Characterization of Li-ion batteries Using Neutron Diffraction and Infrared Imaging Techniques: Success Stories from the High Temperature Materials Laboratory (HTML) User Program

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DOE 2011 Vehicle Technologies Annual Merit Review and Peer Evaluation Meeting

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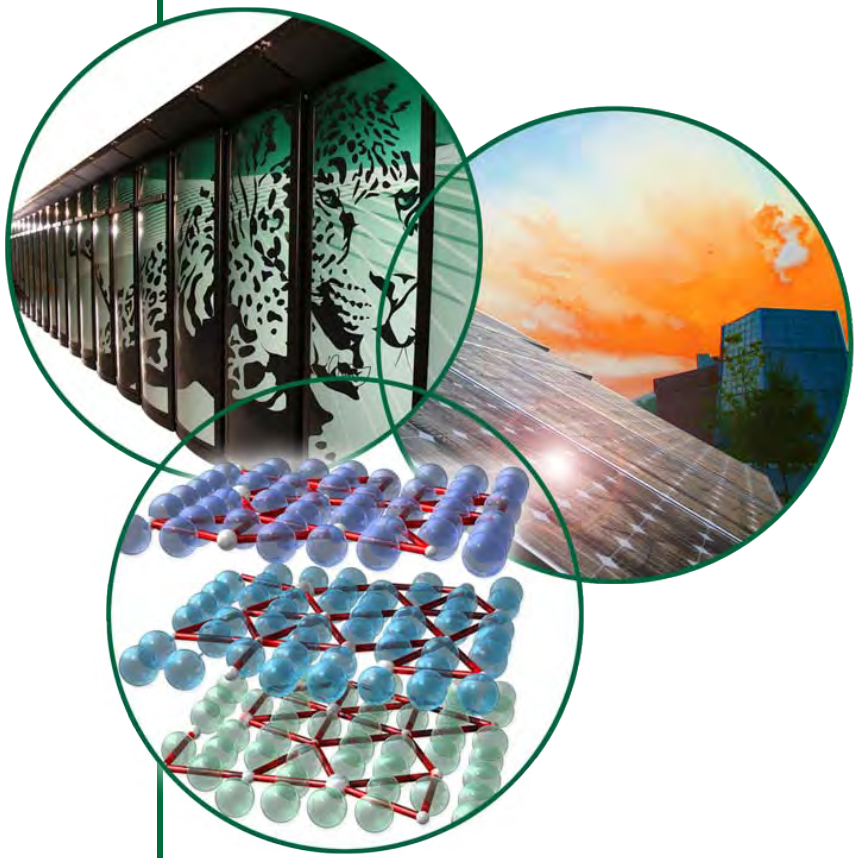

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The HTML User Program: Background

- The HTML is a National User Facility that supports the missions of DOE, EERE and the Vehicle Technologies Program in particular, by working with industry, universities, and other national laboratories to develop energy-efficient technologies that will enable the U.S. to use less petroleum. The HTML is organized into six user centers, which are clusters of highly skilled staff and sophisticated, often one-of-a-kind instruments for materials characterization.
- Access to the HTML User Program is provided through the HTML User Program proposal process. Research proposals are reviewed by a committee and approved based on scientific merit, relevance of the proposed research to the mission of DOE's Vehicle Technologies Program, feasibility, and non-competition with the private sector. Projects have a well-defined scope, and research is completed within 24 months and often involves multiple user visits to the HTML.
- Both nonproprietary and proprietary research is conducted within the HTML User Program. There are generally no charges for nonproprietary research projects, and users conducting nonproprietary research must agree to submit research results for publication in the open, refereed literature. A nonproprietary project is complete when research ends, accompanied by the required publication in the open literature and/or presentation at a professional conference. For proprietary research, the user owns the research data, and all costs at the HTML are paid by the user based on DOE guidelines for ORNL cost recovery.

The HTML User Program – FY2010 Activity

During FY2010, the HTML User Program collaborated with 18 companies, 25 universities, and 6 national laboratories on 68 user projects addressing critical technical barriers to achieving the goals of DOE's Vehicle Technologies Program. There were 96 researchers, 63% of them first-time users, who visited the HTML for a total of 716 research days.

The HTML User Program FY2010 budget was $5,312,400 and allocated as follows:

- Capital equipment: $881,959
- Operations: $4,430,441

Users cost-share their HTML user projects through:

1) direct involvement with HTML staff members during the development of the user project;
2) funding their travel to the HTML to perform research;
3) cost of materials provided by the user or the research performed prior to the user project;
4) collaboration with HTML staff members to analyze the data and publish the results.

The HTML also supports the education and preparation of the next generation of scientists and engineers. During FY2010, students and professors from 25 universities participated in the HTML User Program. Five of those students earned their Ph.D. degree and one earned her M.S. degree based in part on research they conducted through the HTML User Program.

Relevance to the VT Program

- The Vehicle Technologies Program funds the operation of the HTML User Program to maintain world-class expertise and instrumentation capabilities for materials characterization to work with industry, universities and national laboratories toward the goals of the Vehicle Technologies Program. The HTML User Program capabilities at the Oak Ridge National Laboratory support the activities of the Vehicle Technologies Program's subprograms in Lightweight Materials, Propulsion Materials, Energy Storage, Solid State Energy Conversion, Combustion & Emissions Controls, Power Electronics & Electric Motors, and Non-Petroleum Fuels.

- During FY2010, the HTML User Program managed **10** characterization projects in **Energy Storage**, and this poster presentation highlights ***one*** of them. The user project with **General Motors** in this poster presentation addresses the need for developing techniques for non-destructive methods to monitor cell temperature and phase changes during operation. Neutron diffraction and infrared imaging are used to detect early signs of degradation and predict cycle life. Long-term stability and understanding of degradation are very important to General Motors and crucial to large-scale deployment of electric vehicles.

General Motors R&D Center User Project: "Characterization of Li-ion Batteries Using Neutron Diffraction and Infrared Imaging Techniques"

Timeline

- Start date: 10/1/09
- End date: 6/30/11
- % complete: 90%

Barriers

- Performance
- Life

Budget

- Included in the user center allocations from the annual budget of the HTML User Program; users cost-share as noted on slide #3.

Collaborators

- **Users:** General Motors R&D Center Jihui Yang, Robert S. Conell
- **HTML Staff:** Hsin Wang, Camden R. Hubbard and Wei Cai

Collaborations

As a DOE User Facility, the HTML User Program is collaborative in nature. Potential users are assisted with the proposal submission process as necessary, and all research is hands-on with direct involvement from both user and HTML User Program staff researchers. The DOE-required publication of results for non-proprietary projects is also a collaborative effort.

