

Giga Life Cycle: Manufacture of Cells from Recycled EV Li-ion Batteries

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ES205

DE-SC0006336

Overview

Timeline

- Project start: October 2012
- Project end: April 2015
- Percent complete: 99%

Budget

- Total project funding
 - DOE share \$1M
 - Contractor share: N/A
- Funding received in FY14 \$500K
- Funding for FY15: 0

Barriers

- Noted VT Barriers that are addressed with Advanced Battery Recycling
 - Cost: The project shows feasibility to reduce or eliminate the end-of-life cost for recycling advanced lithium-ion chemistries.
 - Rare Earth Minerals: The project successfully recycles lithium and dilute cobalt cathode materials for advanced battery manufacturing.
 - Abuse Tolerance, Reliability and Ruggedness: The project demonstrates successful recycling of 10-20% capacity faded & other severely abused material, which proves ongoing reliability and ruggedness of advanced materials after use and abuse.

Partners

- Oregon State University, Corvallis
 - Elemental, surface and structural analyses
- Dow Kokam (now Xalt)
 - Material source and manufacturing partner
- Project lead – OnTo Technology

Relevance / Objectives

Project Objectives:

1. *Working with a lithium battery manufacturer, demonstrate the number of times that large format lithium-ion, cobalt dilute batteries may be recycled.*
2. *Perform the rejuvenation process on the kilogram scale.*
3. *Assemble recycling prototype process-line.*
4. *Build full cells through manufacturing partner, with capacity > 2Ah.*
5. *Perform the recycling process on another material.*

Relevance to Vehicle Technology Program:

The Advanced Battery Recycling project develops manufacturing qualified material from recycling:

- A. *Increases the durability of materials from generation to generation of battery manufacturing, and*
- B. *Improves the affordability of advanced batteries by increasing the potential value at end-of-life.*
- C. *Helps to protect the Public Health and Environment with efficient recycling*

Impact on Barriers:

The Advanced Battery Recycling project impacts barriers:

- I. *Cost: The projects shows feasibility for a processor to buy end-of-life cobalt dilute batteries, and profitably sell recycled material at projected future costs: shows feasibility to end fees at end-of-life.*
- II. *Rare Earth Minerals: Cobalt and lithium are recovered from the active material and reused in recycled material, and remain in the battery material niche.*
- III. *Abuse Tolerance, Reliability and Ruggedness: Recycling of 50% faded batteries is demonstrated in the project, further demonstrating abuse tolerance of cobalt dilute chemistries, such as NMC, and their durability to be successfully recycled through this technology.*

Milestones

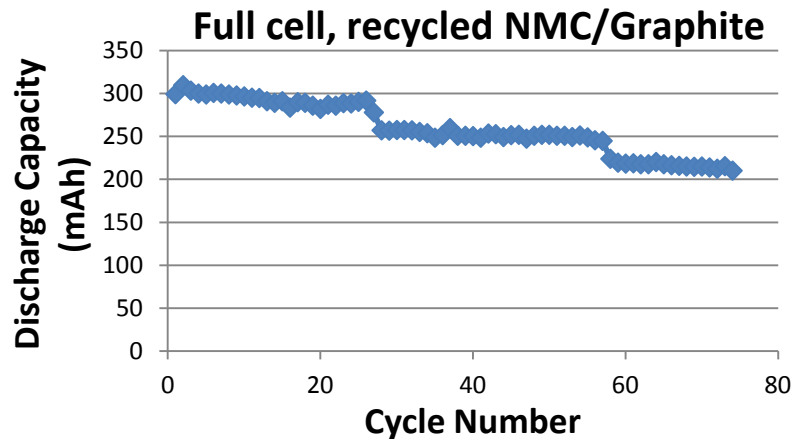
1. *Working with a lithium battery manufacturer, demonstrate the number of times that large format lithium-ion, cobalt dilute batteries may be recycled.*
 - *First iteration is underway.*
 - *Severely faded material is processed and recovers to match standard performance.*
 - *Full cells were manufactured from recycled material and compared to standards.*
2. *Perform the rejuvenation process on the kilogram scale.*
 - *Accomplished in 2012, refined in 2013 and 2014.*
 - *1 L reactor volume can host active material mass of 500g.*
 - *Mass yield of active cathode material is 87%, limited to transfer losses.*
3. *Assemble recycling prototype process-line.*
 - *Accomplished in 2012, refined in 2013 and 2014.*
4. *Build full cells through manufacturing partner, with capacity > 2Ah as requested in the RFP.*
 - *First 2Ah cells built from recycled material are being tested in parallel with standard material.*
 - *High yield allows for a total of six 2.2Ah cells to be made from scrap feedstock of 16Ah.*
5. *Perform the recycling process on another material.*
 - *Accomplished with NMC-lithium metal oxide battery formulation in 2013*
 - *Another cobalt dilute lithium-ion formulation is under evaluation.*

Approach/Strategy

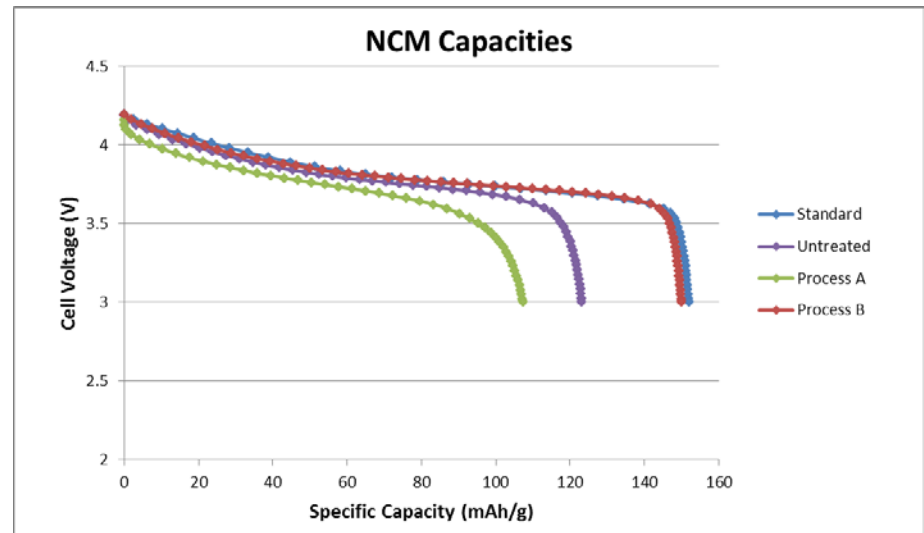
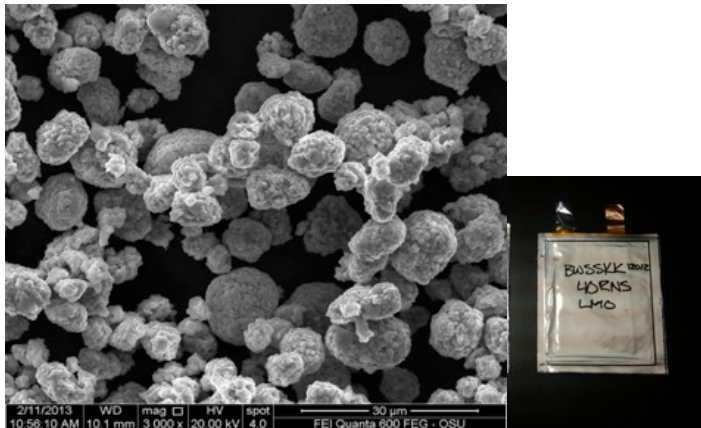
- Unique aspects of the developmental work:
 - Harvest and rejuvenate cathodes from 10-20% & 50% faded cells.
 - Match the performance characteristics with standards.
 - Recycled cells perform like standards: 1400 cycles and 87% capacity retention.
- Developments to address the project's technical barriers:
 - Develop process scale.
 - Reproduce recycled material performance with process scale.
- Project integration with other VT deployment projects:
 - Recycling of NMC EV grade batteries from XALT Energy.
 - Manufacture of new cells with recycled NMC in a relevant, industrial environment.
- Milestones completed from FY14.
 - First professional grade, 2.2 Ah full cells, manufactured in OEM setting.
 - 1400 cycles and counting with 87% capacity retention.
 - Performance matches standard material in side by side testing.

