U.S. DEPARTMENT OF ENERGY'S (DOE)
VEHICLE TECHNOLOGIES OFFICE (VTO)
2020 ANNUAL MERIT REVIEW (AMR)

ReCell
ADVANCED BATTERY RECYCLING

RECELL CENTER FOR ADVANCED BATTERY RECYCLING: CROSSCUTTING EFFORTS

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U.S. DEPARTMENT OF ENERGY | Energy Efficiency & Renewable Energy
VEHICLE TECHNOLOGIES OFFICE

PROJECT OVERVIEW

Timeline

- Project start: October 2018
- Project end: September 2021
- Percent complete: ~50%

Budget

FY19	$4,615k
FY20	$5,150k

Barriers

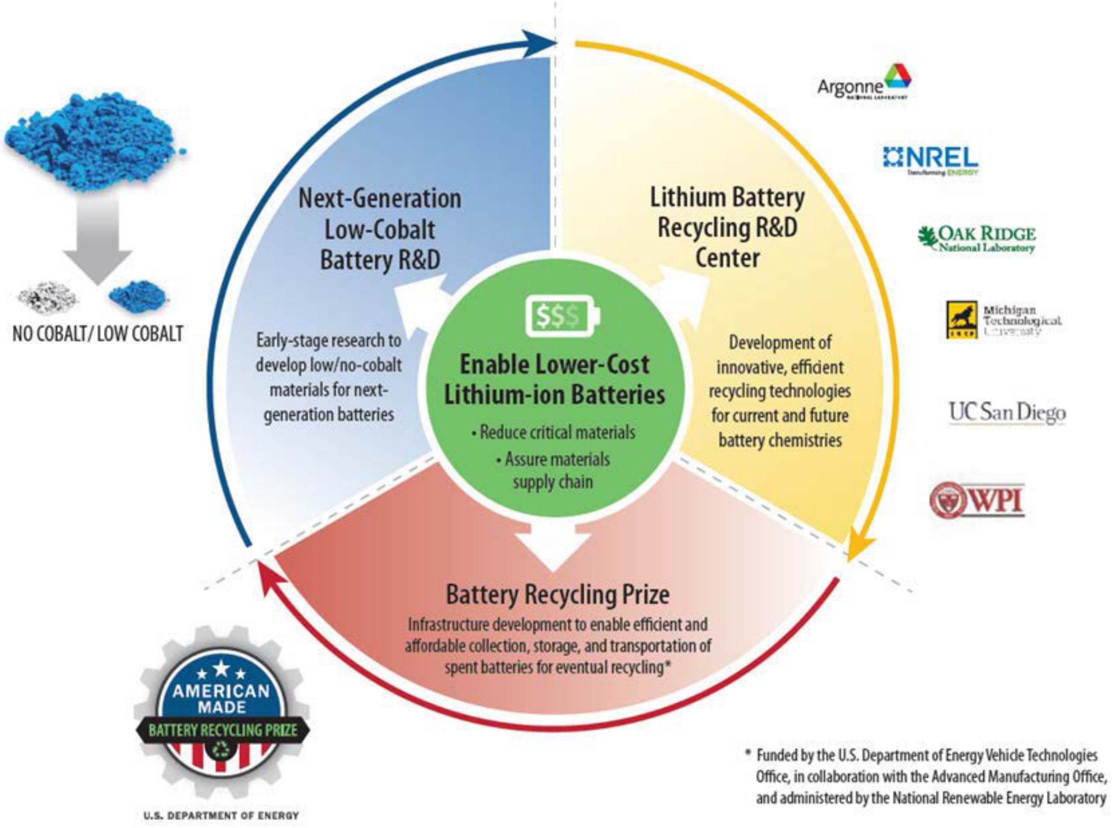
- Recycling and Sustainability
  - Cost to recycle is currently 5-15% of battery cost
  - Material shortage (Li, Co, and Ni)
  - Varying chemistries result in variable backend value

Partners

- Argonne National Laboratory
- National Renewable Energy Laboratory
- Oak Ridge National Laboratory
- University of California, San Diego
- Worcester Polytechnic Institute
- Michigan Technological University
- Industry

RELEVANCE

- Lower cost of batteries
- Enable lower environmental impacts
- Increase our country's energy security



