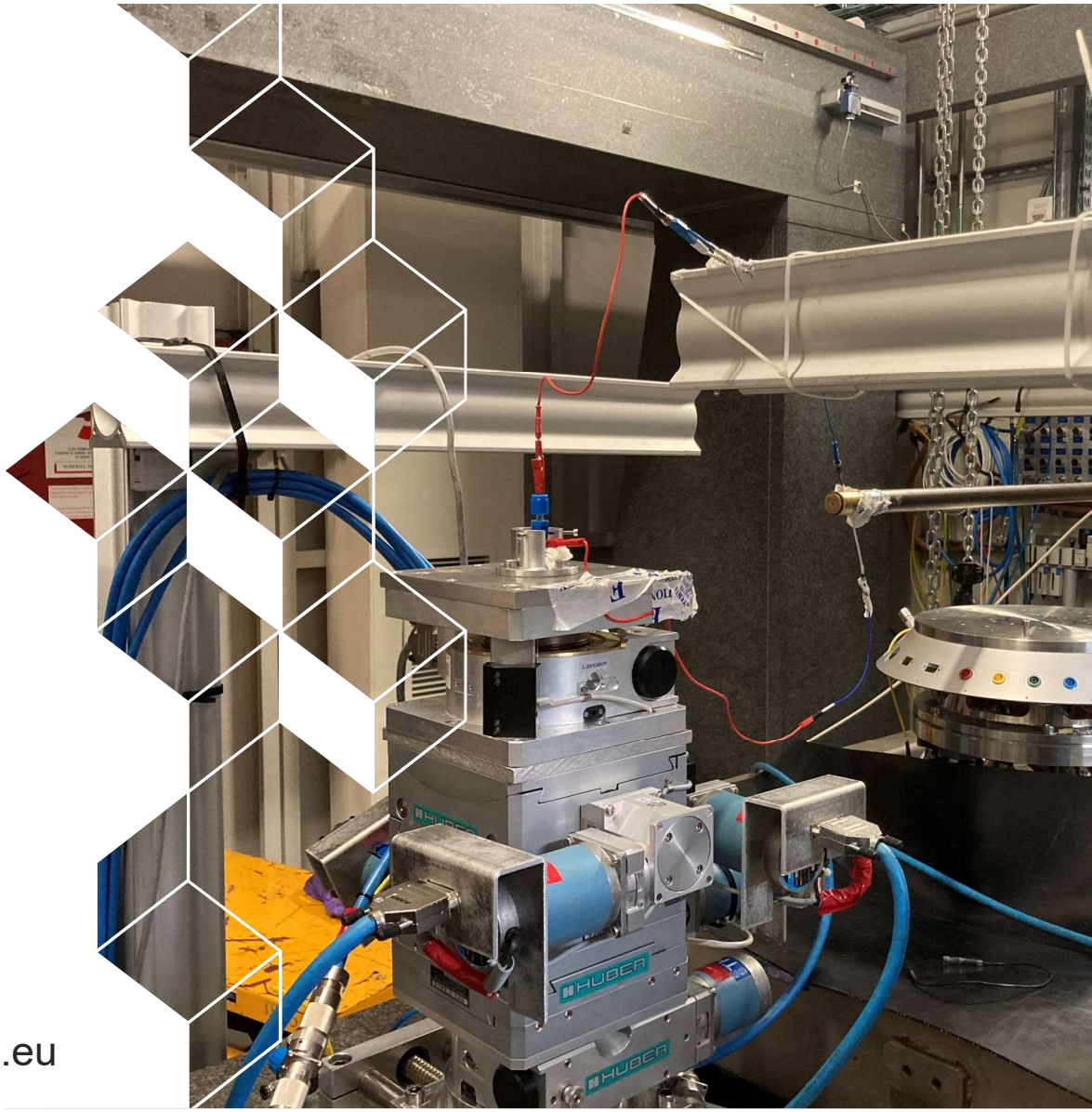
(Operando) studies on battery materials

Sandrine Lyonnard

www.europeanbatteryhub.eu

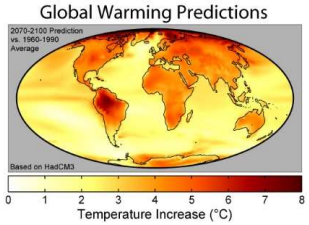


Outline

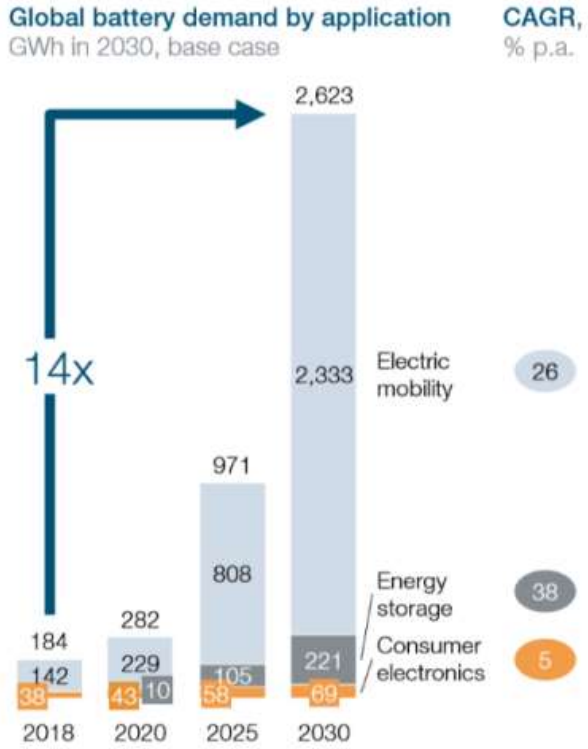
- I - A (not so quick) introduction to battery research challenges
- II - …and why large scale facilities are unique tools
- III - Few examples of x-ray and neutron studies – insisting on multimodal and multitechniques approach: the case of Silicon anodes.
- IV – How can we truly reach a more « holistic » understanding ?
  The European Battery Hub in Grenoble

Li-ion batteries, and beyond

Societal Impact

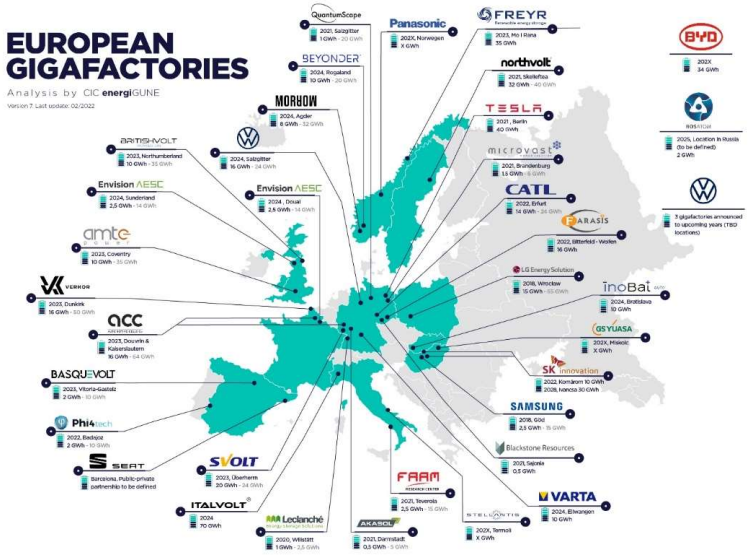


Market growth



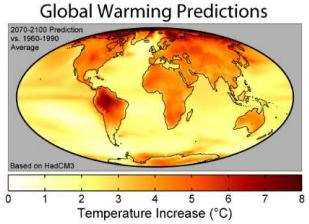
Industrial challenges

- More batteries must be fabricated
- Better batteries must be invented (performances, lifetime, safety, cost)



Li-ion batteries, and beyond

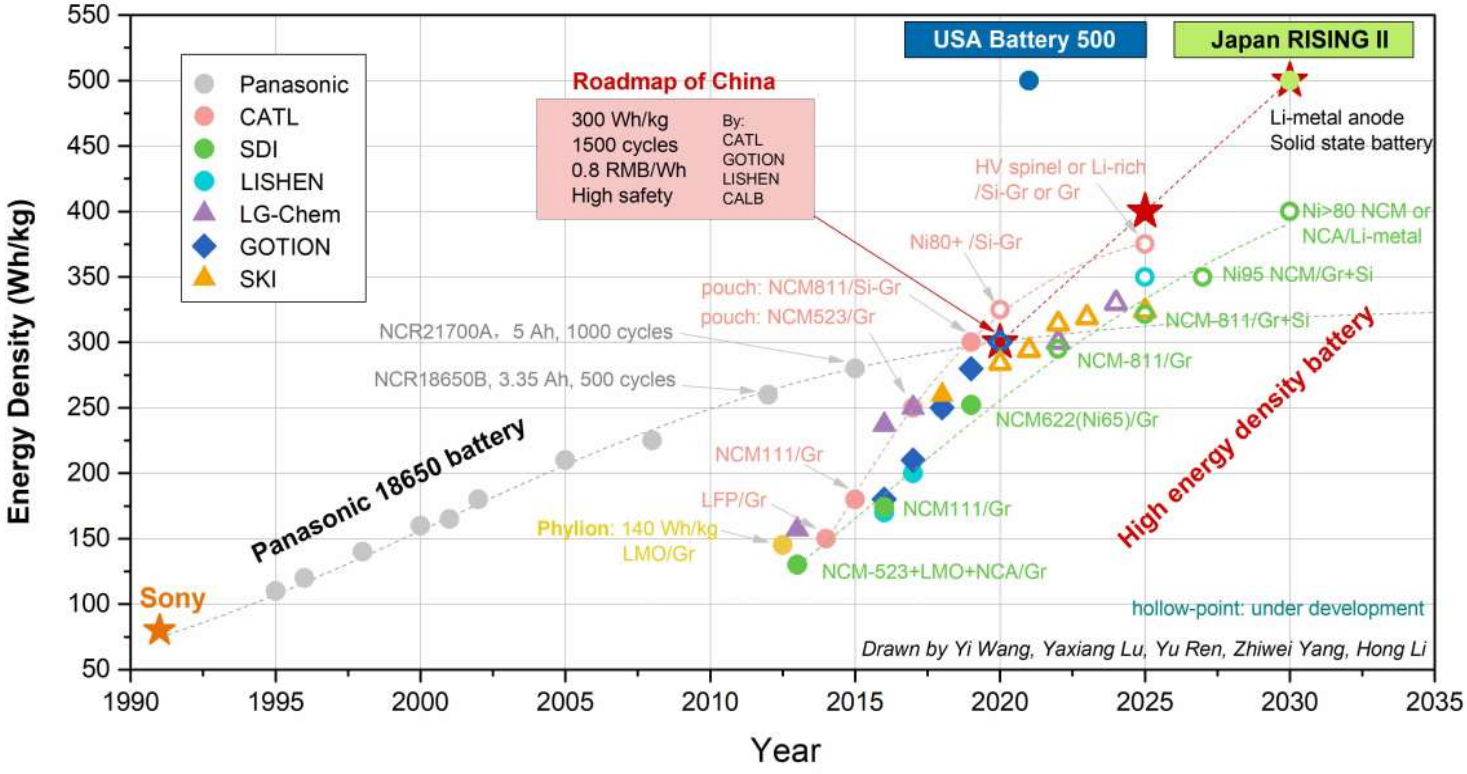
Societal Impact



Lithium-ion batteries are dominating the market

❖ But they are reaching their theoretical limits

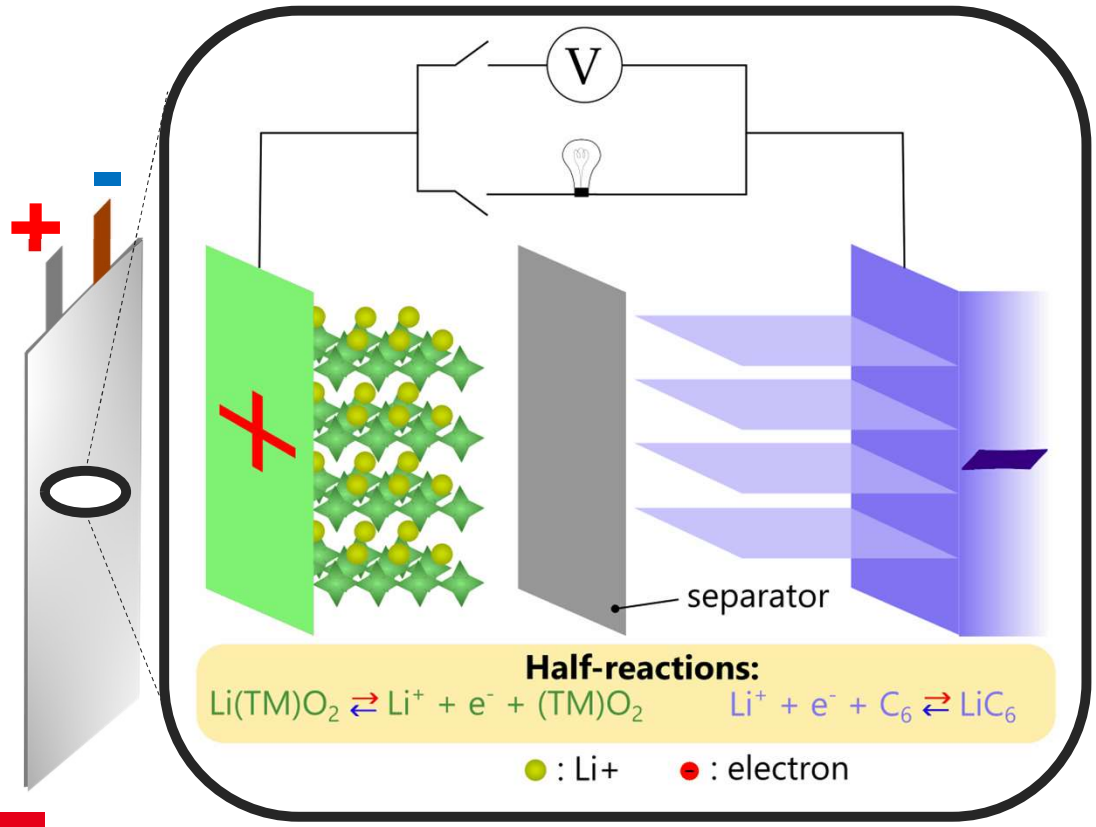
Gravimetric performance of batteries for automotive applications

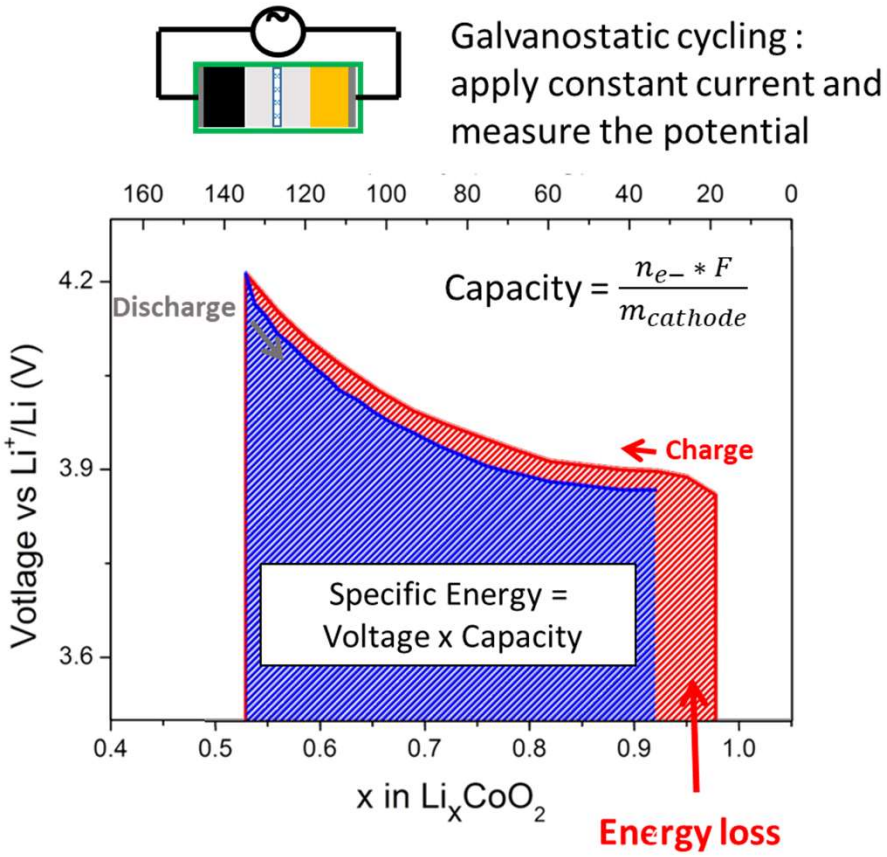


Adapted from Y. Lu, X. Rong, Y.-S. Hu, L. Chen, H. Li, *Energy Storage Mater.* **2019**, *23*, 144

Li-ion batteries, and beyond

A simple energy storage concept but a complex system governed by redox reactions

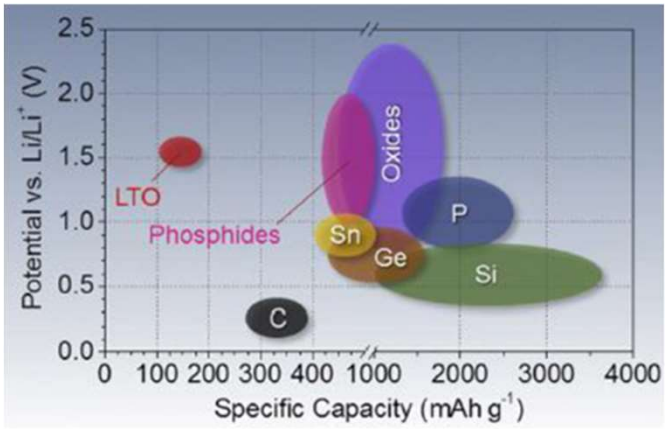




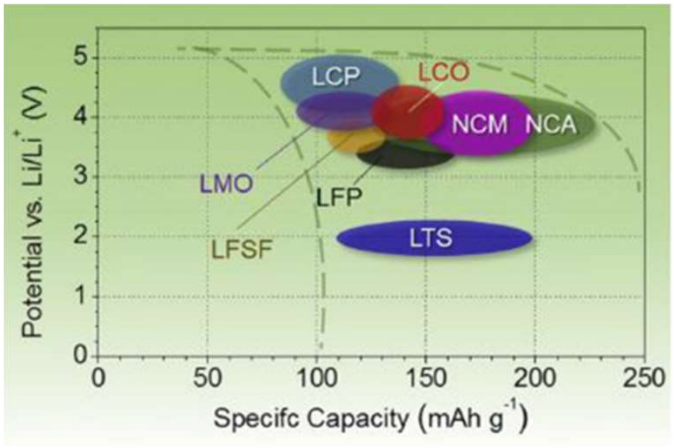
Li-ion batteries, and beyond

- Energy density is maximized by high voltage electrode materials and high specific capacity materials.

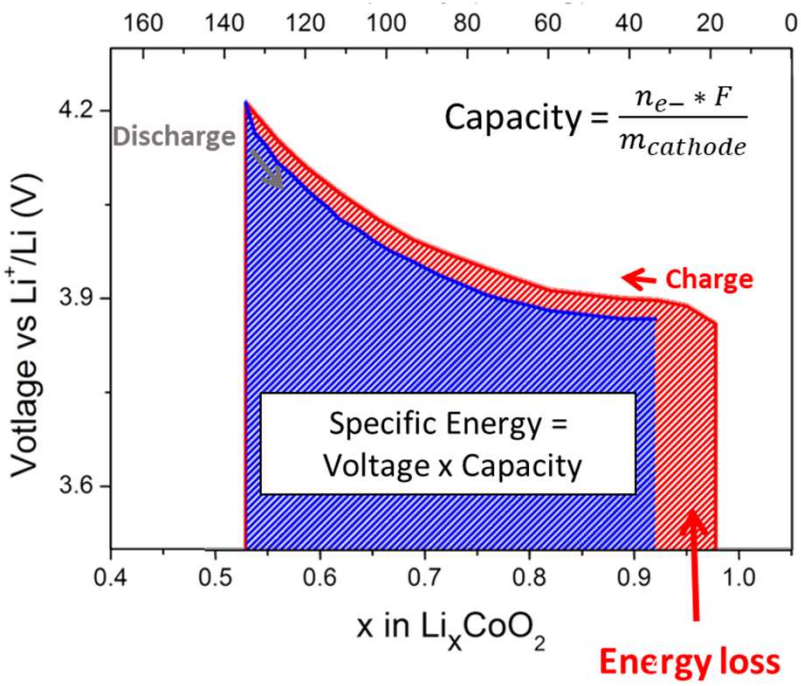
Common anode materials



Intercalation-type cathode materials



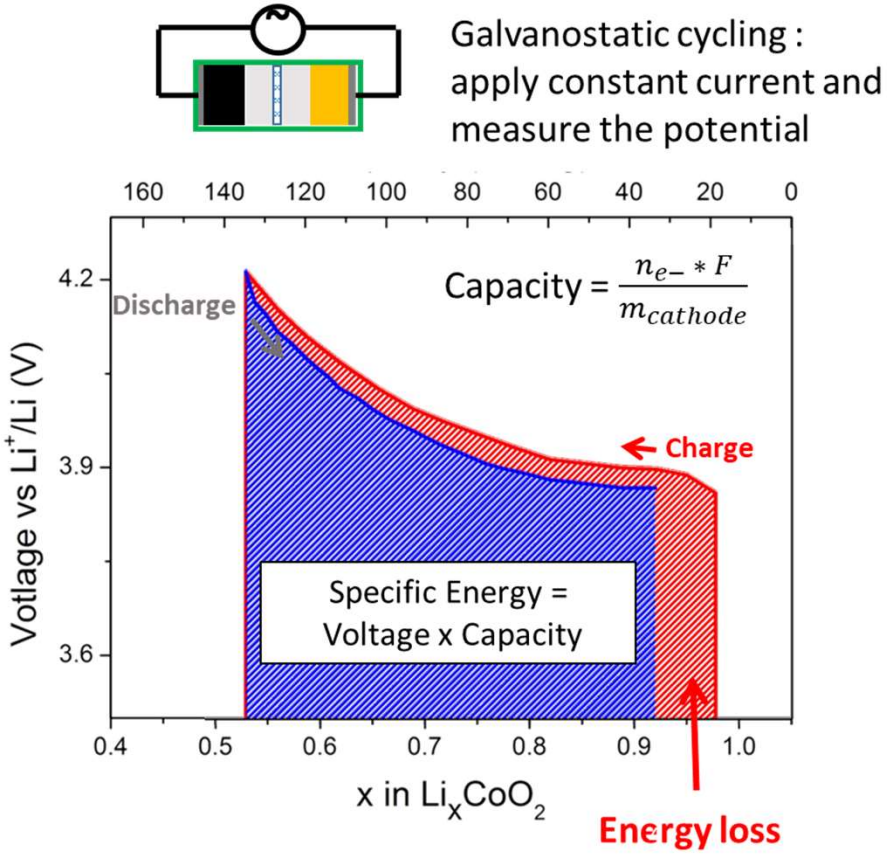
Galvanostatic cycling : apply constant current and measure the potential



Materials Today 18, 252-264 (2015)

Li-ion batteries, and beyond

- Energy density is maximized by high voltage electrode materials and high specific capacity materials.
- Energy density is not the only driver

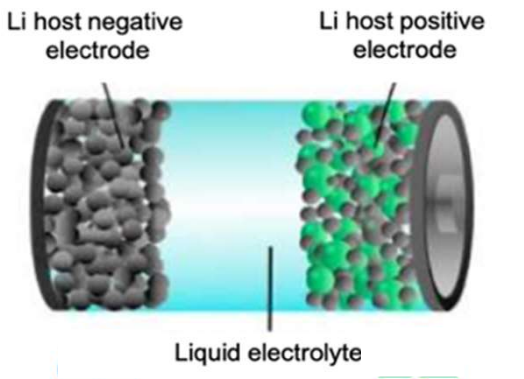


Sustainable Batteries—Quo Vadis? M-M. Titirici, *Adv. Energy Mater.* **2021**, *11*, 2003700

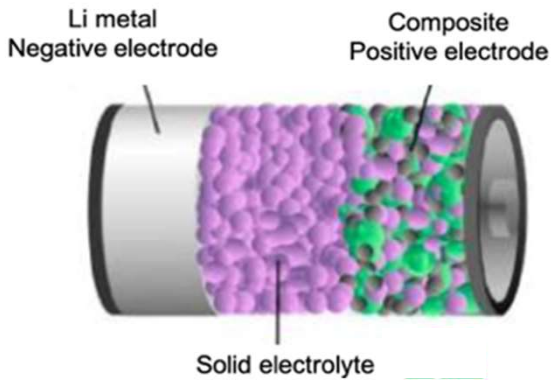
Li-ion batteries, and beyond

- Energy density is maximized by high voltage electrode materials and high specific capacity materials.
- Energy density is not the only driver
- Different types of batteries are needed

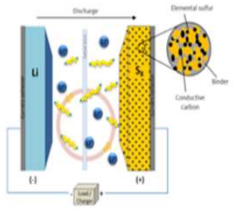
Liquid-type Li-ion and Na-ion batteries



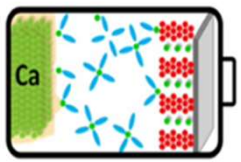
Solid-state batteries



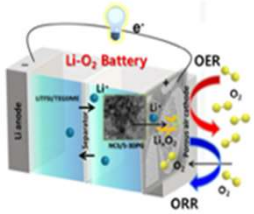
Next generation



Li-S

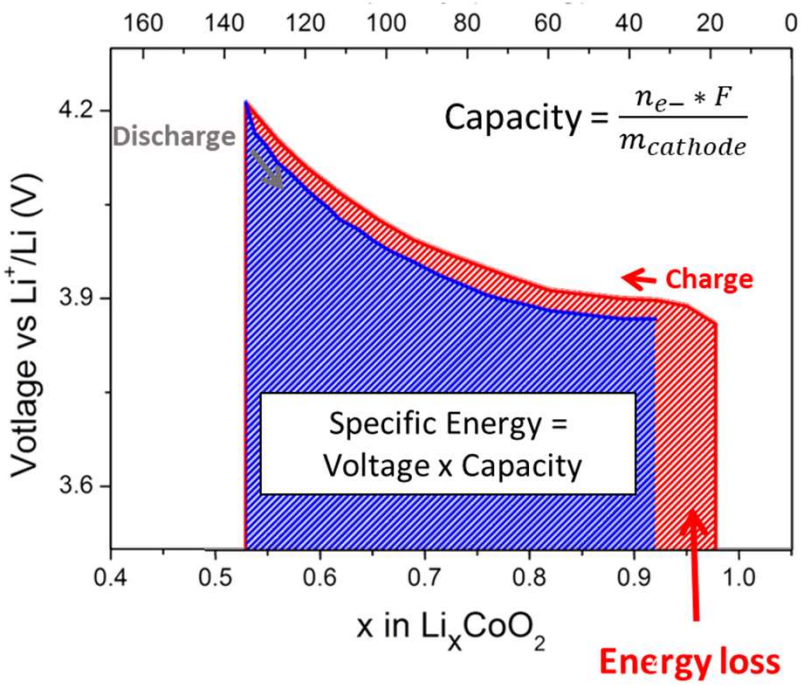


Multivalent



Li_2O_2

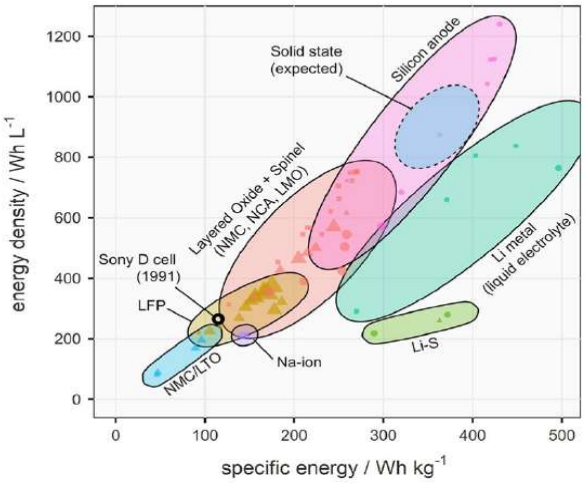
Galvanostatic cycling : apply constant current and measure the potential

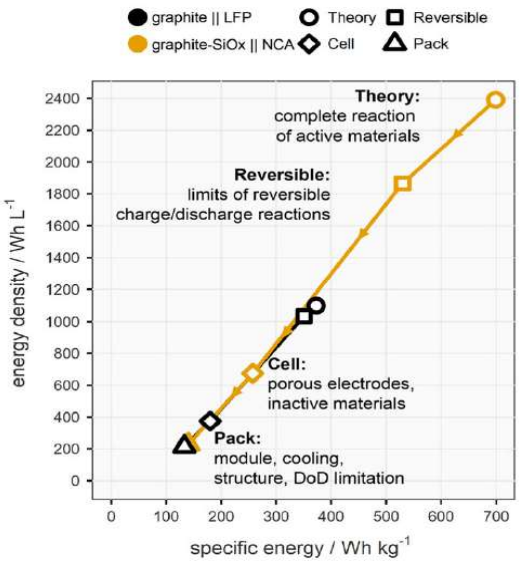


Li-ion batteries, and beyond

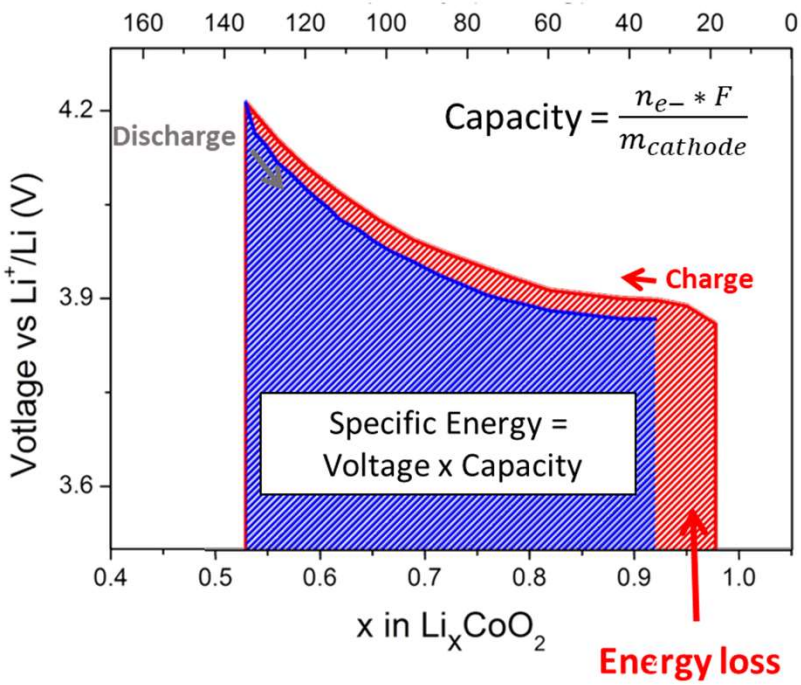
- Energy density is maximized by high voltage electrode materials and high specific capacity materials.
- Energy density is not the only driver
- Different types of batteries are needed

■ Cylindrical ● Pouch ▲ Prismatic





Galvanostatic cycling : apply constant current and measure the potential

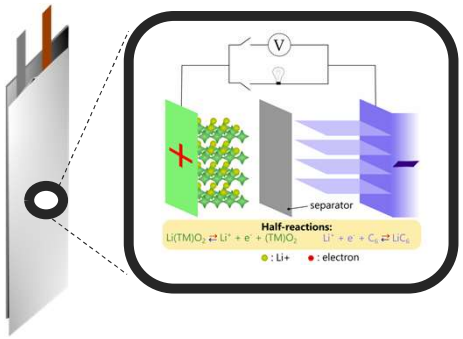


A non-academic perspective on the future of lithium-based batteries, J. T. Frith, M. J. Lacey, U. Ulissi, *Nature Communications,* **14**, 420 (2023)

Major breakthroughs are needed

to invent and produce better batteries

Academic research



A Roadmap for Transforming Research to Invent the Batteries of the Future Designed within the European Large Scale Research Initiative BATTERY 2030+.

