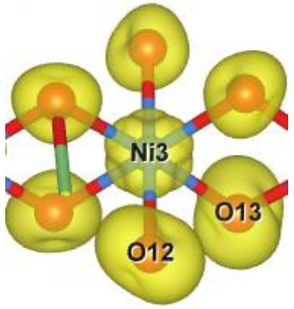

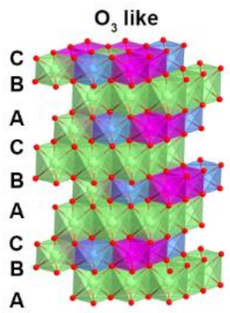

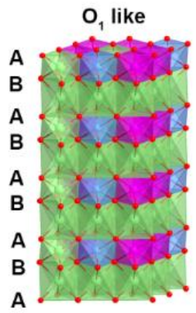

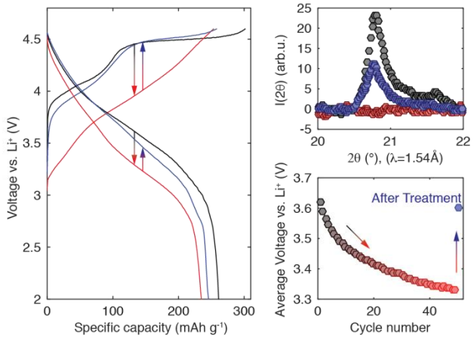

Structure and Voltage Recovery Driven by Defects Elimination in Li-rich Layered Oxide Cathode Materials

Minghao Zhang

Y. Shirley Meng

Department of NanoEngineering

University of California San Diego

Tackle Multi-scale Issues

Professor Ying Shirley Meng

DFT study:
Mr. Thomas Wynn (UCSD)
Dr. Marco Olguin (UCSD)

(S)TEM study:
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Dr. Lijun Wu (BNL)

Modification:
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ND/NPDF study:
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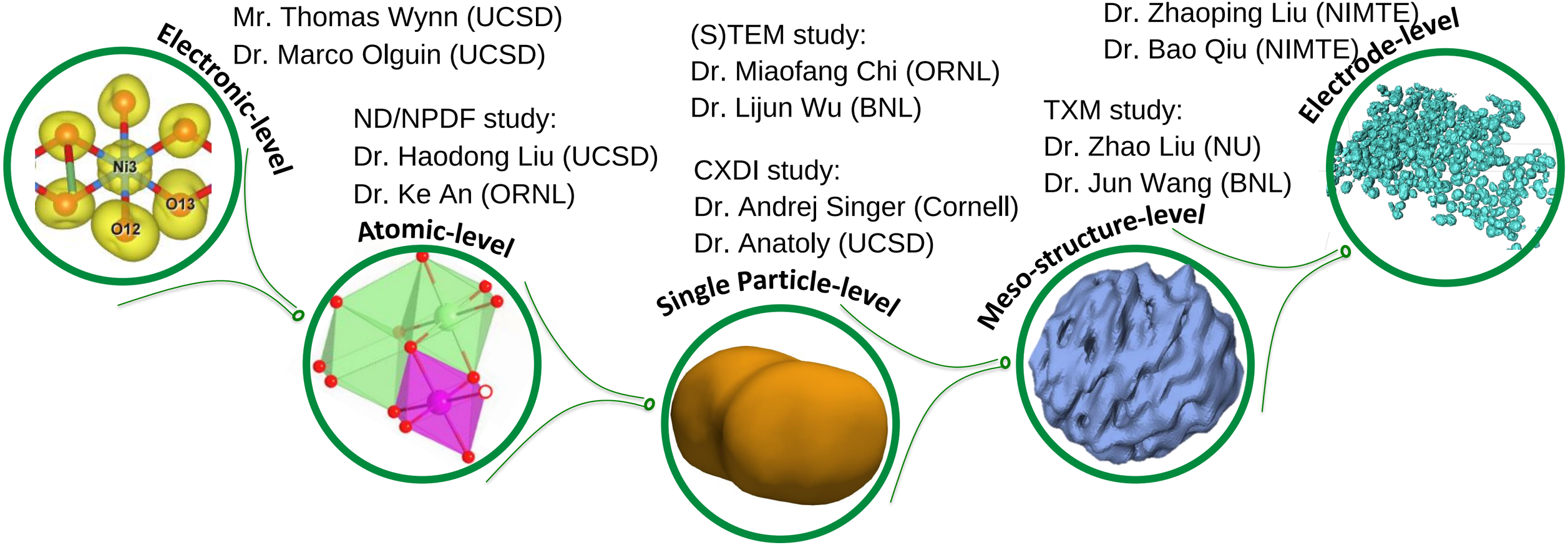
Dr. Ke An (ORNL)

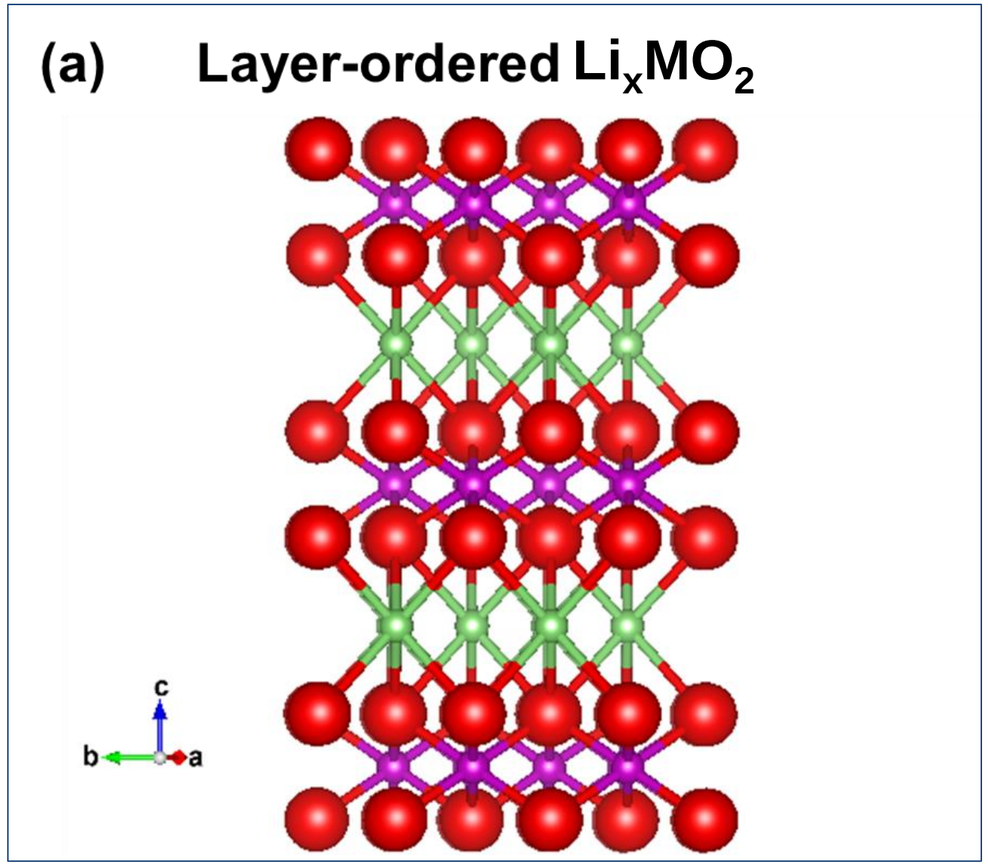
CXDI study:
Dr. Andrej Singer (Cornell)
Dr. Anatoly (UCSD)

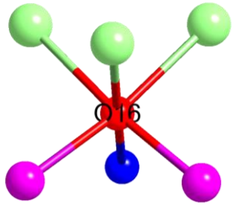
TXM study:
Dr. Zhao Liu (NU)
Dr. Jun Wang (BNL)