Proof of Concept Stage:

Recycling NMC from 13% faded cells



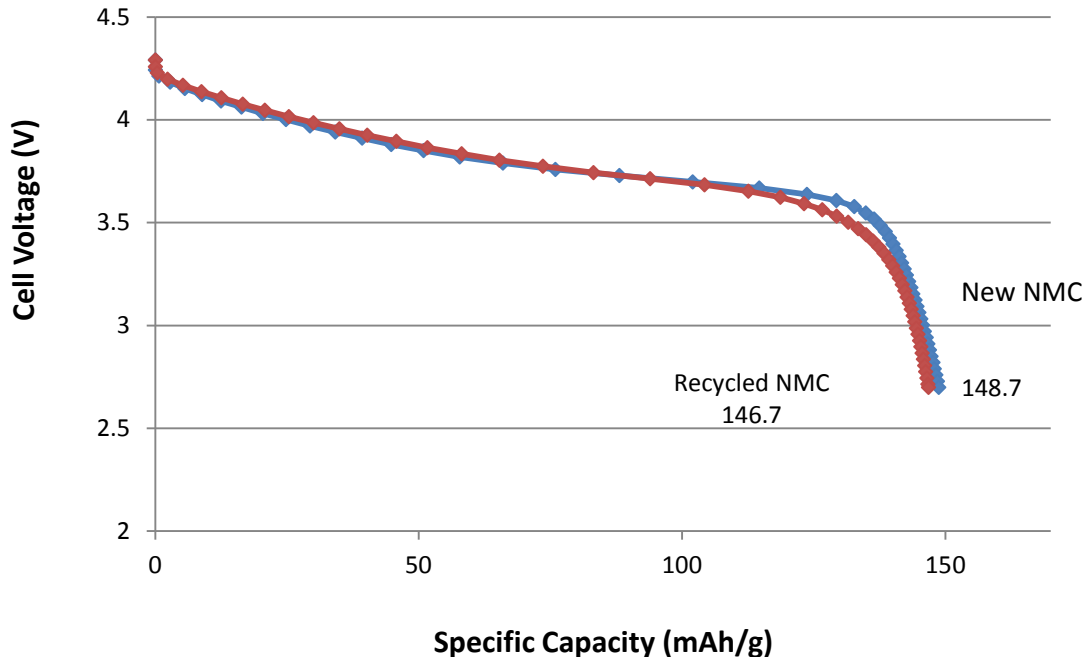
- Recycled NMC (R-NMC) matches specific capacity of standard NMC. (graph below from Q4 '12)
- R-NMC maintains rosette appearance. (SEM Q1 '13)
- First full cell with graphite, made in-house, has good cycle characteristics. (graph to left, representative pouch-cell in photo, Q1 '13)
- Challenges Identified: rate capability of R-NMC was low compared with NMC. Resolution of that challenge is demonstrated in the next technical accomplishment slides.



Technical Accomplishment:

Development of R-NMC rate capability performance Starting from lightly faded cells

Recycled and new NMC C/2 Rate Test comparison
lithium-half-cell (Q4 2013)

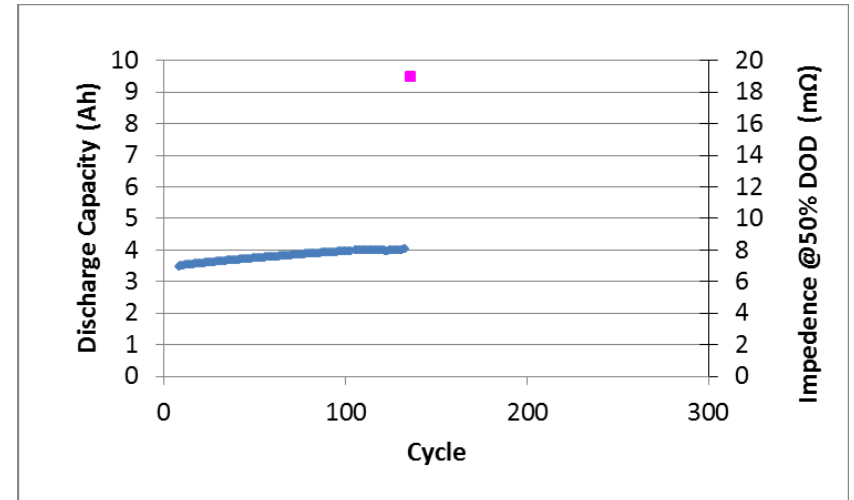
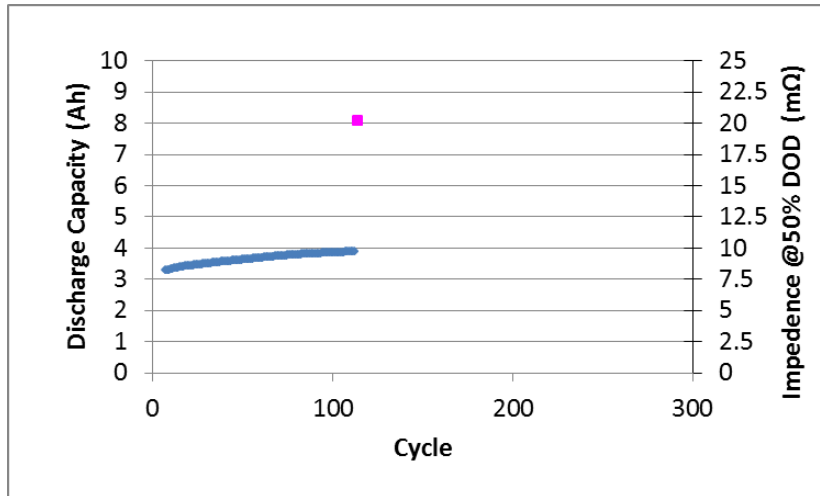


- Recycled NMC (R-NMC) matches specific capacity of standard NMC at C/2. (Q4 '13)
- Resolves challenges identified for rate capability.
- Move onto processing of heavily faded cells at larger scale.