Julia Amici, Pietro Asinari, Elixabete Ayerbe, Philippe Barboux, Pascale Bayle-Guillemaud, R. Jürgen Behm, Maitane Berecibar, Erik Berg, Arghya Bhowmik, Silvia Bodoardo, Ivano E. Castelli, Isidora Cekic-Laskovic, Rune Christensen, Simon Clark, Ralf Diehm, Robert Dominko, Maximilian Fichtner, Alejandro A. Franco, Alexis Grimaud, Nicolas Guillet, Maria Hahlin, Sarah Hartmann, Vincent Heiries, Kersti Hermansson, Andreas Heuer, Saibal Jana, Lara Jabbour, Josef Kallo, Arnulf Latz, Henning Lorrmann, Ole Martin Løvvik, Sandrine Lyonnard, Marcel Meeus, Elie Paillard, Simon Perraud, Tobias Placke, Christian Punckt, Olivier Raccurt, Janna Ruhland, Edel Sheridan, Helge Stein, Jean-Marie Tarascon, Victor Trapp, Tejs Vegge, Marcel Weil, Wolfgang Wenzel, Martin Winter, Andreas Wolf, Kristina Edström

Advanced Energy Materials, 2022, 12, 2102785https://doi.org/10.1002/aenm.202102785

- → Increase performance and durability
- → Optimize materials and manufacturing
- → Enable fast charging
- → Recycling, Second life
- → Develop digital twins
- → Enhanced Battery Management System

Key research questions

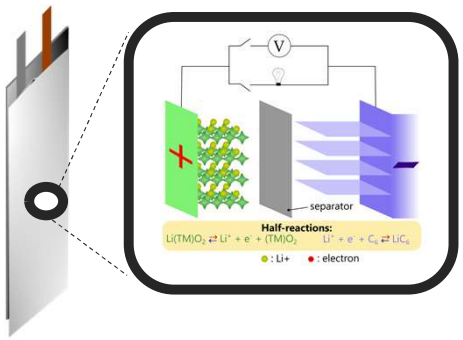
- → Understand **redox and ionic transport** mechanisms
- → Understand the role of **interfaces** and interfacial processes
- → Investigate **degradation** modes
- → Unravel the **roots of aging** and cell failure

- → Measuring reaction heterogeneities & species distributions
- → Tracking new phases, phase transitions, degradation products
- → ……

Major breakthroughs are needed

to invent and produce better batteries

Academic research



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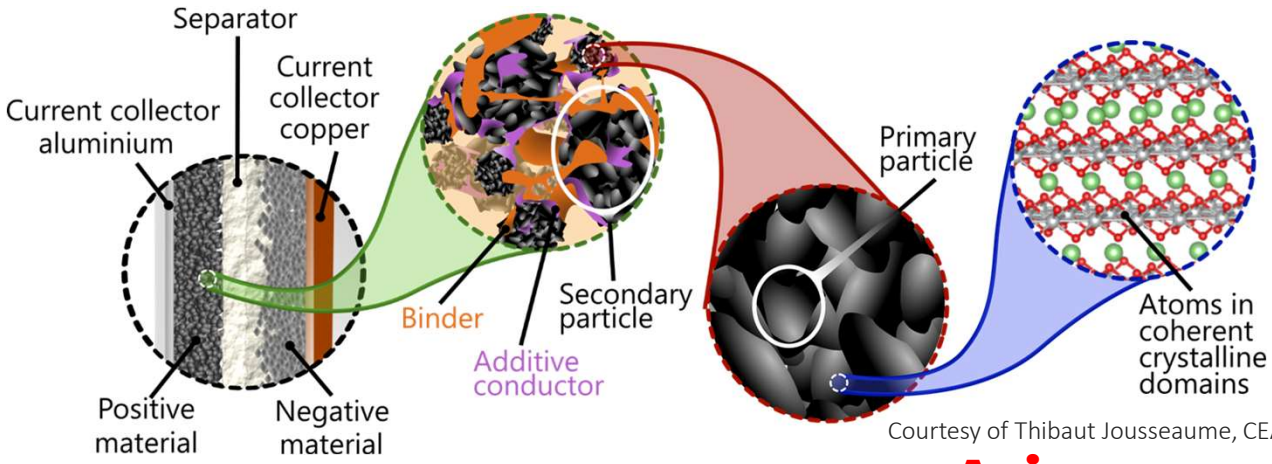
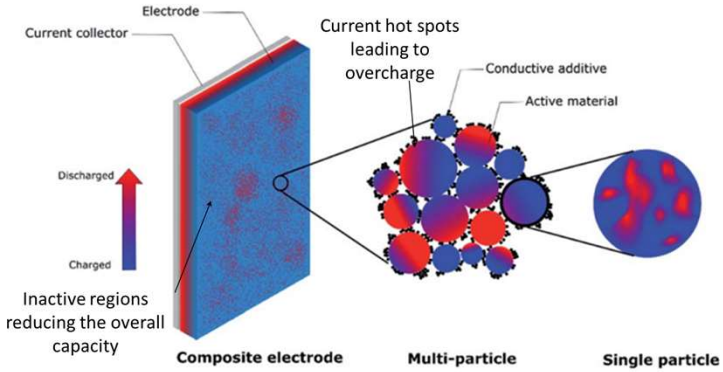
Multi-scale

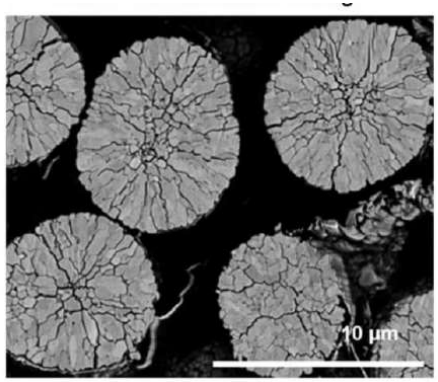


Courtesy of Thibaut Jousseaume, CEA

Heterogeneous

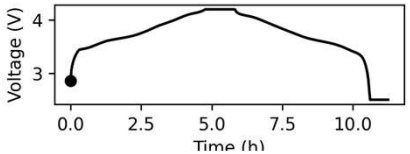


Aging

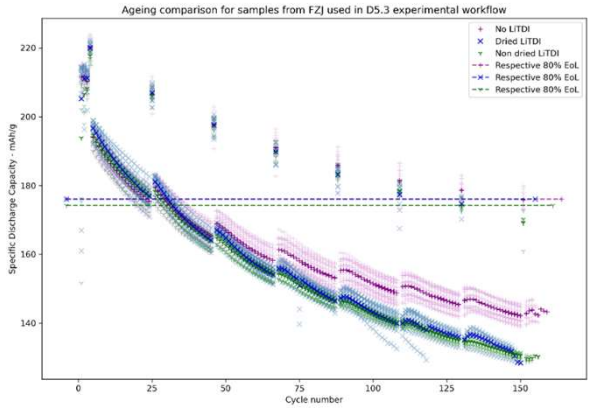


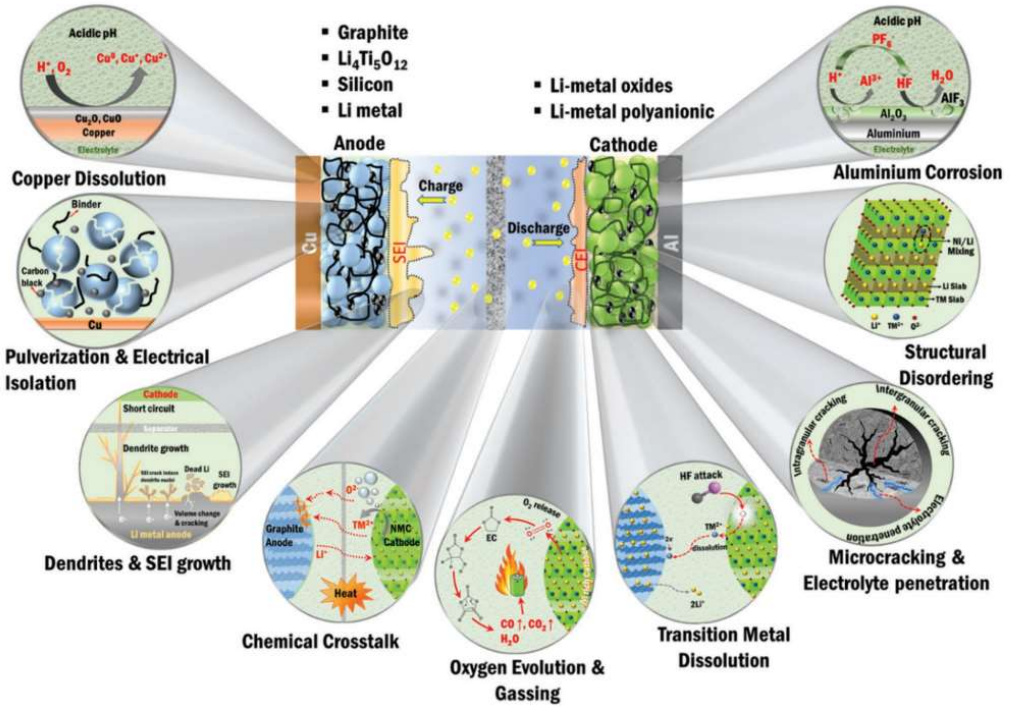
Advanced materials & device characterization

Capturing the dynamics of the system



Electrochemical bevahior





Narayan, R., Laberty-Robert, C., Pelta, J., Tarascon, J.-M., Dominko, R., Self-Healing: An Emerging Technology for Next-Generation Smart Batteries. *Adv. Energy Mater.* 2022, 12, 2102652.

Advanced materials & device characterization

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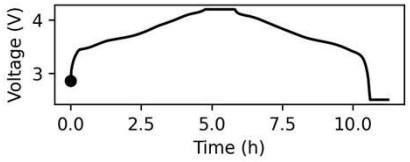
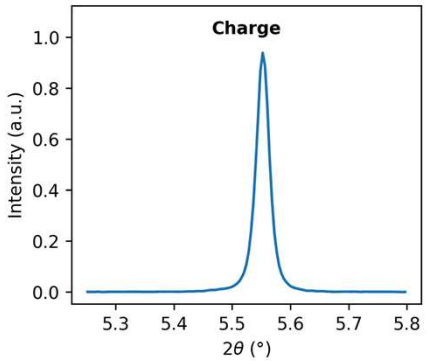
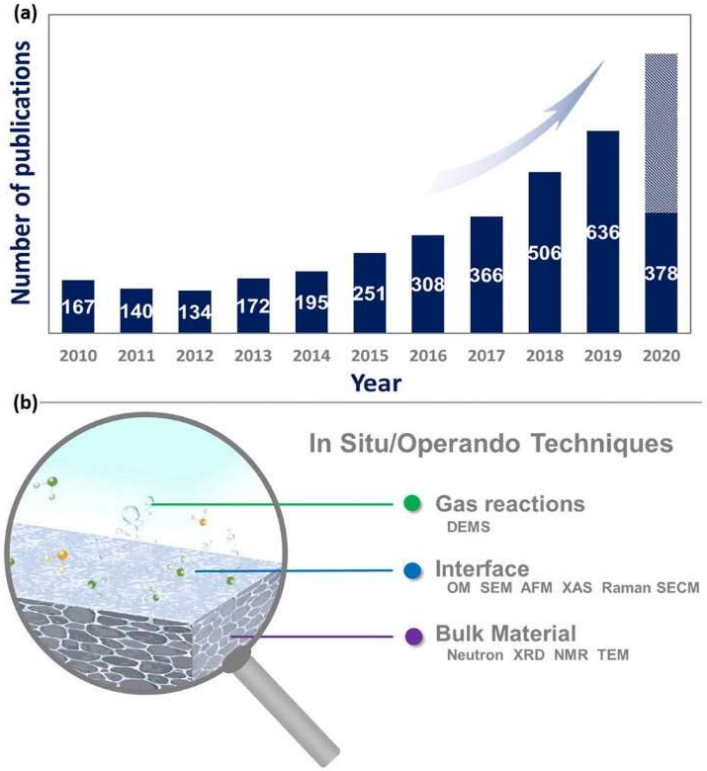


Electrochemical bevahior



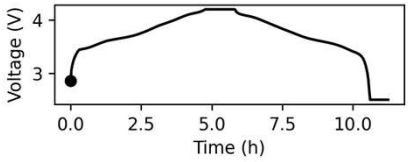
Property evolution

In-situ/operando characterization techniques in lithium-ion batteries and beyond
Journal of Energy Chemistry 59 (2021) 191–211

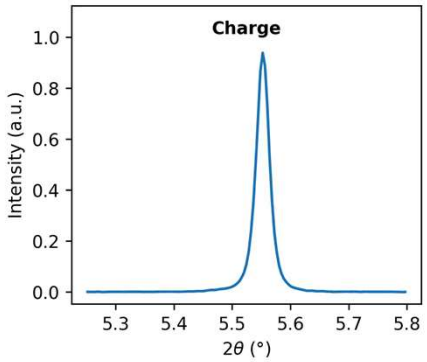


Advanced materials & device characterization

Capturing the dynamics of the system

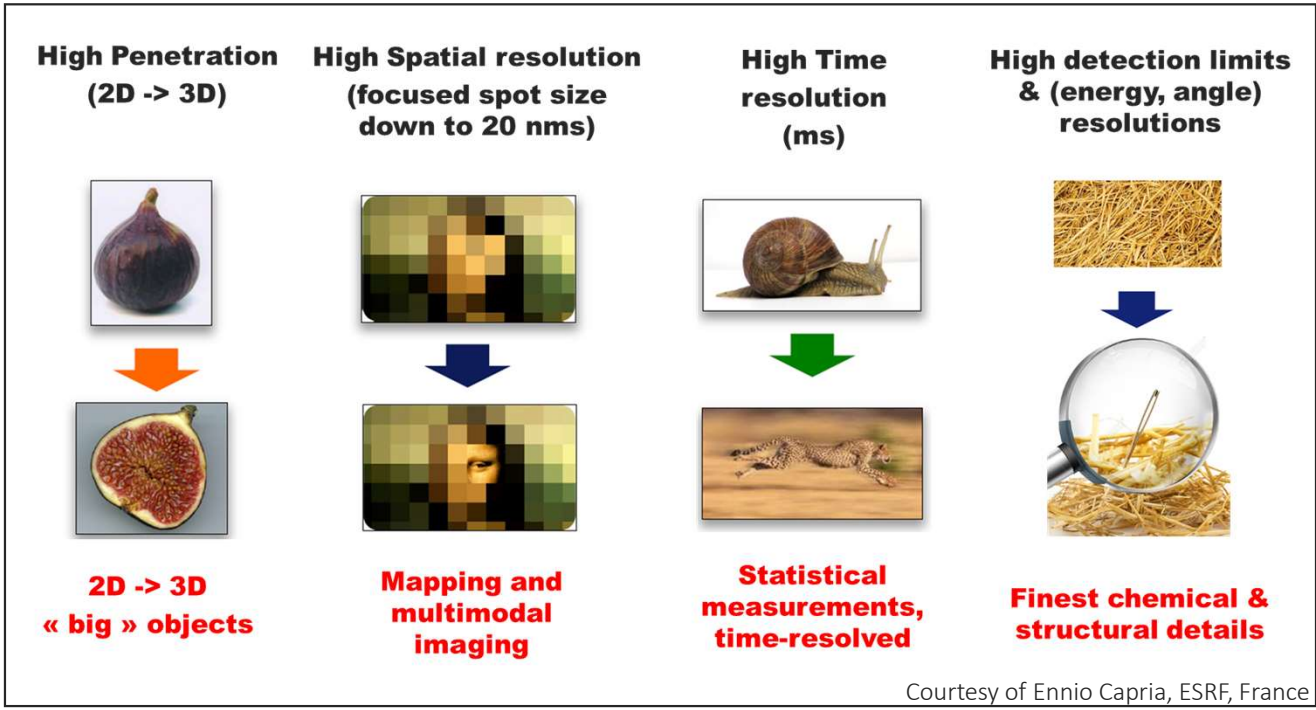


Electrochemical bevahior



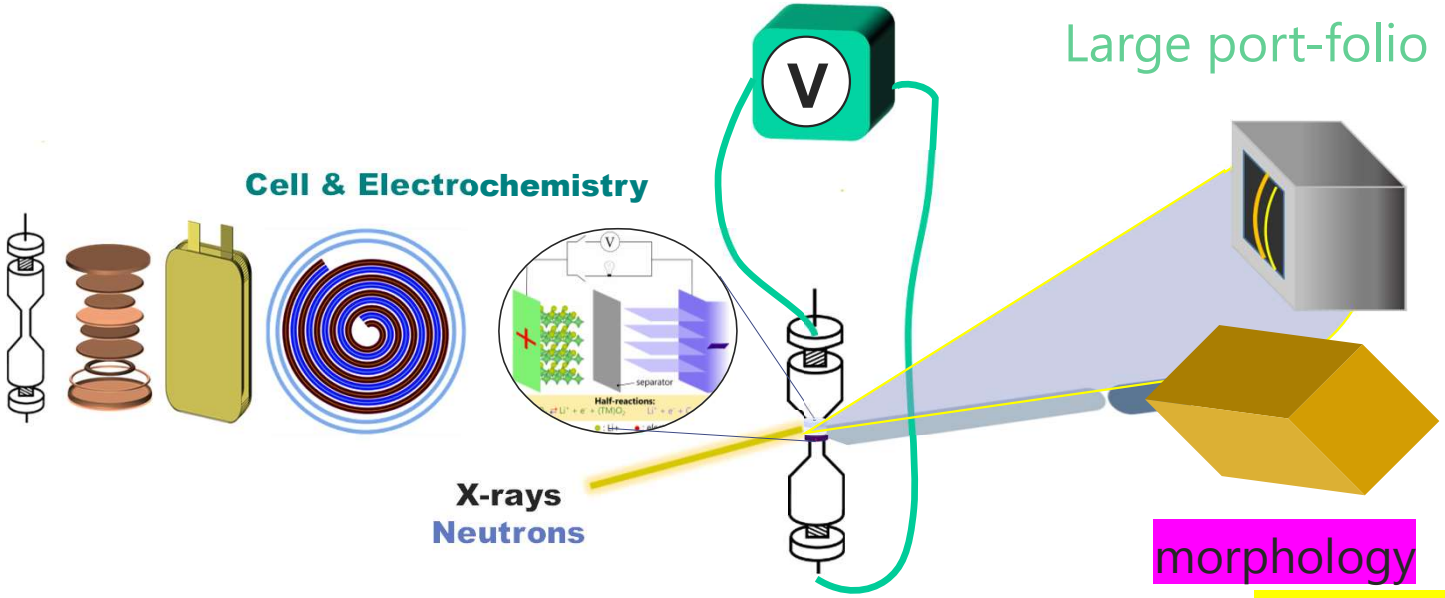
Property evolution

The advantages of neutron & synchrotron techniques



Courtesy of Ennio Capria, ESRF, France

Advanced materials & device characterization



Large port-folio of X / N techniques !

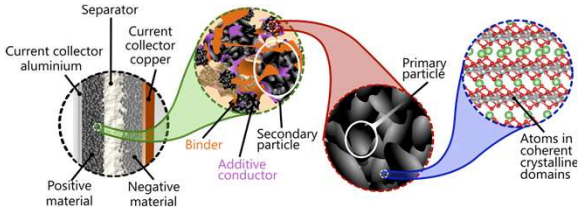
Data
- Spectra
- Patterns
- Images

XAS	(μ)XRD
XANES	μLaue
XES	SAXS
XRS	SWAXS
RIXS	BCDI
XRF	μCT
XPS	nCT
HAXPES	XRD-CT
XTM	SWAXS-CT
...	...
NPD	NI, NDP
SANS	NR, QENS
NPDF	INS

morphology

chemistry

structure



Correlative ex situ & operando characterization for holistic understanding

Capturing heterogeneities : fast spatially-resolved mappings of key properties

Reliable, **representative** and **reproducible** battery cells

Bridge scales from particle to electrode to device level informations !

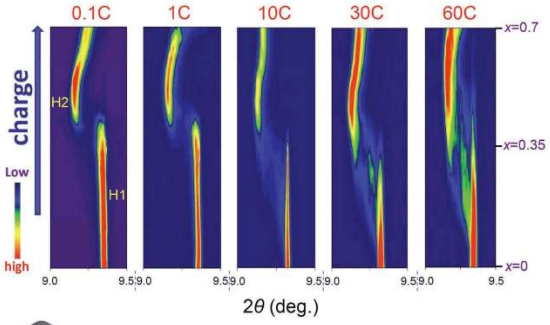
Synchrotron (& neutron) techniques

Sensitivity/Selectivity

Resolving materials transformations

Fast time-resolved in situ X-ray diffraction

High rate induced overpotential is thought to be the driving force for the formation of intermediate Li-poor phase.

