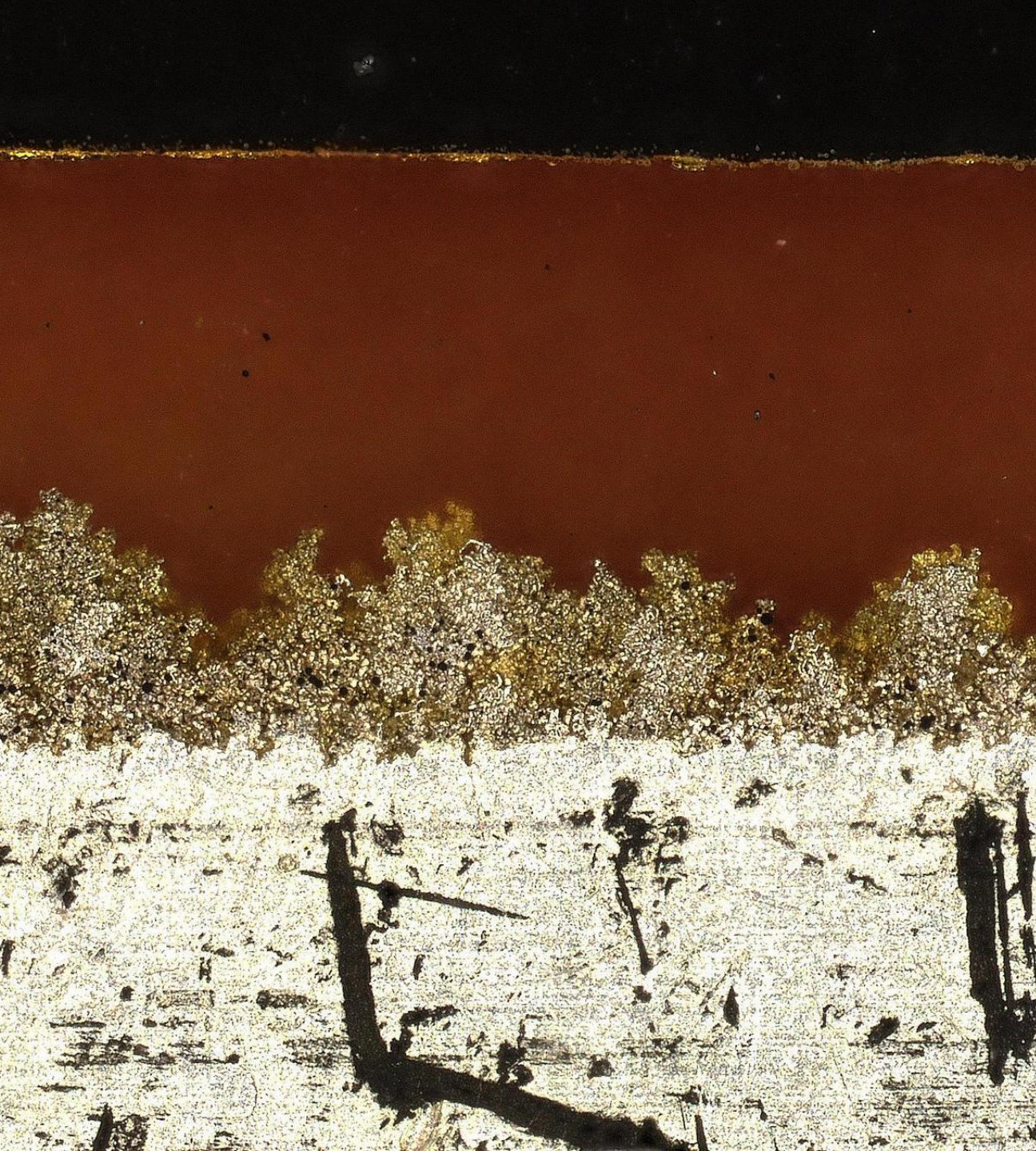




Benchmarking rate capability testing protocols for lithium-sulfur batteries using in-situ synchrotron X-ray tomography

Liam Bird^{1, 2}, Yiheng Shao^{1, 2}, James Le Houx^{2, 3}, James Robinson^{2, 4}, Paul Shearing^{1, 2}

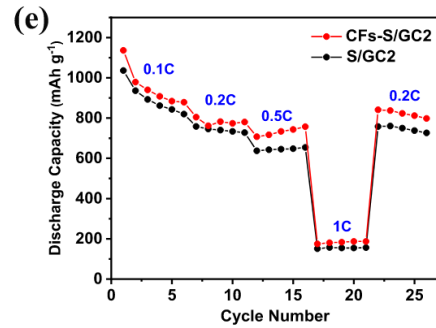
¹ ZERO Institute, University of Oxford

² Faraday Institution

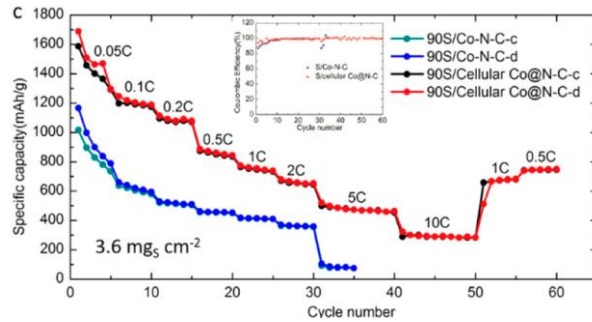
³ University of Greenwich

⁴ Advanced Propulsion Laboratory, University College London

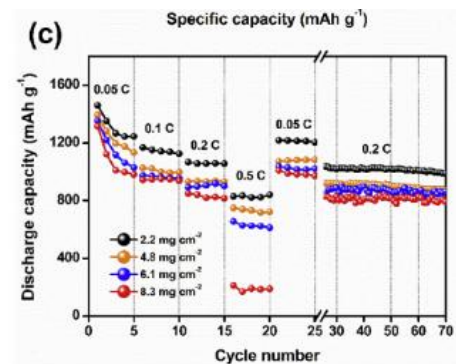
STARTING POINT: RATE CAPABILITY TESTING



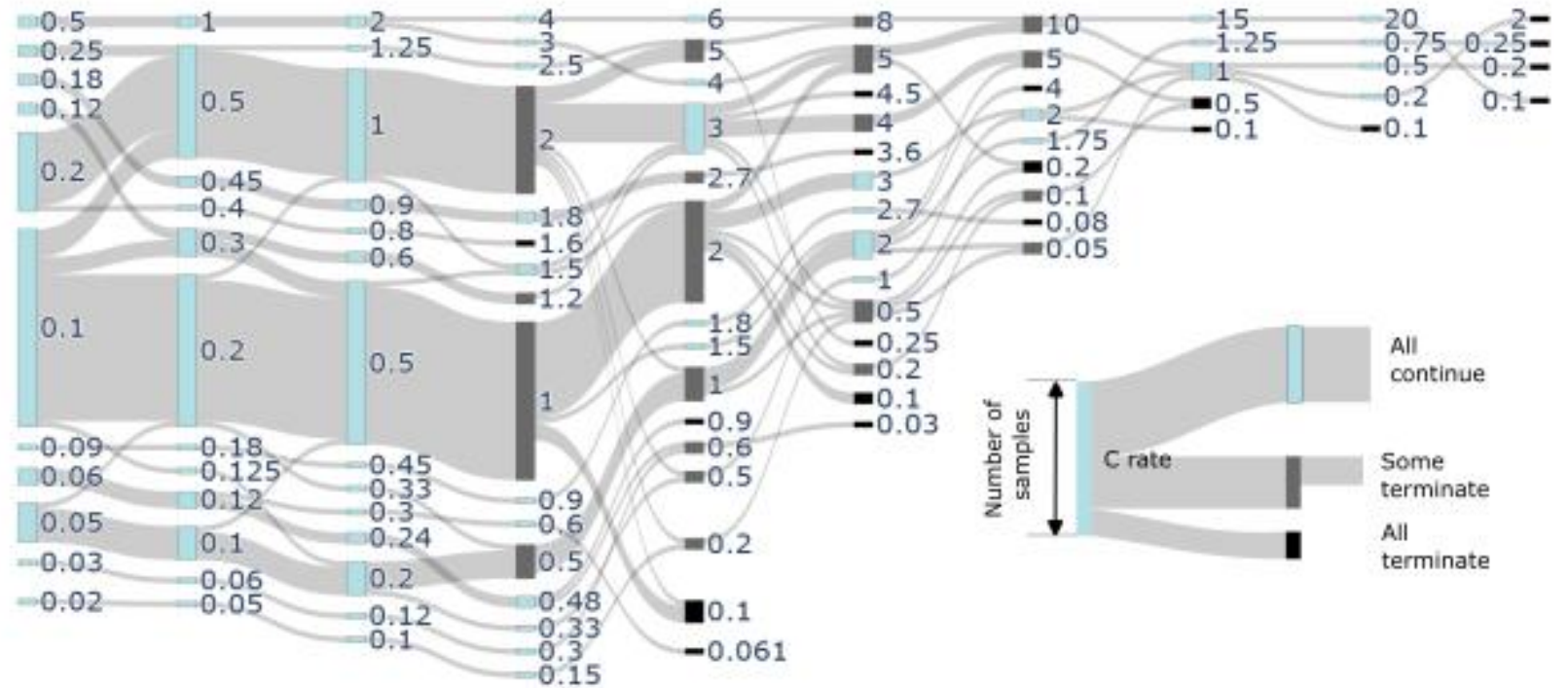
Yan, *et al.* Diam. Relat. Mater., 123, 108888 (2022)



