‘Seeing inside’ energy materials – understanding why and how materials in electrochemical devices fail

Dr Jen Johnstone-Hack

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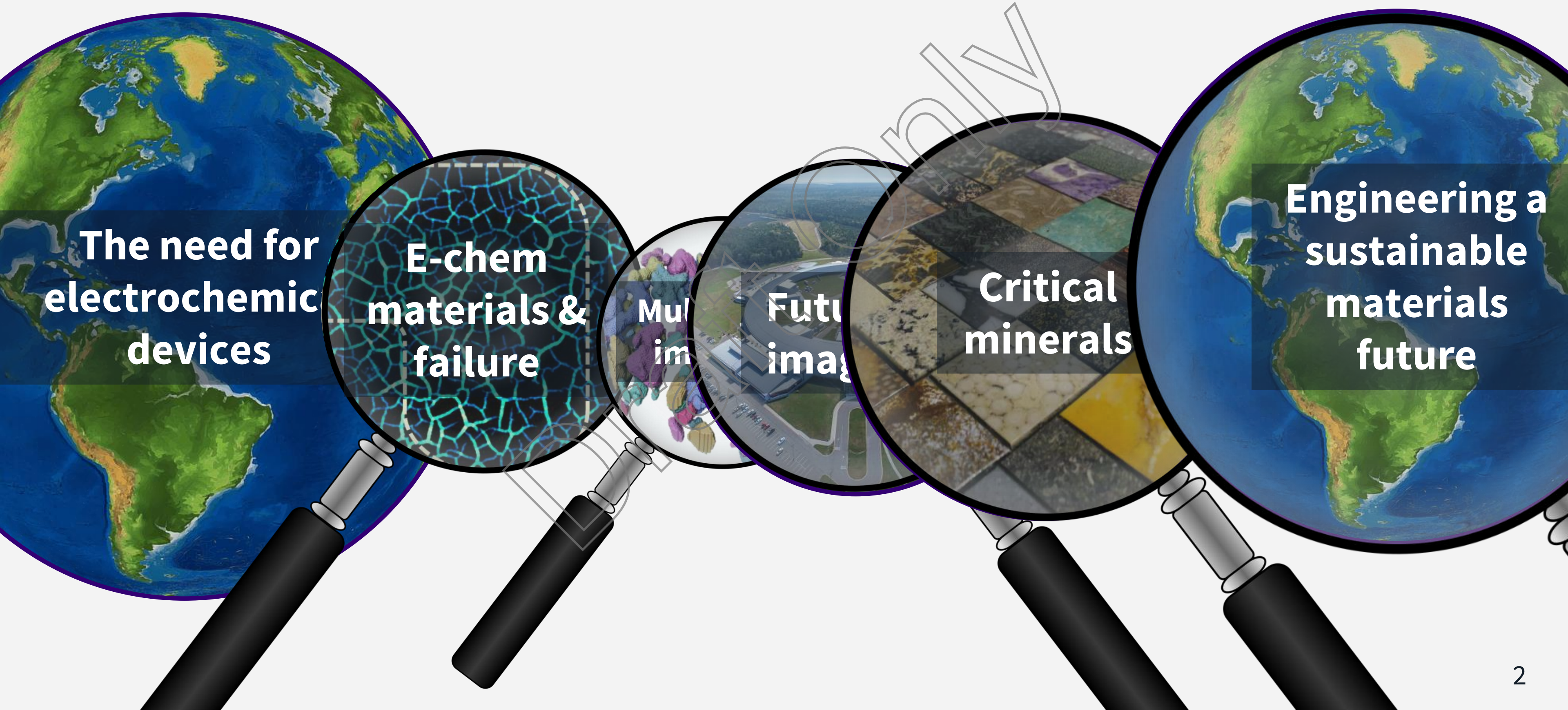
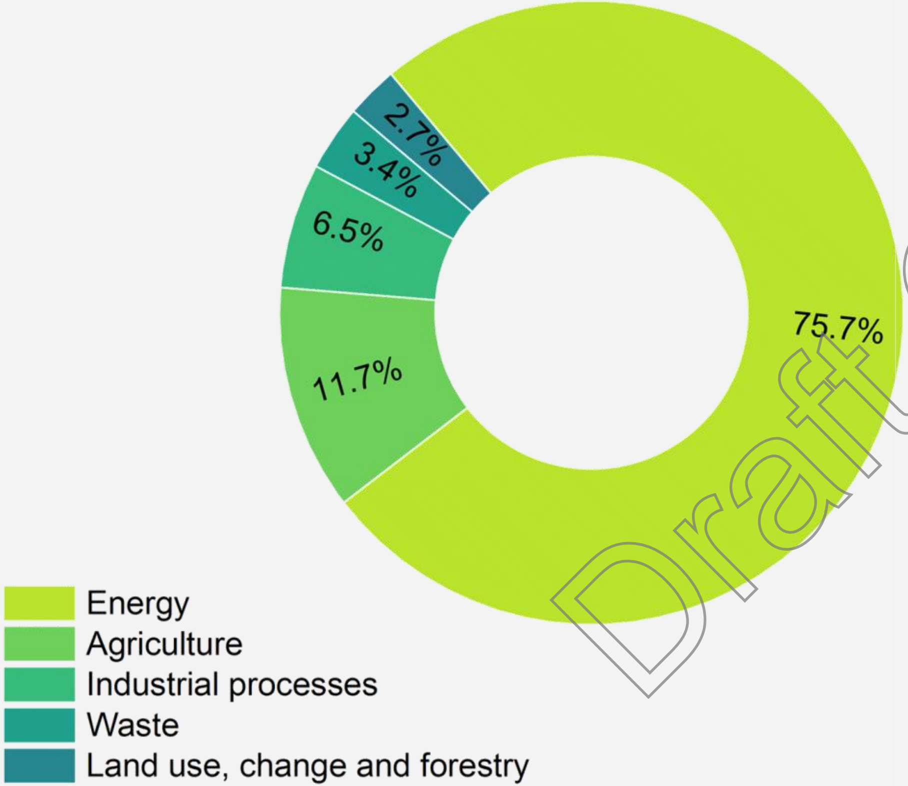
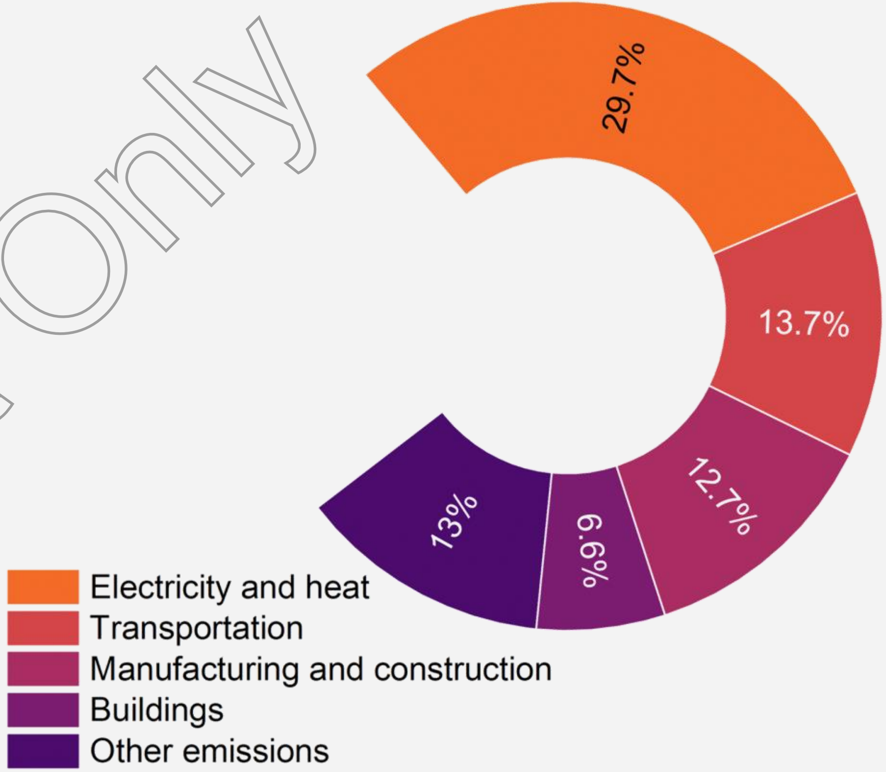
Talk outline



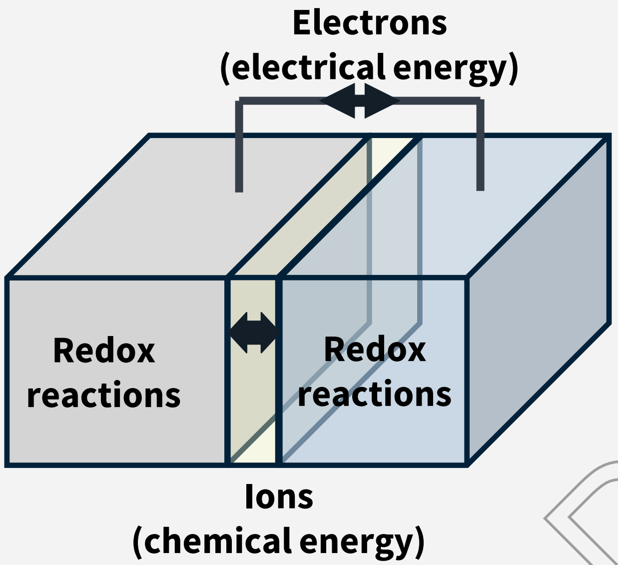
Energy sector contributes 76% of global GHG emissions

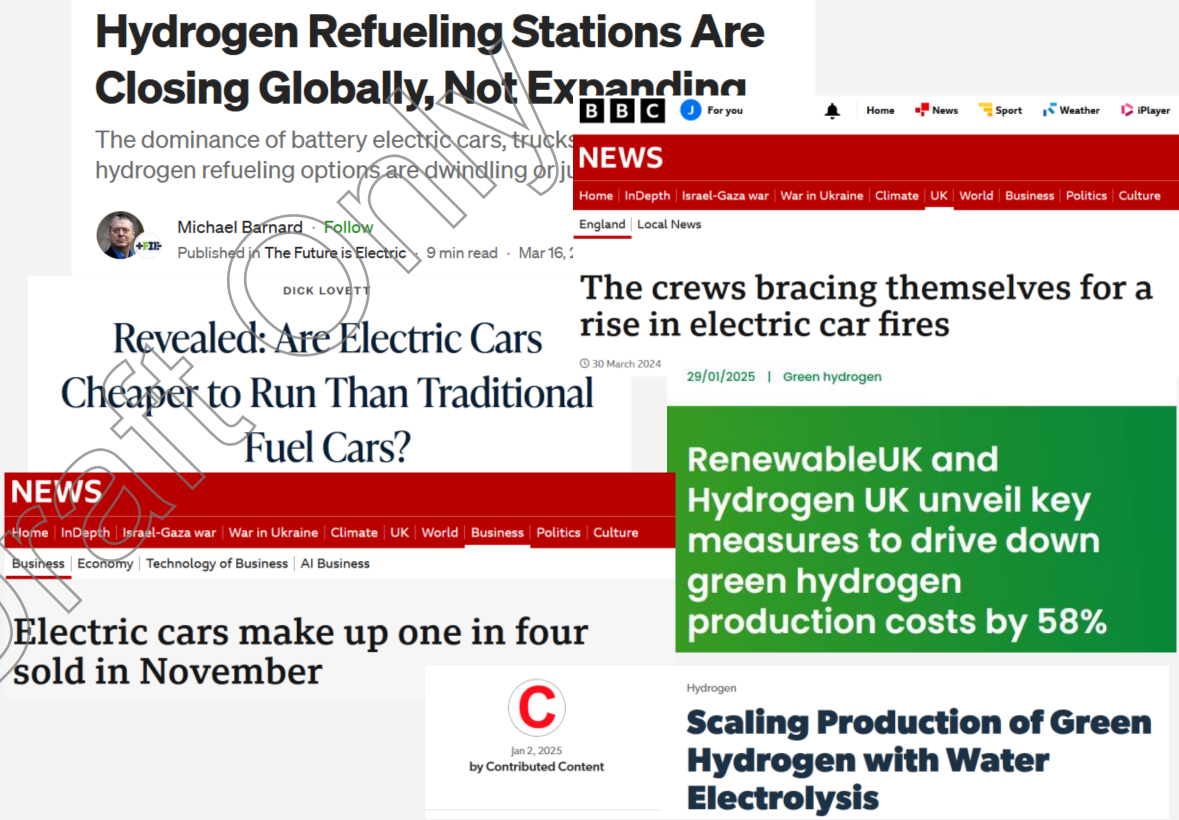




Data source: Climate watch, World GHG Emissions by Sector 2021

Electrochemical technologies have a key role to play…




Hydrogen Refueling Stations Are Closing Globally, Not Expanding

The dominance of battery electric cars, trucks
hydrogen refueling options are dwindling or ju

Michael Barnard · Follow
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DICK LOVETT

Revealed: Are Electric Cars Cheaper to Run Than Traditional Fuel Cars?

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30 March 2024

29/01/2025 | Green hydrogen

RenewableUK and Hydrogen UK unveil key measures to drive down green hydrogen production costs by 58%

NEWS

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Electric cars make up one in four sold in November

C

Jan 2, 2025
by Contributed Content

Hydrogen

Scaling Production of Green Hydrogen with Water Electrolysis

…but getting enough in the right places is hard.

The ‘electrochemical’ (r)evolution?