Addresses program goal to match performance criteria between recycled and standard material, and to work with a lithium-ion battery manufacturer.

Phase II, Increase Scale:

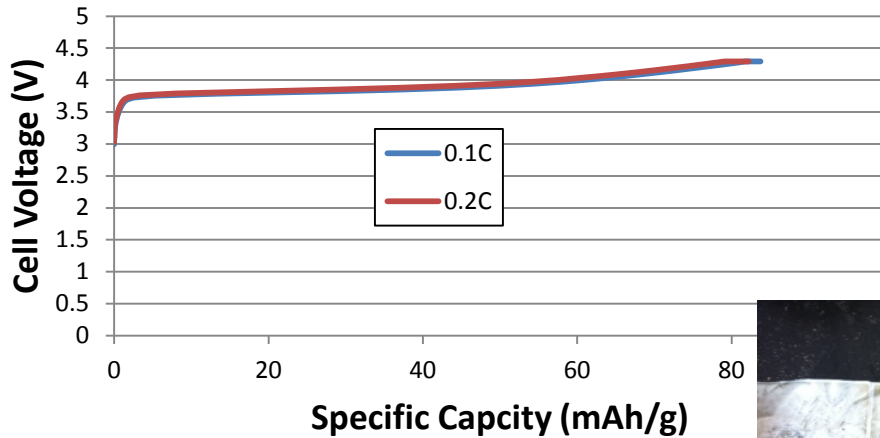
Recycle 50% faded batteries at the kilogram scale.



- The original cell capacity is 8Ah, the faded cells were supplied by XALT and characterized prior to recycling.
- Capacity measurement of 50% faded cells used in the recycle study.
- First cell C-rate cycle-life at 45°C (LHS) and second cell (RHS).
- Cell impedance at 50% SOC from 25A 10s pulse.

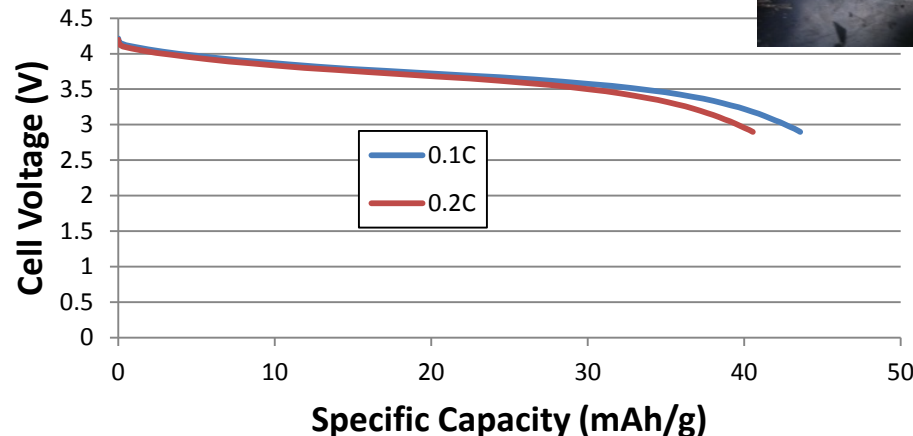
Harvested material characteristics from faded cells: The “before recycling” picture

Unprocessed NMC: Charge



Top photo:
Representative 8Ah cell for the study had 3.9Ah and 4Ah remaining capacity or 50% fade.

Unprocessed NMC: Discharge



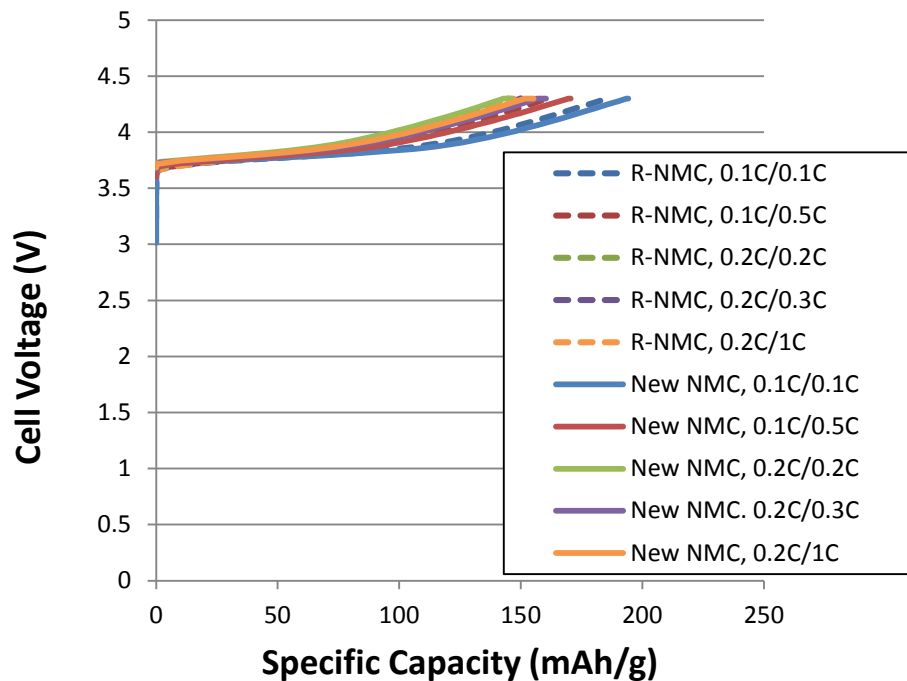
Middle and bottom photo:
Evidence of heavy cycling:
Discolored separator and current collector corrosion.



Specific capacity of the harvested material is 40-45mAh/g, which is less than 1/3 of standard material.