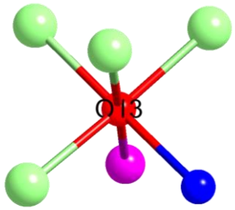


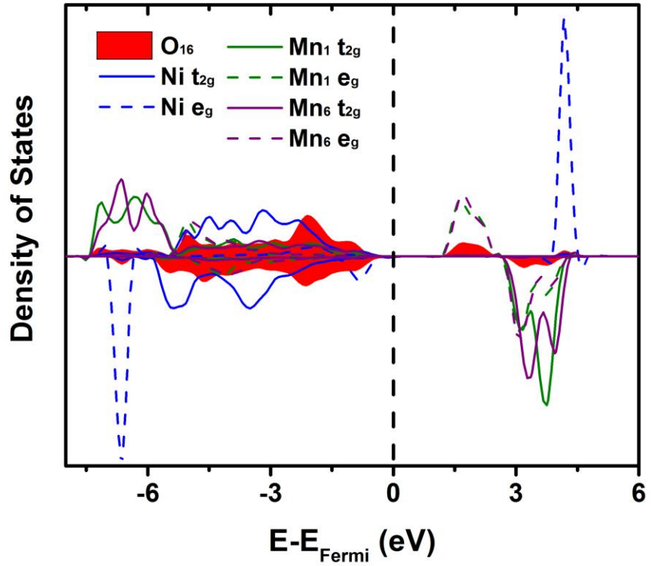
Funding Sources: Energy Efficiency and Renewable Energy, Office of Vehicle Technologies, U.S. Department of Energy under Contract No. DE-AC02-05CH11231, Subcontract No. 7073923 under the Advanced Battery Materials Research (BMR) Program

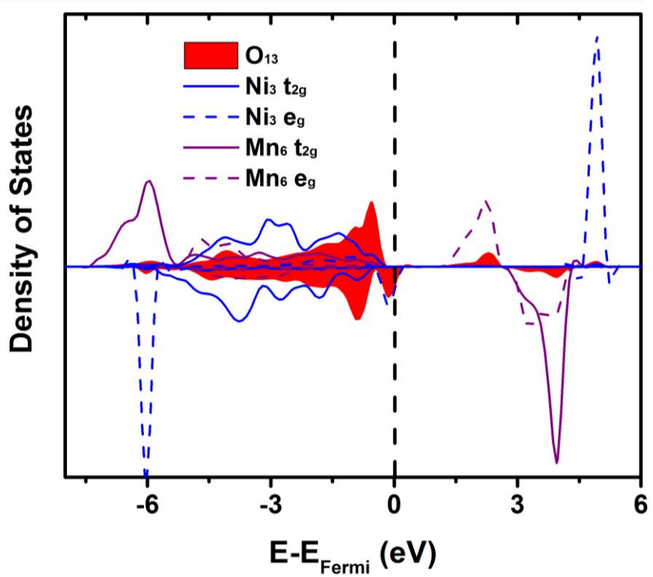
Layered-ordered Structure



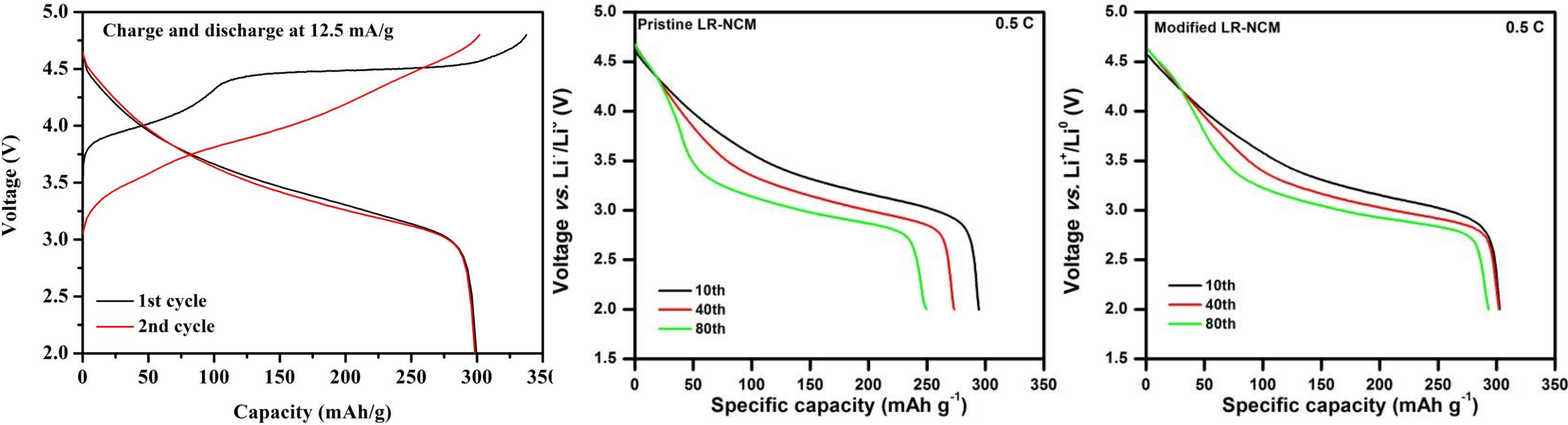








Electrochemical Performance

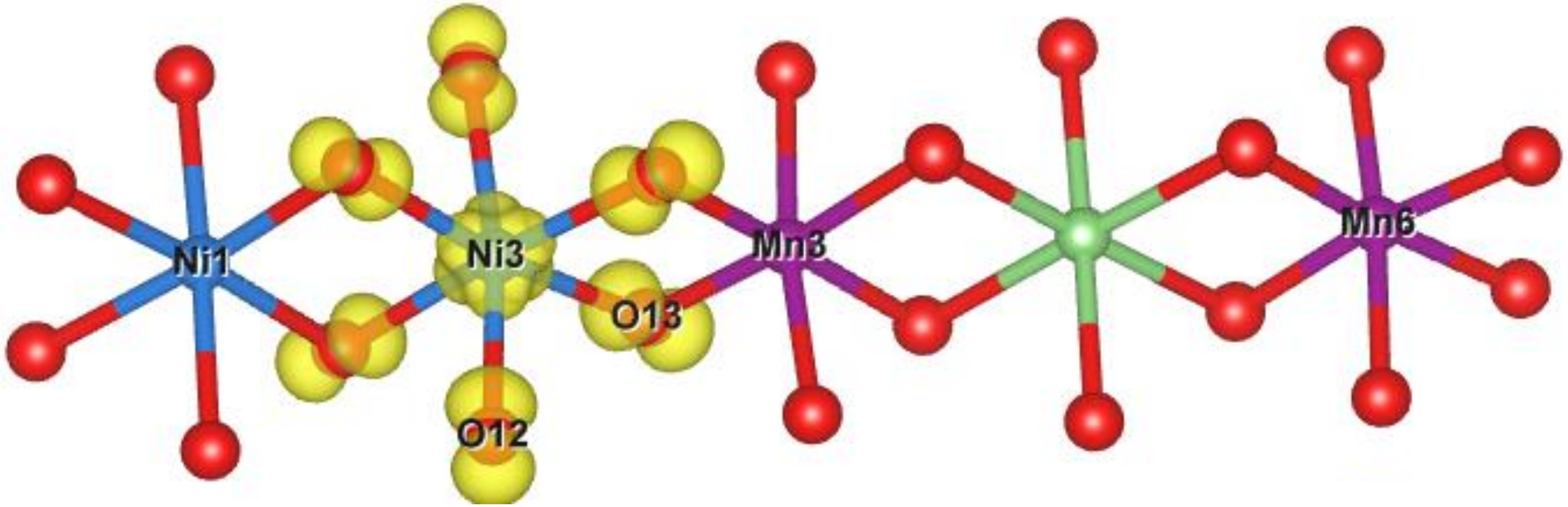


- ❑ Anionic (oxygen) activity is activated in Li-rich layered oxides within the same operating voltage range.
- ❑ Irreversible phase evolution is expected in Li-rich layered oxides after oxygen oxidation.
- ❑ Large irreversible capacity still exists in the first cycle of classical layered oxides.

