Oak Ridge National Laboratory

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Understanding Battery Degradation for Grid Energy Storage Systems

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ID# - 902

U.S. DEPARTMENT of ENERGY

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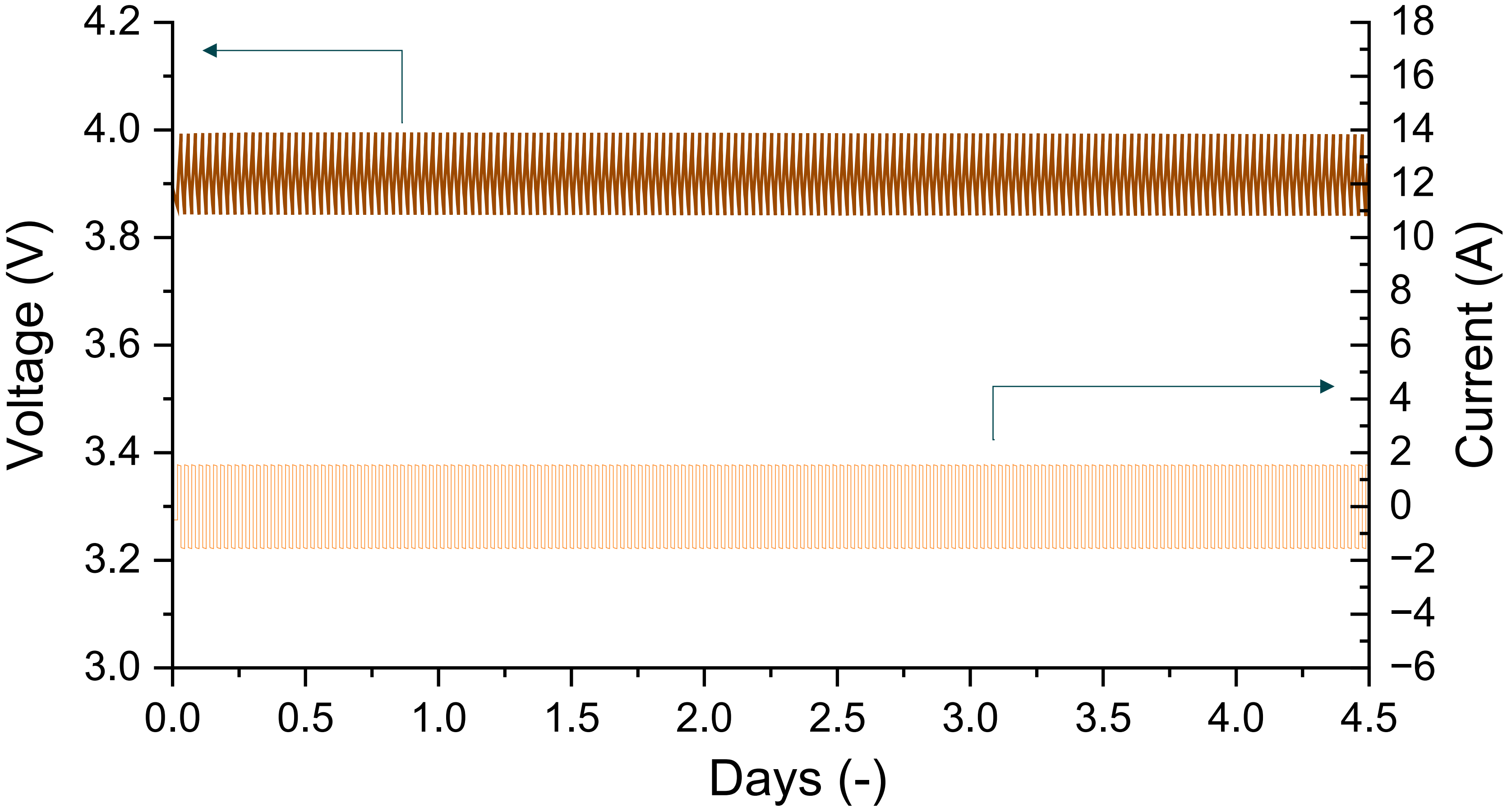
Project Overview



- Current battery performance and lifetime models are based on uniform, factory-new cells tested under simplified conditions

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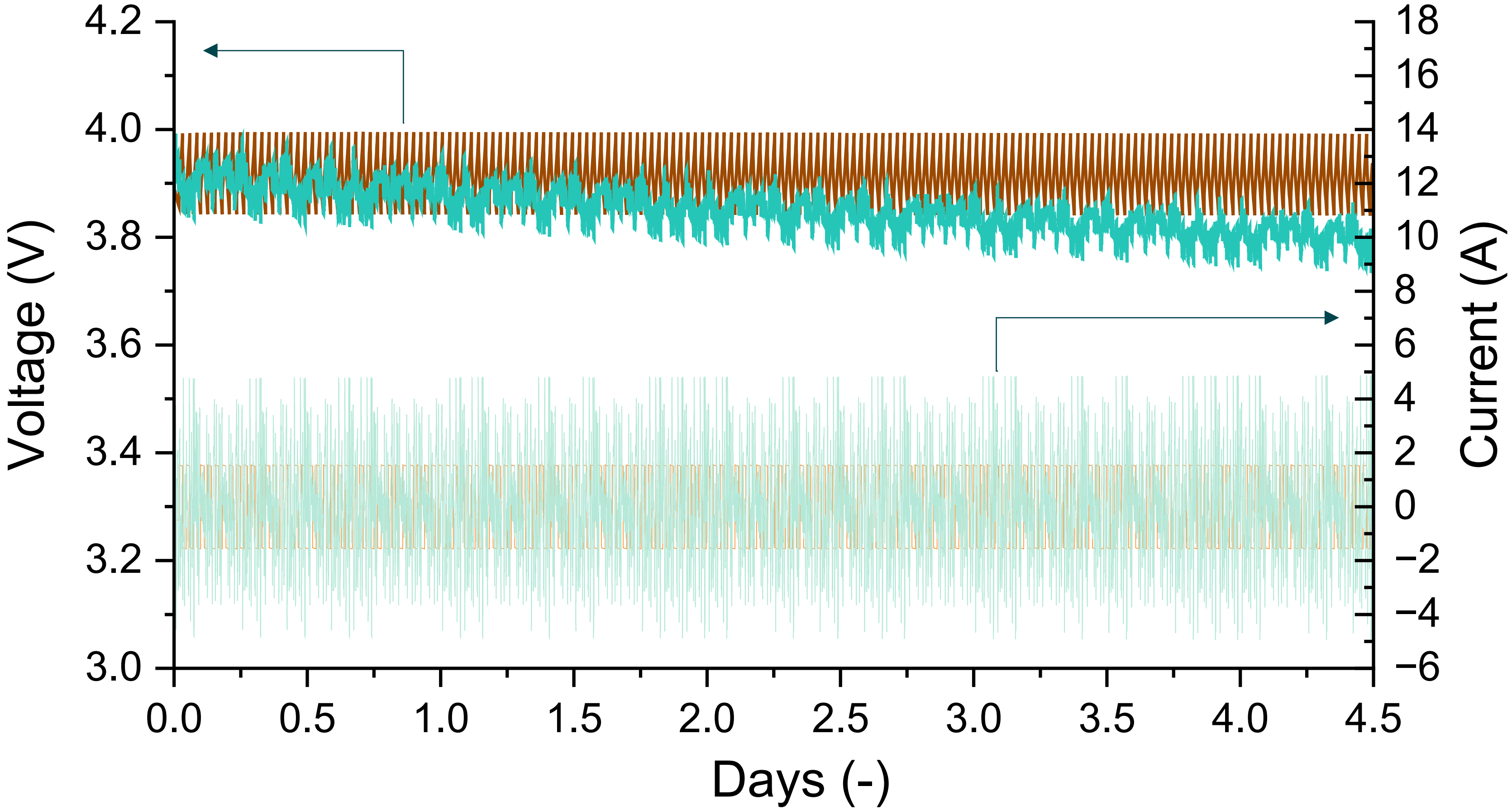
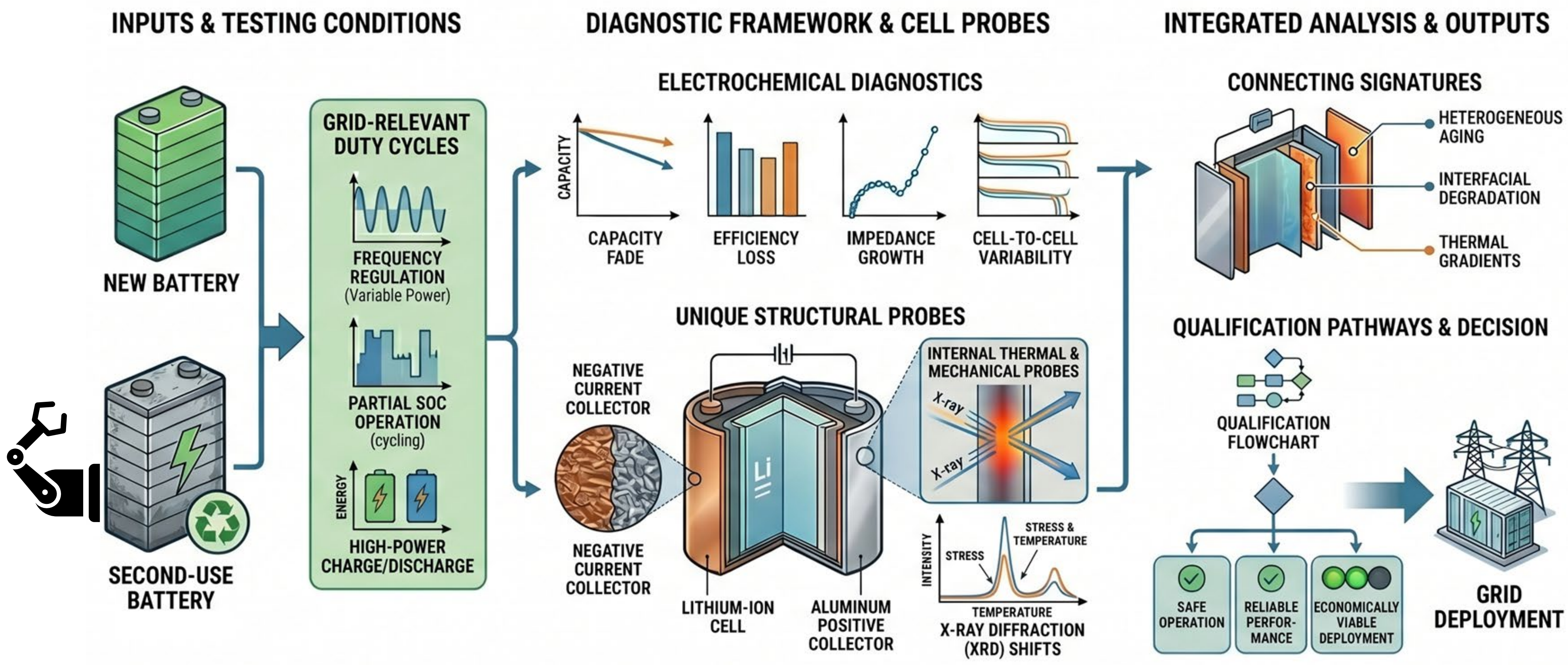
Project Overview



- Identical cells cycled at same "average" power between identical SOCs show distinct behavior
- These models fail to predict the behavior of mixed-history, unevenly degraded, or second-life battery packs under real grid duty cycles

Project Overview

Topic Area: Grid Resilience & Reliability | Vertical: Safety & Reliability
Strategic Pathway: Optimize
Project Duration: FY24-FY26



Develop science-based qualification methods for new, second-use, and end-of-life batteries in grid energy storage applications by linking electrochemical performance, degradation mechanisms, and safety-relevant behavior under realistic grid duty cycles.

