Li-Ion Cell Manufacturing Using Directly Recycled Active Materials

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DOE VTO Annual Merit Review

Project ID: bat356

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Overview

Timeline

- Start: February 2017
- End: January 2019
- NCE: to Oct 2019
- Percent complete: 66 %

Barriers

- Cost/value recovery of current battery recycling processes
- Quality of feedstock for direct recycling
- Complexity and variability of LiB designs

Budget

- \$1.8M total project budget:
  - \$900k DOE
  - \$900k Farasis
- 50 % Cost share

Partners

- Farasis Energy, Inc
- Lawrence Berkeley National Laboratory – R. Kostecki group

Relevance / Objectives

- **Project Goal:** The goal of this project is to develop recycling technology for Li-ion batteries that will enable direct reuse of valuable active materials.
- **Performance Objective:** The objective is to demonstrate the utility of direct recycling technology by producing cells with recycled active materials that have performance within 5% of control cells using pristine versions of the same active materials.
  - Optimized recovered materials and formulations will be used to manufacture large pouch cells (25 Ah) as project deliverables.

Project Milestones

Tasks	Milestone		Project Month	Status
Task 1	1.3.1	Final Report summarizing initial electrochemical testing	24	*Delayed*
Task 2	2.1.1	Acquisition of direct recycling process equipment	3	**Complete**
	2.2.1	Completed installation of direct recycling pilot line	5	**Complete**
	2.3.1	Recovery of 2 kg Positive AM & 1 kg Negative AM from manufacturing residues	8	**Complete**
	2.3.2	Recovery of 2 kg Positive AM & 1 kg Negative AM from EOL cells	14	*50 % Complete*
	2.3.3	Recovery of 2 kg Positive AM & 1 kg Negative AM from EV battery modules	17	*Delayed*
Task 3	3.1.1	Demonstrate density based separation at a scale of 5 kg black mass input	10	*Delayed*
	3.1.2	Improve separation yield to >95%	17	*Delayed*
	3.1.3	Recover direct recycled active materials in greater than 99.9% purity.	17	**Complete**
	3.2.1	Demonstrate recovered active materials with specific capacities and first cycle efficiencies identical to pristine materials	17	**Complete**
	3.2.2	Assessment of economic impact of surface area reduction processing	18	**Complete**
	3.2.3	Report on detailed materials characterization of recycled active materials.	20	*Delayed*
Task 4	4.1.1	Demonstrate separation of mixed spinel/layered oxide cathode material mixtures using density-based separation.	19	*Delayed*
	4.2.1	Demonstrate reconditioning of mixed spinel/layered oxide cathode material mixtures	19	*Delayed*
Task 5	5.1.1	Completion of Cell Build 1	12	*Delayed*
	5.2.1	Completion of Cell Build 2	16	*Delayed*
	5.3.1	Completion of Deliverable Cell Build	21	*Delayed*
	5.3.2	Delivery of controls and cells with > 50% recycled active material content.	22	*Delayed*
Task 6	6.1.1	Delivery of initial test data	24	*Delayed*
	6.2.1	Quantification of impact of recycled active materials on technology lifetime and cost	24	*Delayed*

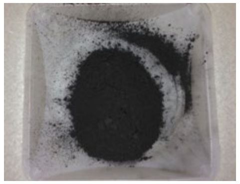
Approach – Direct Recycling Process Overview