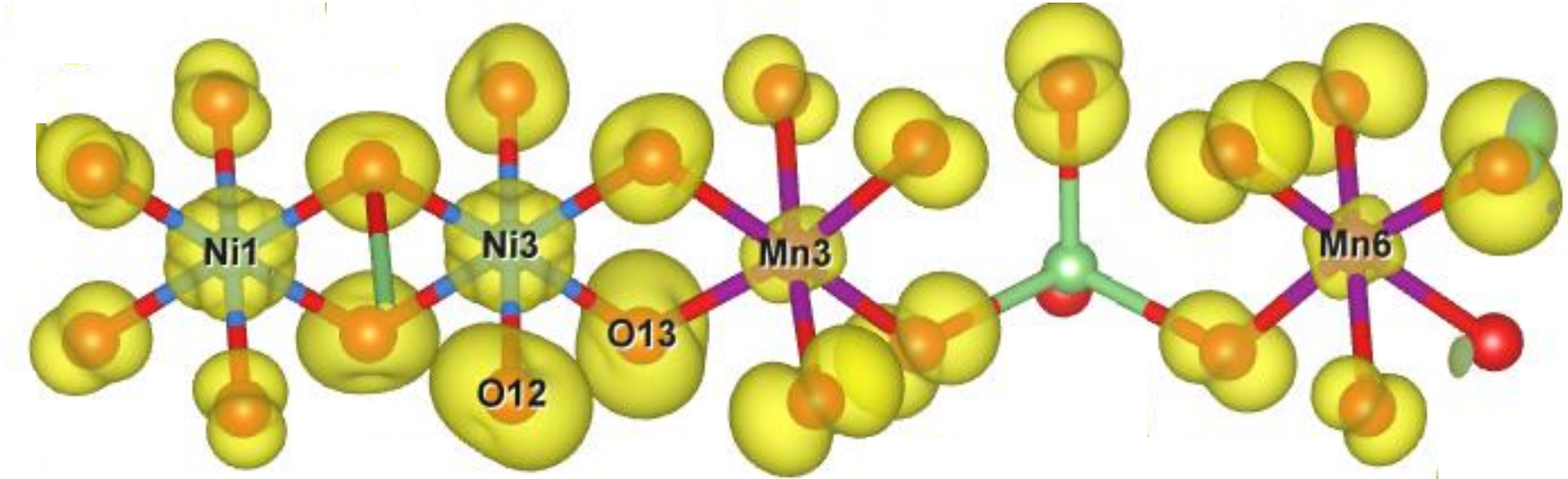
Electronic-level

3D isosurface contour plots of partial charge density (-0.5 eV, 1.5 eV) at an 0.015 electron/Å^3 isosurface value