Why ORNL: Integrated battery diagnostics, advanced characterization, robotic disassembly/qualification, and GRID-C validation capabilities enable ORNL to evaluate batteries from cell-level mechanisms to grid-relevant system behavior.

Impact: Establishes a data-driven qualification pathway for new and second-life batteries, reducing uncertainty in grid deployment by linking baseline tests, realistic duty-cycle response, and abnormal-cell detection within modules/strings.

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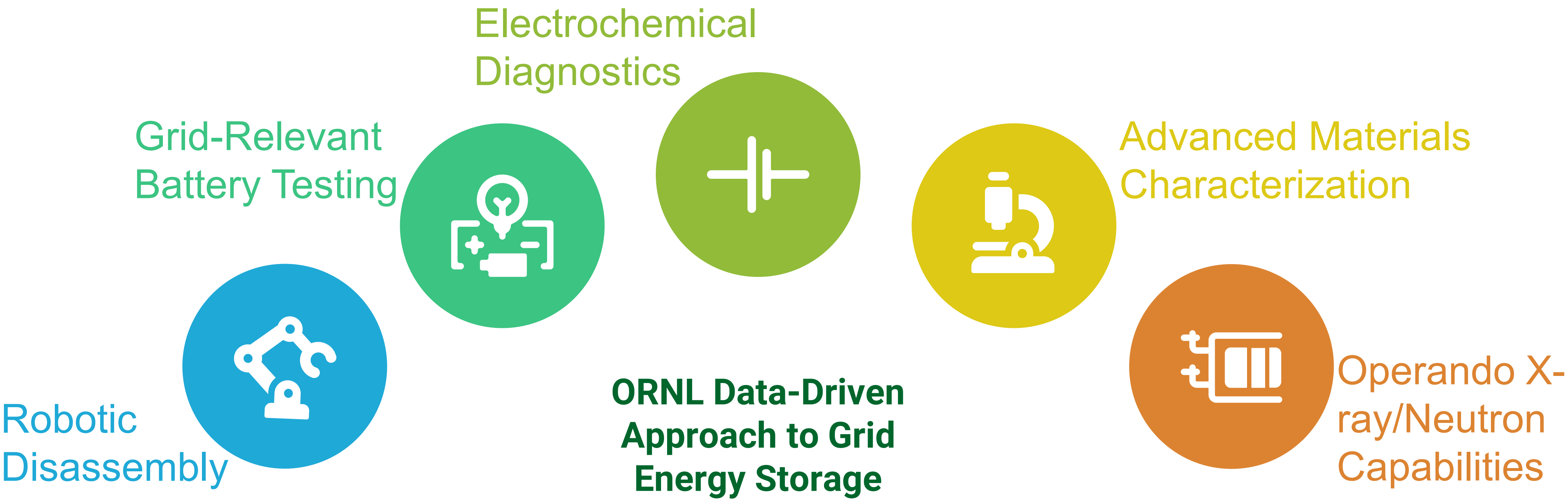
Project Overview



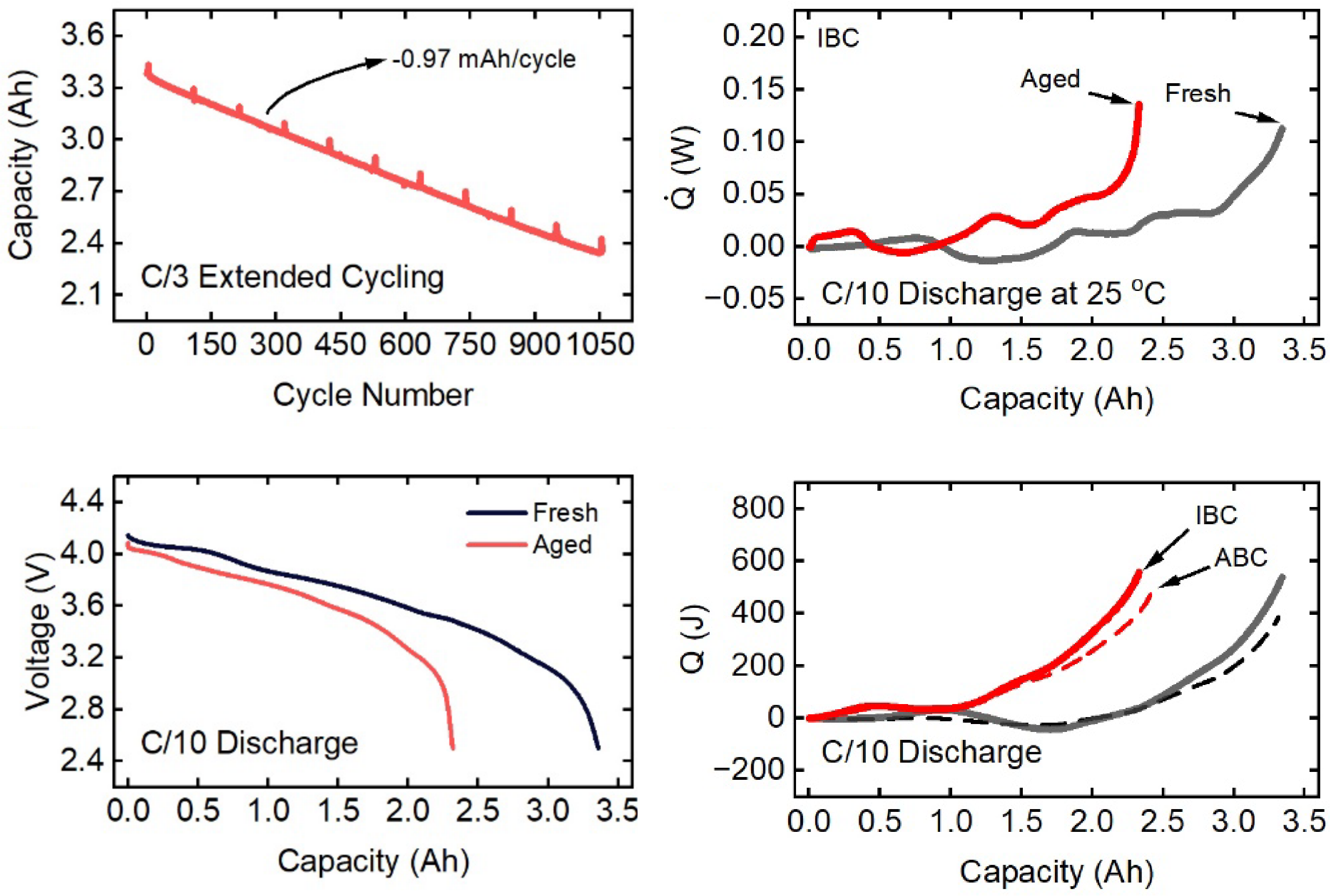
- This work supports reliable and more cost-effective deployment of second-use batteries for grid storage.
- Successful outcomes will benefit utilities, battery integrators, recyclers, regulators, and technology developers by enabling better screening, sorting, module design, and deployment decisions.

Understanding Degradation of Cylindrical Cells[1]

RQ. How do aged cells compare to fresh cells structurally?

A: Aged cells show trapped delithiated domains that may create thermally distinct regions within the electrode. As a result, aged cells may retain higher-risk thermal behavior even when the cell appears discharged by voltage measurements.

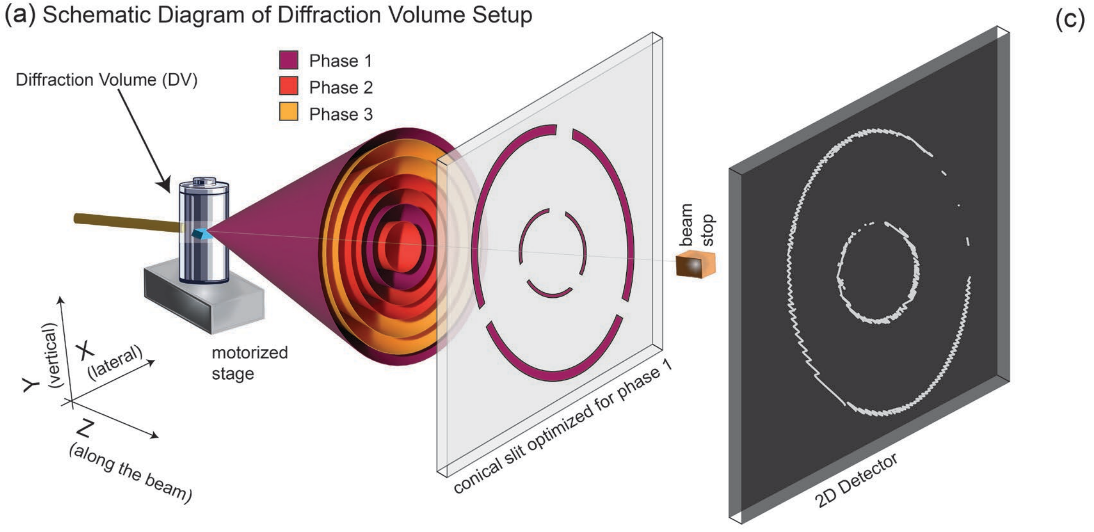
Aged cylindrical cells (~70% RUL) show a very distinct thermal behavior compared to fresh cells