APPROACH

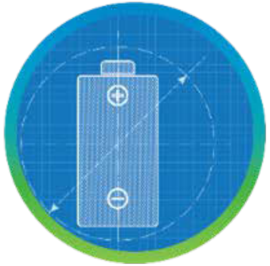
Year 1 – Bench scale testing

Year 2 – Start to scale up unit operations

Year 3 – Finish scale up and show cell to cell recycling

DIRECT CATHODE RECYCLING

OTHER MATERIAL RECOVERY



DESIGN FOR RECYCLING

MODELING AND ANALYSIS

Program Scope

Initiated as powder to cell recycling, but moving toward cell to cell

Does not include battery dismantling, transportation, or 2nd use

PROGRAM MILESTONES

FY19 Q1 Complete Establish the battery recycling center's mission and include its targets and goals

FY19 Q2 Complete Provide an initial progress report on roll-to-roll relithiation

FY19 Q3 Complete Provide an initial progress report on design for recycle initiative

FY19 Q4 Complete Establish the ReCell Center's Battery Recycling Laboratory and Scale-up Facility

FY20 Q1 Complete Electron Backscatter Diffraction data comparison of various chemically delithiated NMC-111 versus pristine NMC-111

FY20 Q2 Complete All five relithiation processes added to EverBatt at lab scale and production scale

FY20 Q3 Ongoing Down-select solvent(s) to separate black mass from current collector and optimize the process conditions to achieve >90% recovery of black mass

FY20 Q4 Ongoing Demonstrate recovery of anode and cathode powders using the new pilot scale froth column

Each Individual project has its own milestones. Though not listed here.

The COVID-19 pandemic has reduced lab time causing delays in completing FY20 milestones

CROSSCUTTING EFFORTS

APPROACH – CROSSCUTTING EFFORTS

- Standardize materials, cell types and test protocols used across different projects to compare different recycling methods (ANL/CAMP)
- Provide technical rationale and diagnostic criteria to determine suitability of specific methods of recycling (ANL/POST-TEST & NREL)
- Serve as quality check by quantifying chemical signatures at end of life and after regeneration (ANL/POST-TEST & NREL)
- Provide feedback to different recycling processes (e.g., re-lithiation parameters) (ALL)
- Provide technical input on quality of regenerated material from different recycling methods for cost metrics (ALL)

Cross Cutting Effort interacts will all Focus Areas under ReCell

APPROACH – CROSSCUTTING EFFORTS

List of crosscutting Projects and Impact

CAMP Facility Support - *Andrew Jansen (ANL)*
Standardizes materials, cell formats and testing protocols across different projects

Post Test Facility Support - *Ira Bloom (ANL)*
Provides additional information regarding the cause of performance degradation, which, previously, could be only inferred

Diagnostics on Aged Materials - *Shriram Santhanagopalan (NREL)*
Develops new tools beyond conventional morphology, phase-purity and surface characterization to ascertain quality of materials recovered from recycling streams

Thermal Analysis - *Matthew Keyser (NREL)*
Benchmarks thermal signatures of fresh versus recycled materials and relate these to cell performance

ESTABLISH ELECTROCHEMICAL PERFORMANCE OF BASELINE MATERIALS (ANL/CAMP)

Accomplishments: Standardize Test Protocols across ReCell Projects

Path I

Characterization Protocols + Cycling that mimic Calendar Life & Cycle Life Aging

- Formation: ReCell-FullCell-Form
  - 3.0 to 4.2 V for all Path I cycles
  - 4 x C/10 Chg & Dchg
- Rate: ReCell-FullCell-Rate
  - 2x C/20, 3x C/10, 3x C/5, then 3x Dchg at C/2, 1C, 2C with C/3 Chg; C/20 trickle Chg for all
- HPPC: ReCell-FullCell-HPPC
  - 1C Chg & Dchg; 5C Dchg & 3.75C Chg pulses
- C/2 Life Cycling: ReCell-FullCell-Cyc
  - 1x C/20, 48x C/3 Chg + C/2 Dchg, 1x HPPC
  - Repeat until <80% capacity retention

Path II

Quick Protocols to stress Cycle Life Aging

- 1C Fast Cycling: ReCell-FullCell-Age
  - 3.0 to 4.2 V for all Path II cycles
  - 4 x C/10 Chg & Dchg (Formation)
  - 500x 1C Chg + Dchg , (C/20 trickle Chg)

Active Materials: Graphite, NMC111, & Delithiated NMC111 (from MERF)