$Li_{12}[Li_2Ni_3Mn_7]O_{24}$

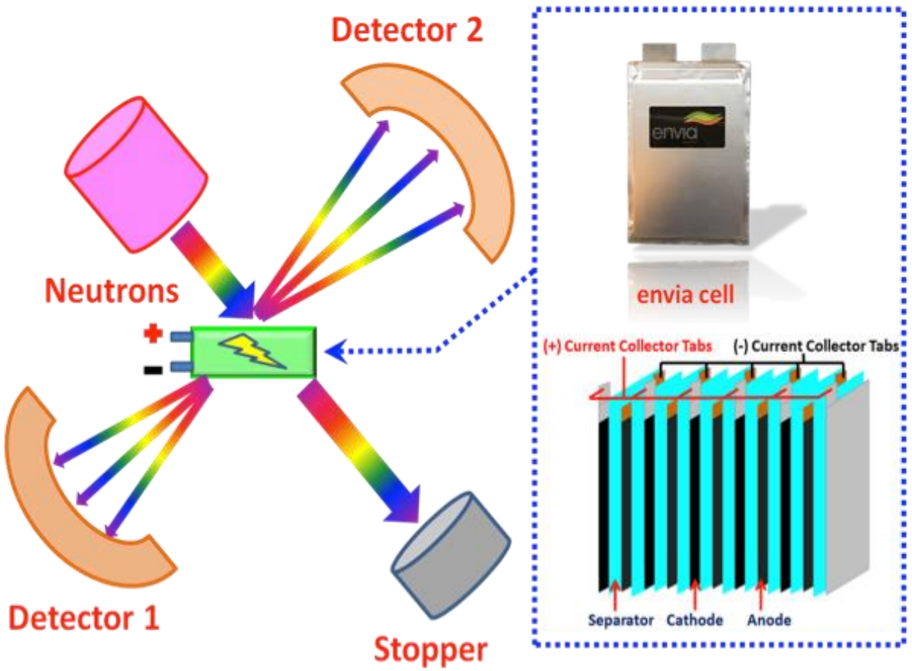


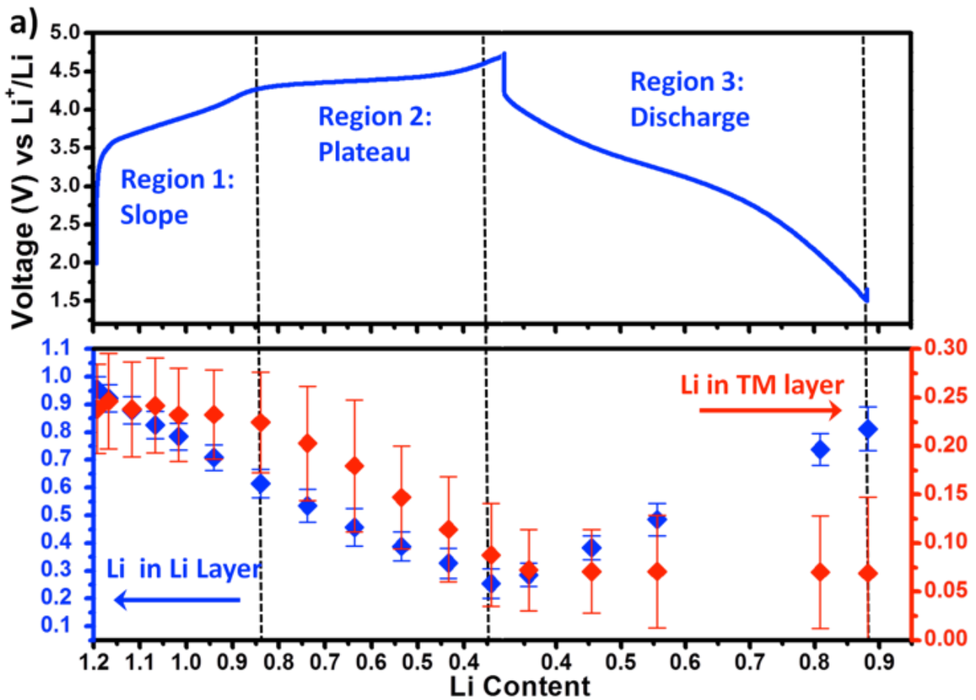
$Li_4[Ni_3Mn_7]O_{24}$



Atomic-level

Operando Neutron Diffraction Characterization

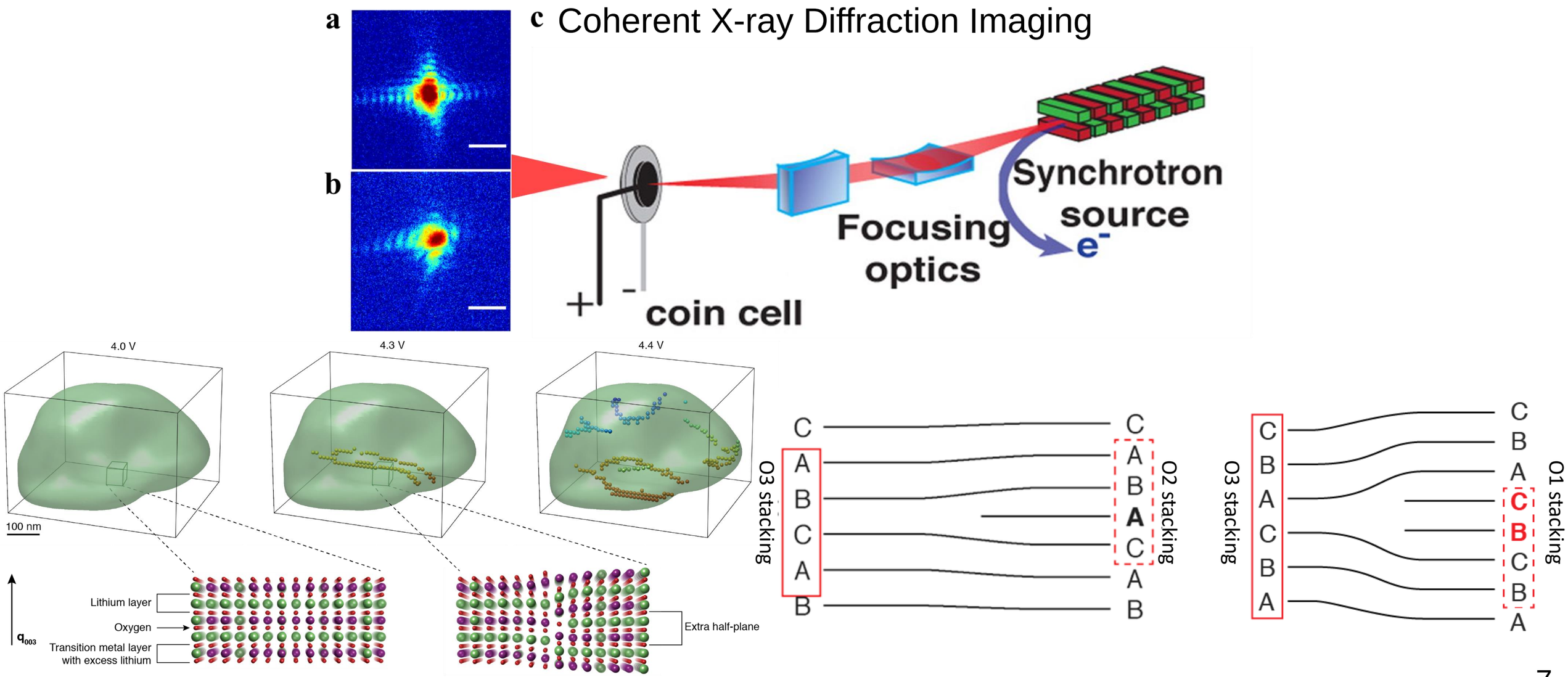




- Li in Li layer is extracted out first.
- Most of Li in TM layer is extracted out during the plateau region.
- Li is inserted back to Li layer during discharge process.

H. D. Liu, Y. S. Meng et al., Adv. Energy Mater. 2016

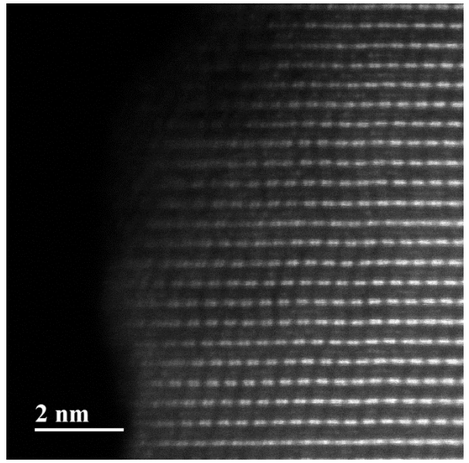
Single Particle-level

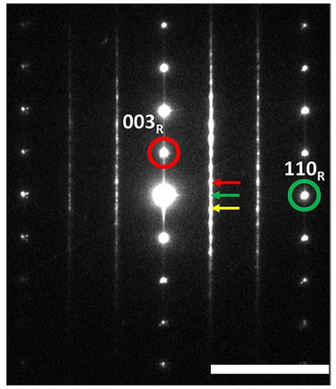


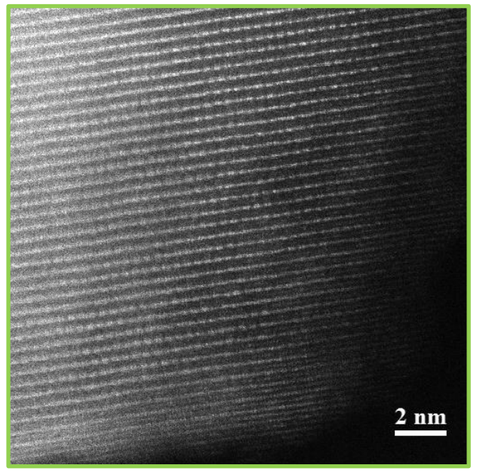
A. Singer, M. Zhang, S. Hy, Y. S. Meng et al., 2018, under review