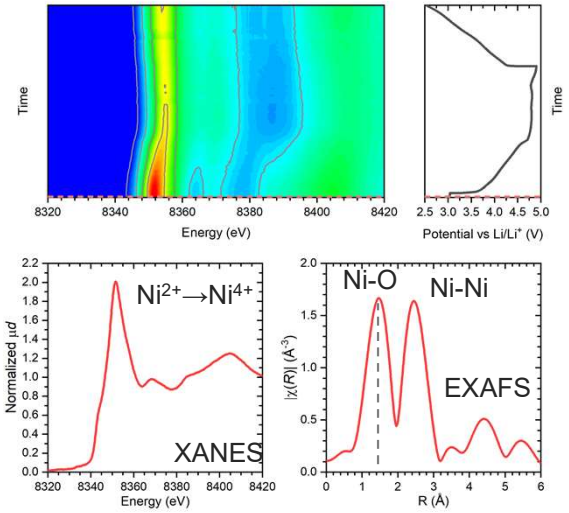


003 diffraction peak of $Li_{1-x}Ni_{1/3}Co_1/3Mn_{1/3}O_2$ with increasing x between x = 0 and 0.7 during the first charge process at different C rates

Zhou, Y. N. et al. High-rate charging induced intermediate phases and structural changes of layer-structured cathode for lithium-ion batteries. Adv. Energy Mater. 6, 1600597 (2016). DOI:10.1002/aenm.201600597

Tracking redox reactions

Operando multi-edge X-ray absorption



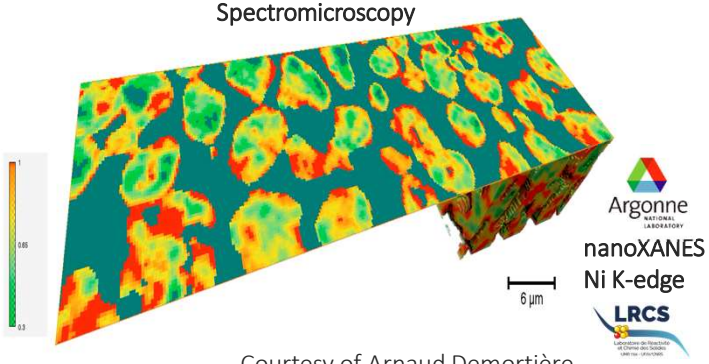
Ni oxidation and concomitant contraction of Ni-O shell

O.Usoltsev et al. Operando Multi-edge XAS to Reveal the Effect of Co in Li- and Mn-rich NMC Li-ion Cathodes, Materials Today Energy, 2025
DOI: 10.1016/j.mtener.2025.101853

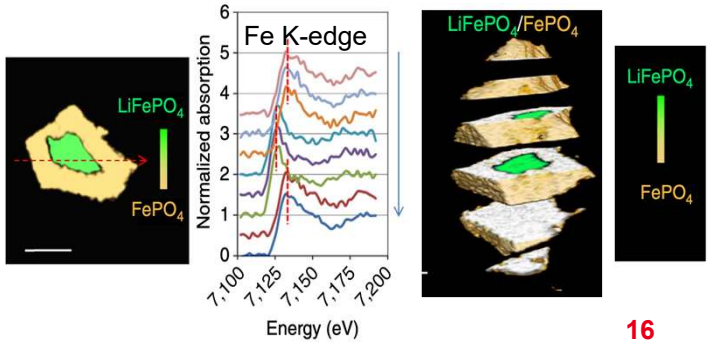
Courtesy of Laura Simonelli

2D/3D mapping of species/phase distribution

Spectromicroscopy



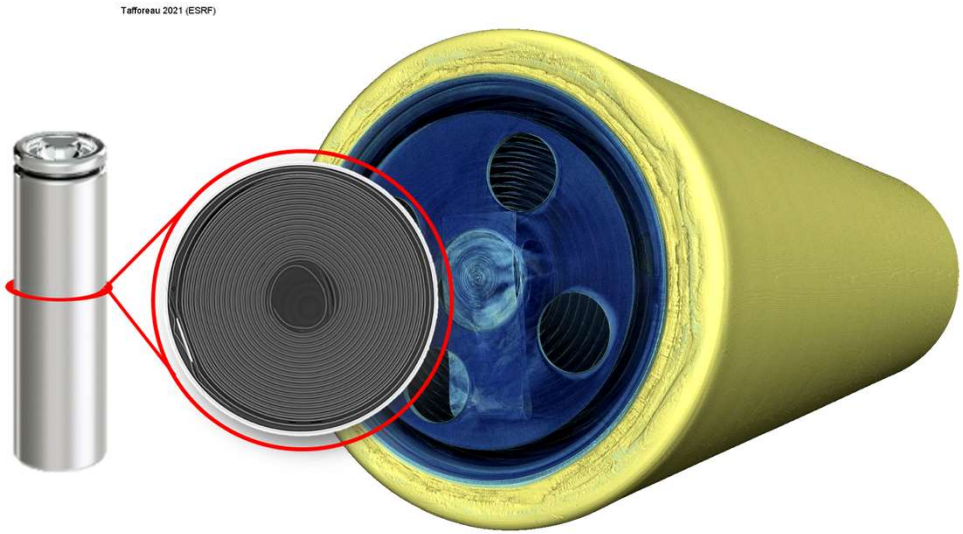
Courtesy of Arnaud Demortière



J. Wang et al., Nature Comm., 7, 12372, 2016

Imaging large format batteries

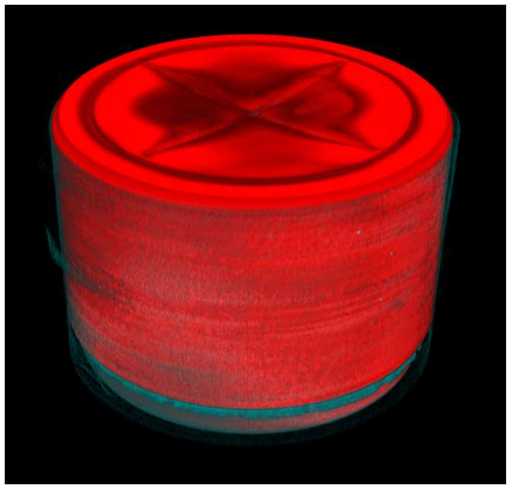
X-ray micro-tomography on Li-ion 18650 battery

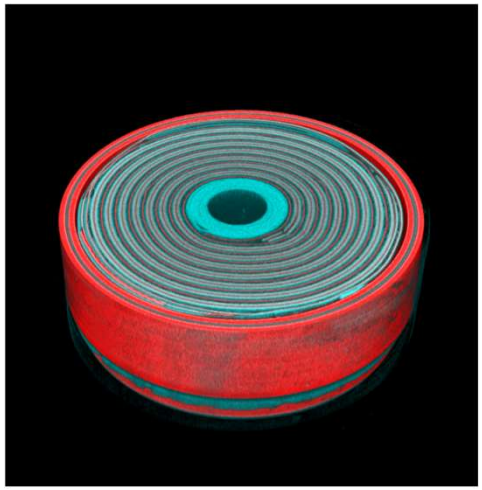


ESRF
The European Synchrotron

Courtesy of BM05 staff at ESRF, Paul Tafforeau and Franhofer EZRT for the images.

Neutron Tomography on small cylindrical cells combined with μCT





X-ray attenuating (metals)

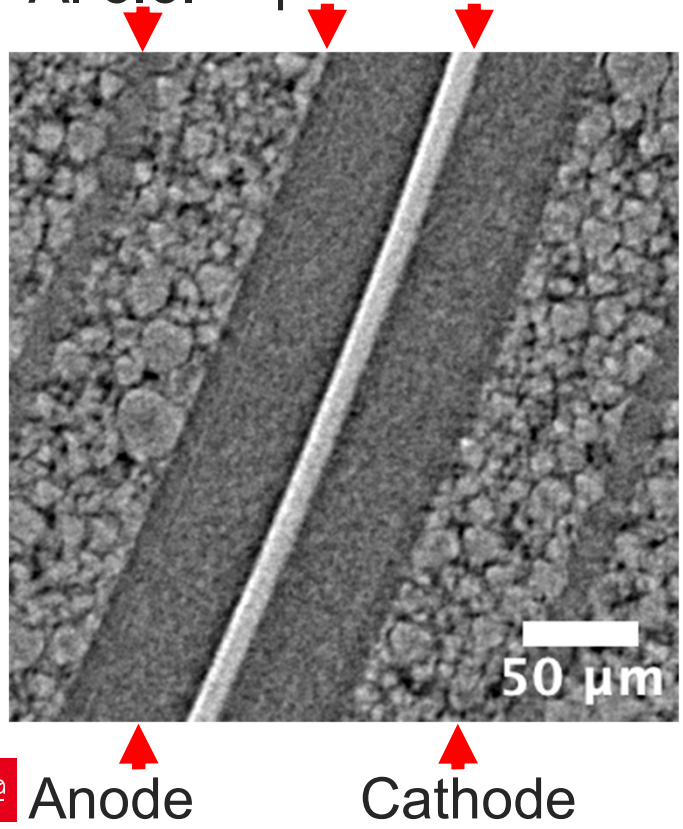
Neutron attenuating (Li, H)

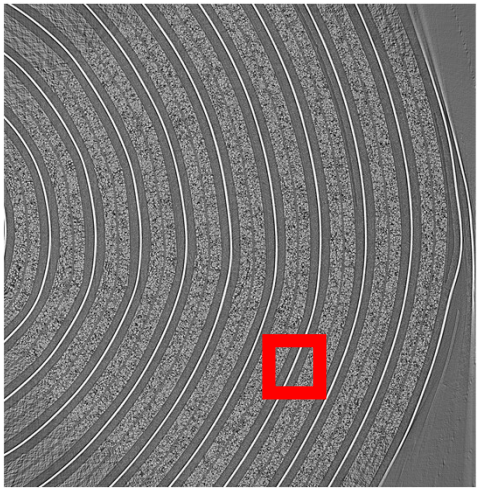
NEUTRONS FOR SOCIETY

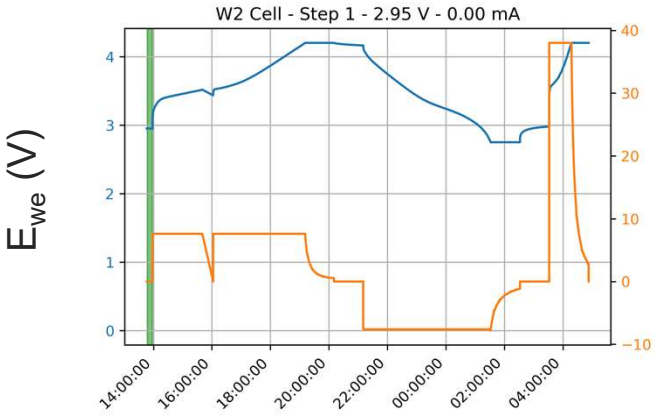
E. Lübke et al, Energy Environ. Sci. **17**, 5048-5059 (2024).

Imaging large format batteries

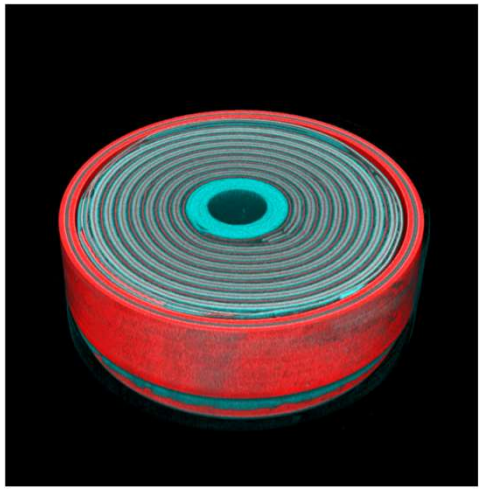
High resolution phase-contrast x-ray tomography BM05 at ESRF







Neutron Tomography on small cylindrical cells combined with µCT

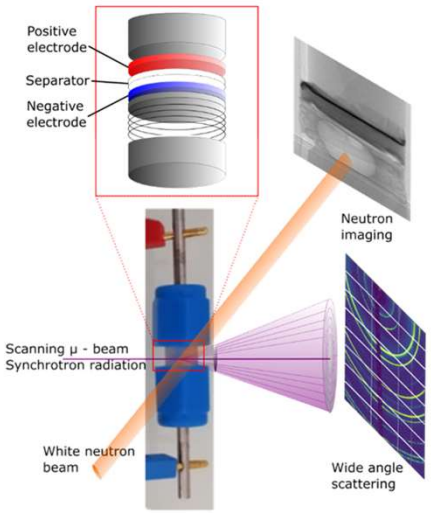


E. Lübke et al, Energy Environ. Sci. **17**, 5048-5059 (2024)
E. Lübke et al, in review.

Imaging custom batteries

Swagelok-type
custom cell

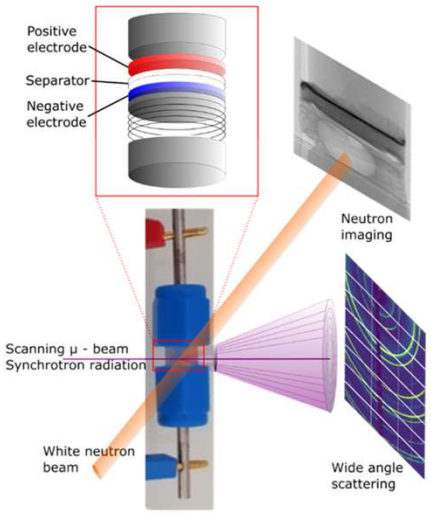
Diameter = 1 to 3 mm



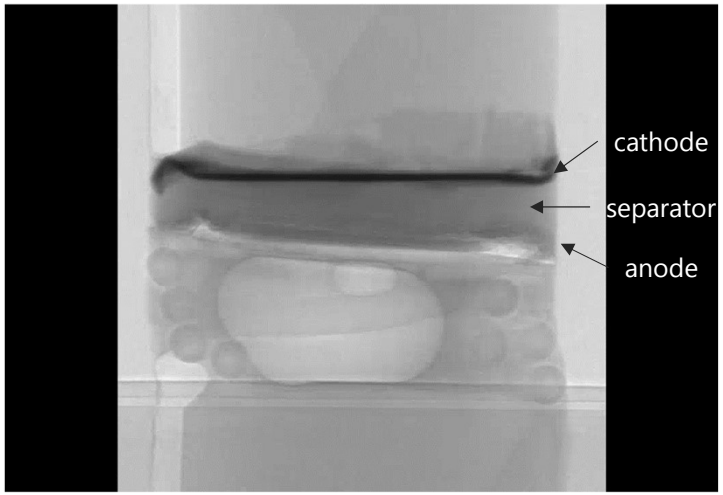
Imaging custom batteries

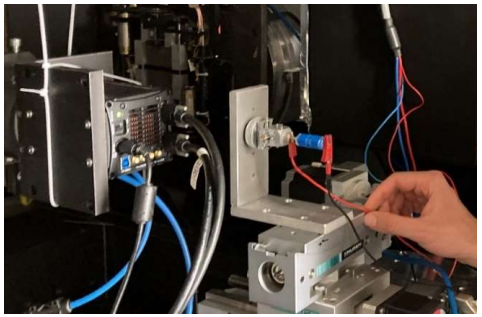
Swagelok-type
custom cell

Diameter = 1 to 3 mm



Neutron Radiography
Sensitive to ^{6}Li concentration





NEUTRONS FOR SOCIETY

NeXT

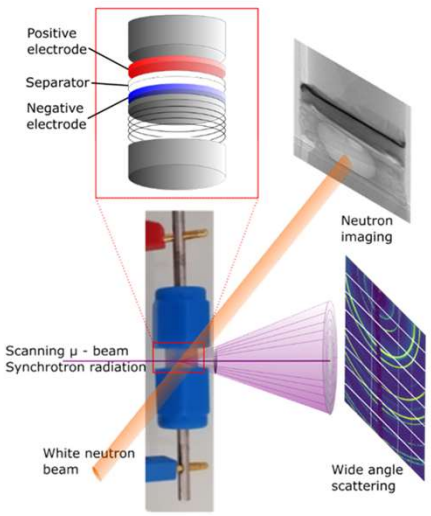
Pixel size : 2.7 μm
Spatial resolution : 10 μm
Time resolution : 30 seconds/frame

Oney et al, in preparation.

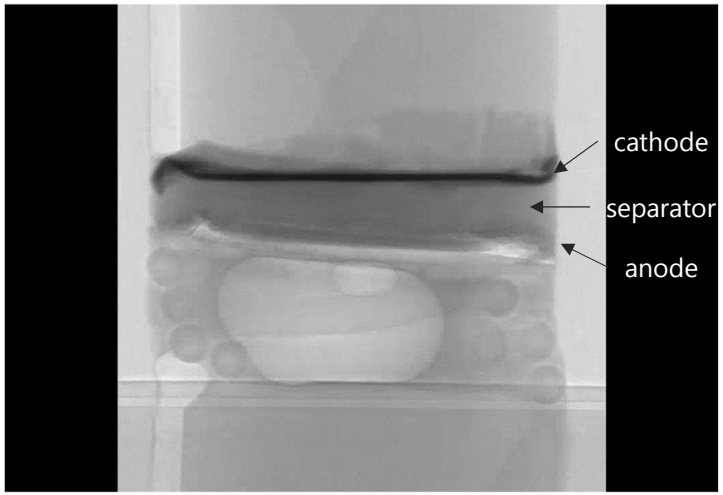
Imaging custom batteries

Swagelok-type
custom cell

Diameter = 1 to 3 mm



Neutron Radiography
Sensitive to ^{6}Li concentration

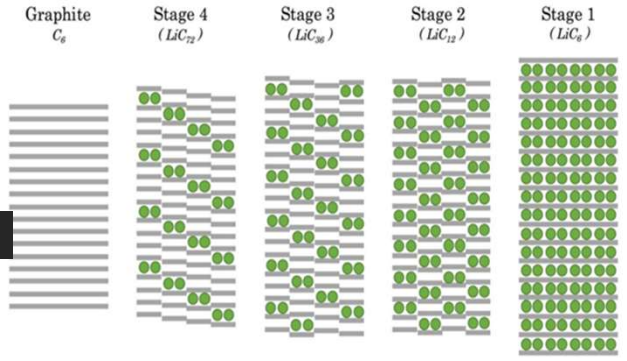


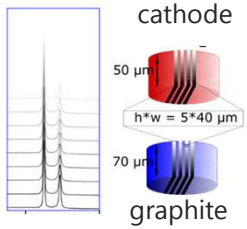
NEUTRONS FOR SOCIETY

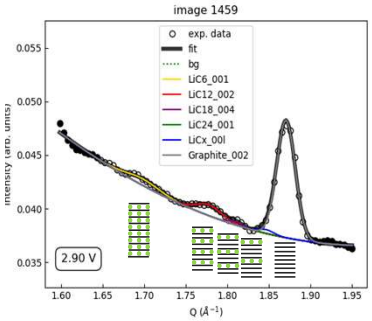
NeXT

Spatial resolution : 10 μm
Time resolution : 2 minutes

Scanning synchrotron microXRD
Lithium concentrations x in Li_xC_6





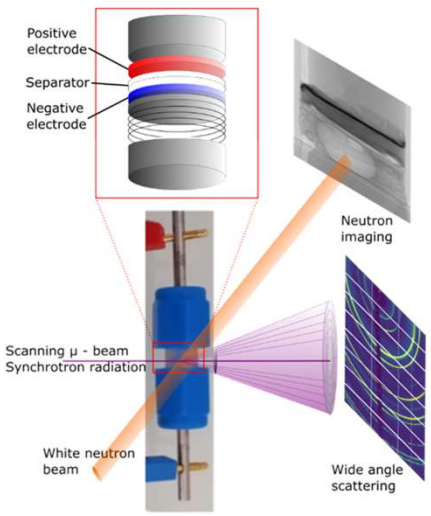


Oney et al, in preparation.

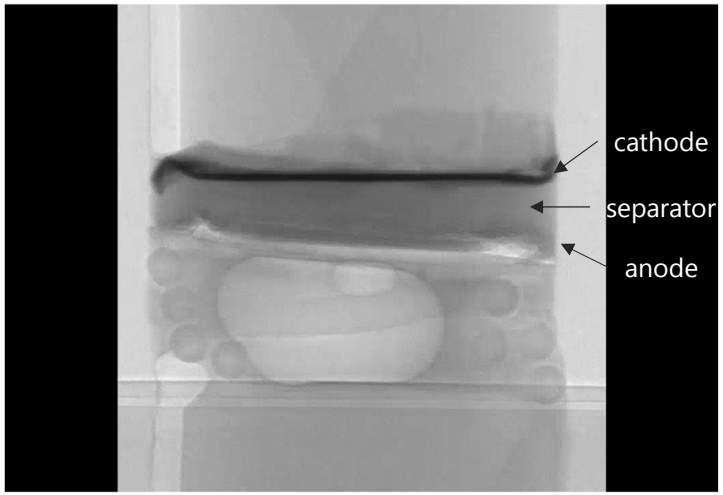
Imaging custom batteries

Swagelok-type
custom cell

Diameter = 1 to 3 mm



Neutron Radiography
Sensitive to ^{6}Li concentration



NEUTRONS FOR SOCIETY

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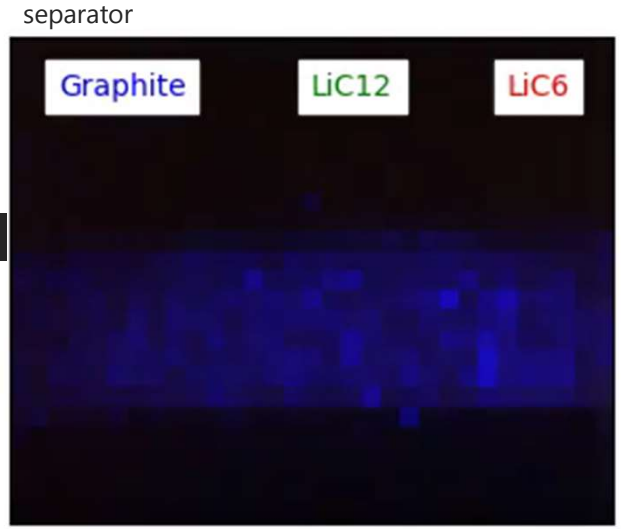


Courtesy of Samuel Tardif, CEA

Beam size (vertical) ~ 1 to 5 μm
Time resolution z-scans ≈ 1 min

Tardif et al, J. Mater. Chem. A, 2021, 9, 4281
Oney et al, Adv. Energy Materials, 2025, e02032

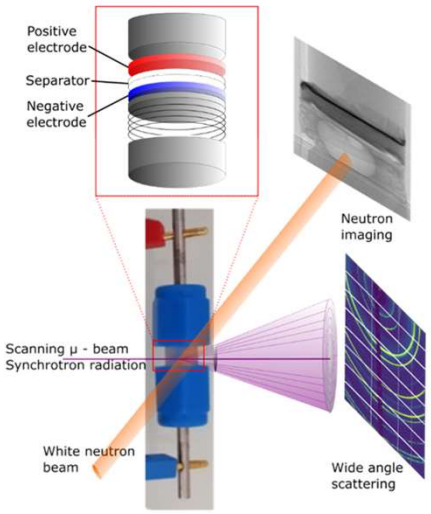
Imaging custom batteries

$$\mathrm{NAAD}(x_{\mathrm{Li}}) = \frac{1}{L}\int_{z=0}^{z=L}\frac{|x_{Li}(z) - \langle x_{\mathrm{Li}}\rangle_z|}{\langle x_{\mathrm{Li}}\rangle_z}dz$$

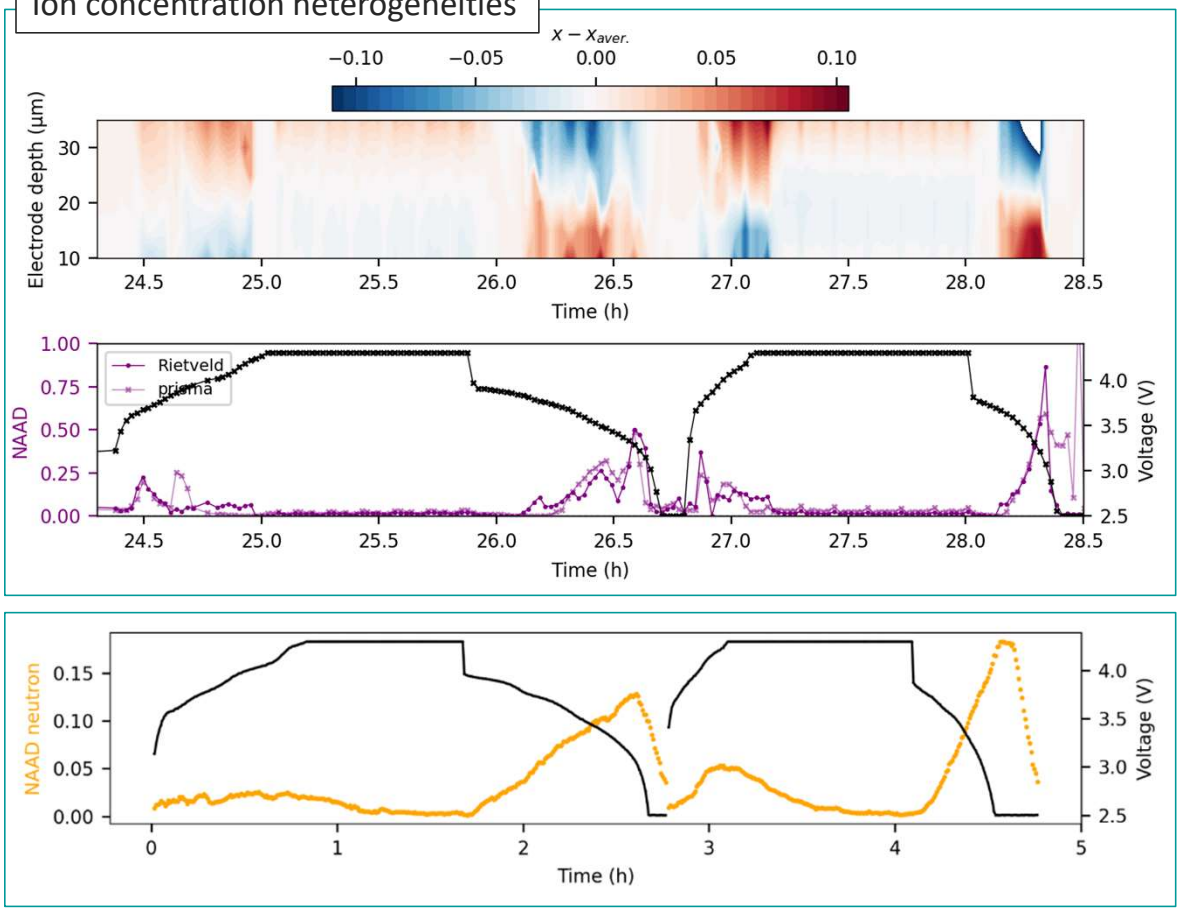
S. Tardif *et al*, J. Mat. Chem. A, 2021, 9, 4281-4290.

Swagelok-type custom cell

Diameter = 1 to 3 mm



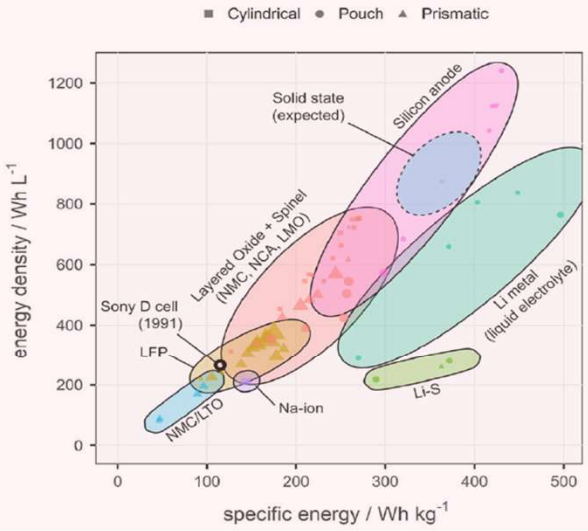
Ion concentration heterogeneities



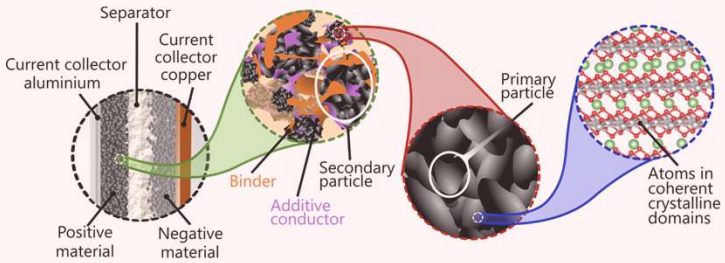
Oney, Jacquet, Helfen, Lyonnard et al, in prep.

