RECELL–DIRECT CATHODE RECYCLING: MATERIAL SEPARATION AND PREPARATION

Project ID: bat464

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Annual Merit Review
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PROJECT OVERVIEW

Timeline

- Project start: October 2018
- Project end: September 2021
- Percent complete: ~90%

Budget

FY19	$4,615k
FY20	$5,150k
FY21	$4,915k

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

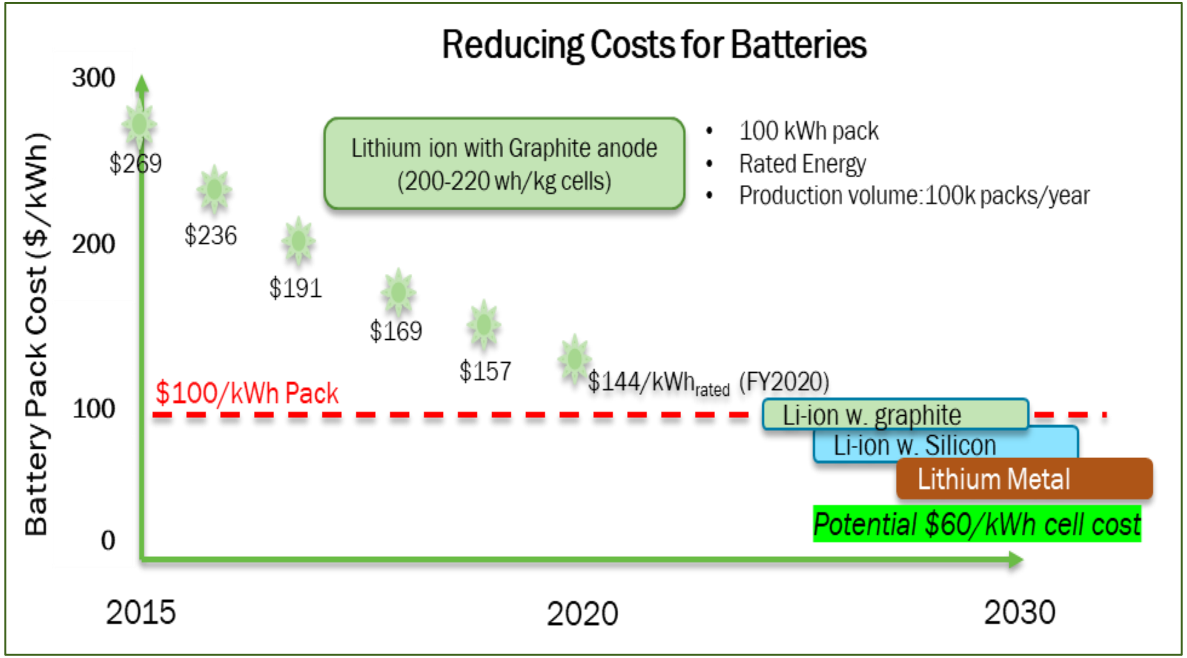
Partners

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

ReCell ADVANCED BATTERY RECYCLING

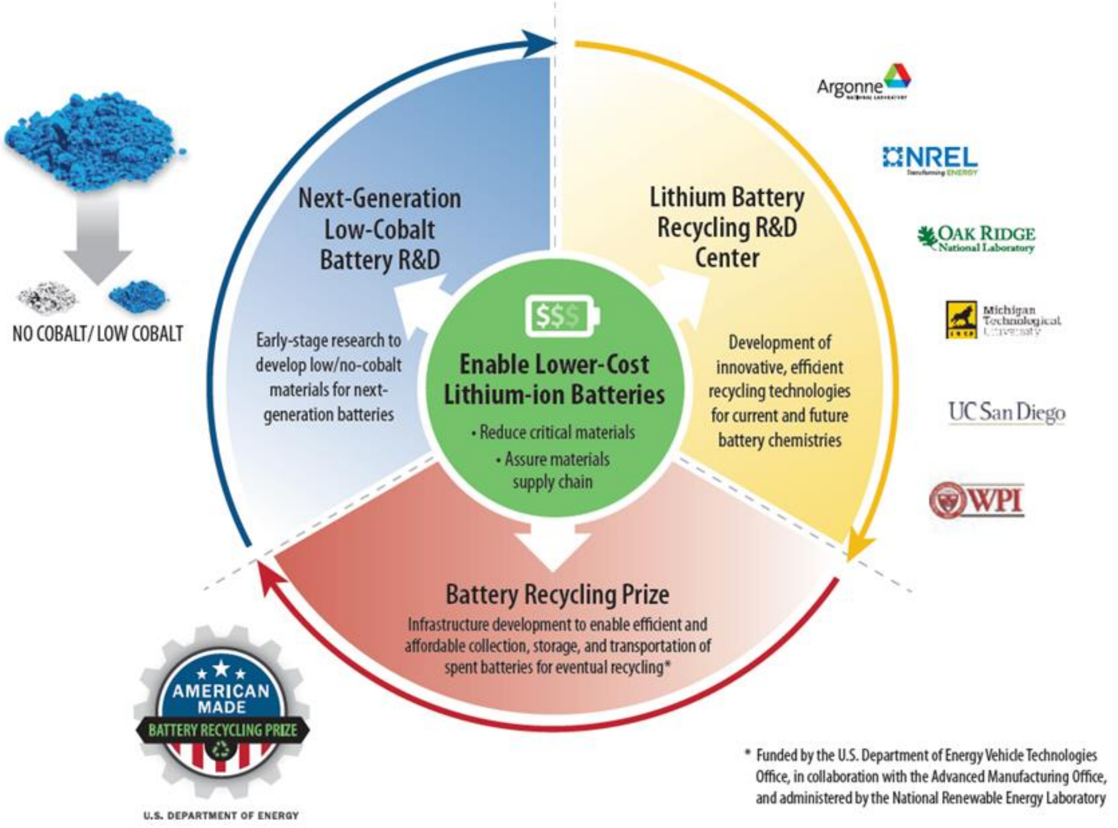
RELEVANCE

By 2025, reduce the cost of EV battery packs to less than $100/kWh with technologies that significantly reduce or eliminate the dependency on critical materials (such as cobalt) and utilize recycled material feedstocks.