Half Cell Protocols similar with adjusted voltage windows

COMPLETED BASELINE MATRIX IN COIN CELLS

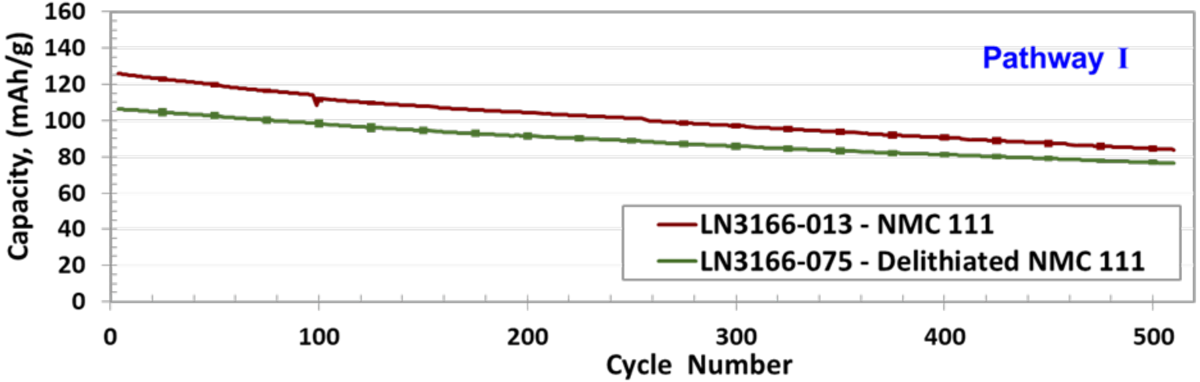
Accomplishments: Assembled & Tested in ≥4 Coin Cells Per Set at 30°C

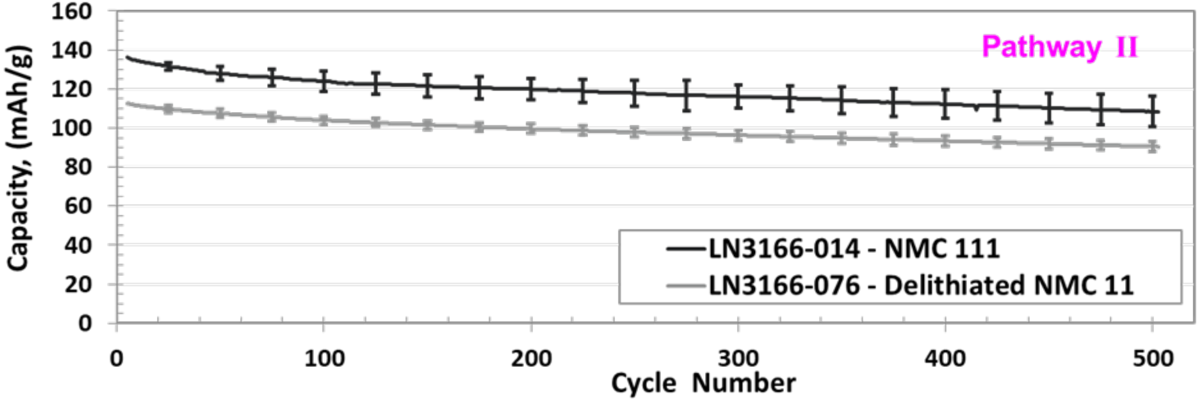
Anode	Cathode	Gen2 Electrolyte	Test Path	Testing Protocol Order			
				Profile 1	Profile 2	Profile 3	Profile 4
Li	NMC 111	Flooded	I	ReCell-Cathode-Half-Form	ReCell-Cathode-Half-Rate	-	-
Li	NMC 111	Flooded	II	ReCell-Cathode-Half-Form	ReCell-Cathode-Half-Cyc	-	-
Li	Delithiated NMC 111	Flooded	I	ReCell-Cathode-Half-Form	ReCell-Cathode-Half-Rate	-	-
Li	Delithiated NMC 111	Flooded	II	ReCell-Cathode-Half-Form	ReCell-Cathode-Half-Cyc	-	-
1520P	Li	Flooded	I	ReCell-Gr-Half-Form	ReCell-Gr-Half-Rate	-	-
1520P	Li	Flooded	II	ReCell-Gr-Half-Form	ReCell-Gr-Half-Cyc	-	-
1520P	NMC 111	29 µL	I	ReCell-FullCell-Form	ReCell-FullCell-Rate	ReCell-FullCell-HPPC	ReCell-FullCell-Cyc
1520P	NMC 111	29 µL	II	ReCell-FullCell-Age	-	-	-
1520P	Delithiated NMC 111	29 µL	I	ReCell-FullCell-Form	ReCell-FullCell-Rate	ReCell-FullCell-HPPC	ReCell-FullCell-Cyc
1520P	Delithiated NMC 111	29 µL	II	ReCell-FullCell-Age	-	-	-

CHEMICAL DELITHIATION PROCESS DID NOT DAMAGE NMC, ASIDE FROM LOWER CAPACITY

Accomplishments: Cycling performance of chemically delithiated cathodes

- This delithiated baseline cathode is suitable for use in processes being developed for relithiation of recycled cathode materials.
- Half-cell results also matched expectations.
- Impedance was also unchanged.