Discharged cells

Shredding

Electrolyte extraction

Sieving



Black Mass

Density Separation

Graphite

$LiMO_x$

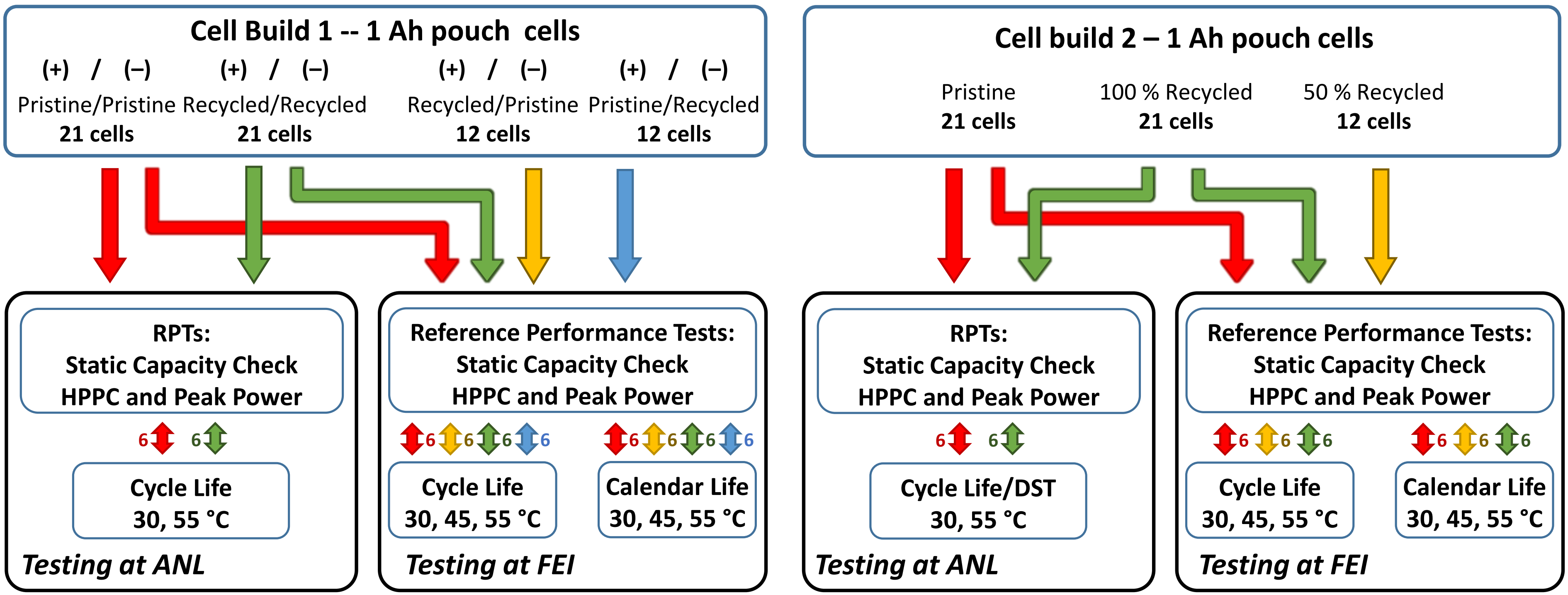
Purification

Regen-eration

Recycled Materials

- Direct recycling uses only physical separation processes
- Active materials are recovered essentially intact, thus capturing some of the value added during original material synthesis
- Chemical purification and re-lithiation are performed under relatively mild conditions with low energy intensity

Approach – Cell Builds



- Intermediate cell build 1 examines different combinations of recycled and pristine active material electrodes to test for cell performance sensitivity to either recycled active material.
- Intermediate cell build 2 provides additional data to optimize blending of recycled and pristine active materials for the final deliverables.

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Approach - Complexity

Recycling Feedstocks

This project evaluates multiple possible inputs for direct recycling:

- **Electrode production scrap**
- **Formed cells**
- **Entire battery modules**

Cell Chemistries

- **First large-scale feedstock for commercial scale recycling NCM111 is the main focus for process development and deliverables**
- **Additional process development is being performed for more complex mixed active material cathodes: LMO+NCM**
- **Other cathode chemistries (NCM523, LFP, …) will be evaluated for compatibility with optimized processes**

Technical Accomplishments- Electrode Scrap Recovery Process

Electrode / Material → *shred* → Scraps → *thermal1* → Activated Scraps → *classify* → NMC$_{crude}$ → *thermal2* → rNMC

classify ↓ Metal foils

1. Electrode scrap and dried out slurry residue are milled to reduce scrap to coarse shreds.

2. The scrap is heated at moderate temperature to decompose the binder. The fine powders are separated from the current collector scrap using vibratory screen or air classifier.

3. The crude active material is fired at higher temperature to remove residues and improve powder characteristics.

- The above process has been optimized and applied to recover graphite, NMC111, and NMC523 in kg quantities.