Zhou, *et al.* Materials Letters 276 128243 (2020)

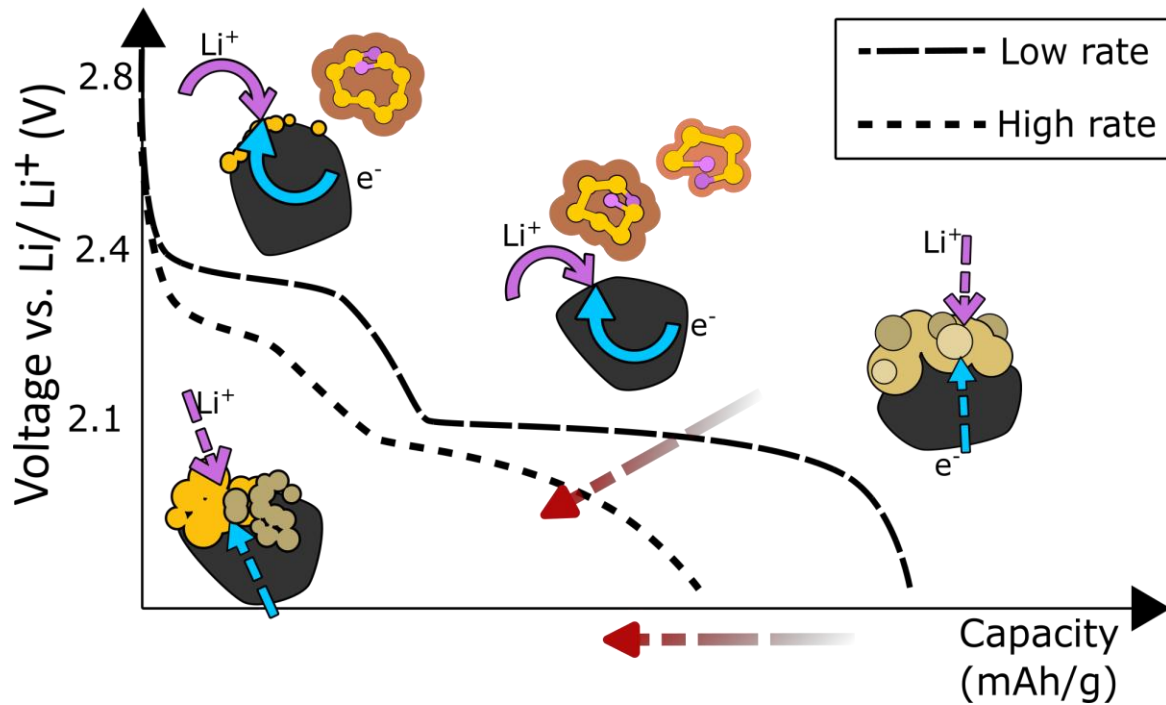


Zhao, *et al.* Carbon 128 138 (2018)



Data aggregated from 75 Li-S articles reporting rate capability testing
Bird, *et al.* ACS Materials Lett., 6(12), 5363, (2024)

EFFECTS OF RATE CAPABILITY TESTING



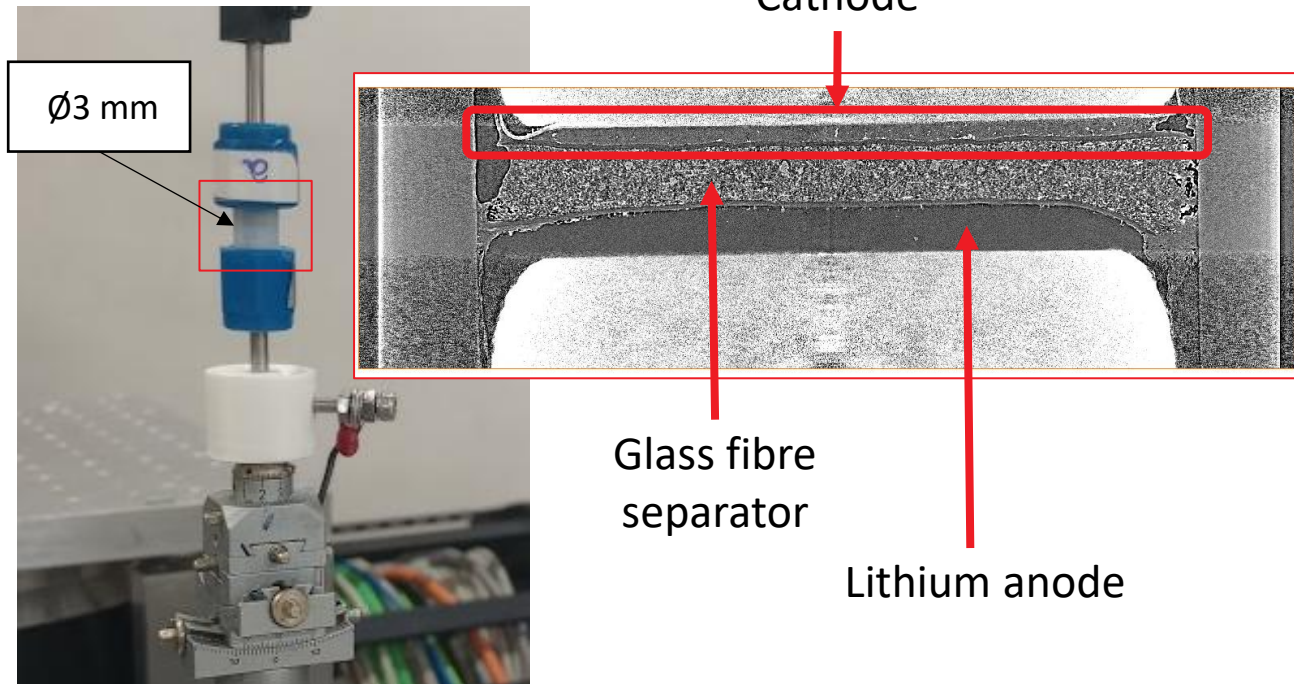
- Is capacity at high rate after incremental increase representative of long-term cycling at a single high rate?
 - Capacity
 - Overpotential

Influence of sulfur/ Li_2S morphology and distribution
- How does the (re)-distribution of sulfur affect capacity retention following rate increase?
- What can be learnt from capacity recovery when changing from higher to lower rate?
- Is the current density, or the increment between current densities, more informative?