Collaborators on the user project reported in this presentation:

General Motors R&D Center

Dr. Jihui Yang, Laboratory Group Manager & Senior Research Scientist

Robert S. Conell, Staff Researcher

HTML User Center

Hsin Wang, Camden Hubbard, and Wei Cai

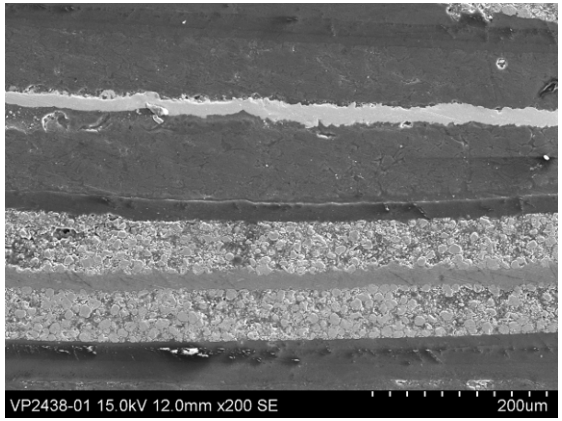
Residual Stress and Thermography & Thermophysical Properties User Centers

General Motors R&D Center User Project

Research Problem: To determine the local temperature variation during charging and discharging and to explore phase changes in electrodes using *in situ* neutron mapping; information needed for GM to estimate life and reliability of full-scale battery system.

Technical Approach: Utilize the unique capabilities of the HTML User Program for *in situ* studies. A high-speed infrared camera was used for temperature mapping and the neutron strain mapping facility was used for *in situ* phase monitoring during charging and discharging.

Materials



Devices

Vehicle Application

Photo (center) and schematic courtesy of General Motors R&D Center.

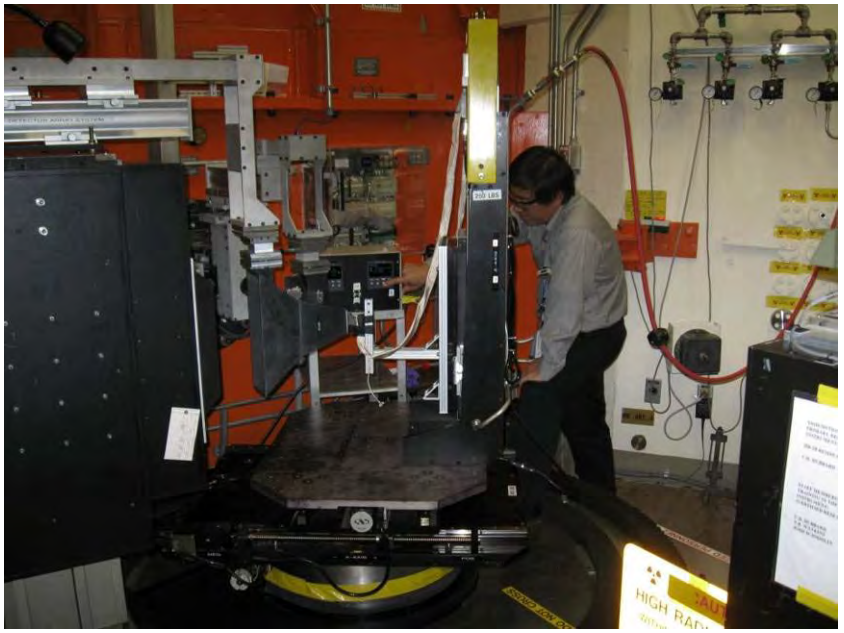
General Motors R&D Center User Project: Milestones

- Obtain temperature distribution of Li-ion cells during normal charging and discharging operation (completed) ✓
- Conduct *in situ* monitoring and mapping of internal phase changes on production cells during charging and discharging (completed) ✓
- Report the results of the user project via joint papers and presentations (in progress)

15 A-hr cell enclosed in pouch used for this study

General Motors R&D Center User Project Approach - Utilize Unique HTML Capabilities and Expertise

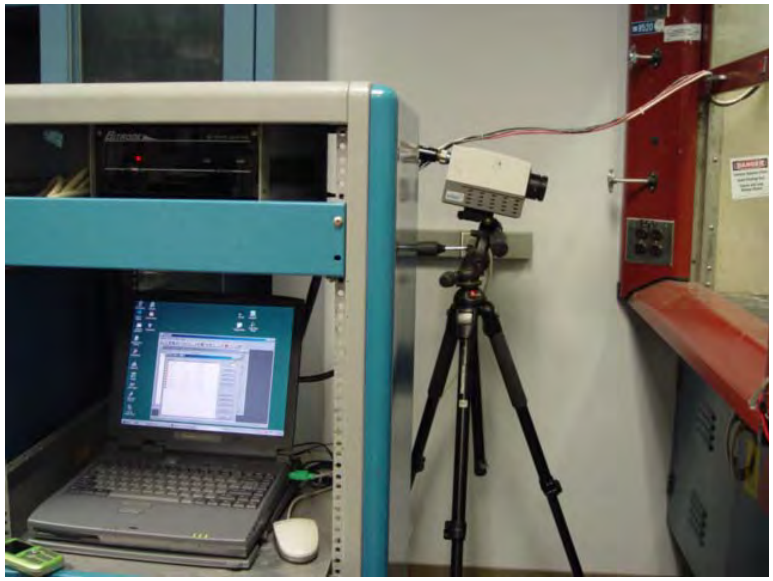
FLIR High-speed high resolution infrared Camera: 256 x 320 pixels, 0.02K resolution



High Flux Isotope Reactor (HFIR) beam line: neutron strain mapping facility

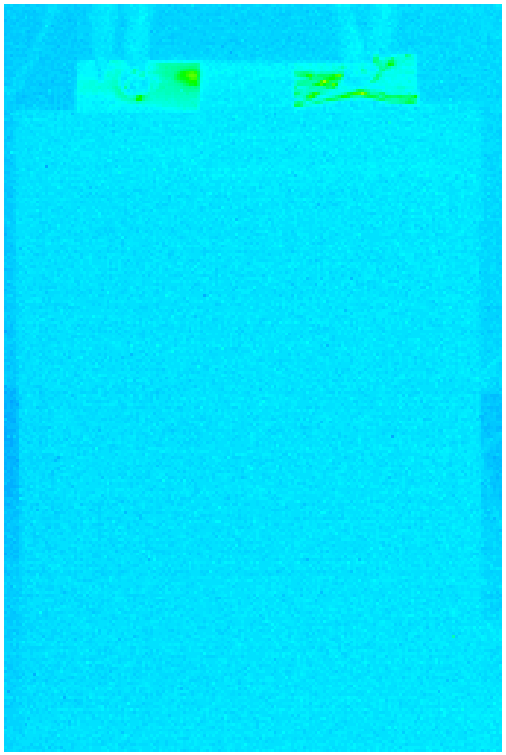
GM utilized the world-class capabilities at the HTML User Program to determine temperature variation and phase changes during normal cell operation.