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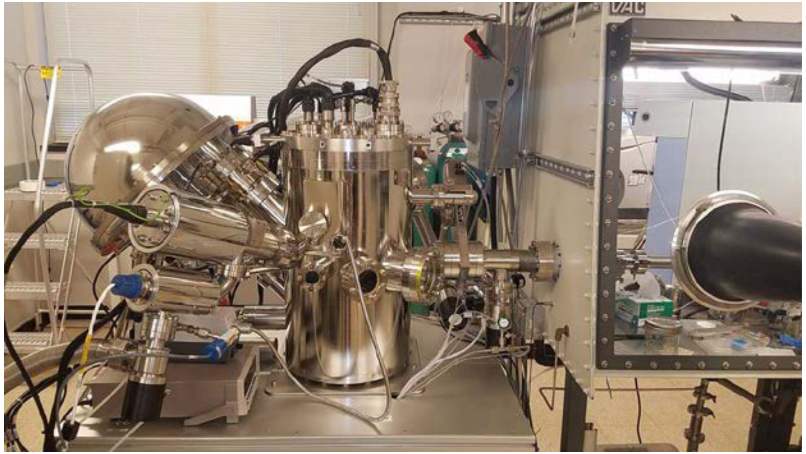
POST TEST FACILITY SUPPORT (ANL)

- Post-test diagnostics of aged batteries provide additional information regarding the cause of performance degradation, which, previously, could be only inferred
- The facility combines microscopy, spectroscopy and chromatography in a controlled-atmosphere glovebox to characterize materials without air exposure.
- This effort helps identify issues in recycled materials (e.g., how well a given recycling process separates an initial mixture of cathode, anode, supporting foils and casing materials.



Post-test diagnostics toolset:

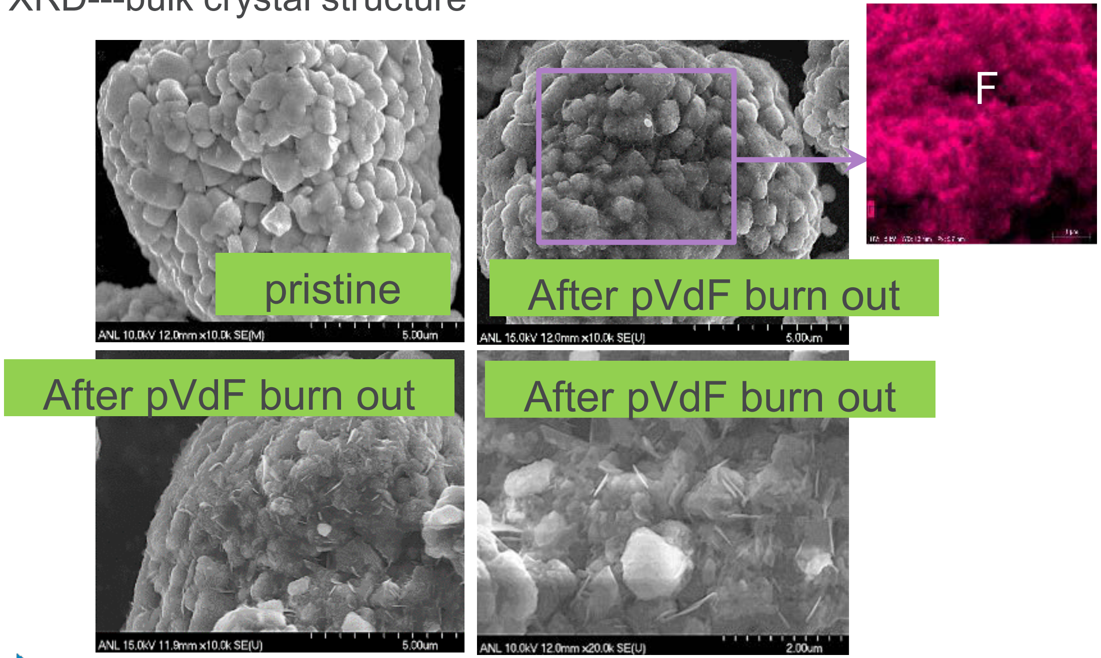
—FT-IR spectroscopy
—Raman spectroscopy
—Optical and scanning-electron microscopy
—Electrochemical impedance spectroscopy
—X-ray photoelectron spectroscopy
—High Pressure Liquid Chromatography
—Gel Permeation Chromatography
—TGA-GC/MS
—Half-cell fabrication and test equipment