IN-SITU SYNCHROTRON X-RAY TOMOGRAPHY

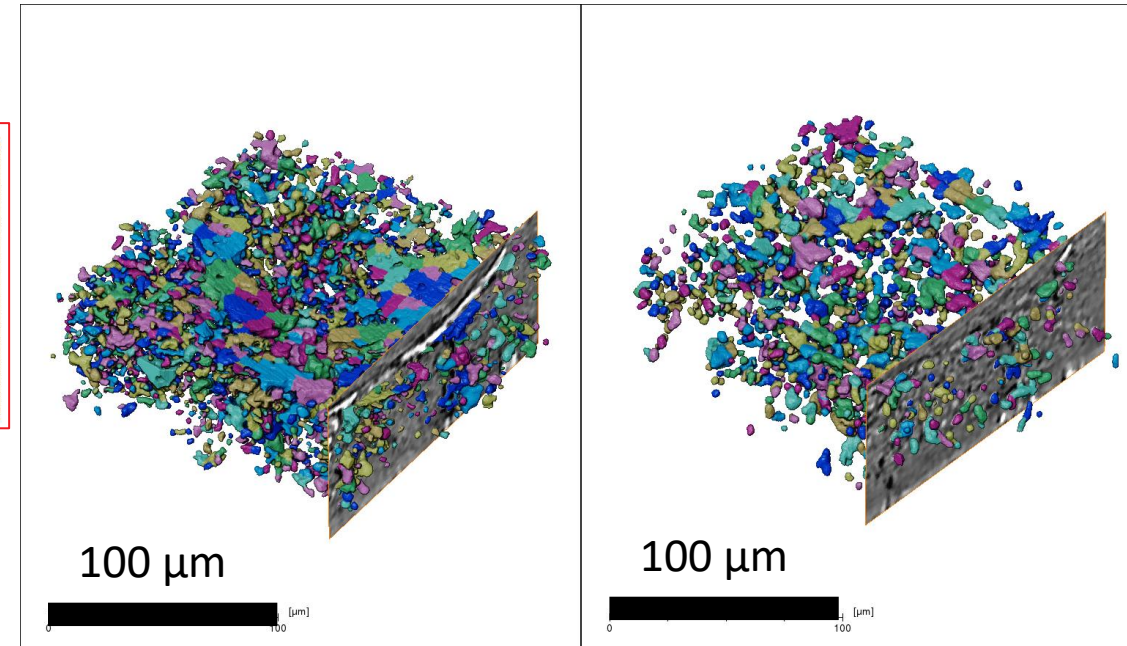


I-13 Diamond Light Source

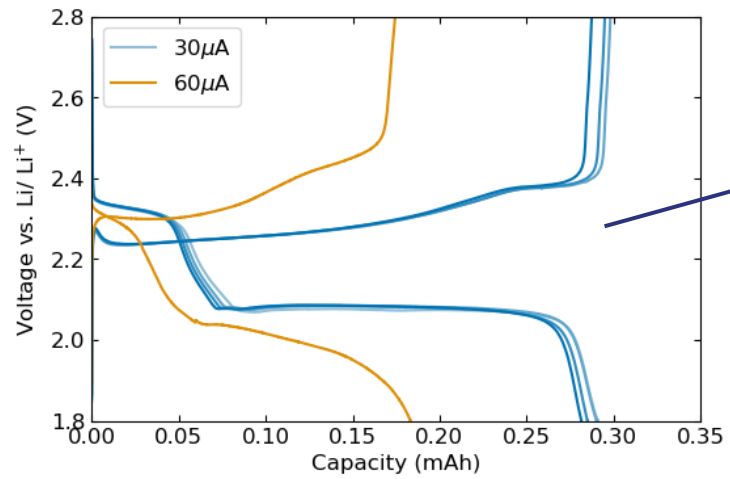


$5 \times 30 \mu\text{A} \dots$

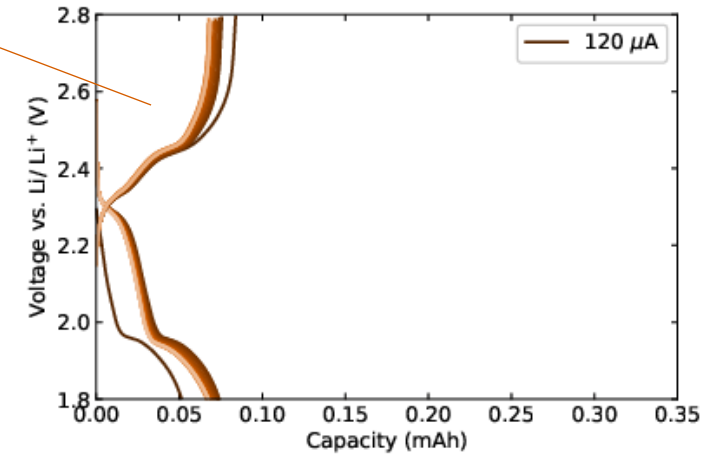
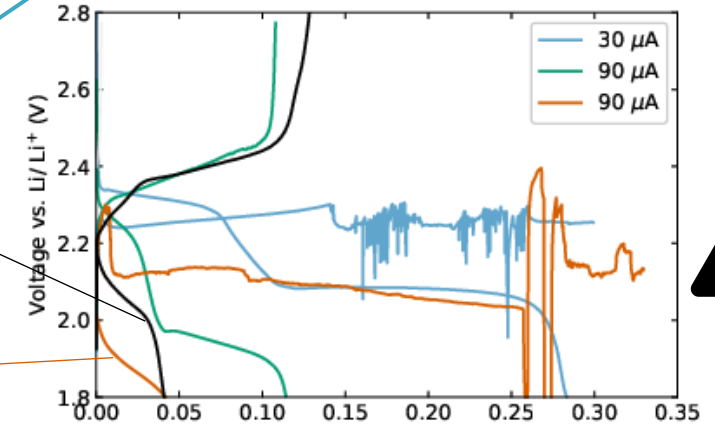
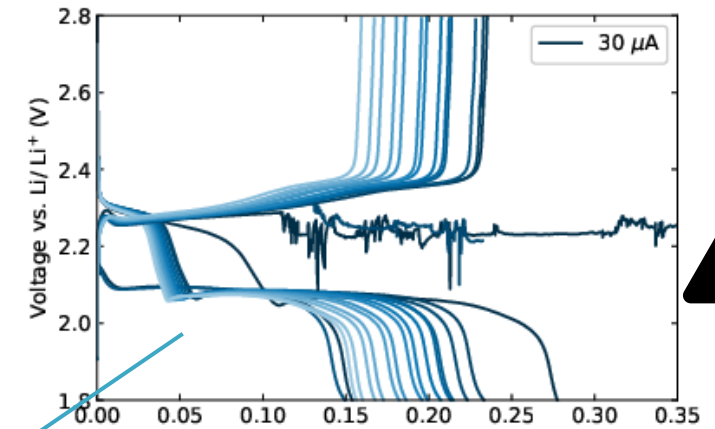
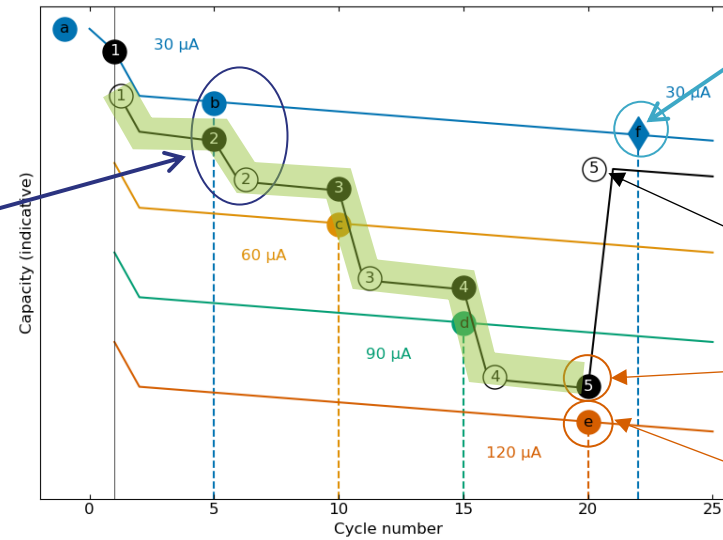
$\dots + 1 \times 60 \mu\text{A}$



