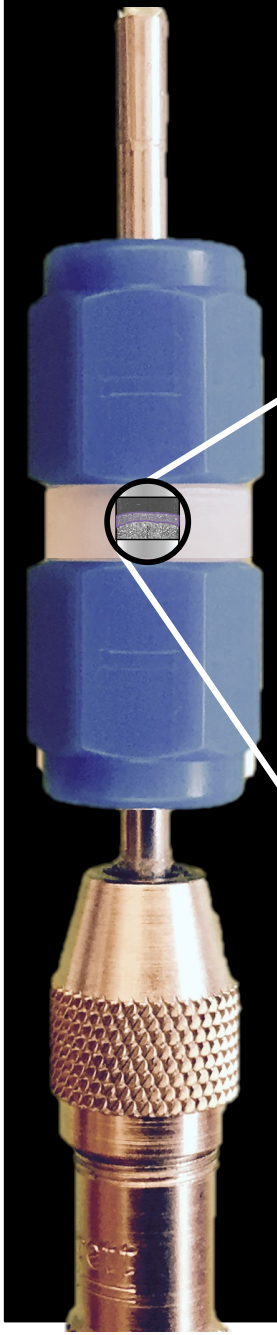


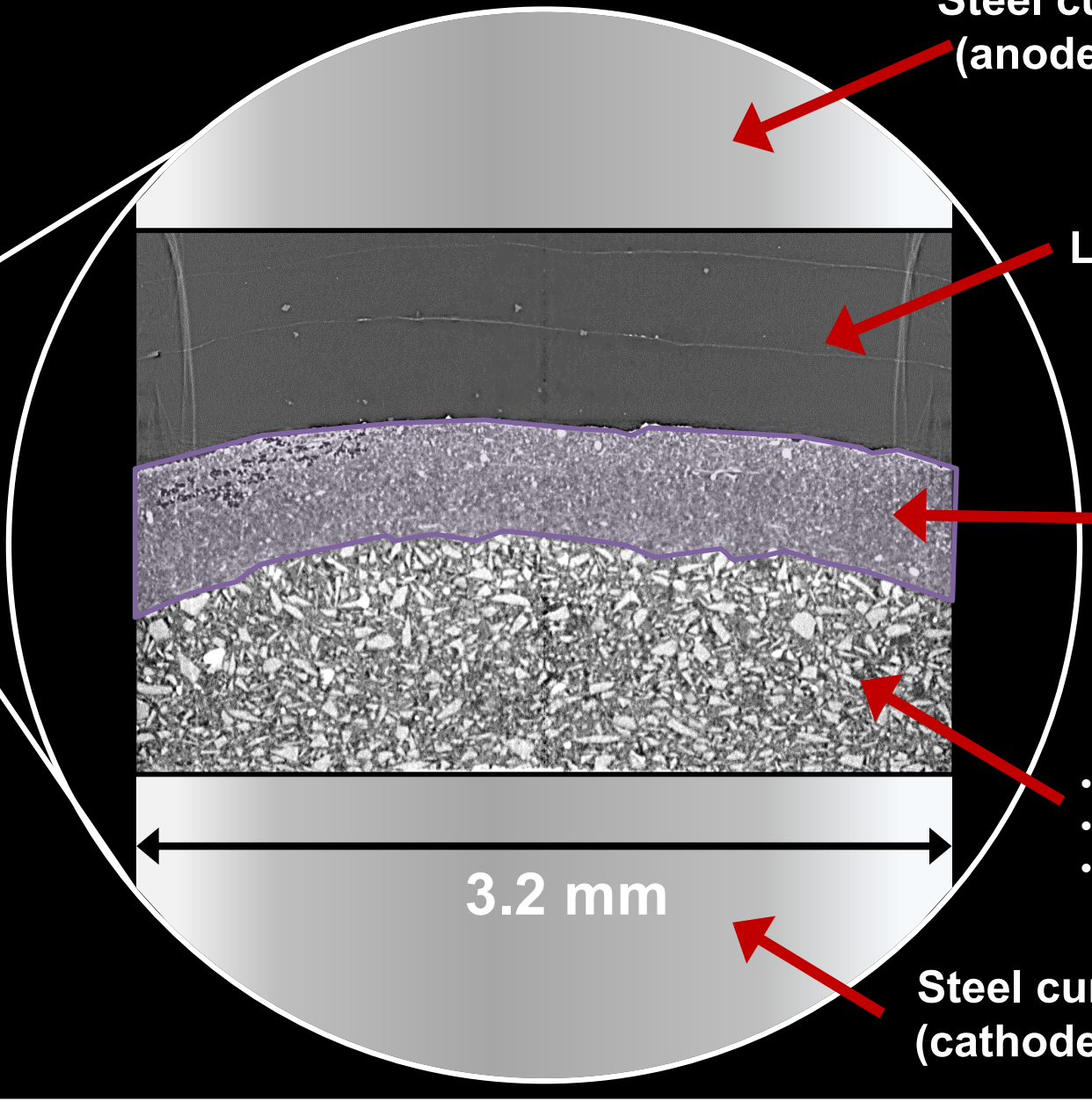


Components suffer irreversible damage and loss of electrical contact

UCL

Swagelok Battery Test Cells





Steel current collector (anode at discharge)

Lithium metal foil

Separator

Silicon-based electrode, w% :

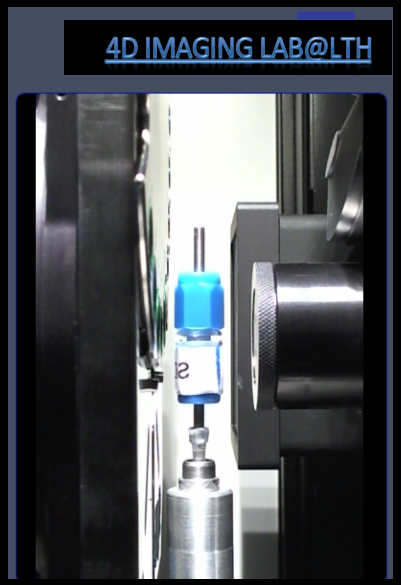
- 80% **Si**
- 10% conductive carbon
- 10% PVDF

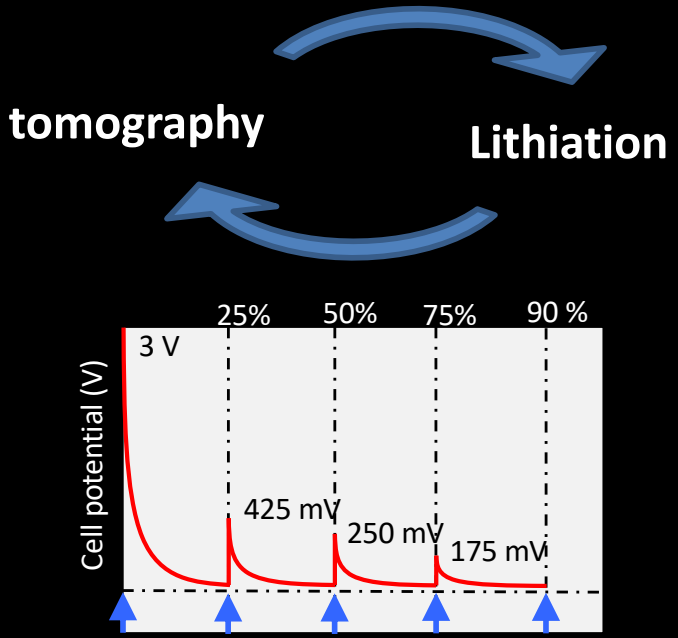
3.2 mm

Steel current collector (cathode at discharge)

4D IMAGING

- **Lab tomography**

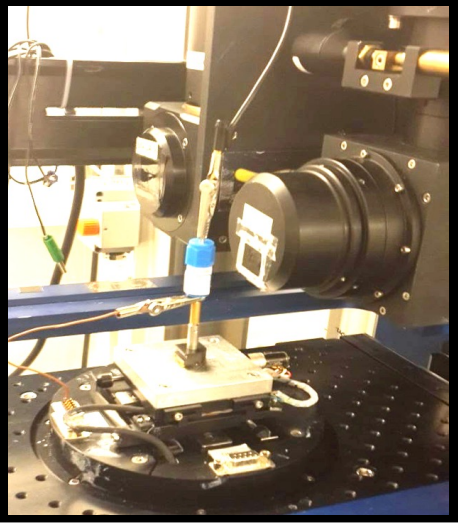


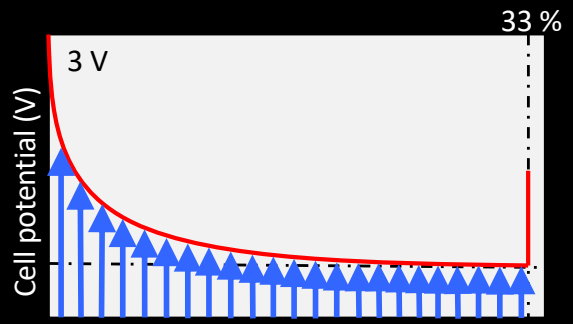


"ex-situ" discharging (+ charging)
&
"extended 4D" imaging

- Optimal for discontinuous (*ex situ*) acquisitions in repeated charge/discharge cycles over long periods