CHARACTERIZING CHANGES INDUCED BY RECOVERY PROCESS

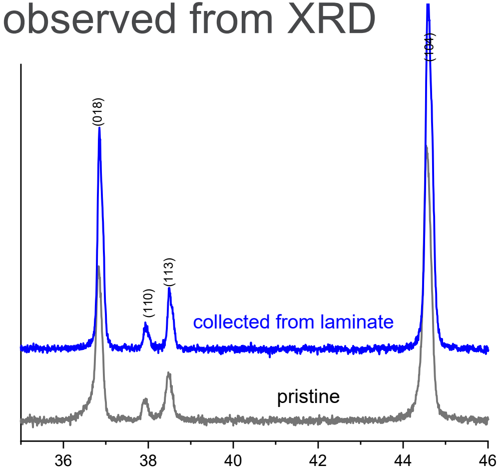
Accomplishments – Effect of Binder Burnout

Comparison of pristine vs recycled materials:

- SEM/EDX---particle morphology and rough size; local composition
- XRD---bulk crystal structure



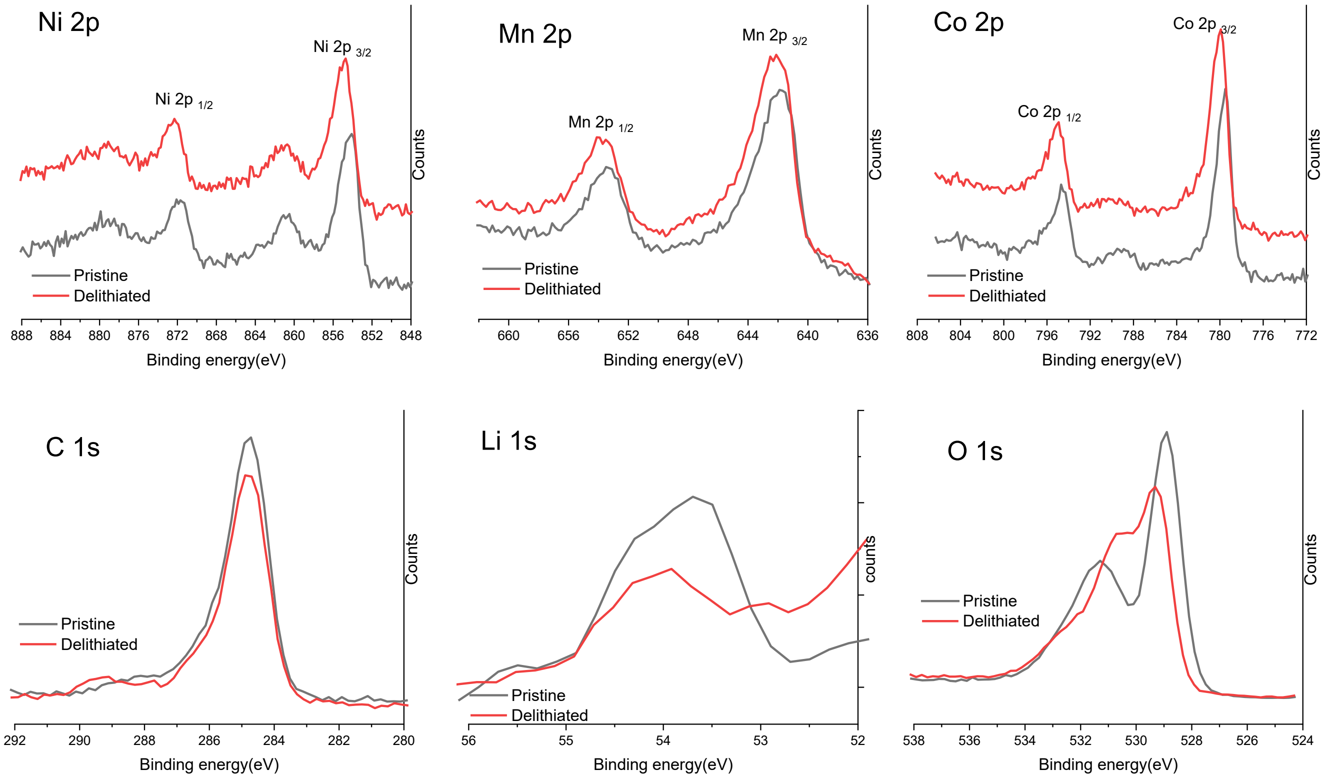
- SEM analysis reveals the surface of the particles was covered by thin platelets after burning out the binder.
- EDX results reveal that the platelets rich in fluorine.
- No significant changes observed from XRD