ELECTROCHEMISTRY

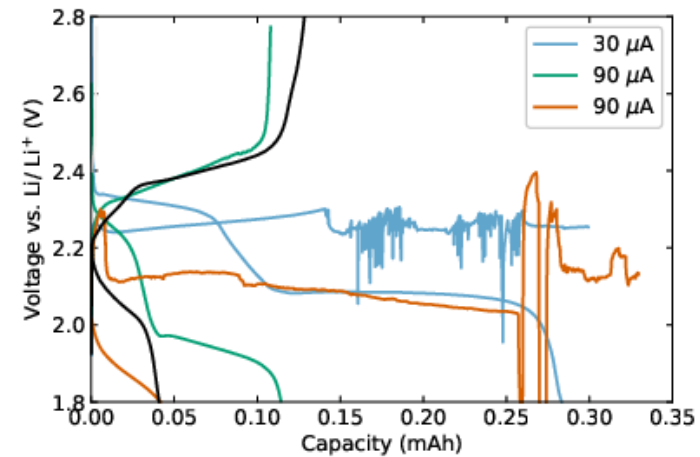
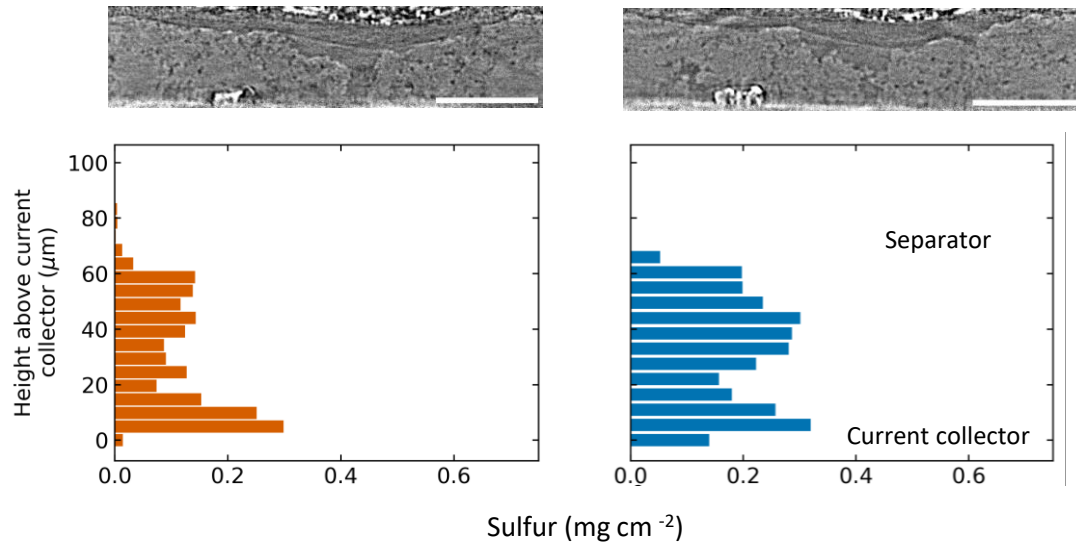
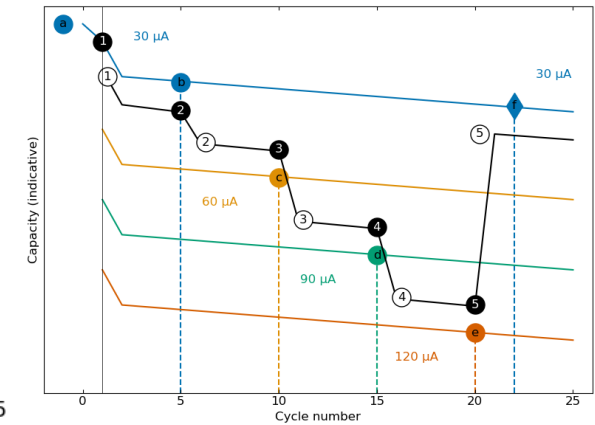
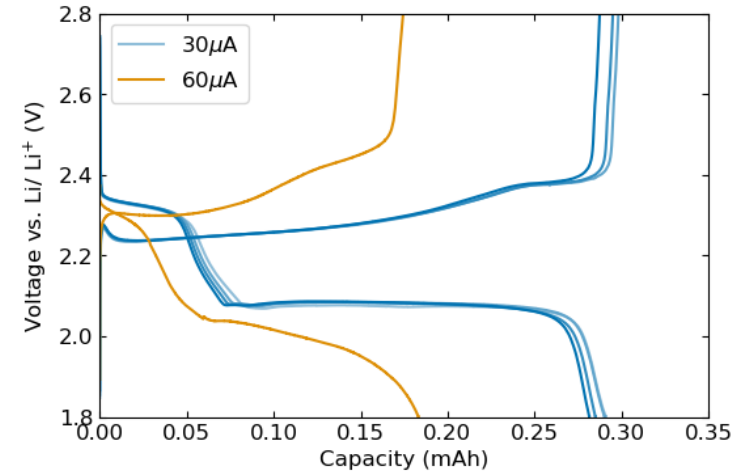
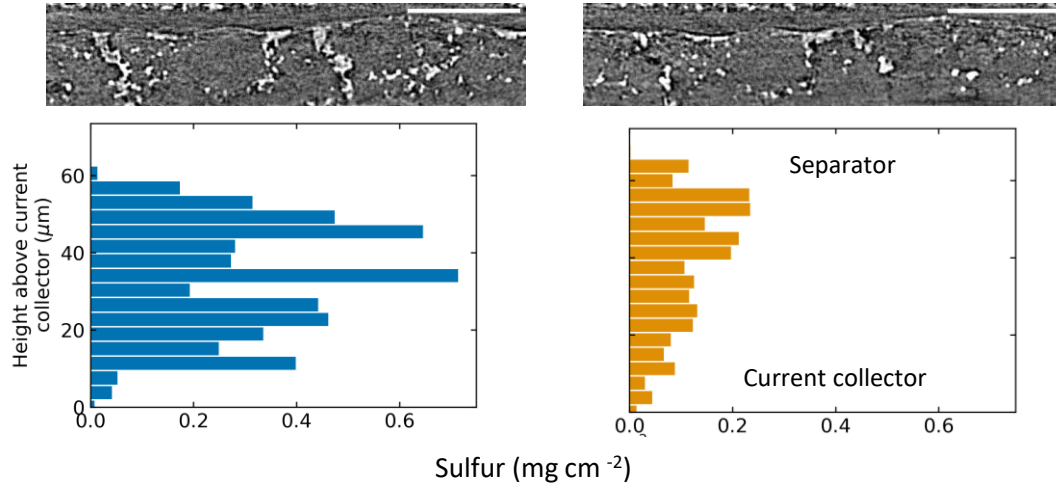


