Lithium-Ion Cell Manufacturing Using Directly Recycled Active Materials

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DOE VTO Annual Merit Review

Project ID: BAT356

Overview

Timeline

- Start: February 2017
- End: January 2019
- No Cost Extension: to June 2021
- Percent complete: 100%

Barriers

- Recycling and Sustainability
  - Recycling Li-ion batteries is currently expensive, representing 5-15% of the total technology cost.
  - Critical material availability (Co, Li, Ni, graphite).
  - Variable backend value for diverse battery designs in the market today.

Budget

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

Partners

- Project Lead: 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 high-value 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.
- **Relevance:**
  Optimized recycling processes decrease energy storage technology lifetime cost and maintain availability of critical materials. Experience with implementing recycled active materials informs future battery designs to improve recycling process efficiency.

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	**Complete**
	2.3.3	Recovery of 2 kg Positive AM & 1 kg Negative AM from EV battery modules	17	**Complete**
Task 3	3.1.1	Demonstrate density based separation at a scale of 5 kg black mass input	10	**Complete**
	3.1.2	Improve separation yield to >95%	17	**Complete**
	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	**Complete**
Task 4	4.1.1	Demonstrate separation of mixed spinel/layered oxide cathode material mixtures using density-based separation.	19	**Complete**
	4.2.1	Demonstrate reconditioning of mixed spinel/layered oxide cathode material mixtures	19	**Complete**
Task 5	5.1.1	Completion of Cell Build 1	12	**Complete**
	5.2.1	Completion of Cell Build 2	16	**Complete**
	5.3.1	Completion of Deliverable Cell Build	21	**Complete**
	5.3.2	Delivery of controls and cells with > 20% recycled active material content.	22	**Complete**
Task 6	6.1.1	Delivery of initial test data	24	**Complete**
	6.2.1	Quantification of impact of recycled active materials on technology lifetime and cost	24	**Complete**

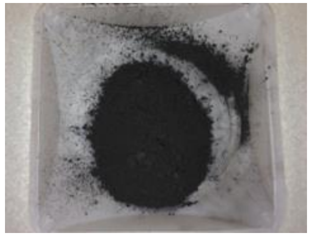
Approach – Direct Recycling Process Overview