Matching problems and solutions

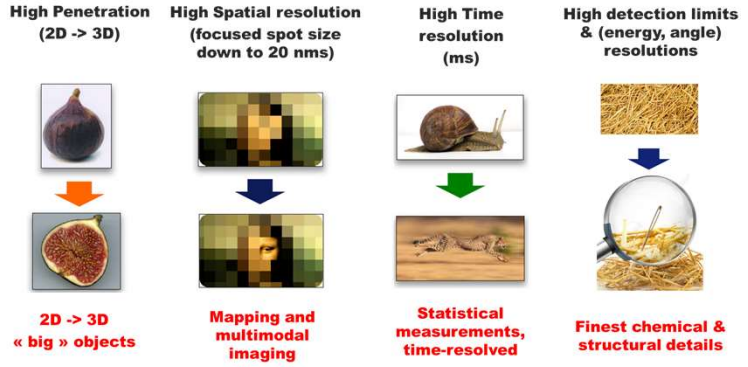
Many chemistries, materials and cells

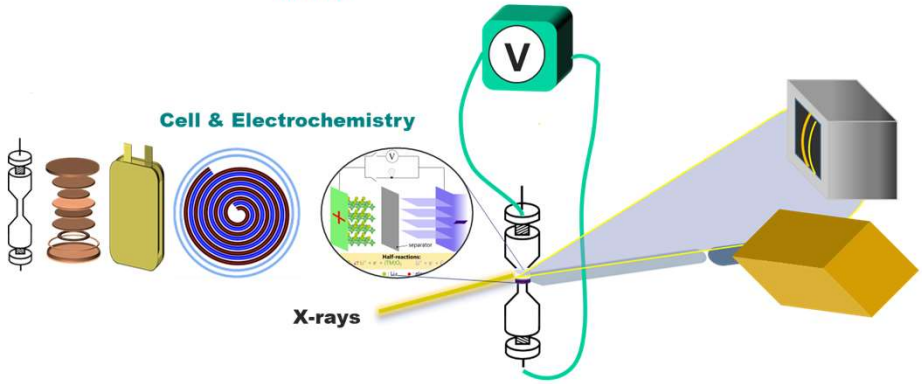


Multiscale processes (reaction, degradation)



X-rays and neutrons are great !
Many techniques are useful



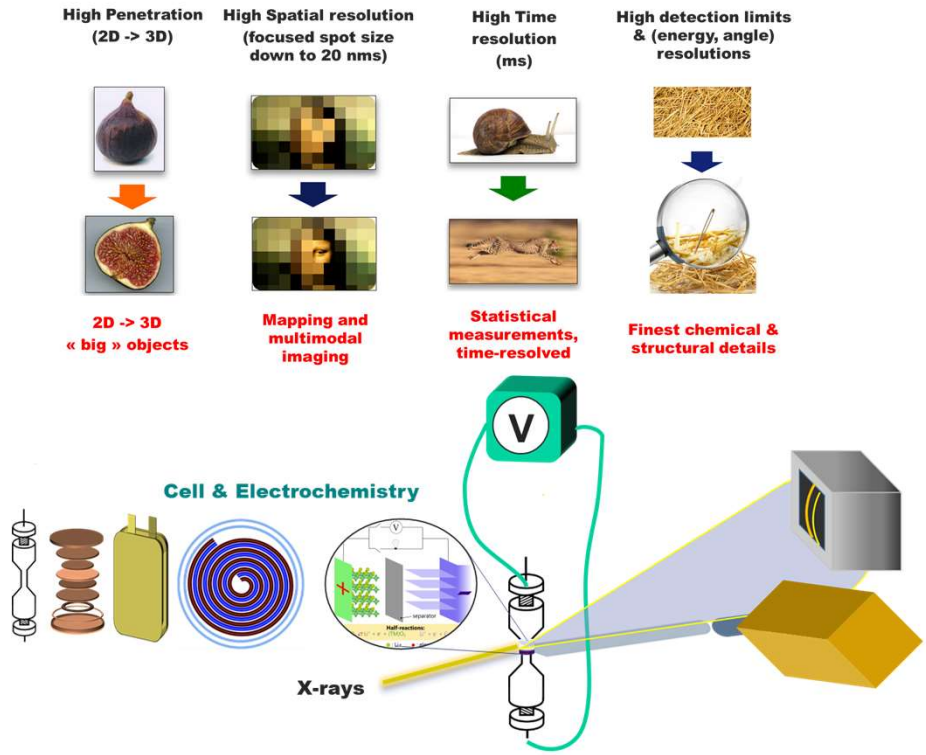


structure chemistry morphology

Matching problems and solutions

The case of silicon-based anodes

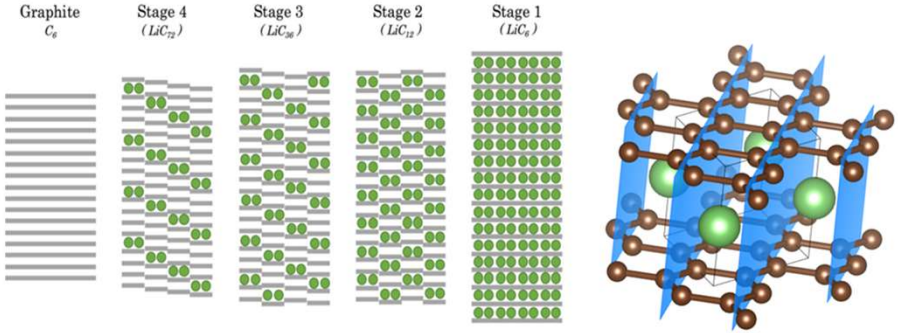
X-rays and neutrons are great !
Many techniques are useful



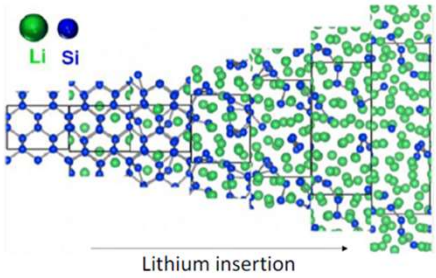
structure chemistry morphology

The case of silicon-based anodes

Graphite Intercalation mechanism



Silicon Alloying Mechanism

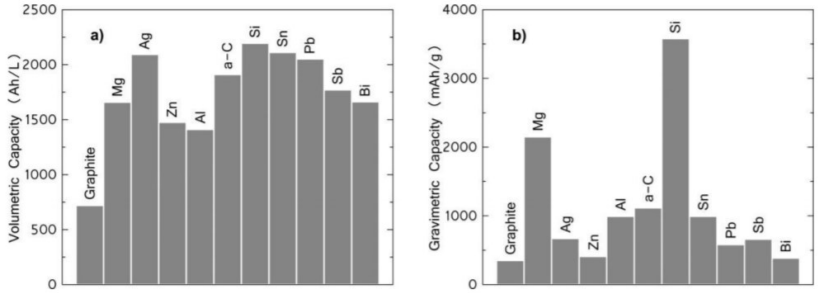


Si -> $Li_{3.75}Si$

Silicon has a very high specific capacity causing mechanical degradation of the electrode (volume changes, SEI, strain, lithium trapping)

J. Am. Chem. Soc. **134**, 14362–14374 (2012)

Si = best gravimetric and volumetric capacity



Anode	graphite	silicium
Theoretical specific capacity (mAh/g)	372	3579
Potential vs Li/Li+ (V)	0,1	0,4
Type	Intercalation	Alliage
Lithiated phases	LiC_6	$Li_{15}Si_4$

Obrovac, M. N. & Chevrier, V. L. *Chem. Rev.* **114**, 11444–11502 (2014).

The case of silicon-based anodes

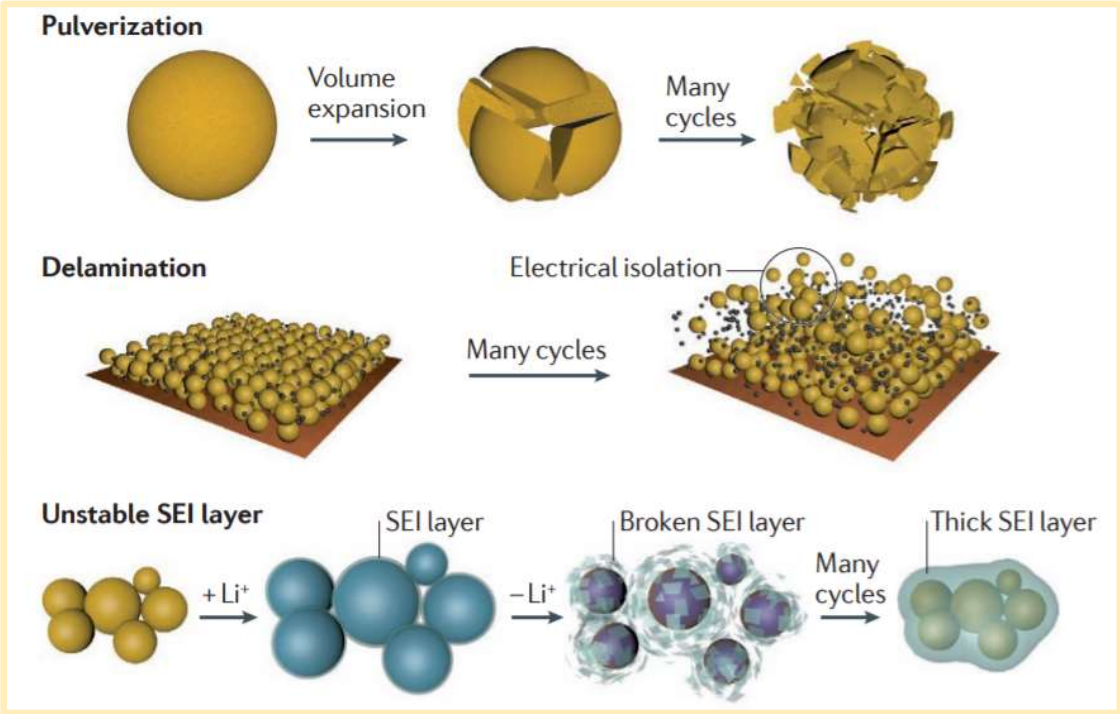
Increasing the amount of Si in « real » batteries is a challenge

How can we mitigate the enormous volume changes and irreversible degradations to enable a viable high silicon content-based high energy density Li-ion technology ?

How to access the SEI and Particles Morphology changes in real time at ultimate spatial scales and possibly inside a commercial cell using an advanced silicon anode ????

High first-cycle irreversible capacity +

Alloying process: **300%** volume expansion

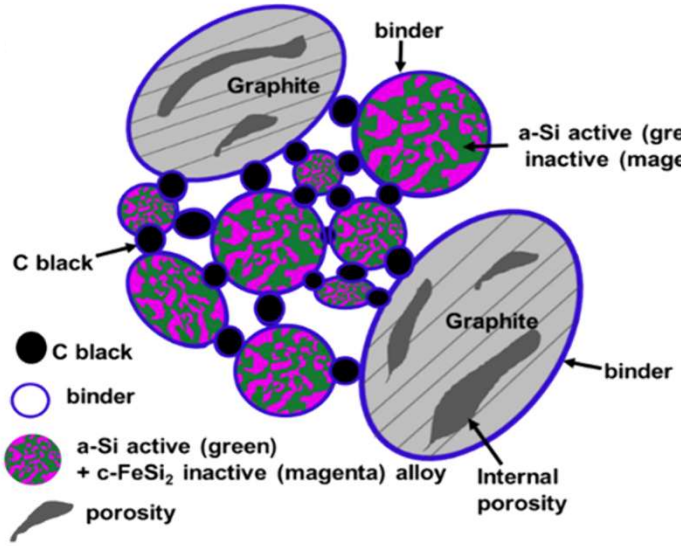


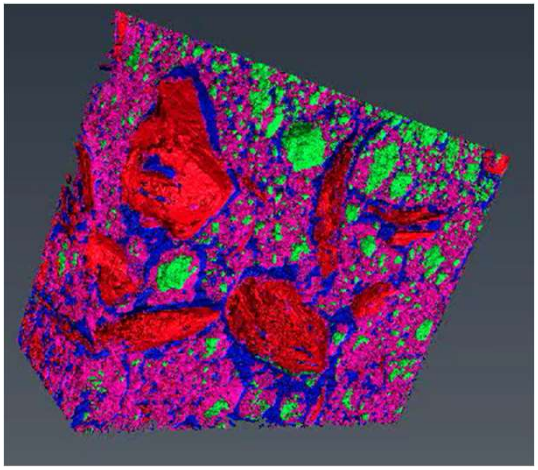
J. W. Choi, D. Aurbach, *Nature Reviews Materials*. **1**, 1–16 (2016).

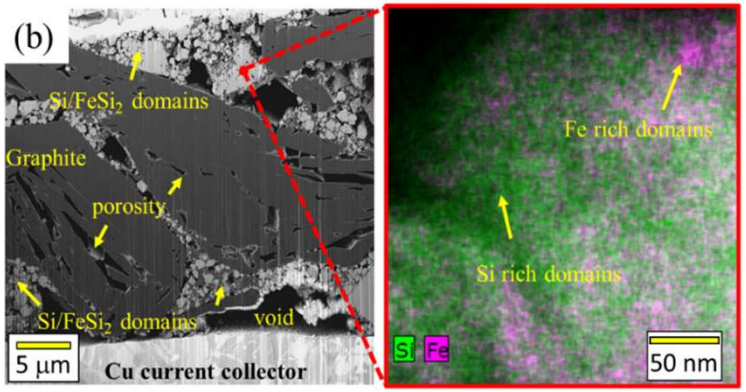
Graphite-Silicon composite anodes

hold promises to increase the energy density while maintaining long-term cyclability

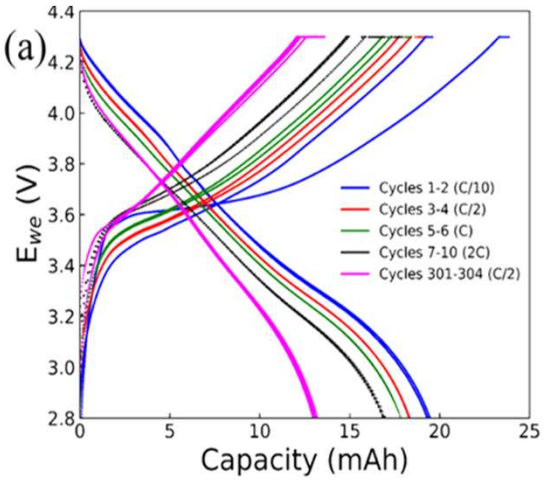
50% Si-comp, 41% graphite, 7%LiPAA, 2% Super P
Si-comp : 20% graphite, 25% a-Si, 55% c-FeSi2







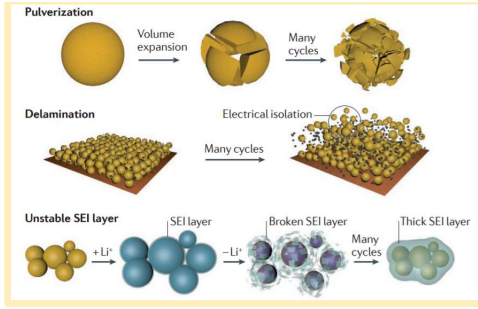
70% capacity loss after 300 cycles



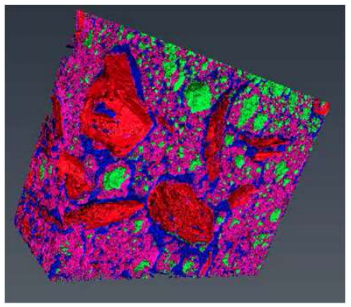
Kumar e*t al*, Small, **2020**, 16 (11), 1906812

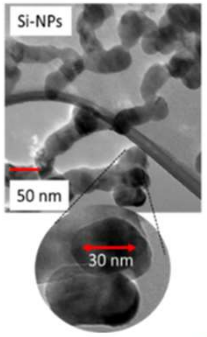
Graphite-Silicon composite anodes

structure chemistry morphology



Real Model

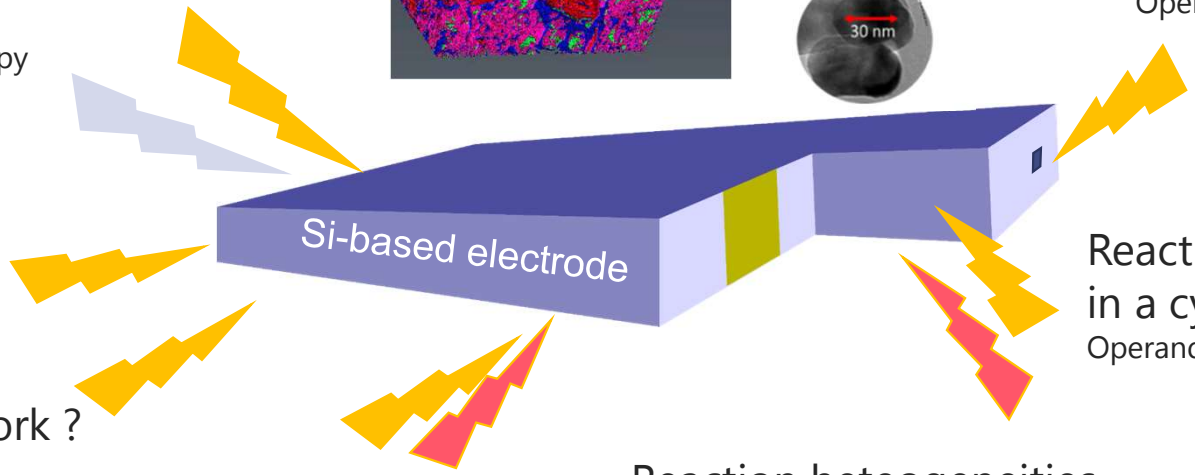




SEI growth and morphology ?
Operando X-ray Reflectivity

Electron microscopy
Lab spectroscopy

SEI composition ?
Ex situ X-ray Raman Scattering



Local particle/electrode scale activity ?
Operando nanoholotomography

Reaction heterogeneities in a cylindrical cell ?
Operando SWAXS-CT and NI

Porous network ?
MicroCT

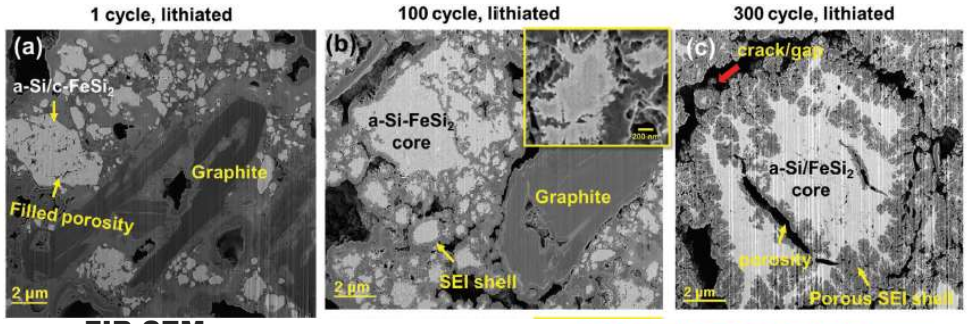
Particle swelling ?
Operando SWAXS/SANS

Reaction heteogeneities in a pouch cell ?
Operando scanning SWAXS

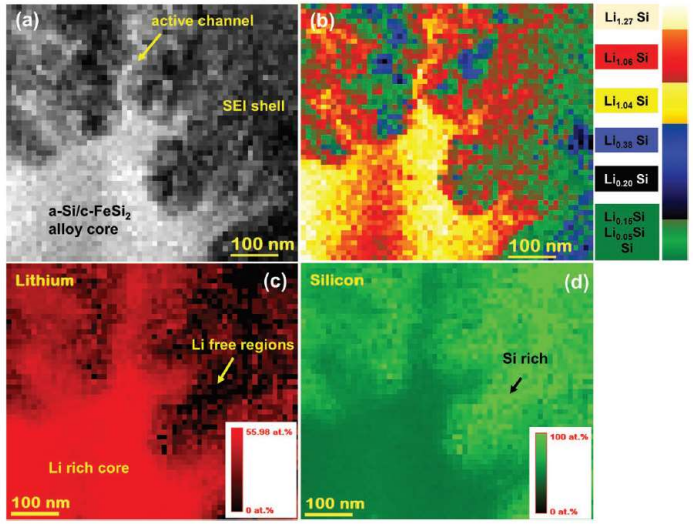
Results by other ex situ techniques

FIB-SEM showing SEI formation & growth

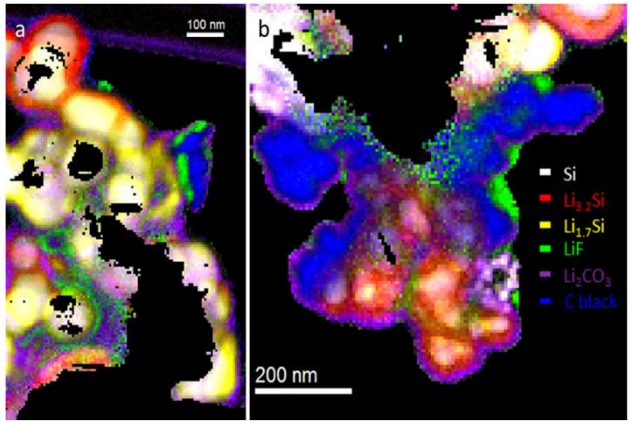
P. Kumar *et al*, Small (2020), 16, 1906812



FIB-SEM



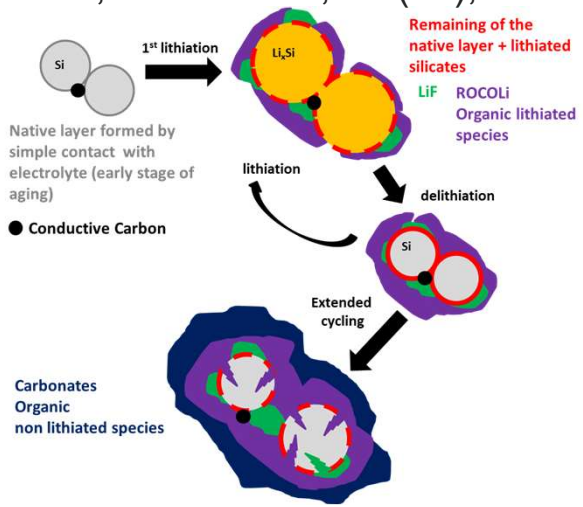
STEM-EELS
700 cycles
Delithiated



STEM-EELS

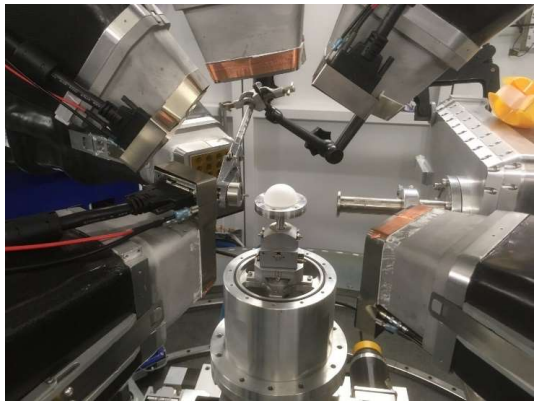
Silicon nanoparticles ~ 100 nms

Boniface *et al*, Nanoletters, 16 (12), 7381-7388, 2016.

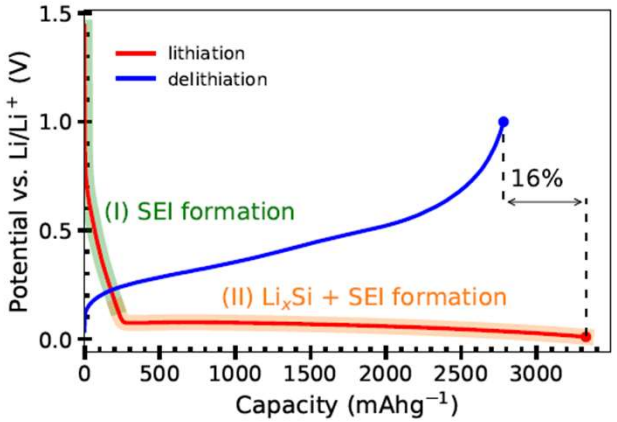


SEI composition

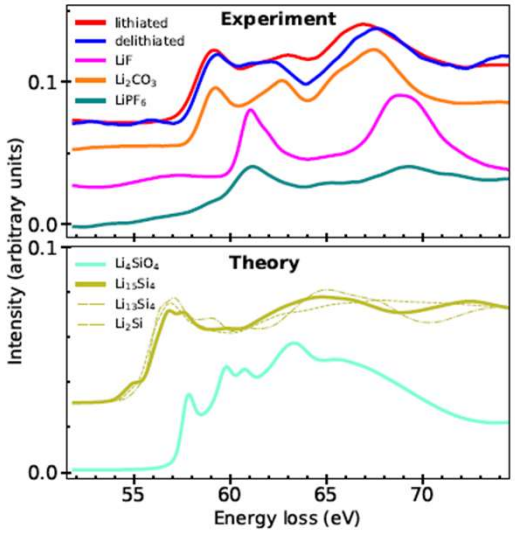
Ex situ X-ray Raman Scattering
ID20 at ESRF

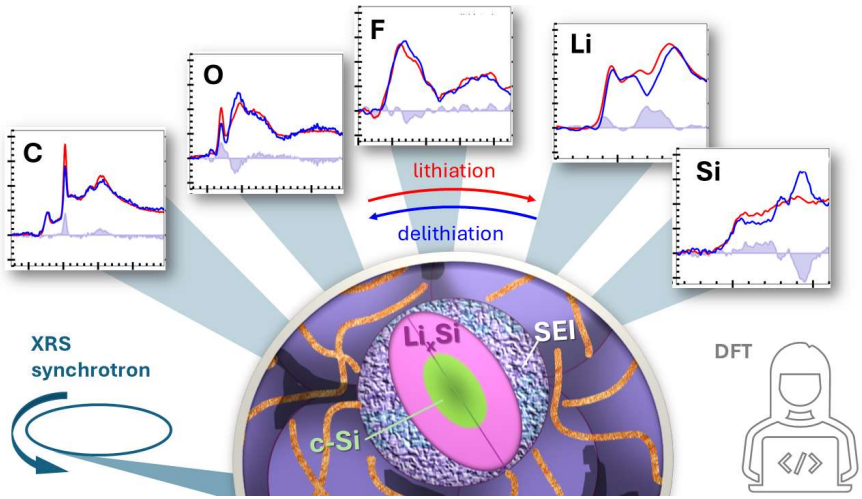


Model Si-Nanoparticles electrodes



Using experimental and DFT-based reference spectra to identify and quantify species





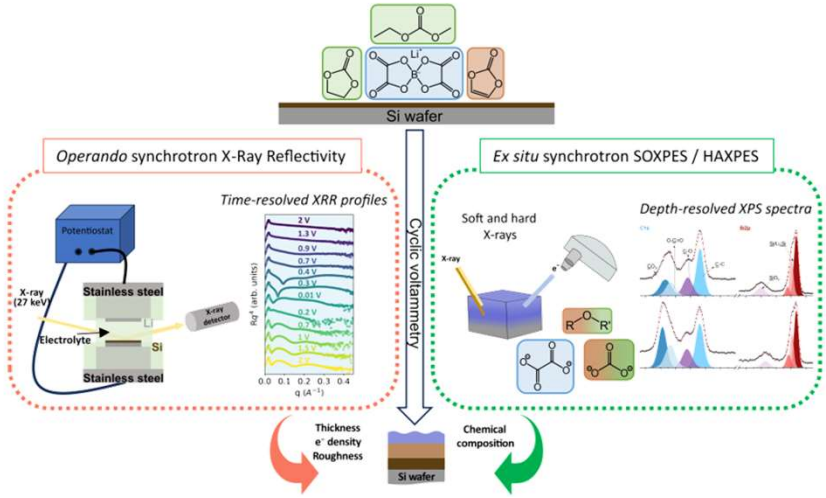
- **SEI species** are mostly LiF and Li_2CO_3, also POxFy and SiOxF
- The SEI **partially dissolves** upon delithiation up to 35%
- **Lithium is not fully removed** from the anode in the delithiated state, even after only one cycle.
- The **irreversible lithium loss** due to trapping in alloyed Si regions is quantified at 17% of Li unavailable for reactionsBla

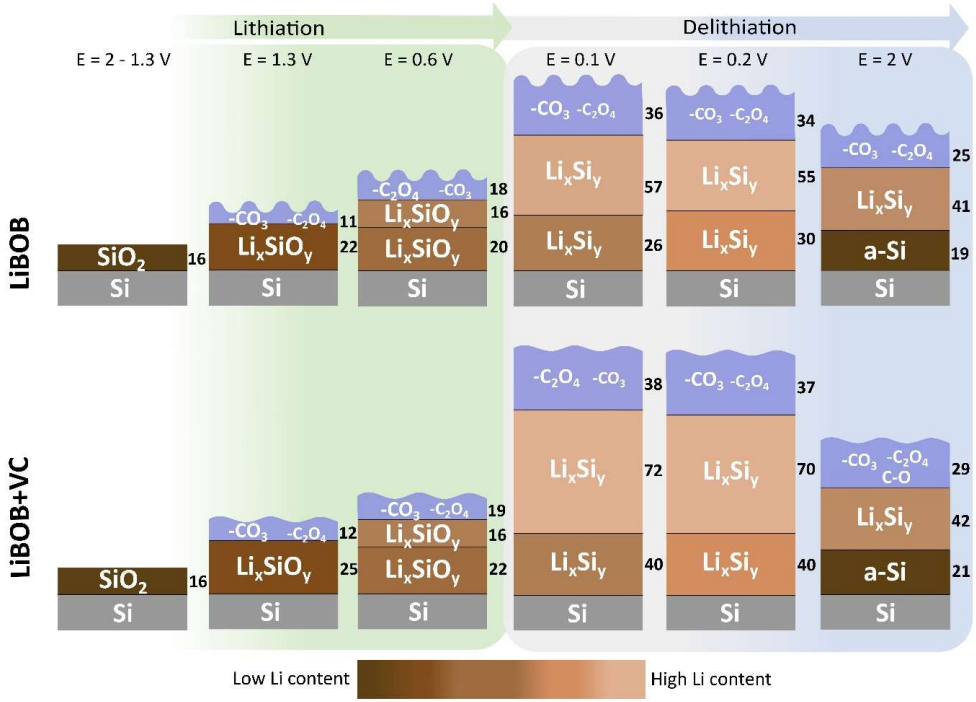
Hernandez Bertran et al, Understanding the irreversible lithium loss in silicon anodes using multi-edge X-ray scattering Analysis, Chem. Mater. 2025, 37, 3648−3660

SEI growth and morphology

Combining operando X-ray Reflectivity (BM32 at ESRF) and ex situ HAXPES/SOXPES

- Model wafer-type system
- Fluorine-free electrolytes w/w.o. VC additive





- **Distinct stages in the SEI evolution** during (de)lithiation
- VC addition leads to a slightly **thicker and smoother SEI** with lower electron density composed of carbonates mainly.
- Without VC the layer is a mixture of ester/carbonates/ether species that, suggests **continuous decomposition** of the BOB anion.
- At the end of the delithiation, L3 is an amorphous lithium-free silicon layer, while some lithium remains trapped in L2.