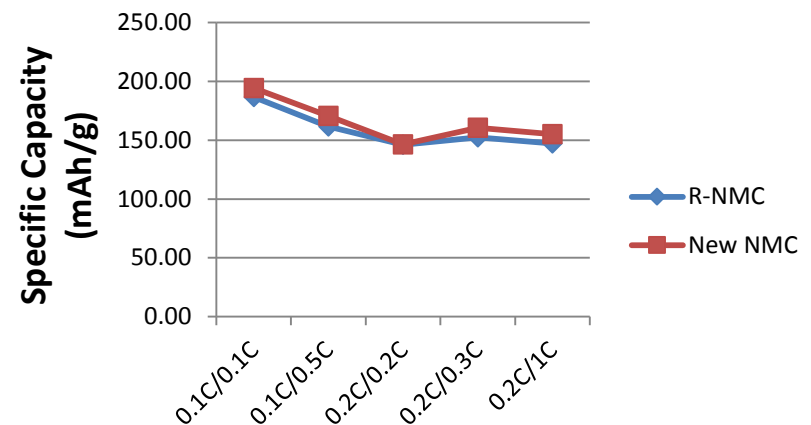
Technical Accomplishment:

Develop recycling process that produces high rate & capacity R-NMC
 Comparative charge data, **source material is 50% capacity faded**



Specific Charge Capacities (mAh/g)		
Cycle (Rate)	R-NMC	New
1: 0.1C	187	194
2: 0.1C	162	171
3: 0.2C	146	146
4: 0.2C	152	160
5: 0.2C	147	155

Matches rate capability to within 4% of new material.

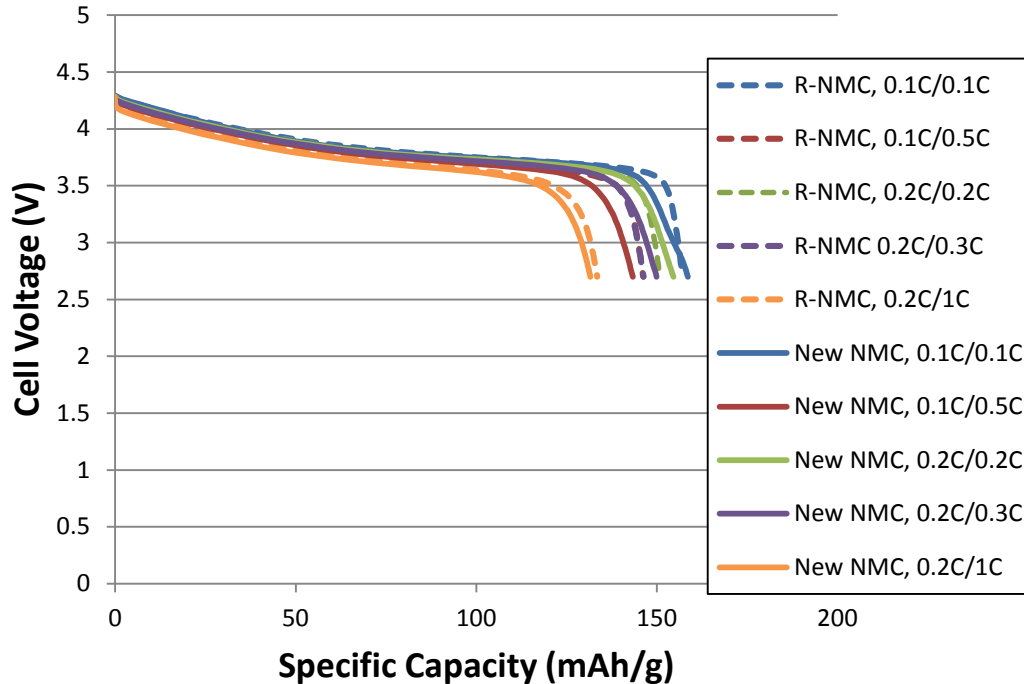


Addresses program goal to match performance criteria between recycled and standard material, develop a kilogram scale reactor for recycling, and to work with a lithium-ion battery manufacturer. (successful prequalification for manufacture).

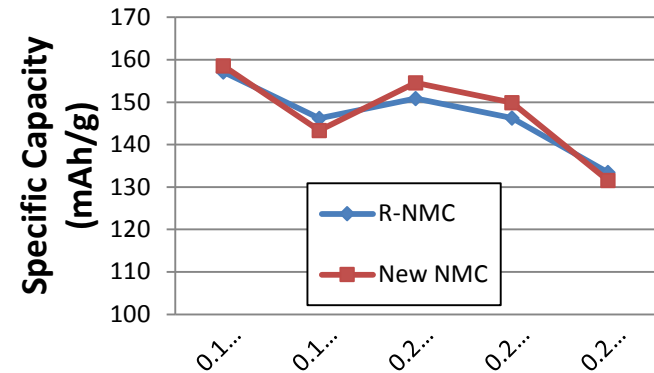
Technical Accomplishment:

Match the high rate & capacity of R-NMC with standard material

Discharge Data, source material is 50% capacity faded



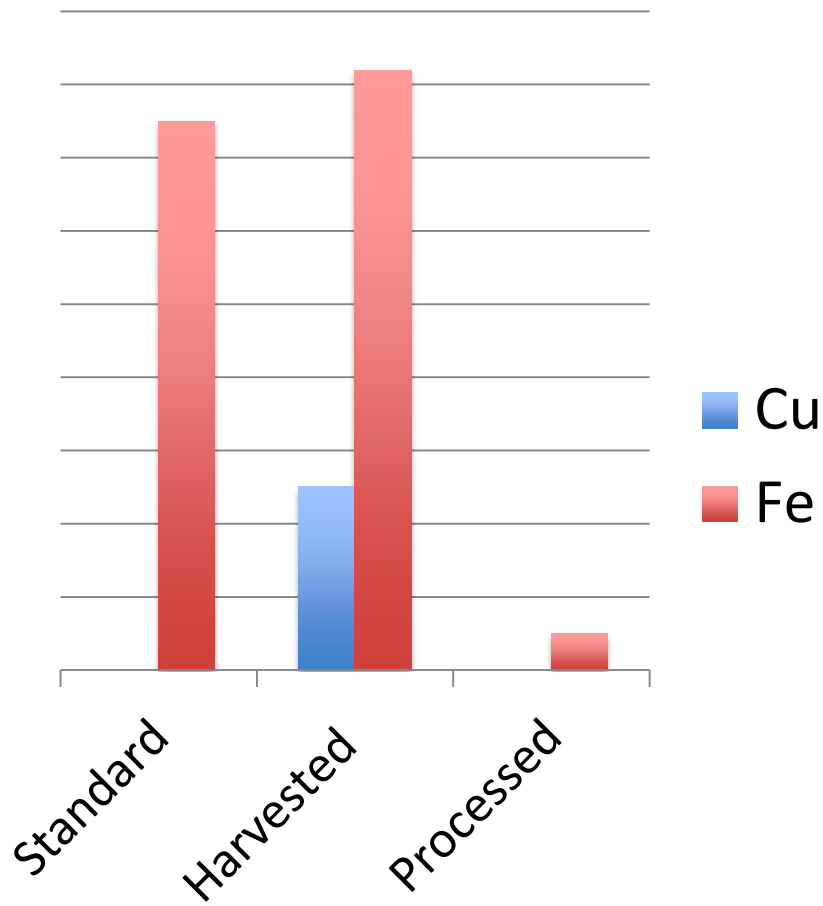
Specific Discharge Capacities (mAh/g)		
Cycle (Rate)	R-NMC	New NMC
1: 0.1C	157	159
2: 0.5C	146	143
3: 0.2C	151	155
4: 0.3C	146	150
5: 1C	133	132



Matches rate capability between recycled and new material, within 2%.