- **Synchrotron tomography**



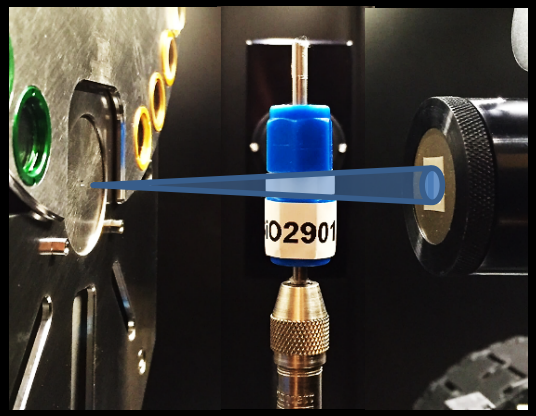


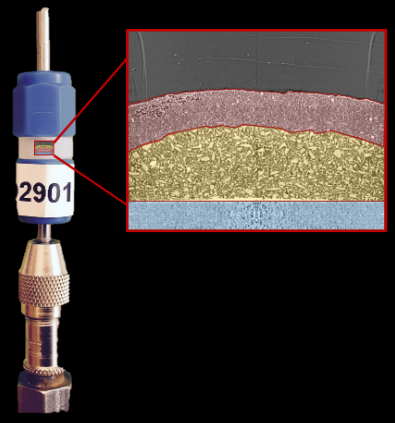
- Continuous acquisition throughout cell cycling test (**ca. 8.2 mins acquisition**)

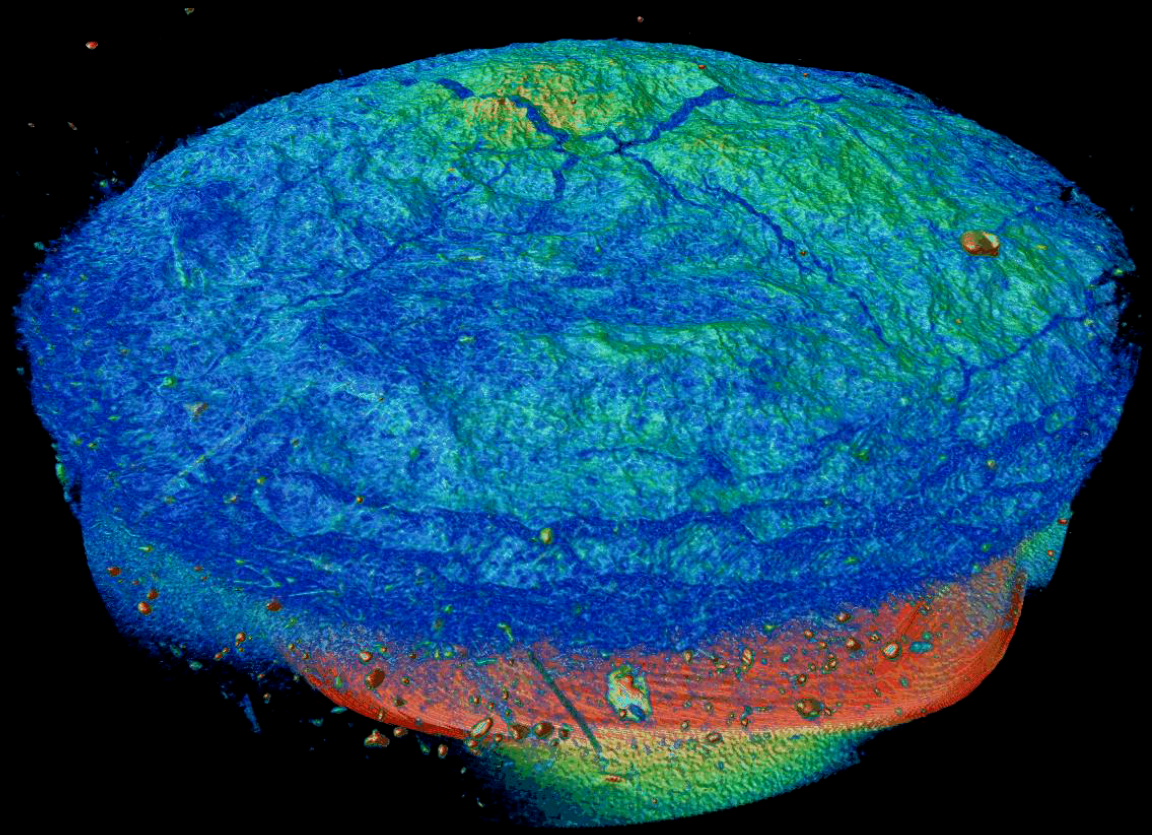
Fully in-situ + operando

- Faster, higher resolution:
  →optimal for continuous in-situ tests
- Limited time and limited access:
  →Inconvenient for repeated charge-discharge cycles

TOMOGRAPHY@4DIMAGING LAB



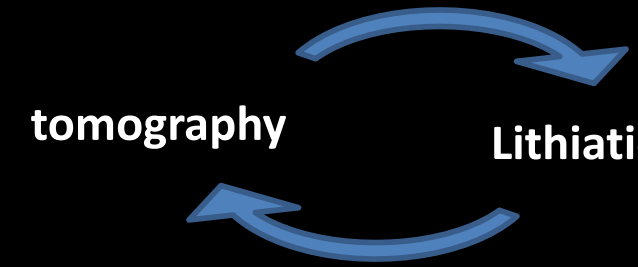




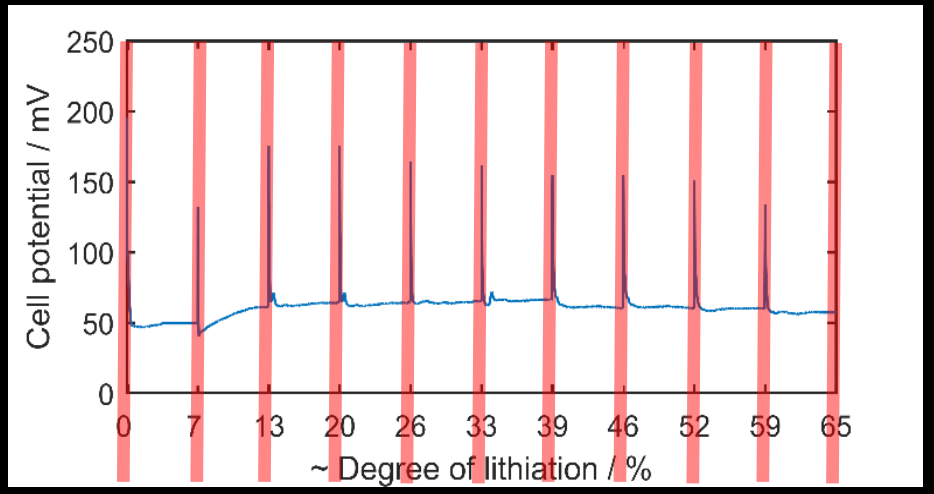
4D IMAGING LAB@LTH

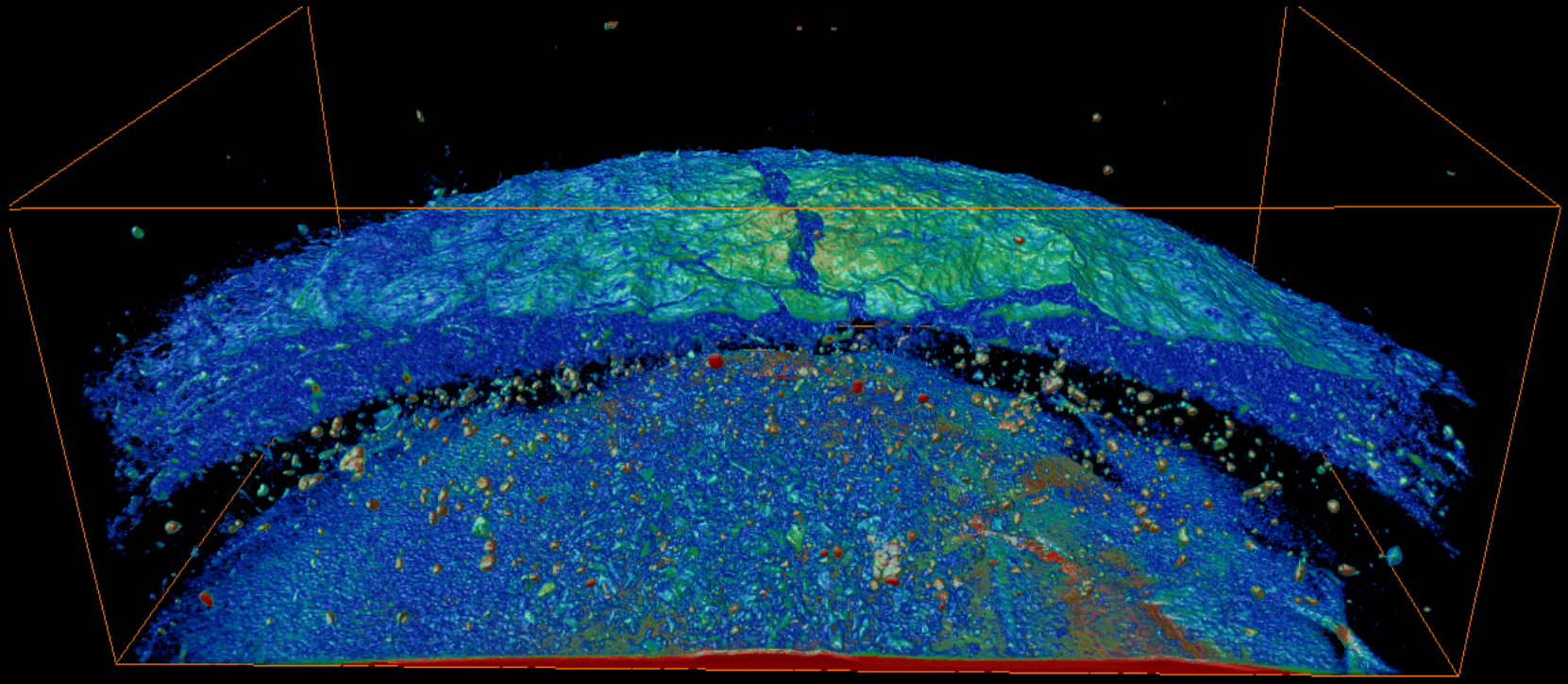
Paz Garcia et al., 2016, J. Power Sources