General Motors R&D Center User Project: Accomplishments – Infrared Imaging

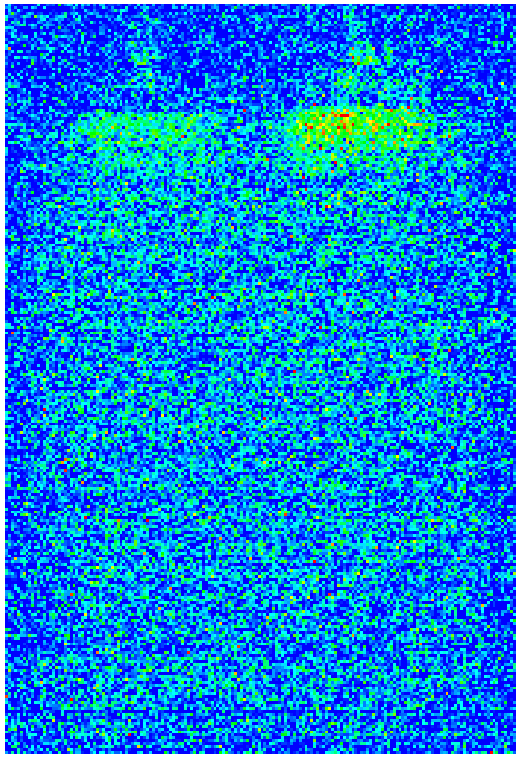


Infrared Imaging during 25A discharging/charging

- Maximum temperature change was below 8°C.
- A slight temperature change (<1°C) at the tabs was observed.



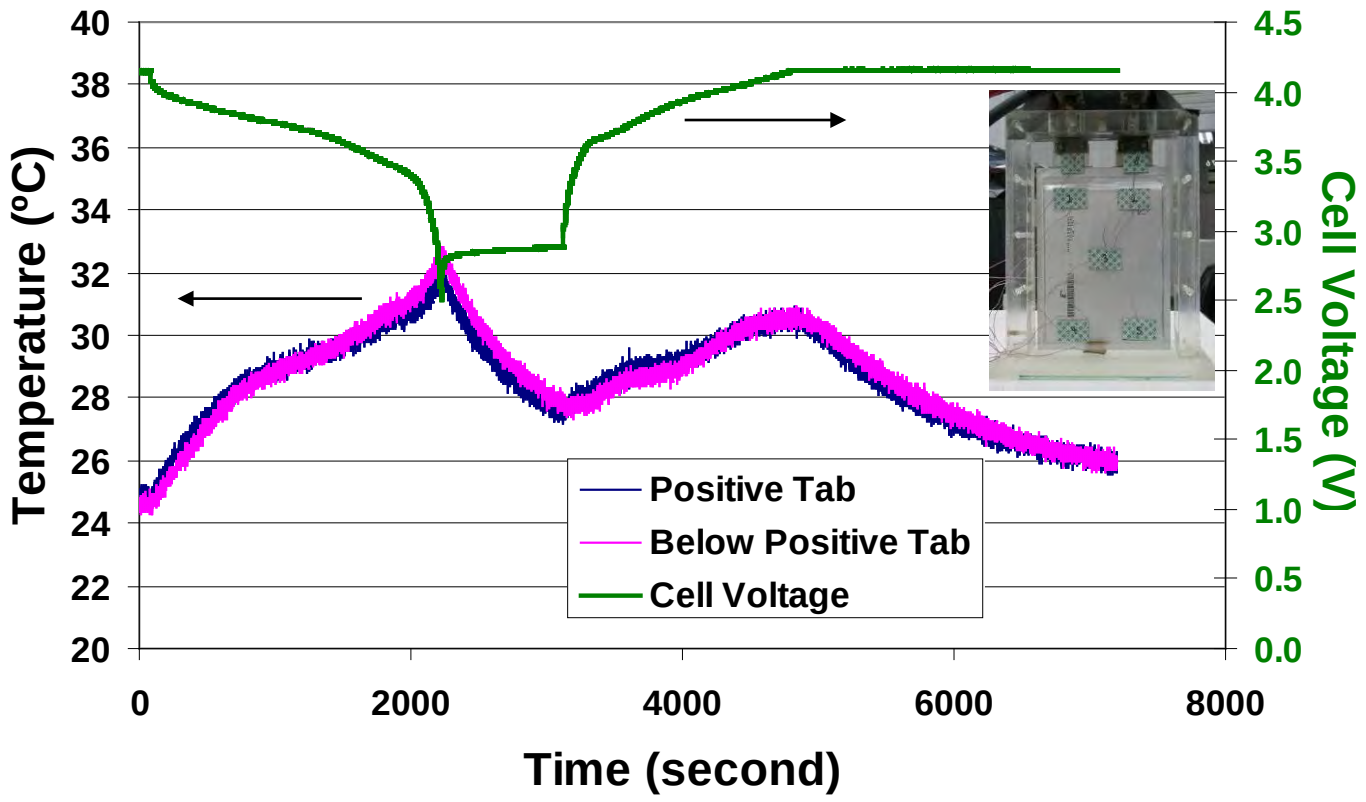
Cell temperatures were very uniform.



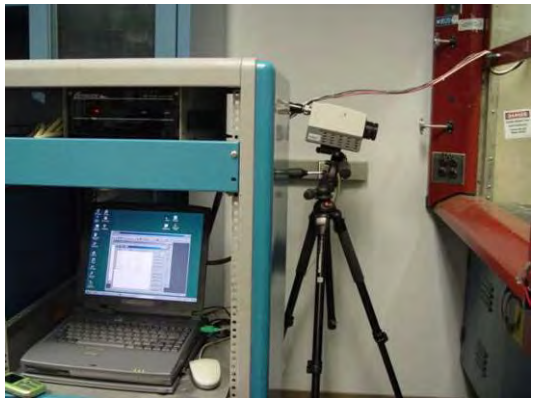
Maps of cell temperature changes taken every 5 sec were very uniform.

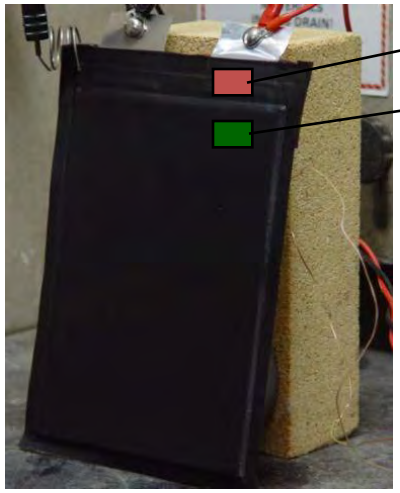
General Motors R&D Center User Project: Accomplishments – IR Imaging Results Matched with 7 Thermocouple Values