Addresses program goal to match performance criteria between recycled and standard material, develop a kilogram scale reactor for recycling, and to work with a lithium-ion battery manufacturer. (successful prequalification for manufacture).

Low Trace Metal Content in Recycled Cathode



- Relative trace content of Fe and Cu in cathode
 - Standard
 - Harvested
 - Final processing
- Recycled cathode is cleaner than standard cathode.

Technical Accomplishment:

Cells built from recycled material

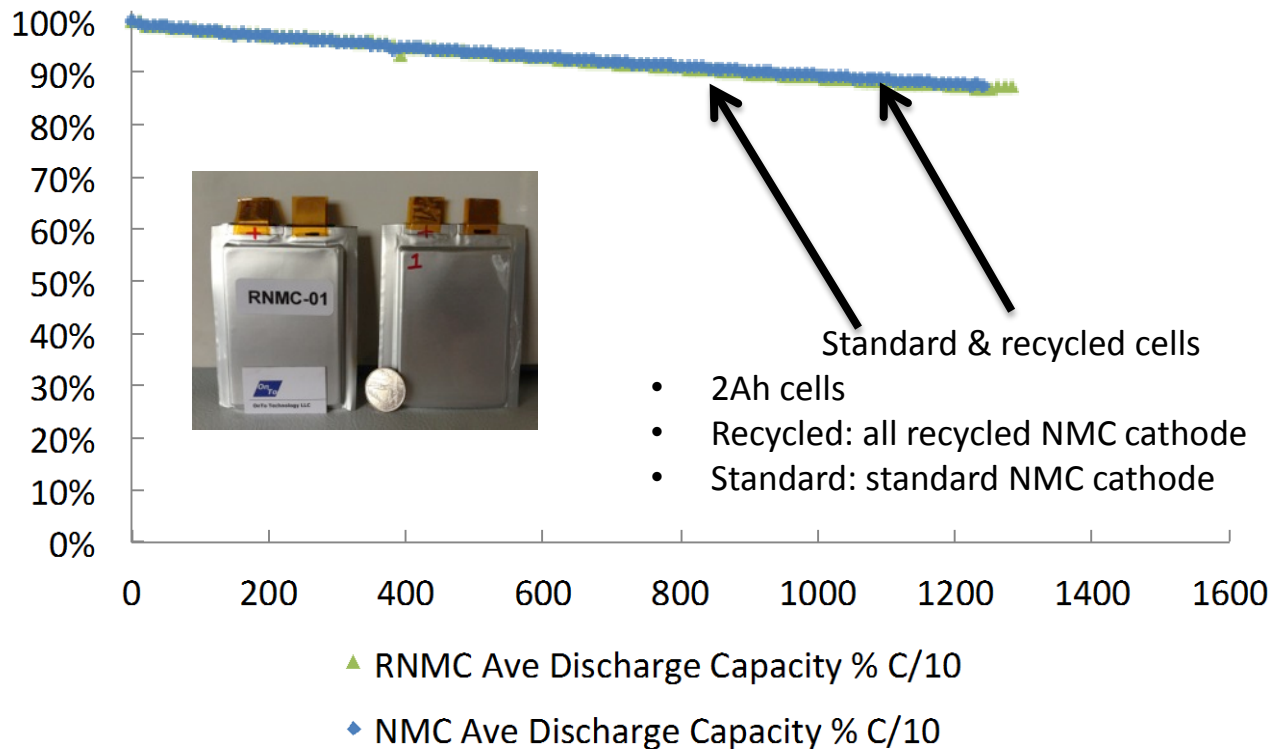
Standard graphite/R-NMC, 2.2 Ah capacity, meets cell criteria from RFP



- *source material is 50% capacity faded*
- Six recycled material cells with 2.2Ah capacity @ 0.5C @ 30°C
- Five standard cells with 2Ah capacity @ 0.5C @ 30°C.
- Same coating specifications, one extra cathode/anode sheet in the recycled cell to provide an offset in capacity vs. cycle # comparison.
- Cycling is underway: C/2 / C/2 (CC charge/CC discharge), 4.2-2.7V, 30°C

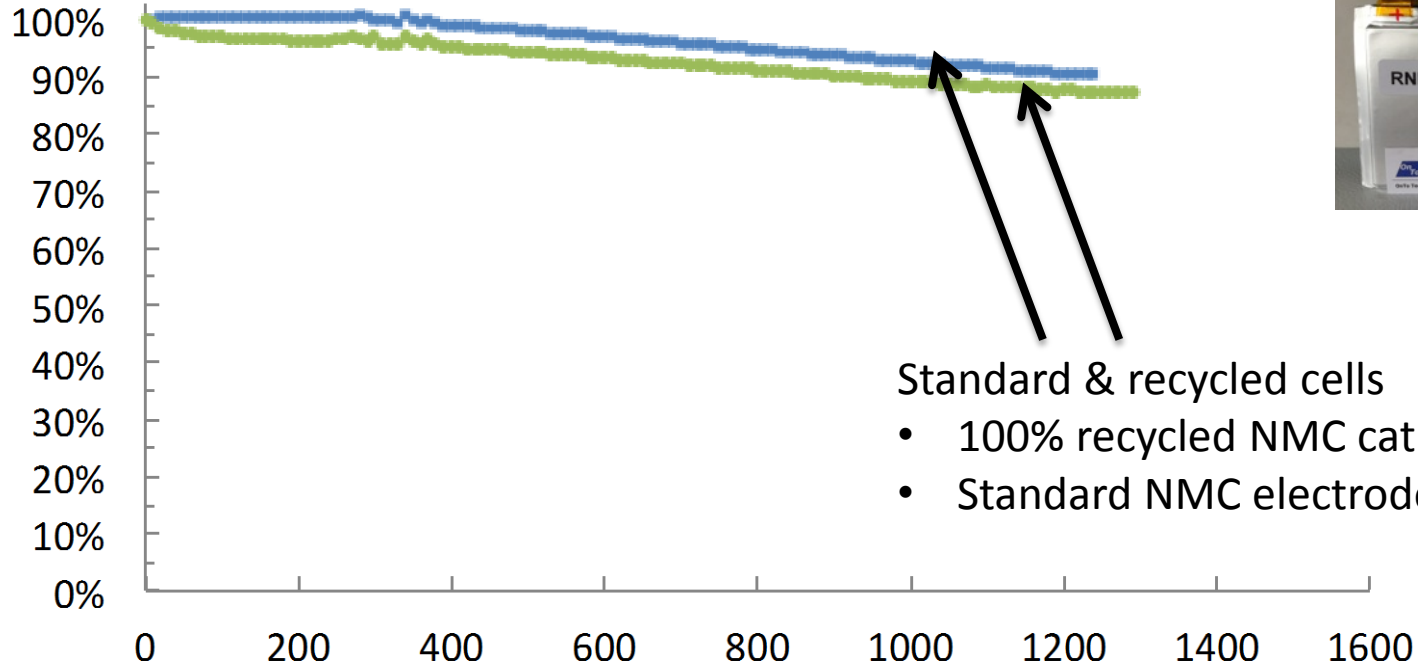
Addresses program goal to have an OEM build full cells from recycled and standard material. Two, faded 8Ah cells were recycled, the material was used to produce six 2.2Ah cells; demonstrates high yield, losses due to material transfer. Addresses VT cost barrier for end-of-life disposal/destruction fees. Demonstrates feasibility to recycle material for battery manufacturing; addresses rare earth material, and abuse tolerance.