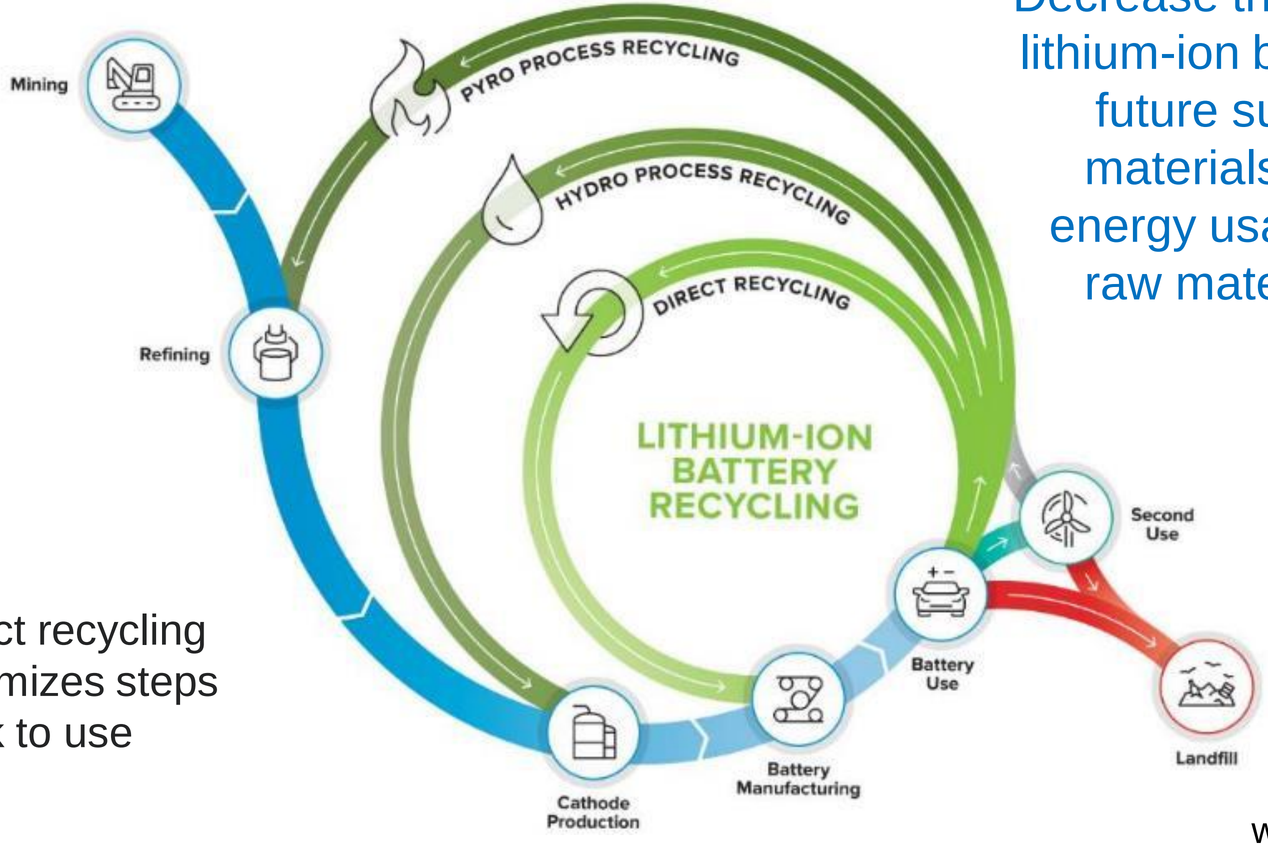


RELEVANCE

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



APPROACH



ReCell's Mission:

Decrease the cost of recycling lithium-ion batteries to ensure future supply of critical materials and decrease energy usage compared to raw material production

Direct recycling minimizes steps back to use

www.ReCellCenter.org

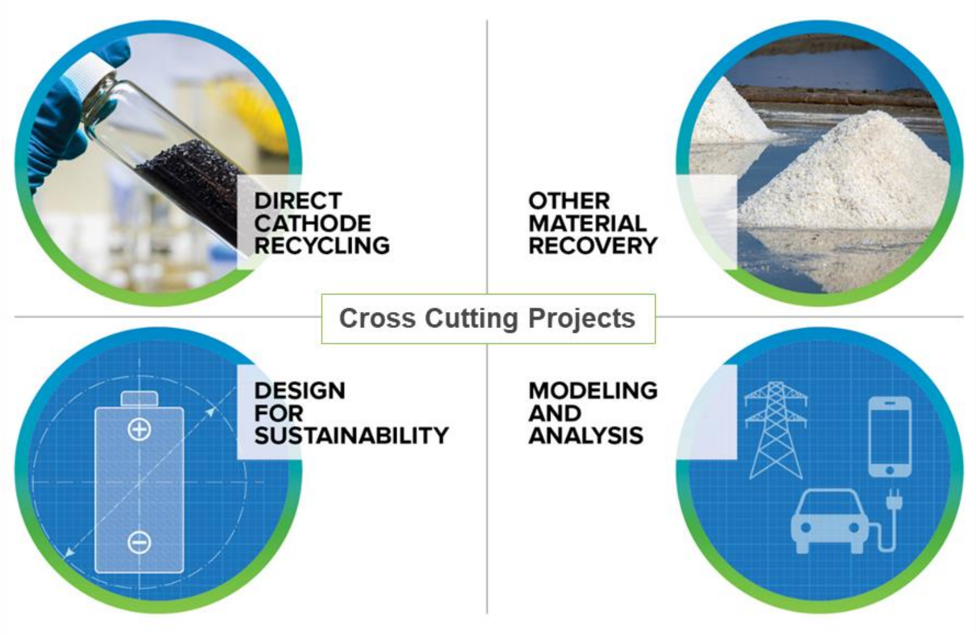
ReCell ADVANCED BATTERY RECYCLING

APPROACH

Year 1 – Bench scale testing: Powder-to-Cell → Year 2 – Start to scale up unit operations → Year 3 – Finish scale up and show cell-to-cell recycling

- Binder Removal
- Cathode/ Cathode Separation
- Relithiation
- Cathode Upcycling
- Impurity Impact

- Cell Design for Rejuvenation



- Cell Shredding
- Electrode Delamination
- Anode/ Cathode Separation
- Electrolyte Component Recovery