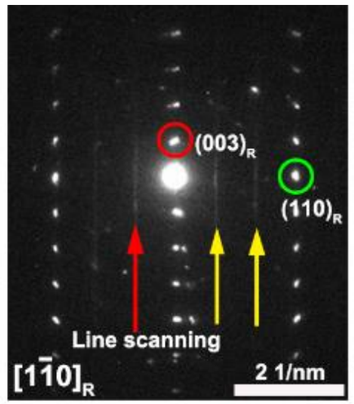
Single Particle-level

Pristine



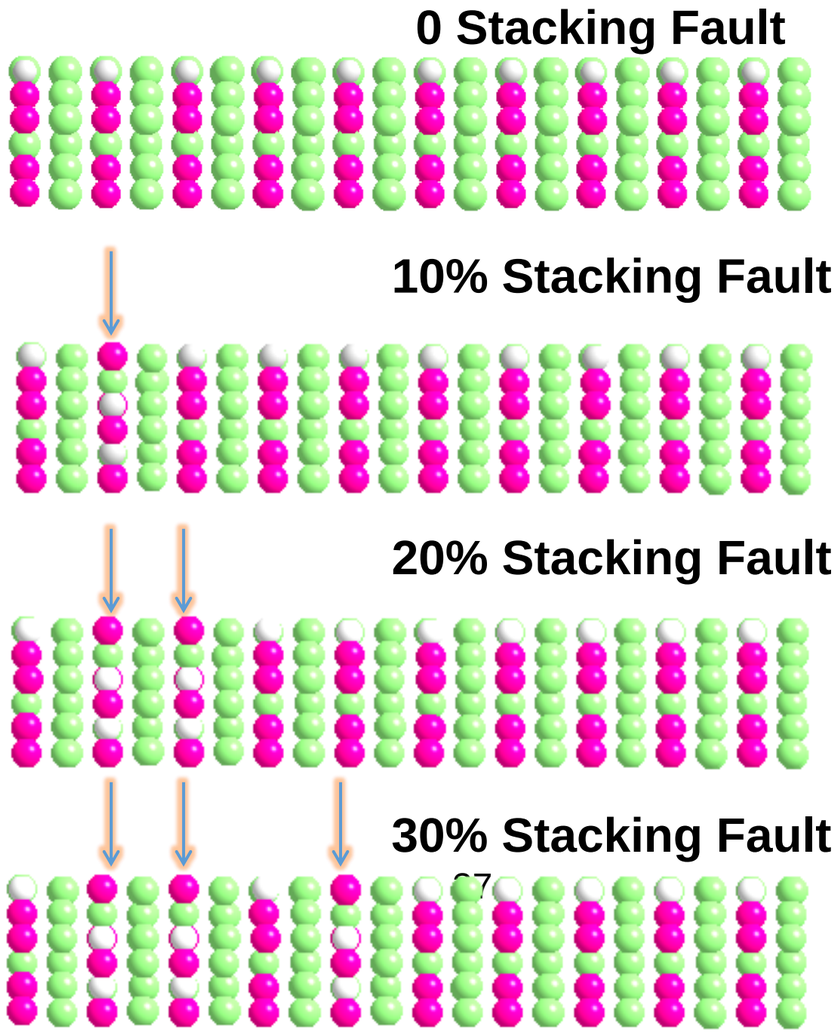


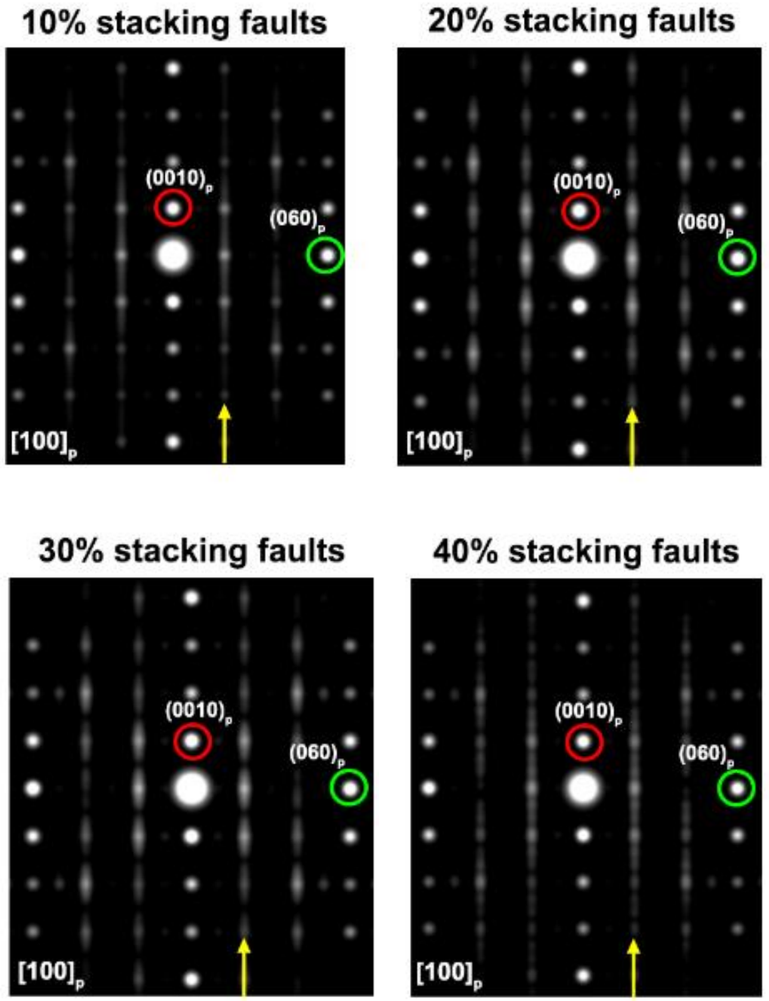




After 1 cycle

Experimental

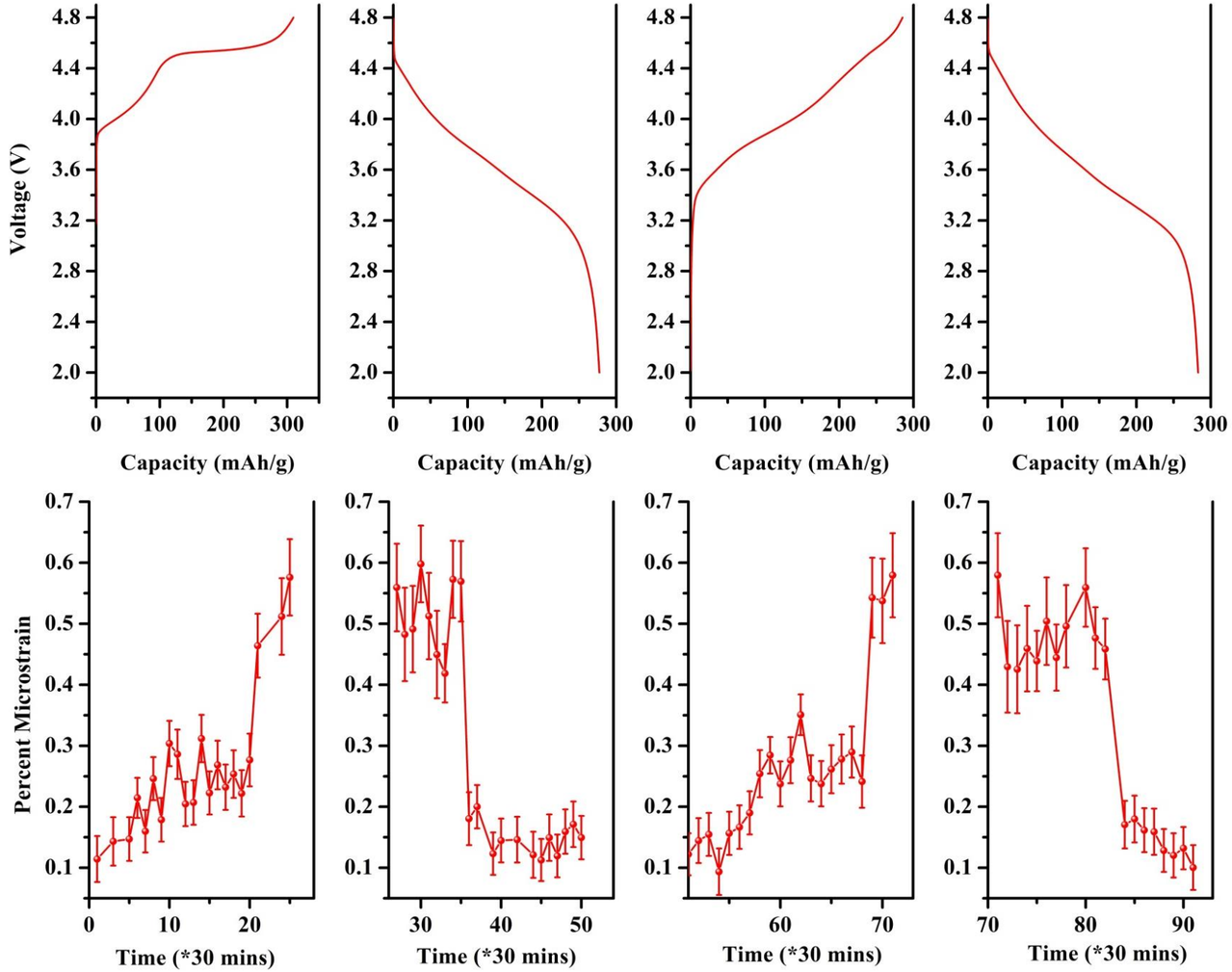


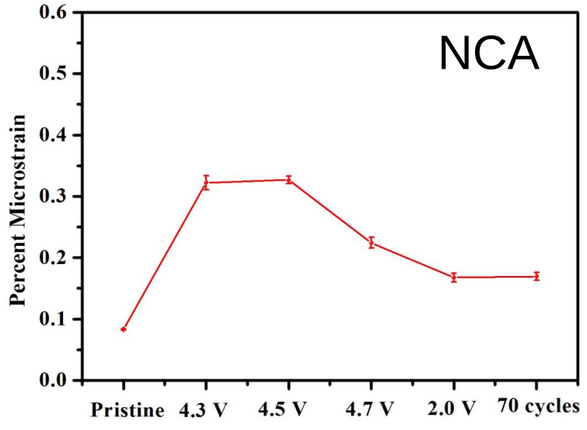


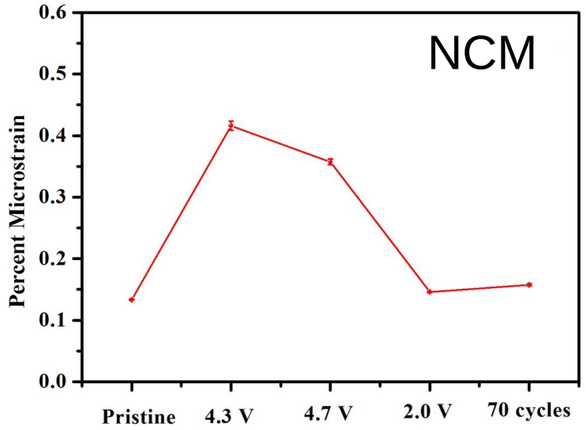
Simulated

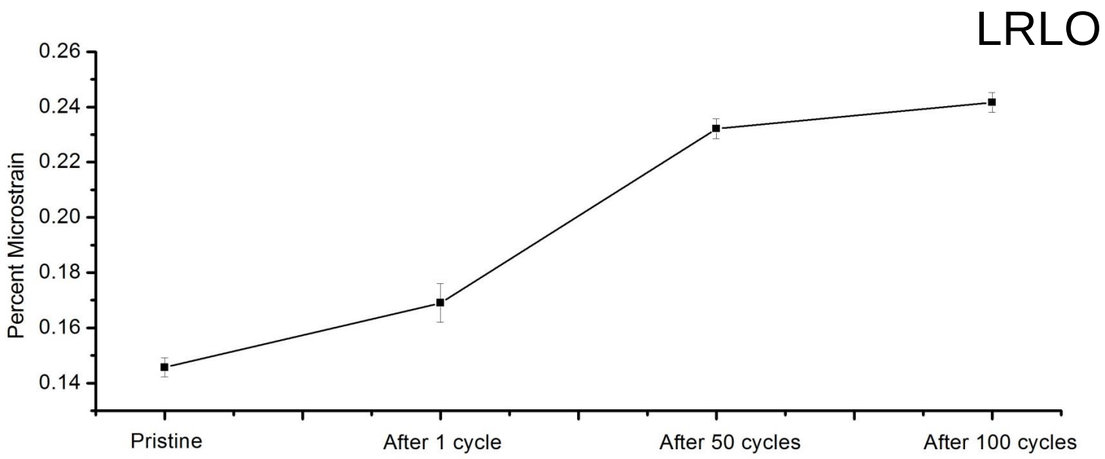
B. Qiu+, M. Zhang+, Y. S. Meng et al., 2018, in submission