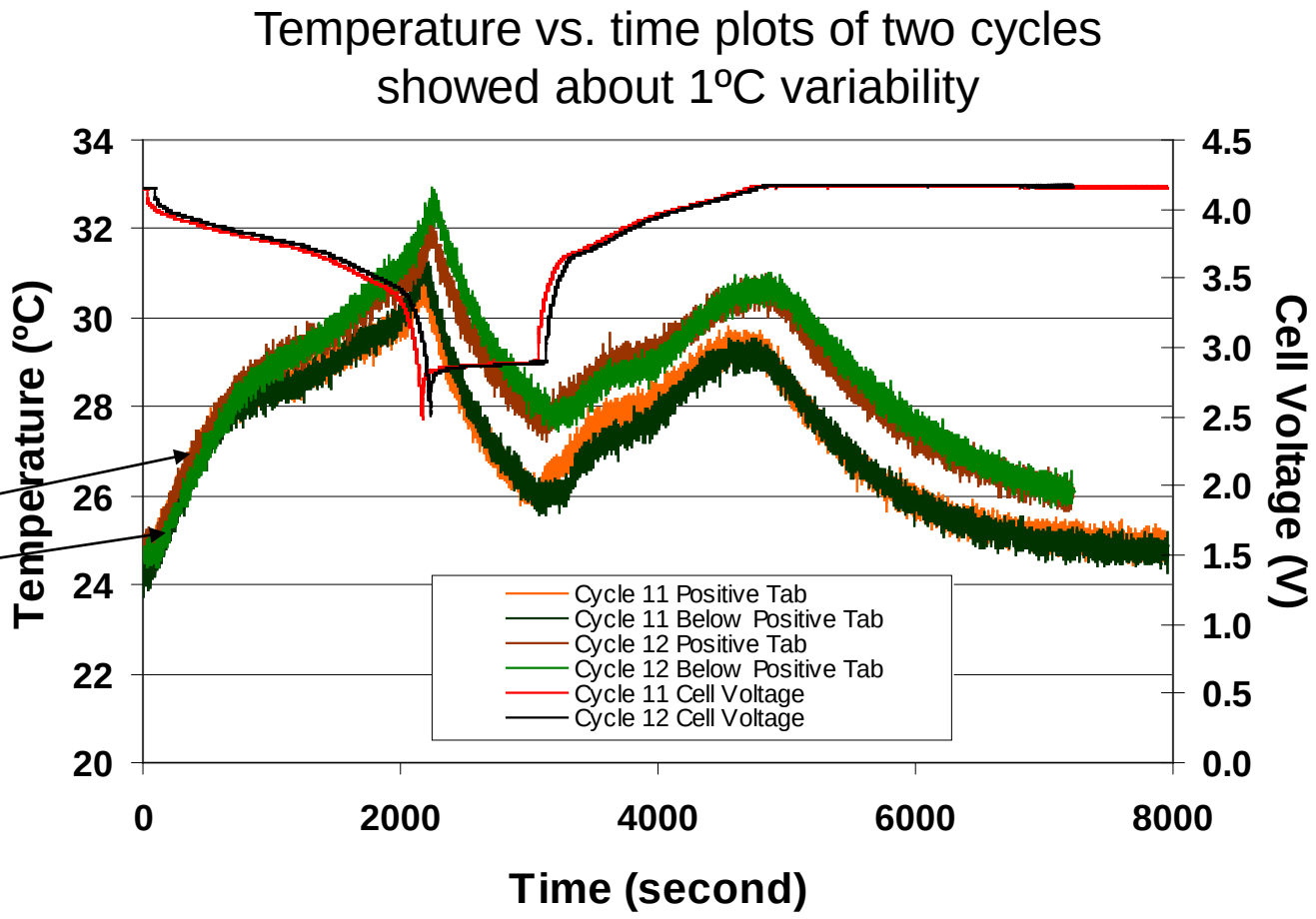
Maximum temperature rise was below 8ºC at the end of discharge.



General Motors R&D Center User Project: Accomplishments – Infrared Imaging Results of Two Consecutive Cycles







General Motors R&D Center User Project: Accomplishments – *In situ* Neutron Diffraction

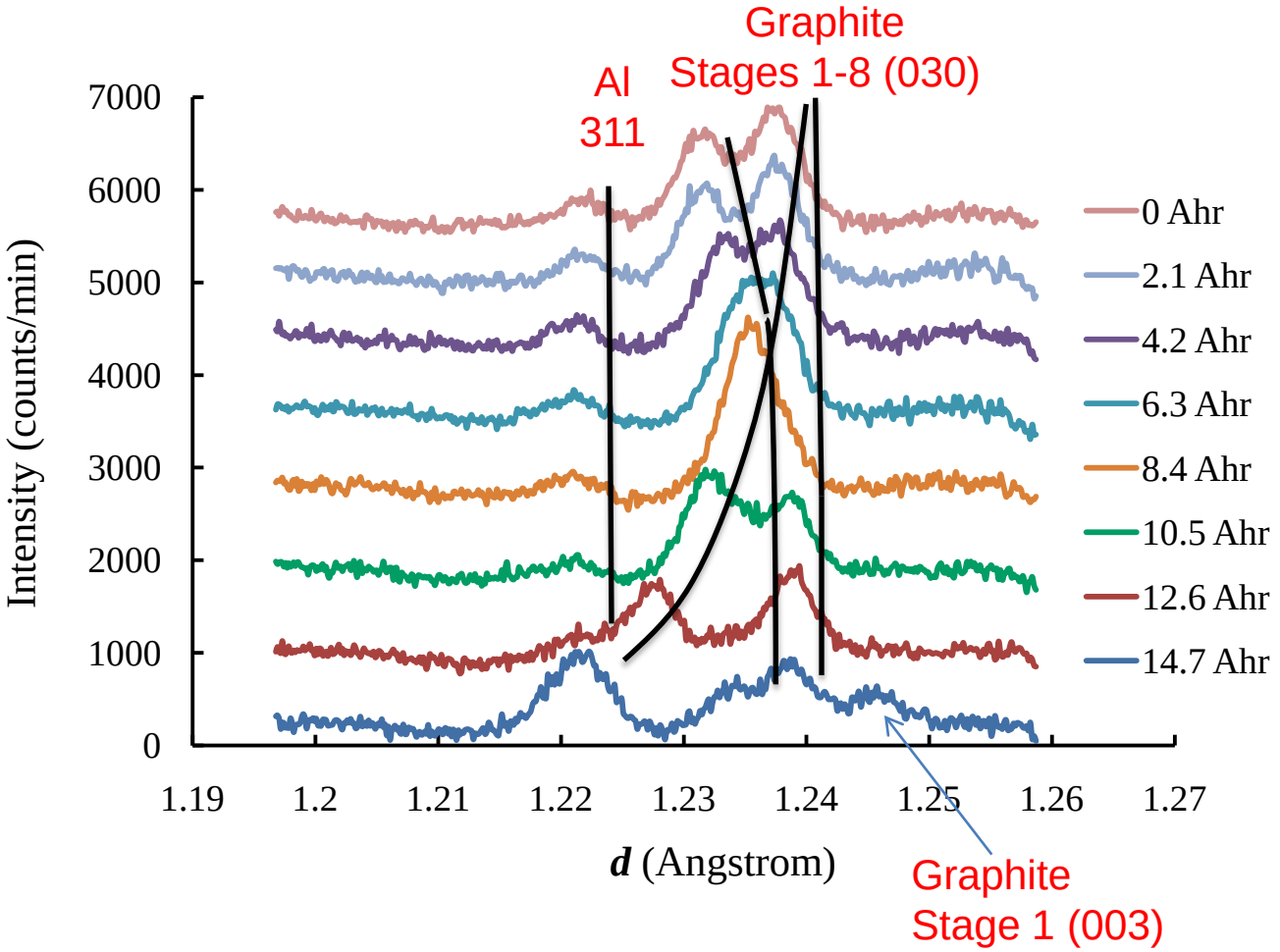
In situ neutron diffraction study of a production cell during charging or discharging was conducted for 3 selected d-spacing ranges at NRSF2–HFIR*. Each region showed diffraction peaks associated with the phases in cathodes, anodes and current collectors.