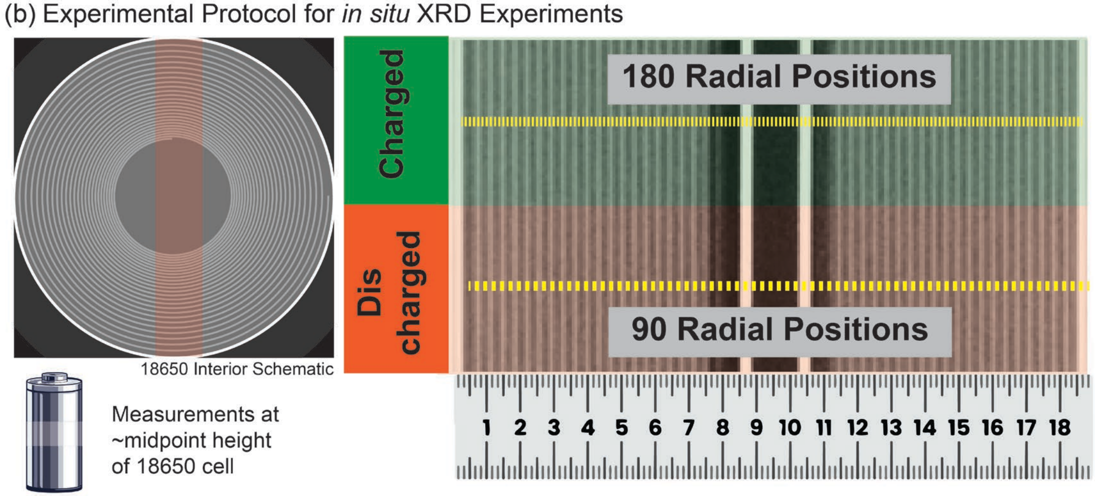


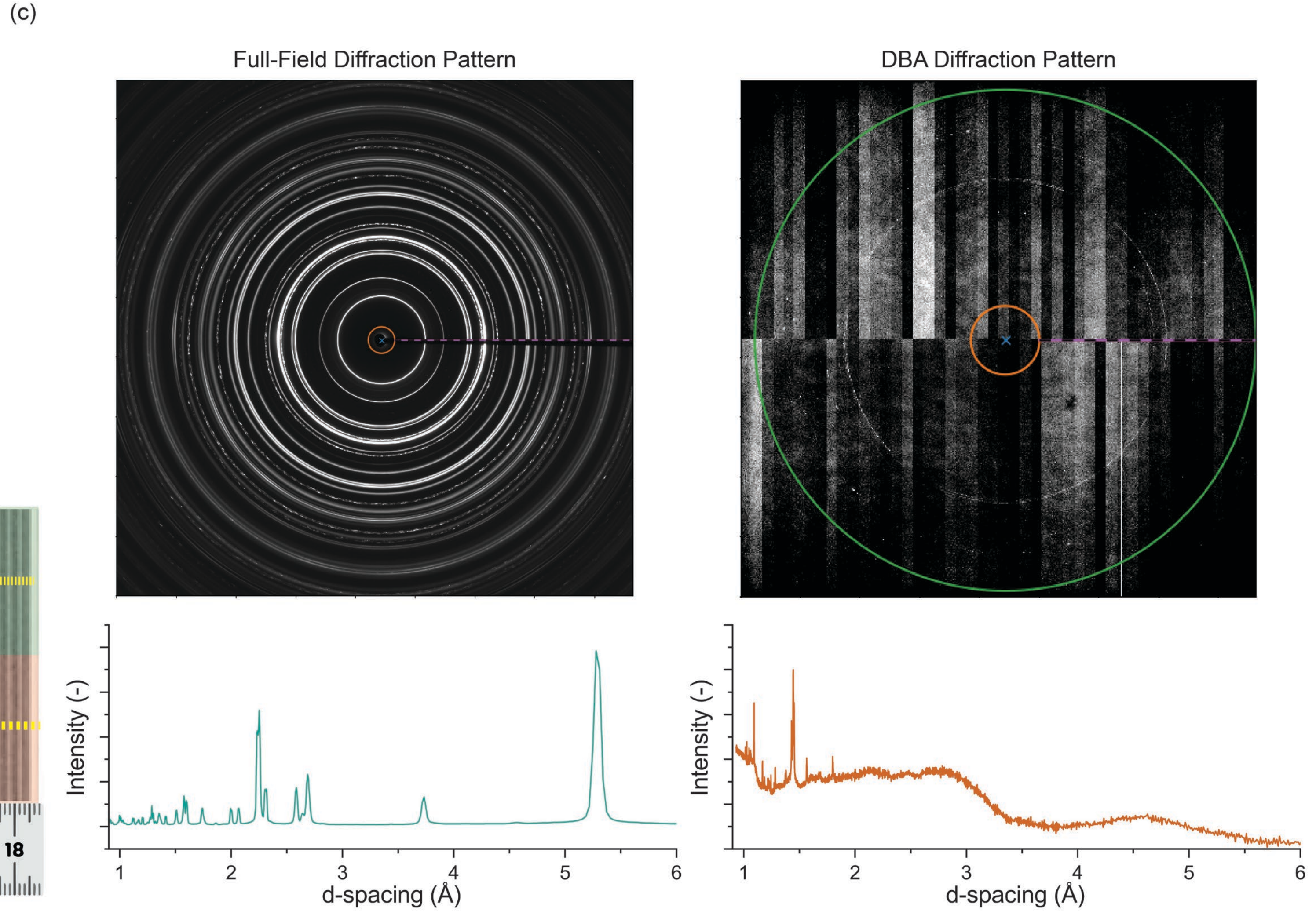
Chemistry: $LiNi_xCo_yAl_zO_2$/Graphite+Si
Configuration: Cylindrical Cell
Individual Cell Nominal Capacity: 3.4 Ah

- Long term cycling carried out at C/3
- Isothermal and adiabatic calorimetry measurements indicate lower accessible capacity as well as increased heat release from the aged cells

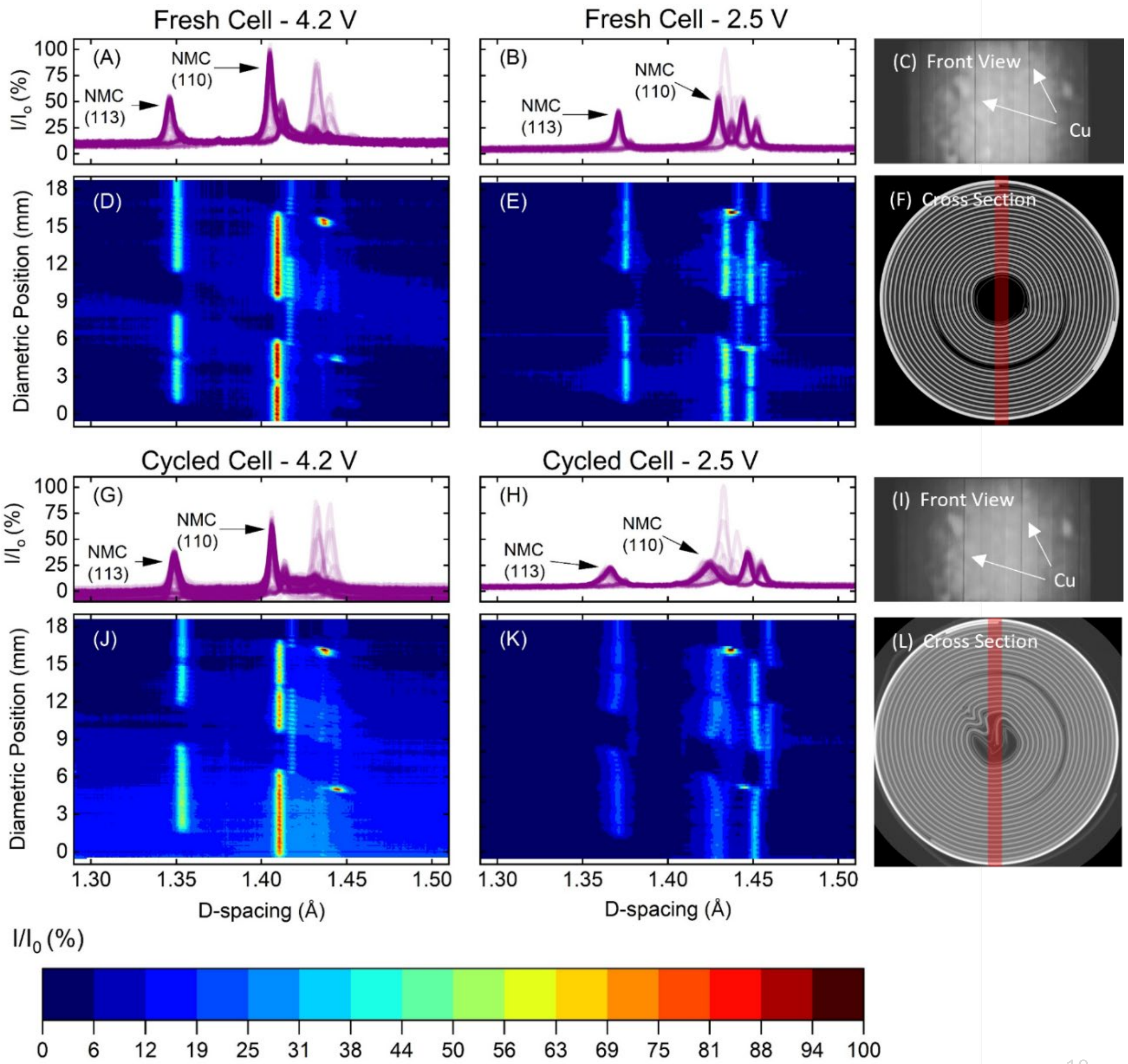
Synchrotron Diffraction with a Conical Slit Setup | Optical approach to isolating local regions (20 x 20 x 150 μm) within the cell