Discharged cells

Shredding

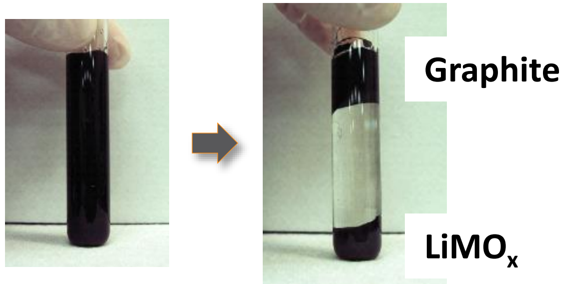
Electrolyte extraction

Sieving



Black Mass

Density Separation



Purification

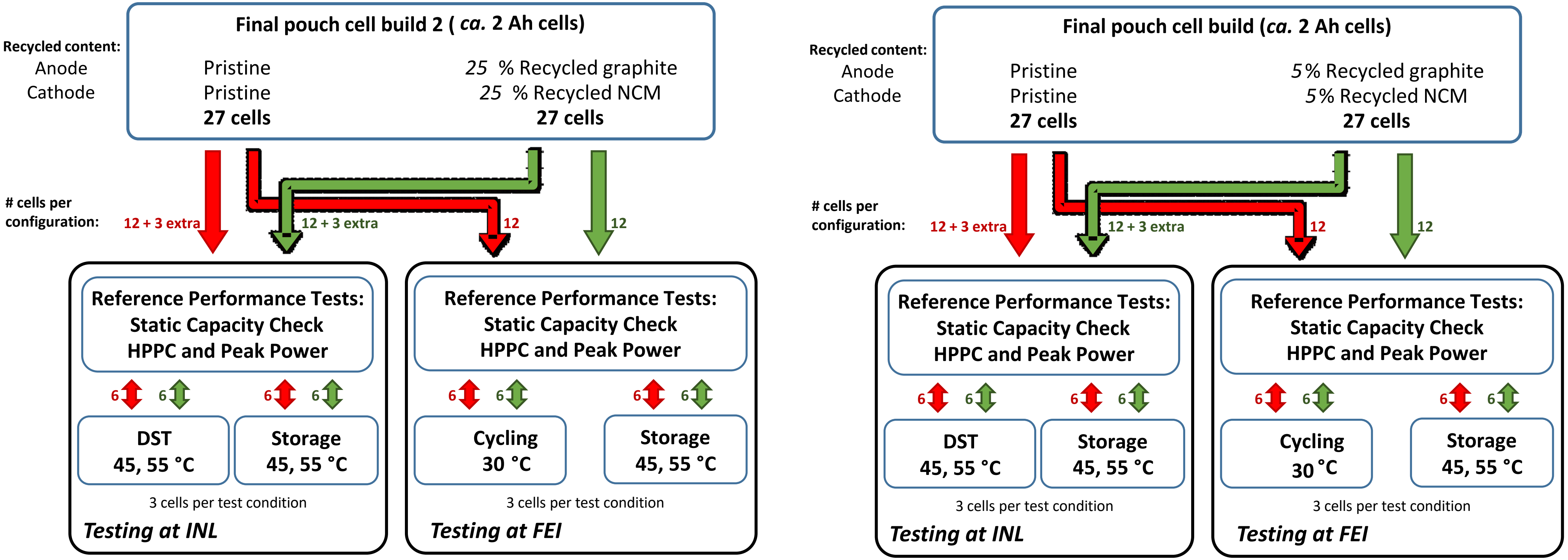
Regen-eration

Recycled Materials

Direct Recycling Approach Highlights

- Direct recycling uses minimal processing to recover materials and restore them for reuse.
- 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 2 is designed to examine the effects of blending higher recycled active materials with their pristine counterparts.
- The final, deliverable cell build will implement likely industry use cases of recycled active materials with a focus on minimizing performance differences of cells using recycled active materials.