CHARACTERIZING CHANGES INDUCED BY RECOVERY PROCESS

Accomplishments: XPS Analysis of Chemically Delithiated Cathodes

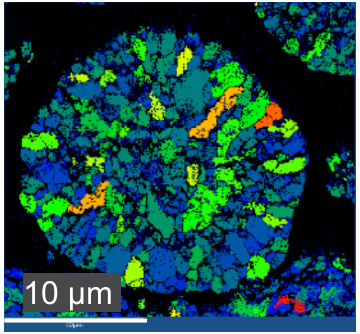
- TM shifts to a little higher binding energy and wider peaks for chemically delithiated cathode.
- This indicates slightly larger percentage of higher oxidation states of Ni, Mn and Co existing on the powder surface after chemical de-lithiation, which also revealed by clear changes in the surface oxygen.



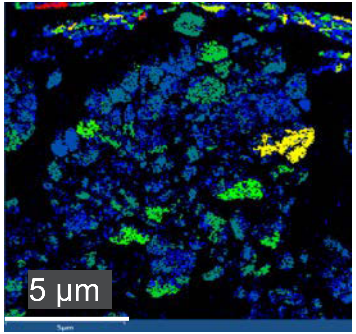
END OF LIFE MATERIALS DIAGNOSTICS (NREL)

Accomplishments: *Developing new characterization techniques*

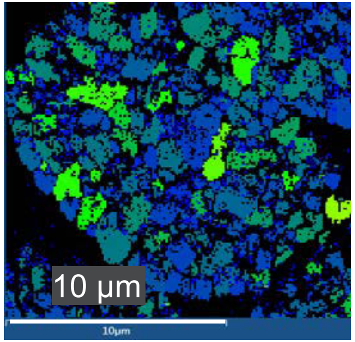
Electron Back Scatter Diffraction (EBSD)



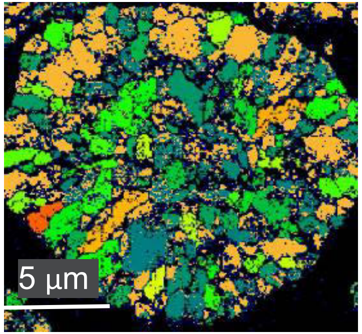
Baseline NMC-111



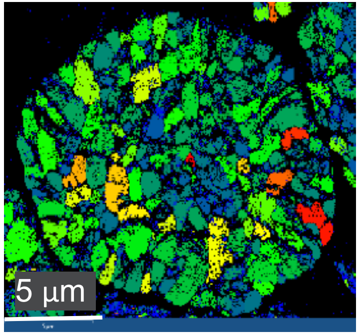
Fresh



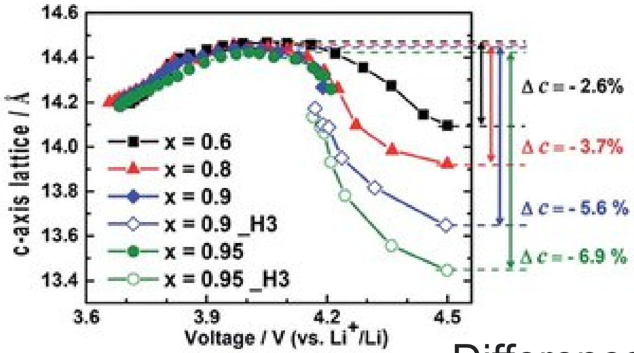
20% Capacity Loss



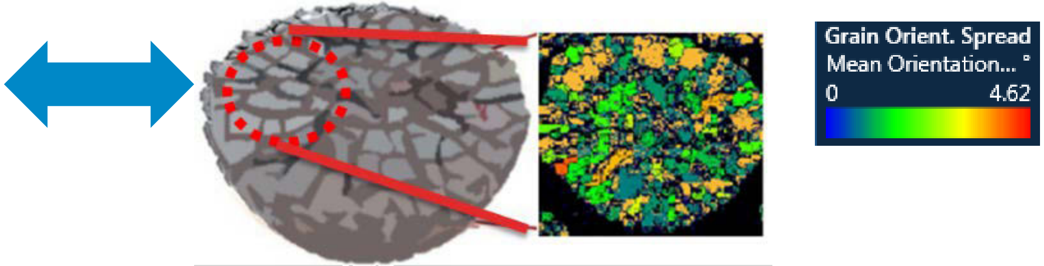
40% Capacity Loss



Chemically Delithiated



Chemical Vs Electrochemical Delithiation



Differences in mean orientation within the primary particles (see color scale) are a measure of difficulty with re-lithiation