TOMOGRAPHY@4DIMAGING LAB



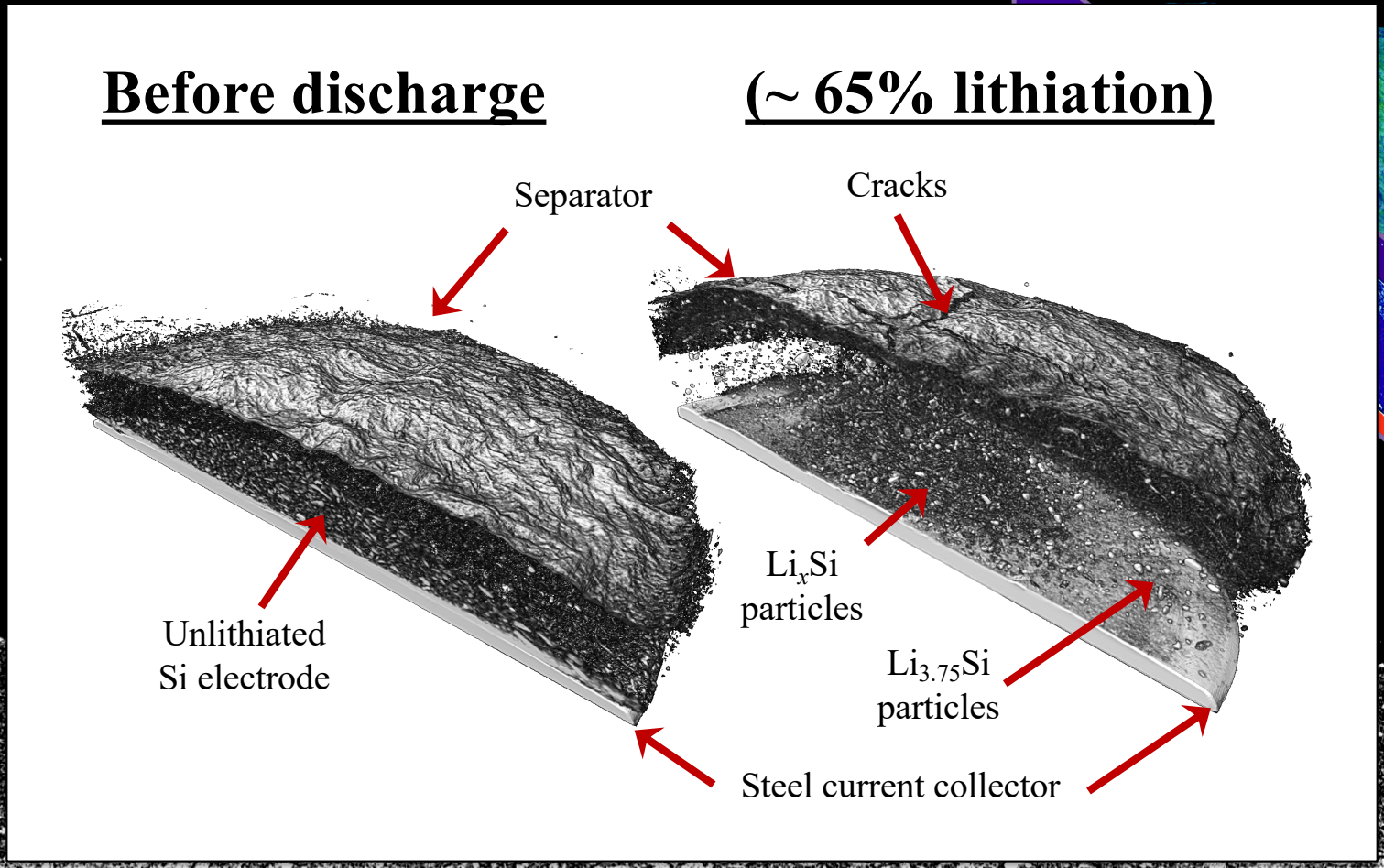
"ex-situ" discharging and "extended 4D" imaging





Paz Garcia et al., 2016, J. Power Sources

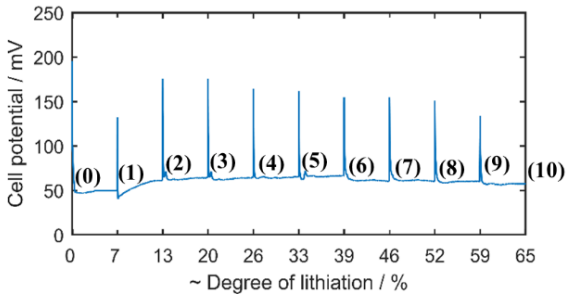
TIMELAPSE EVOLUTION DURING DISCHARGE



Paz Garcia et al., 2016, J. Power Sources

QUANTIFICATION

- **Grey-scale changes** → "lithiation"
- **Digital Volume Correlation** → Strain / volume change



Paz Garcia et al., 2016, J. Power Sources

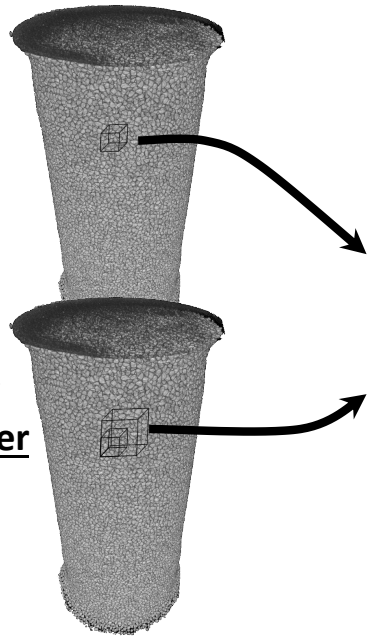
Digital Volume Correlation (DVC)

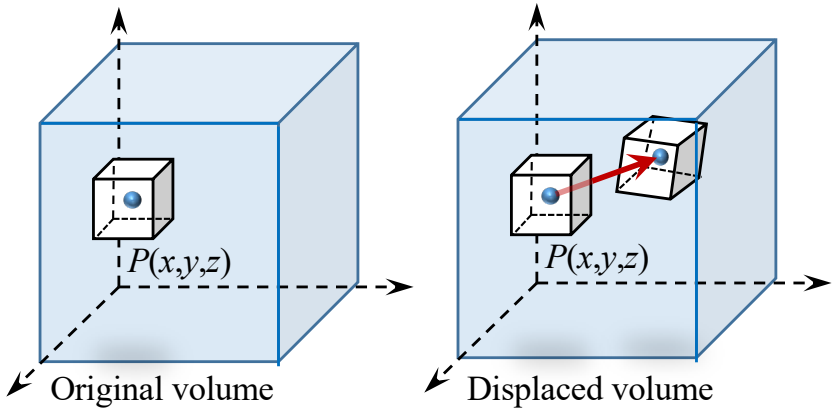
(in-house code "TomoWarp2")

(See Tudisco et al., 2018)

X-ray tomography image volume **before** loading

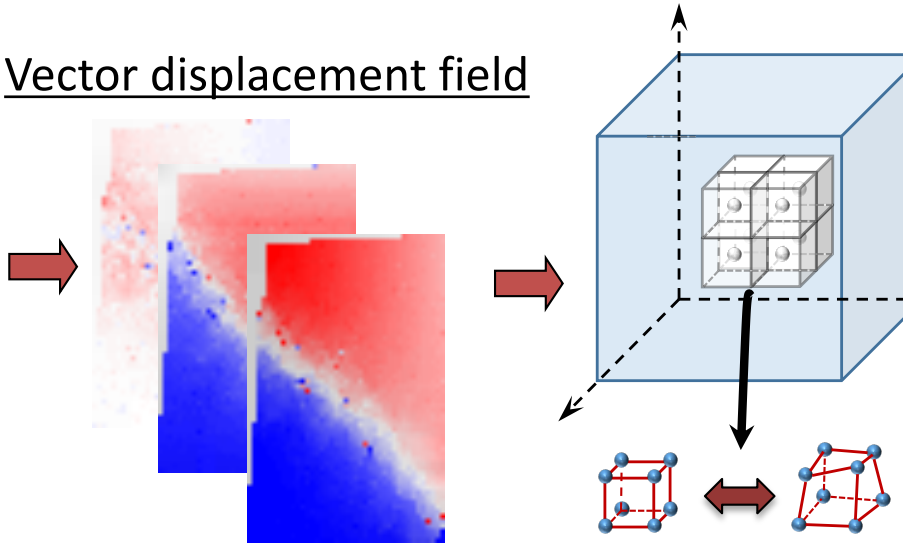
X-ray tomography image volume **after** loading





3D search 3D for transformation vector based on best image correlation → displacements (+ rotation / distortion)

Vector displacement field



Continuum hypothesis

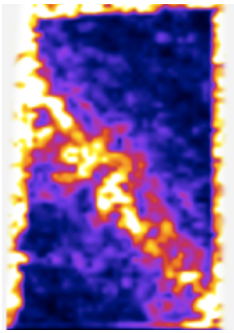
Full 3D strain tensor field

$$\varepsilon_{ij} = \begin{pmatrix} \varepsilon_{xx} & \varepsilon_{xy} & \varepsilon_{xz} \\ \varepsilon_{yx} & \varepsilon_{yy} & \varepsilon_{yz} \\ \varepsilon_{zx} & \varepsilon_{zy} & \varepsilon_{zz} \end{pmatrix}$$