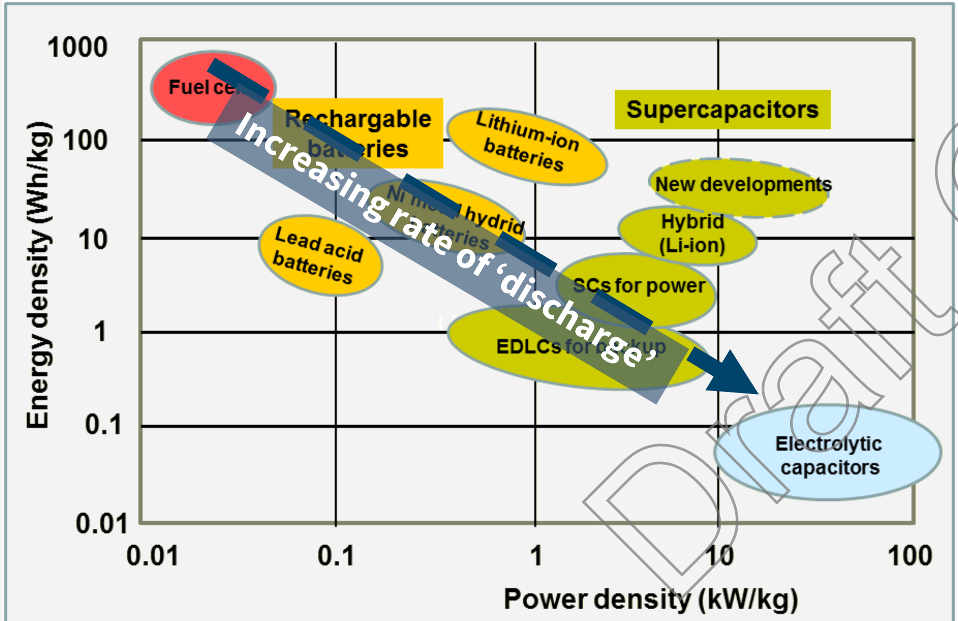
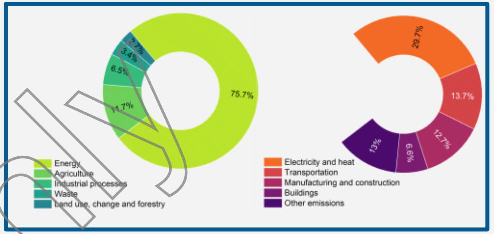
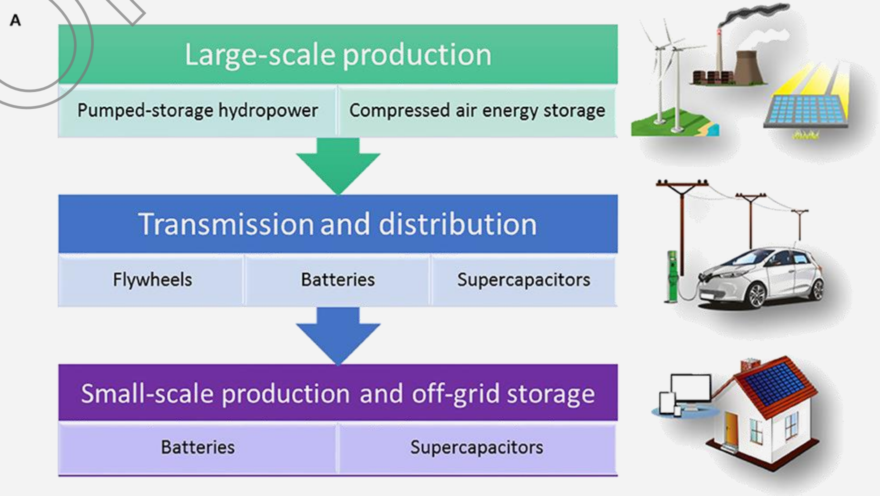


Image: Elcap/CC0 1.0

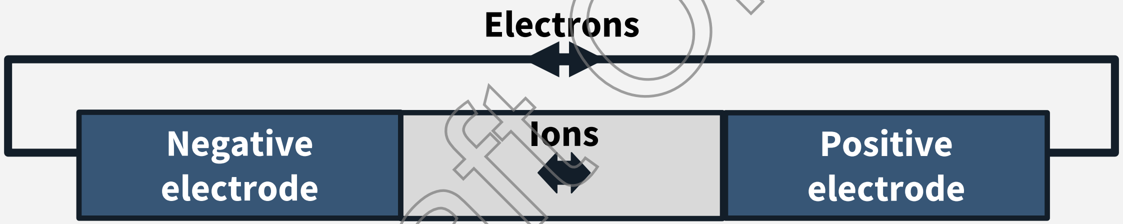




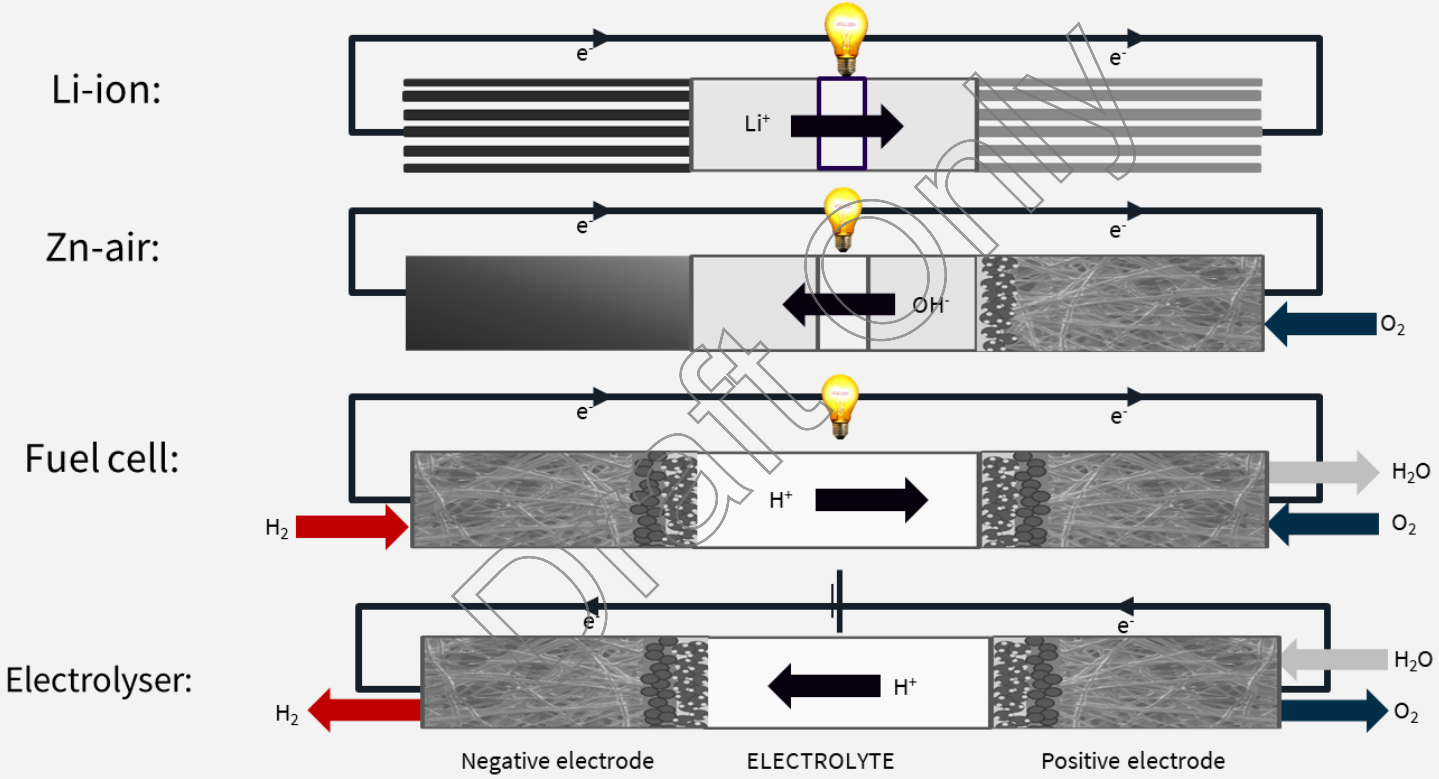
Adapted from: J. Castro-Gutiérrez, A. Celzard and V. Fierro, *Front. Mater.*, 7 (2020) 217, DOI: 10.3389/fmats.2020.00217

We can tune the materials properties of the electrochemical technology to the target application.

Change the materials and change the technology



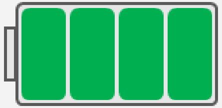
Change the materials and change the technology



Adapted from: T. Heenan, C. Tan, J. Hack et al., Materials Today, 2019, 31, 69-85

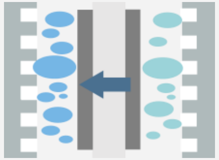
Same materials challenges for different technologies

'Battery people'



Diffusion
Conductivity
Resistance and impedance
Kinetics
Thermodynamics
Chemical compatibility
Degradation
Multiscale time and length processes

'Electrolyser/ fuel cell people'

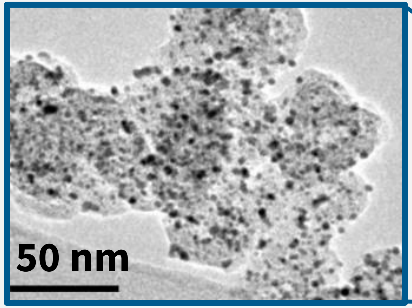


Two key challenges for characterisation with imaging…

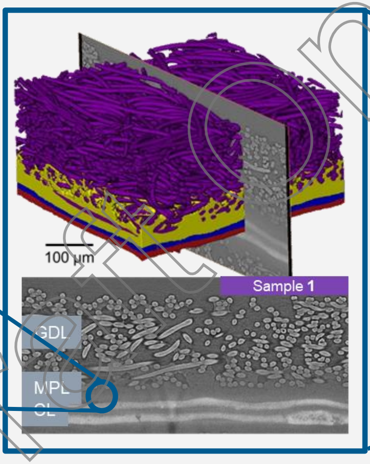
1. They are ‘multiscale’, spanning length and time scales

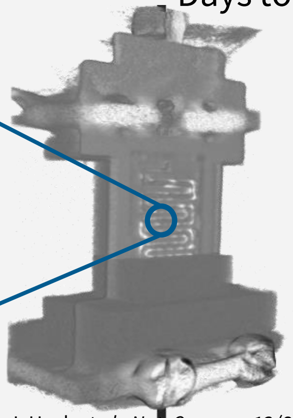
Fractions of seconds

Days to years



J. Hack *et al.*, *Electrochim. Acta,* 352 (2020) 136464, https://doi.org/10.1016/j.electacta.2020.136464





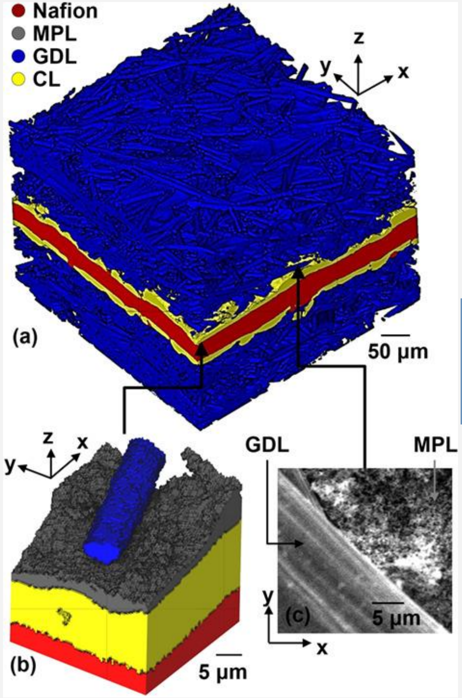
R. Ziesche, J. Hack *et al.*, *Nat. Comms.*, *13* (2022), https://doi.org/10.1038/s41467-022-29313-5

