Lithium-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
- No Cost Extension: to June 2020
- Percent complete: 85 %

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	*15 % 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	*80 % 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	*10 % Complete*
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	*50 % Complete*
Task 5	5.1.1	Completion of Cell Build 1	12	**Complete**
	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	*33 % Complete*
	6.2.1	Quantification of impact of recycled active materials on technology lifetime and cost	24	*25 % Complete*

Approach – Direct Recycling Process Overview

Direct Recycling Approach Highlights

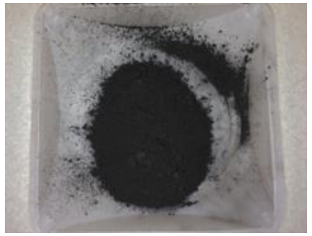
- Direct recycling uses minimal processing to recover materials and restore them for reuse.
- Active materials are recovered essentially intact, thus capturing some of the value added during original material synthesis
- Chemical purification and re-lithiation are performed under relatively mild conditions with low energy intensity

Discharged cells

Shredding

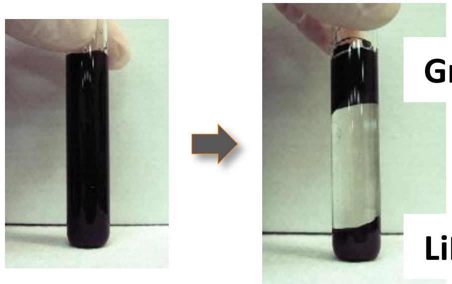
Electrolyte extraction

Sieving



Black Mass

Density Separation



Graphite

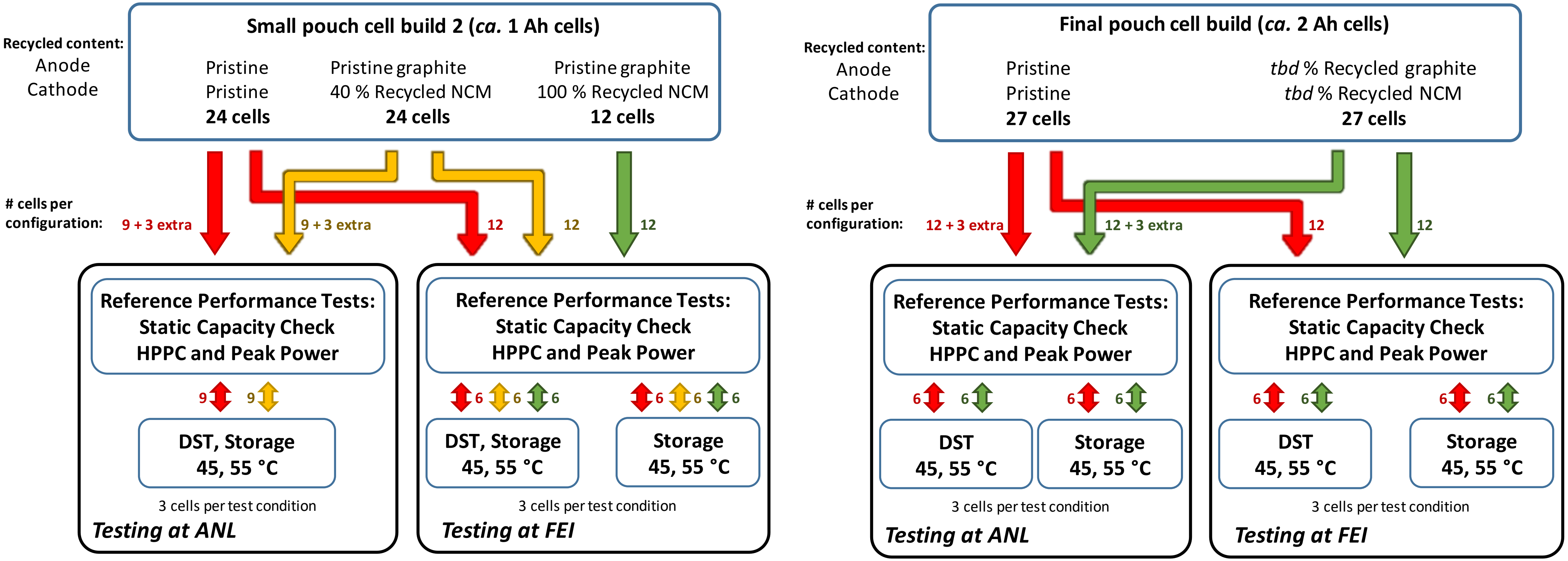
$LiMO_x$

Purification

Regen-eration

Recycled Materials

Approach – Cell Builds



- Intermediate cell build 2 is designed to examine the effects of blending 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.

Approach - Complexity

Recycling Feedstocks

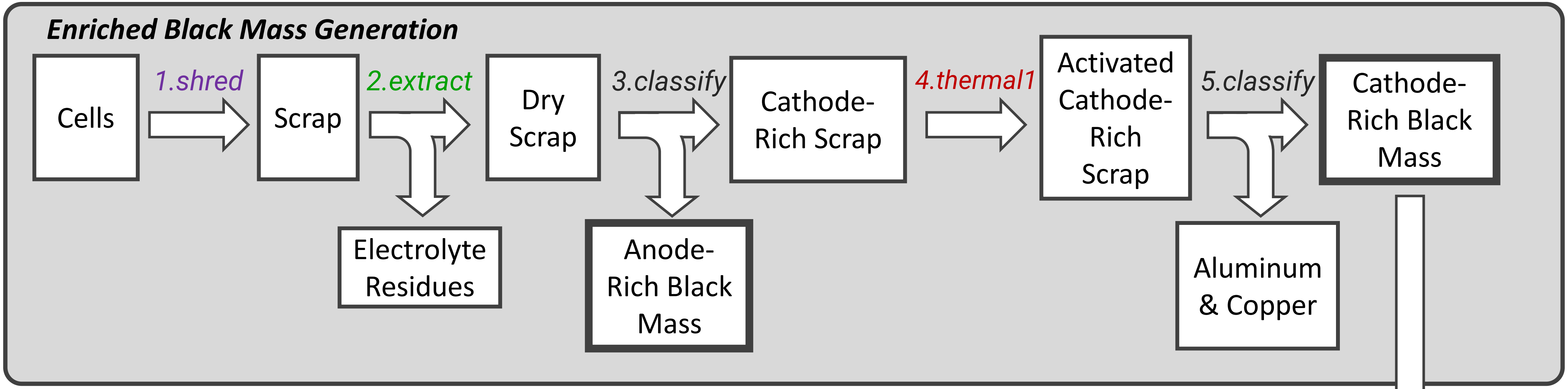
This project evaluates multiple possible inputs for direct recycling:

- Electrode production scrap
- Formed cells
- Entire battery modules

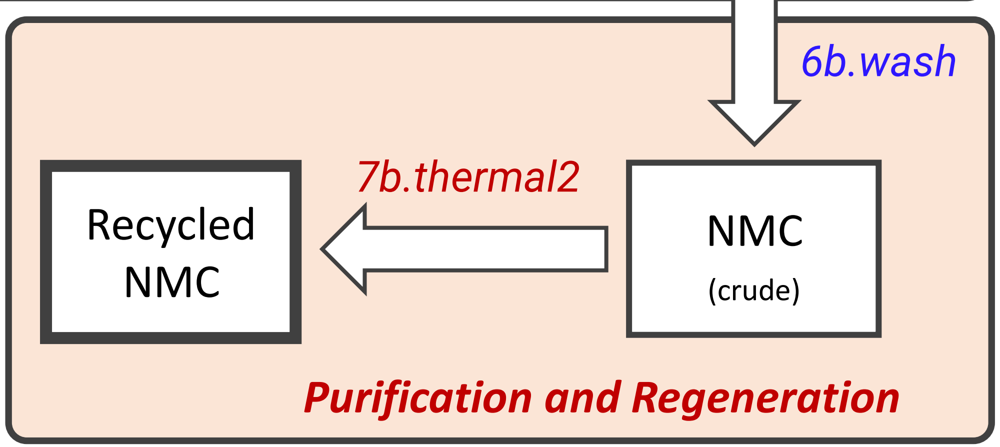
Cell Chemistries

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

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



- Process optimization for whole cell feedstock resulted in a modified route that can produce black mass that is enriched in one active material or the other.
- 3 kg of recycled NMC111 was recovered using this process to provide the necessary materials for Cell Build 2.



