

In situ

Microscop

Gas

Heating

Cooling

Thermal

rradiation

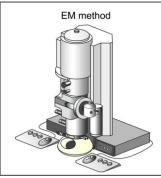
Mechanical

Atmosphere

ombinatorial

iaht

Liquid



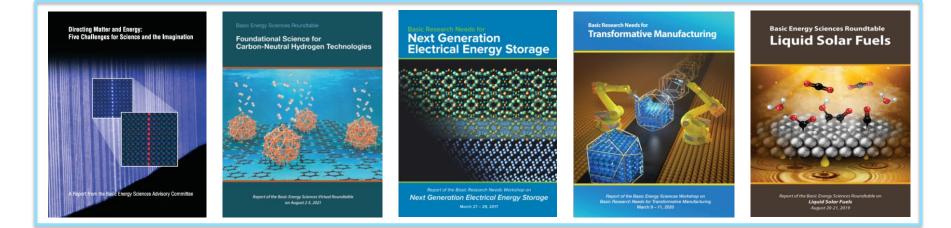
Operando ElectronMicroscopy for EnergyGeneration, Use, and Storage

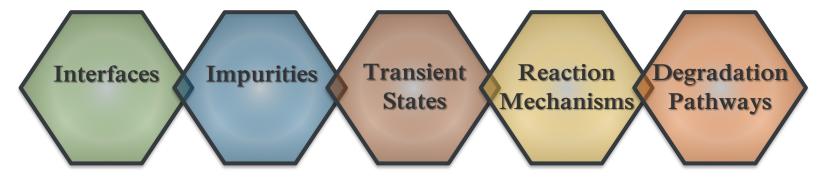
Katherine Jungjohann

Group Manager, Analytical Microscopy and Imaging Sciences

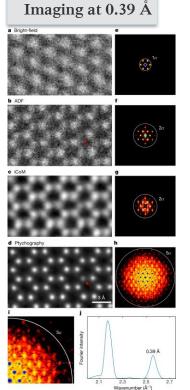
BESAC Virtual Meeting July 14th, 2022

Atomic Scale Foundational Energy Phenomena



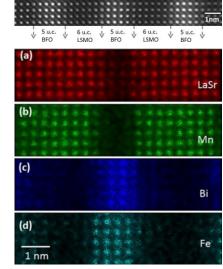


Why Electron Microscopy? Atomic Scale Detail

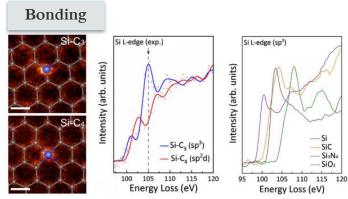


Jiang et al. Nature 559, 343 (2018)

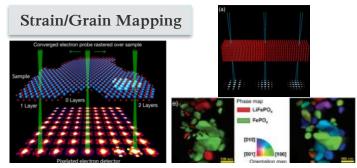
Composition Maps



Lu et al. Appl. Phys. Lett. 102, 17311 (2013)



Zhou et al. Phys. Rev. Lett. 109, 206803 (2012)



Ophus et al. Microsc. Microanal. 25, 563 (2019)

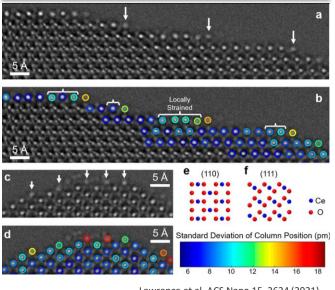
Obtain this information for operando phenomena in energy systems?