‘Nanoscale’
Atoms and particles

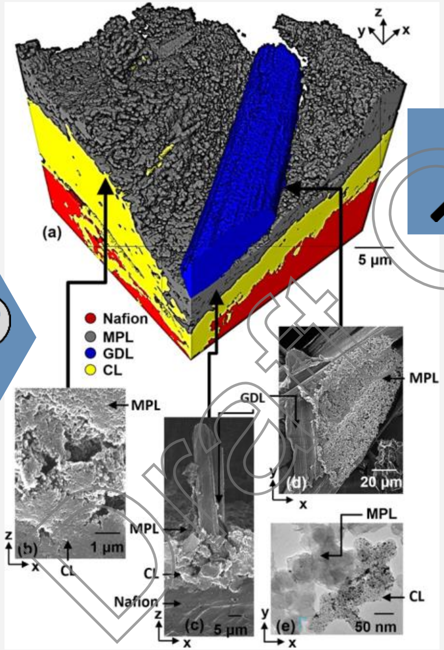
‘Microscale’
Electrodes and layers

‘Macroscale’
Devices

Multiscale imaging can help us access different features

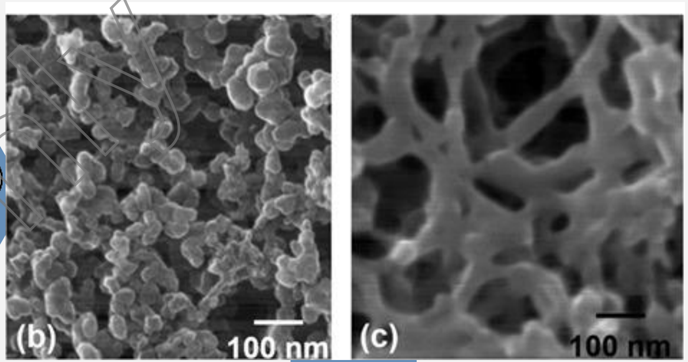


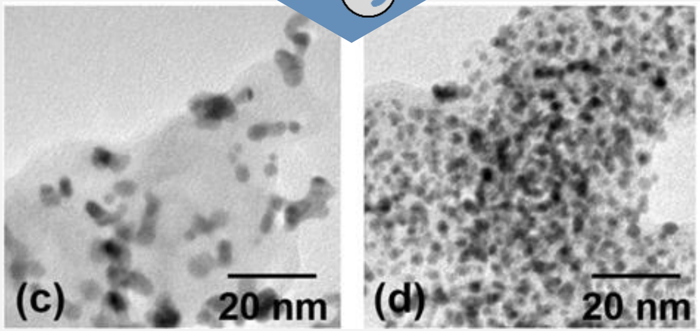
X-ray micro-CT



X-ray nano-CT

SEM

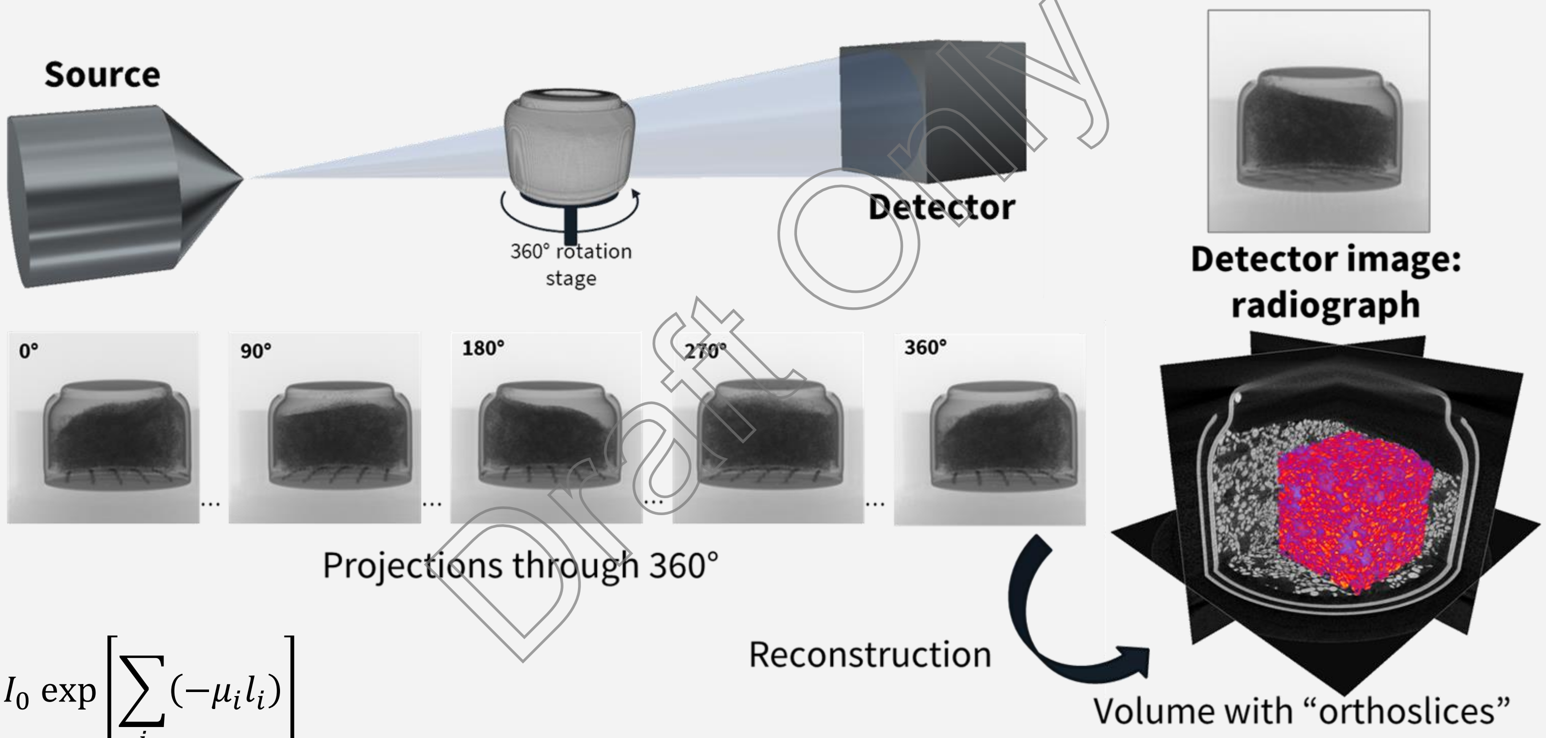




TEM

Q Meyer, J Hack *et al.*, Fuel Cells, 19 (2019) 35-42.

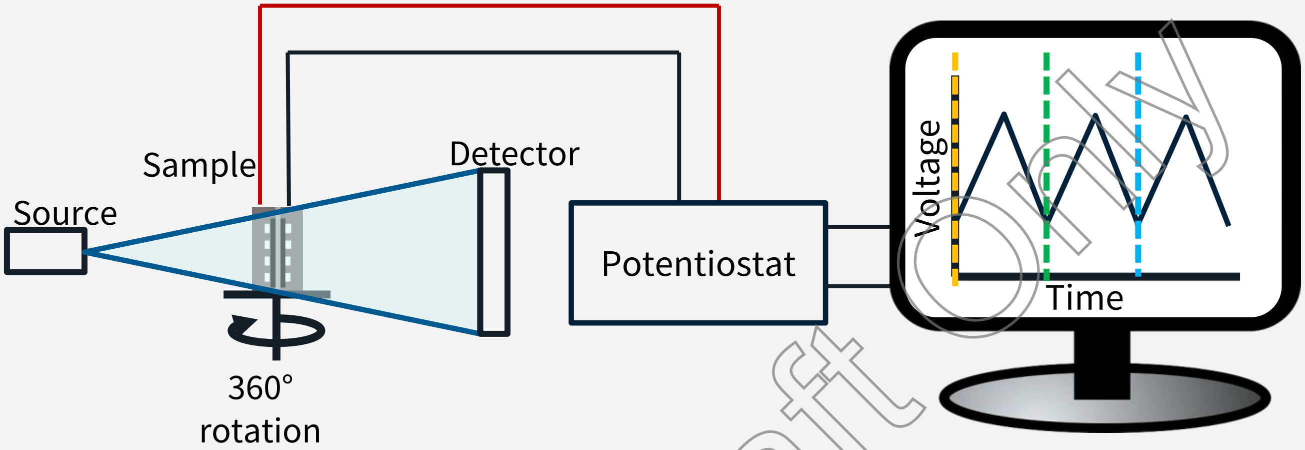
‘Seeing inside’ devices with 3D (and 4D) imaging



$$I = I_0 \exp\left[\sum_i (-\mu_i l_i)\right]$$

Beer-Lambert law for attenuation

Choosing whether to do ex-situ, in-situ or operando



Ideally, we can study the same location/ feature/ material over the course of the degradation test, cycling lifetime – but this depends on the source, the instrument, the cell design.

Imaging dataset; t = 0

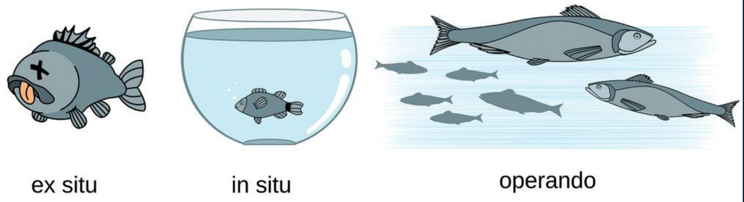
…

Imaging dataset; t = 0 + x

…

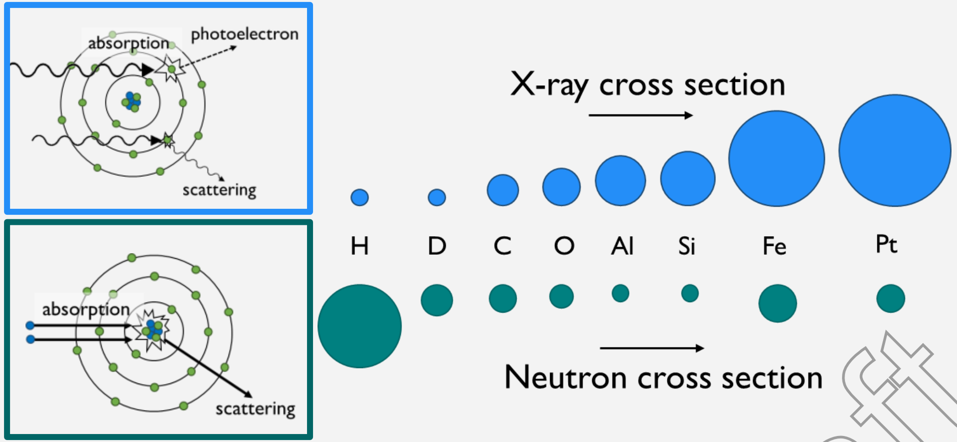
Imaging dataset; t = 0 + y

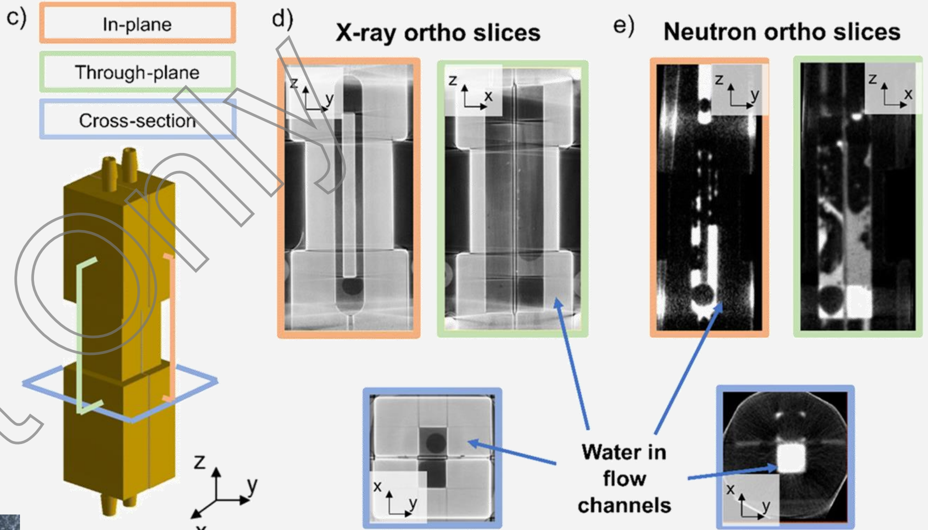
Etc…



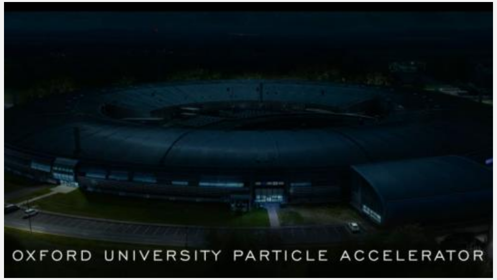
N. Schlüter, P. Novák, D. Schröder, *Adv. Energ. Mater.*, 12 (2022) 2200708, https://doi.org/10.1002/aenm.202200708

X-rays and neutrons are complementary





S. J. Altus, B. J. Inkson and J. Hack, *J. Mater. Chem. A,* 12 (2024) 23364-23391, 10.1039/D4TA02885F

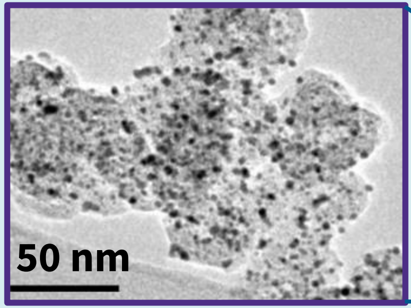


Synchrotrons for if you need neutrons or to go really fast!

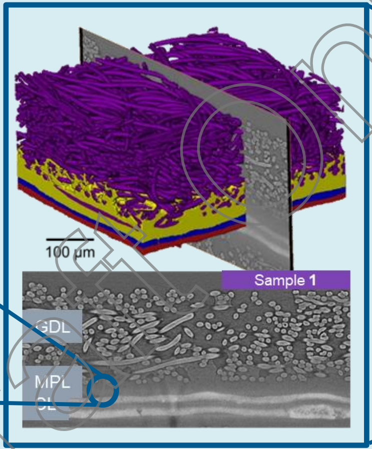
	X-rays	Neutrons
High-resolution	✓ <µm-features	× <mm-features
High-speed	✓ Scans < 1s	↗ Improving!
Imaging light elements (H/Li)	×	✓