Strain invariants
- volumetric and shear strains

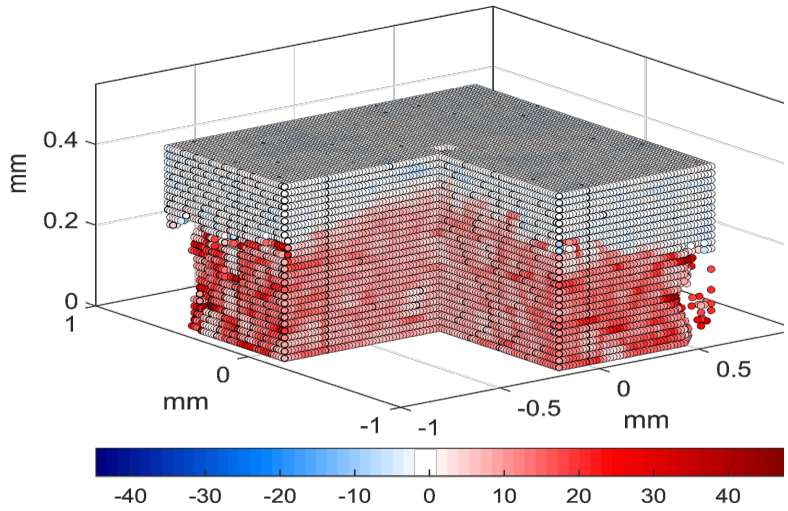
$$\varepsilon_v = \varepsilon_1 + \varepsilon_2 + \varepsilon_3$$

$$\varepsilon_s = \sqrt{\left(\frac{\varepsilon_1 - \varepsilon_2}{2}\right)^2 + \left(\frac{\varepsilon_1 - \varepsilon_3}{2}\right)^2 + \left(\frac{\varepsilon_2 - \varepsilon_3}{2}\right)^2}$$