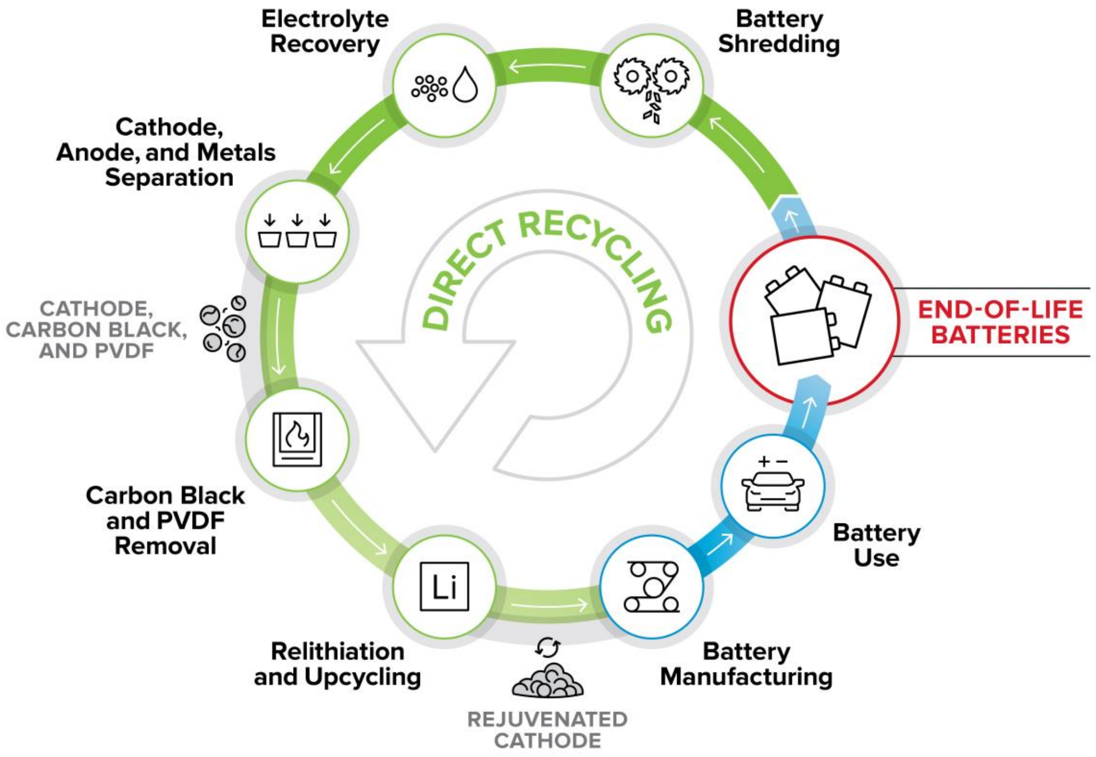
- EverBatt (TEA/LCA)
- LIBRA (Supply Chain Modeling)

Program does not include battery dismantling, transportation, or 2^{nd} use

APPROACH

Typical Direct Recycling Process Flow

- Multiple processes investigated to mitigate risk
- Continual review of new project ideas
- End projects that are not showing promise in cost and performance
- These processes can benefit other recycling processes



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MILESTONES

FY20 Q3	Complete	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	Complete	Demonstrate recovery of anode and cathode powders using the new pilot scale froth column
FY21 Q1	Complete	Preliminary report of sensitivity analysis of battery recycling in the LIBRA model focusing on outputs including the number of recycling plants built and the percentage of batteries recycled over time.
FY21 Q2	Complete	Demonstrate 30% graphene yield from spent anode using a Taylor Vortex Reactor
FY21 Q3	Ongoing	Final report on performance and cost modeling of directly recycled manufacturing scrap
FY21 Q4	Ongoing	Provide preliminary cost analysis, yield, and efficiency on the separation-relithiation conditions on NMC spent electrodes via solvent-based dual process

Each Individual project has its own milestones, though not listed here.

COVID-19 has reduced lab time and may cause delays in completing FY22 milestones

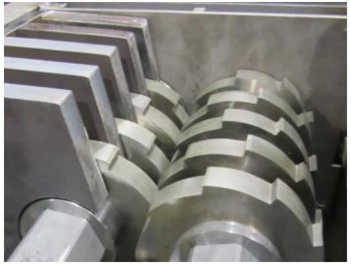
APPROACH TO SEPARATION AND PREPROCESSING

- Minimize contamination
- Recover as many materials as possible
- Keep processing costs low
- Reduce waste streams
- Understand the role of remaining contaminants
- Develop solutions to problematic contamination
- Develop multiple solutions to increase likelihood that a complete working process can be developed