Pre-cycled in lab



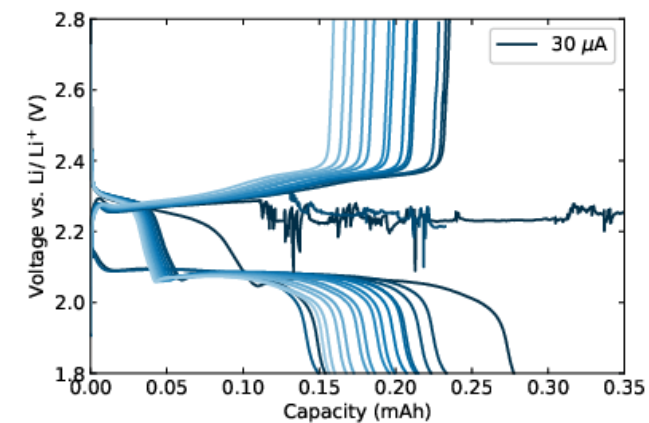
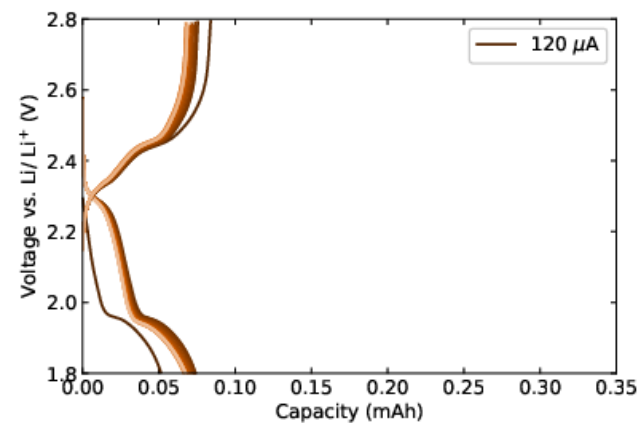
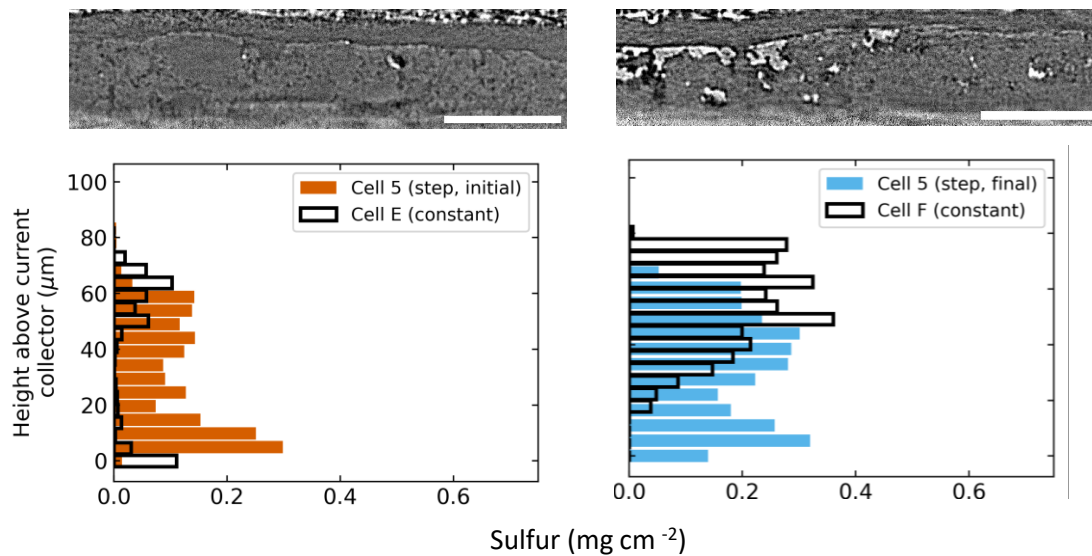
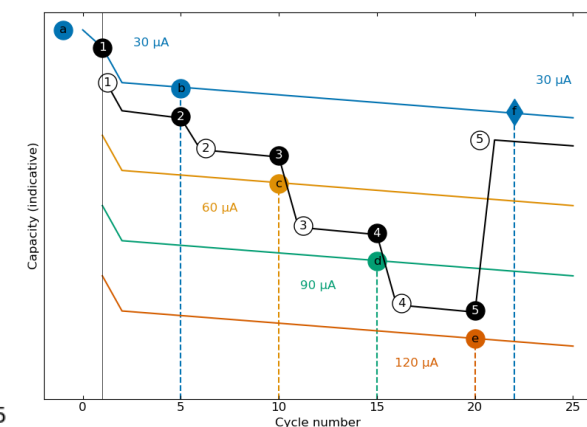
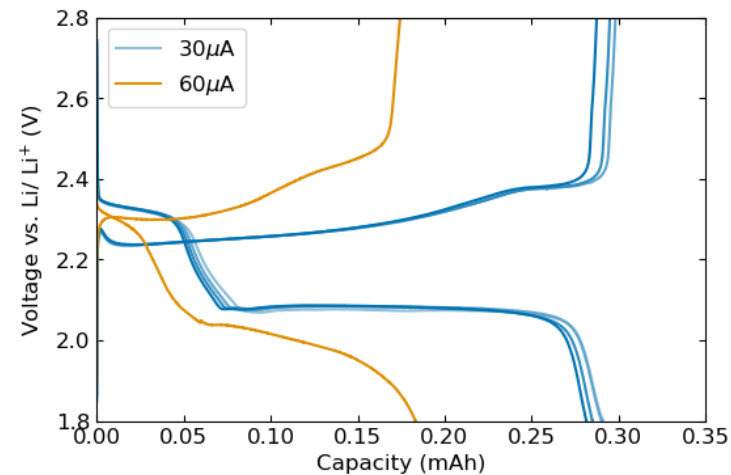
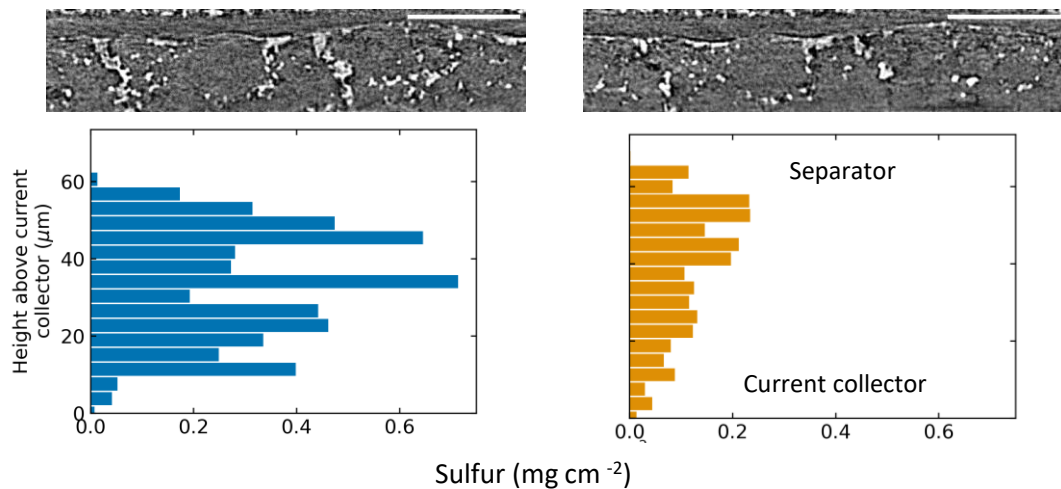
THROUGH-THICKNESS DISTRIBUTION: INCREMENTAL

All scalebars 100 μm

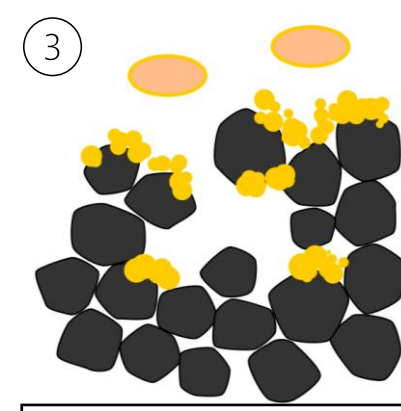
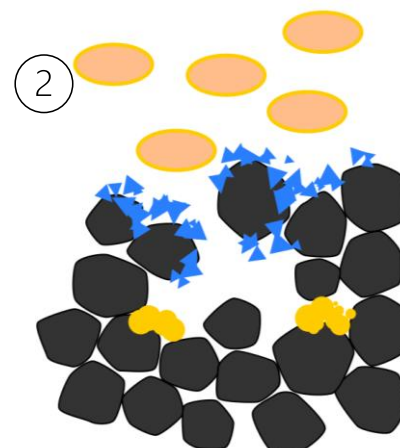
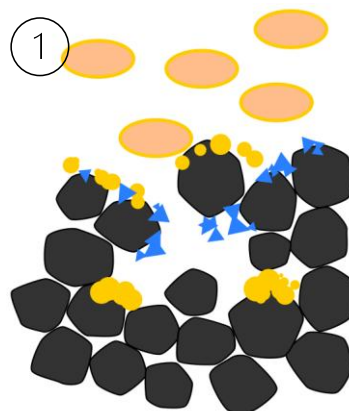
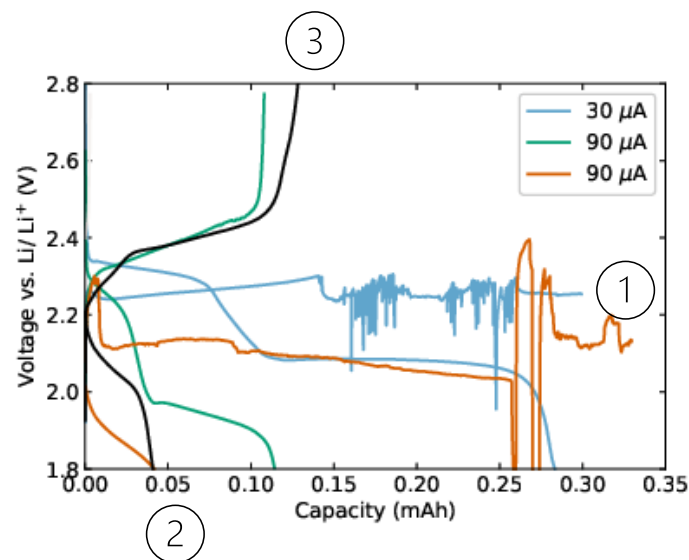
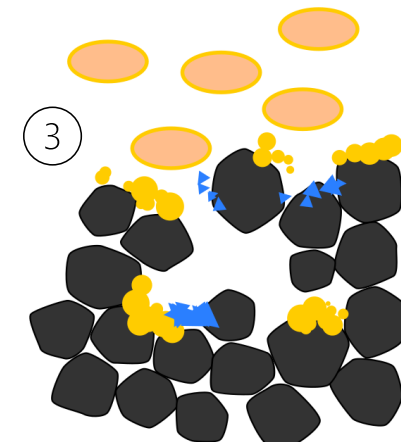
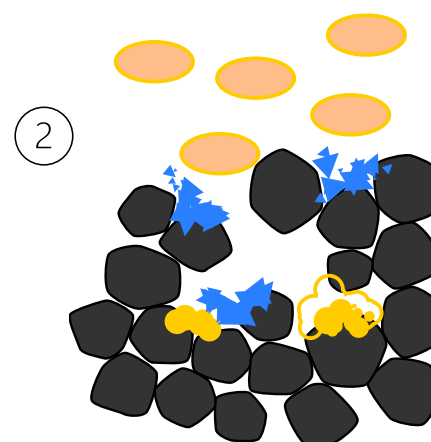
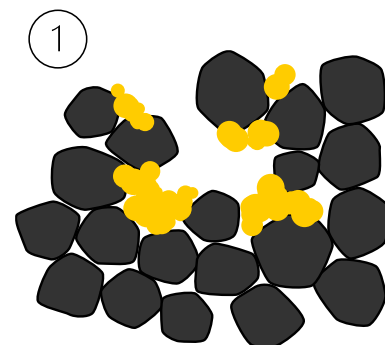
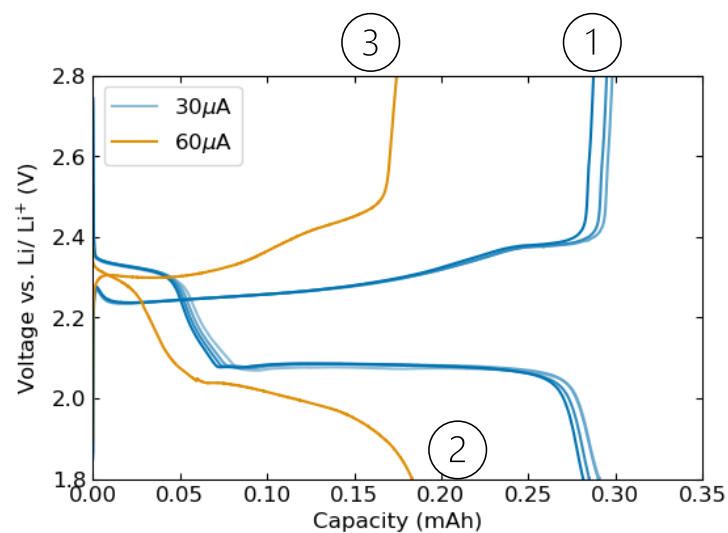