Lu et al, Formation and Evolution of the Solid Electrolyte Interphase on Silicon Electrodes from Fluorine-Free Electrolytes

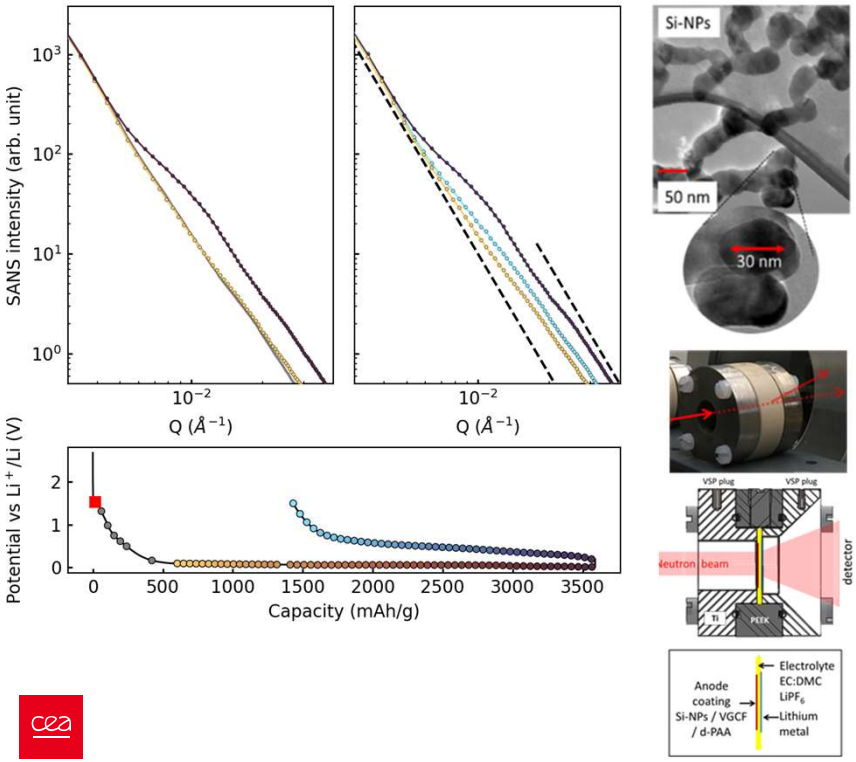
Accessing silicon nanoscale expansion ?

Small angle scattering techniques SAXS/SANS

Accessing silicon nanoscale expansion ?

Small angle scattering techniques SAXS/SANS

Model Silicon Nanoparticles Study on D22 at ILL using custom operando cell in half-cell (against Li metal)

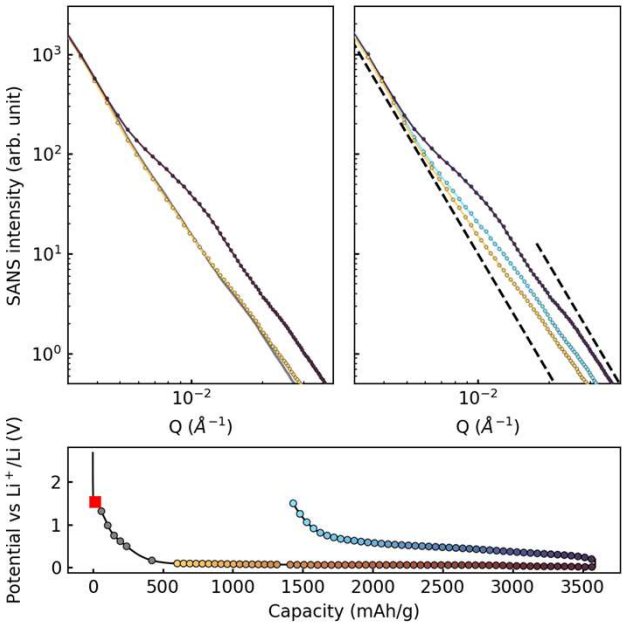


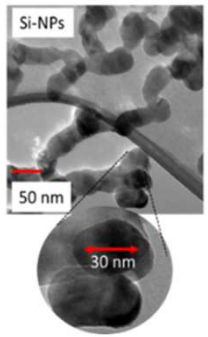
Accessing silicon nanoscale expansion ?

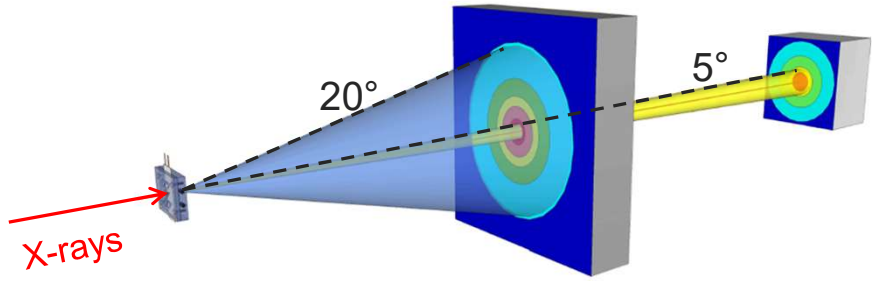
Small angle scattering techniques SAXS/SANS

French Beamline BM02 @ ESRF
A unique combined WAXS/SAXS set-up

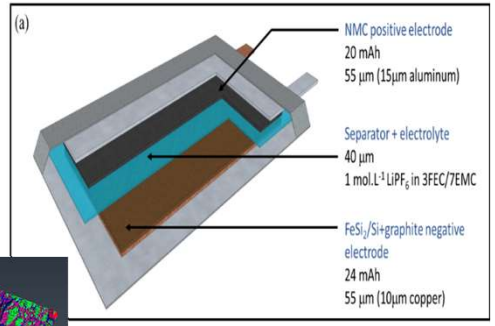
Model Silicon Nanoparticles Study on D22 at ILL using custom operando cell in half-cell (against Li metal)

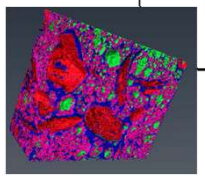




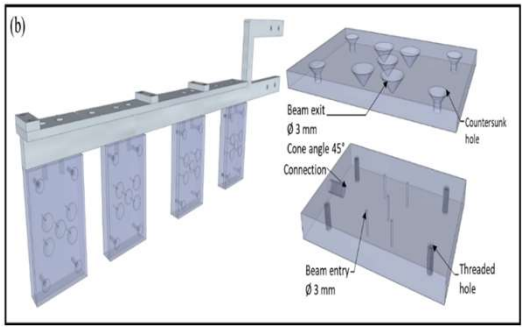


Cycling in full cell (pouch geometry)





Operando experiments on several cells simultaneously (pristine and aged)



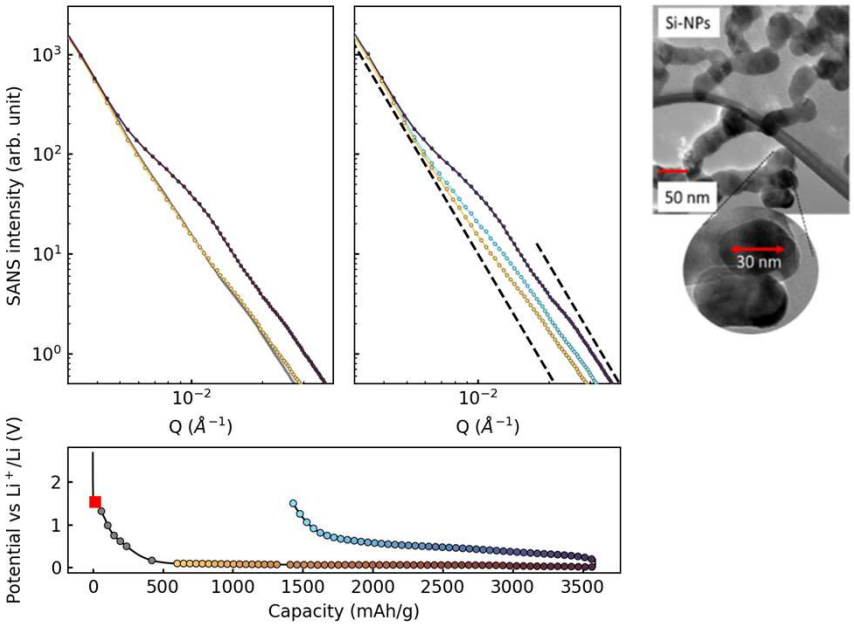
C. Berhaut *et al*, ACS Nano (2019), 13, 10, 11538-11551

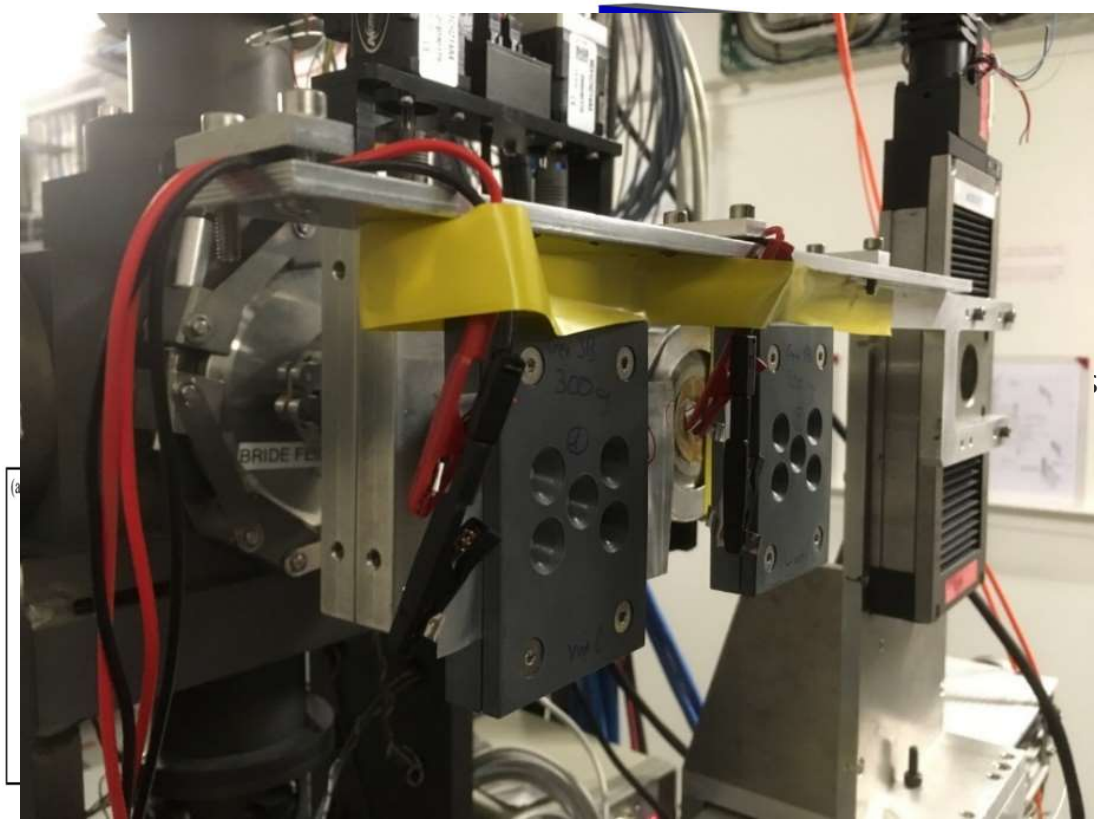
Accessing silicon nanoscale expansion ?

Small angle scattering techniques SAXS/SANS

French Beamline BM02 @ ESRF
A unique combined WAXS/SAXS set-up

Model Silicon Nanoparticles Study on D22 at ILL using custom operando cell in half-cell (against Li metal)

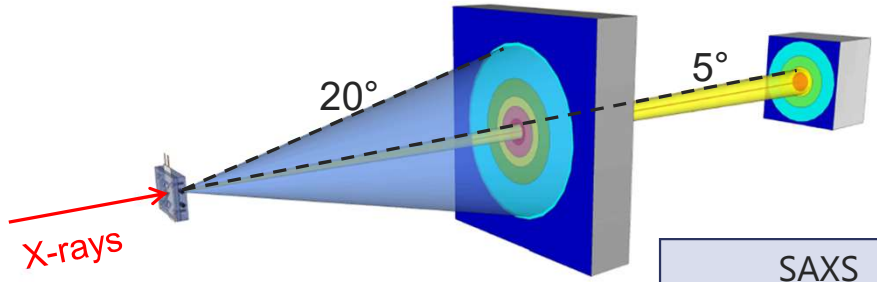




C. Berhaut *et al*, ACS Nano (2019), 13, 10, 11538-11551

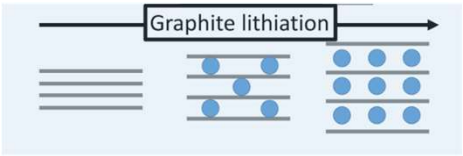
Silicon nanoscale expansion

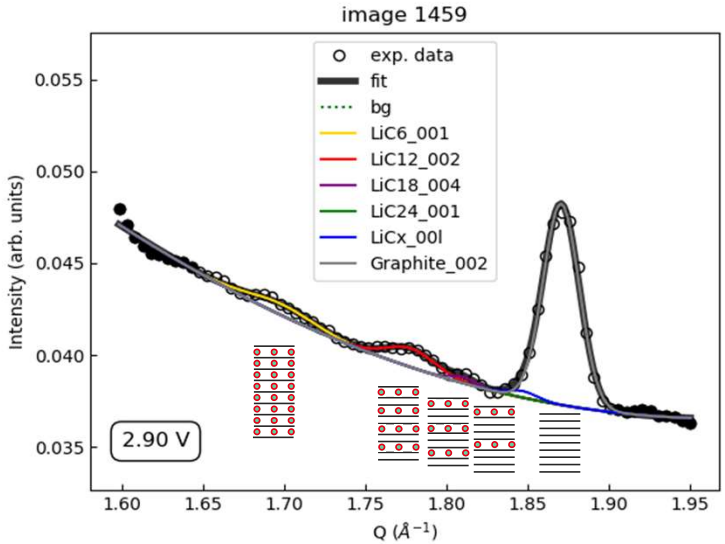
French Beamline BM02 @ ESRF
A unique combined WAXS/SAXS set-up

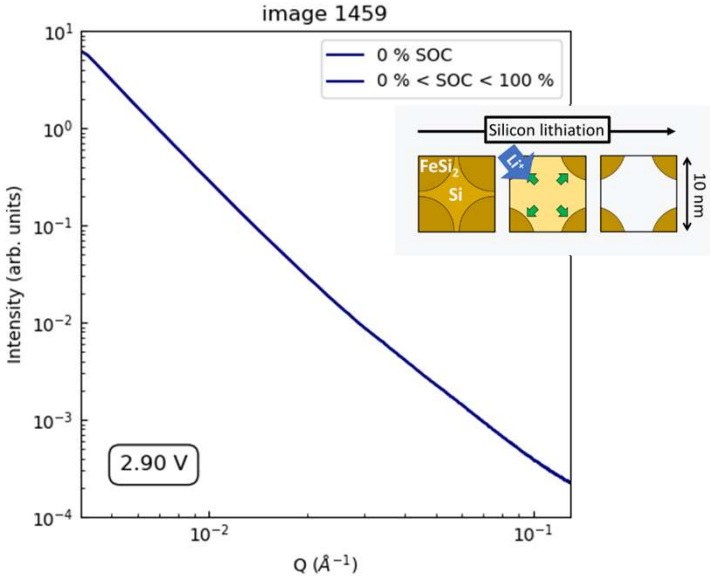


SAXS
Nanoscale - Silicon

WAXS
Atomic scale - Graphite

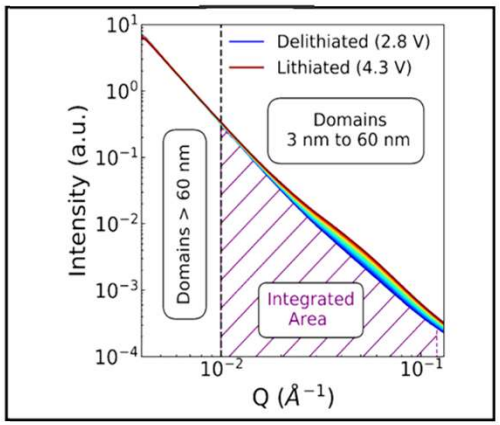


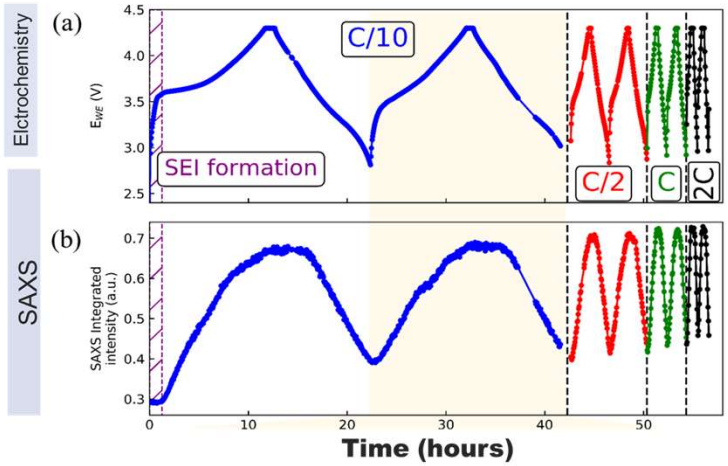


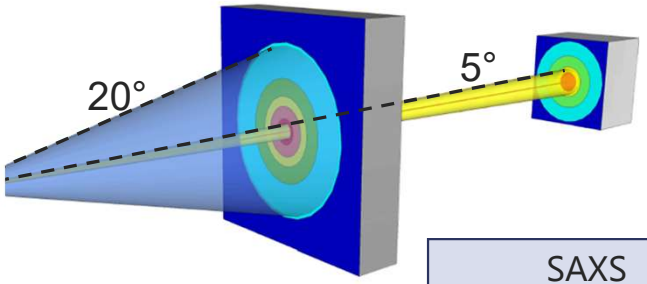


Silicon nanoscale expansion

Qualitative analysis

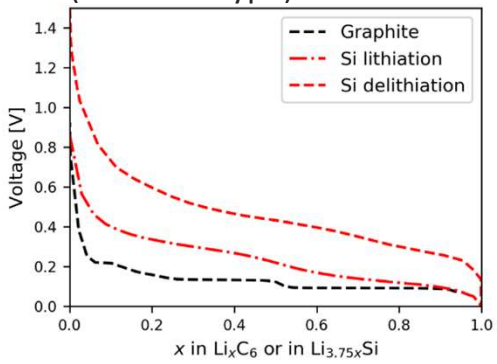


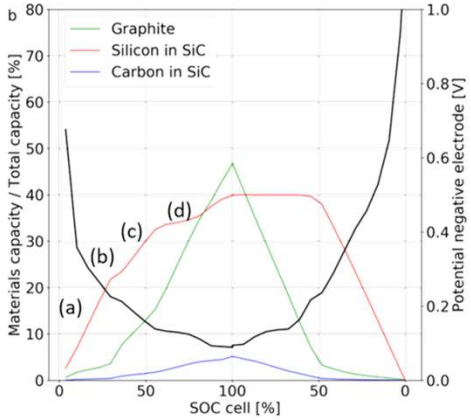


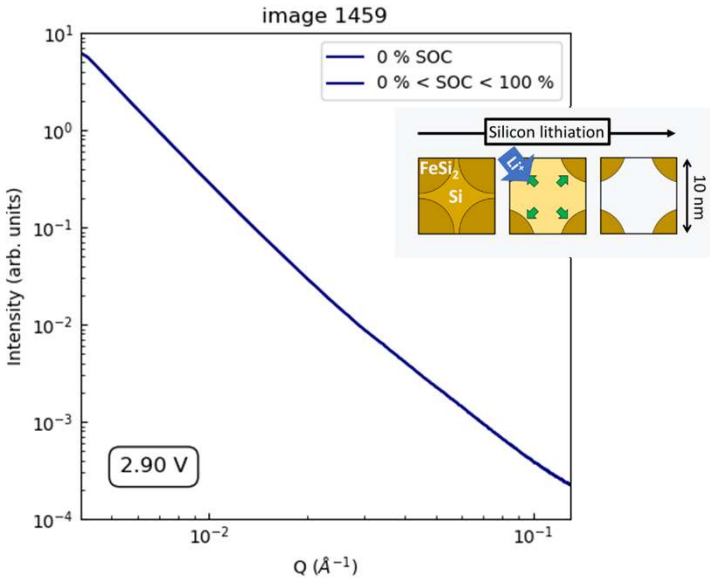


SAXS
Nanoscale - Silicon

Porous electrode modelling (Newman-type)



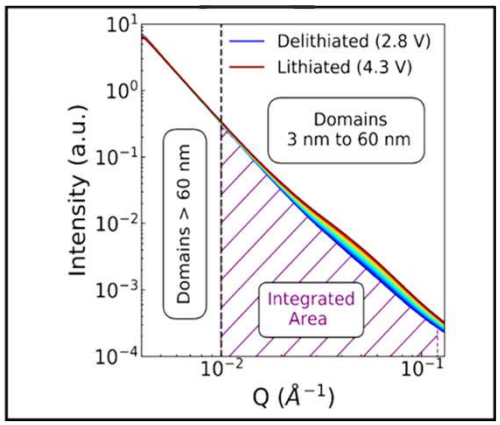




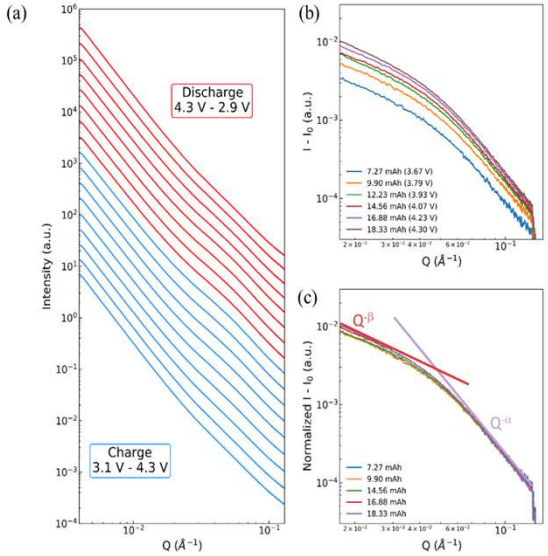
P-F Lory *et al* 2020 *J. Electrochem. Soc.* **167** 120506

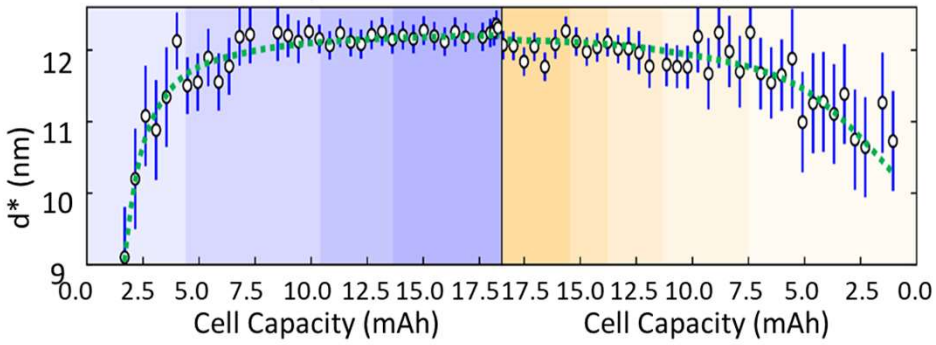
Silicon nanoscale expansion

Qualitative analysis



Quantitative analysis





I(Q) α **$(\Delta\rho)^2$** × P(Q) × S(Q)

$(\Delta r)=(\rho_{LixSi}-\rho_{medium})$
Contrast, highly sensitive to Li

P(Q) form factor
(size/shape Si domains)

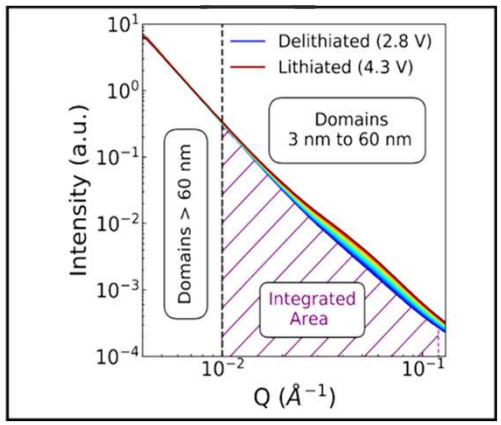
S(Q) structure factor
(organization)

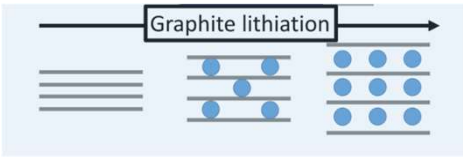
d* = 2p/Q* is a model-free characteristic structural fingerprint of the materials nanoscale organization

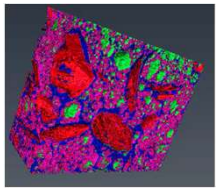
- **Dynamic nanoscale swelling** and shrinking of silicon
- **Non-linear behavior** with capacity
- **Non reversible**

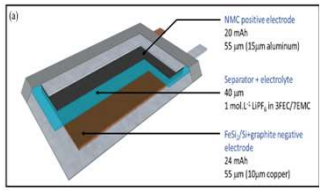
Silicon nanoscale expansion

Qualitative analysis

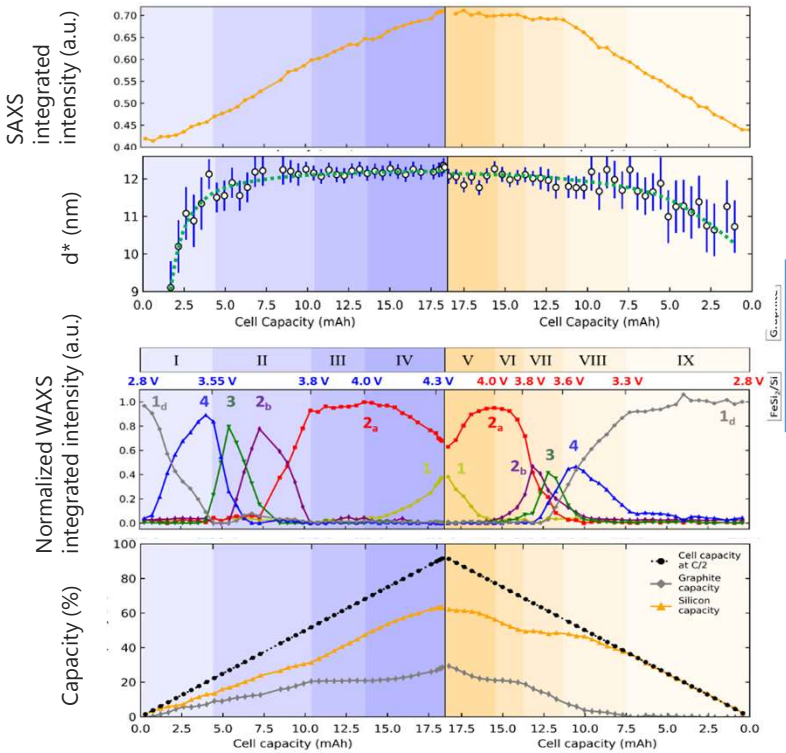


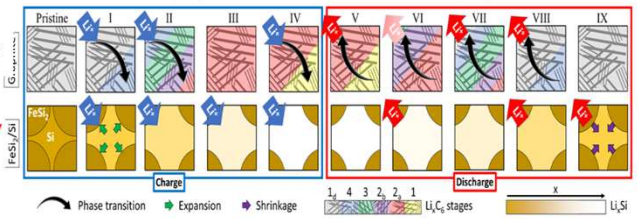






- Combining SAXS with WAXS gives a **full understanding of the sequential reaction mechanism** between silicon and graphite, not symetric between lithiation and delithiation

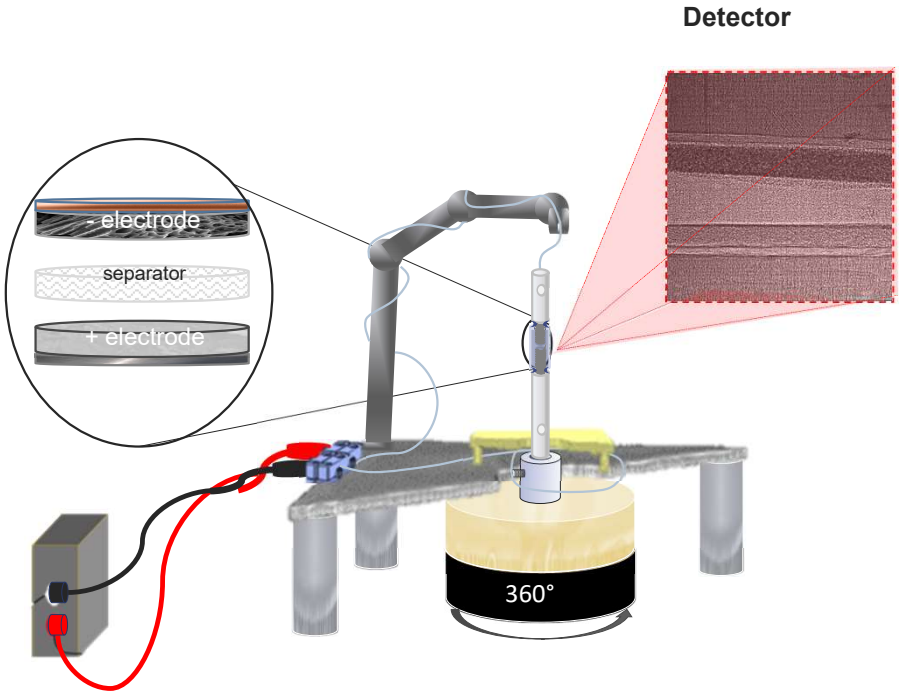




Bridging particle-electrode scales ?