Single Particle-level









Defects Generation and Voltage Decay

O3 stacking

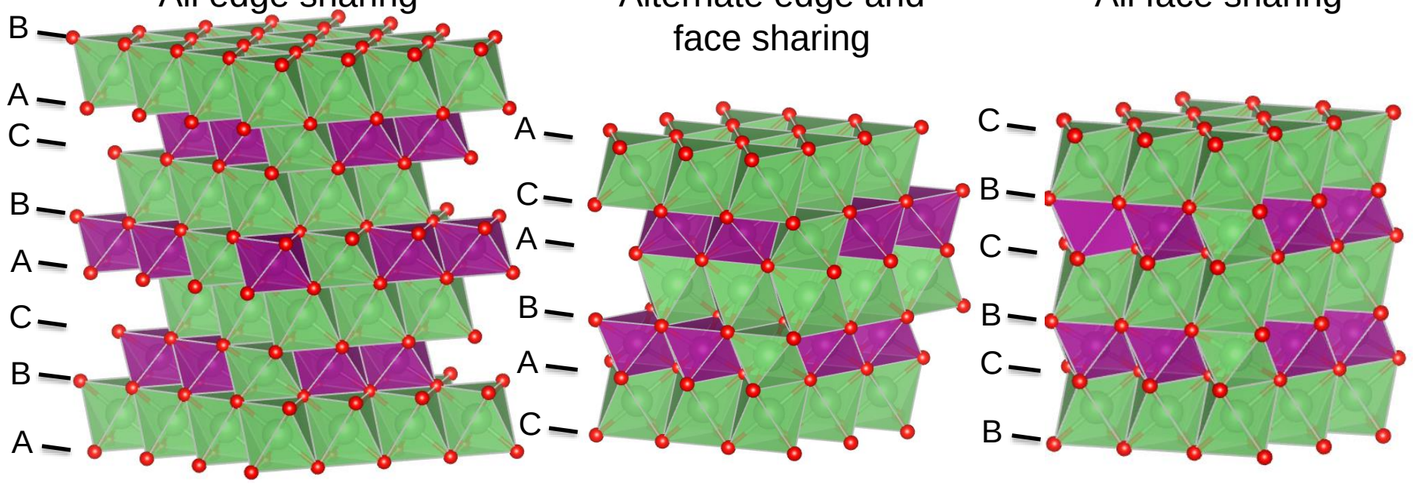
All edge sharing

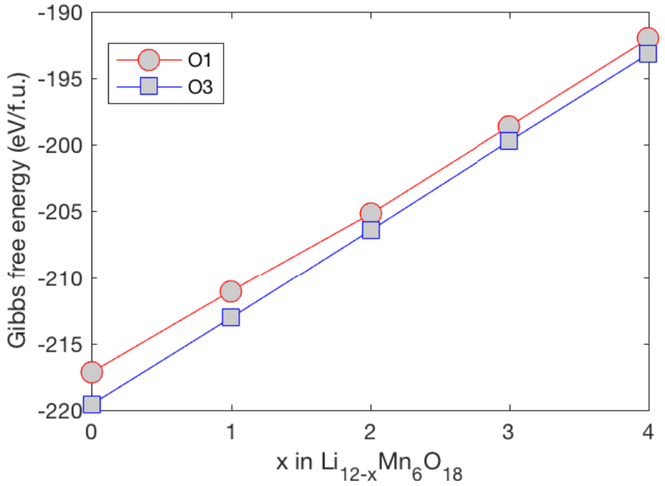
O2 stacking

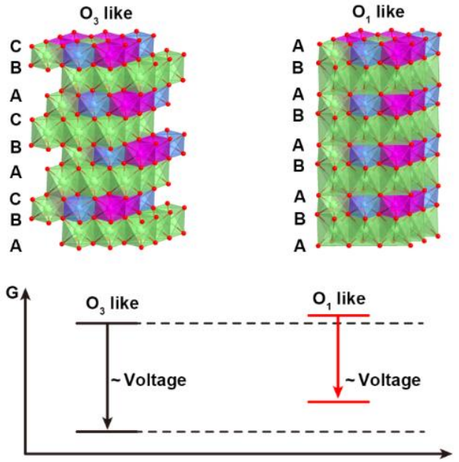
Alternate edge and face sharing

O1 stacking

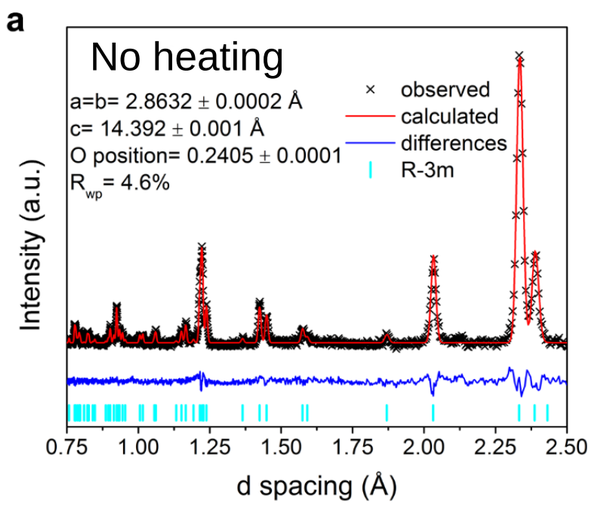
All face sharing