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Approach - Complexity

Recycling Feedstocks

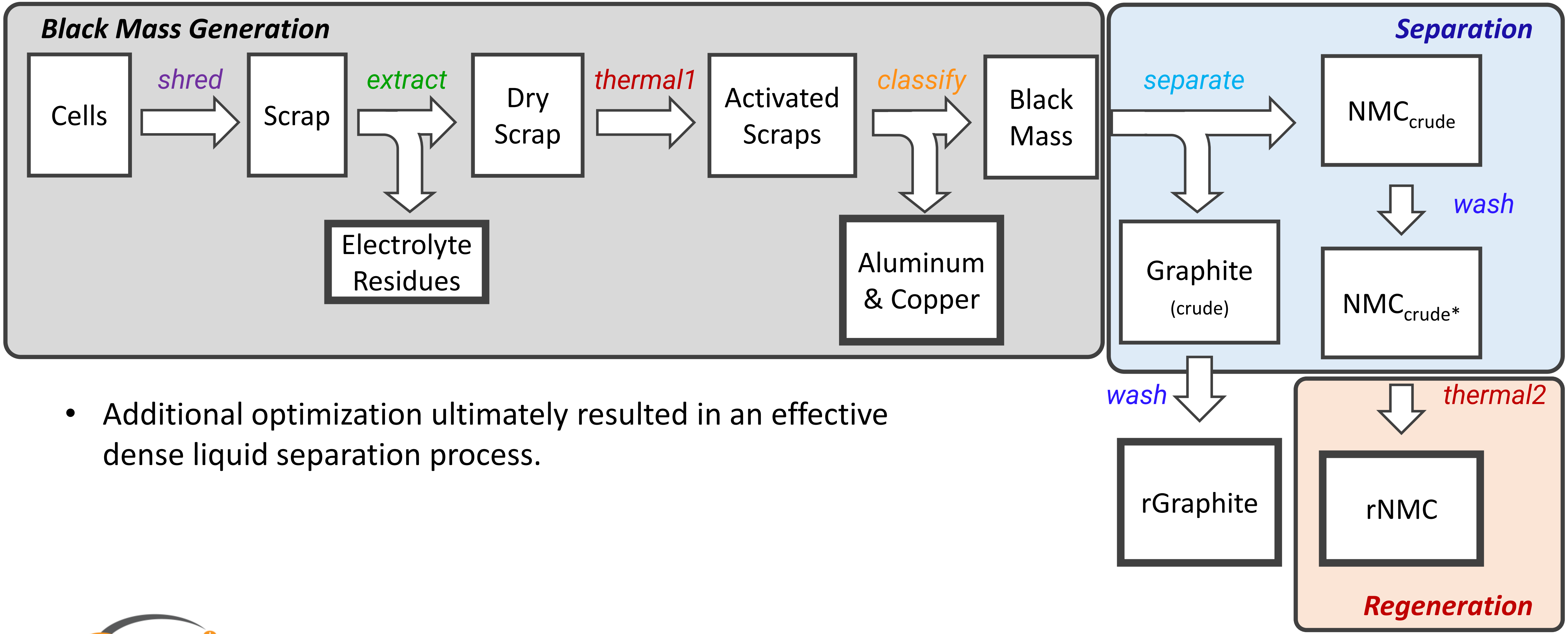
This project evaluates multiple possible inputs for direct recycling:

- Electrode production scrap
- Formed cells

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, *e.g.* LMO+NCM**

Technical Accomplishment – Recovery Scale-Up
Modified Process for Whole-Cell Feedstock



- Additional optimization ultimately resulted in an effective dense liquid separation process.

Technical Accomplishment – Recovery Scale-Up Whole Cell Feedstock Recycled NMC111 Properties