	Neutron wavelength (angstrom)	2theta range	d-spacing range (angstrom)	Number of steps in SOC during discharge or charge	Probable phases
Region 1	1.73	86.8-92.6	1.20-1.26	24 steps during charge 7 steps during discharge 35 steps during discharge	Al (311) $Li_xMn_2O_4$ Li_xC_6 (?)
Region 2	1.73	92.5-97.2	1.15-1.20	9 steps during discharge	Li_xC_6 $Li_x(Co_{1/3},Ni_{1/3},Mn_{1/3})O_2$
Region 3	2.66	87.0-110.0	1.62-1.93	10 steps during discharge	Cu (200) Li_xC_6 Li_xMnO_2 $Li_xMn_2O_4$

* **NRSF2** = 2nd generation neutron residual stress mapping facility; **HFIR** = High Flux Isotope Reactor

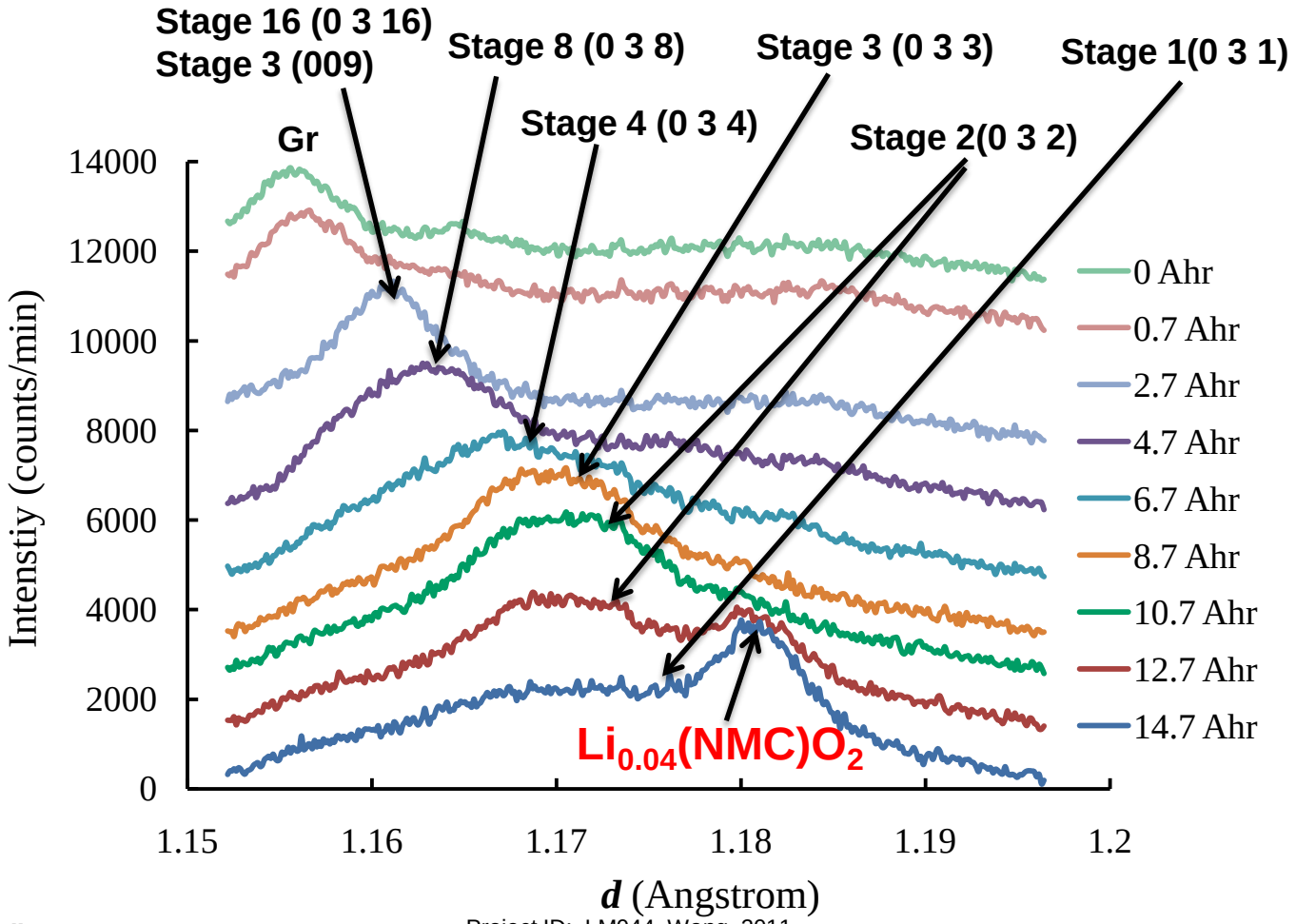
General Motors R&D Center User Project: Accomplishments – *In situ* Neutron Diffraction

Neutron diffraction peaks in Region #1 between 1.20 – 1.26 Å as a function of cell capacity. Data were collected in 7 steps during discharging.