The question : what limits the performance ?
Can we determine the (local) chemomechanics ?

Operando nanoholotomography



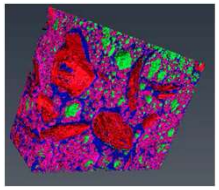
V. Vanpeene et al. Comparative Study of the Quantitative Analysis of Battery Materials with X-ray Nanotomography: From Ex Situ toward Operando Measurements, ACS Nano, 2025. doi.org/10.1021/acsnano.4c16419

Nanoscale imaging

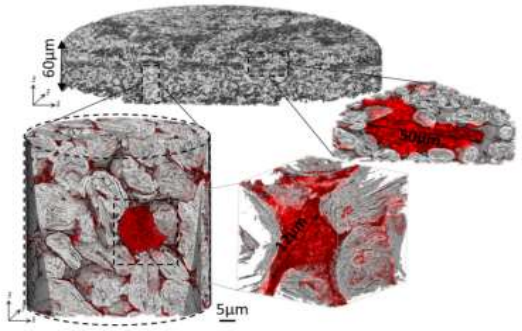

Victor Vanpeene


Julie Villanova

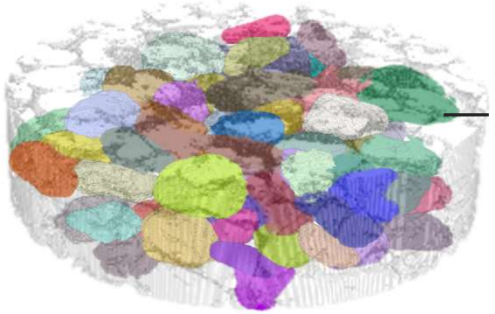

Olga Stamati



Silicon clusters



Particle-by-particle labellisation



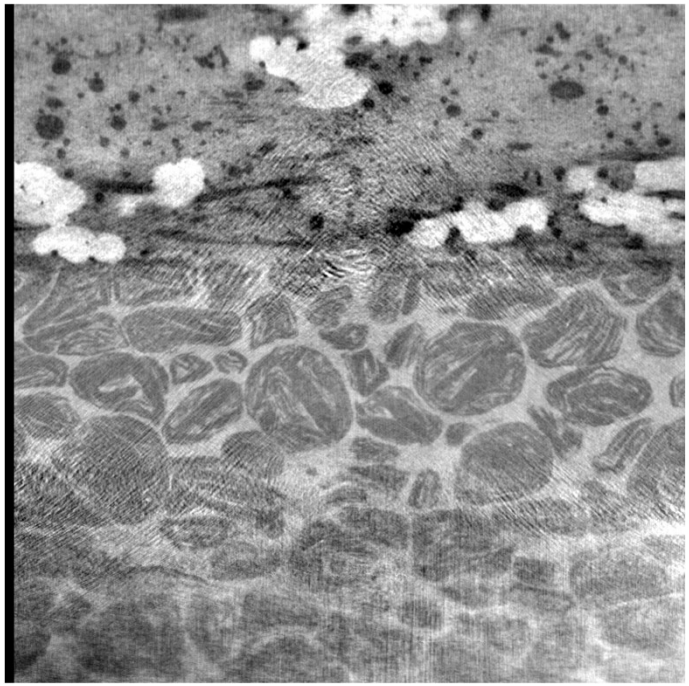
Nanoscale imaging


Victor Vanpeene

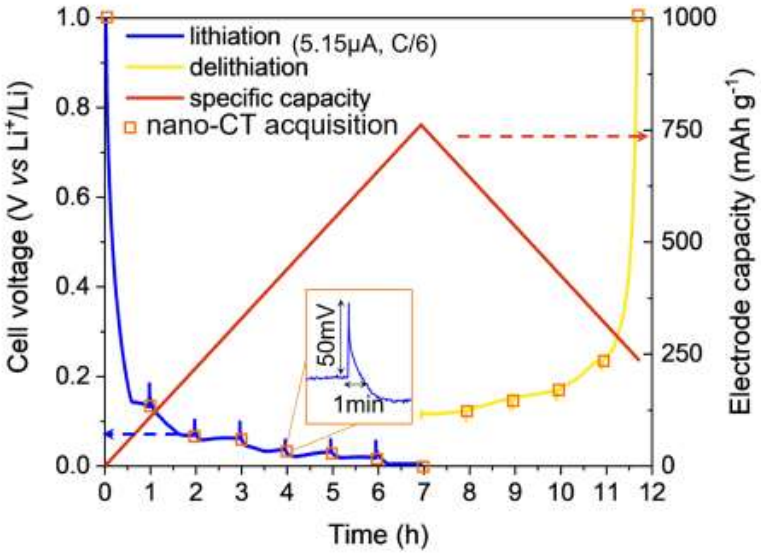

Julie Villanova


Olga Stamati

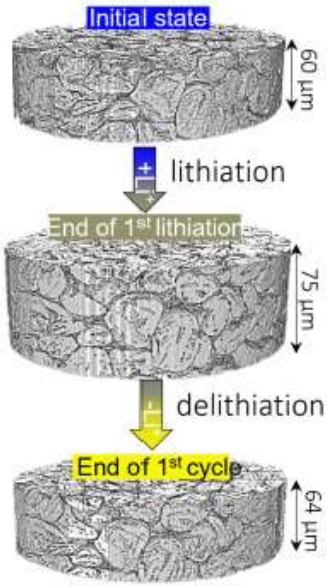
Nanoholotomography ID16b
Field of view 102x102µm^2
Voxel size 50 nm



Formation cycle C/6
Gr/Si composite anode



- Capturing volume changes



Imaged volume 5.3 10^{-3} mm^3

Victor Vanpeene*,a, Olga Stamati, Francois Cadiou, Quentin Jacquet, Julie Villanova, Sandrine Lyonnard*. *4D operando X-ray nano-holo-tomography reveals multiscale chemomechanics in Silicon-Graphite anode*, arXiv:2508.06413

Nanoscale imaging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lithiation delithiation

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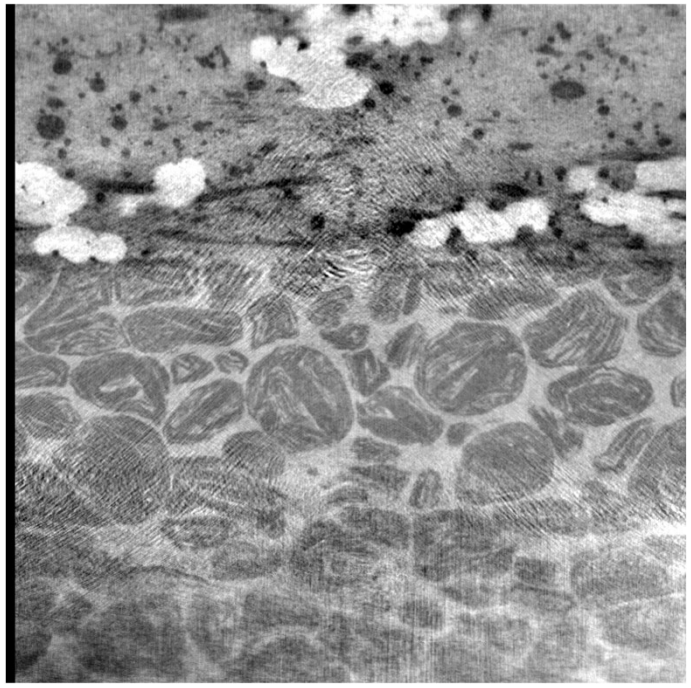
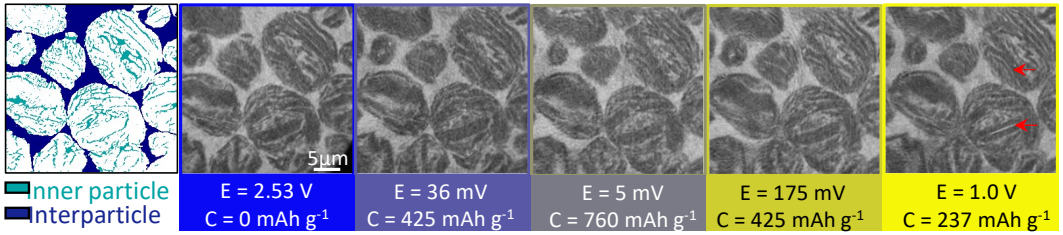
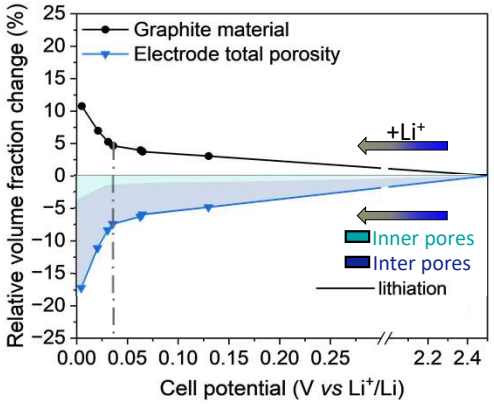
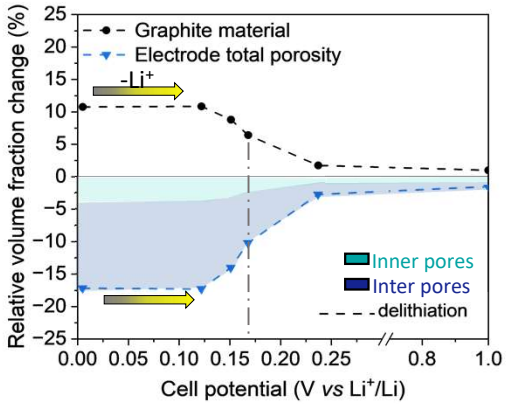
Field of view 102x102µm²
Voxel size 50 nm



○ *Quantifying graphite inner-outer pores evolution*







Victor Vanpeene*,a, Olga Stamati, Francois Cadiou, Quentin Jacquet, Julie Villanova, Sandrine Lyonnard*. *4D operando X-ray nano-holo-tomography reveals multiscale chemomechanics in Silicon-Graphite anode*, arXiv:2508.06413

Nanoscale imaging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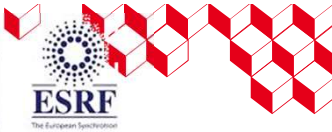
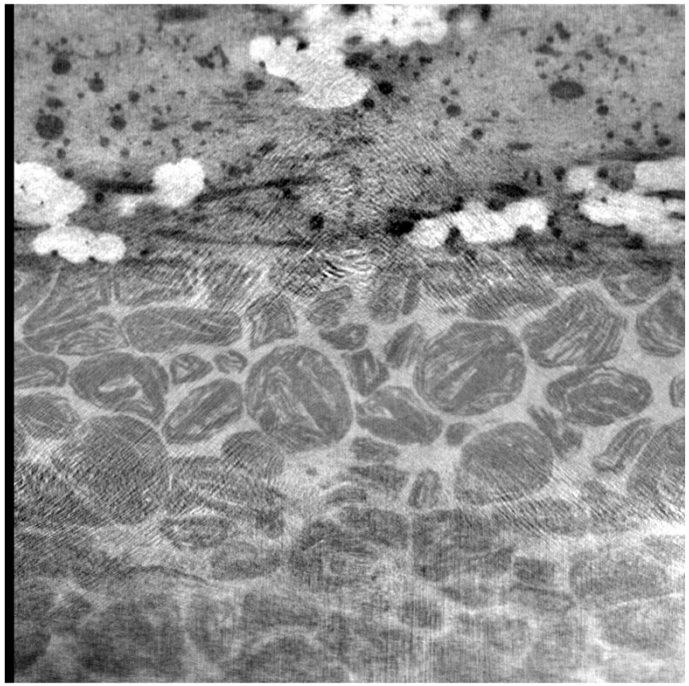
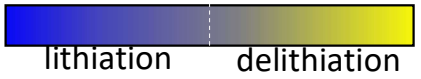
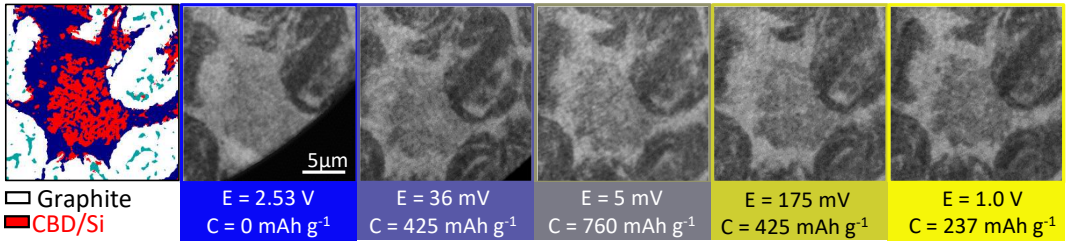


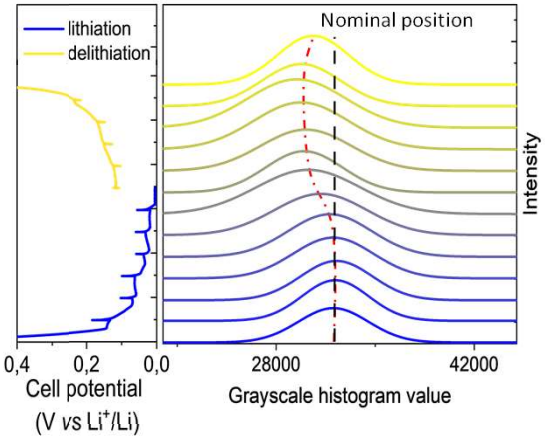
Nanoholotomography ID16b
Field of view 102x102µm²
Voxel size 50 nm

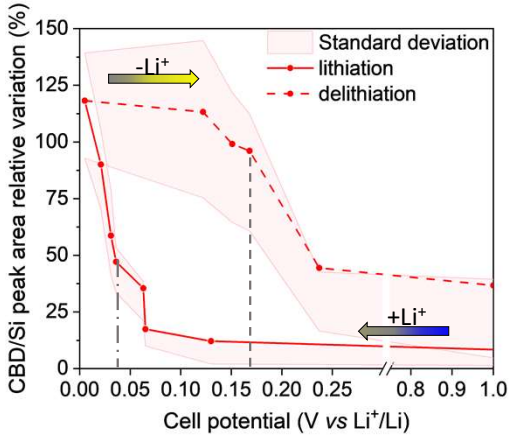


○ *Visualizing silicon phase activity*









Nanoscale imaging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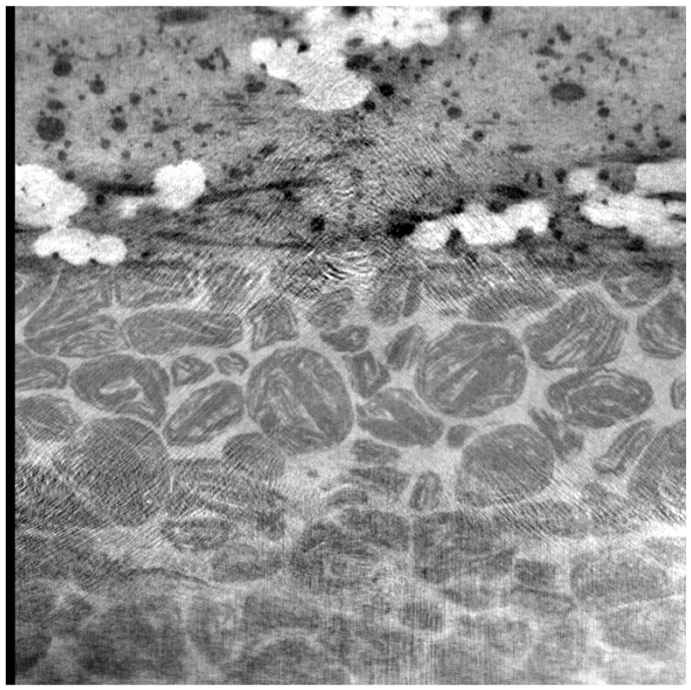
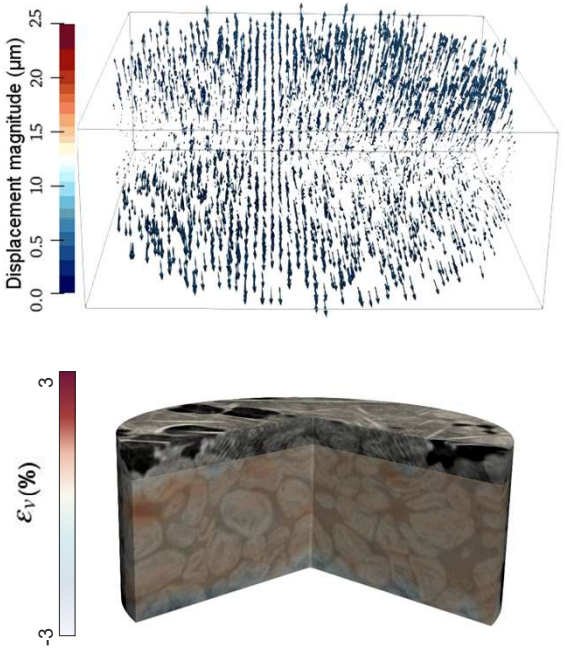
Nanoholotomography ID16b
Field of view 102x102µm^2
Voxel size 50 nm

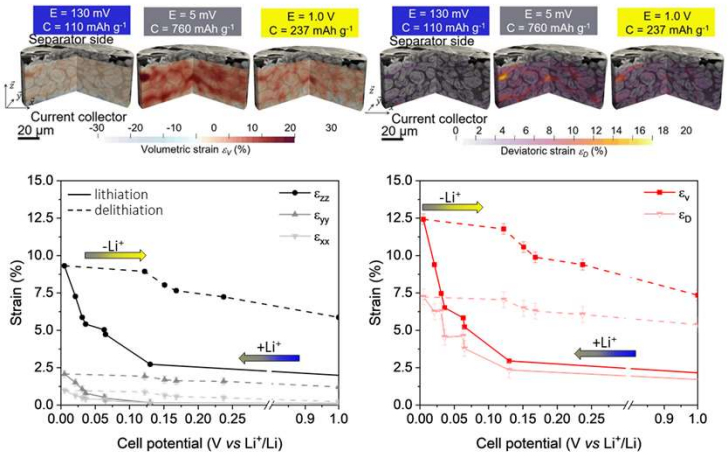


Digital Volume Correlations
Electrode-scale deformations
Displacements and strain maps



Time frame 1h

- Heterogeneous strain
- Presence of hot spots



Victor Vanpeene*,a, Olga Stamati, Francois Cadiou, Quentin Jacquet, Julie Villanova, Sandrine Lyonnard*. *4D operando X-ray nano-holo-tomography reveals multiscale chemomechanics in Silicon-Graphite anode*, arXiv:2508.06413

Nanoscale imaging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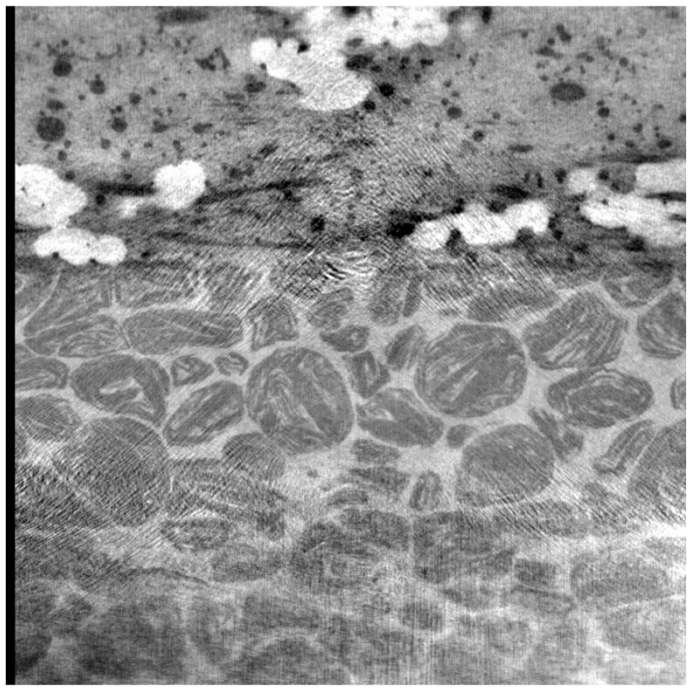
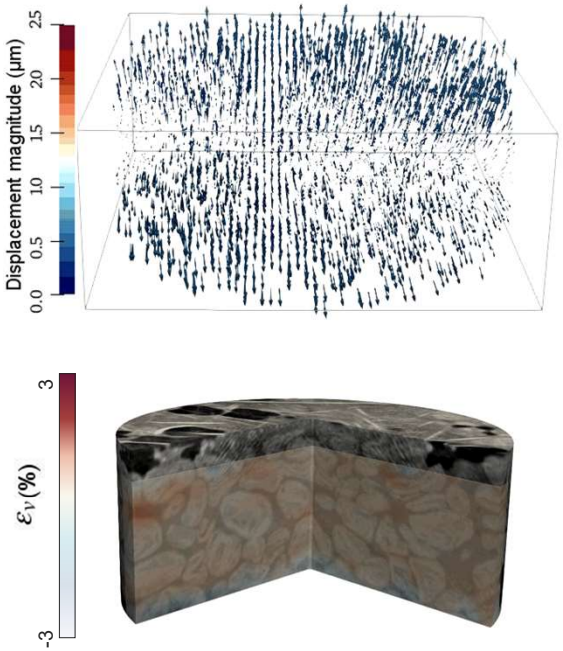
Nanoholotomography ID16b
Field of view 102x102µm²
Voxel size 50 nm

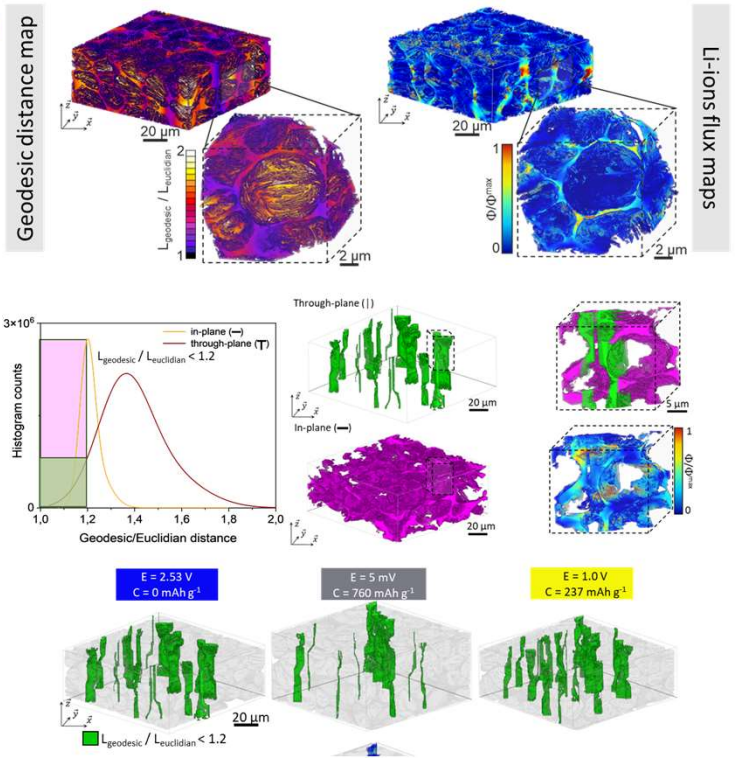


Digital Volume Correlations
Electrode-scale deformations
Displacements and strain maps



Time frame 1h

- Evidence of « fast track channels »



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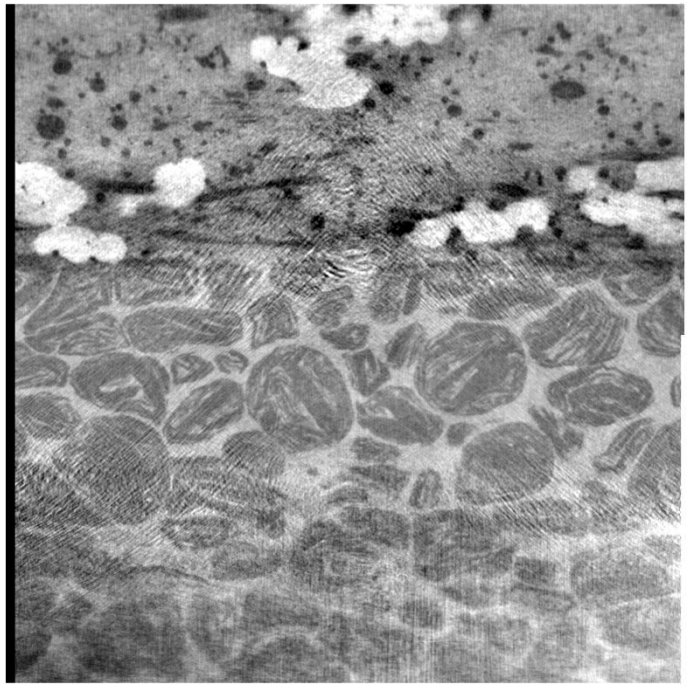
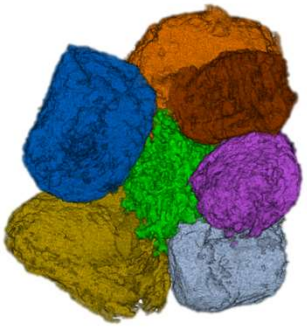
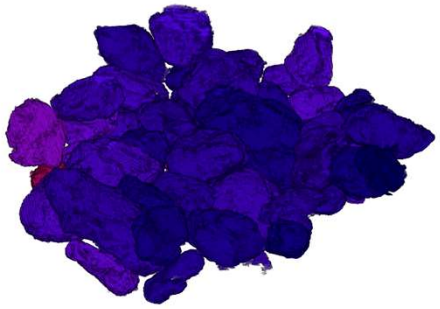
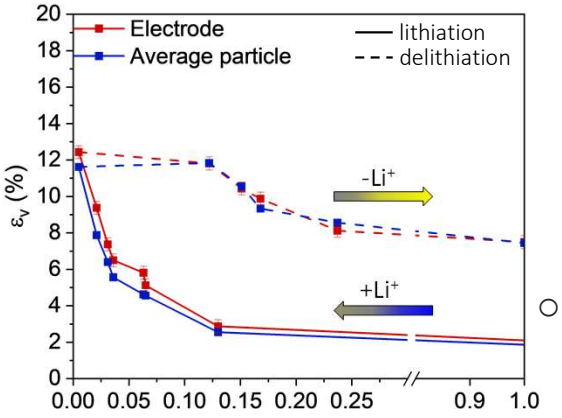
Field of view 102x102µm²
Voxel size 50 nm