Technical Accomplishments- Recycled NMC111 Properties

Positive Active Material Properties Gap Chart Electrode Scrap Feedstock				
Characteristic	**Units**	**QC Spec**	**Virgin**	**100% Recycled**
Particle size (D_{50})	μm	9 - 14	11	12
Tap Density	(g/cm^3)	≥ 2.0	2.5	2.3
Reversible capacity (4.2 - 3.0 V vs. Li/Li+, 0.1 C)	(mAh/g)	145	150	145
Specific Surface Area (BET method)	(m^2/g)	0.15 - 0.55	0.23	0.28
First cycle efficiency	%	≥ 88	91	88
Impurities*	%w/w	Na < 0.08 Mg < 0.02 Ca < 0.02 Fe < 0.012 Cu < 0.005 Al: *no spec* F: *no spec*	0.11 < Na < 1 Mg < 0.026 Ca < 0.07 Fe = 0.022 Cu < 0.0014 Al < 0.072 F < 0.001	0.11 < Na < 1 Mg < 0.026 Ca < 0.07 Fe = 0.1 Cu < 0.0014 Al < 0.072 F = 0.85
pH assay	-log [H^+]	10.7 – 11.7	11.0	11.1

Technical Accomplishments- Recycled Graphite Properties

Negative Active Material Properties Gap Chart Electrode Scrap Feedstock				
Characteristic	**Units**	**QC Spec**	**Virgin**	**100% Recycled**
Particle size (D_{50})	mm	15	15	14
Tap Density	(g/cm^3)	≥ 1.0	1.1	0.97
Reversible capacity	(mAh/g)	≥ 306	322	315
First Cycle Efficiency	%	≥ 90	89	88
Specific Surface Area	(m^2/g)	1.0-1.6	1.3	1.6
Ash content	%w/w	≤ 0.05	< 0.1	0.6

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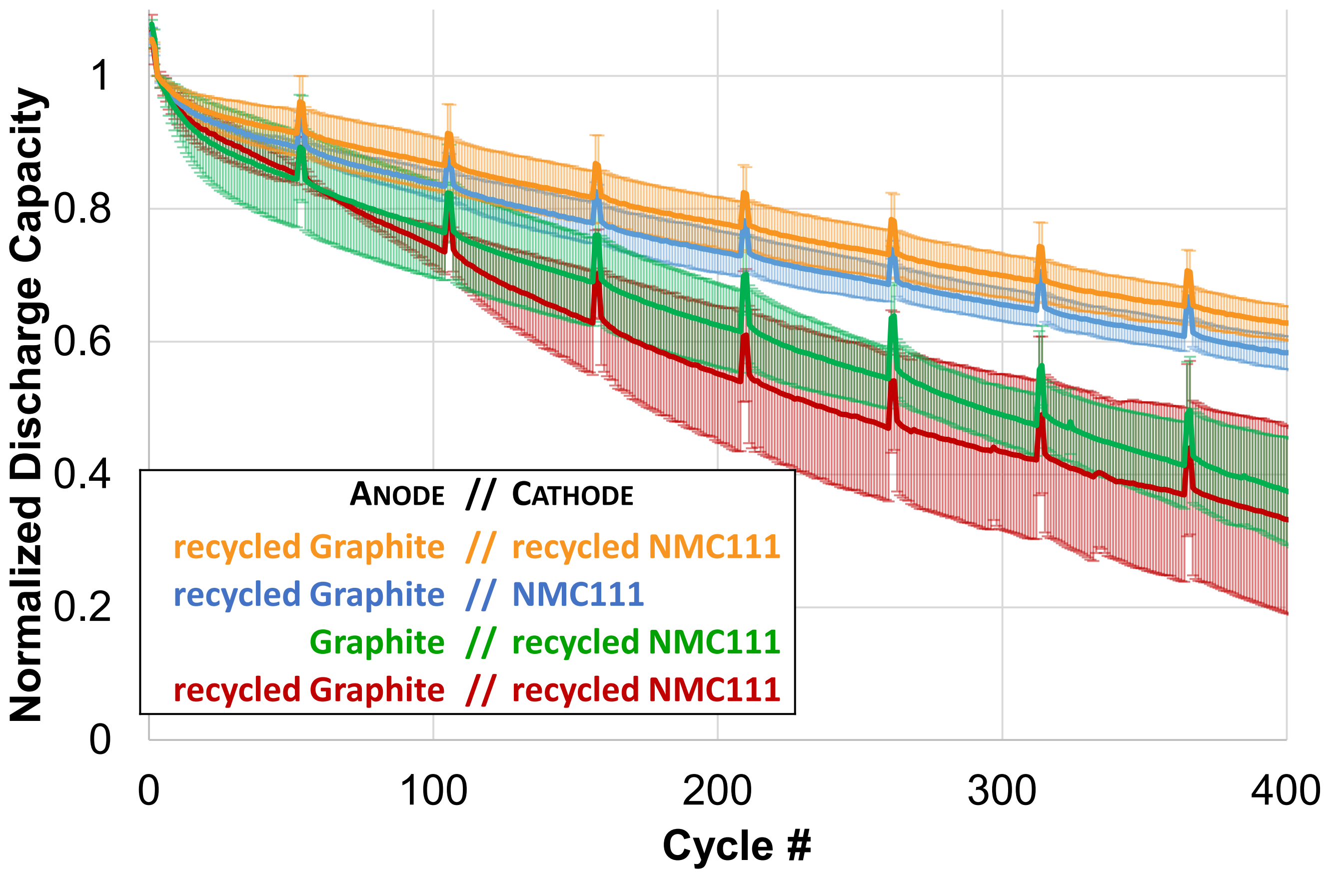
Technical Accomplishments- Recycled/Pristine Materials Comparison