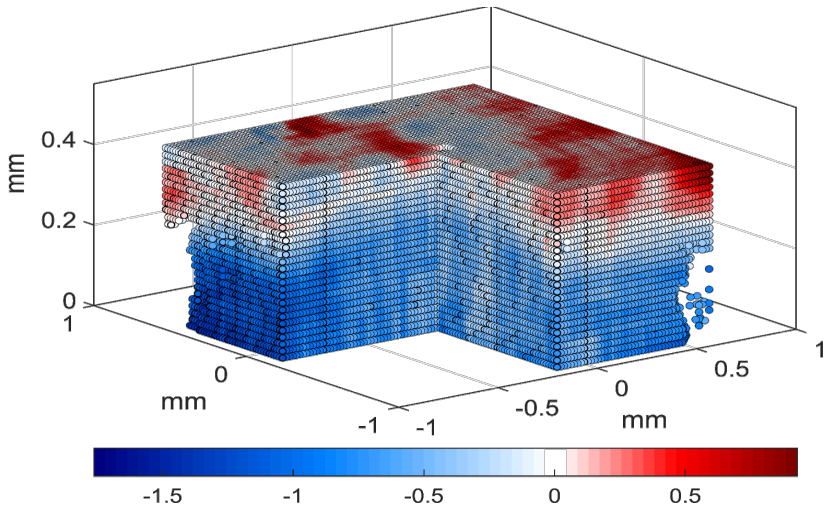


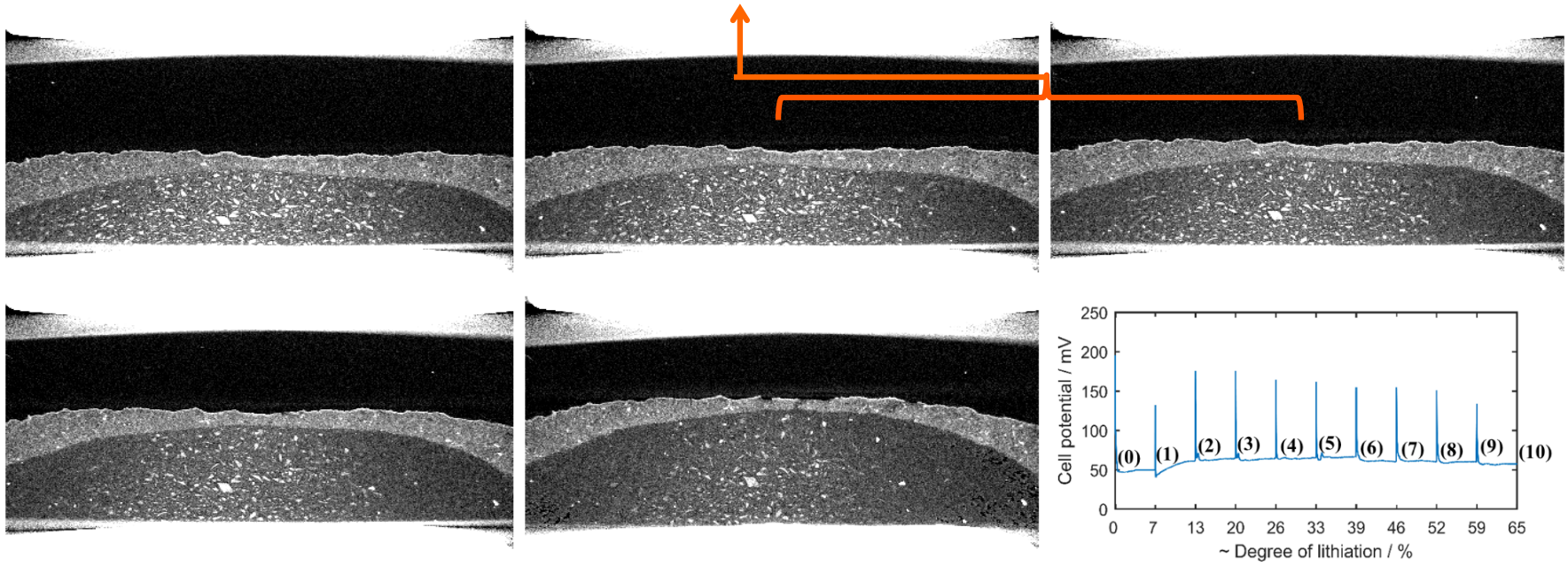
QUANTIFICATION

Volume changes



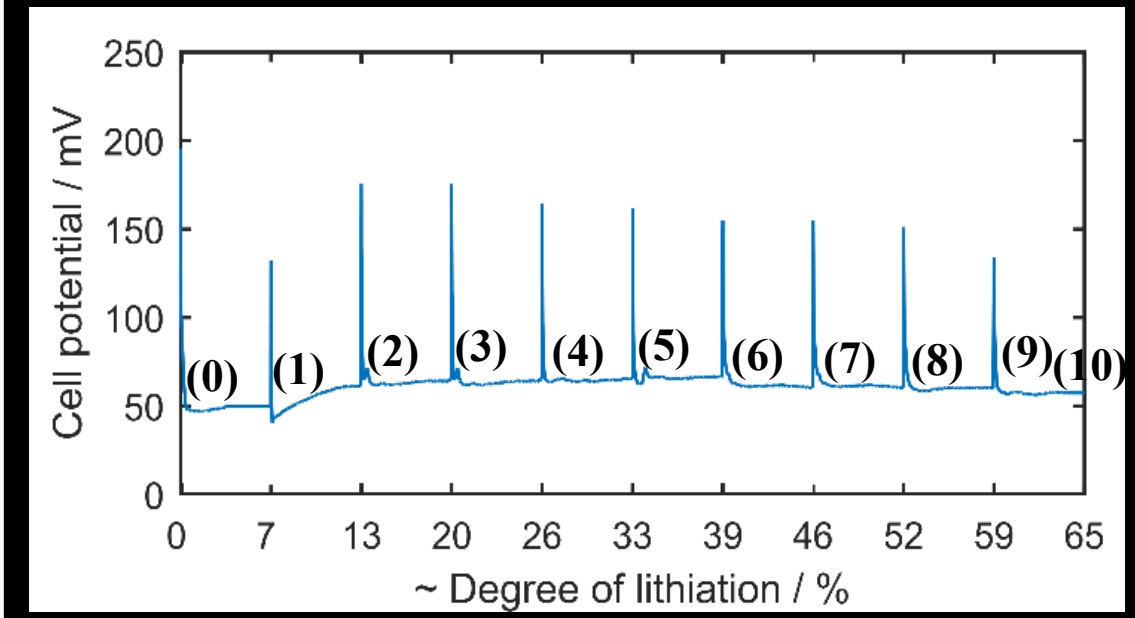
Grey-scale changes

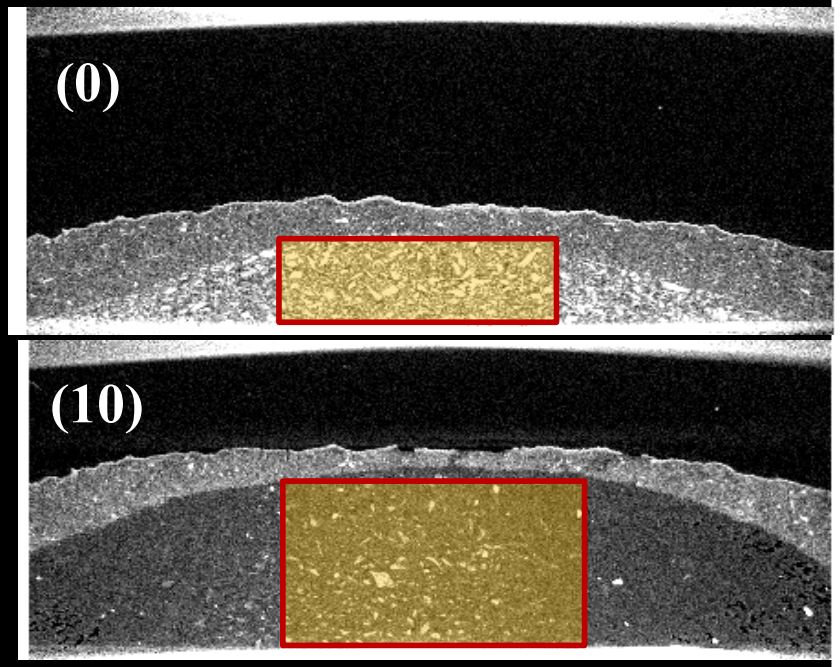


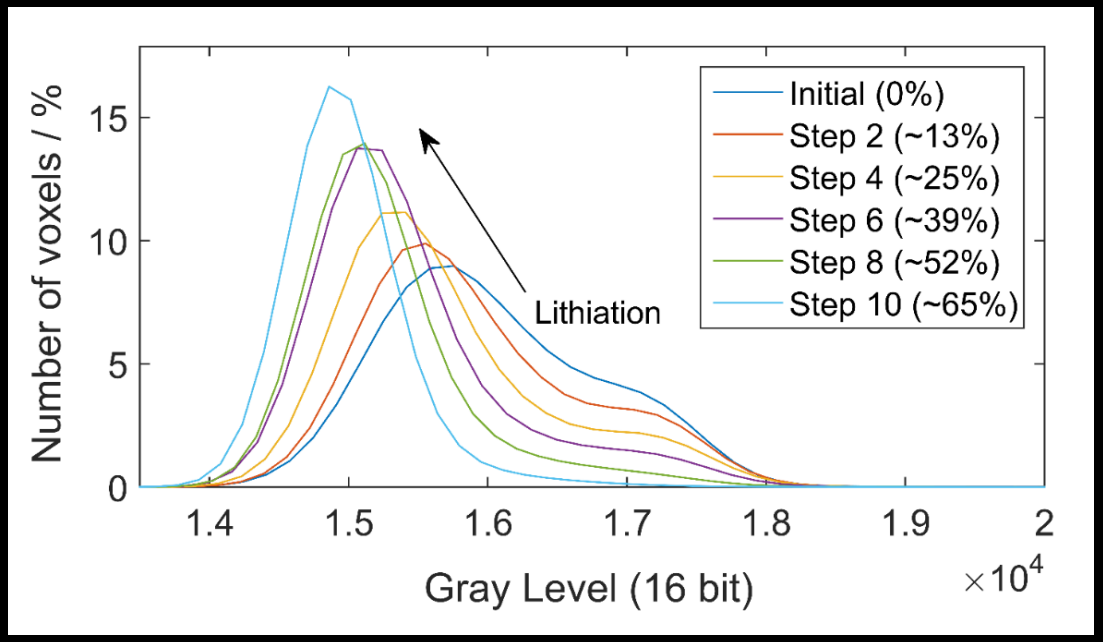


Paz Garcia et al., 2016, J. Power Sources

Quantification: Grey-Scale Changes (Lithiation)



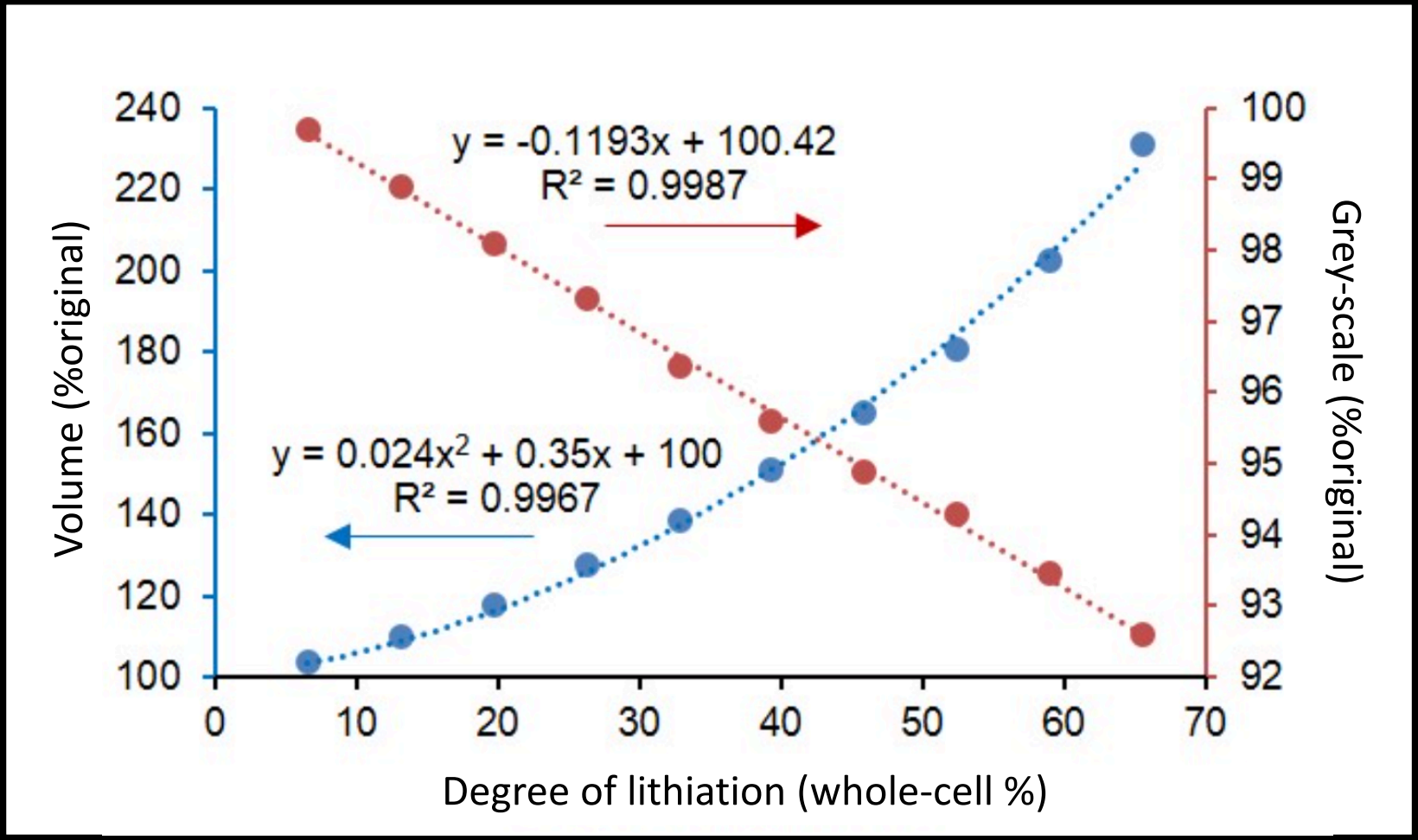




Paz Garcia et al., 2016, J. Power Sources

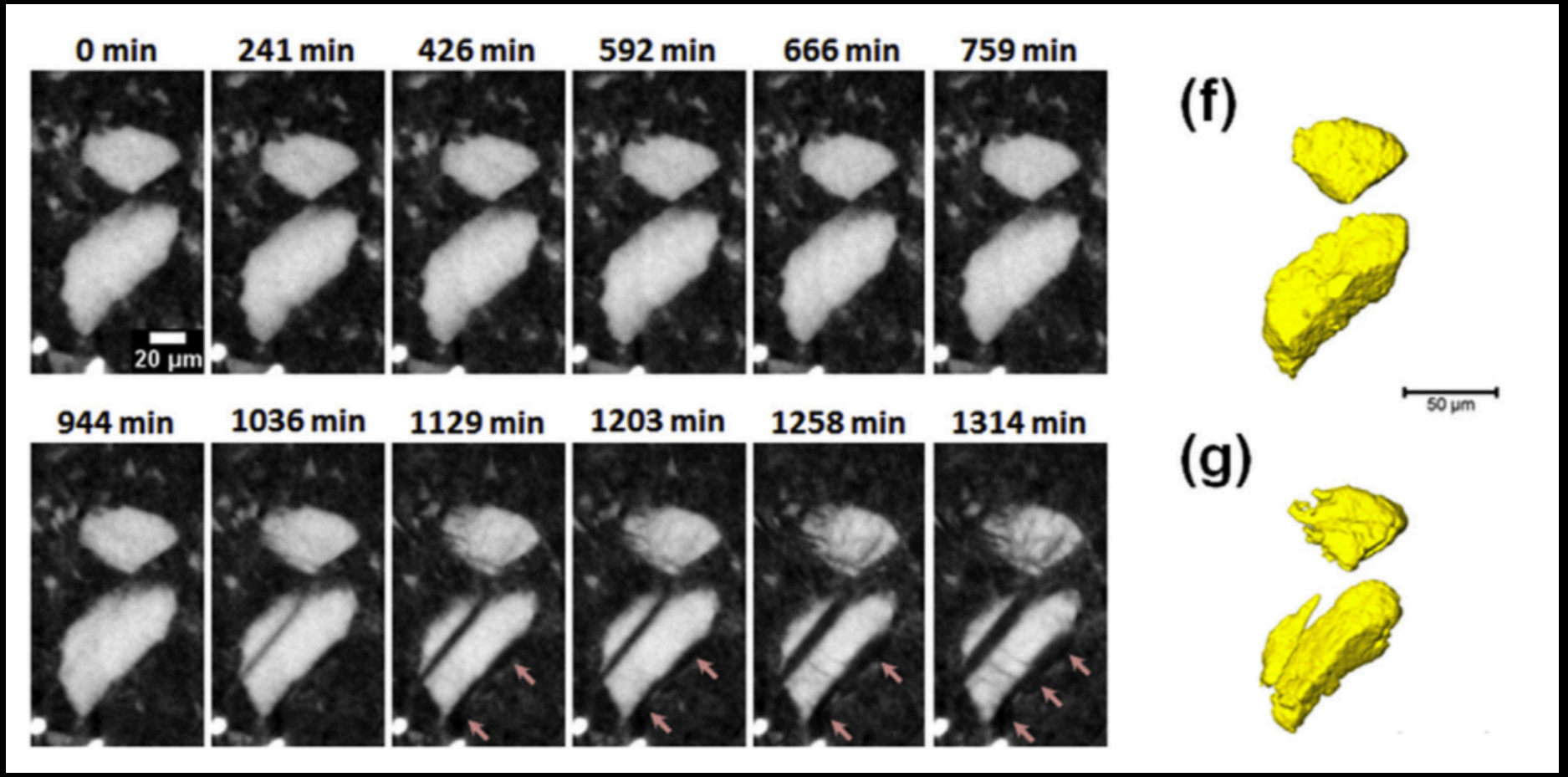
STRAIN VS GREYSCALE CHANGE

- Average volume/grey-scale change in Si electrode from 4D image analysis versus average degree of lithiation (based on the theoretical capacity of the cell)