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

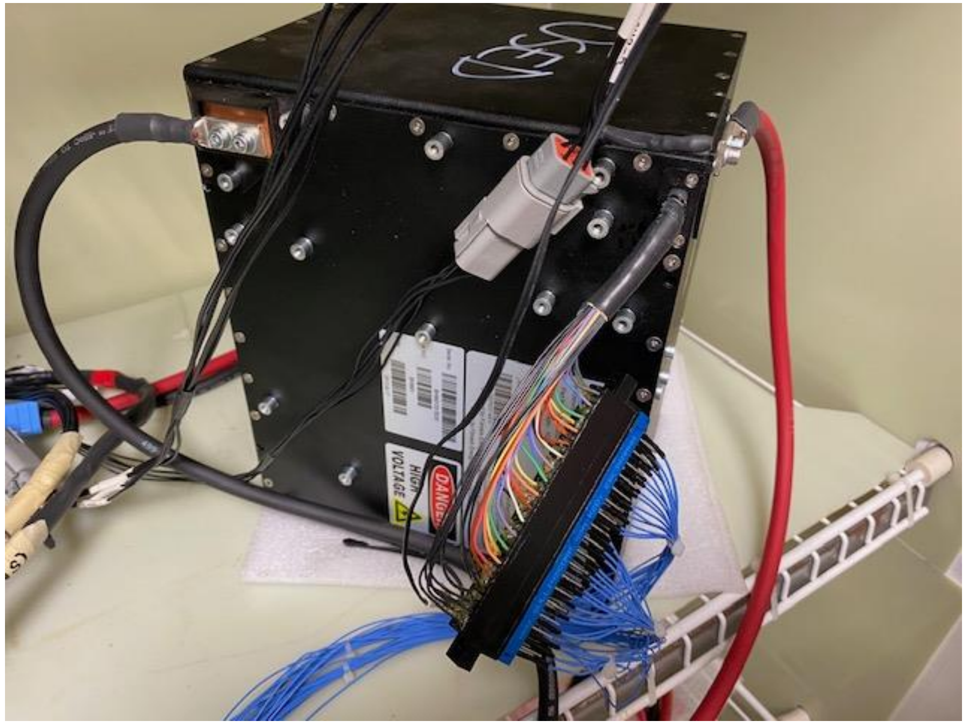
Positive Active Material Properties Gap Chart					
Characteristic	**Units**	**QC Spec**	**Virgin**	**Recovered from cells**	**Maximum %Recycled in Blend**
Particle size (D_{50})	um	9 - 14	11	10	100
Tap Density	(g/cm^3)	≥ 2.0	2.5	2.1	100
Reversible capacity (4.2 - 3.0 V vs. Li/Li+, 0.1 C)	(mAh/g)	145	150	130	**28**
Specific Surface Area (BET method)	(m^2/g)	0.15 - 0.55	0.23	0.71	65
First cycle efficiency	%	≥ 88	91	89	100
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 < 0.02 Ca < 0.05 Fe < 0.02 **Cu = 1.4** Al < 0.05 **F = 0.055**	--
pH assay	-log [H^+]	10.7 – 11.7	11.0	12.0	*Nonlinear*

- Direct recycled NMC recovered from whole cell feedstock has similar physical properties as pristine powder.
- Chemical contamination is problematic, but not inherent to the process. Further development will produce higher purity materials.

Technical Accomplishment – Recovery Scale-Up Module Feedstock

- An EOL 2.6 kWh module (70% of initial capacity) was obtained from Farasis inventory and evaluated electrochemically.