Operando Electron Microscopy



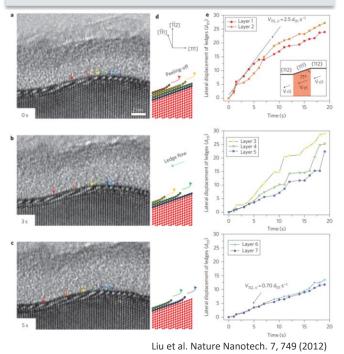
Atomic Scale Operando Electron Microscopy

CeO₂ Catalyst : Atomic Displacements



Lawrence et al. ACS Nano 15, 2624 (2021)

Silicon Nanowire: Lithiation



Atomic-scale operando imaging is simpler with electron beam resistant materials/interfaces

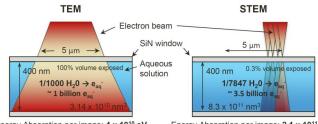
Limitations with Atomic Scale Operando EM

Beam Effects

Radiolysis of Liquids

Lithium Metal

 $H_2O \rightarrow 2.7 e_{aq}^- + 2.7 H^+ + 0.61 H + 0.43 H_2 + 2.87 OH + 0.61 H_2O_2 + 0.026 HO_2$



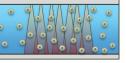
Energy Absorption per image: 4 x 10¹⁰ eV Energy Absorption per image: 2.1 x 10¹¹ eV * not drawn to scale, 40 pA beam current 0.00422 e_{ag}⁻/nm³ 0.0318 e_{ag}-/nm³





0.2 µm

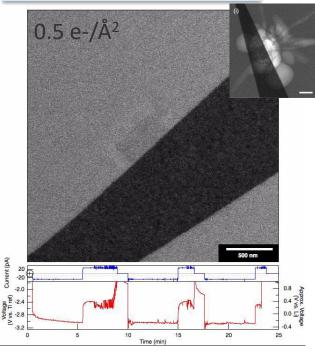




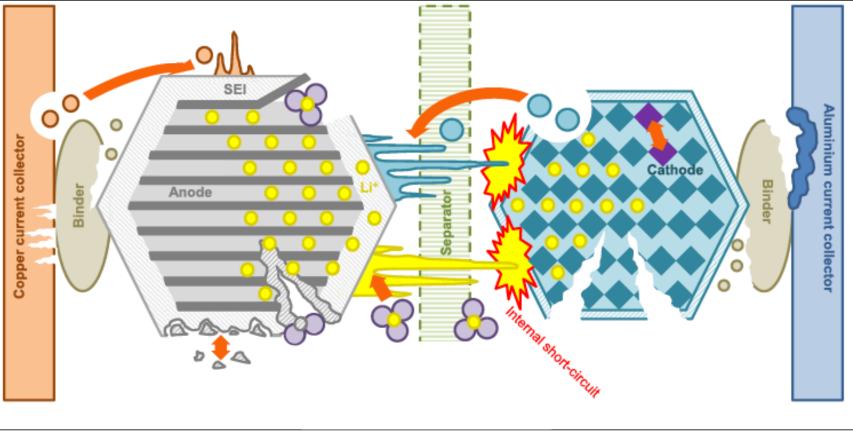
Current (pA)