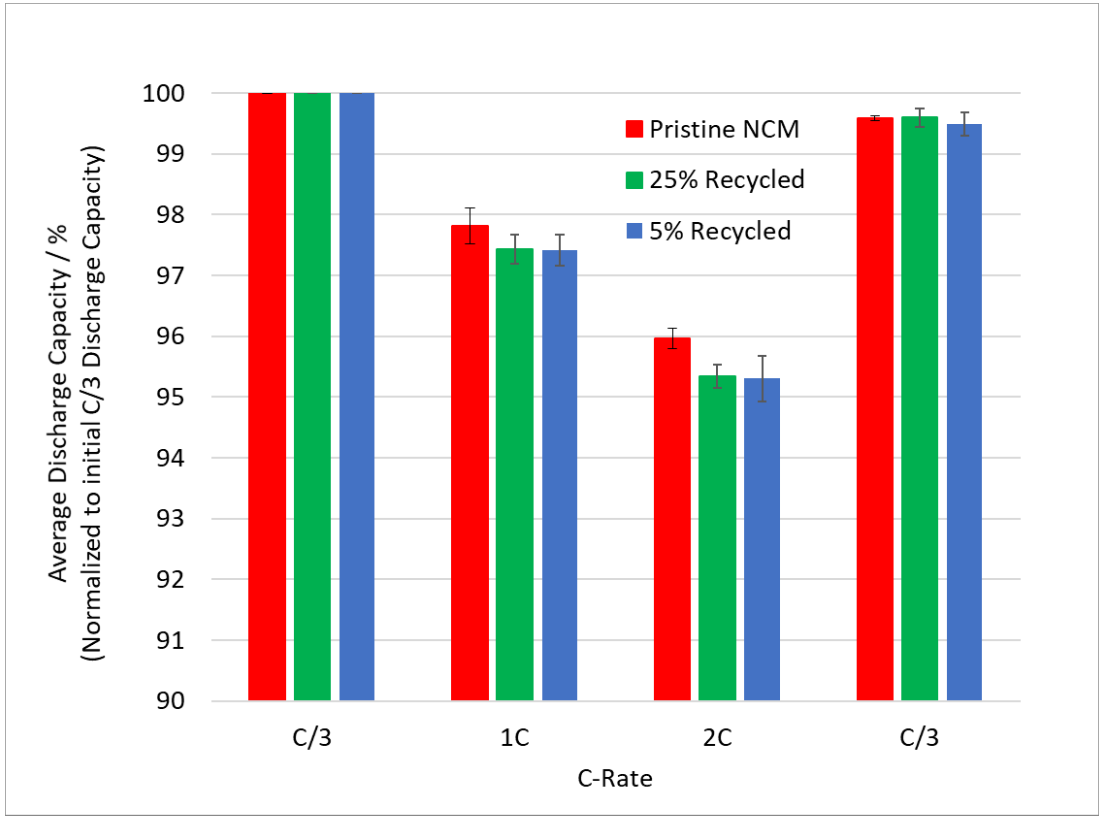
Positive Active Material Properties Gap Chart					
Characteristic	**Units**	**QC Spec**	**Pristine**	**Cell Build 2 Blend (25% Recycled)**	**Cell Build 3 Blend (5% Recycled)**
Particle size (D_{50})	um	9 - 14	11	6.6	8.3
Tap Density	(g/cm^3)	≥ 2.0	2.5	2.3	2.4
Reversible Capacity (4.2 - 3.0 V vs. Li/Li+, 0.1 C)	(mAh/g)	145	150	149	149
Specific Surface Area (BET method)	(m^2/g)	0.15 - 0.55	0.23	0.41	0.24