Multi-modal correlative imaging is the ideal

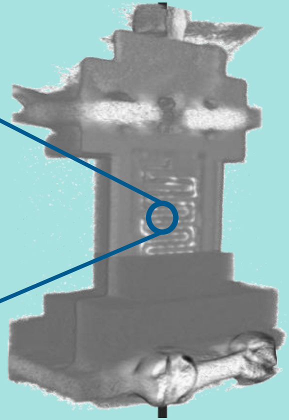
X-rays



J. Hack *et al.*, *Electrochim. Acta,* 352 (2020) 136464,
https://doi.org/10.1016/j.electacta.2020.136464



Neutrons

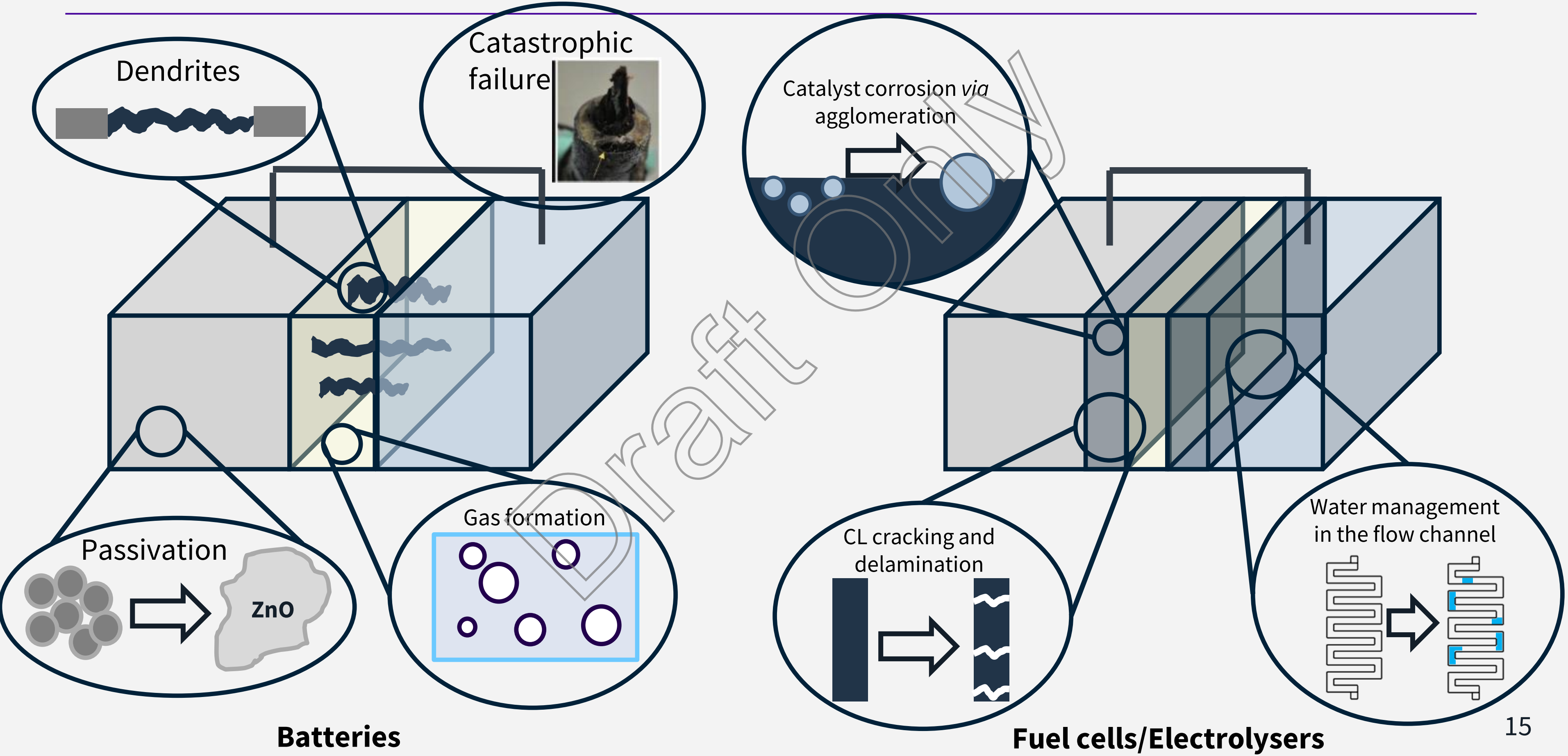


‘Nanoscale’
Atoms and particles

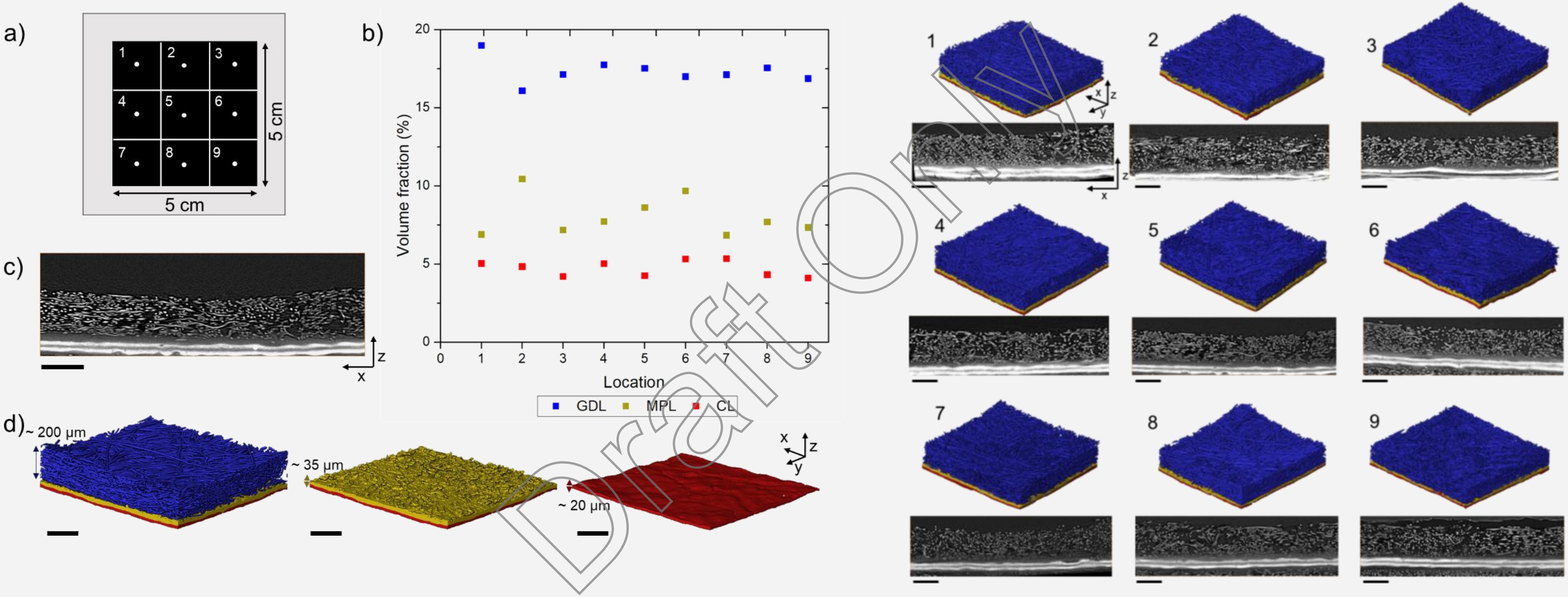
‘Microscale’
Electrodes and layers

‘Macroscale’
Devices

2. They all have failure modes across length scales (at hidden interfaces)



Fuel cells as an example – deep dive into inhomogeneity



- Fuel cell catalyst layers (CLs) are very inhomogeneous from the start.
- Some fluctuation in volume of each phase across the MEA.

J. Hack *et al.*, *J. Electrochem. Soc.*, 167 (2020) 013545, 10.1149/1945-7111/ab6983

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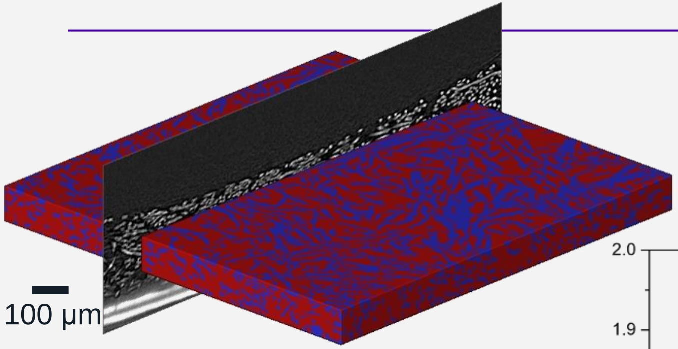
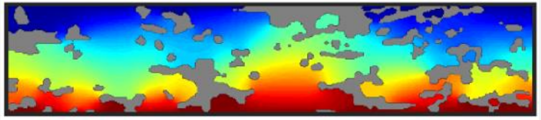
Multiscale inhomogeneity is present from the start



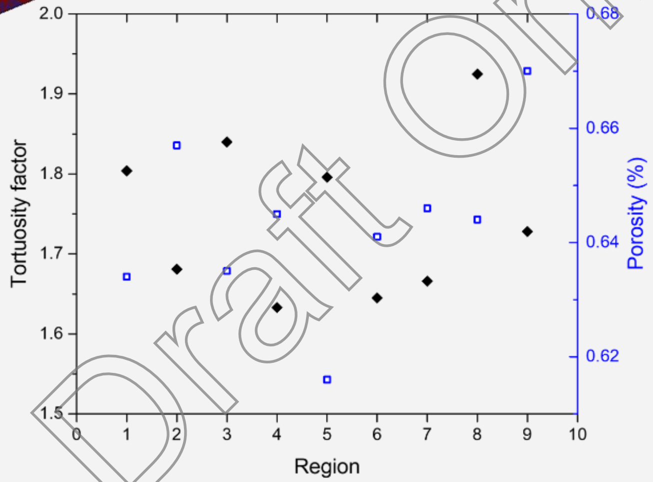
Calculate tortuosity factor

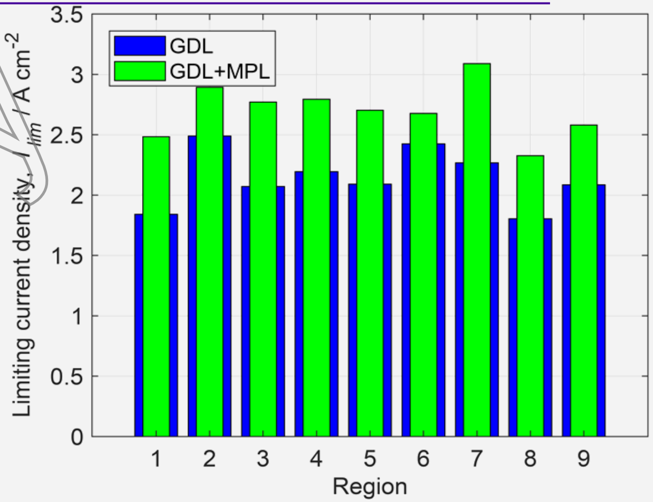


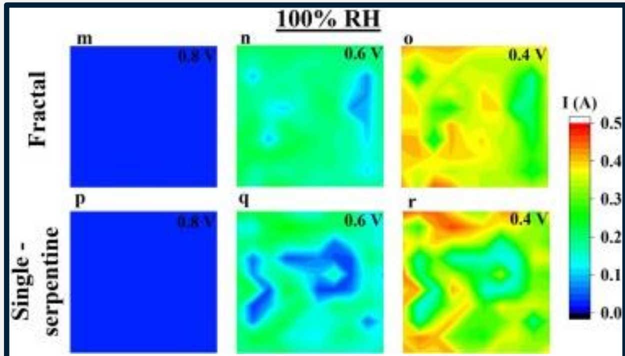
$$\tau = \varepsilon \cdot \frac{D}{D^{eff}}$$

(Cooper *et al.*, *SoftwareX* (2016))

Leading to inhomogeneous predicted current distributions across the electrode.



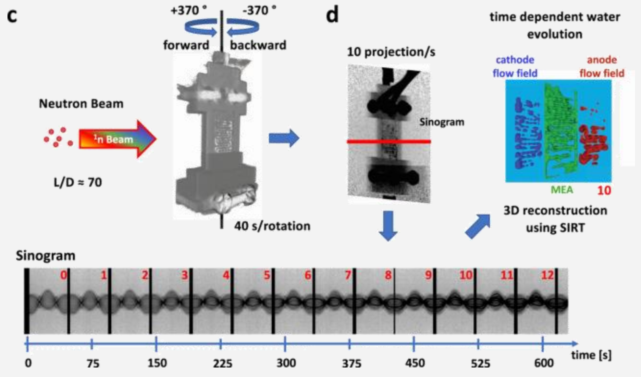


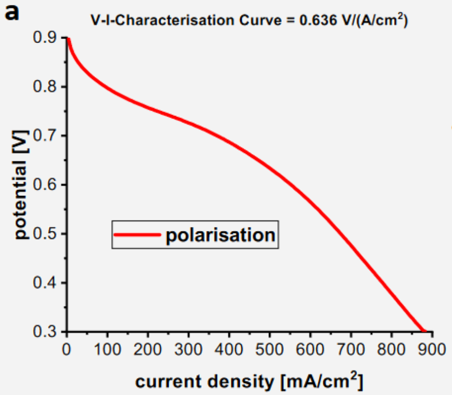


V. S. Bethapudi, J. Hack *et al.*, *Energy. Conv. Manag.*, 250 (2021) 114924. https://doi.org/10.1016/j.enconman.2021.114924

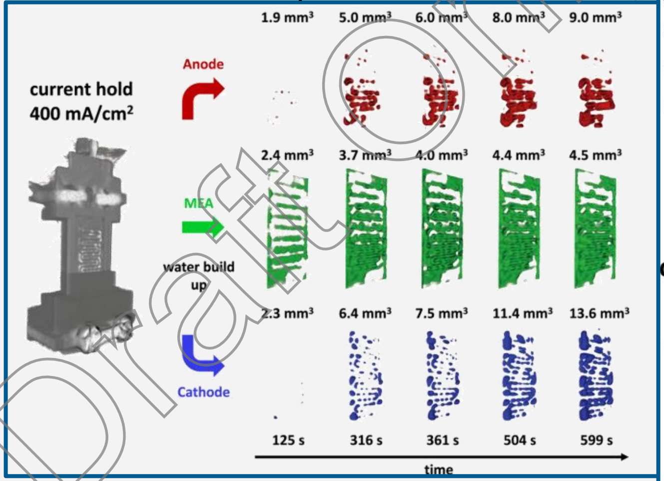
J. Hack *et al.*, *J. Electrochem. Soc.*, 167 (2020) 013545, 10.1149/1945-7111/ab6983

The way water is produced is also non-uniform…

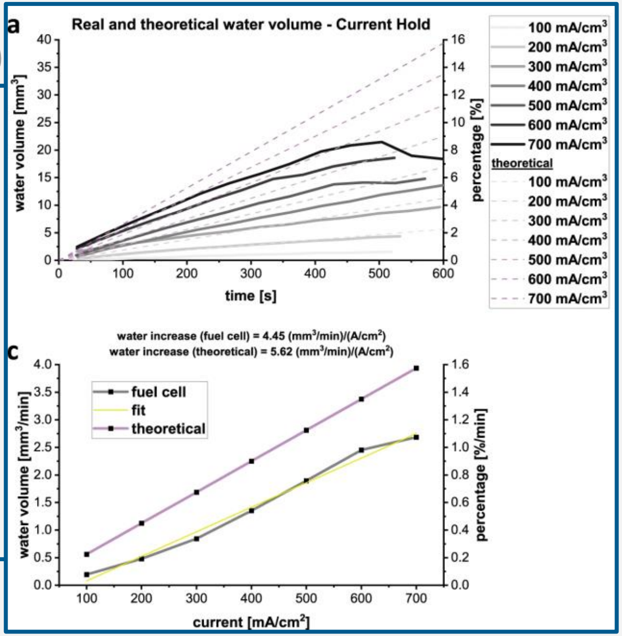




Qualitative information about each component



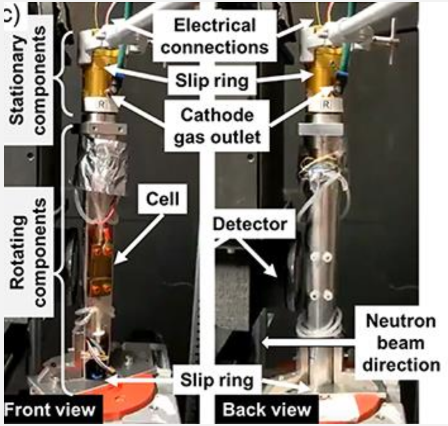
Plus quantification of water volume (and removal rate) in the cell

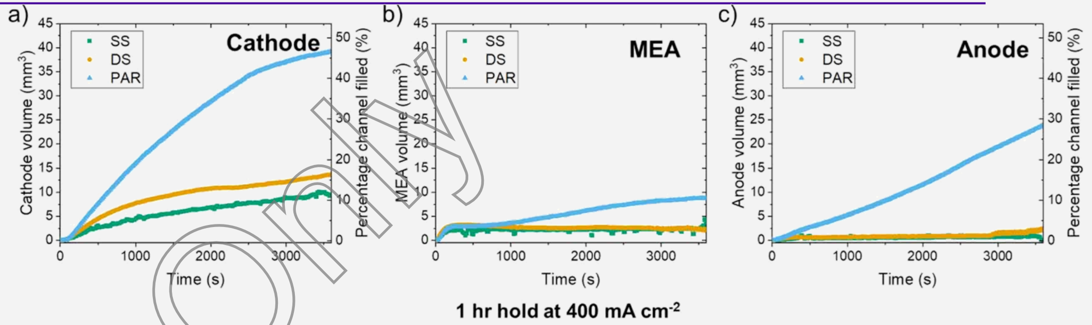


High-speed for neutrons = 36 s per scan!