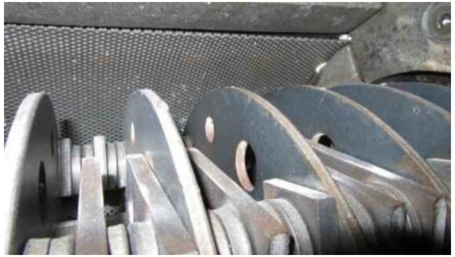
ReCell ADVANCED BATTERY RECYCLING

HAMMERMILLING *vs*. SHREDDING

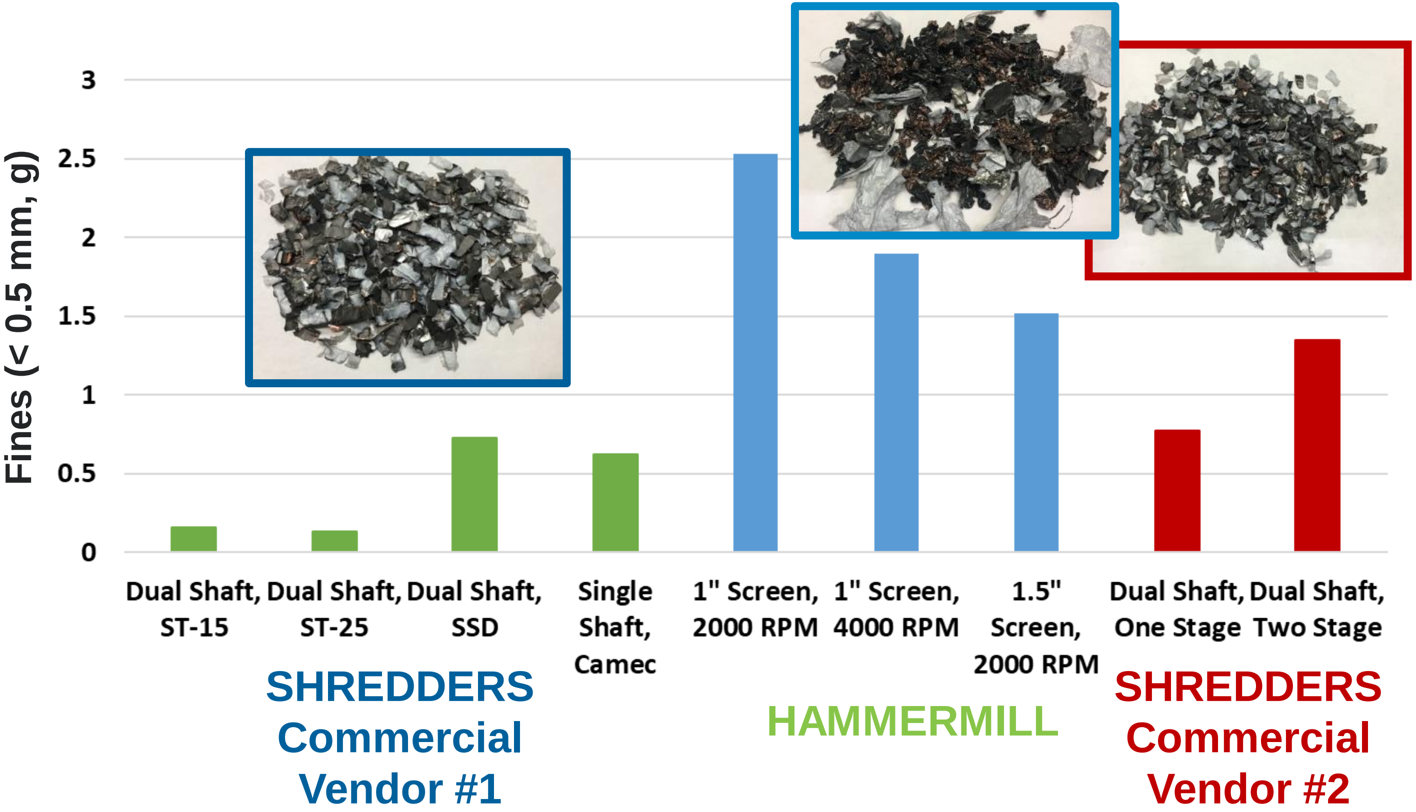
Fine Particle Contamination from Size Reduced, Dry Pouch Cells