First cycle efficiency	%	≥ 88	91	90	89
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 < 0.05 W < 0.05 **Cu = 0.36** Al < 0.072 **F = 0.12**	Na < 0.05 Mg < 0.05 Ca < 0.05 Fe < 0.05 W = 0.009 **Cu = 0.42** Al < 0.05 **F = 0.04**
pH assay	$-\log [H^+]$	10.7 – 11.7	11.0	11.2	11.2

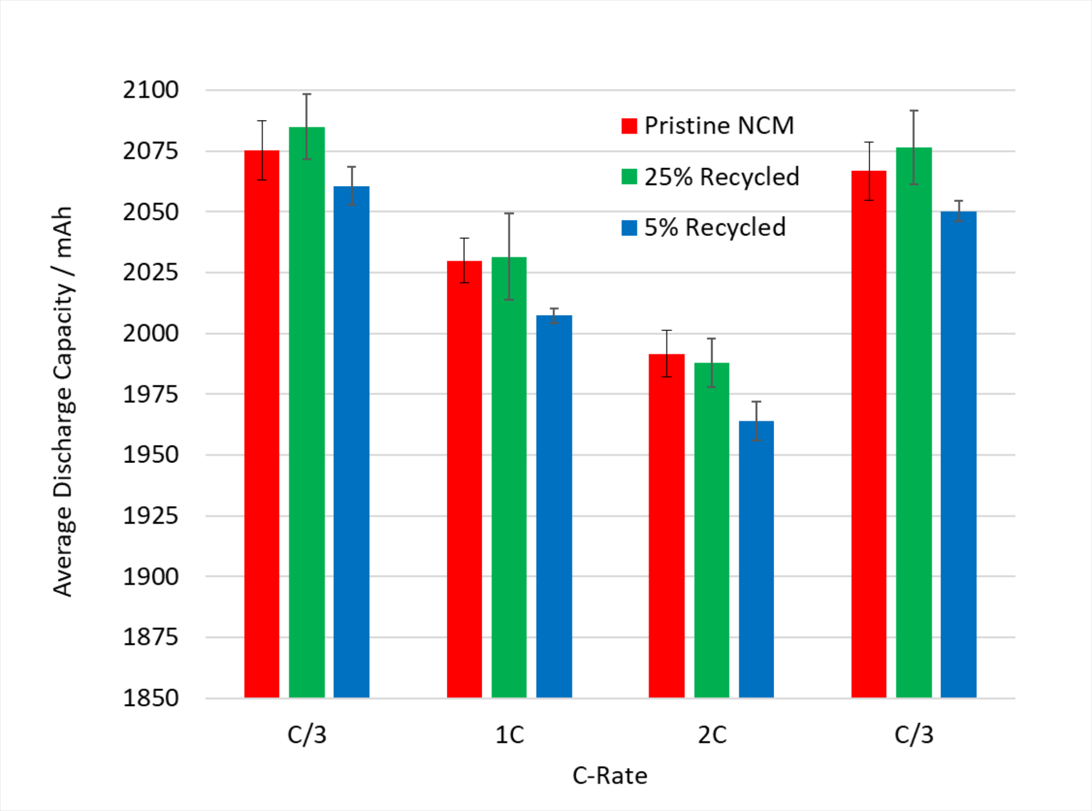
Technical Accomplishment – Cell Manufacturing Cell Builds 1, 2 & 3