THROUGH-THICKNESS DISTRIBUTION: INCREMENTAL VS. CONSTANT

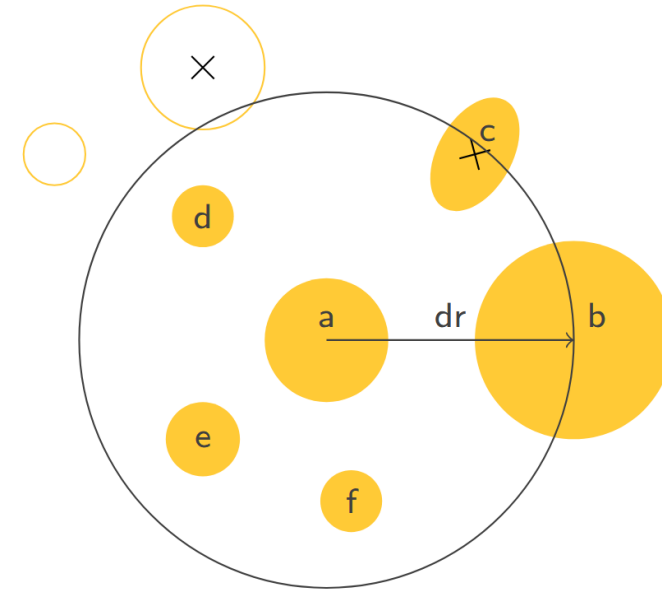
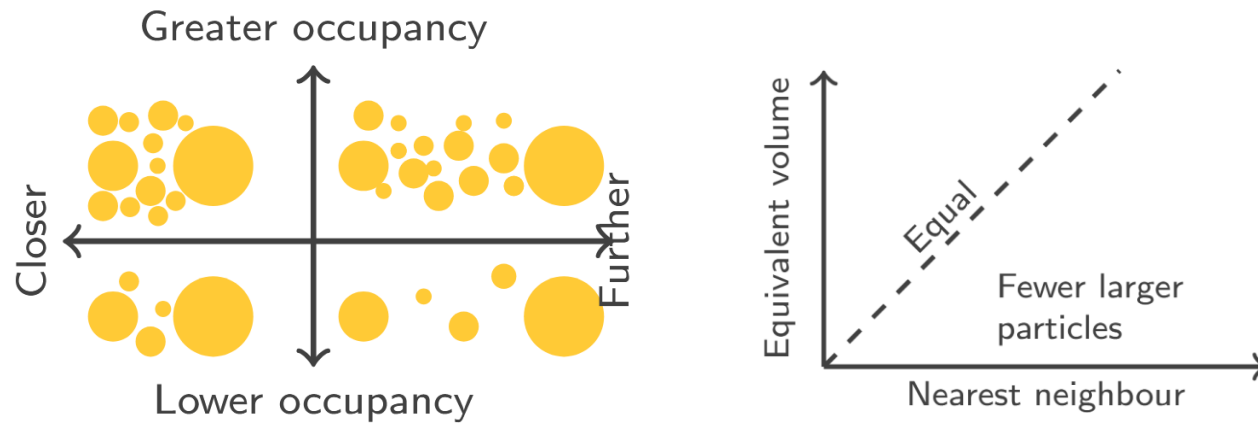
All scalebars 100 μm



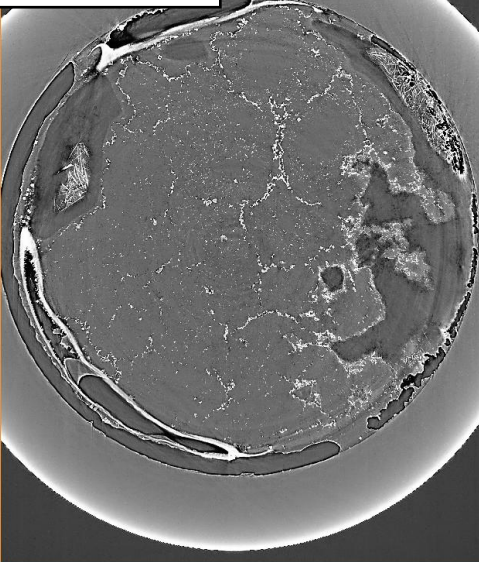
DISSOLUTION VS. PRECIPITATION



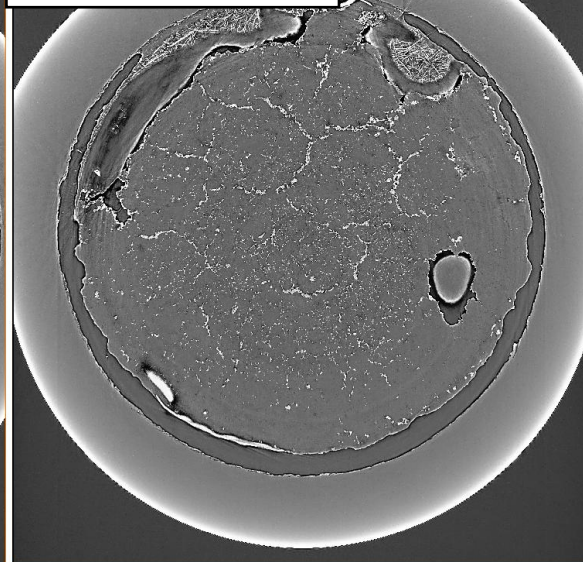
HOMOGENEITY METRIC



5 × 30 μA...