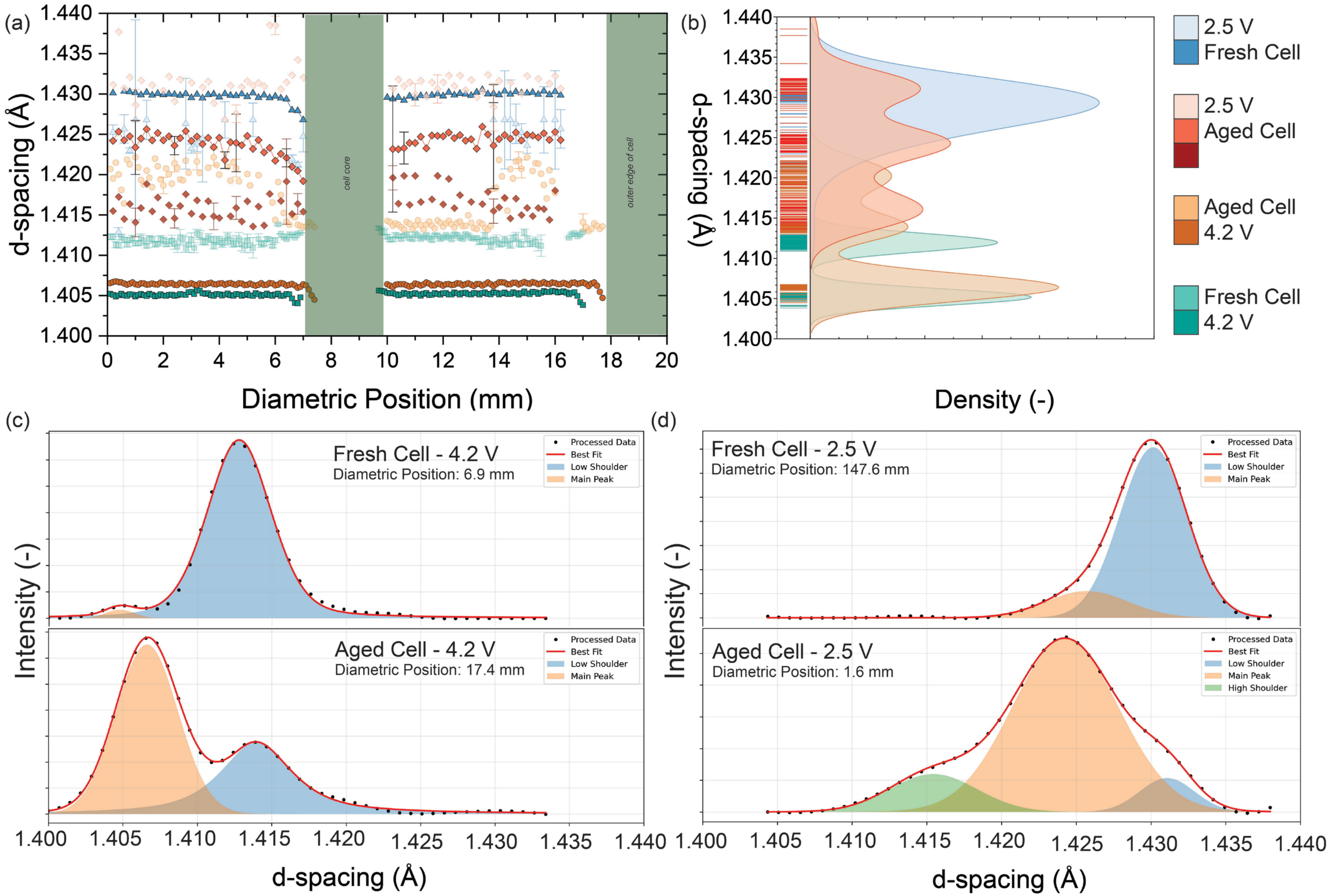


- This is what the data-set looks like
- We collected diffraction patterns across the diameter of the 18650 cell at the charged and discharged state
- Visual comparison of the data-sets show that the aged cell at discharge shows the most significant difference in the structural response



Analysis of the (113) Peak from the Diffraction Data-set Provides Key Insight

- In the aged cell (70% remaining capacity), some cathode regions show d-spacing values similar to the charged state, even after full discharge.
- This behavior is not observed in the fresh cell, suggesting it arises from aging-induced heterogeneity.
- Likely causes include electrically isolated regions or transport-limited domains.
- These trapped delithiated domains may create thermally distinct regions within the electrode.
- As a result, aged cells may retain higher-risk thermal behavior even when the cell appears discharge by voltage.



Qualifying "Second-Life" Battery Systems for Grid Storage[2-3]

RQ. How to design sub-system for integrating "end-of-life" cells?

A: Pack configuration plays a huge role in dictating the electrochemical performance of "second life" systems.

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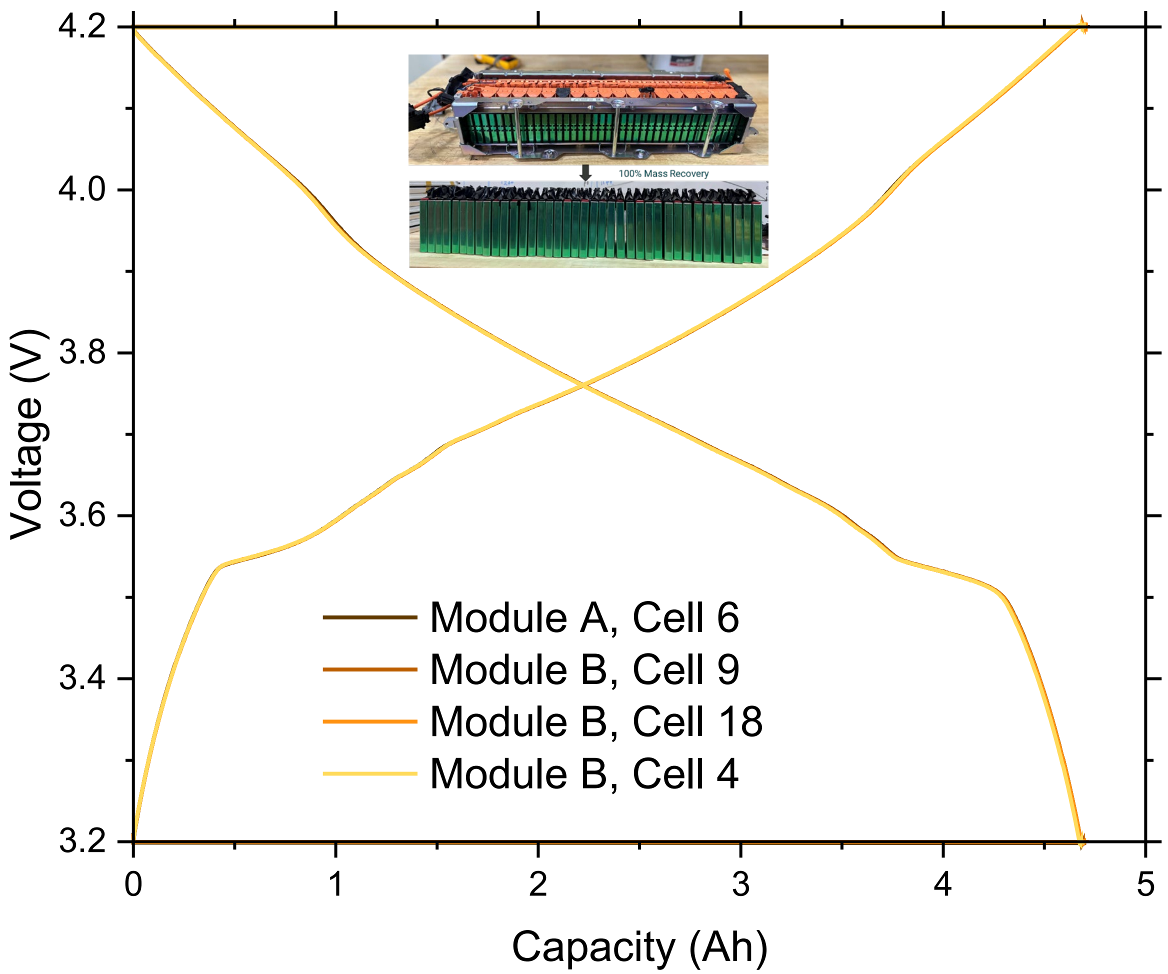
Subtleties of Grouping Cells : Discharge Capacity and Polarization are Independent from Naïve Measurements