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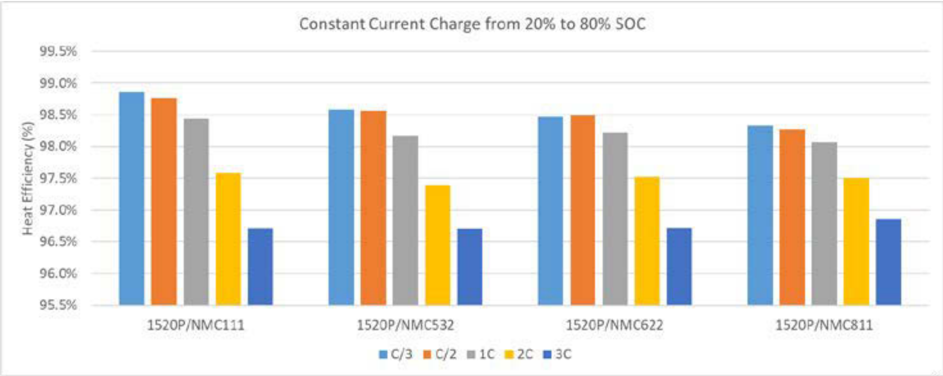
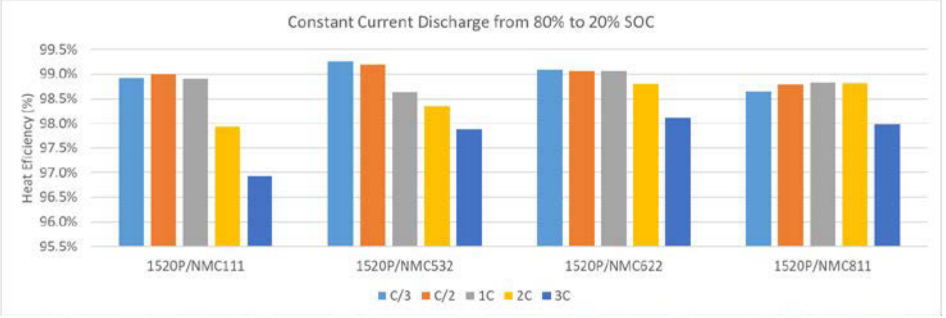
THERMAL CHARACTERIZATION (NREL)

Accomplishment: Quantified heat generation of graphite/NMC at the beginning of life. Data will be used to assess material quality from future recycling techniques.

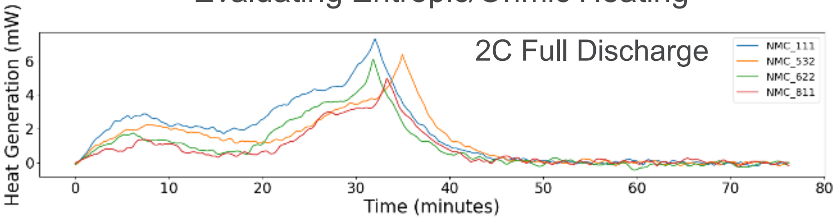
1520P/NMC – Partial Charge/Discharge

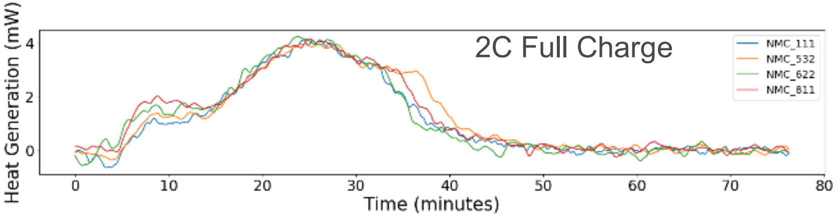




Cathode Material	Anode Material
NMC – 111	1506T
NMC – 532	1506T
NMC – 622	1506T
NMC – 811	1506T
NMC – 111	1520P
NMC – 532	1520P
NMC – 622	1520P
NMC – 811	1520P