- Module dismantling for material recovery is in progress, but currently paused pending restart of operations at US facililty.



Technical Accomplishment – Cell Manufacturing Cell Builds 1 & 2

- Cell build 1 was completed in July 2019.
  - No differences in processability of recycled NMC as compared to pristine NMC.
  - The lower specific capacity of recycled NMC is reflected in overall lower cell capacities for these cells.
  - Lower specific capacity and FCE of recycled NMC is reflected in overall lower cell capacities for these cells.
  - Recycled NMC cells have higher initial impedance; recycled graphite contributes as well.

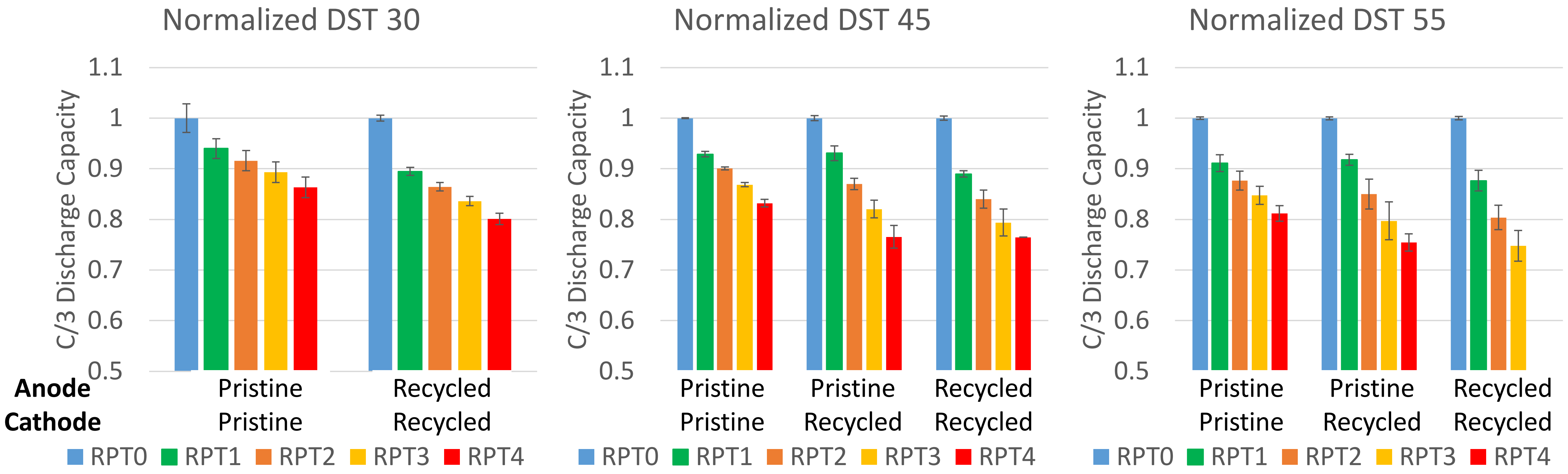
- Cell build 2 is currently pending material deliveries impacted by COVID-19 closures.

Cell Build 1 QC Metrics per Material Configuration:

Material Configuration		Capacity		First Cycle Efficiency		30s DCiR (40,50,60% DOD avg)	
Anode //	Cathode	mAh	+/-	%	+/-	mOhm	+/-
pristine	Pristine	630	12	90.1	1.1	71	5
pristine	**recycled**	591	9	88.6	1.0	99	17
recycled	**recycled**	587	14	87.4	1.8	106	24

Technical Accomplishment – Electrochemical Testing
Cell Build 1 Testing

Static Capacity Check Reference Performance Test (RPT) for Dynamic Stress Test (DST) Cell Groups



- Cells with both recycled graphite and recycled NCM had the fastest rate of capacity loss
- Cells with NCM as the only recycled component are intermediate.
- There is a general trend of increasing rate of capacity loss with increasing temperature across all cell groups.