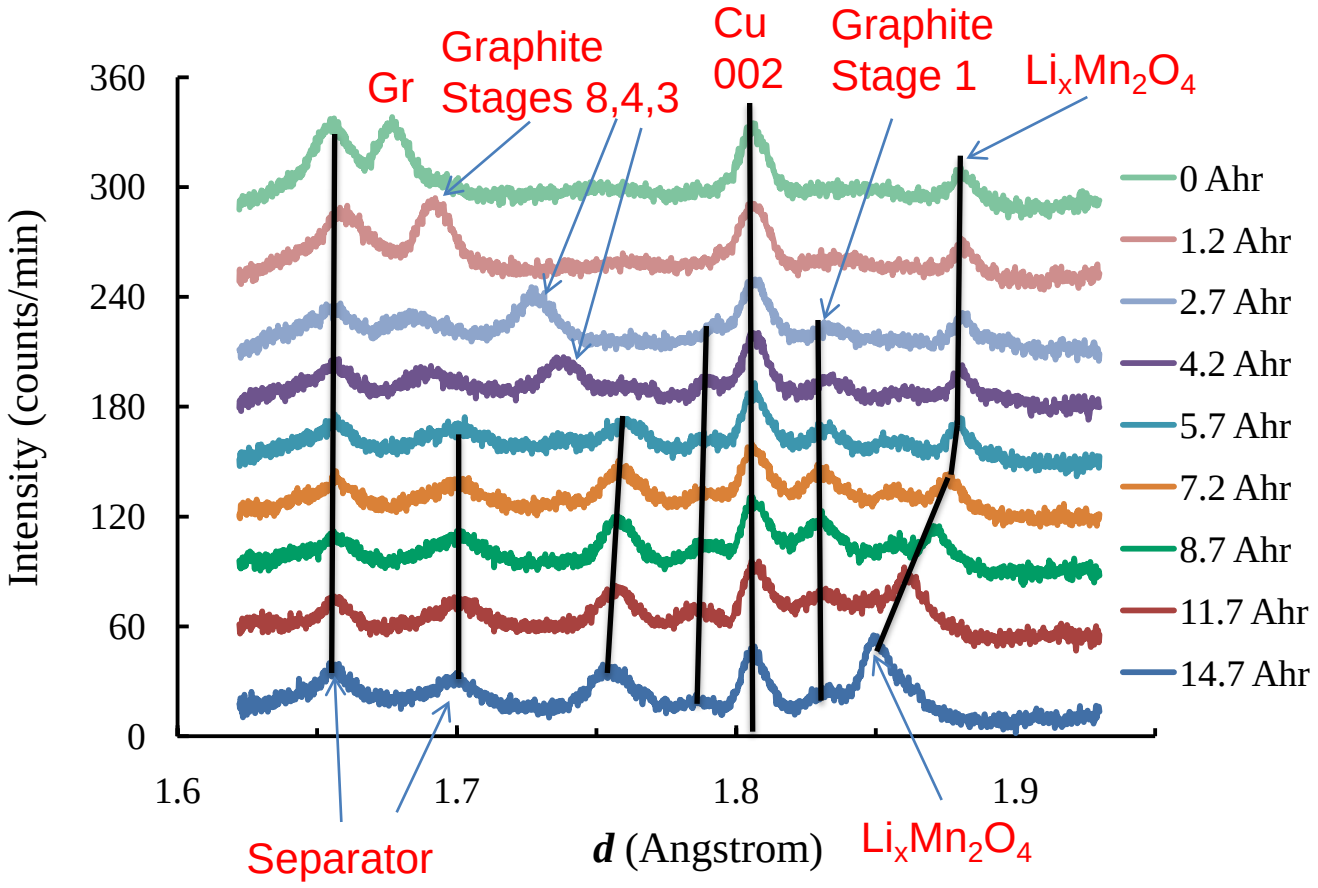
General Motors R&D Center User Project: Accomplishments – *In situ* Neutron Diffraction

Neutron diffraction peaks in Region #2 between 1.15 – 1.20 Å as a function of cell capacity. Data were collected in 9 steps during discharging.



General Motors R&D Center User Project: Accomplishments – *In situ* Neutron Diffraction

Neutron diffraction peaks in Region #3 between 1.62 – 1.93 Å as a function of cell capacity. Data were collected in 9 steps during discharging.



General Motors R&D Center User Project: Summary

- GM utilized the unique combination of world-class characterization capabilities and expertise available through the HTML User Program, located in one convenient location, to non-destructively study the performance of Li-ion batteries for extended-range electric vehicles.
- The use of a high-speed, high-resolution infrared camera and the HFIR NRSF2 neutron beam line managed by the HTML User Program enabled *in situ* monitoring of temperature changes and phase changes of production cells during charging and discharging
- The results obtained from this HTML User Program project will enable GM to develop a better understanding of cell performance, which will result in cells with improved performance and durability. The results from this project contribute toward achieving the goals of the Vehicle Technologies Program's Energy Storage activity.

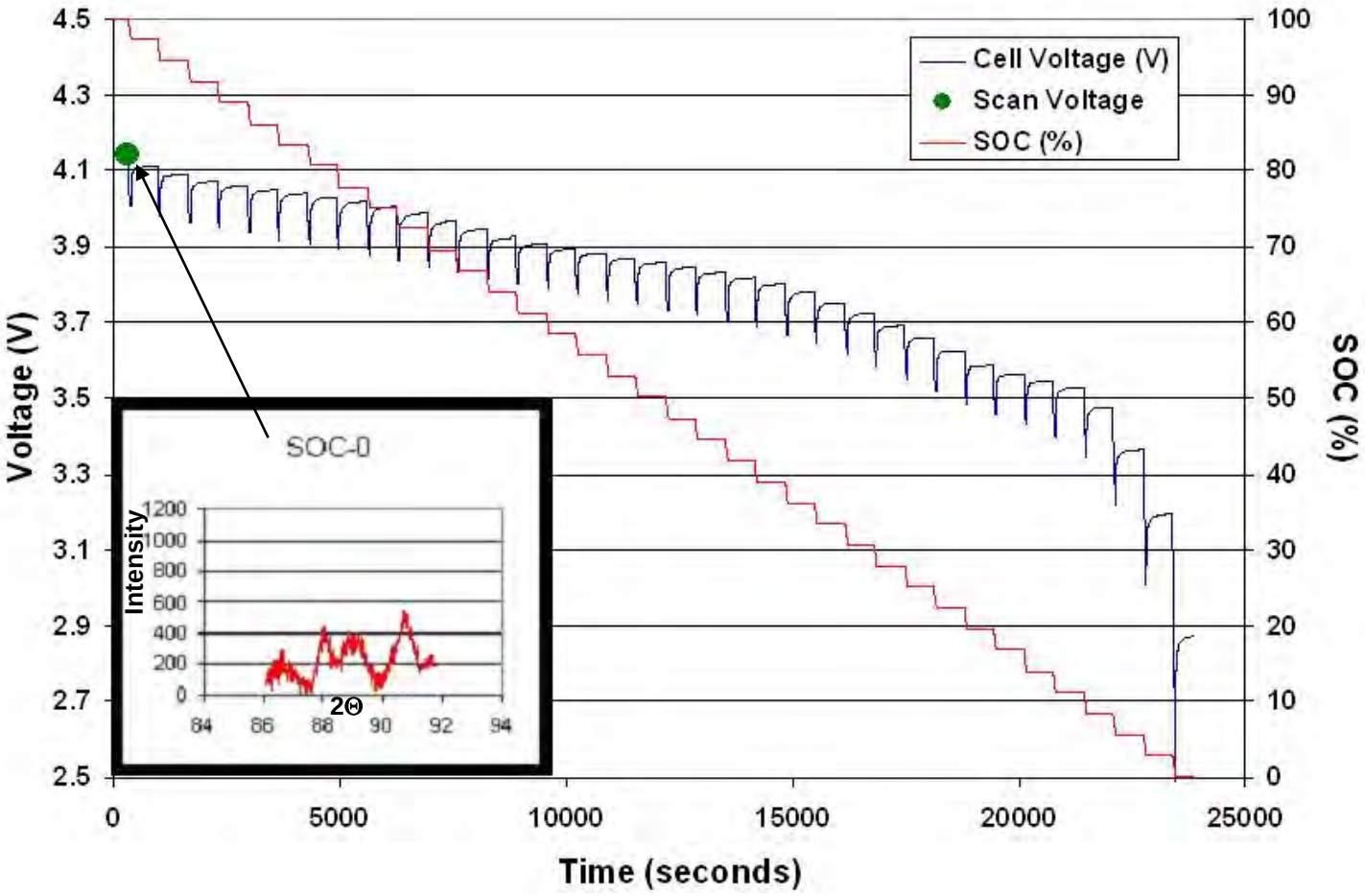
General Motors R&D Center User Project: Future Work