- Cell build 1 was completed in July 2019 and delivered to National Labs
- Cell builds 2 and 3 were completed in March and delivered to National Lab
  - 80 Control cells (2.08 Ah, 100% pristine NCM111)
  - 40 Build 2 cells (2.07 Ah, 25% recycled NCM111 / 75% pristine NCM111)
  - 40 Build 3 cells (2.07 Ah, 5% recycled NCM111 / 95% pristine NCM111)
- BOL characterization has been done at Farasis
  - Rate capability at C/3, 1C, 2C rates
  - HPPC and DC-IR analysis

Technical Accomplishment – Cell Builds 2 and 3 BOL Characterization: Rate Capability

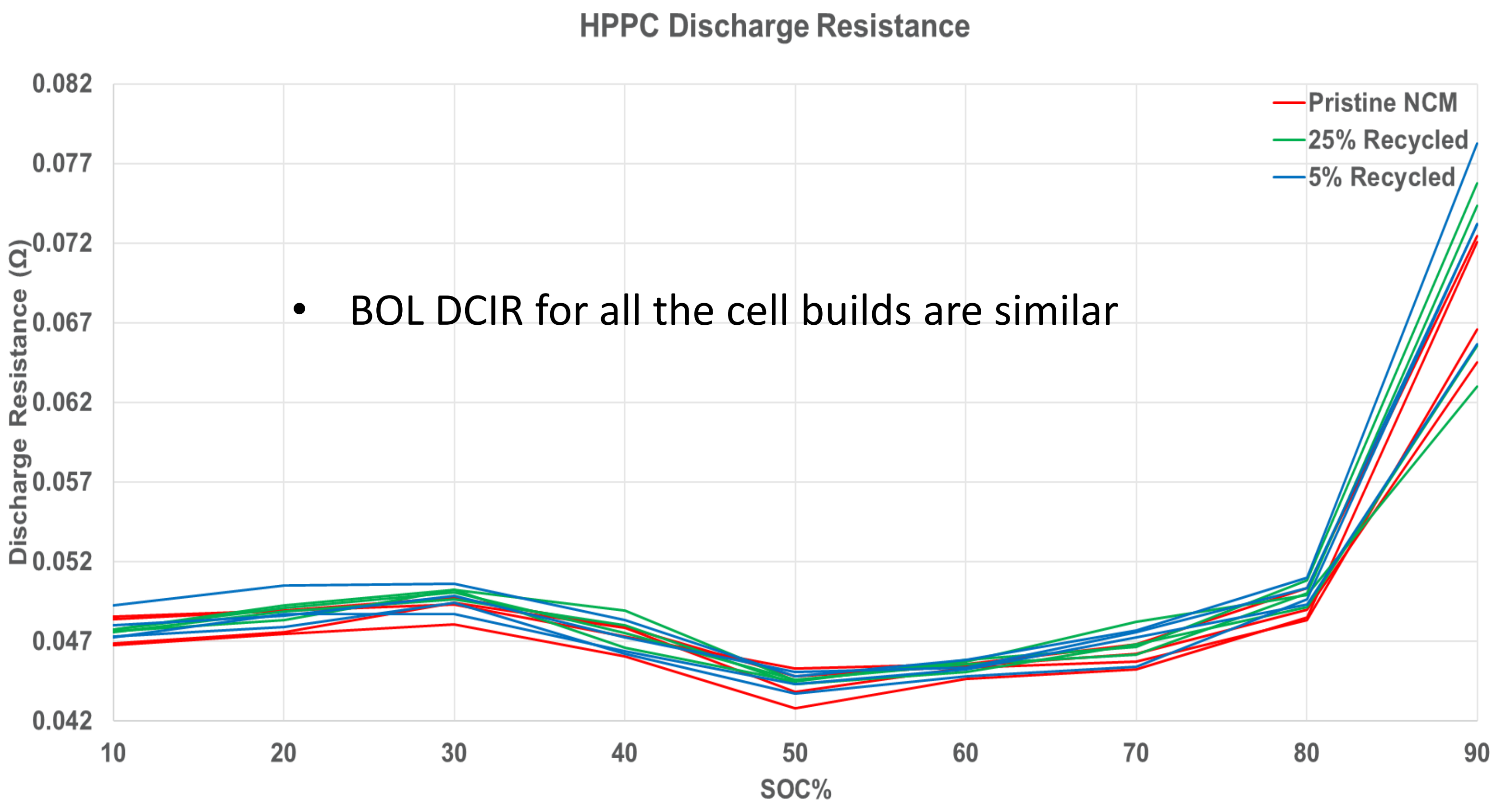
Test Conditions: 30°C, C/3 charge to 4.20 V, discharge to 2.75 V at C/3, 1C, 2C, and C/3 rates





Technical Accomplishment – Cell Builds 2 and 3 BOL Characterization: DCIR

Test Conditions: 30°C, C/3 charge to 4.20 V; discharge Vmin 2.75 V
C/3 discharge for 10% SOC, then 30s 2C pulse to calculate DC-IR