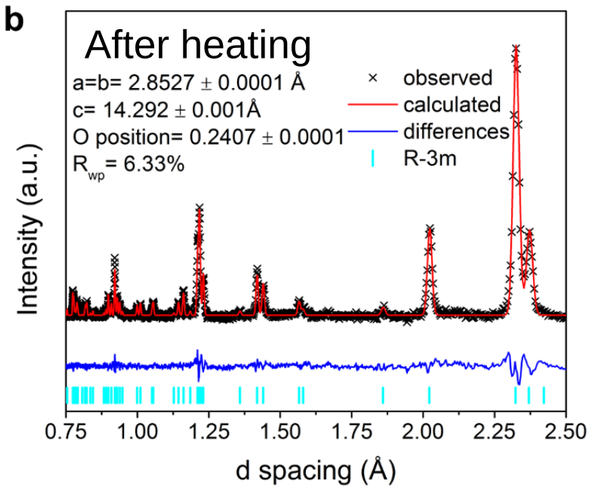


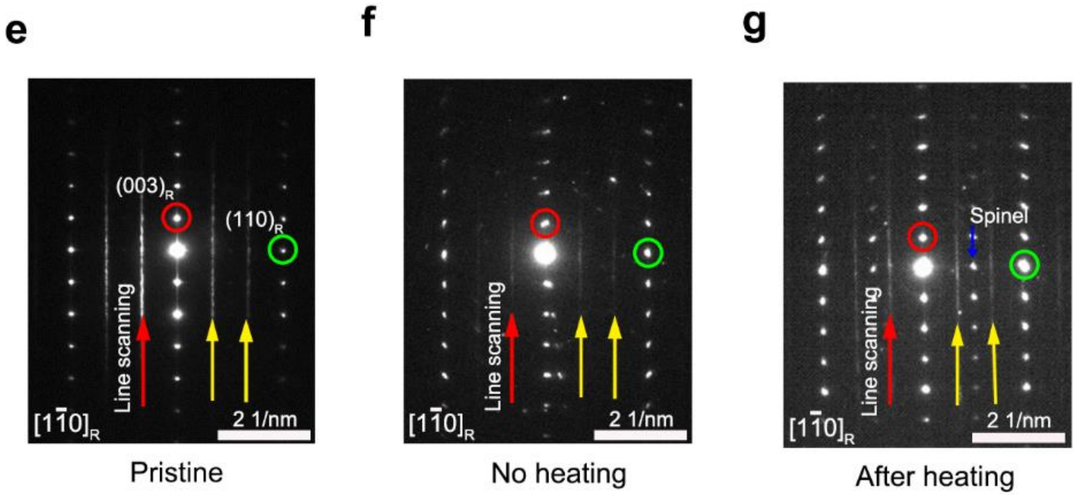


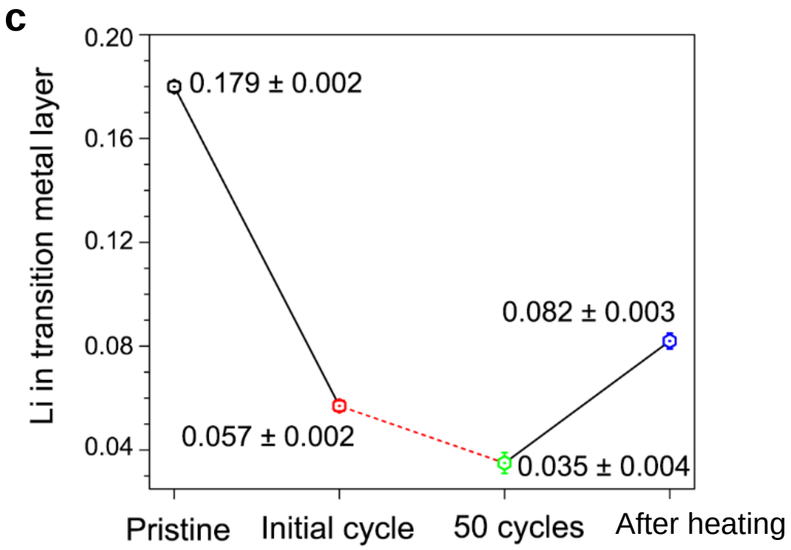


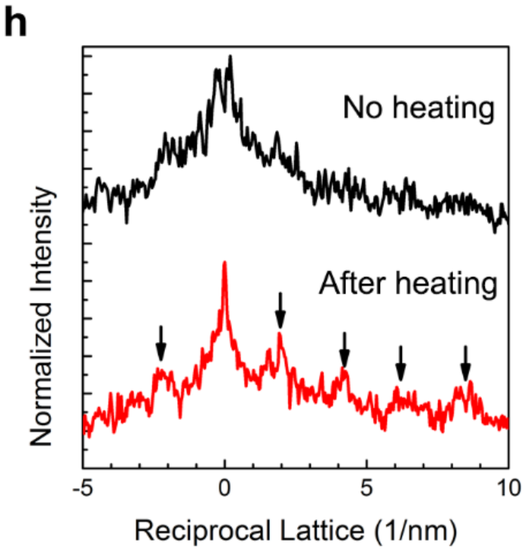
Defects Elimination Driven by Heat



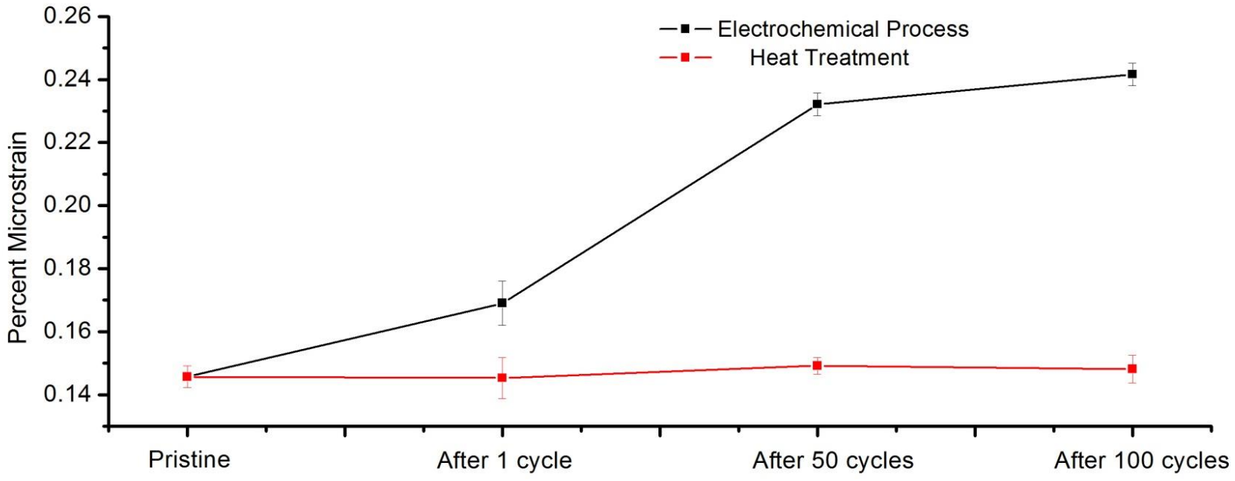


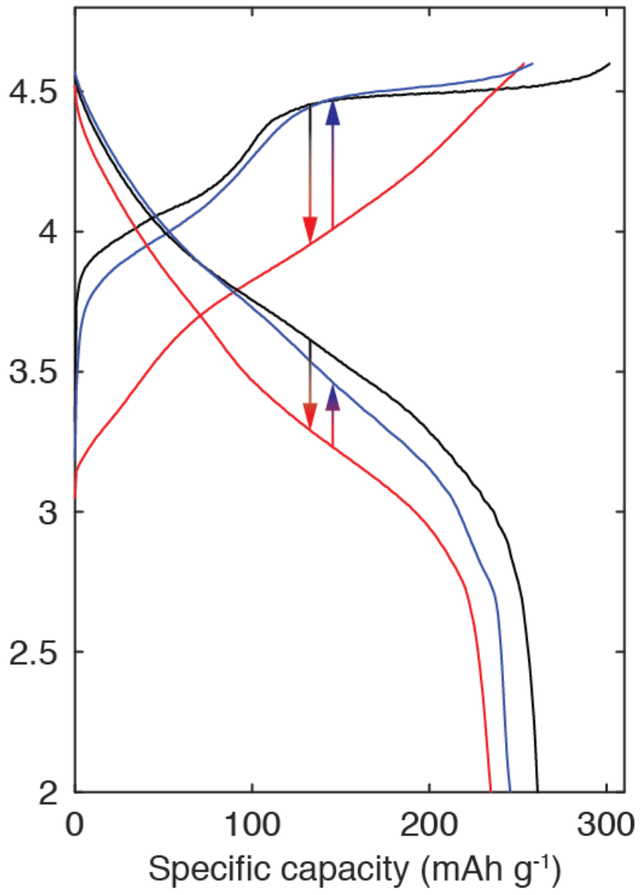


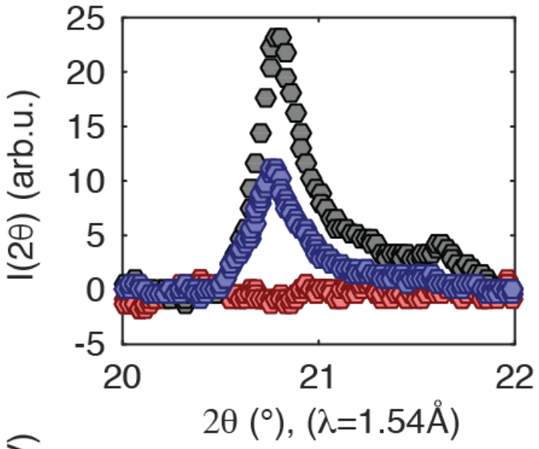


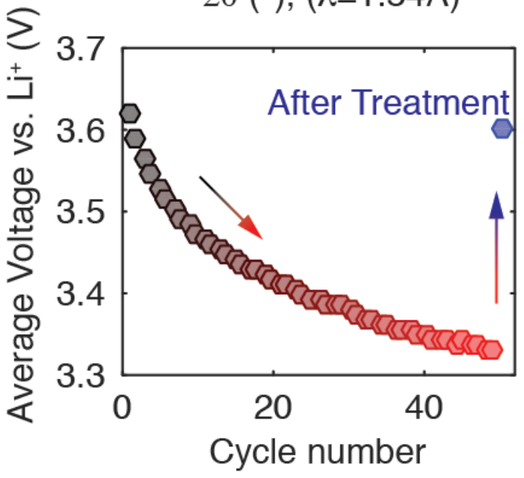