Li Electrodeposition Structure

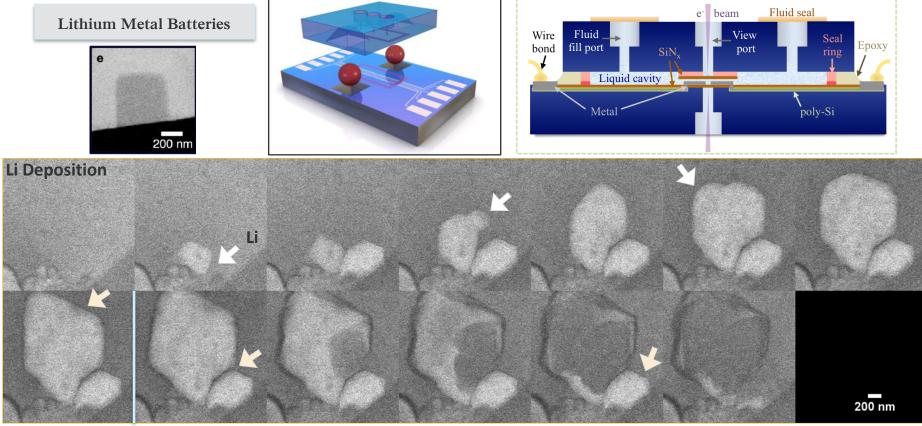


Battery Degradation Pathways



Operando Information Unlocks Pathways



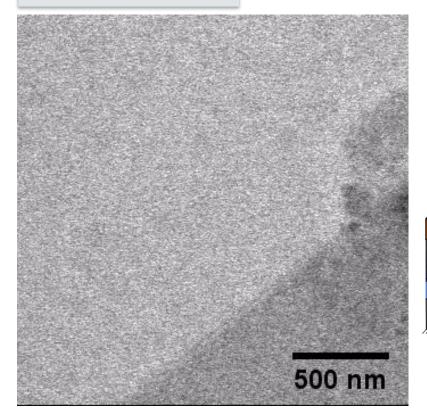


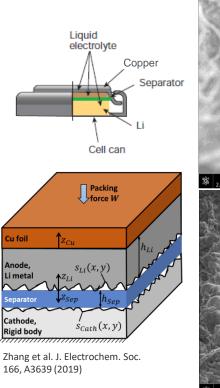
Li Stripping

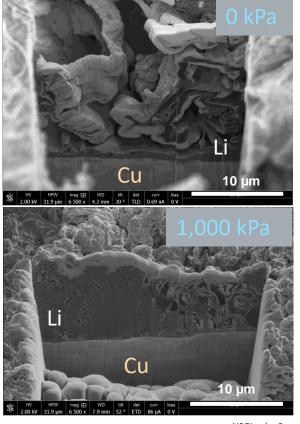
Operando Information Unlocks Pathways

JEESR

Lithium Metal Batteries



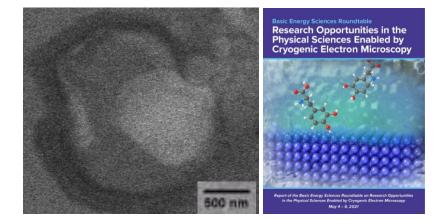


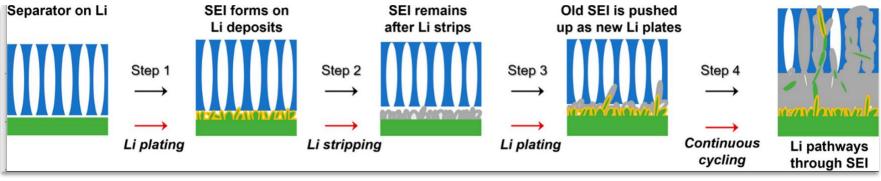


Operando EM Supported by Cryo EM

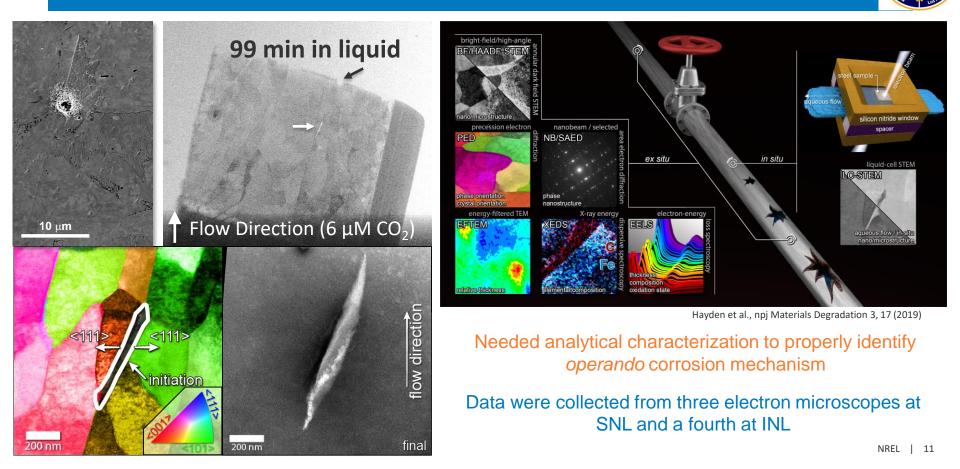


Harrison et al., iScience 24, 103394 (2021)