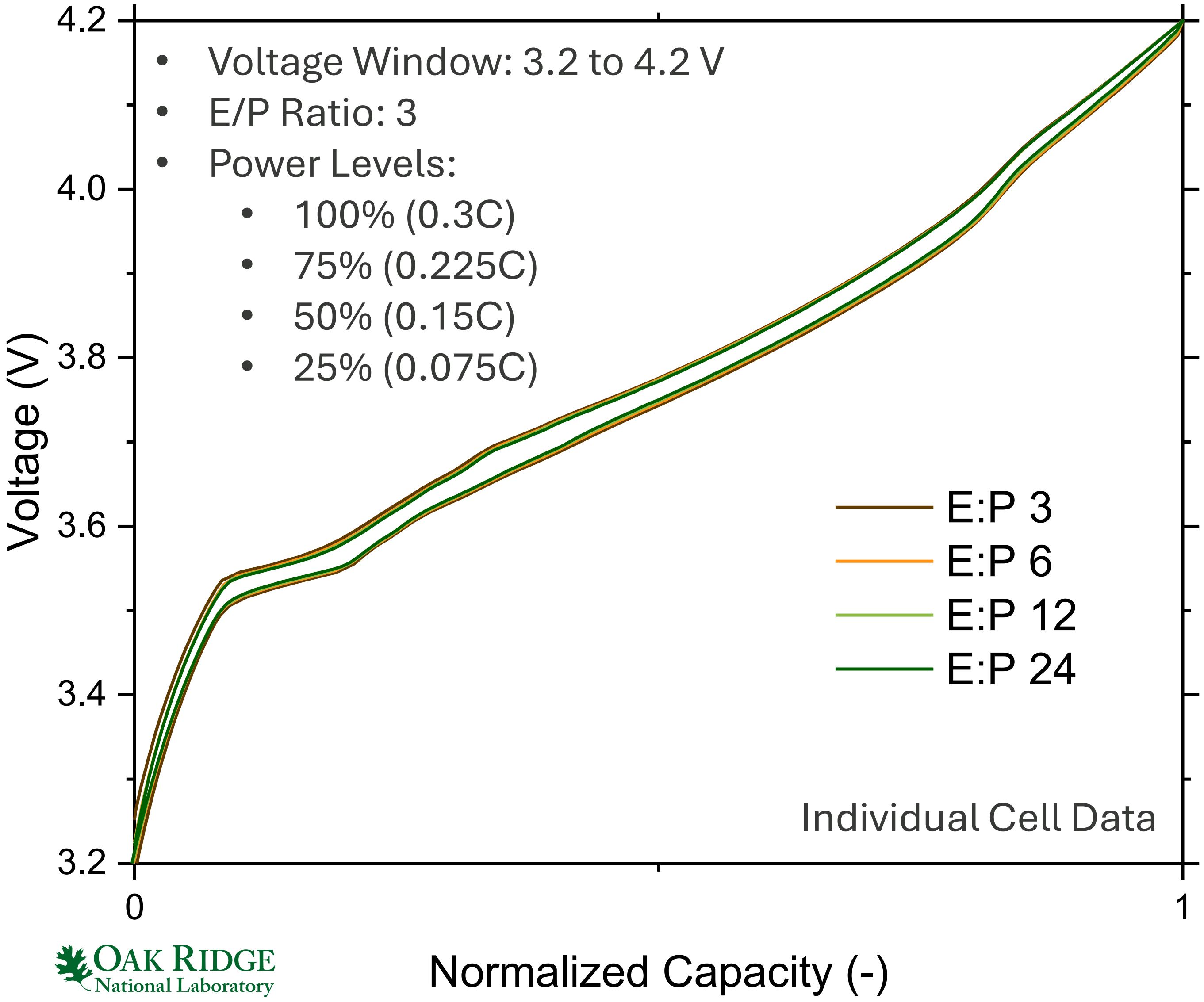


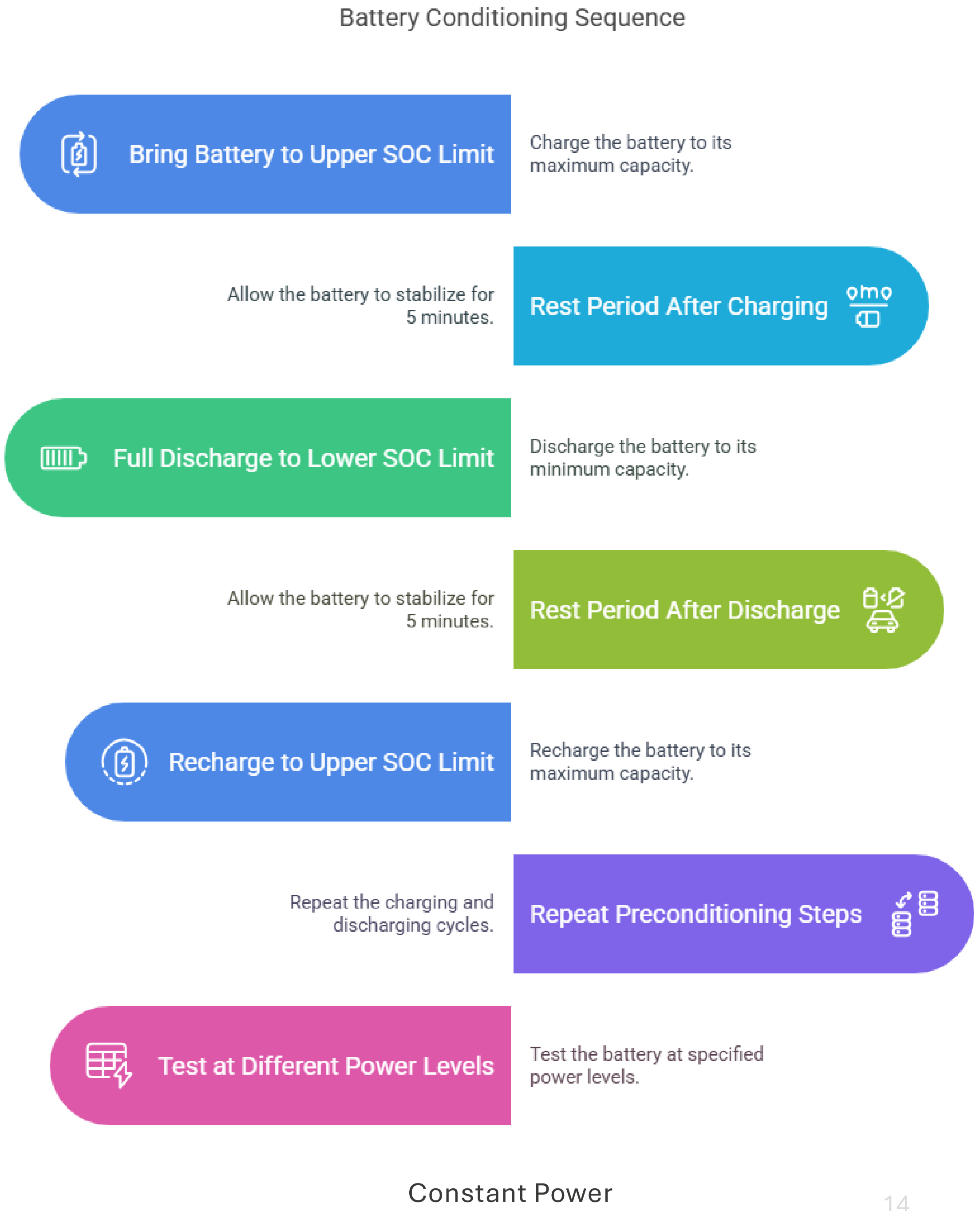
Chemistry: NMC-Graphite
Configuration: Prismatic Cell
Individual Cell Nominal Capacity: 5 Ah

- Identical Discharge Capacity and Discharge Polarization;
- However, their measured OCV and estimated charge transfer resistance (R1) from impedance fitting are not identical
- This is the best-case scenario - all other grouping scenarios add complexity

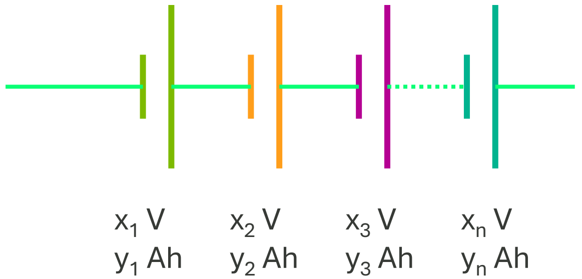
Cell Information	OCV (V)	R1 (mΩ)
Module A, Cell 6	$3.28\pm5\times10^{-5}$	7.34
Module B, Cell 9	$3.27\pm6\times10^{-4}$	7.12
Module B, Cell 18	$3.27\pm2\times10^{-3}$	7.94
Module B, Cell 4	$3.27\pm4\times10^{-3}$	6.87