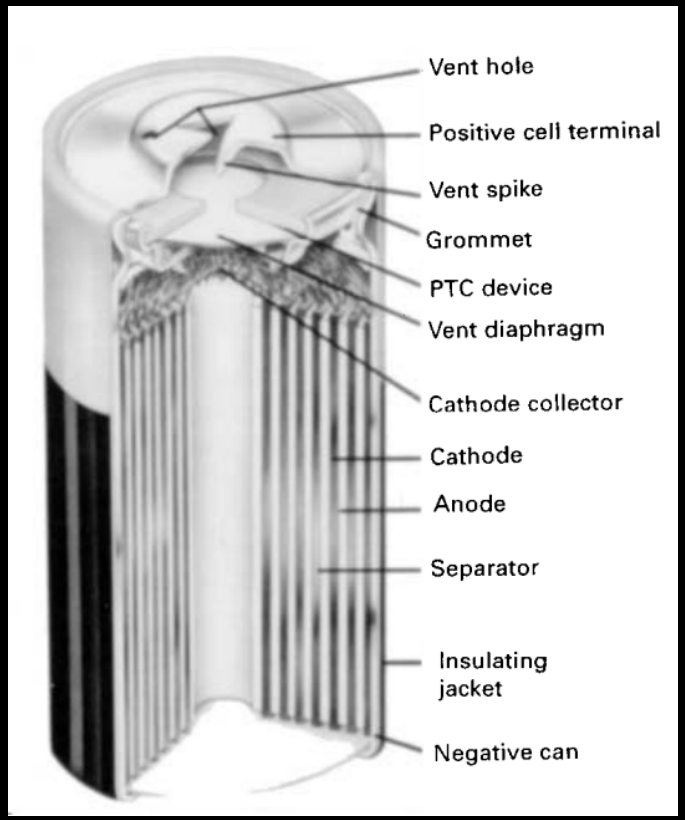
Paz Garcia et al., 2016, J. Power Sources

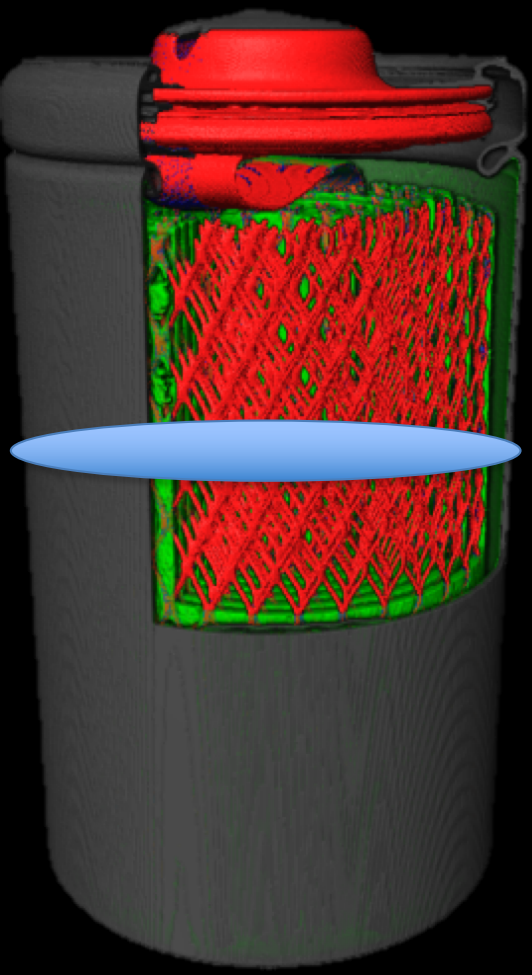
GRAIN SCALE ANALYSIS



Taiwo et al., 2017, J. Power Sources

Deformation during charge /discharge of a CR2 Li-ion battery





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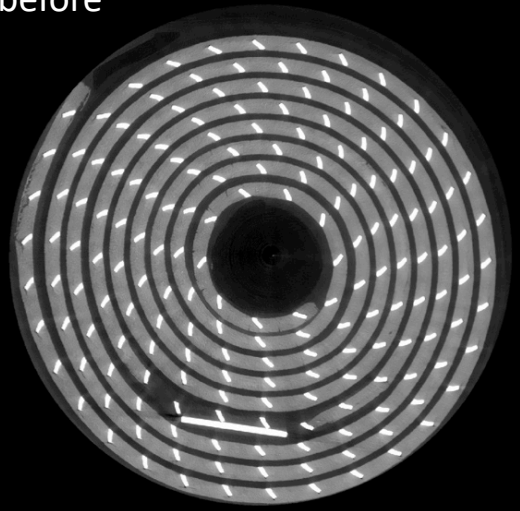

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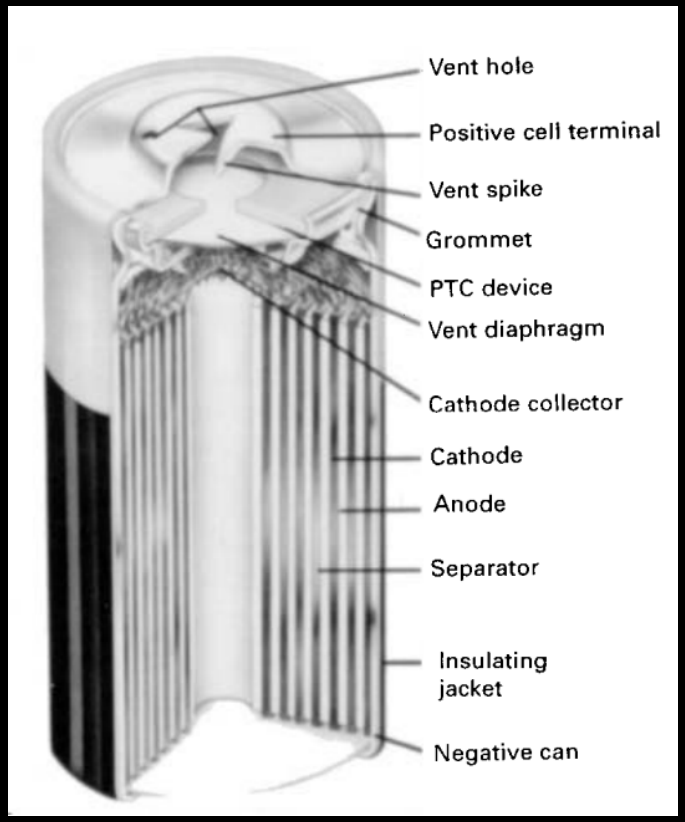
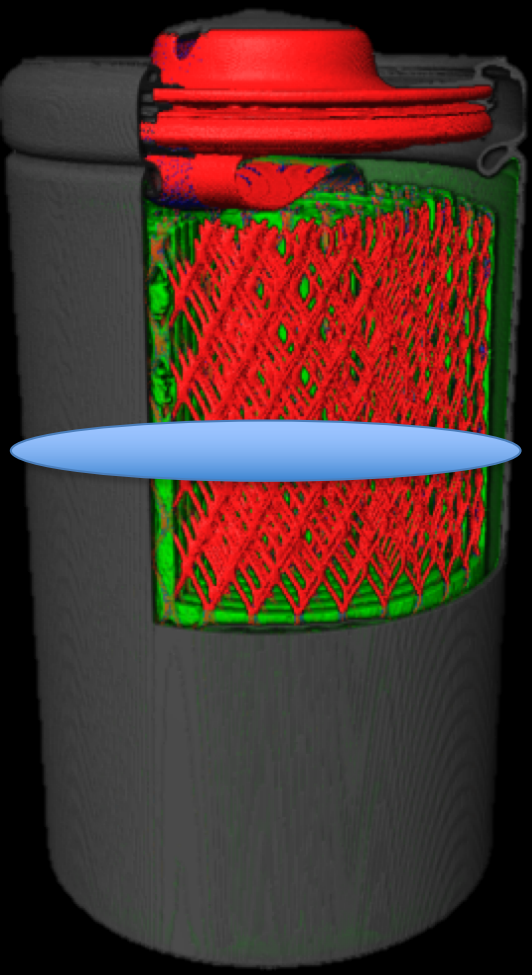
Finegan et al., 2016, Advanced Science