Testing in *ca.* 8 mAh coin cells: 2.75 V - 4.2 V; C/8 charge, C/4 discharge, cycle at C/20 every 50 cycles.

- In each case and in combination, cells with recycled active materials have better cycle life capacity retention and lower rate of impedance growth than the pristine versions.
- Opportunistic material engineering during recycling may be responsible for improved performance characteristics.

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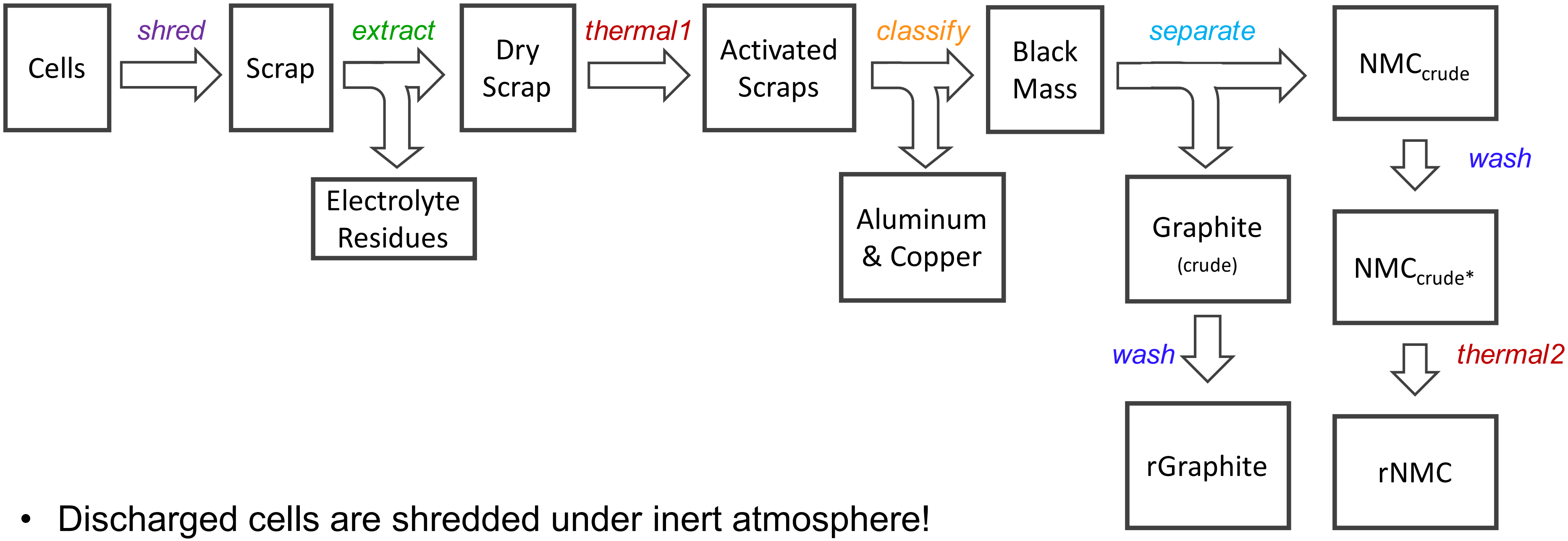
Technical Progress- Processing Whole Cell Feedstock