Significant New Results for Recycled NMC Lithium-ion Batteries



Addresses program goal to have an OEM build full cells from recycled and standard material. Demonstrates feasibility to recycle material for battery manufacturing; addresses conservation/reuse of lithium; recycling of faded cells demonstrates the ruggedness and abuse tolerance for advanced materials.

Cycle Life comparison at C/2



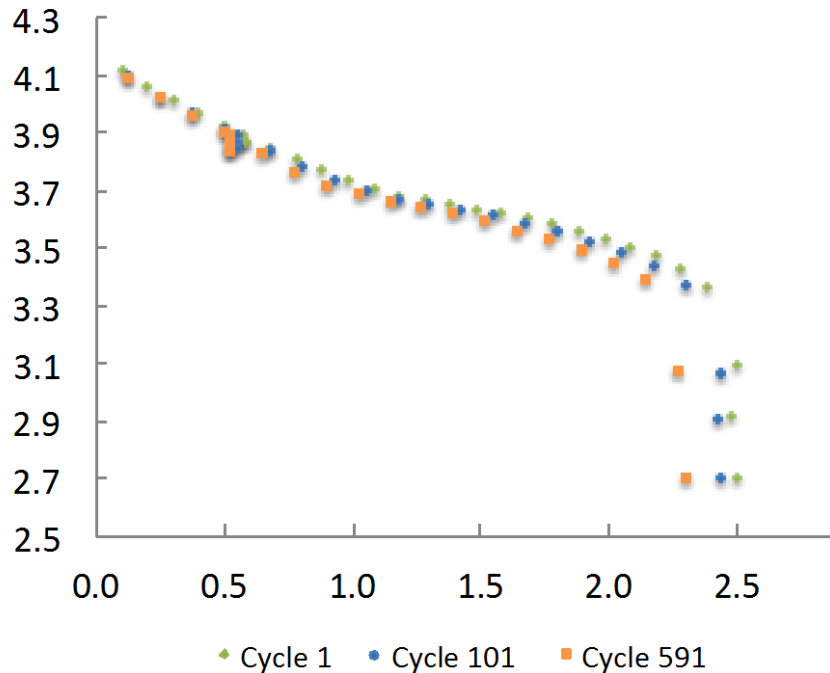
Standard & recycled cells

- 100% recycled NMC cathode
- Standard NMC electrode

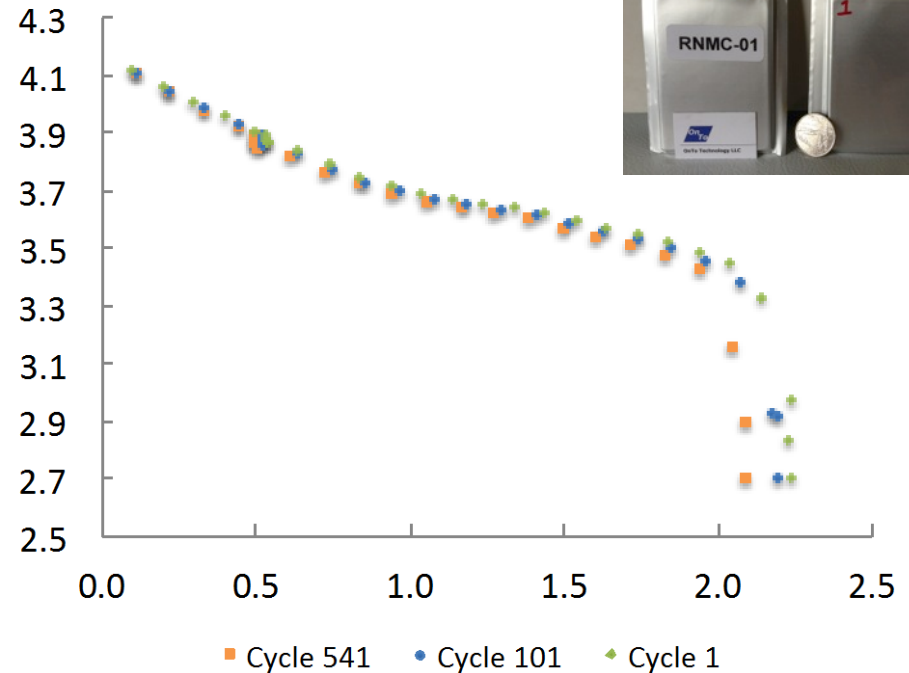
- NMC Ave % Discharge Capacity C/2
- RNMC Ave % Discharge Capacity C/2

Voltage / V vs. Discharge Capacity / Ah

Discharge profile for Recycled and Standard NMC

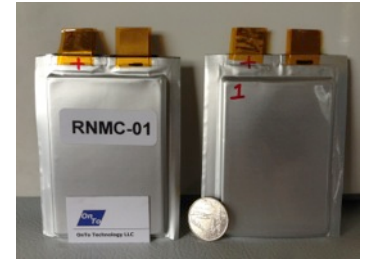


Recycled NMC, Cell #1, C/10



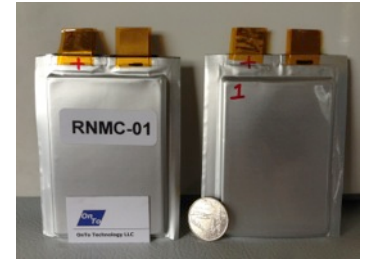
Standard NMC, Cell #4, C/10

Discharge voltage vs. capacity of recycled material matches standard



Performance of Recycled and Standard NMC

- Voltage profile is the same for standard and recycled.
- The recycled C/2 fade is slightly greater than the standard. Both retain 87% after ~1400 cycles.
- The C/10 fade is identical for both recycled and standard material.