- HTML User Program projects are expected to be completed within 24 months.
- The experimental phase of this project has been completed, and its objectives have been met. Work is in progress to complete three papers containing results from this investigation.
- GM plans to continue its collaboration with the HTML through new projects submitted to the HTML User Program that help achieve goals set by the DOE Office of Vehicle Technologies.

Technical Back-up Slides

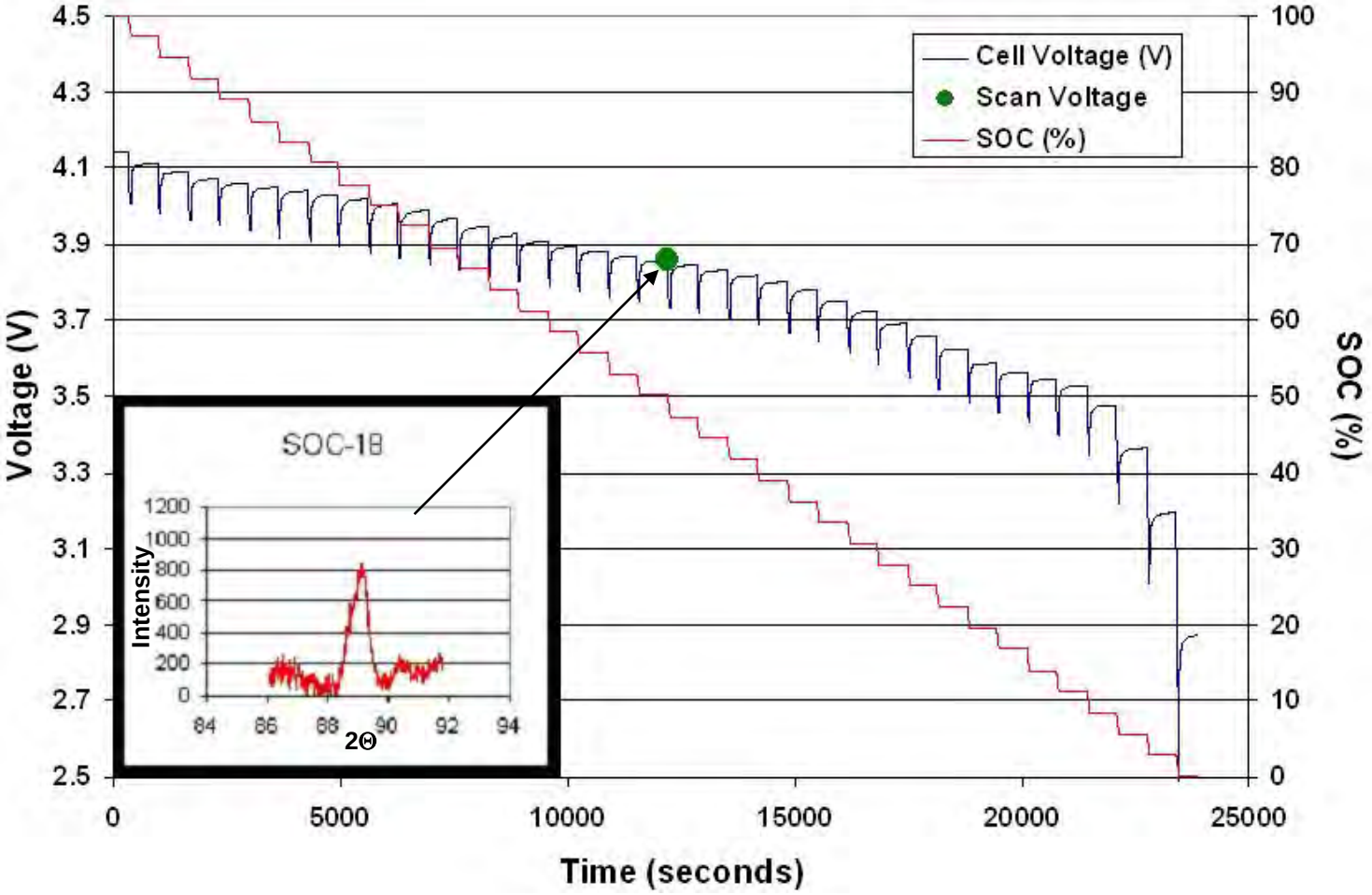
General Motors R&D Center User Project: Selected Neutron Diffraction Pattern

Neutron diffraction (Region 1) during discharging:
Neutron pattern for start-point of discharge