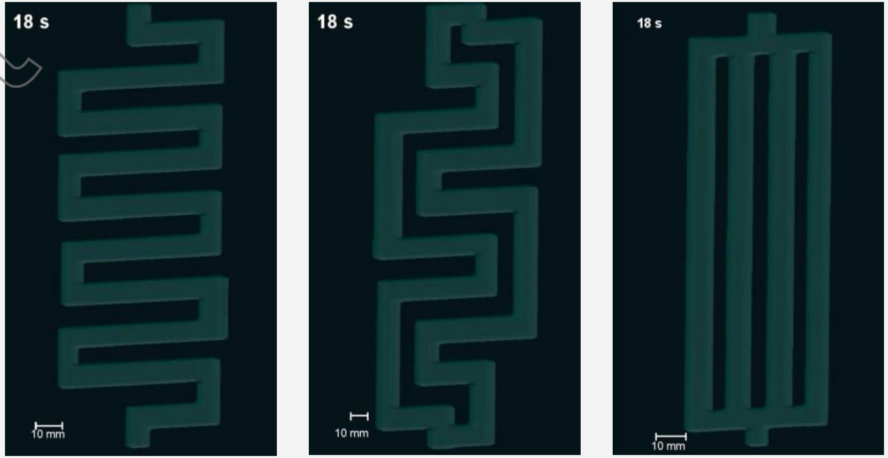
R. F. Ziesche, J. Hack *et al.*, *Nat. Comms.*, 13 (2022) 1616. https://doi.org/10.1038/s41467-022-29313-5

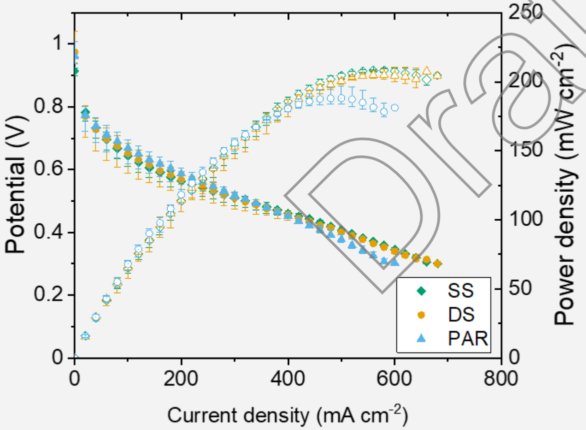
And the flow field design greatly influences water distribution





Results of *operando* neutron CT @ 400 mA cm^{-2}





NEUTRONS FOR SOCIETY

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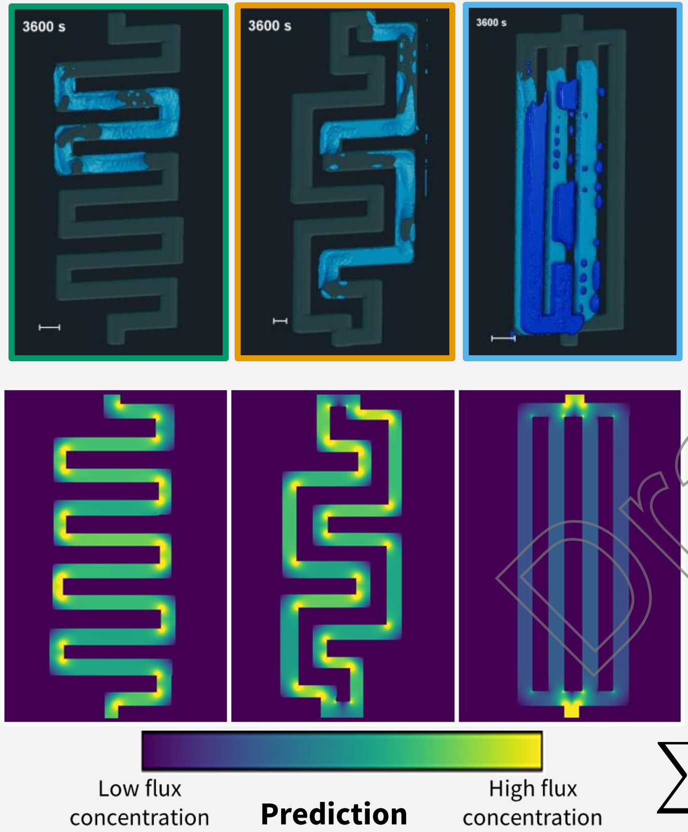
J. Hack *et al.*, *J. Phys. Energy*, 6 (2024) 025021. 10.1088/2515-7655/ad3984

Even higher-speed for neutrons = 18 s per scan!

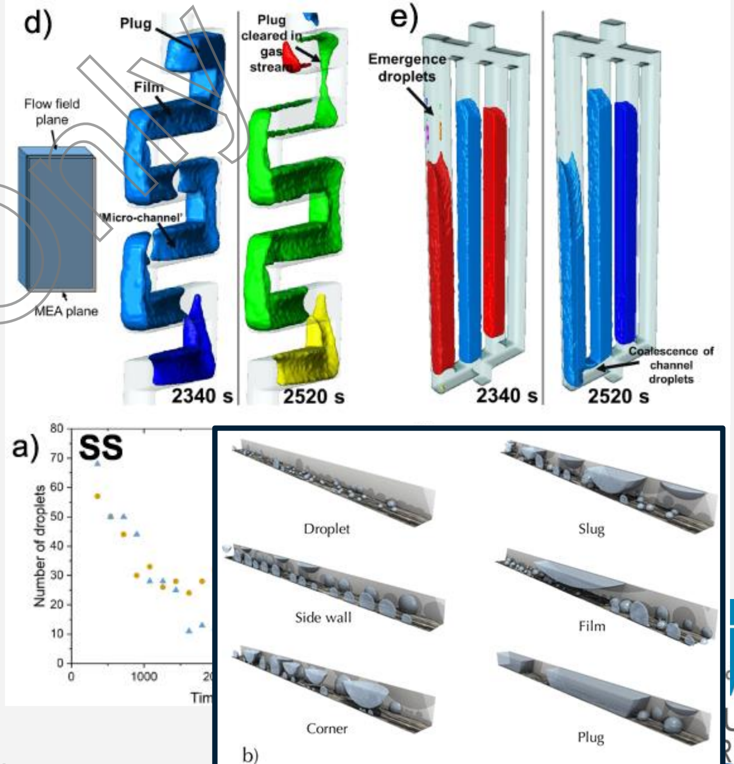
We can also understand how individual droplets evolve

Results of *operando* neutron CT (after 3600s @ 400 mA cm^{-2})



$$\sum A_{in} v_{in} = \sum A_{out} v_{out}$$

Understanding droplet type and evolution

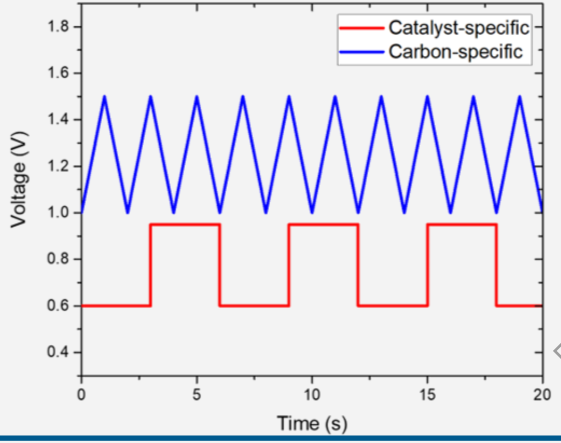


D. Niblett, S. M. Holmes, V. Niasar, *ACS Appl. Energy. Mater.*, 4 (2021) 10514-10533.

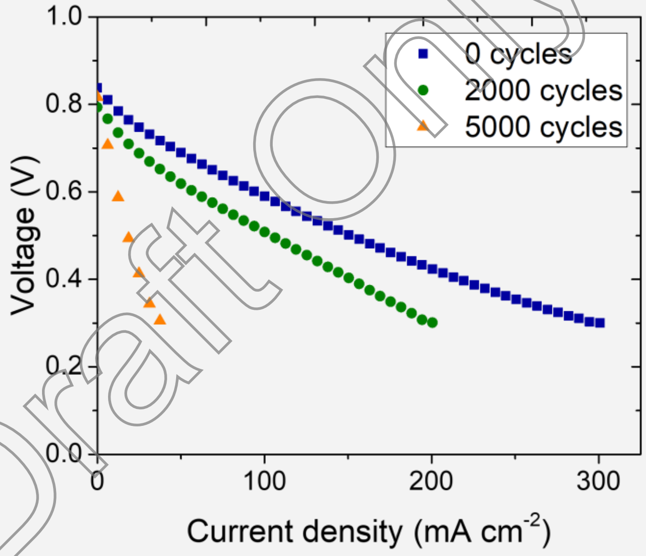
J. Hack *et al.*, *J. Phys. Energy*, 6 (2024) 025021. 10.1088/2515-7655/ad3984

Inhomogeneous start leads to an inhomogeneous end

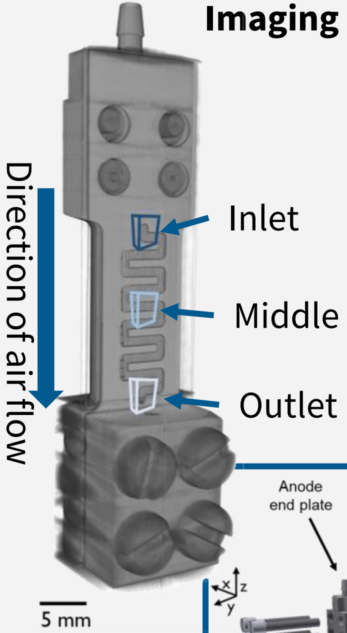
Accelerated stress tests (ASTs) allow us to degrade cells on a reasonable time scale – i.e. hours to days (not 10 years!)

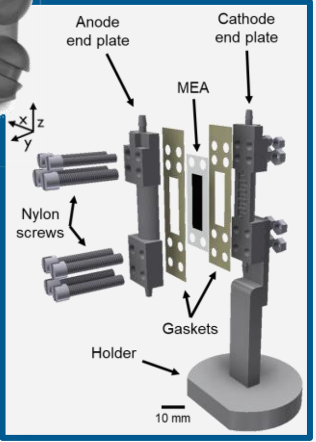


Electrochemistry



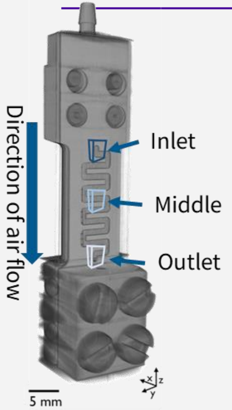
Imaging



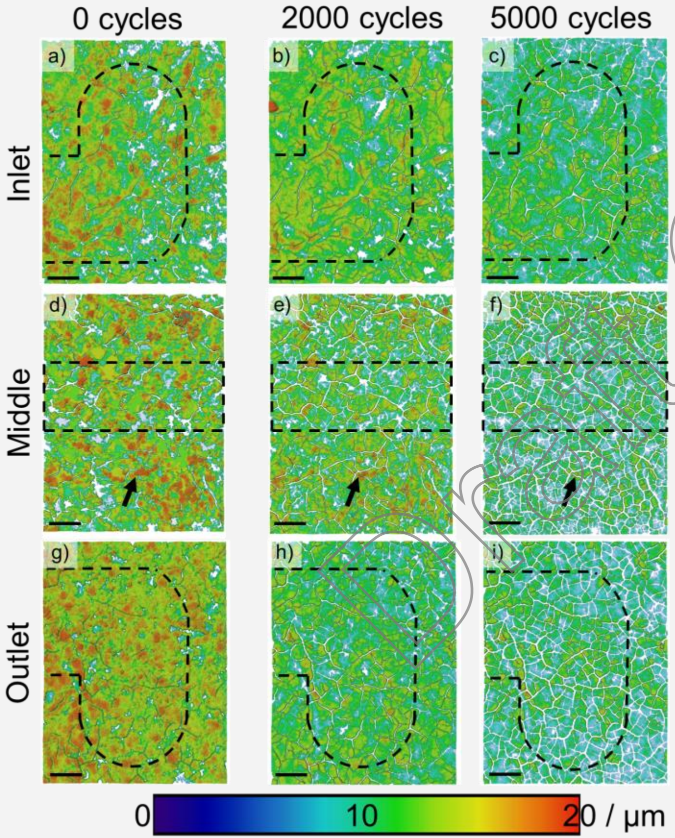


J. Hack et al., Electrochim. Acta, 352 (2020), 136464.
https://doi.org/10.1016/j.electacta.2020.136464

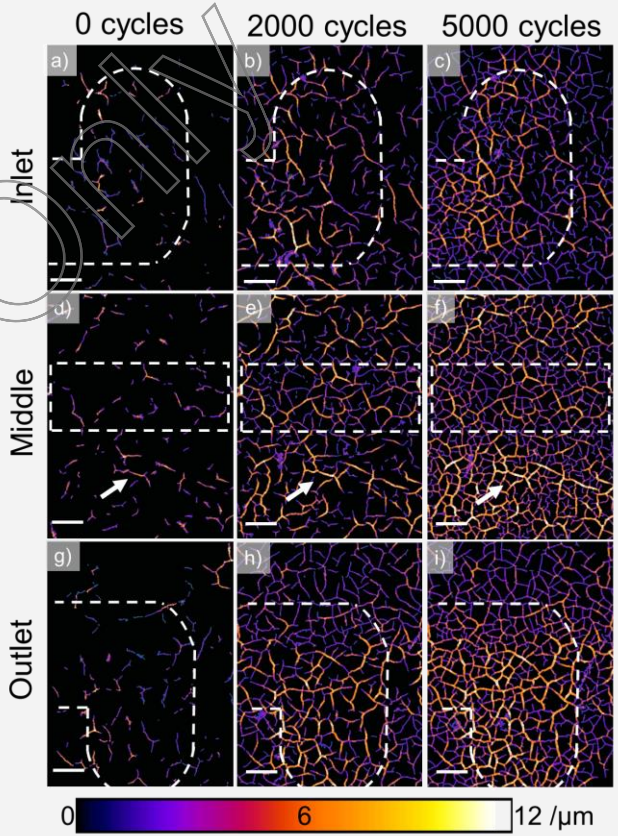
Extent of electrochemical-induced cracking varies along the flow channel