Patents for Processing & Battery Deactivation:

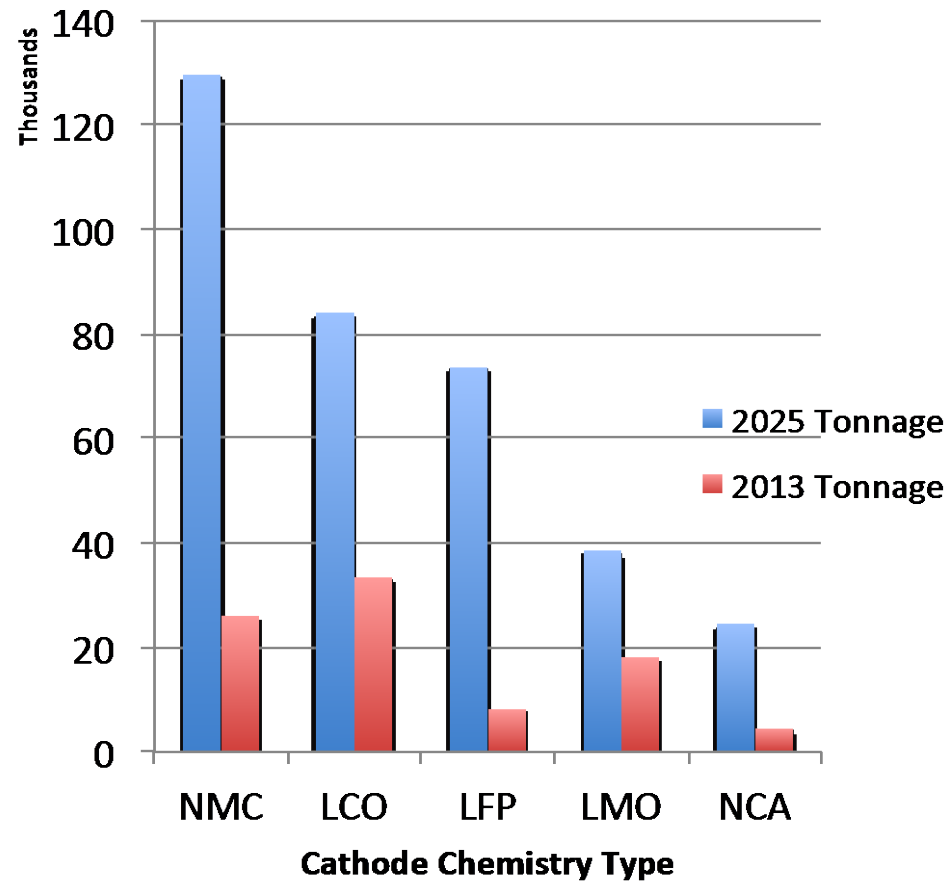
U.S. Patent #	Title (assignee)
8,846,225	Reintroduction of lithium into recycled battery materials (Sloop)
8,823,329	Discharging of Batteries (Sloop)
8,497,030	Recycling Batteries Having Basic Electrolytes (Sloop)



- # 8,846,225 reduced to practice in this work.
- Features:
 - Ease: does not require solid-state reaction calibration.
 - Flexibility: applicable to most metal oxides.
 - Priority: early dates over other direct methods.
- Advantages:
 - Low cost.
 - Larger market capability.
- Benefits:
 - A more profitable, efficient and service-oriented recycling industry.
 - Elimination of costs in the EV market: such as disposal/recycling fees, and expensive replacement of materials
 - Eliminate \$25-33M in life cycle costs per 10,000 vehicles.

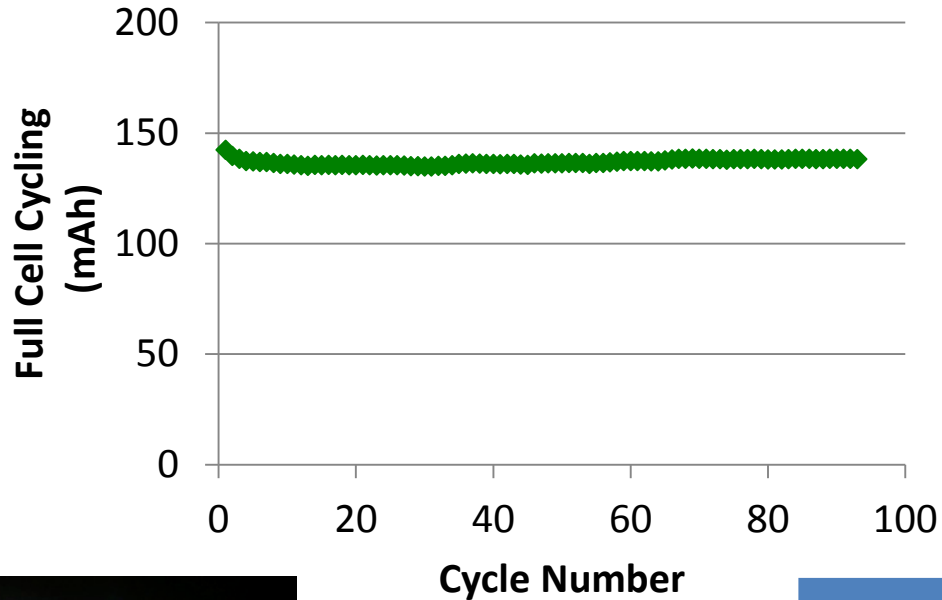
Impact: Save Millions on Future Annual EV costs

- Li-ion growth through 2025.
 - NMC: Li nickel manganese cobalt oxide
 - LCO: Li cobalt oxide
 - LFP: Li iron phosphate
 - LMO: Li manganese oxide “spinel”
 - NCA: Li nickel cobalt aluminum oxide
- Cathode market value estimate.
 - 2013: \$2.4B
 - 2025: \$7.3B
 - 5% recycled material by 2025 has a cathode market potential of >\$370M
- Recycling Opportunity per 10,000 EV packs. The significance is as follows:
 - Disposal cost elimination: \$25M
 - new material for sale: \$8M
 - Total estimated positive impact: \$33M



Proof of concept on another system:

Mixed Oxide and LMO from Nissan, faded by 20% before processing.