Dual Shaft Shredder



Hammer Mill



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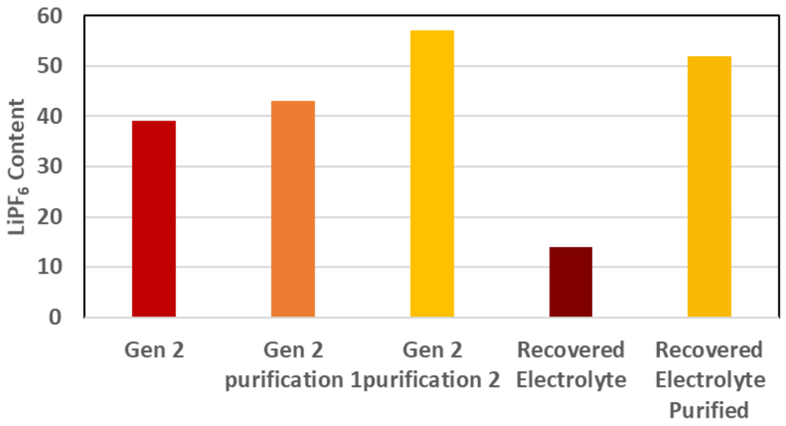
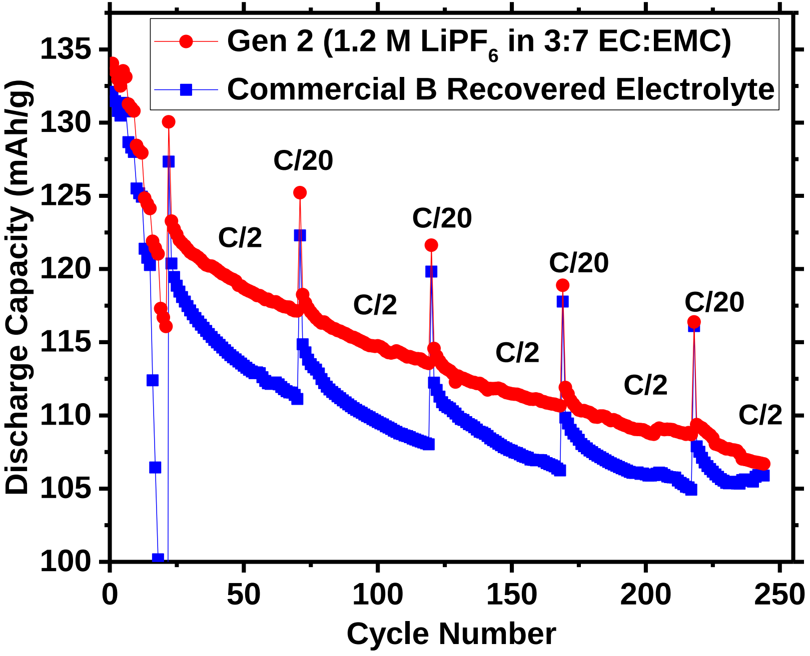
RECOVERING ELECTROLYTE

- Electrolyte can be recovered using additional carbonate solvent after shredding
- Tested different end-of-life commercial electrolytes to see if they could be recovered and made into a new cell
- Recovered electrolytes perform close to baseline electrolytes
- Tested different purification methods





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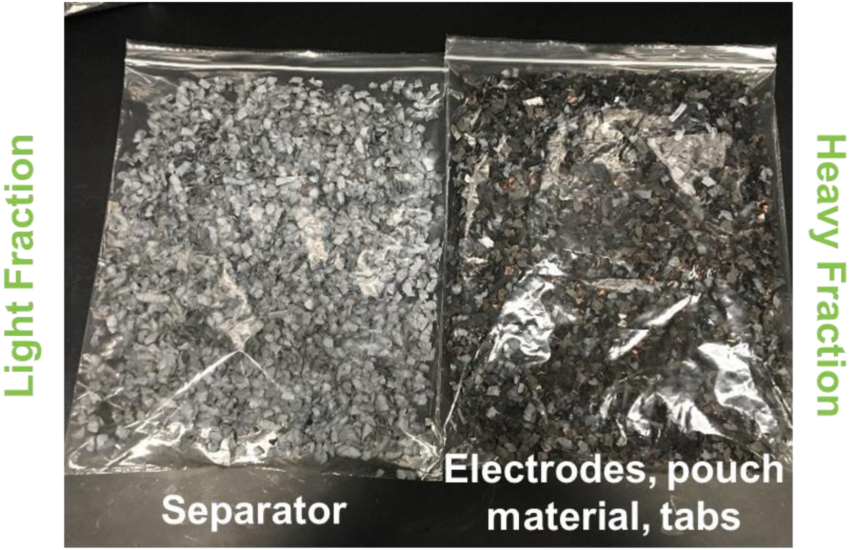
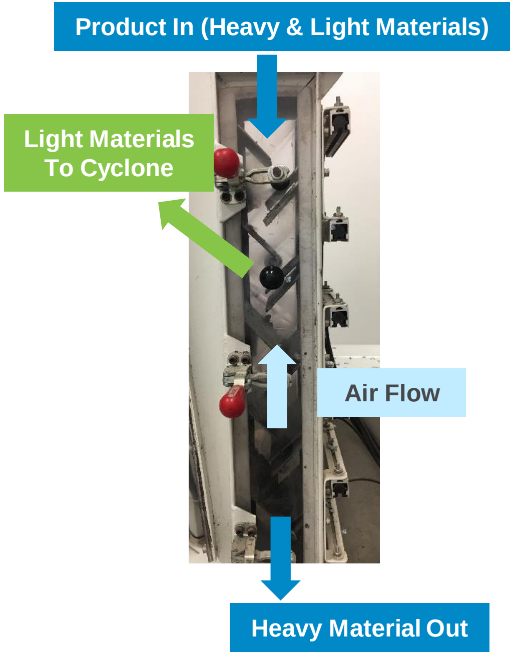
REMOVING SEPARATOR