Deformation during charge /discharge of a CR2 Li-ion battery





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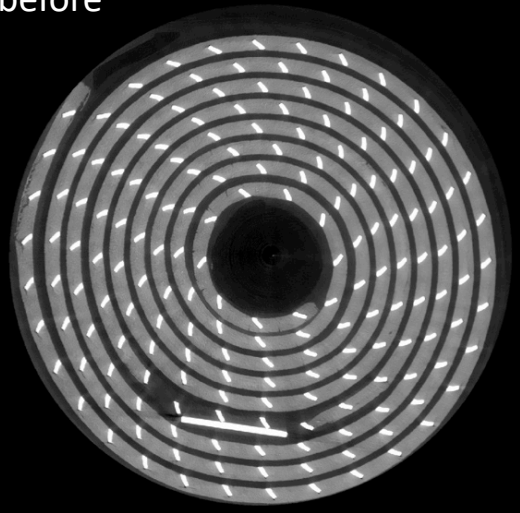
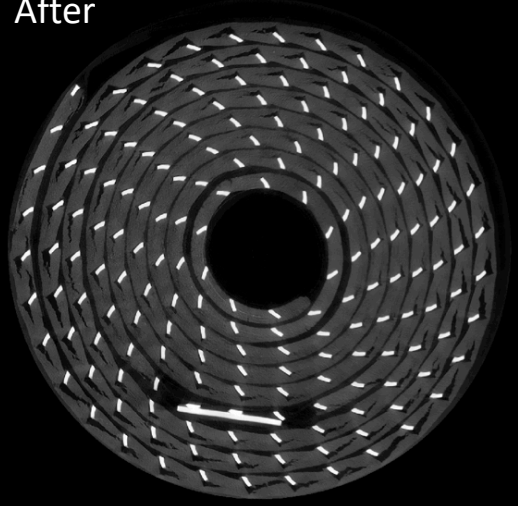


After



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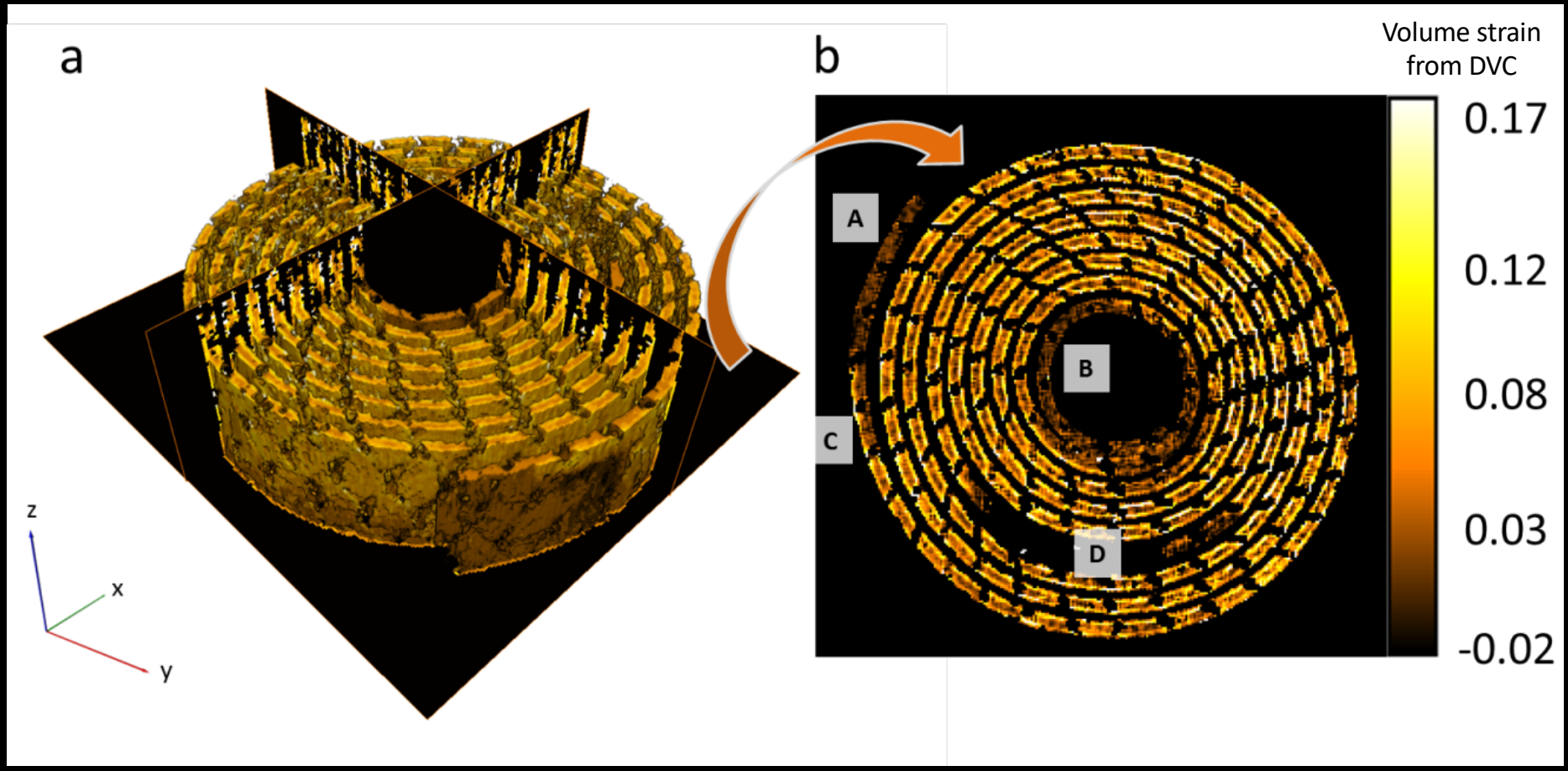
Finegan et al., 2016, Advanced Science

Deformation during charge /discharge of a CR2 Li-ion battery

In-operandi x-ray tomography during charging

Quantification: Volume change from DVC



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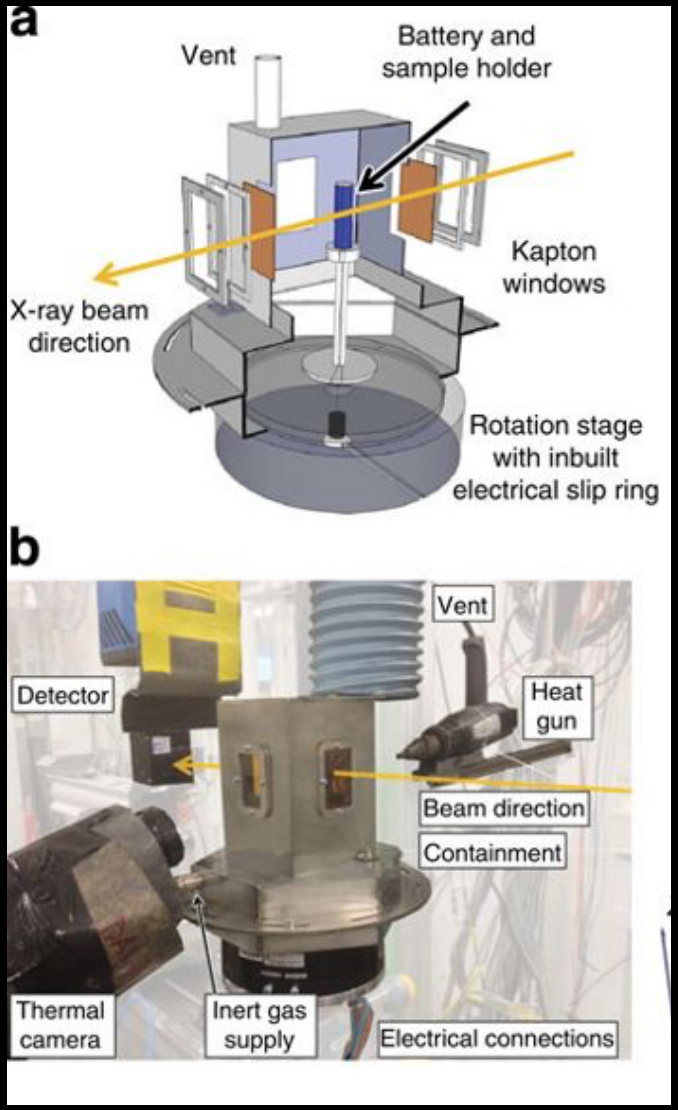
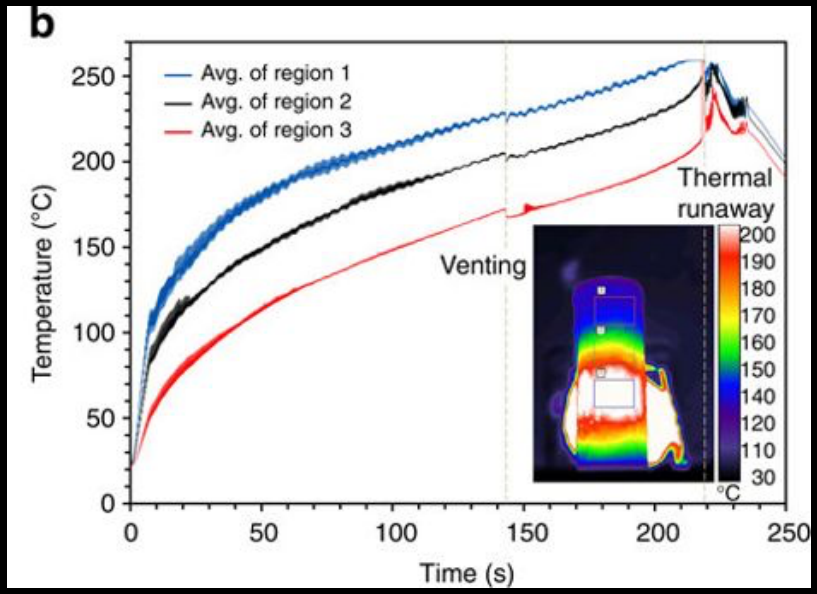
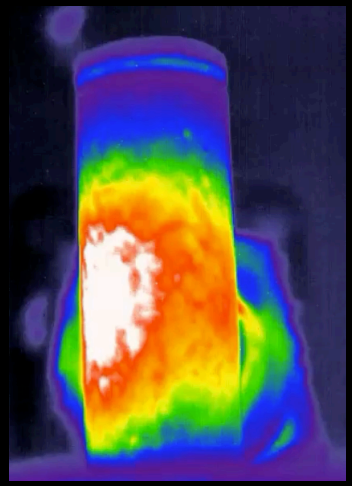
Thermal runaway