CL thickness decreases



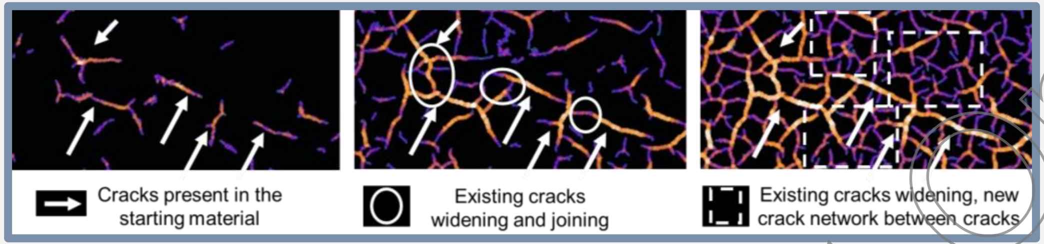
Extent of cracking increases

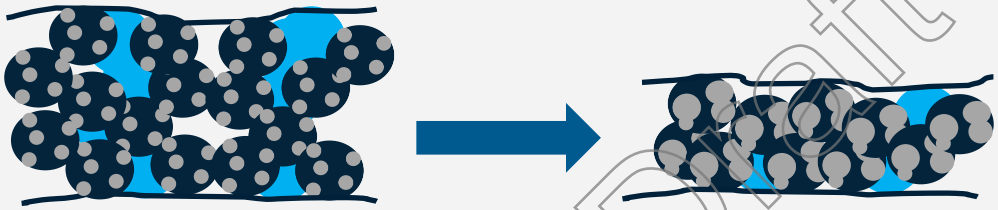


J. Hack et al., Electrochim. Acta, 352 (2020), 136464. https://doi.org/10.1016/j.electacta.2020.136464

This leads to a collapse of the CL structure and degradation on the nanoscale

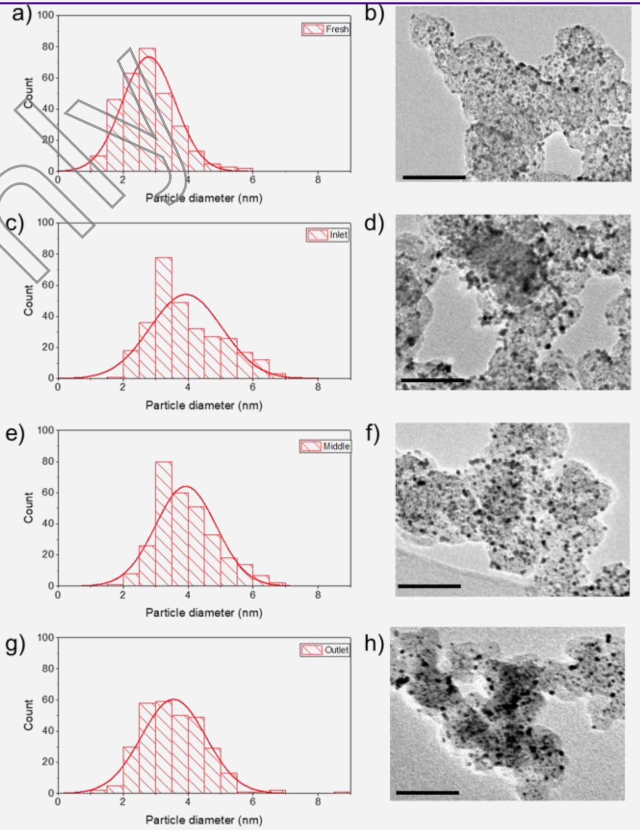
Microscale crack network evolution…





Fresh CL:
- Connected pore network
- Effective water removal
- Good electrical contact

Degraded CL:
- Poorly-connected pore network
- Poor water removal
- Agglomerated catalyst

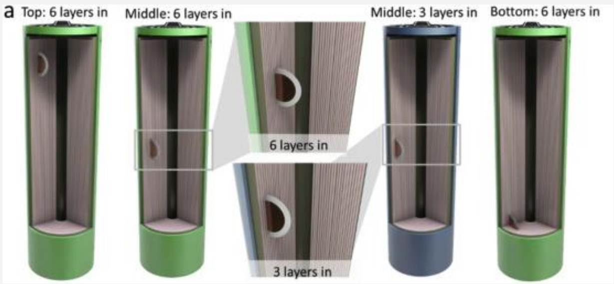


As a result of carbon-support and catalyst degradation at the nanoscale.

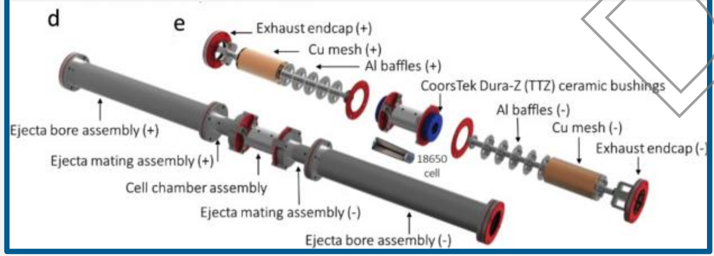
J. Hack et al., Electrochim. Acta, 352 (2020), 136464. https://doi.org/10.1016/j.electacta.2020.136464

Using X-rays to go *really* fast – studying Li-ion battery safety

Internal short-circuiting (ISC) device used to mimic the effect of a defect.

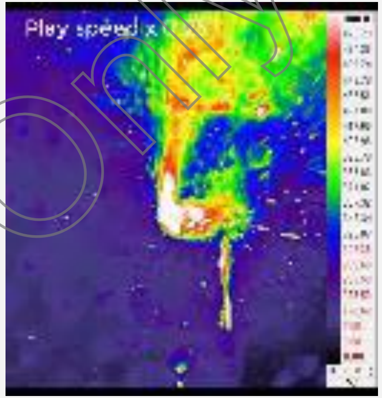


Custom-built calorimeter allowed for correlation of high-speed imaging to thermal behaviour of cells.

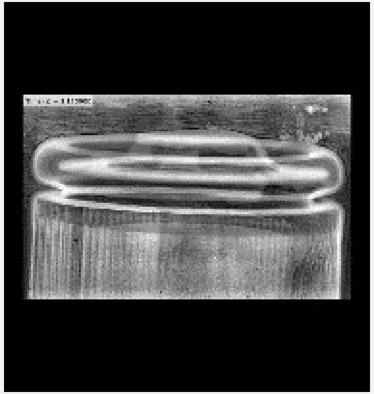


Example: Placing ISC device near the top of the cell.

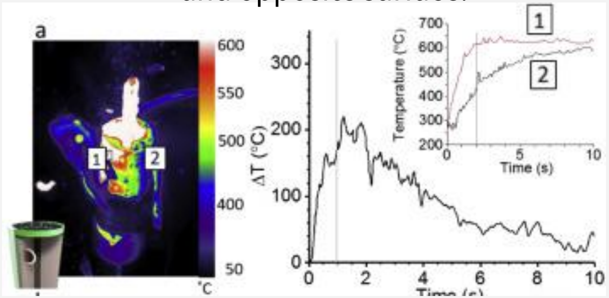
Thermal imaging (50 fps) shows breach of casing.



Visualised by high-speed radiography (>2000 fps).



And >200 °C difference between surface near ISC device and opposite surface.

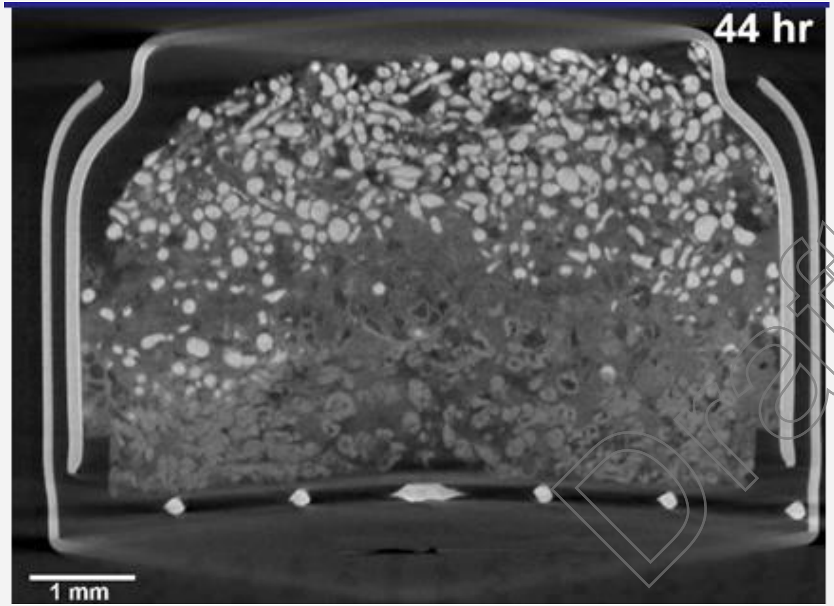


D. Finegan *et al.*, 417 (2019) 29-41, https://doi.org/10.1016/j.jpowsour.2019.01.077

Or much slower to study alternative cell chemistries

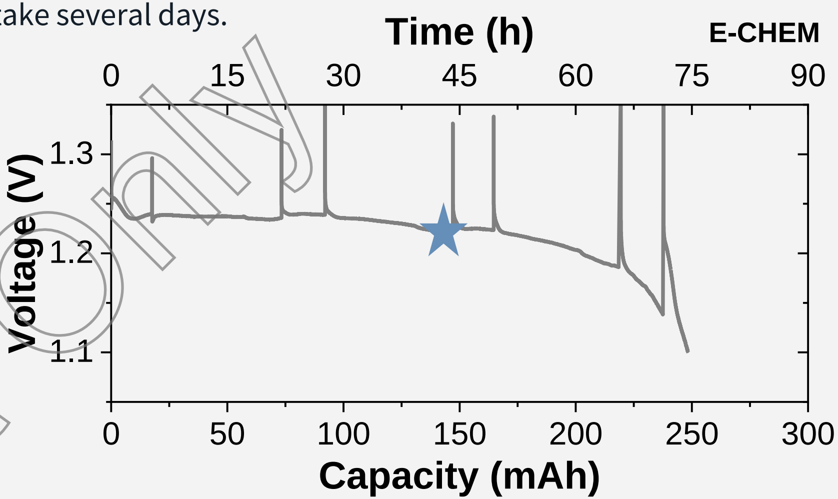
Whereas Li-ion batteries can discharge in less than 1 h, zinc-air batteries take several days.

ORTHOSLICES

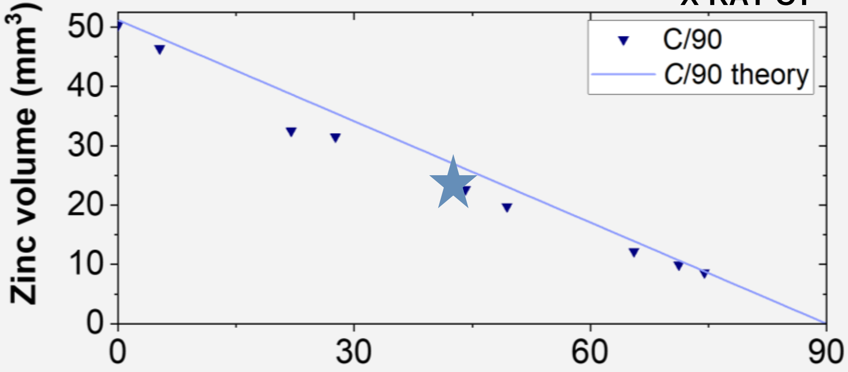


$$Zn + \frac{1}{2}O_2 \rightarrow ZnO$$

E-CHEM

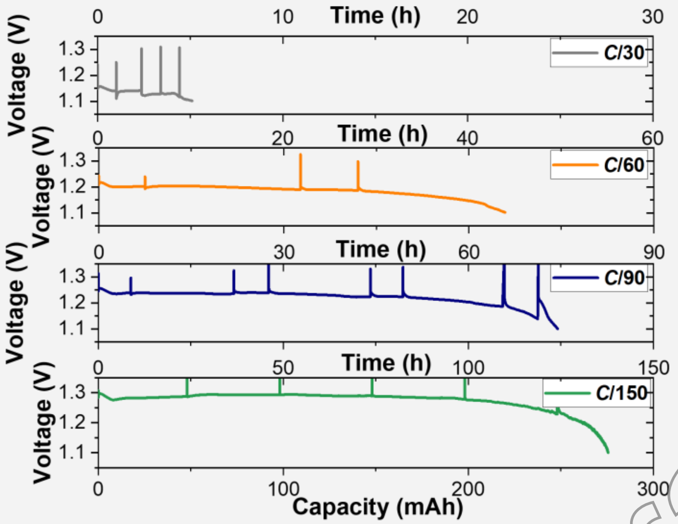


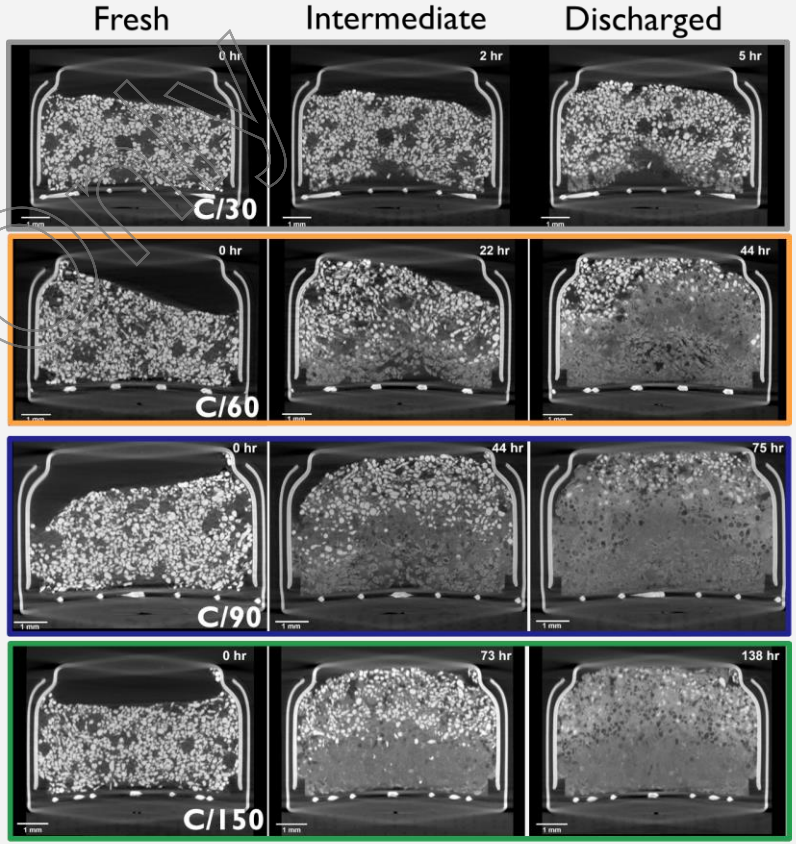
X-RAY CT

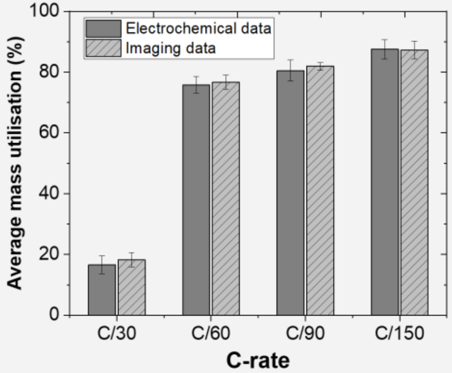


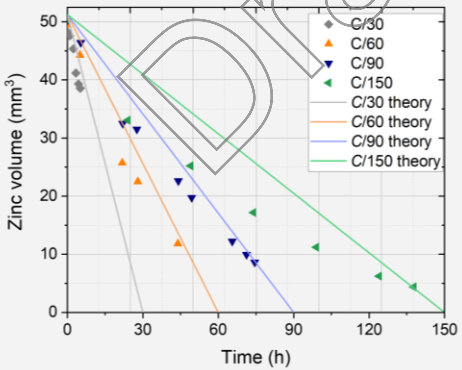
J. Hack *et al.*, *J. Phys. Mater.*, 5 (2022), 014001. 10.1088/2515-7639/ac3f9a

ZnO passivation and poor conductivity are a key barrier for realizing rechargeable zinc-air batteries





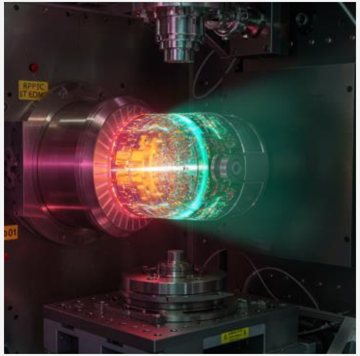




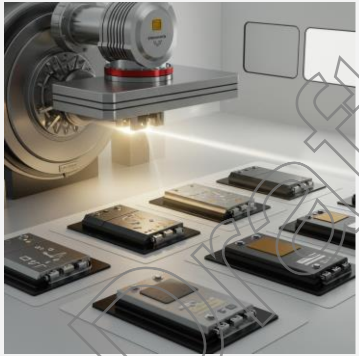
Where do we need to go next with imaging?