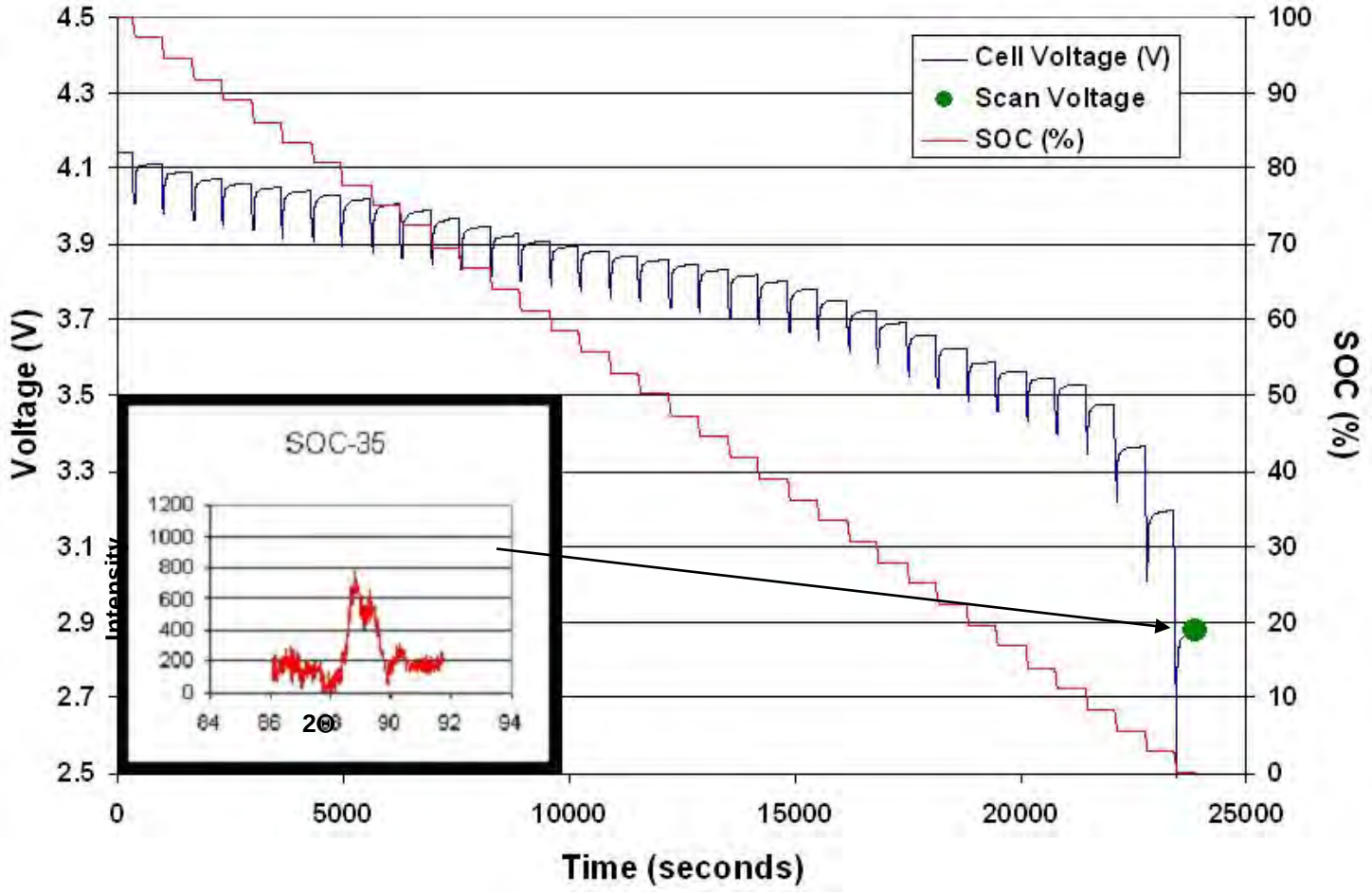
General Motors R&D Center User Project: Selected Neutron Diffraction Pattern

Neutron diffraction (Region 1) during discharging:
Neutron pattern for mid-point of discharge



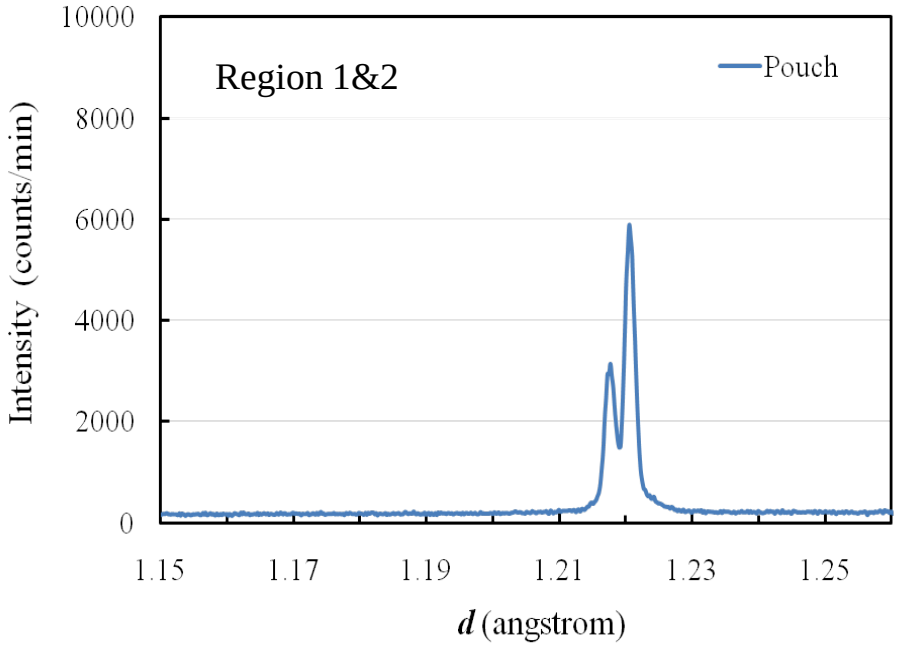
General Motors R&D Center User Project: Selected Neutron Diffraction Pattern

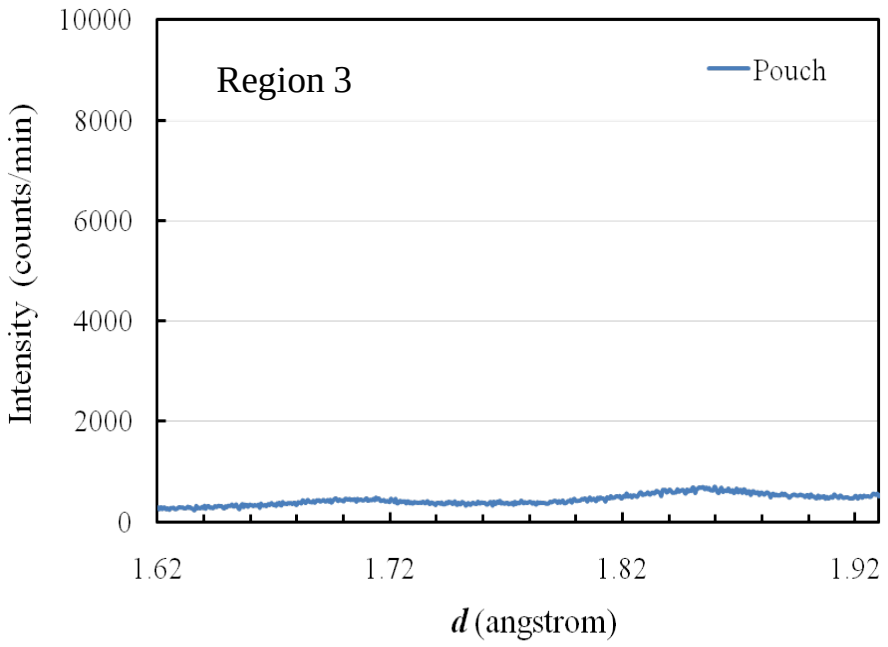
Neutron diffraction (Region 1) during discharging:
Neutron pattern for end-point of discharge



General Motors R&D Center User Project: Neutron Diffraction Patterns of the Pouch

Neutron diffraction of pouch showing two peaks in Region 1, indicating the presence of aluminum. Region 3 only showed two small humps related to the pouch polymer. By limiting the gage volume during the *in situ* tests to a region inside the cell, the effect of the pouch was minimized.





General Motors R&D Center User Project: Neutron Diffraction Pattern of the Separator

Neutron diffraction of separator showing several small peaks in Regions 1 & 2. Region 3 showed two larger peaks along with 1-2 small peaks. They are related to ceramic coatings. The *in situ* monitoring gage volume includes the separator.