- Discharged cells are shredded under inert atmosphere!
- Unoptimized recovery yield of rNMC is about 50%.
- Graphite is currently not being recovered from this process.

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Technical Accomplishments- Recycled NMC111 Properties

Positive Active Material Properties Gap Chart
Whole Cell Feedstock

Characteristic	Units	QC Spec	Virgin	Recovered from cells
Particle size (D_{50})	µm	9 - 14	11	12
Tap Density	(g/cm^3)	≥ 2.0	2.5	2.2
Reversible capacity (4.2 - 3.0 V vs. Li/Li+, 0.1 C)	(mAh/g)	145	150	120
Specific Surface Area (BET method)	(m^2/g)	0.15 - 0.55	0.23	0.66
First cycle efficiency	%	≥ 88	91	86
Impurities*	%w/w	Na < 0.08 Mg < 0.02 Ca < 0.02 Fe < 0.012 Cu < 0.005 Al: *no spec* F: *no spec*	0.11 < Na < 1 Mg < 0.026 Ca < 0.07 Fe = 0.022 Cu < 0.0014 Al < 0.072 F < 0.001	*Metals quantification pending* F = 1.6
pH assay	-log [H^+]	10.7 – 11.7	11.0	12.0

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Responses to Reviewers' Comments

- How to ensure the fully restored Li inventory in each different NMC resource. Residual Li in NMC could vary significantly from each different recycled battery.
  - Elemental analysis of feedstock composition, in conjunction with knowledge from the battery OEM, will be needed to determine proper Li addition levels. Currently, we use the above approaches in addition to refinement of Li reagent stoichiometry as a process variable.
- …trade-offs between anode thermal treatment temperature effects on Gr oxidation and removing SEI components. The requirements of Gr yield and SEI layer removal seem to be in competition with each other.
  - This can be addressed by controlling the atmosphere in the furnace which will alter the ratio of thermal depolymerization (SEI-breakdown) vs. combustion reaction (nanocarbon and graphite oxidation).
- More collaborators than just LBNL will likely be needed for the extensive materials characterization that will be needed to address the material complexity issues…
  - We have run up against this limitation as there is always the need for more characterization! We have added some additional characterization techniques in-house and are leveraging capabilities at other institutions via informal collaborations.

Collaborations

- **Lawrence Berkeley National Laboratory (Robert Kostecki)**

  Advanced chemical diagnostics and materials characterization to guide recycling process development.

Challenges and Barriers

- **Feedstock complexity leading to unexpected interactions during active material separations in dense liquid media.**
- **Cell manufacturing lead times at our pilot facility are unexpectedly long.**

Proposed Future Research

- **Extend direct recycling process to additional feedstocks –complete modules**
- **Complete deliverable cell builds and testing to fully quantify impact of recycled materials on technology lifetime**
- **Evaluate process compatibility with other cell chemistries**

Summary

- A recycling process for direct recovery and reuse of Li-ion battery active materials has been scaled up and is being applied to multiple commercially-relevant feedstocks.
- Kilogram-scale recovery from electrode scrap feedstocks is complete and produced materials with performance characteristics very similar to pristine materials.
- Process refinement and scale-up for formed cell and EOL-module feedstocks is in progress.