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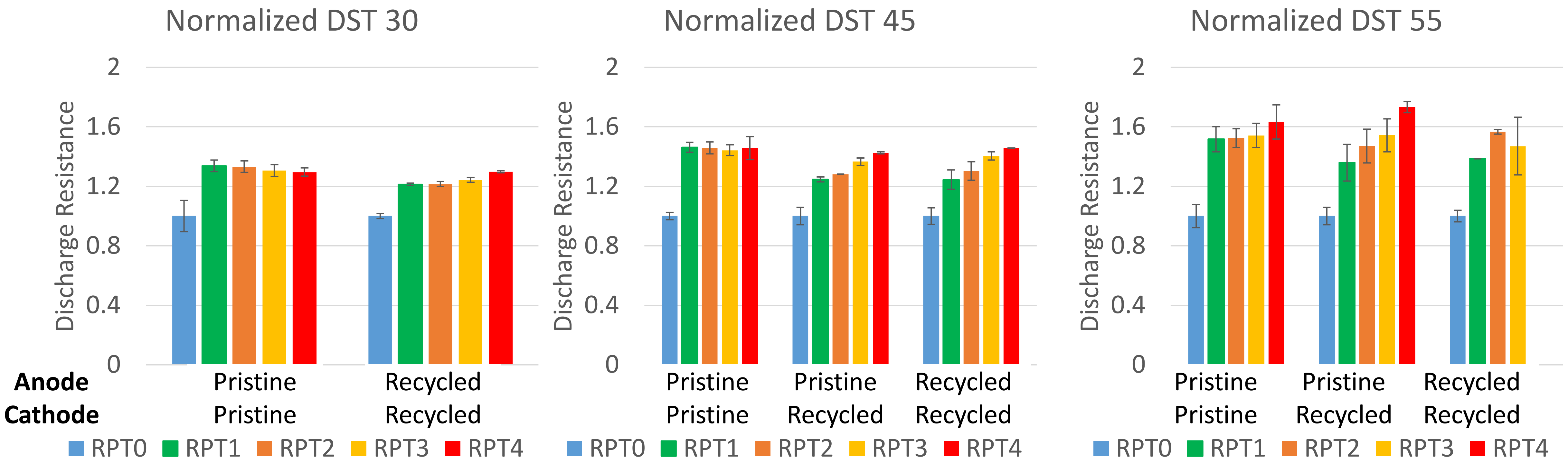
Technical Accomplishment – Electrochemical Testing
Cell Build 1 Testing

Hybrid Pulse Power Characterization (HPPC) RPT for DST Cell Groups



- Use of recycled active materials leads to higher resistance; impacts of individual materials are additive.
- Relative rates of impedance growth are similar.
- The impedance growth trends are different for recycled material cells.

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Technical Accomplishment – Electrochemical Testing Cell Build 1 Testing Summary

Cell Build 1 Summary

- Cells with both recycled NMC and recycled graphite have the highest rates of capacity fade and impedance increase.
- The differences in performance can be tied to the difference in material properties as illustrated in the gap charts.
- Cells with NCM as the only recycled component have a higher rate of capacity loss compared to pristine controls under cycling test conditions, but no significant difference in capacity fade for the storage cells.
- Across all material configurations, storage cells has higher rate of impedance increase as compared to cycling cells with the same materials.
- The shape of the impedance growth curve is different for pristine cells as compared to cells with recycled active material content, which suggests some differences in cell chemistry evolution.

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.

Proposed Future Research

- Complete material recovery from module feedstock for Cell Build 3.
- Complete deliverable cell builds and testing.
- Integrate process model with electrochemical test data to quantify impact of recycled materials on technology lifetime.

Summary

- Cell Build 1 manufacturing and testing is complete and generated data that will help quantify the impact of material purity on device performance.
- A recycling process for direct recovery and reuse of Li-ion battery active materials was used to recover 3 kg of recycled NMC111 from whole-cell feedstock.
- Kilogram-scale recovery of materials from module feedstocks is in progress.
- Remaining material recovery operations and cell builds have been delayed by COVID-19.