- BOL DCIR for all the cell builds are similar

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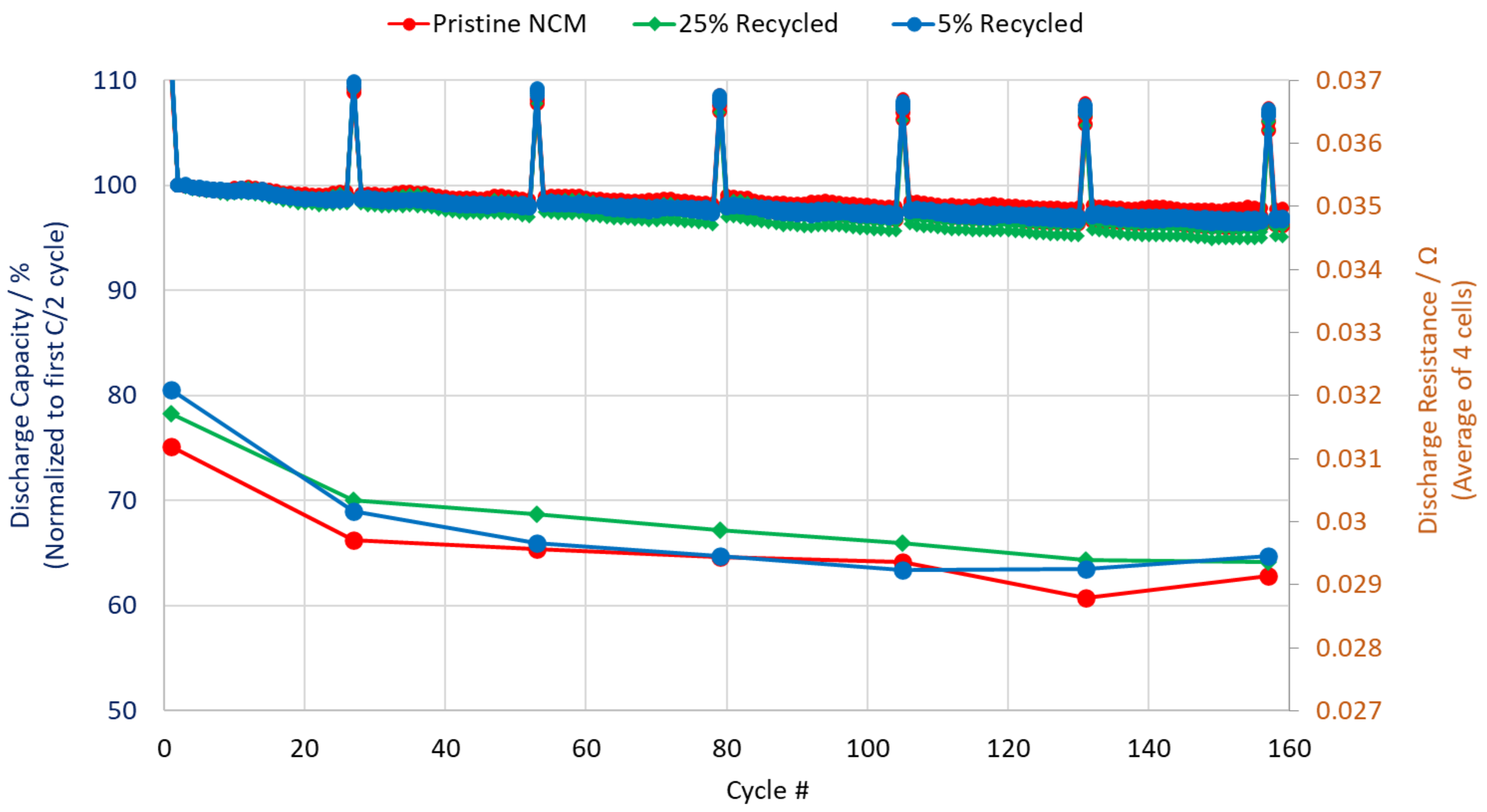
Technical Accomplishment – Cell Builds 2 and 3 BOL Characterization: Cycle Performance

Test Conditions: 30°C, C/2 charge to 4.20 V; C/2 discharge to 2.75 V
C/3 capacity check every 25 cycles with 30s 2C pulse at 50% SOC to measure DC-IR