PNNL-SANDIA Report Preconditioning Steps





Series and Parallel Configuration of Battery Architectures



System Voltage = $\sum_{i=1}^{n} x_i \;\; V$

System Capacity = $\min(y_i) \; Ah$

Voltage Amplification

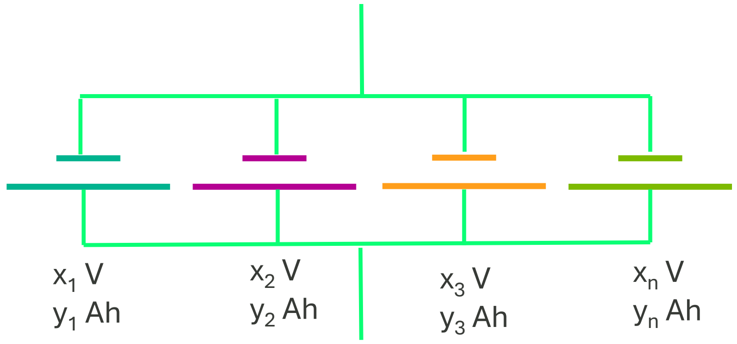
Enhanced Compatibility

Uniform Current Distribution

Limited Capacity Utilization

Dependency on All Batteries

Overcharging and Imbalanced Discharge



System Voltage = $\mathrm{mean}(x_i) \; V$

System Capacity = $\sum_{i=1}^{n} y_i \;\; Ah$

Increased Capacity

Uniform Voltage Output

Balanced Performance

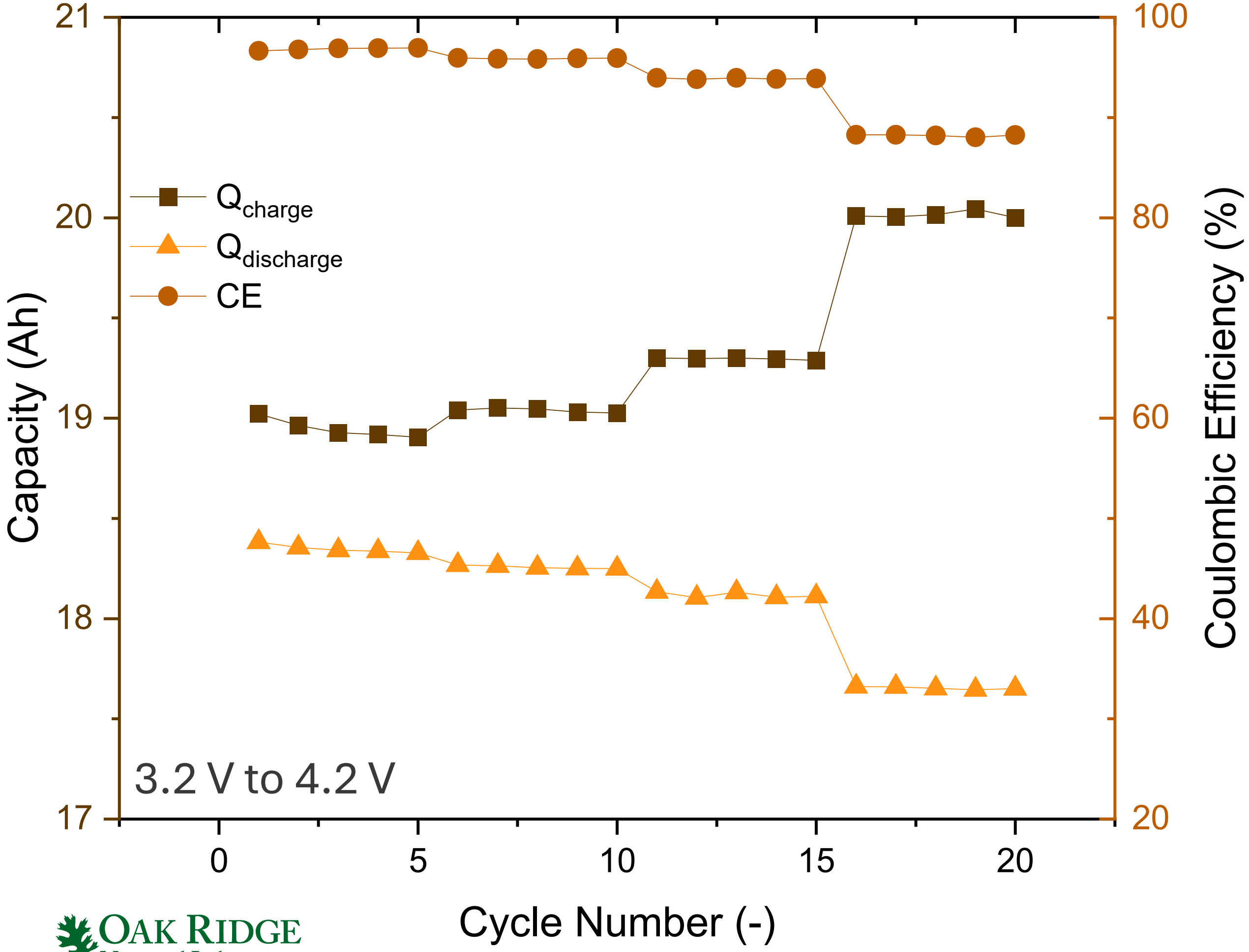
Complex Management

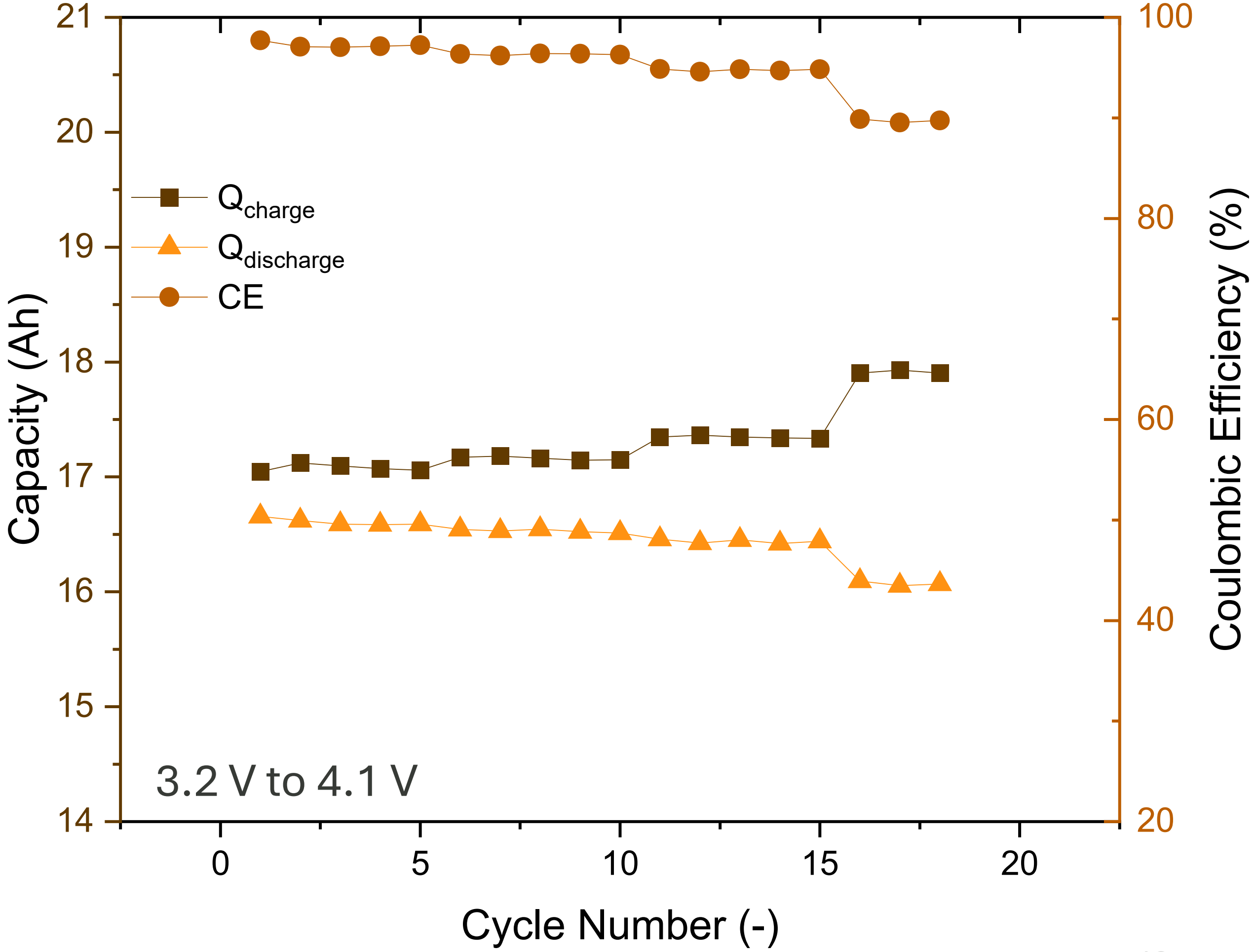
Enhanced Reliability

Compatibility Challenges

Parallel Configuration Dominates Module and Sections in current systems we observe

Power Conditioning Cycles (E:P-3, E:P-6, E:P-12, E:P-24; 5 cycles each) | Group in Parallel Behaves Weird and the Behavior is Cutoff Voltage Independent





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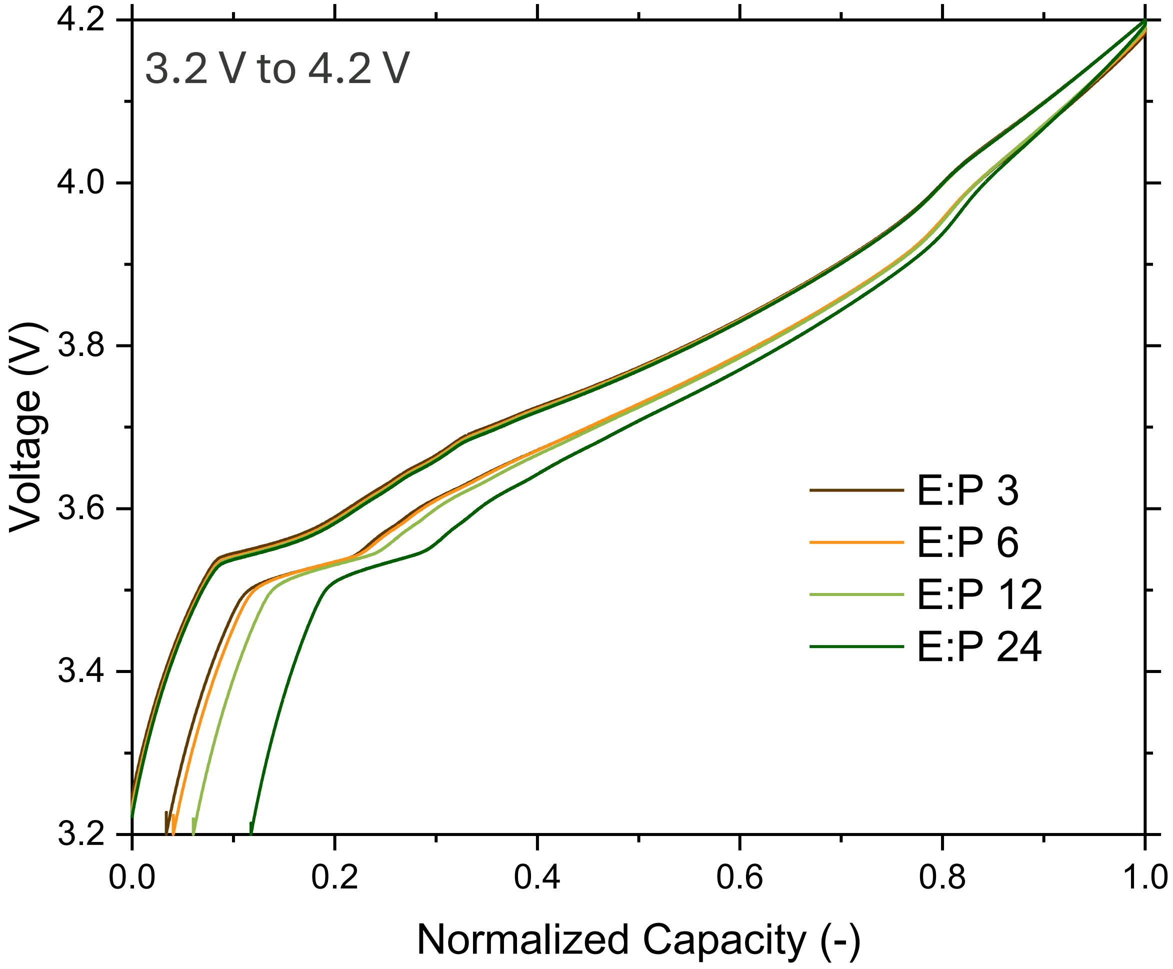
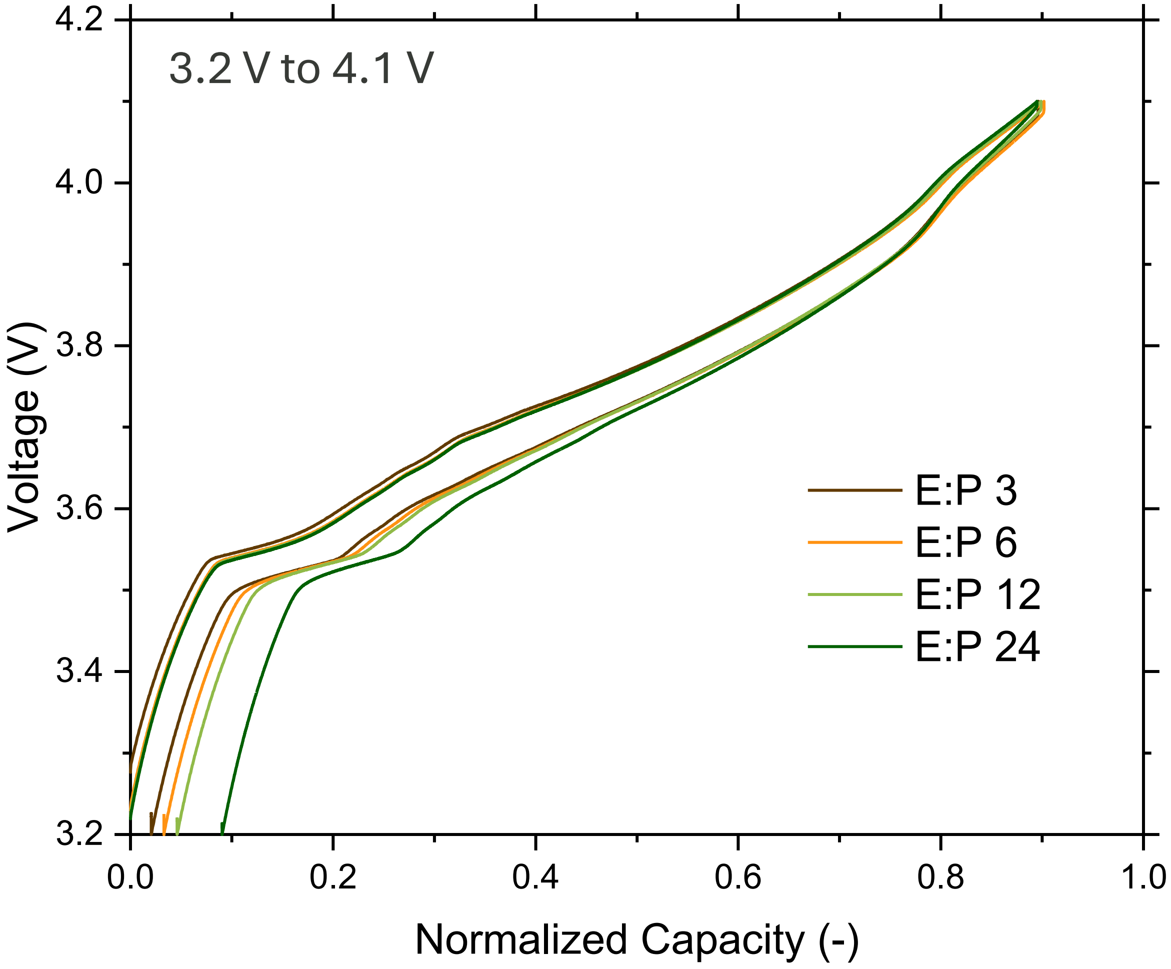
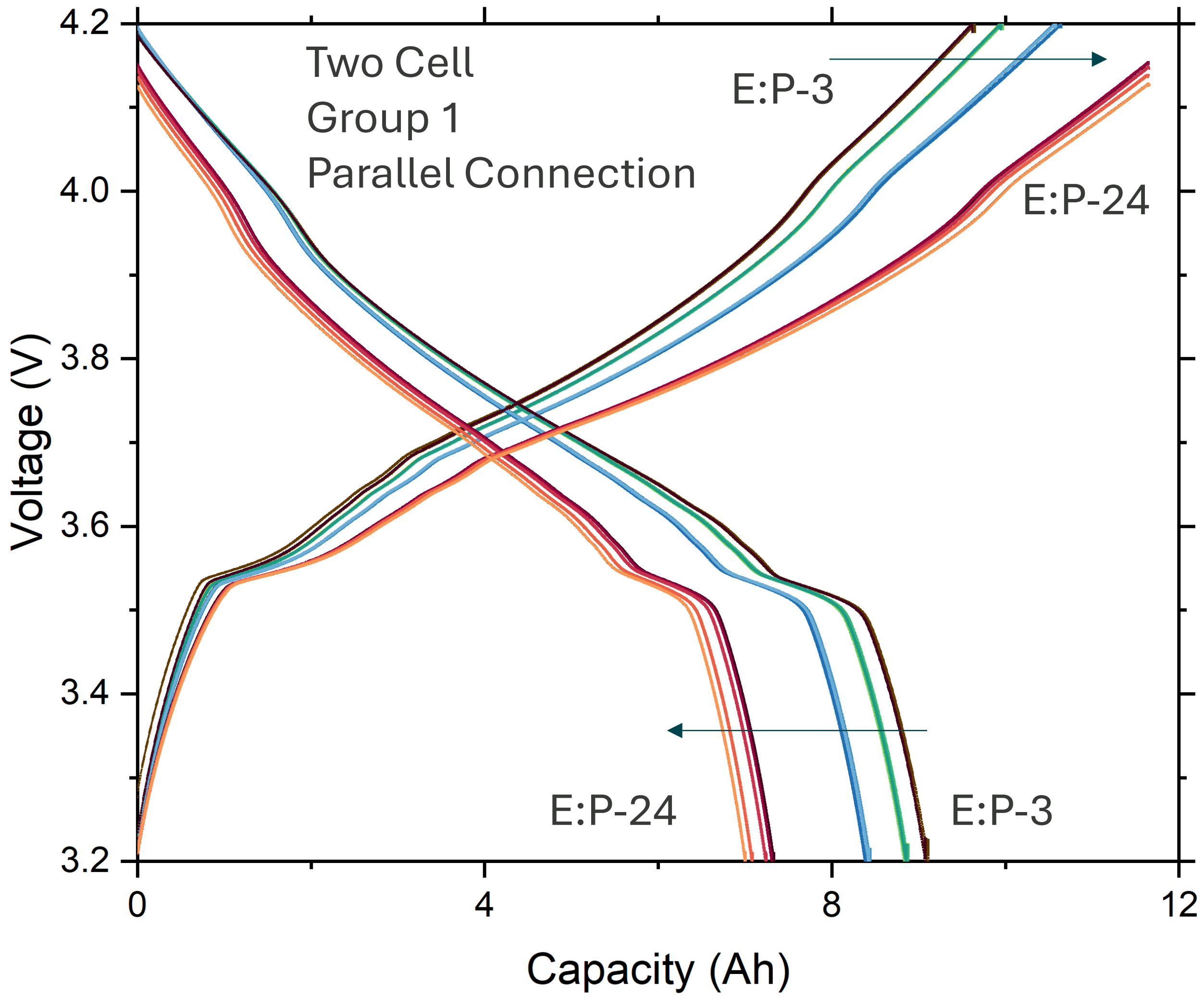
Normalized Capacity Analysis