Addresses program goal for recycling an additional cobalt dilute chemistry. This is a mixed oxide typical of a large fleet EV.



System	mAh/g	Rate
Standard/ Li	130	C/20
Recycled/ Li	130	C/20

Collaborations

- XALT Energy
 - Contributor.
 - Industry within the VT Program.
 - Critical collaborator supplying new and faded cells, standards and recycled. material manufacturing test runs.
- Nissan-Sumitomo
 - Contributor.
 - Industry outside of the VT Program.
 - Provided relevant battery packs for recycle testing.
- Chrysler
 - Contributor.
 - Industry within the VT Program.
 - Provided relevant battery packs for recycle testing.
- Oregon State University
 - Subcontractor.
 - University, outside of the VT program.
 - Performed material characterization and analyses.
- Willamette University
 - Subcontractor.
 - University outside of the VT program.
 - Performed material characterization and provided undergraduate internships.

Remaining Challenges and Barriers

- Will a second recycling iteration be as successful as the first?
- Complete description of the use and abuse limits for direct recycling.
- Commercialization of manufacturing with recycled material.

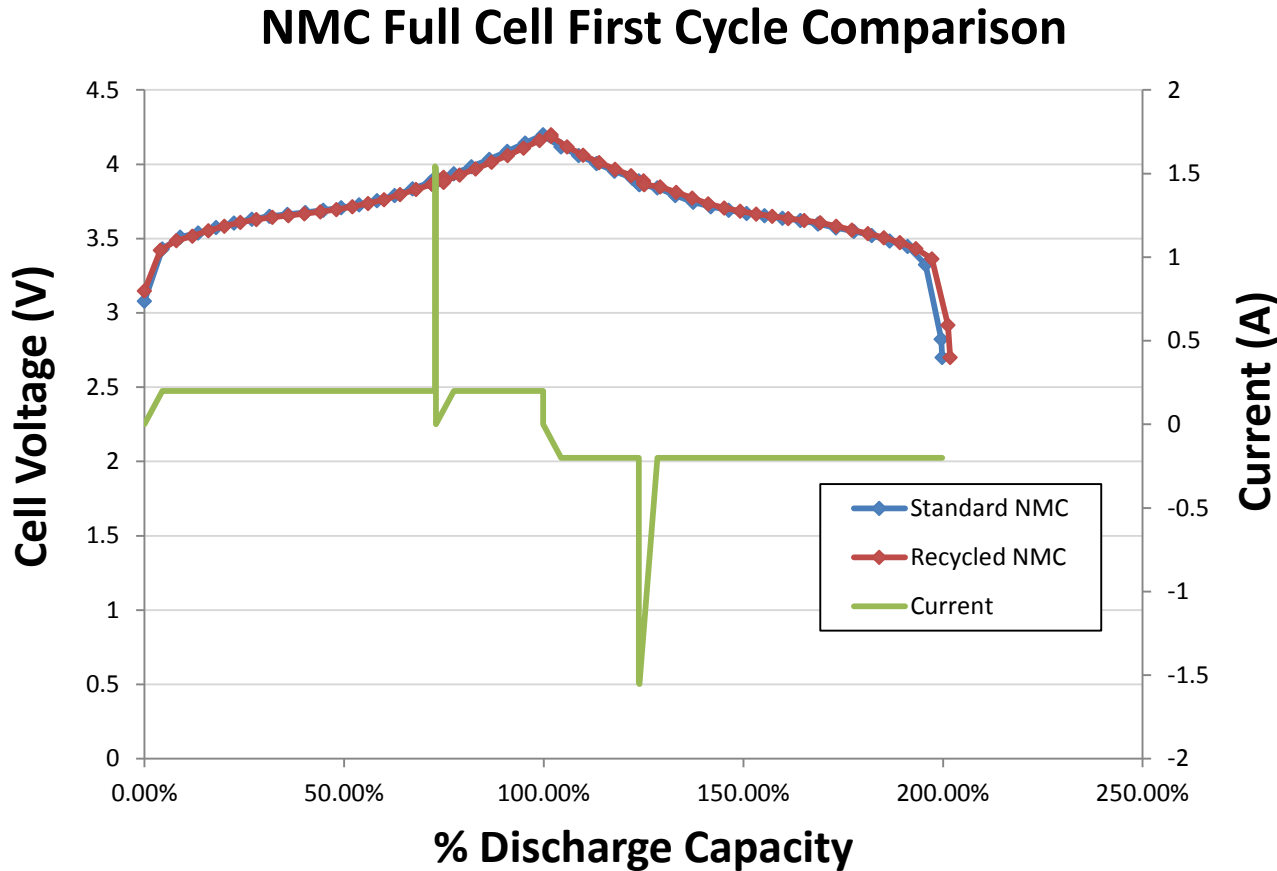
Proposed Future Work

- Harvest and rejuvenation of R-NMC cells for 3rd round manufacturing.
 - After side-by-side capacity fade comparison.
 - After post test analysis
- Develop recycling of mixed oxides demonstrated herein.
- Application of battery models to these cycle life results.
- Material characterization of R-NMC cells.
 - Techniques will be evaluated to remove potential contaminants that may be found in the recycled materials.
- Commercial manufacture of cells using recycled material.

Summary

- **Relevance:** Innovative recycling of high performance cobalt dilute lithium-ion chemistries can help achieve VT goals to reduce cost, demonstrate advanced materials durability, address critical material concerns, and help to protect the public health and environment.
- **Approach:** Teams with a manufacturing partner to qualify recycled material.
- **Technical Accomplishments:**
 - Heavily faded NMC cells were harvested and rejuvenated to produce R-NMC with charge/discharge characteristics equivalent to standard NMC.
 - Performed the process on liter scale reactor, representing a 10x scale of the proof of concept system.
 - Produced the first ever recycled vehicle grade lithium-ion batteries with 2.2Ah capacity.
 - Recycled and standard cells demonstrate EV capability.
- **Collaborations:** First ever manufacturing efforts with recycled material in an OEM setting with XALT Energy. Recycling development of EV grade, faded cells from manufacturers including XALT, Chrysler and Nissan. Undergraduate research efforts supported with internships with Willamette University; Graduate research efforts support the project with Oregon State University.
- **Future Work:** Commercialization of recycling and manufacture, application of the process towards other chemistries, characterization of recycled materials and development of the understanding of life-limitations for advanced battery materials.

Charge/discharge profile for test cells



- 2.2 Ah cells
- Arbin Instruments cycler.
- CD C/2 rate will be used to fade the cells
- Every tenth cycle
 - CD C/10
 - Impedance
 - Power pulse, (shown)
- A self discharge test will be performed after 25 cycles
 - 50°C, one week
 - 10°C, one week