...+ 1 × 60 μA



dr: distance to nearest **larger** particle

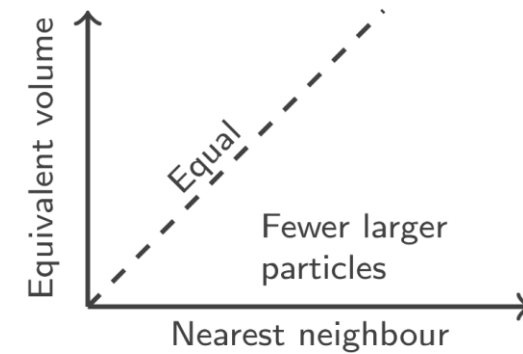
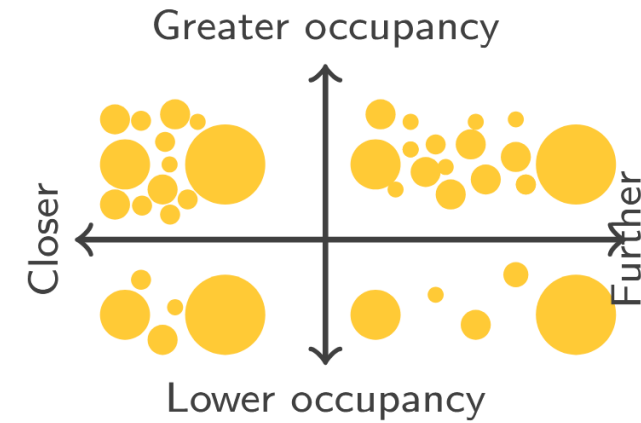
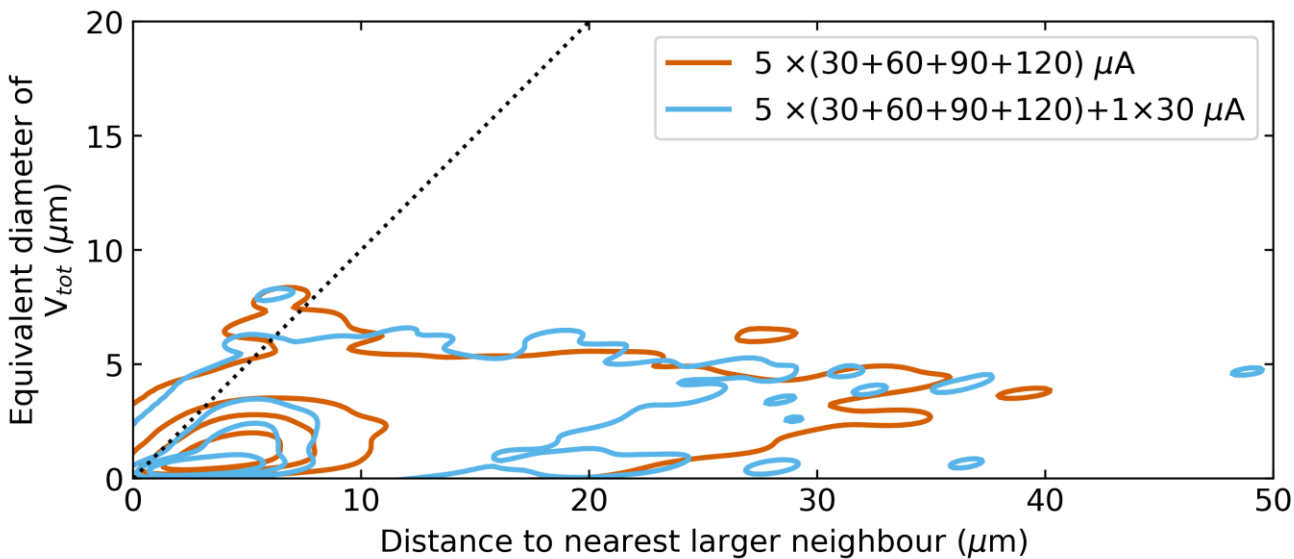
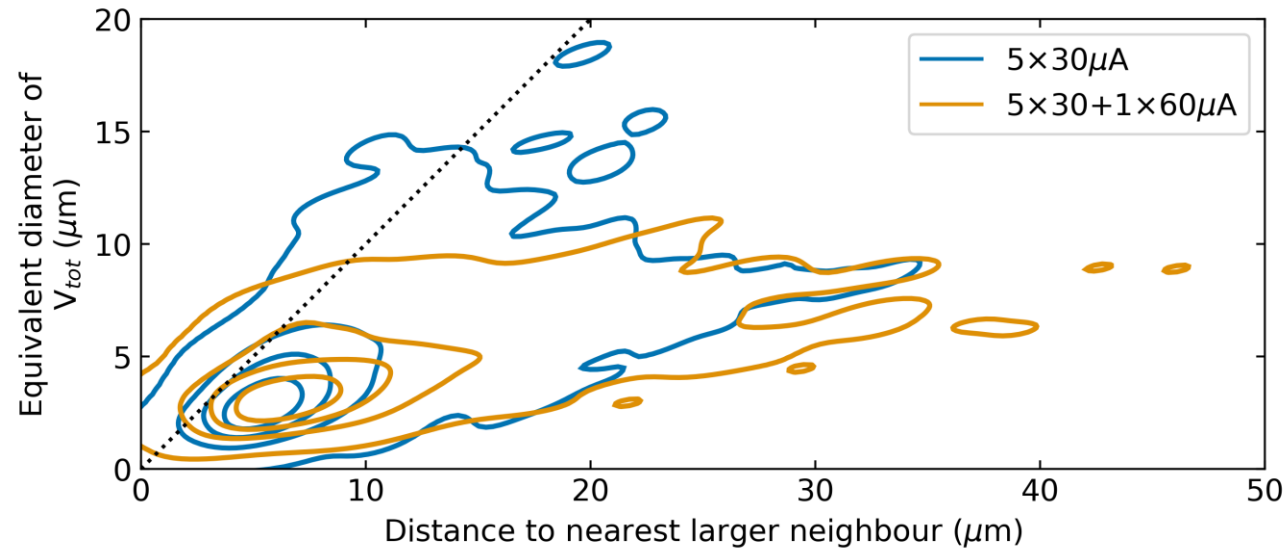
- Includes $a \rightarrow b$ (excludes $b \leftarrow a$)

V_{tot} : total volume of particles with centroid within dr of particle a

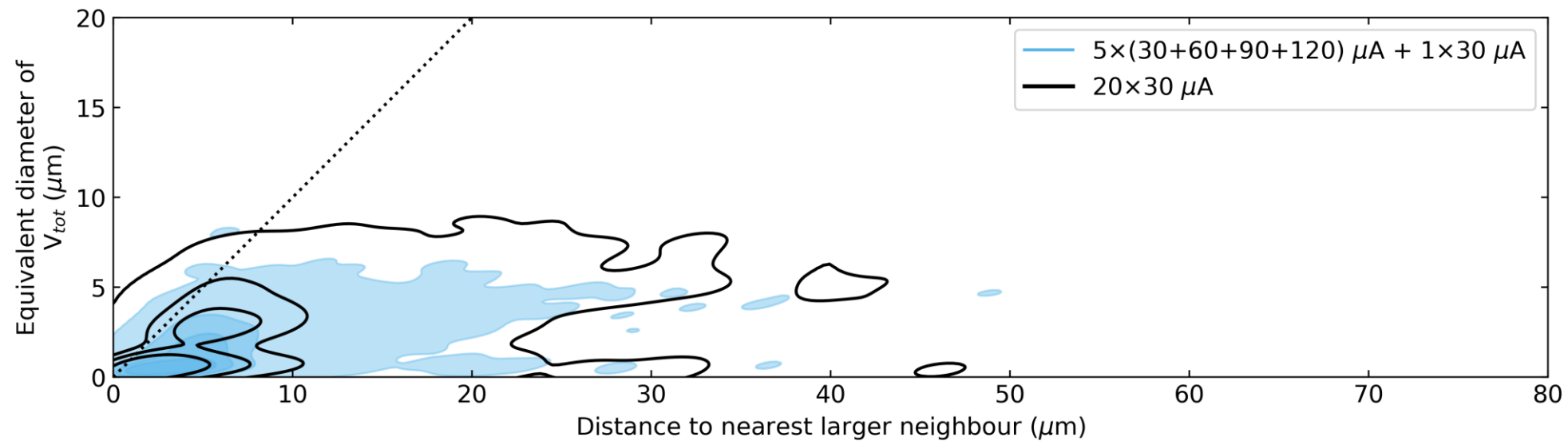
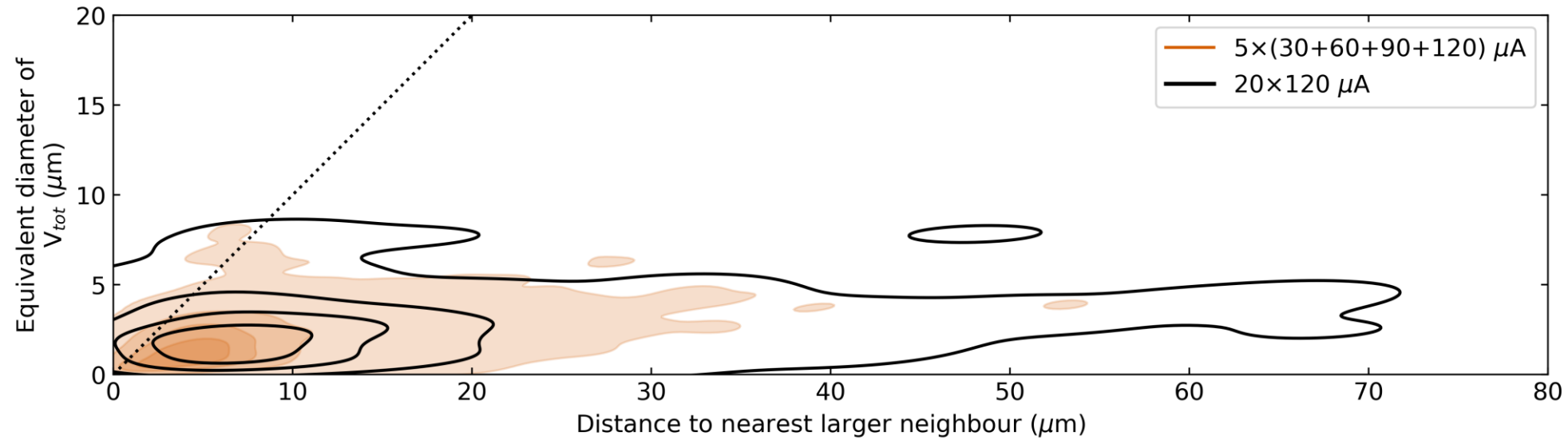
- Used as equivalent diameter:

$$\text{Eq Diam} = \left(\frac{V_{\text{tot}}}{4\pi} \right)^{1/3}$$

HOMOGENEITY METRIC: INCREMENTAL



HOMOGENEITY METRIC: INCREMENTAL VS. CONSTANT



NEXT STEPS

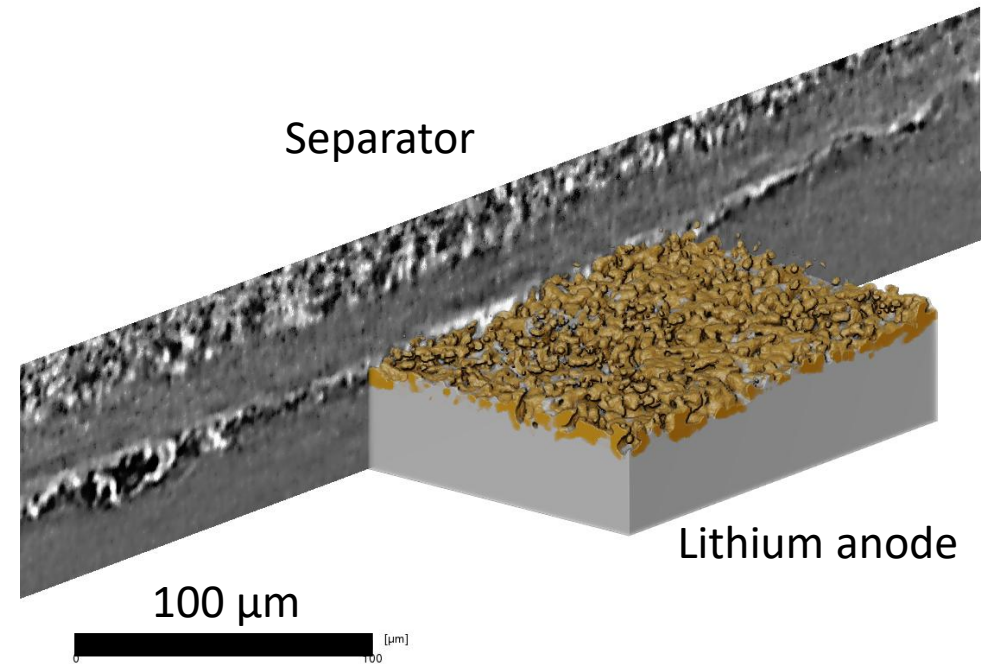


XCT data analysis

- Data series available within single discharge cycle: aim to track change in homogeneity as a function of SOC
- Overcharge and anode deposition:
 - Cells available with single cycle
 - Compare deposit volume with Coulomb counting
- Low magnification data available: apply heterogeneity factor at electrode level
- Tortuosity: change in tortuosity following deposition/dissolution

Electrochemistry

- GITT: Diffusion coefficient as function of SOC



META-ANALYSIS DATABASE



github.com/LiamBird/meta-characterisation

```
In [4]: meta = MetaChecklistTab(test=False)
        meta.tab
```

Out[4]:

Save reload

Article details

Reporting checklist

Characterisation

Save values

No values saved this session

Cell format

Electrode processing

Galvanostatic

Temperature reported during galvanostatic/ CV/ EIS?

☒ Not reported
 ☐ Constant - non room temp
 ☐ Room temp
 ☐ Under test

How is the theoretical capacity calculated?

☐ Active material
 ☐ Areal/ mass
 ☐ Cell
 ☐ Slow initial/ formation

Is a pre-activation/ formation cycle reported?

Rate

No. cycles

Other

Galvanostatic cycling summary (use overall max and min values for all samples)

Voltage cutoffs

Min

Max

☐ Multiple ranges reported

C Rates

Min

Max

Current density

Min

Max

☒ mA/g
 ☐ mA/cm²

CC/CV (constant voltage) step reported?

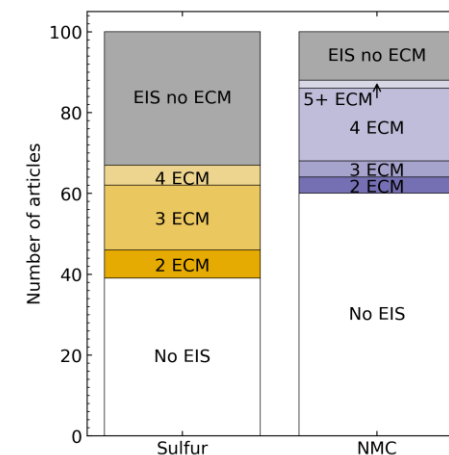
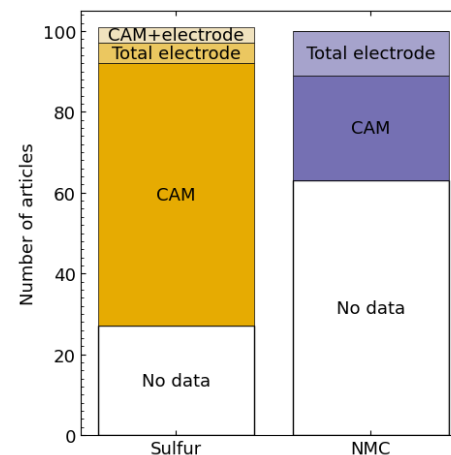
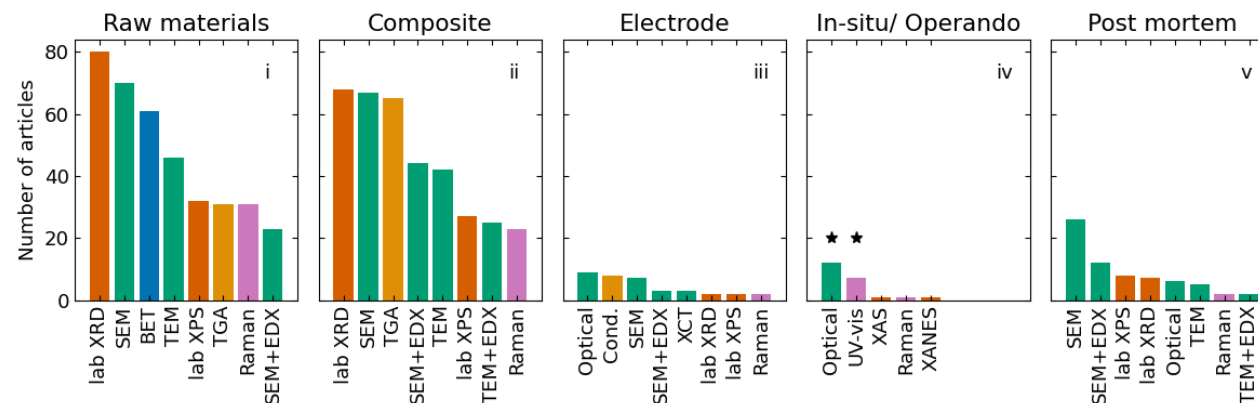
☐ CV during charge
 ☐ CV during discharge

Multiple cells shown with statistics?

☐ Yes (in article)
 ☐ Yes (In SI)
 ☒ No

Cyclic voltammetry (CV)

Electrochem Impedance Spectroscopy (EIS)



Submitted: Discover Electrochemistry (Special Issue "[Raising Standards in the Characterisation of Electrochemical Energy Storage and Conversion Devices](#)".)

ACKNOWLEDGEMENTS



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Science & Technology Facilities Council
ISIS Neutron and Muon Source

Isabel Antony
Dr. James Le Houx



Dr. Shashi Marathe
Dr. Zihan Song
Dr. Kaz Wanelik



Science and
Technology
Facilities Council