Evaluating Entropic/Ohmic Heating





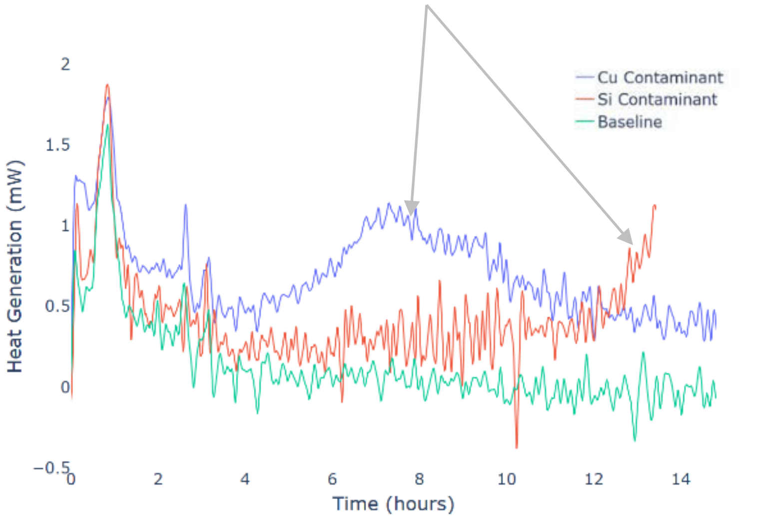
THERMAL CHARACTERIZATION (NREL)

Accomplishment: Calorimetry used to identify contaminants unintentionally introduced into batteries from recycling processes.

Cells with known contaminants tested in calorimeter.

Cell	Cathode Material	Cathode Impurity	Anode Material	Anode Impurity
Baseline (CFF-B41A)	NMC111 (90%)	NA	Graphite 1520P (91.83%)	NA
LN3166-066	NMC111 (89%)	Cu 1%	Graphite 1520P (91.83%)	NA
LN3166-067	NMC111 (89%)	Fe 1%	Graphite 1520P (91.83%)	NA
LN3166-068	NMC111 (89%)	Al 1%	Graphite 1520P (91.83%)	NA
LN3166-069	NMC111 (89%)	Mg 1%	Graphite 1520P (91.83%)	NA
LN3166-070	NMC111 (89%)	Si 1%	Graphite 1520P (91.83%)	NA
LN3166-071	NMC111 (90%)	NA	Graphite 1520P (90.83%)	Fe 1%
LN3166-072	NMC111 (90%)	NA	Graphite 1520P (90.83%)	Al 1%
LN3166-073	NMC111 (90%)	NA	Graphite 1520P (90.83%)	Mg 1%
LN3166-074	NMC111 (90%)	NA	Graphite 1520P (90.83%)	Cu 1%

When atypical materials (contaminants) are introduced to the cathode, the heat signature during formation can be used to identify the contaminant. This technology can be used to assess the efficacy of the recycling processes under ReCell.



1% of known contaminant added to cathode slurry

SUMMARY

- The CAMP Facility actively supports the ReCell Center by fabricating and testing numerous coin cells and providing baseline materials and single-layer pouch cells.
- We have established a thorough test matrix and protocols to evaluate baseline active materials of interest to the ReCell Center in FY 2019-2020.
- These protocols and the testing results are uploaded to the ReCell Center website as they become available (from FY 2020 2nd Quarter) for the general public's use.
- Extensive characterization and quality screening for end-of-life cathodes is performed under the crosscutting efforts to support the individual projects.
- Newer diagnostic tools examine degradation parameters beyond surface area, particle size, averaged XRD parameters help compare cathodes from different recycling streams.
- Together with the heat signatures, these diagnostic tools will provide confidence in re-introducing these materials into new cells.
- For more information please see www.recellcenter.org, where our Quarterly Reports are posted.

FUTURE WORK

- CAMP will continue to support the different projects under ReCell by providing cells, materials and data to the technoeconomic analysis tasks.
- Proposed future work for Post Test includes characterizing recycled materials from the different recovery processes. As the recycling processes mature, cells will be constructed from the materials and materials will be sent to Post-Test for additional characterization data to identify factors that influence the loss of performance.
- Planned work for end of life diagnostics include quantifying the relationship between spread in mean grain orientation and lattice strains, relate these to heat-treatment or other pre-processing steps are required before relithiation.
- The thermal task will benchmark heat signatures for different NMCs under different cycling conditions and develop mechanistic relations between the thermal cycle inefficiencies and structural breakdown of the cathode material.
- Calorimeter technology will be used to identify contaminants in cells which will be used to assess the efficacy of the recycling processes developed under ReCell.

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RECELL RECYCLING TOWN HALL

FRIDAY, JUNE 5, 2020 FROM 1:00 TO 3:00 (CENTRAL)

To continue the discussion the ReCell team will hold an interactive town hall meeting.

Please join us at the BlueJeans session shown below and ask questions through Slido

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https://bluejeans.com/749203749/9534?src=htmlEmail

Phone
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Meeting ID: 749 203 749

Slido Q/A website

Computer or Smart Phone
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Event Code
“recell”

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