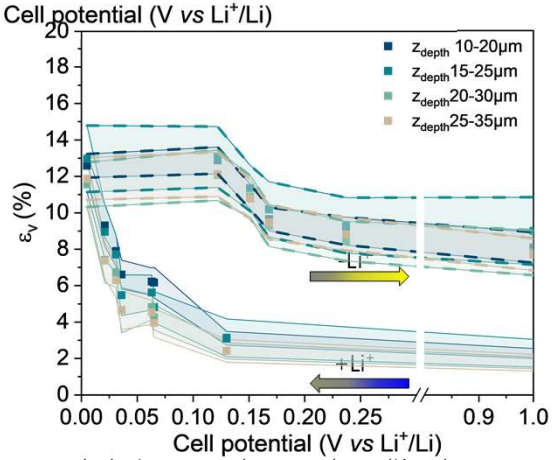
Particle-by-particle labellisation
Local Digital Volume Correlations
Local State-of-charge
Local activity/reversibility







- Shows the heterogeneity at particle level



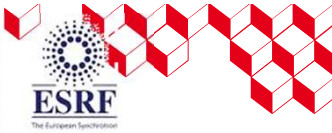
Victor Vanpeene*,a, Olga Stamati, Francois Cadiou, Quentin Jacquet, Julie Villanova, Sandrine Lyonnard*. *4D operando X-ray nano-holo-tomography reveals multiscale chemomechanics in Silicon-Graphite anode*, arXiv:2508.06413

Nanoscale imaging

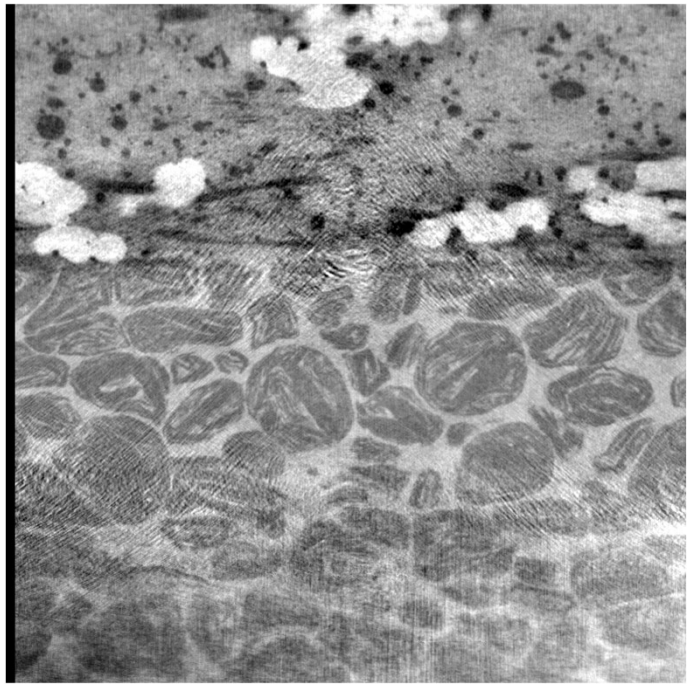

Victor Vanpeene


Julie Villanova

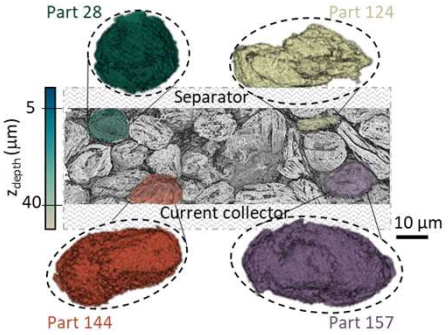

Olga Stamati

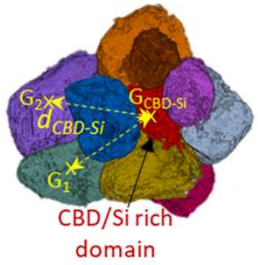


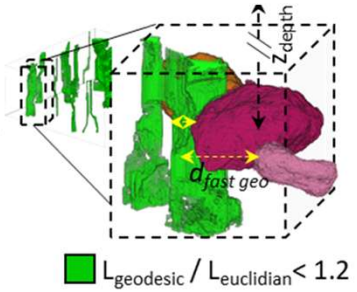
Nanoholotomography ID16b
Field of view 102x102µm²
Voxel size 50 nm

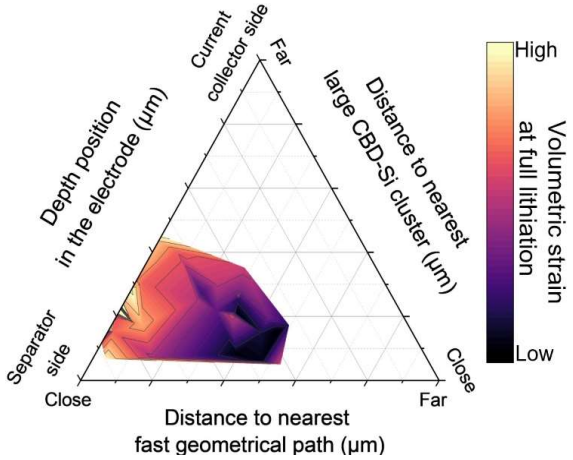


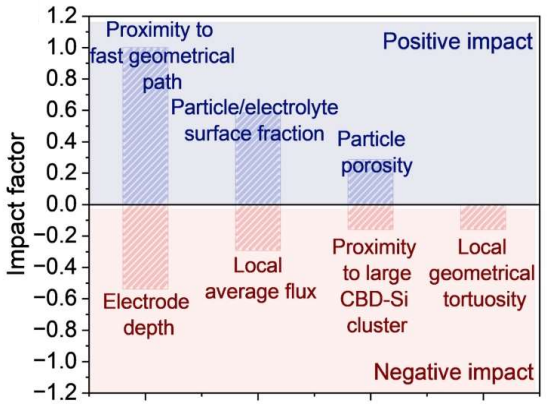
- **Main key factors are influencing the chemomechanics**











Victor Vanpeene*,a, Olga Stamati, Francois Cadiou, Quentin Ja
chemomechanics in Silicon-Graphite anode, arXiv:2508.06413

What about the consequences of having silicon in a « real » cell ?

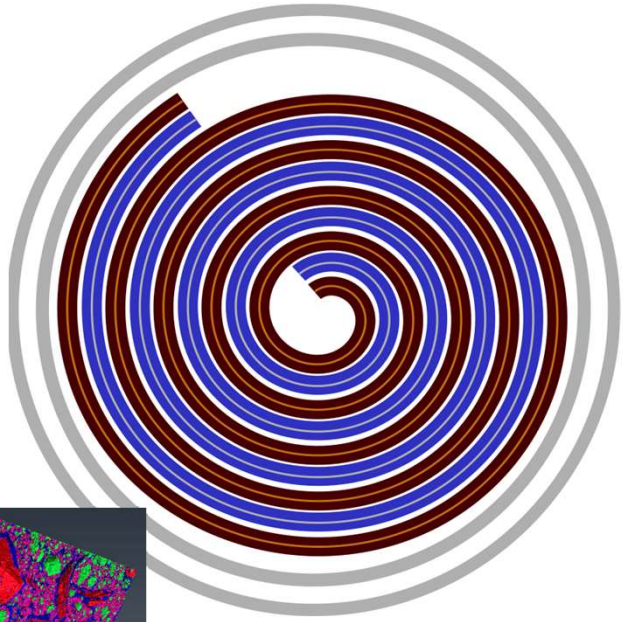
Cylindrical industry-grade cell

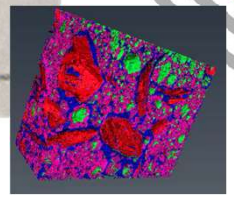
Coll. VARTA MICRO INNOVATION
Coll. ILL (L. Helfen) ; ESRF (J. Drnec)

E. Lübke

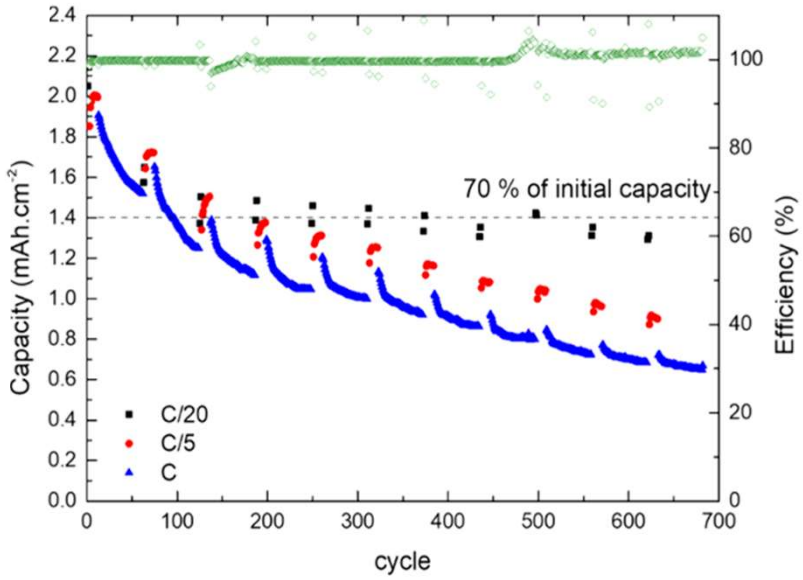
Industry-grade cylindrical Li-ion cell

Silicon-Graphite anode ; NMC622 cathode





30 mAh



E. Lübke et al, Energy Environ. Sci. **17**, 5048-5059 (2024).

Cylindrical industry-grade cell

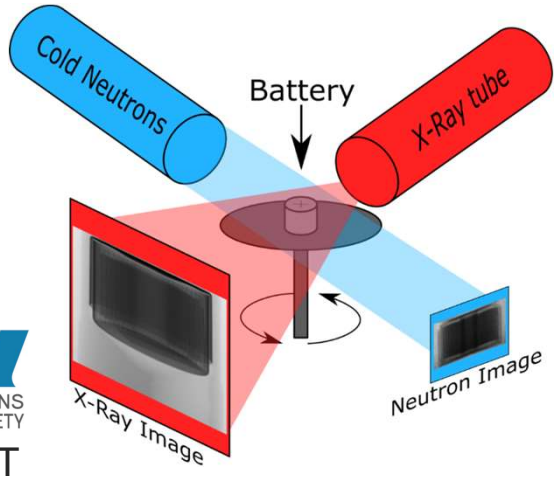
E. Lübke

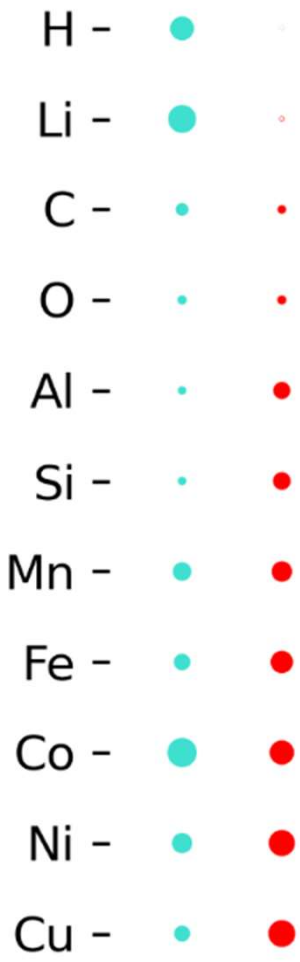
Multimodal study of fresh and aged cells using X-rays and neutron techniques

Lukas Helfen

NEUTRONS FOR SOCIETY

NeXT





Neutron attenuation coefficient

X-Ray attenuation coefficient

E. Lübke et al, Energy Environ. Sci. **17**, 5048-5059 (2024).

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NEUTRONS FOR SOCIETY

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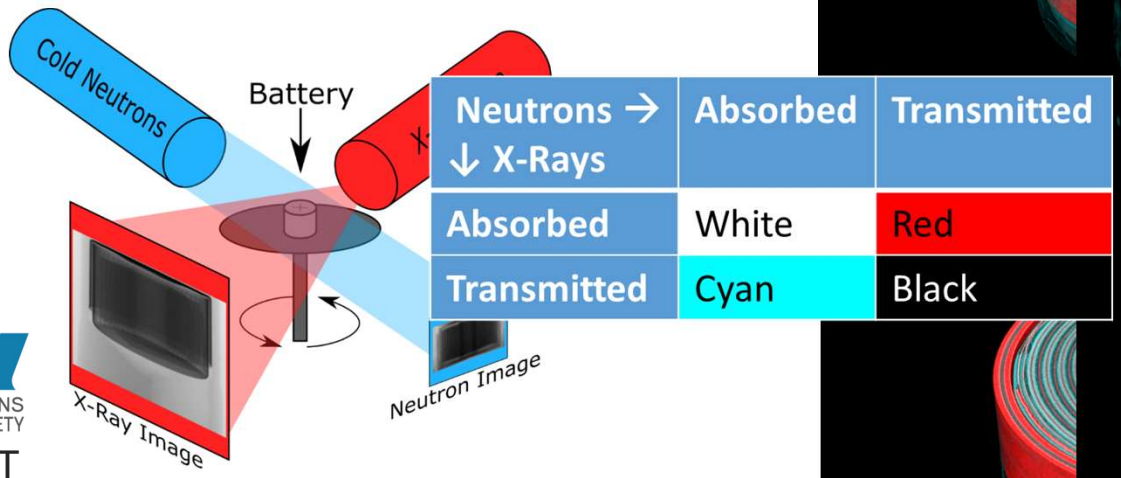
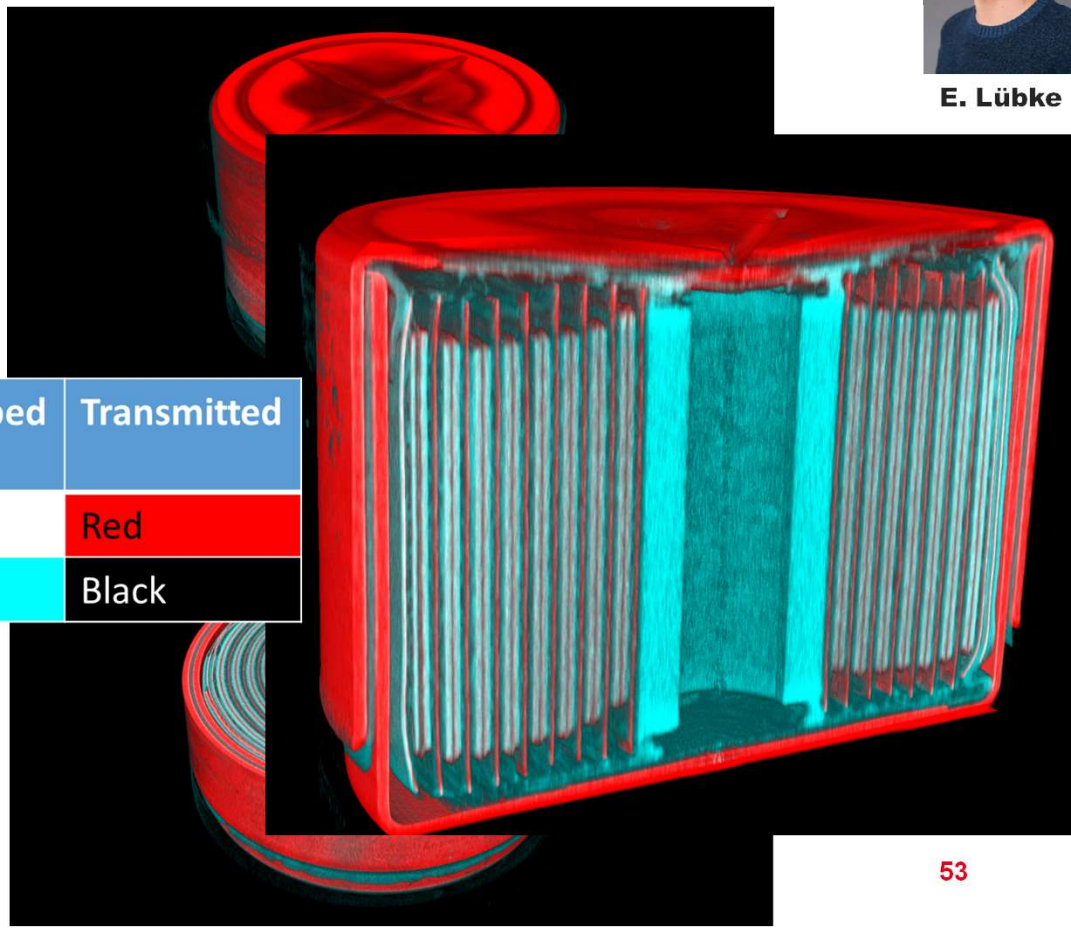


Neutrons → / ↓ X-Rays	Absorbed	Transmitted
Absorbed	White	Red
Transmitted	Cyan	Black



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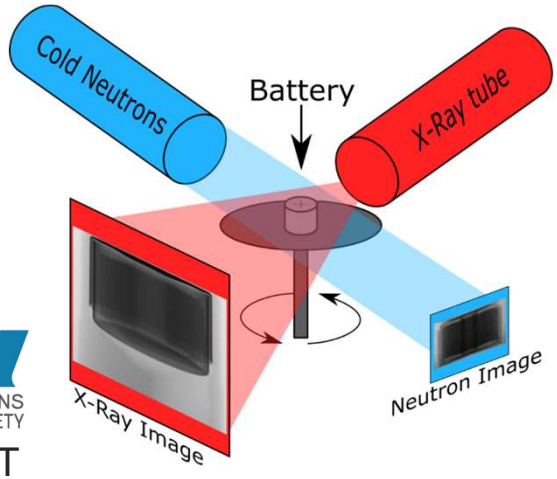
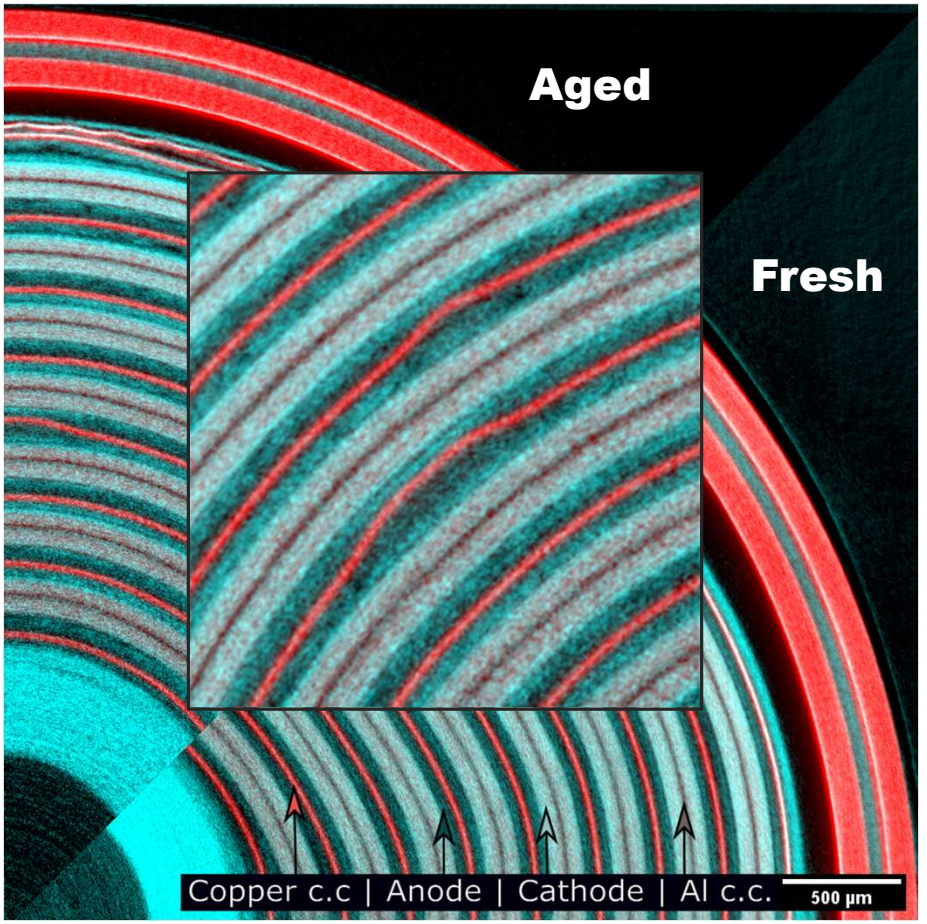




Cu

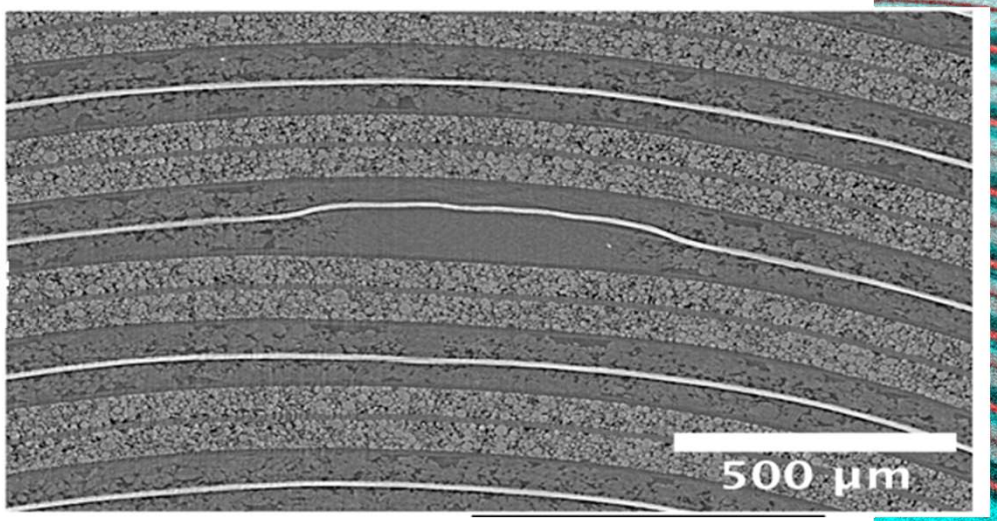
Li

E. Lübke et al, Energy Environ. Sci. **17**, 5048-5059 (2024).

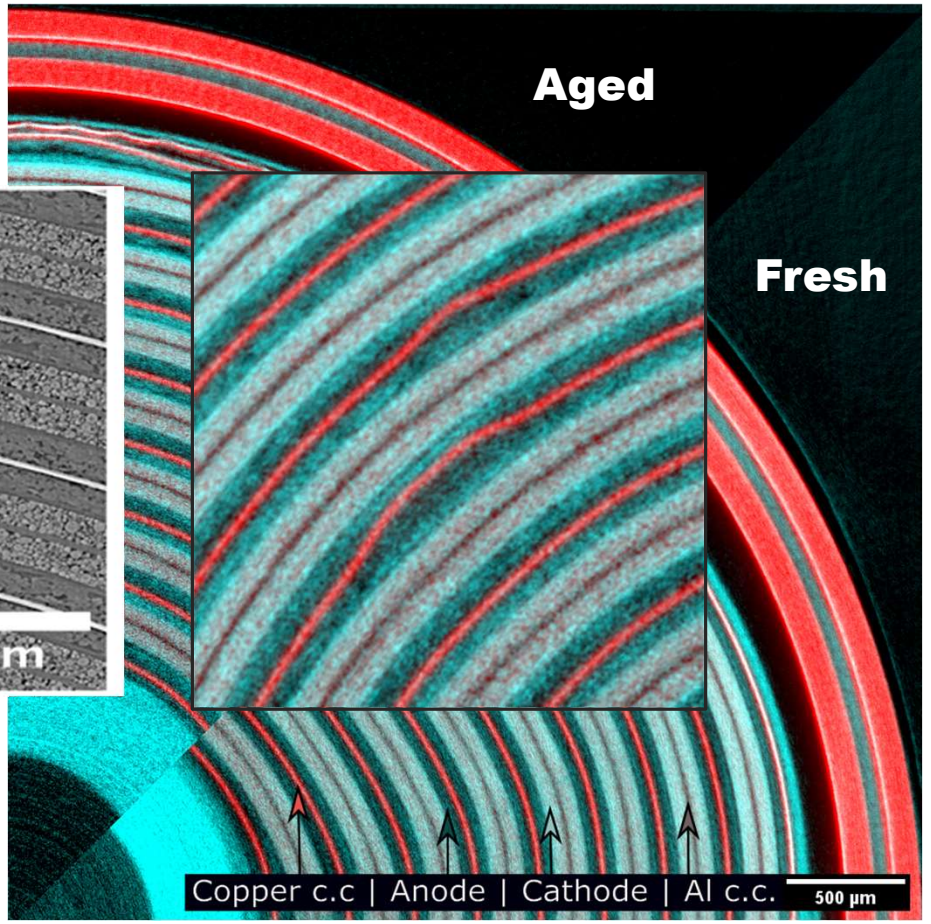
Cylindrical industry-grade cell

E. Lübke

High resolution synchrotron tomography (BM05 at ESRF)