Thermal runaway

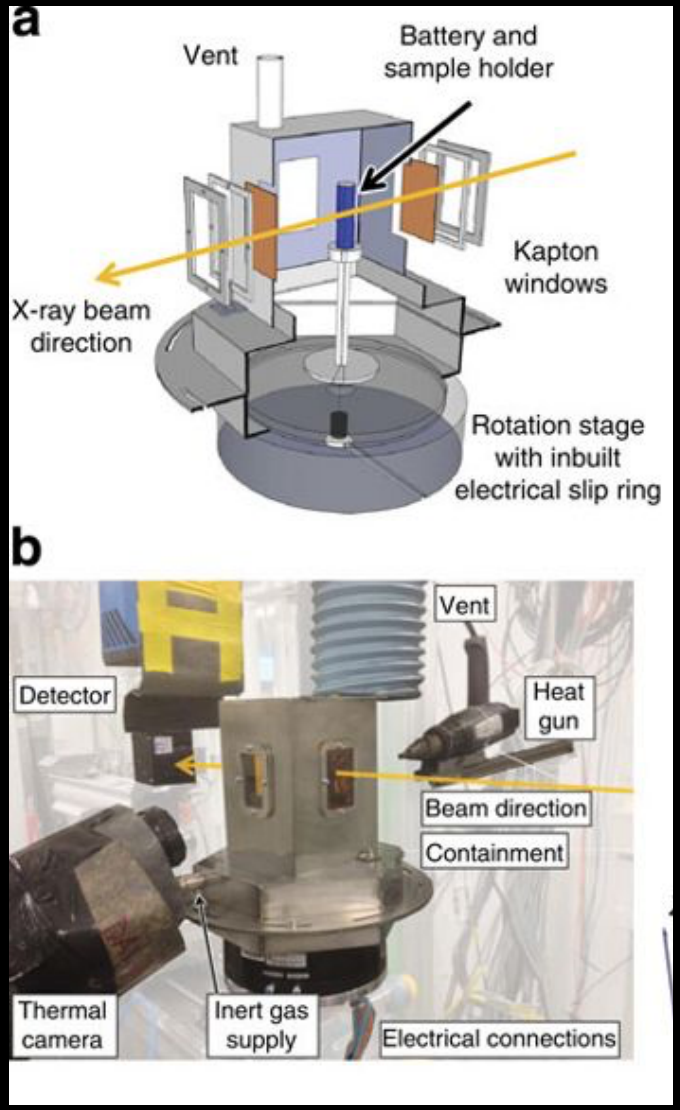


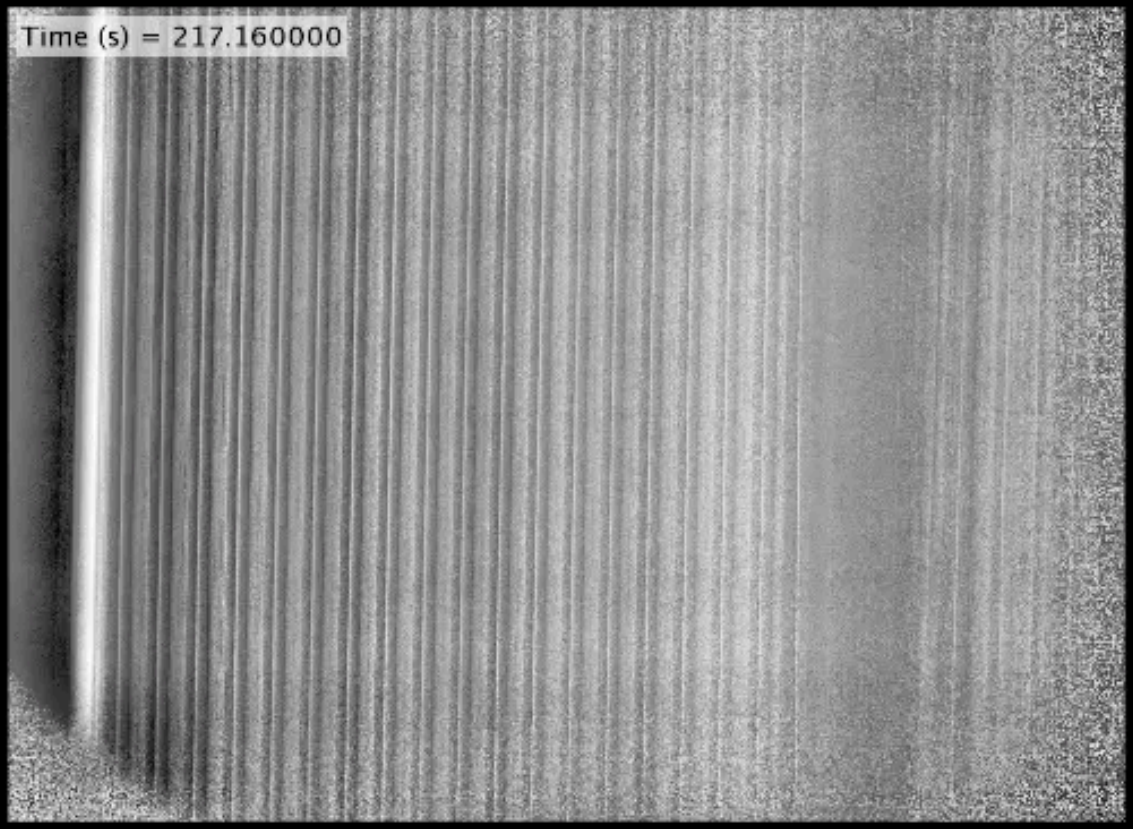




In-operando high-speed tomography of lithium-ion batteries during thermal runaway
Finegan, et al., 2015, Nature Comm., 6

Thermal runaway





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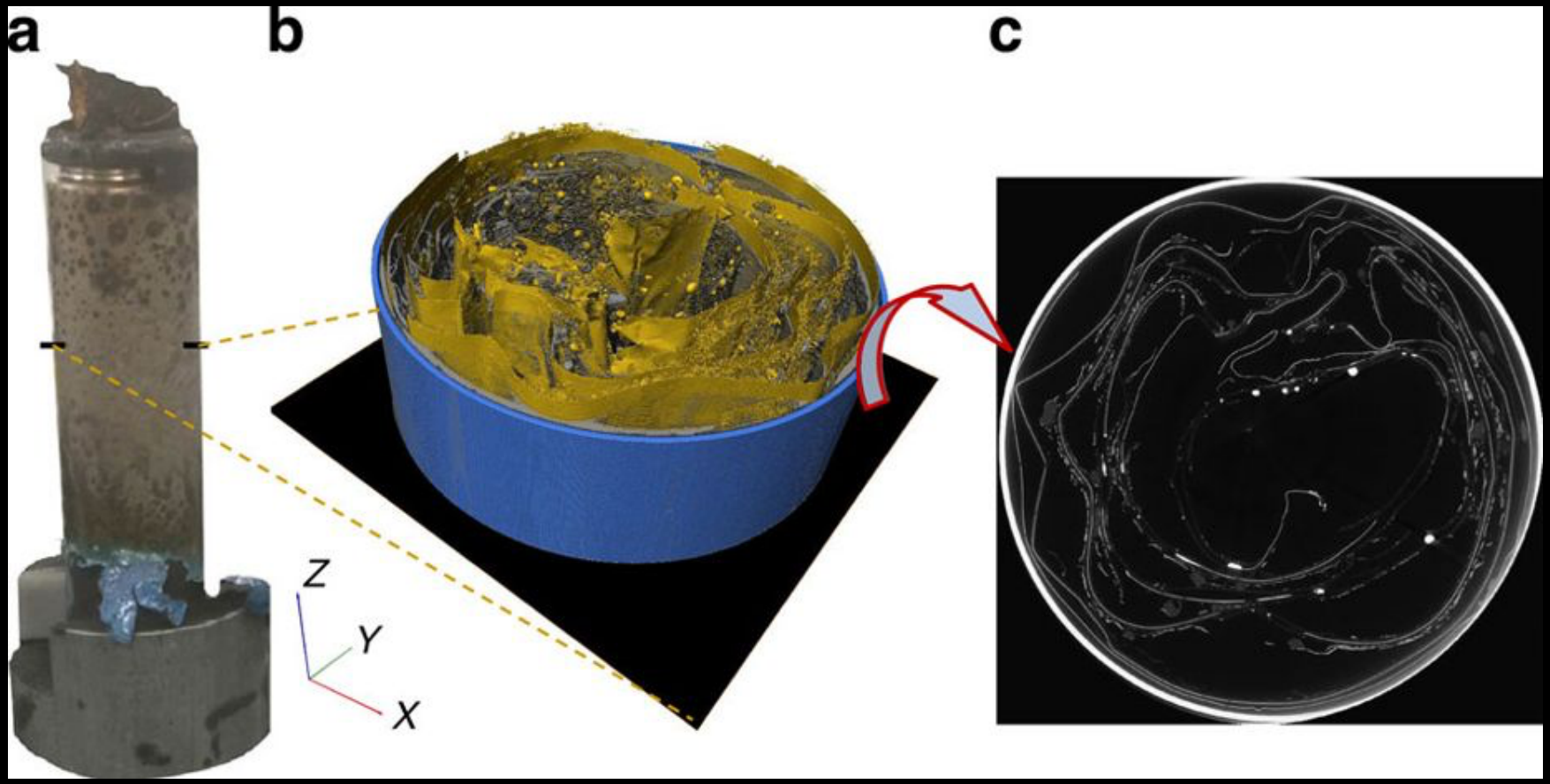

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4D IMAGING OF CHARGING AND DISCHARGING PROCESSES IN LITHIUM-ION BATTERIES

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