Aspirator and Cyclone

- Separated components using an aspirator/cyclone by controlling air flow
- Light, non-bound plastics can be easily removed



Some separator is still present in bound clumps of shredder residue



ReCell ADVANCED BATTERY RECYCLING

SOLVENT BASED ELECTRODE RECOVERY

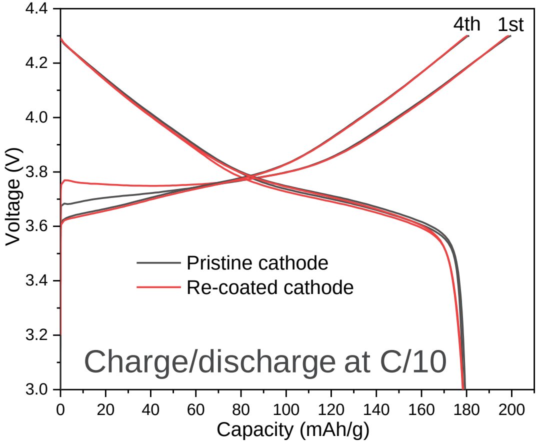
Cathode Scrap

SolveX

Re-coat

New cell

- Demonstrated the direct recycling and re-manufacturing of cathode scraps using SolveX process
- Re-processed the recovered cathode films for new cathode coating
- Proved that SolveX process does no damage to the active materials, does not corrode the current collectors, and has negligible influence on electrochemical performance



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SOLVENT BASED ELECTRODE RECOVERY

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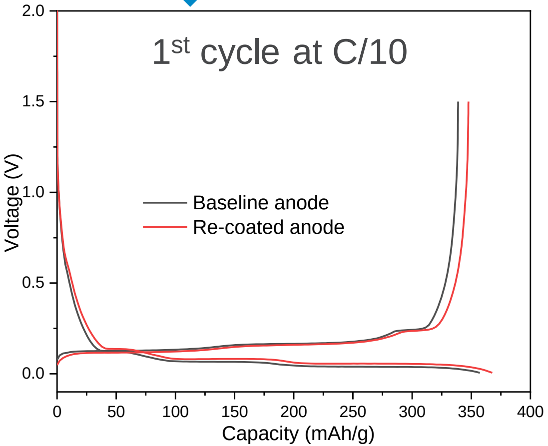
New cell