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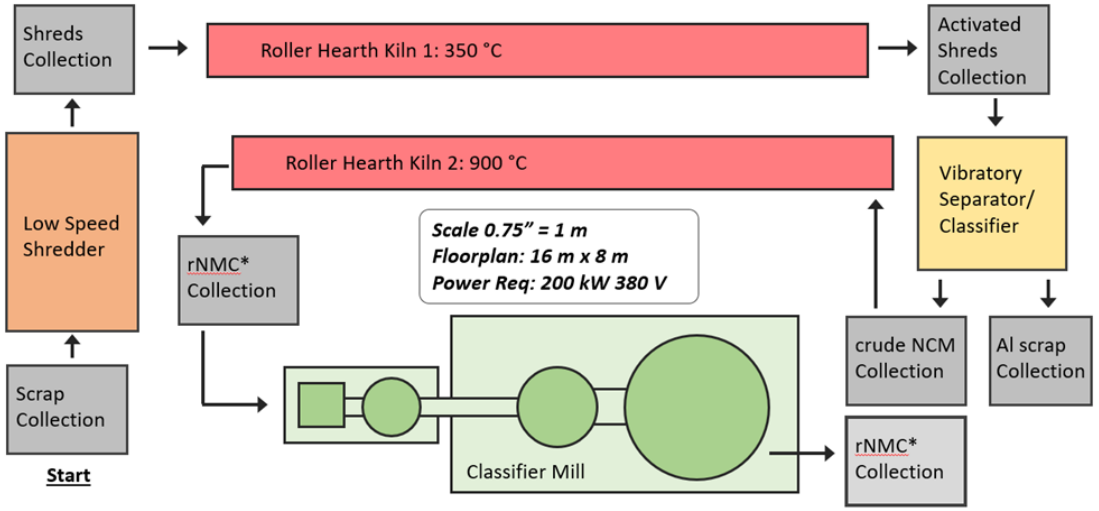
Recycling Cost Model Update
Example Implementation at 20 GWh Cell Manufacturing Facility

- Direct Recycling cost model has been updated with a focus on recovery of cathode electrode scrap materials from cell manufacturing processes
- Economic value was calculated using the following assumptions:
  - 20 GWh annual cell production rate
  - NCM811 pristine cathode cost: $26/kg
  - Cathode electrode scrap rate: 5%
  - Direct Recycling process efficiency: 90%
- Capital investment and operating costs are estimated to be relatively low, and at the 20 GWh scale the economic value of recycling cathode electrode scrap is significant



	China	US	EU
NCM Recovered (mt, annually)	1,324		
NCM Cost per metric ton (mt)	$ 26,000		
Capital Investment	$ 749,837	937,296	1,124,755
Annual Cost (Total, Amortized)	$ 551,186	1,872,614	2,478,932
Cost of Recovered NCM ($/mt)	$ 416	1,414	1,872
Cost of 4.5/95.5 NCM Blend (mt)	$ 24,849	24,894	24,914
Cathode Cost Reduction ($/mt)	$ 1,151	1,106	1,086
Battery Cost Reduction ($/kWh)	$ 1.69	1.63	1.60

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Responses to Reviewers' Comments

- "It was not completely clear to the reviewer if the process will be supporting the battery production facility, thus the feedstock will be relatively uniform, not obsoleted, or if the process t will be used for recycling end-of-life (EOL) spent batteries. In the latter case, the issue of high variability of the feedstock and obsolete chemistries should be expected."

Response: This project addresses multiple possible use cases for direct recycled active materials including both manufacturing scrap- and EOL- feedstocks. The ultimate goal is to identify the impact of feedstock quality/complexity on the properties of materials recovered using direct recycling processes and to use this information to guide future battery design to improve recyclability.

- "…recovering 1-2-kilogram (kg) quantities of the materials does not allow one to fully assess the method performance and quality of the output materials against the feedstock variability…"

Response: This project represents a scaling-up of processes that had only been previously executed at small scale; as direct recycling matures, it will be possible to move to larger scale. There are numerous issues related to feedstock stability and batch sampling that can only be effectively resolved at- or near-commercial scale.

Collaborations

- Lawrence Berkeley National Laboratory (Robert Kostecki)
  - Subcontractor for advanced chemical diagnostics and materials characterization to guide recycling process development.
  - Essential collaboration to fully understand interaction of recycled materials' structure with resulting device performance.

Challenges and Barriers

- Module-level feedstocks include additional components and materials that interact with recycling process differently than manufacturing scrap feedstocks.
- Difficult separations in the direct recycling process lead to low purity of recycled active materials recovered at kg-scale.

Summary

- Cell Build 1 manufacturing and testing is complete and generated data that will help quantify the impact of material purity on device performance.
- Cell Build 2 and 3 delivered to INL for the testing. Started the testing at Farasis.
- Cycle Life:
  - Pristine >5% recycled > 25% recycled