Voltage Recovery

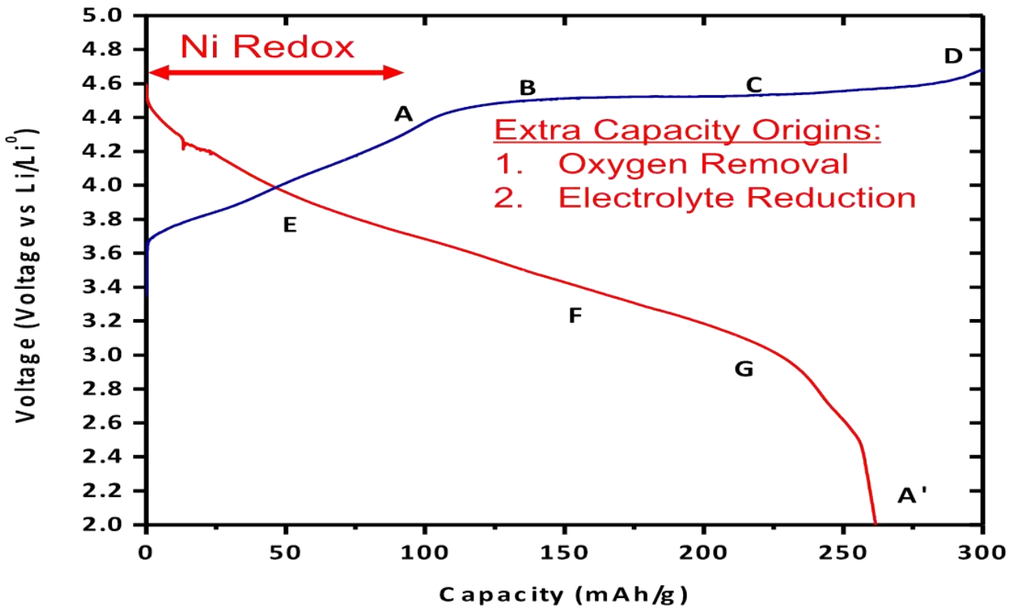




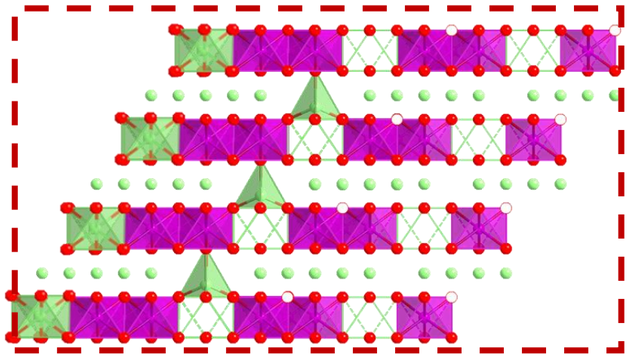




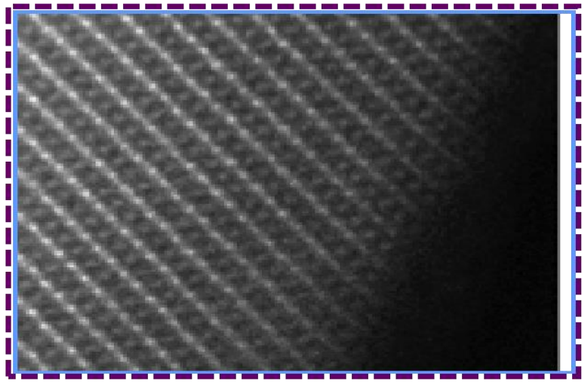
Summary and Outlook



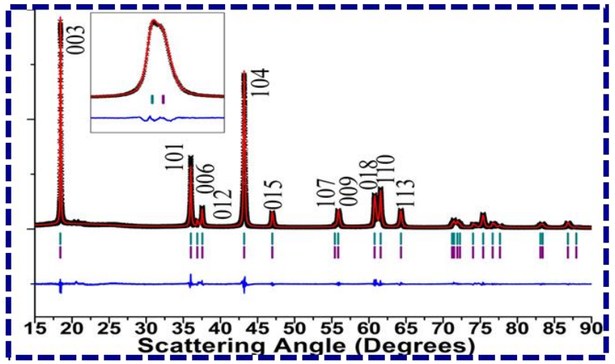
Atomistic



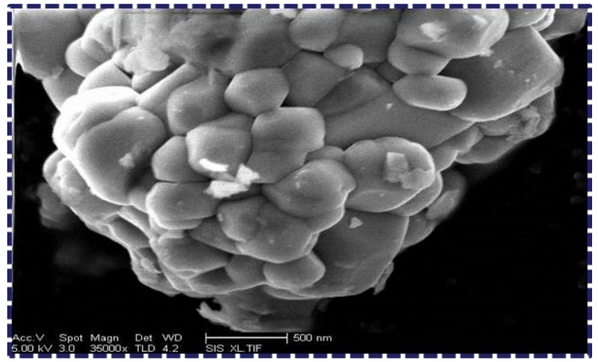
Interface/Surface



Structural



Microstructural



THANK YOU