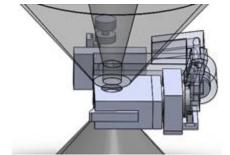
Often Need Capabilities of Many Optimized EMs



Operando Electron Microscopy at NSRCs



Extreme Environment TEM





Center for Integrated Nanotechnologies Sandia National Laboratories Los Alamos National Laboratory

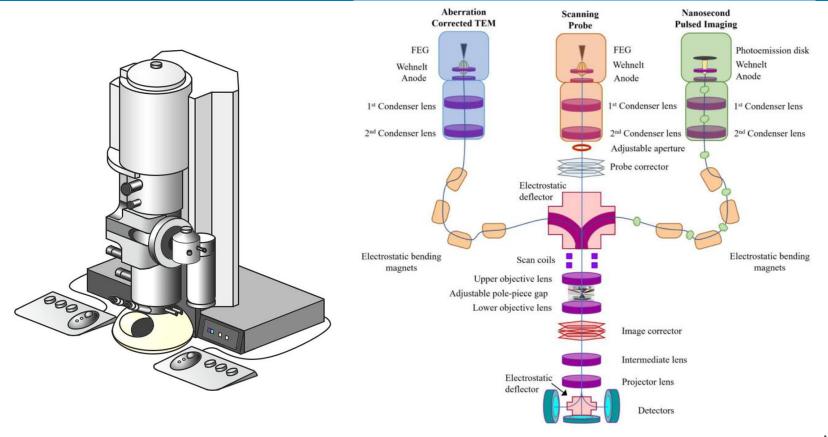


Center for Nanophase Materials Sciences Oak Ridge National Laboratory

Liquid-He MAC-STEM

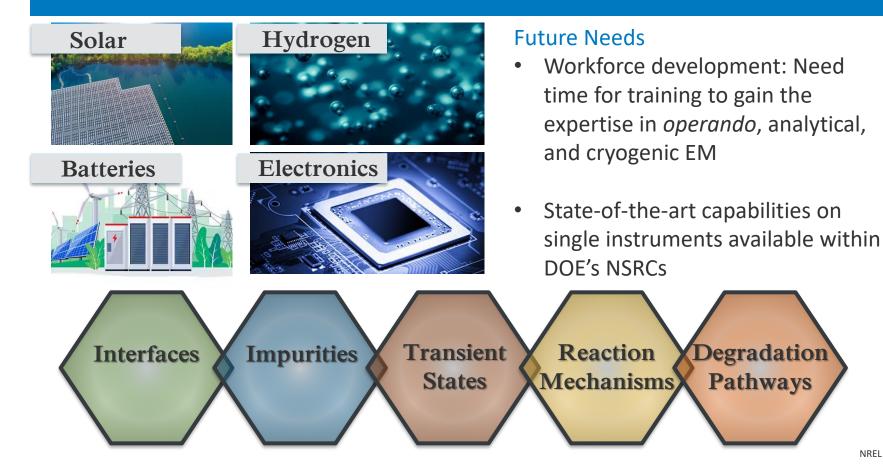


Optimized Operando Electron Microscope



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Concluding Remarks





Renae Gannon









Dave Johnson



Steven Randolph



Daniel Long



Katharine Harrison Laura Merrill Kevin Zavadil **Khalid Hattar** Andrew Leenheer



Steven Hayden

Thank You BES!

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