Synchrotron sources and lab-based X-ray CT instruments have been continuously improving. Now we can…

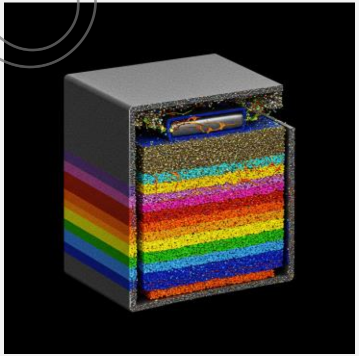
Go faster



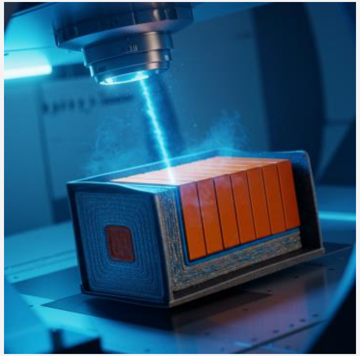
Scan more samples



Achieve higher resolutions



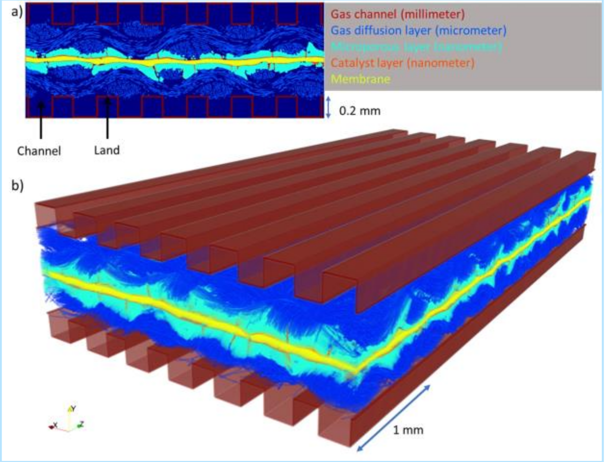
Generate more data!



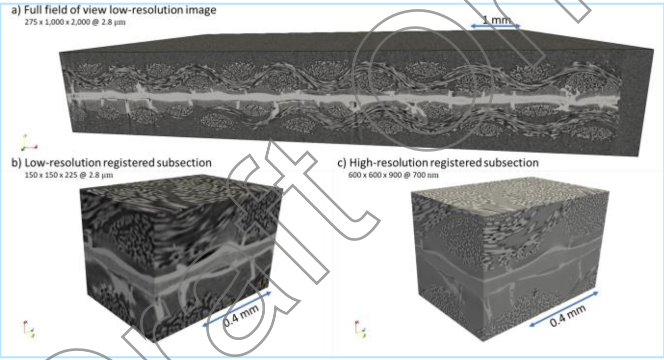
Images courtesy of Google Gemini

This means it's possible to achieve 'super-resolution'

One super-resolved image obtained with lab-based CT!

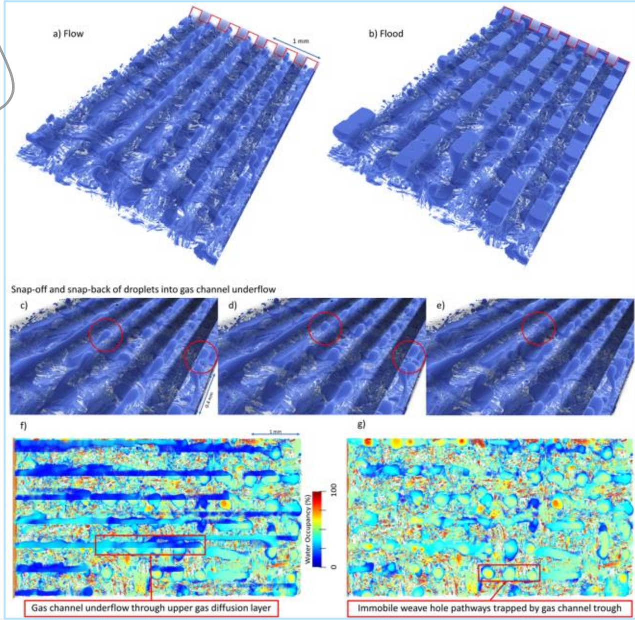


Comprising: low-res, large FOV + high-res, narrow FOV, trained using AI



Wang *et al.*, *Nat. Comms.*, 14 (2023) 745

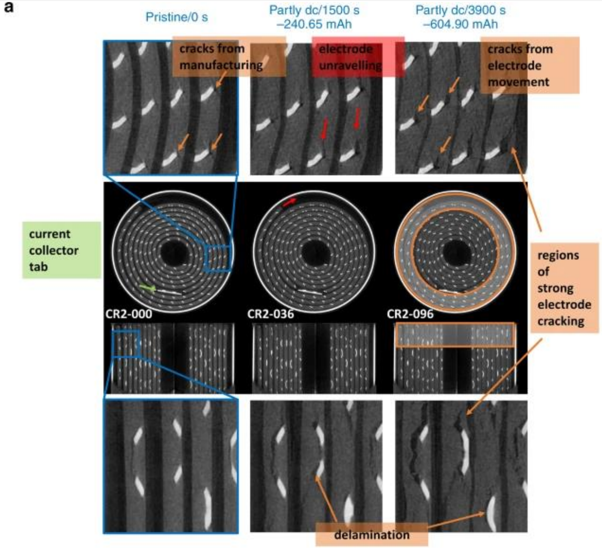
Enabling correlative multiscale model generation



Low-res scans collected in ~2 hours, whereas high-res scan took ~11 hours!

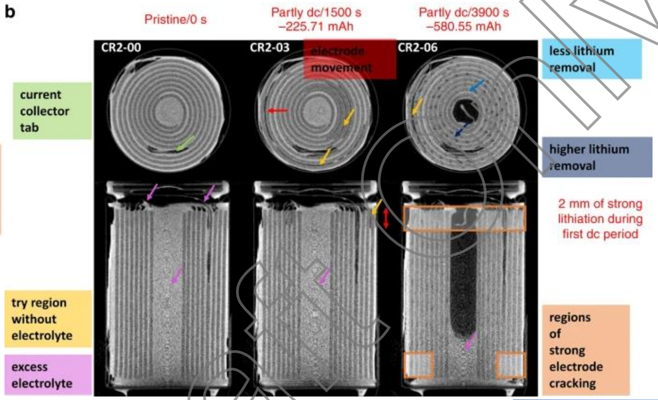
Could multi-modal be a solution instead… or as well?

And we can start moving towards multimodal imaging

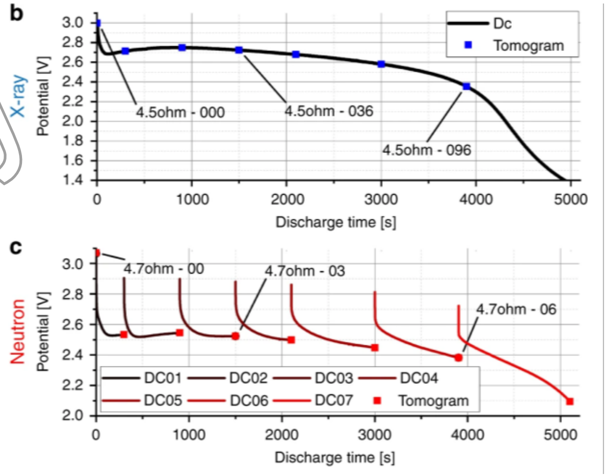


High-speed X-ray tomography

Neutron tomography for visualising lithium



Ziesche *et al.*, *Nat. Comms.*, 11 (2020) 777



X-ray tomo in 2.8 s!, 10.87 µm voxel size.
Neutron tomo in 8 h, 12.9 µm pixel size.

ILL neutron facility

*other national facilities are available!

We should start using more data and repeating things (which is a big challenge for inhomogeneous materials found in electrochemical devices)

- How can we condense >5 TB of beamtime data into one journal article?
- How can we rigorously analyse all this data, consider no two electrochemical devices perform the same?

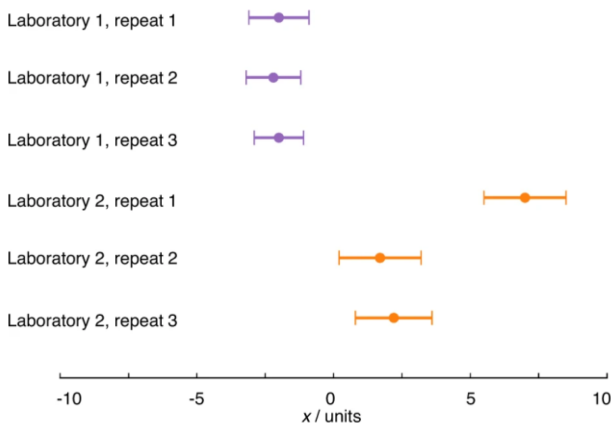
Journal article with 5-8 figures

Comment | Open access | Published: 11 November 2022

Error, reproducibility and uncertainty in experiments for electrochemical energy technologies

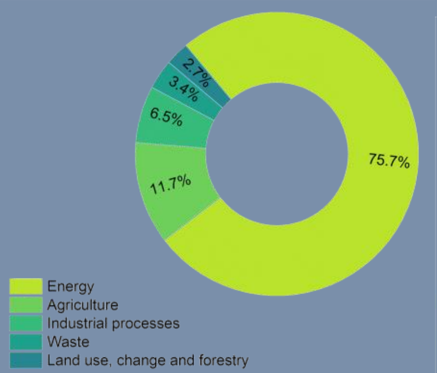
Graham Smith & Edmund J. F. Dickinson

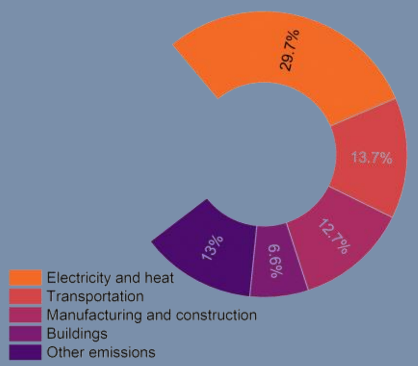
Nature Communications **13**, Article number: 6832 (2022) | Cite this article



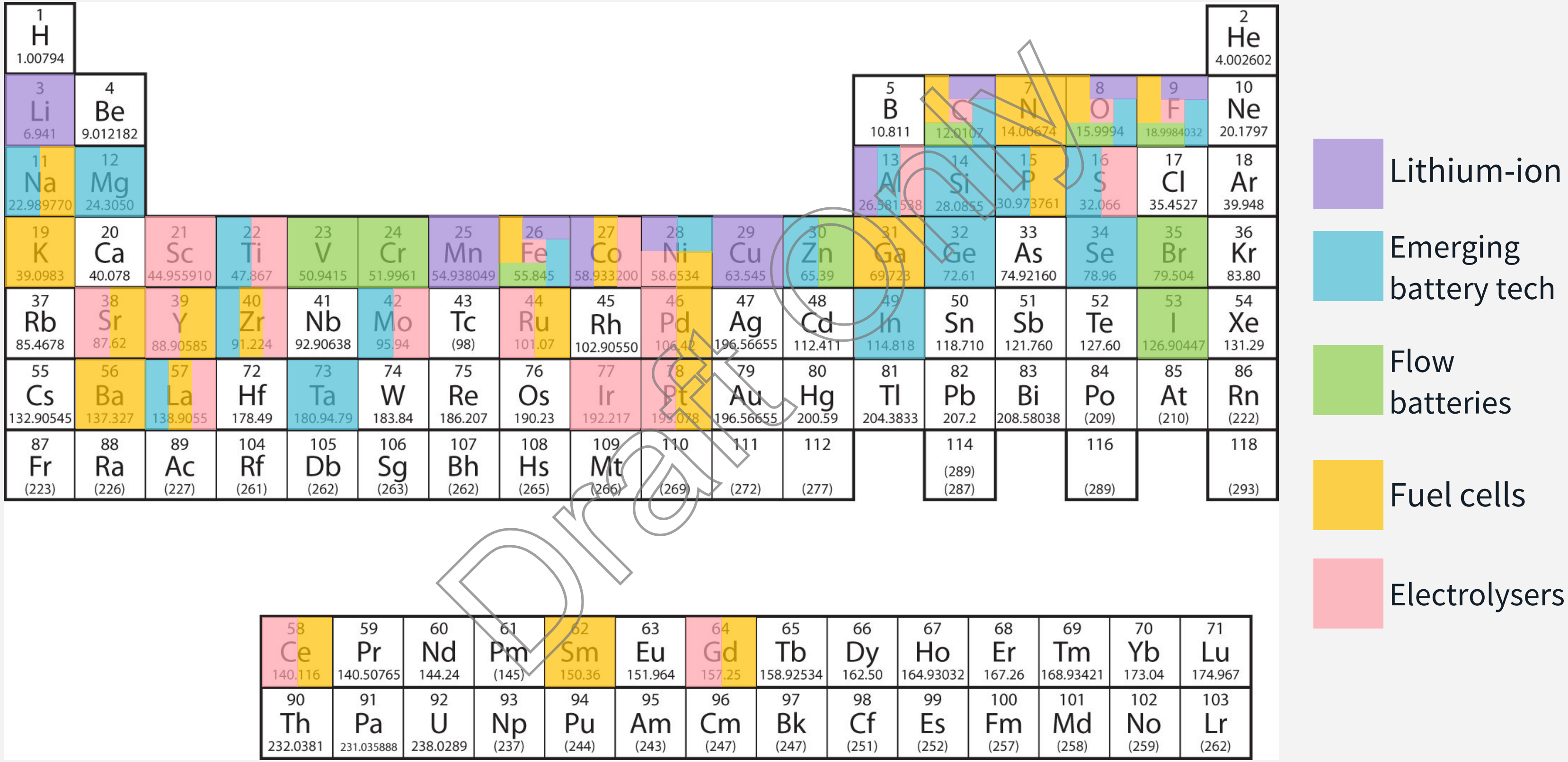
Smith and Dickinson., *Nat. Comms.*, 13 (2022) 6832