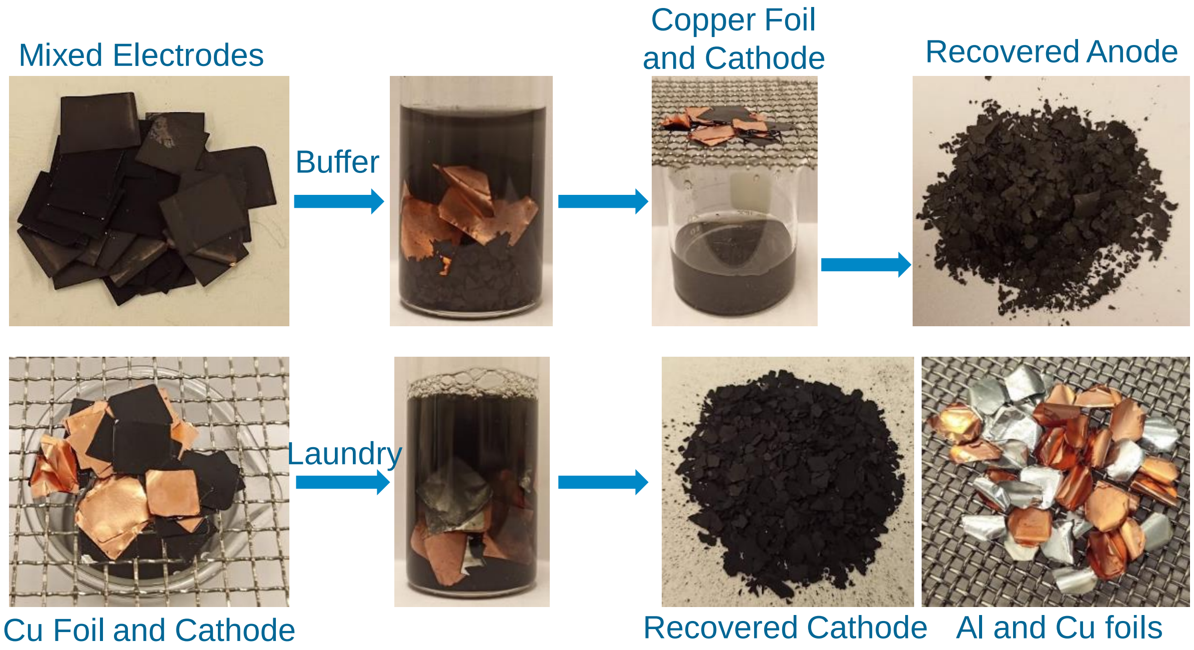


- Demonstrated the recovery of anode material using SolveX process
- Reprocessed the recovered anode material into new anode coating
- Results show that SolveX process does not damage the active materials
- recovered materials have similar electrochemical performance to virgin material

ReCell ADVANCED BATTERY RECYCLING

AQUEOUS SEQUENTIAL SEPARATION

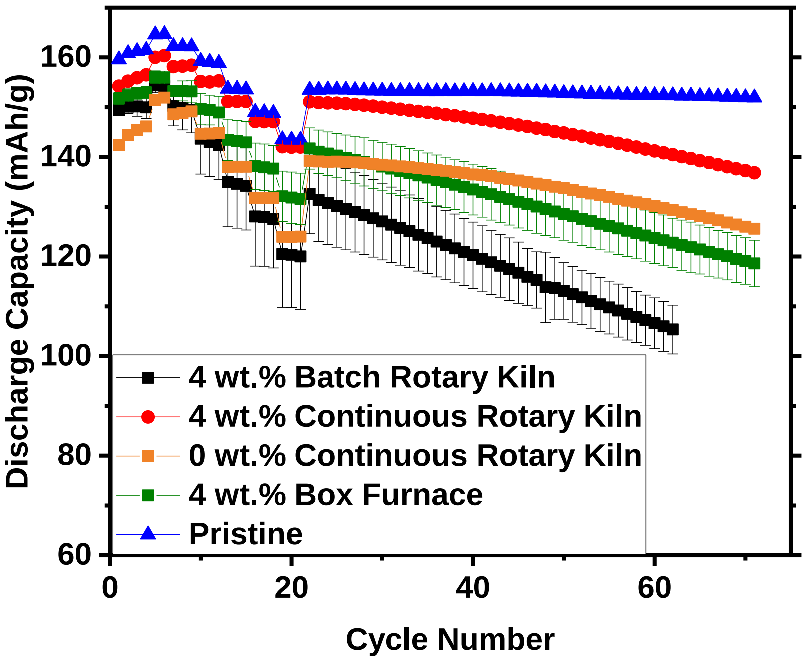
- Separate electrode materials from foils to sequentially recover anode from cathode
- Reduced cost and simplicity of a single mixing vessel process
- Identified the optimal composition of the aqueous solution containing buffer and additive X