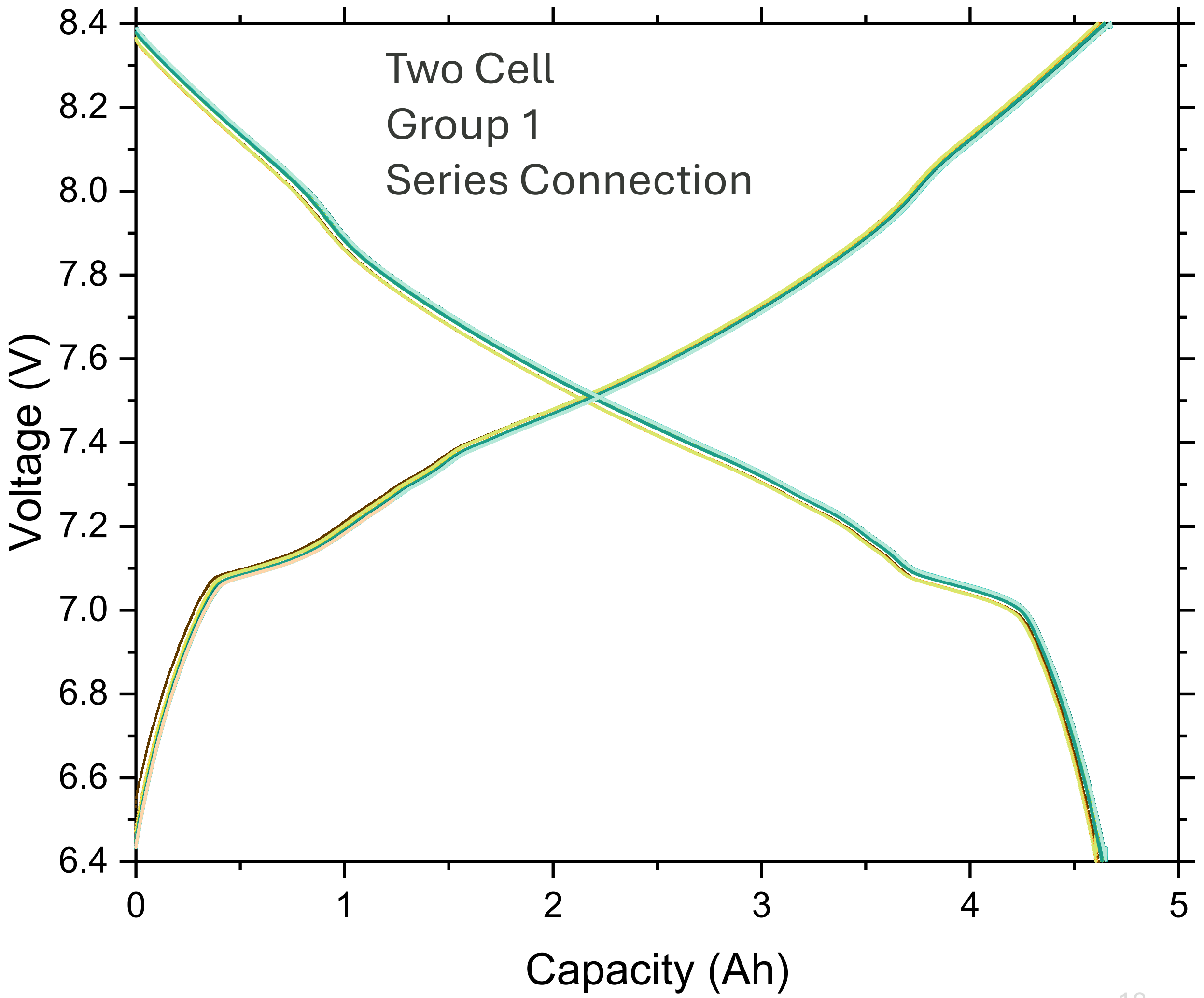




- We get better reversibility with lower value of upper cut-off voltage
- “Normal Polarization” behavior lasts longer during discharge

Two Cell Group Testing | Parallel Configuration Limits Performance | Series Configuration Eliminates the Challenge!