And we should be trying to do this sustainably…



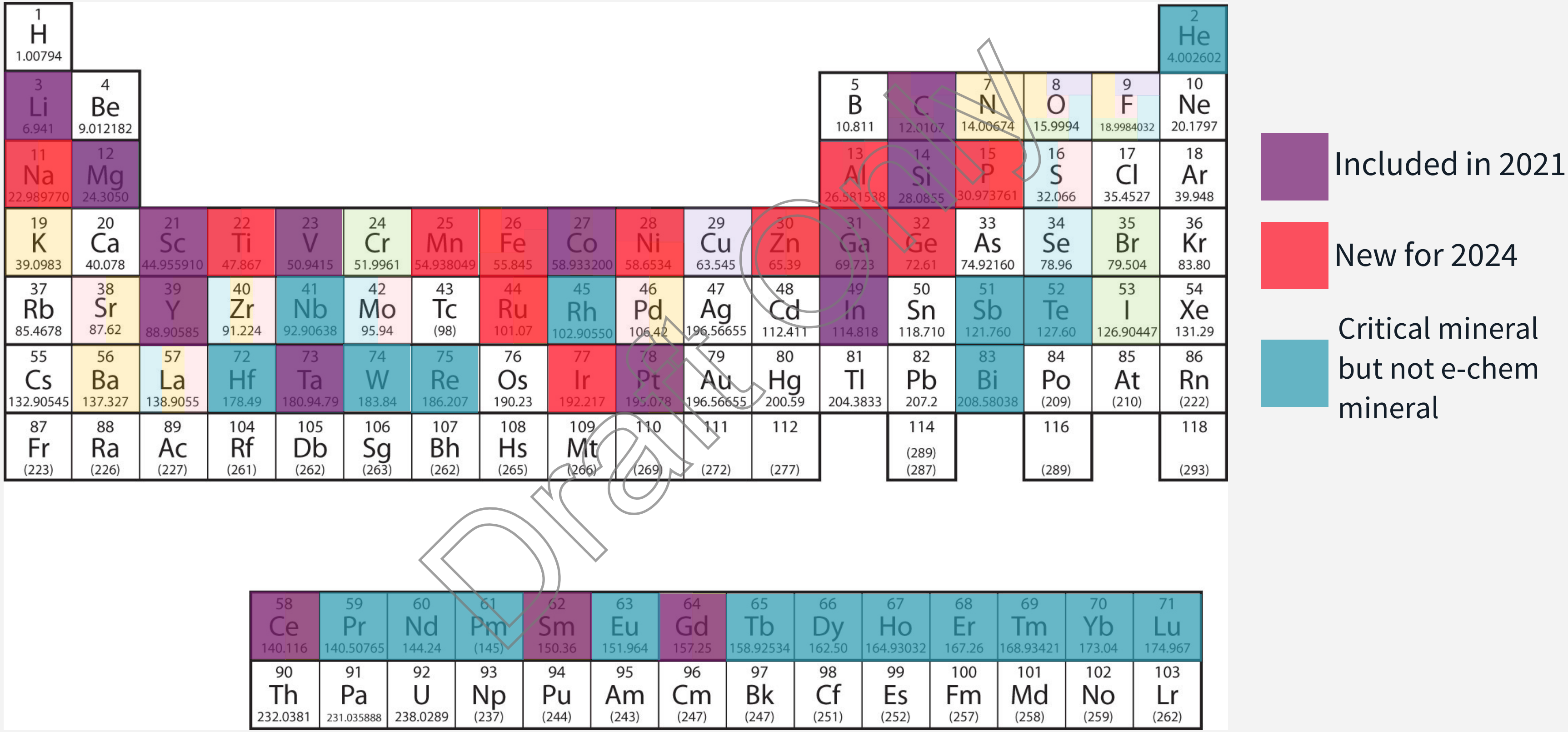


Critical materials: a critical time for electrochemical technologies?



Useful resource: https://criticalmaterials.energypolicy.columbia.edu/

Critical minerals list: (even more?) critical for electrochemical technologies?

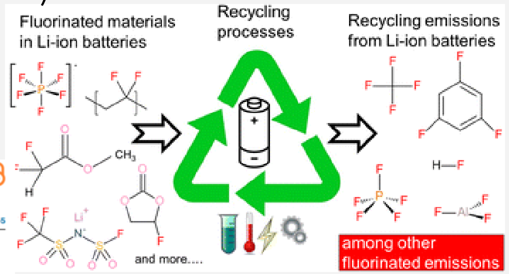


Data from: UK Critical Minerals Intelligence Centre, Criticailty Assessment 2024
https://www.ukcmic.org/downloads/reports/ukcmic-2024-criticality-assessment.pdf

And not forgetting the environmental challenges too…

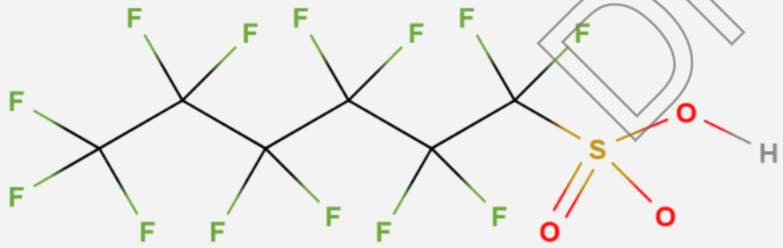
$$-(CF_2-CF_2)_6-CF_2-CF-$$
$$\qquad\qquad\qquad\qquad | $$
$$\qquad\qquad\qquad\qquad O-CF_2-CF_2-SO_3H$$

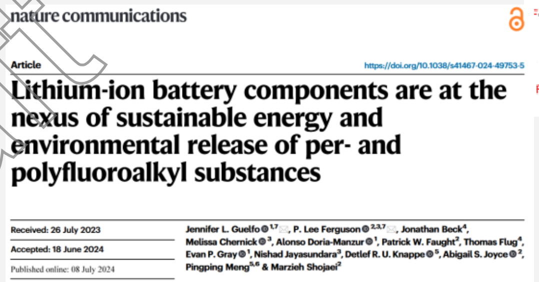
Nafion® in fuel cells and electrolysers: perfluorosulfonic acid (PFSA) ionomer



Rensmo, Benskin *et al.*, *Environ. Sci.: Processes Impacts* (2023)

PFAS = Per- and polyfluoroalkyl substances are everywhere!… Including in energy storage and conversion.




nature communications

Article

https://doi.org/10.1038/s41467-024-49753-5

Lithium-ion battery components are at the nexus of sustainable energy and environmental release of per- and polyfluoroalkyl substances

Received: 26 July 2023

Accepted: 18 June 2024

Published online: 08 July 2024

Jennifer L. Guelfo[1,7], P. Lee Ferguson[2,3,7], Jonathan Beck[4], Melissa Chernick[3], Alonso Doria-Manzur[1], Patrick W. Faught[2], Thomas Flug[4], Evan P. Gray[1], Nishad Jayasundara[3], Detlef R. U. Knappe[5], Abigail S. Joyce[2], Pingping Meng[5,6] & Marzieh Shojaei[2]

Challenges include:

- Current analogues don't perform as well or meet KPIs.
- Legislation and regulation is currently unclear.
- End-of-life handling and recycling not established.

Four possible (materials) strategies for addressing the critical minerals challenge…

The 'million-mile' battery, Harlow, Dahn *et al*. (2019)

A Wide Range of Testing Results on an Excellent Lithium-Ion Cell Chemistry to be used as Benchmarks for New Battery Technologies

Jessie E. Harlow,[1,2] Xiaowei Ma,[1,2] Jing Li,[1,2] Eric Logan,[1,2] Yulong Liu,[1,2] Ning Zhang,[1,2] Lin Ma,[1,2] Stephen L. Glazier,[1,2] Marc M. E. Cormier,[1,2] Matthew Genovese,[1,2,*] Samuel Buteau,[1,2] Andrew Cameron,[1,2] Jamie E. Stark,[1,2] and J. R. Dahn[1,2,**,z]

[1]Department of Physics and Atmospheric Science, Dalhousie University, Halifax, N.S B3H 4R2, Canada
[2]Department of Chemistry, Dalhousie University, Halifax, N.S B3H 4R2, Canada

Press release 11/14/2023

Heraeus and Sibanye-Stillwater introduce a ruthenium-based catalyst to reduce reliance on iridium for PEM water electrolysis

Analysis | Open access | Published: 13 January 2025

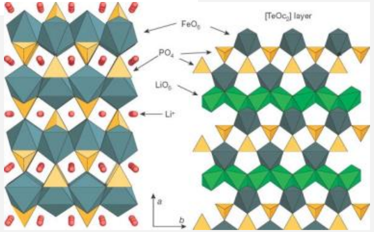
Critically assessing sodium-ion technology roadmaps and scenarios for techno-economic competitiveness against lithium-ion batteries

Adrian Yao, Sally M. Benson & William C. Chueh

Nature Energy (2025) | Cite this article

S. J. Altus, B. J. Inkson and J. Hack, *J. Mater. Chem. A*, 12 (2024) 23364-23391, 10.1039/D4TA02885F

LFP/LMFP cathodes



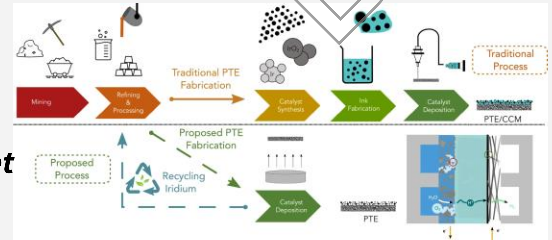
Tarascon & Armand, Nature, 414 (2001) 359-367.

UK research programmes in battery recycling

THE FARADAY INSTITUTION

ReLiB
REUSE & RECYCLING OF LITHIUM ION BATTERIES

'Ionomer-free and recyclable', Lee, Peng *et al.*, *Nat. Comms.* (2023).

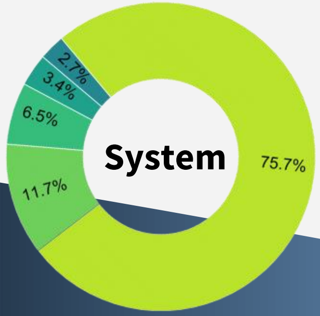


US DoE Technical Targets for Proton Exchange Membrane Electrolysis

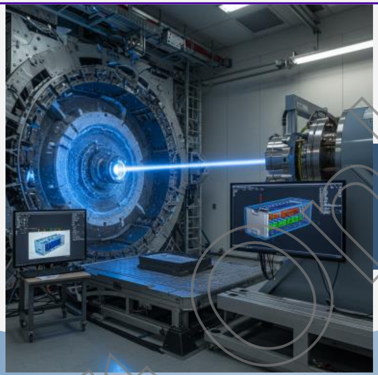
Characteristic	Units	2022 Status	2026 Targets	Ultimate Targets
Total Platinum Group Metal Content (both electrodes combined)	mg/cm^2	3.0	0.5	0.125
	g/kW	0.8	0.1	0.03

https://www.energy.gov/eere/fuelcells/technical-targets-proton-exchange-membrane-electrolysis

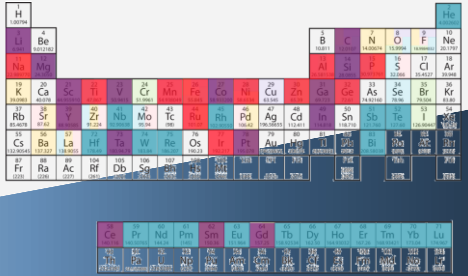
Summary



Next generation imaging



Acknowledge criticality



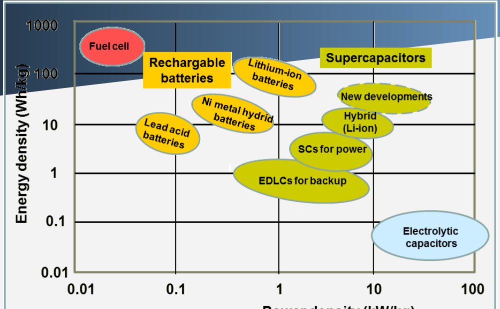
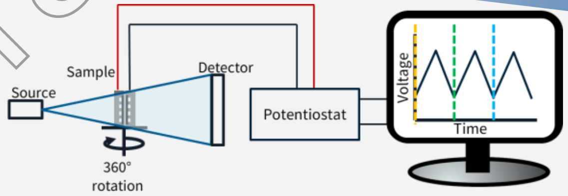


Image: Elcap/CC0 1.0

Devices



Imaging dataset; t = 0 … Imaging dataset; t = 0 + x … Imaging dataset; t = 0 + y Etc…

Multiscale imaging

Address a new set of challenges

Many thanks to

[Group picture coming soon!]

Royal Academy of Engineering

UKRI Engineering and Physical Sciences Research Council

HZB Helmholtz Zentrum Berlin

electrochemical innovation lab

HENRY ROYCE INSTITUTE

ILL NEUTRONS FOR SOCIETY

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uc3m Universidad Carlos III de Madrid