110 keV – 300 nms voxel size – 10 minutes



Cu

Li

E. Lübke et al, Energy Environ. Sci. **17**, 5048-5059 (2024).

Cylindrical industry-grade cell

E. Lübke

Operando WAXS-CT ID31@ESRF

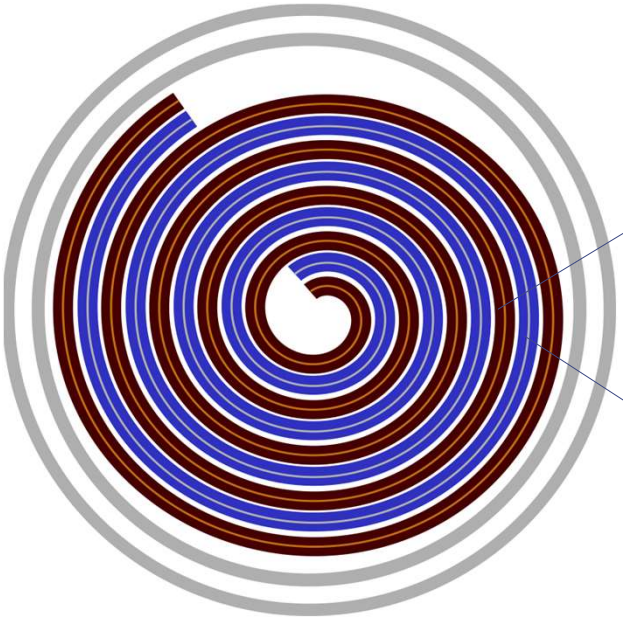
Graphite → LiC_{12} → LiC_6

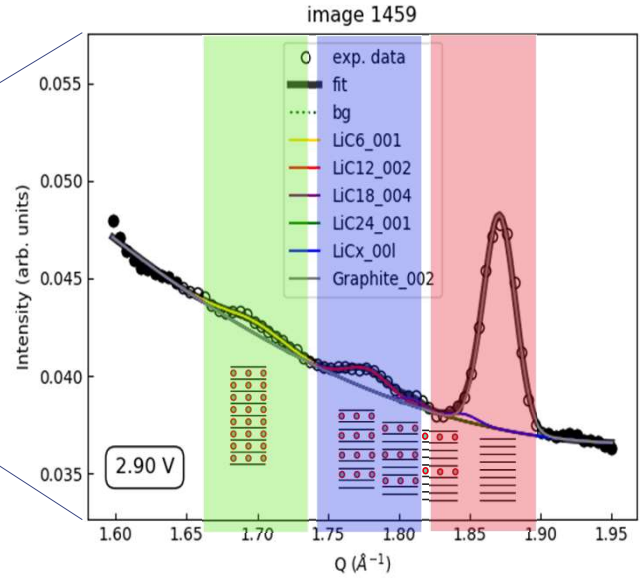
70 keV

Beam 5*10 μm

1 slice-CT

in 5 minutes



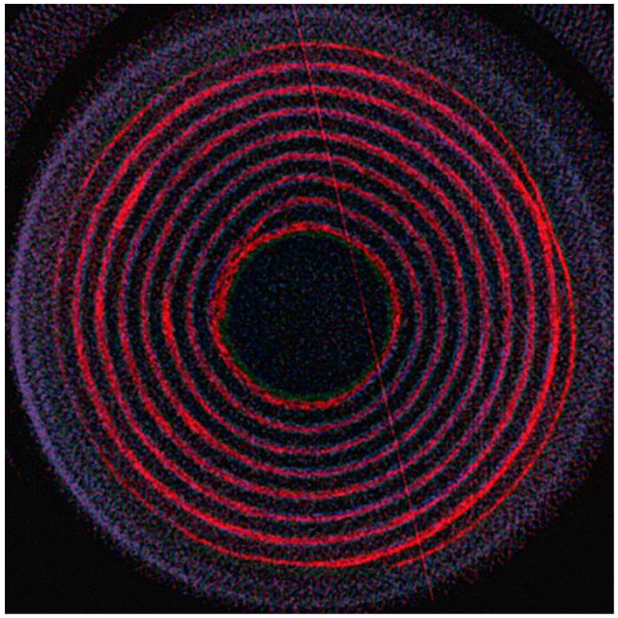


J. Drnec

M. Mirolo

E. Lübke et al, Energy Environ. Sci. **17**, 5048-5059 (2024).

Cylindrical industry-grade cell



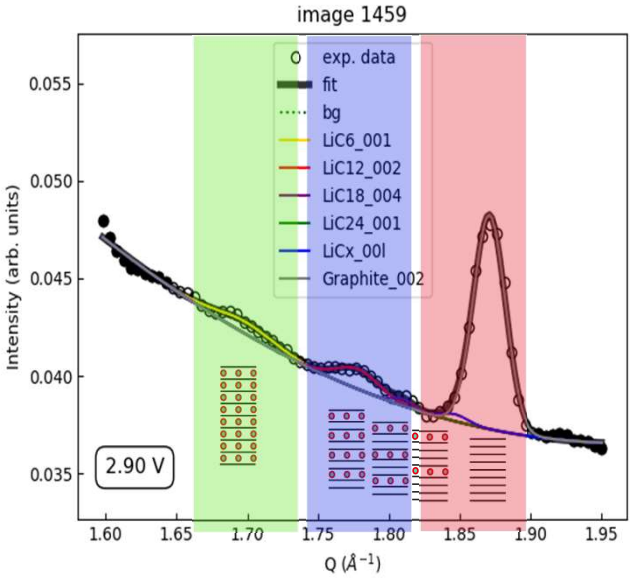
Operando WAXS-CT ID31@ESRF

70 keV

Beam 5*10 μm

1 slice-CT

in 5 minutes



E. Lübke

J. Drnec

M. Mirolo

E. Lübke et al, Energy Environ. Sci. **17**, 5048-5059 (2024).

Cylindrical industry-grade cell

E. Lübke

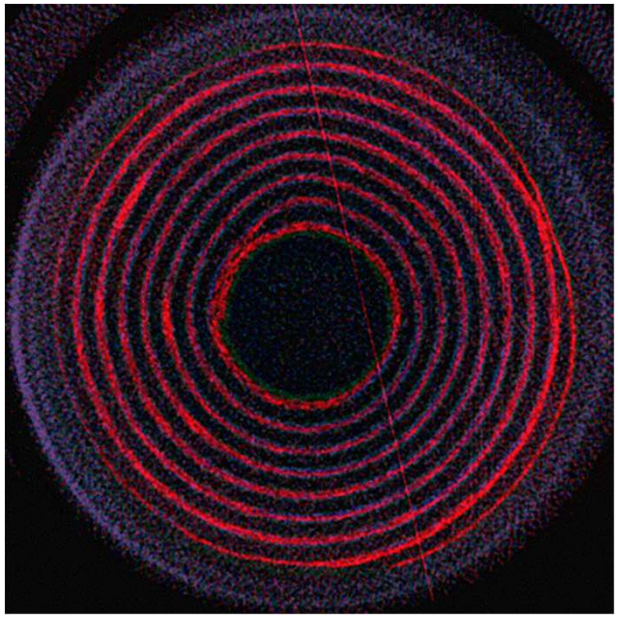
70 keV

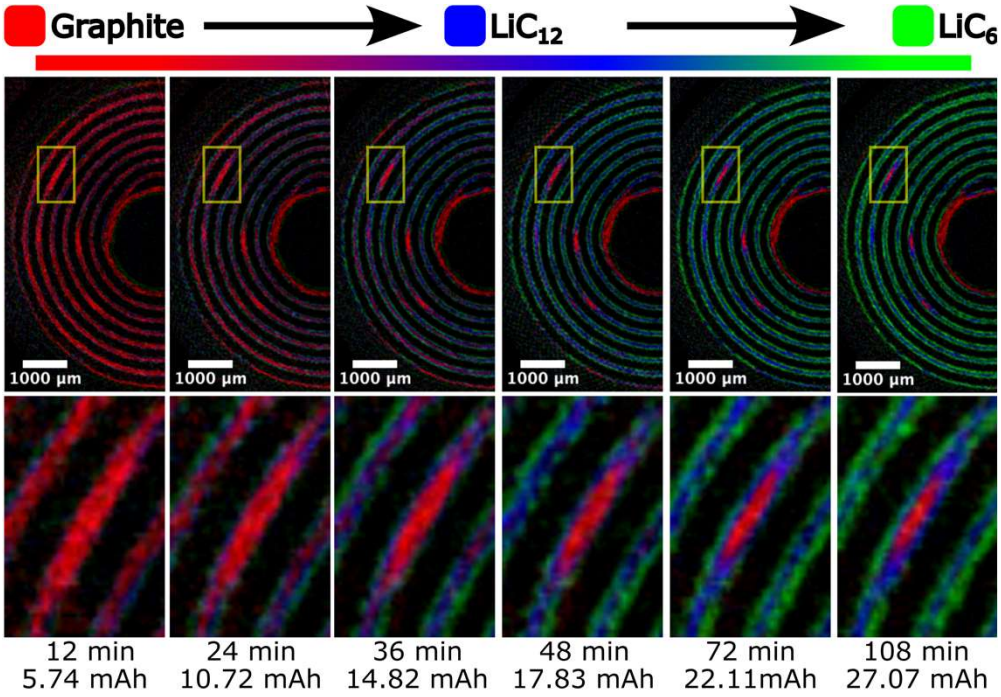
Beam 5*10 μm

1 slice-CT

in 5 minutes

Operando WAXS-CT ID31@ESRF





E. Lübke et al, Energy Environ. Sci. **17**, 5048-5059 (2024).

Cylindrical industry-grade cell

E. Lübke

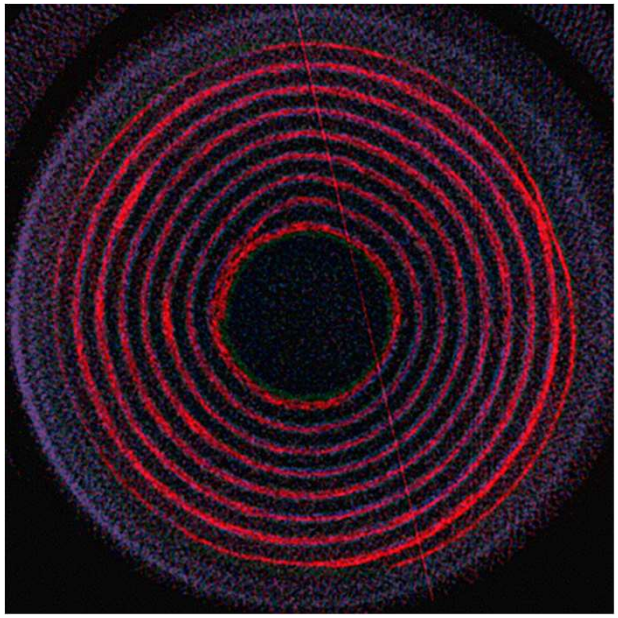
Operando WAXS-CT ID31@ESRF

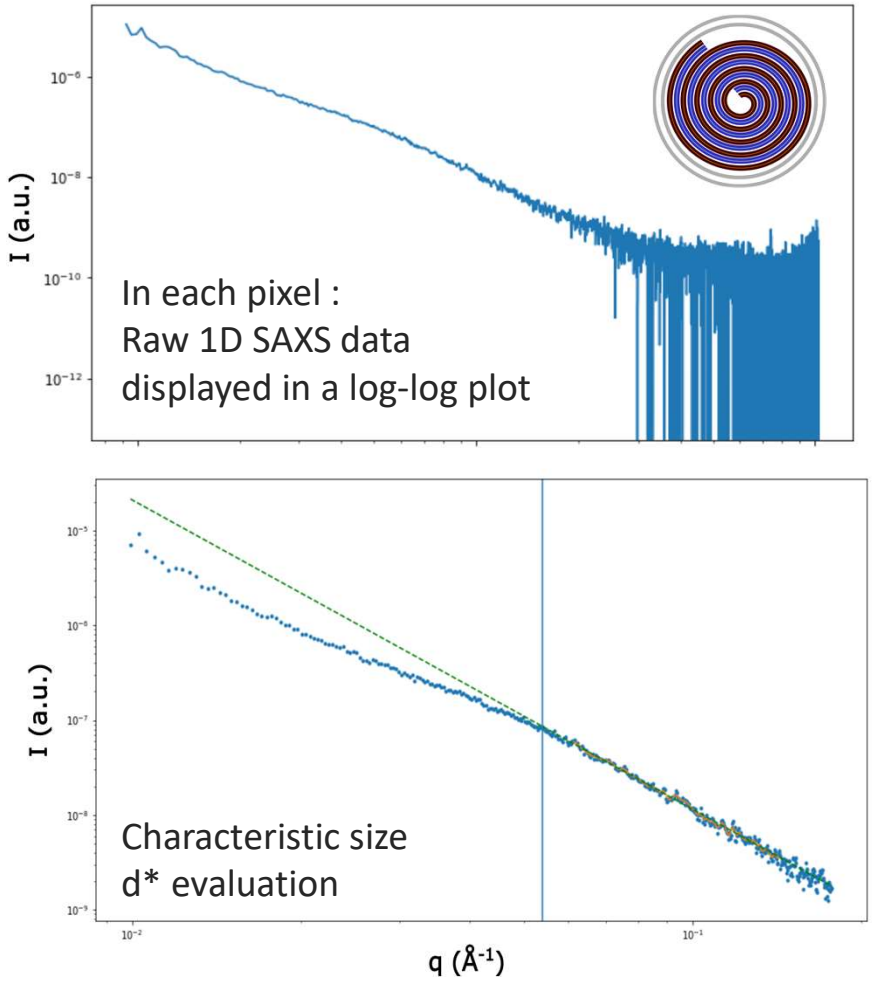
70 keV

Beam 5*10 μm

1 slice-CT

in 5 minutes





E. Lübke et al, Energy Environ. Sci. **17**, 5048-5059 (2024).

Cylindrical industry-grade cell

E. Lübke

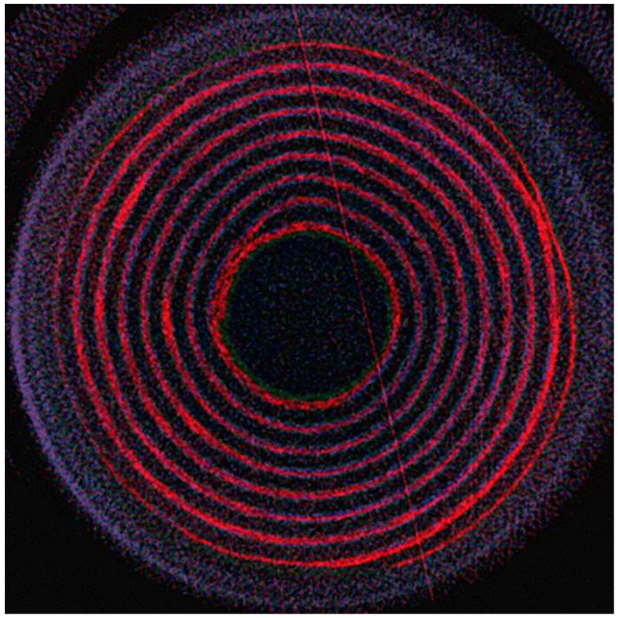
Operando WAXS-CT ID31@ESRF

70 keV

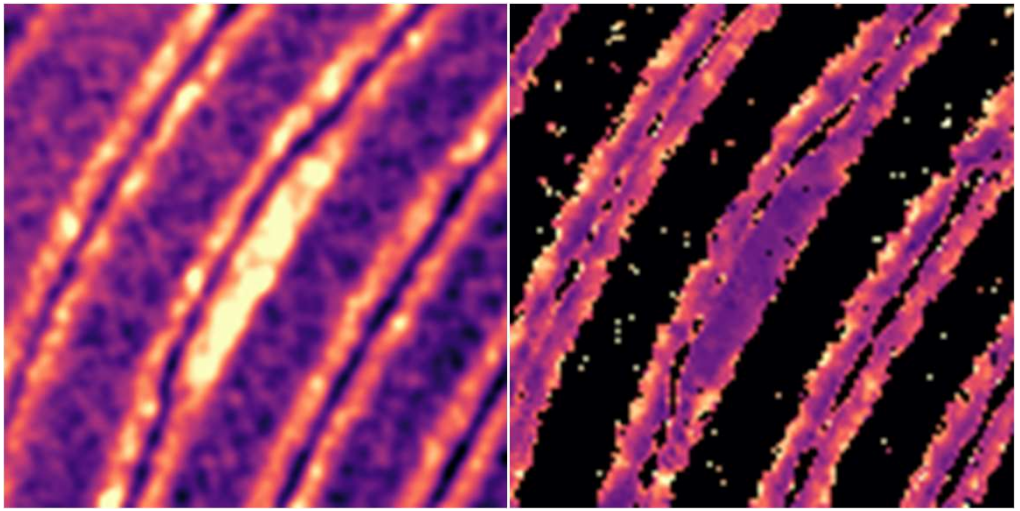
Beam 5*10 μm

1 slice-CT

in 5 minutes



SAXS-CT ID31@ESRF



Intensity ~ Si-content

Si expansion

The defects are due to localized Silicon agglomeration

E. Lübke et al, Energy Environ. Sci. **17**, 5048-5059 (2024).

Cylindrical industry-grade cell

E. Lübke

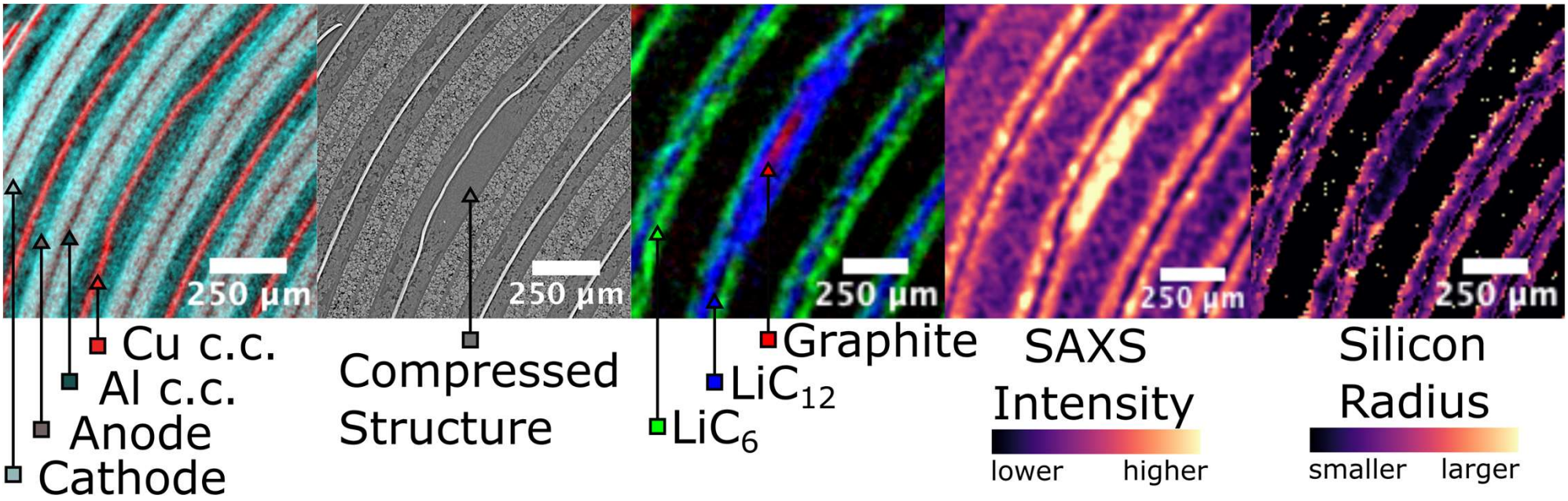
- Correlative analysis unravels the origin of local defects

NeXT:
Deformed areas
Different Li-distribution

High-Res XCT:
Compressed microstructure

WAXS-CT:
Lower graphite lithiation
Slower kinetics

SAXS-CT:
Silicon material accumulation
Lower Si-expansion

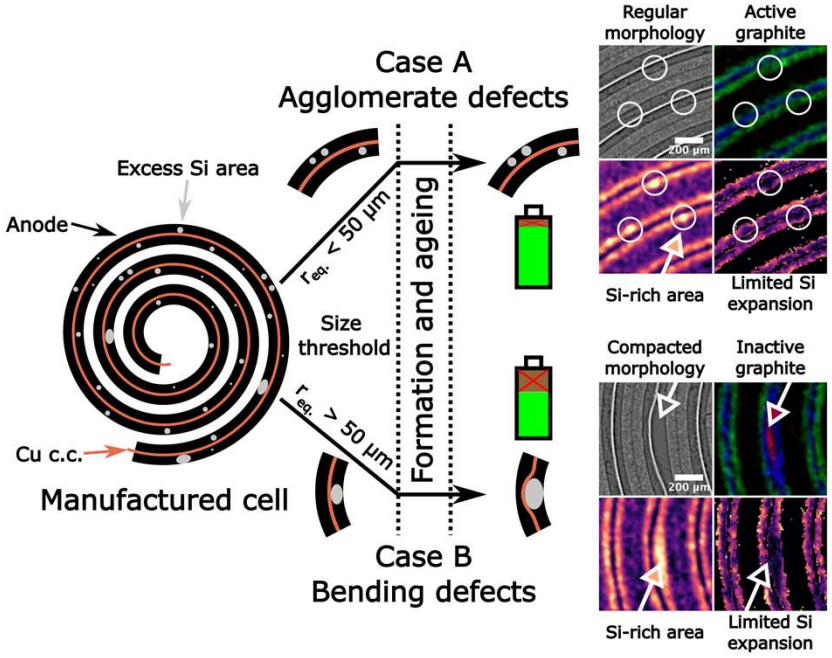


E. Lübke et al, Energy Environ. Sci. **17**, 5048-5059 (2024).

Cylindrical industry-grade cell

E. Lübke

- Correlative analysis unravels the origin of local defects

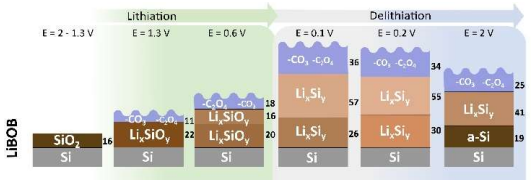


Main Results

- Macroscopic deformations of current collector from 'new'.
- Severe collapse of anode microporosity in defective regions.
- Disrupted lithium diffusion pathways.
- Accumulation of Li and Si caused by Si aggregates during electrode production
- Critical aggregate size is 50 micron above which cell structure/functioning is compromised.

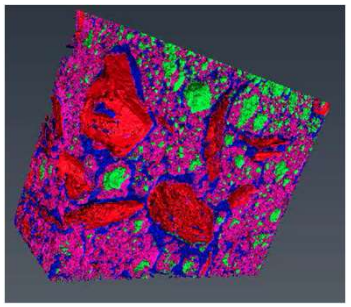
E. Lübke et al, Energy Environ. Sci. **17**, 5048-5059 (2024).

Graphite-Silicon composite anodes

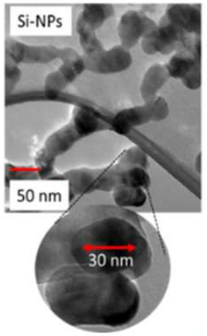


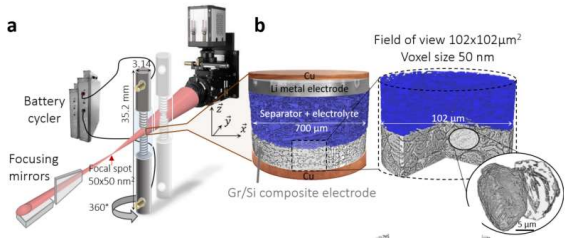
SEI growth and morphology ?
Operando X-ray Reflectivity

Real



Model

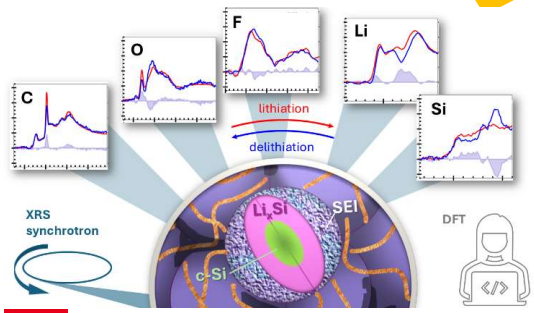




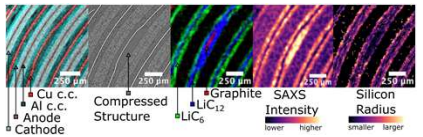
Local particle/electrode scale activity ?
Operando nanoholotomography

Electron microscopy

SEI composition ?
Ex situ X-ray Raman Scattering



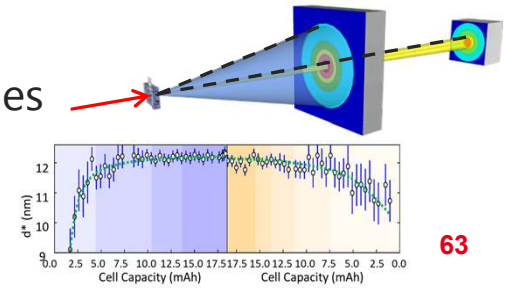
Si-based electrode



Reaction heterogeneities in a cylindrical cell ?
Operando SWAXS-CT and NI

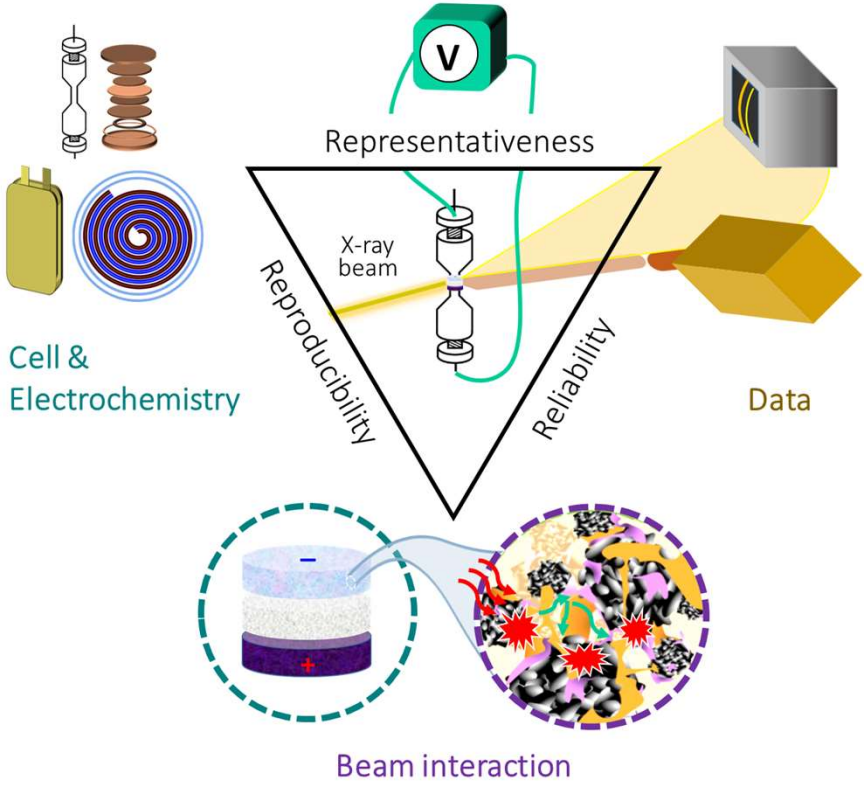
Particle swelling ?
Operando SWAXS/SANS

Reaction heteogeneities in a pouch cell ?
Operando scanning SWAXS



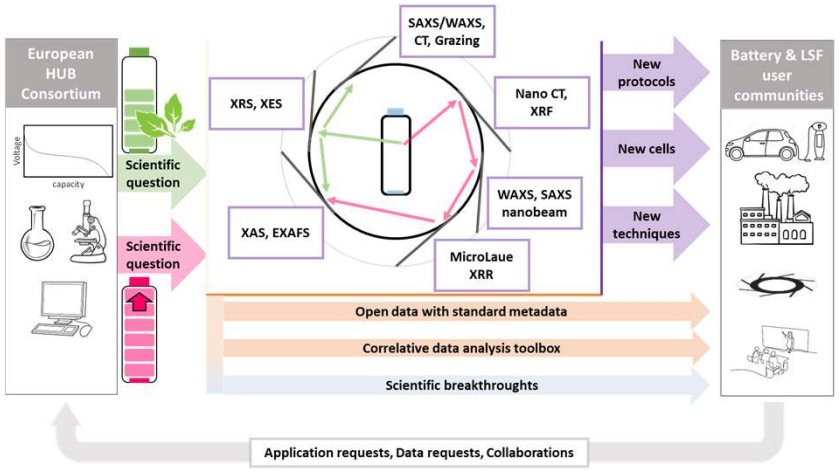
Reinventing the way to do battery research

A novel infrastructure for synchrotron (neutron) studies in Grenoble



Beamtime access
Standardization
Protocols

EUROPEAN BATTERY HUB

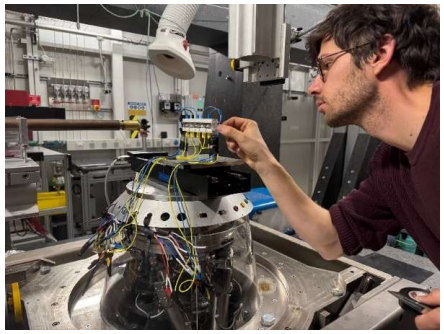


www.europeanbatteryhub.eu

Now launching also the Neutron Battery Hub at ILL !!!!!
with quentin.berrod@cea.fr

J. Drnec and S. Lyonnard. Battery research needs more reliable, representative and reproducible synchrotron characterizations. *Nature Nanotechnology*, 2025
DOI: 10.1038/s41565-025-01921-4

PIs : L. Stievano, A. Matic, D. Bresser, M. Bianchini, L. Daniel, J. Drnec



X. Brems

All Hub members

V. Vanpeene, Q. Jacquet, S. Tardif, Q. Berrod, X. Brems, J. Togonon, G. Oney, A. Olgo, S. Ngoun, M. Fransson, S. Mitra

THANK YOU

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ID26 – B. Deflet
BM32 – S. Tardif, J-S. Micha
ID16b – V. Vanpeene, J. Villanova, O. Stamati
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ID15 – G. Vaughan, M. Di Michiele
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BM26 – M. Rosenthal BM23 – C. Atzori
ID11 – C. Giacobbe ID22 – O. Gjonnes Grendal

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D33 – B. Cubitt
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E. Lübke