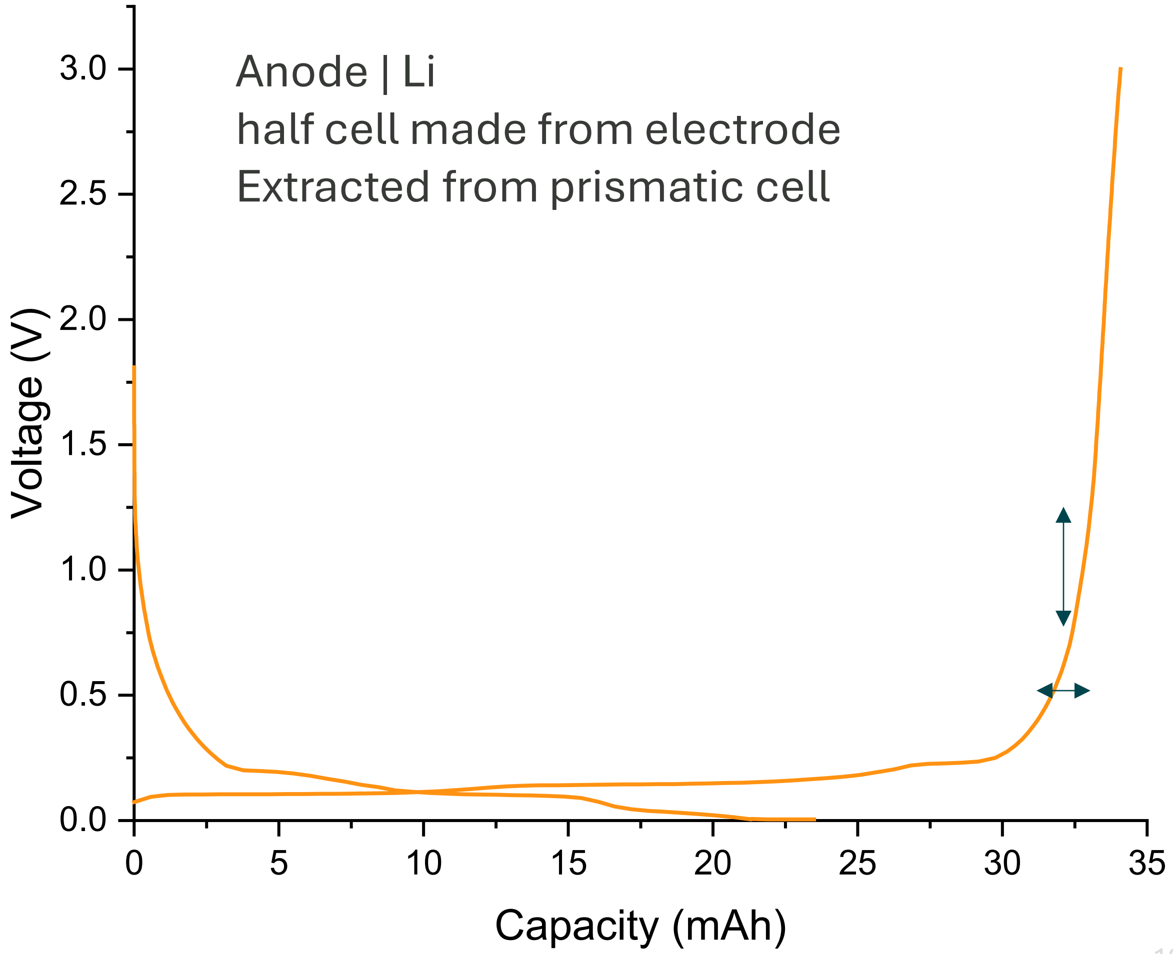


Working Hypothesis

$$i_i = \frac{V_{bus} - OCV_i(SOC_i)}{R_i}$$

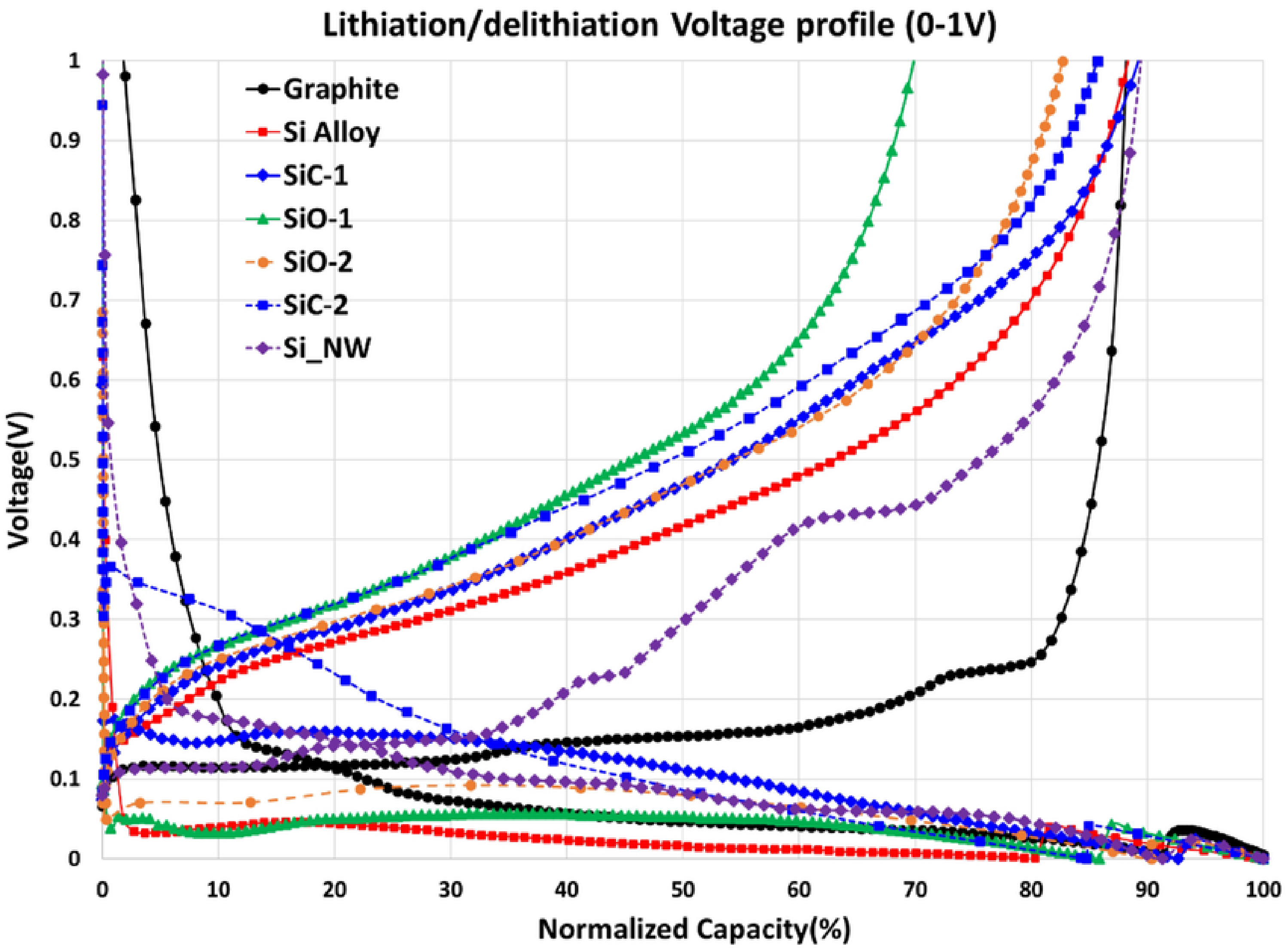
Current through i[th] cell depends on the difference between the bus bar voltage and the OCV of the cell at the local SOC

- Slippage of anode w.r.t cathode in aged cells likely causes the observed behavior
- At high SOC of anode, a small change in lithiation content leads to a LARGE change in voltage
- As you discharge the parallel group, the lower-OCV / higher-IR cell(s) can begin drawing current from the healthier ones, dragging pack voltage down and burning energy internally. The load "sees" less net current → capacity appears severely limited
- Series connection forces the same current through all cells eliminating this behavior



Zoom-Out | Why We Think This Is Important

- Si containing anode blends are highly common in current commercial Li-ion batteries
- All these blend anodes – show polarization curves and end of lithiation that are very distinct from graphite
- Thus, our observations on integration of these “end-of-life” cells have potentially far-reaching implications once all these cells containing Si/SiOx blends come into the grid market as end-of-life cells



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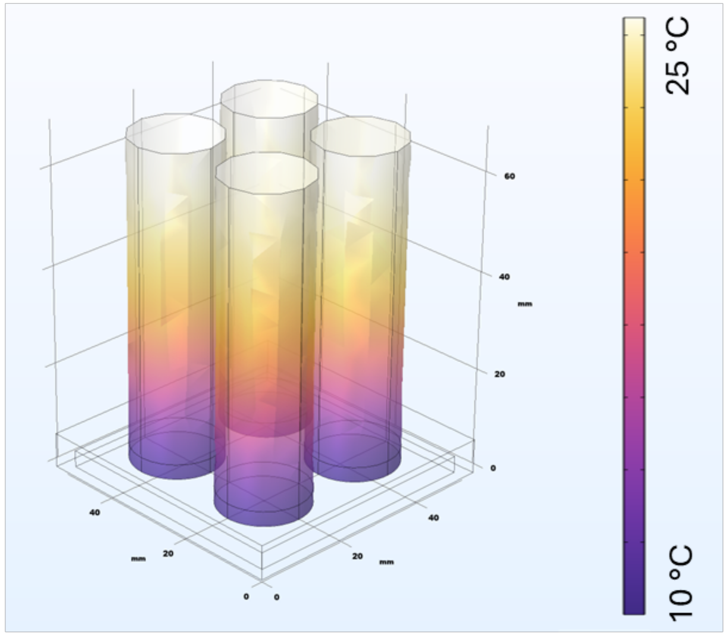
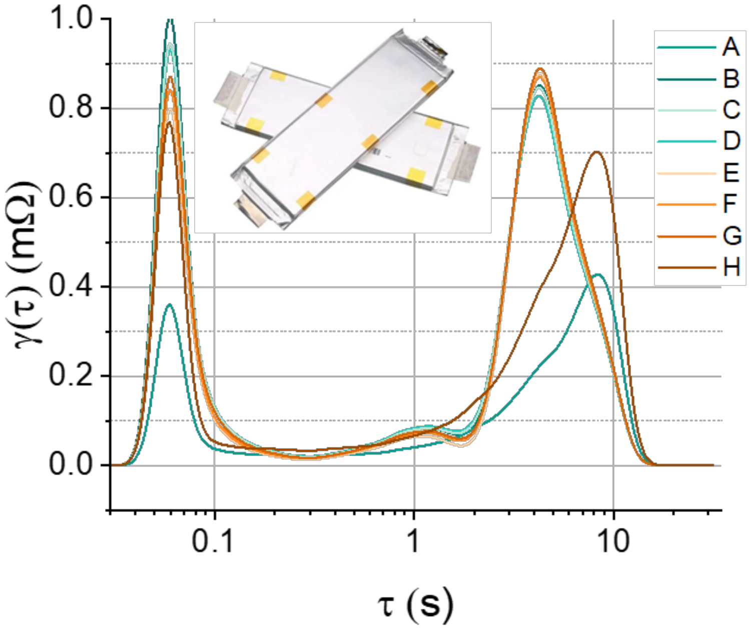
Ongoing Work Teaser

- Evaluating cells with an axial thermal gradient to simulate bottom cooling found in grid storage systems
- Continuing work on developing strategies to qualify batteries and modules for grid systems by incorporating different chemistries and form factors
- Evaluating thermal behavior of cells under different ambient conditions using Al, and Cu diffraction signals as a temperature probe from within the cell

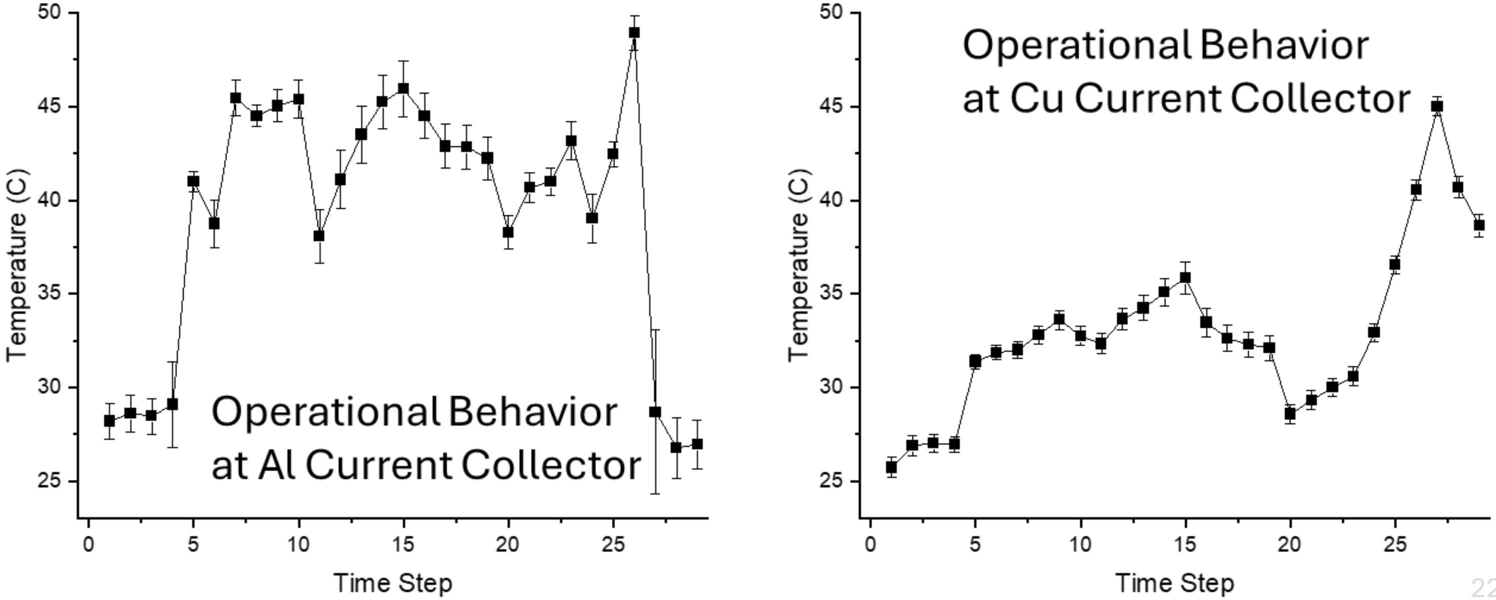
(a) Thermal Gradient Experiments



(b) Diagnostics for Large Cells and Modules[4]



(c) Operando Assessment of Thermal Behavior using Synchrotron Diffraction



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3. Considerations for Qualification of Second-Life Batteries for Grid Applications. Part II: Sub-system Assembly and Repurposed Module Diagnostics. In Preparation.
4. Lessons from Impedance Spectroscopy Studies of Battery Sub-systems for Fault Identification. In Preparation