- Developed a process to separate electrode materials from current collectors in a water solution

THERMAL BINDER REMOVAL IN A CONTINUOUS ROTARY KILN

- Continuous rotary kiln process results in improved performance compared to a batch rotary kiln or a box furnace
- Easily scalable and simple



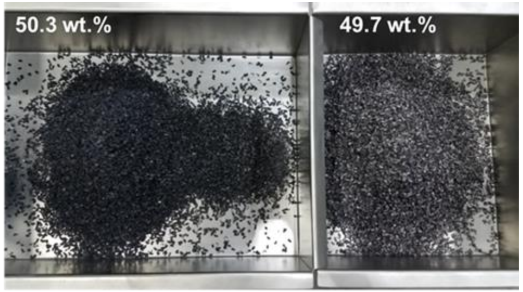
NMC 111 mixed with 3 wt.% PVDF and 5 wt.% carbon black processed at 500°C with 0 or 4 wt.% $LiOH \cdot H_2O$

ReCell ADVANCED BATTERY RECYCLING

SEPARATION OF ANODE AND CATHODE

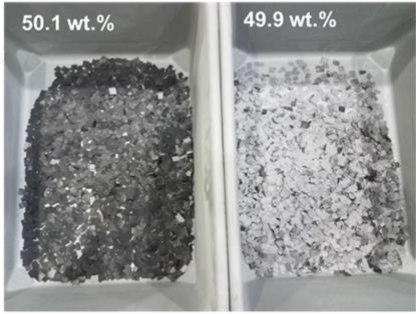
- A separation technique has been used to separate a 1:1 mixture (by weight) of anode on copper foil from cathode on aluminum foil
- Three size fractions (similar to those obtained after shredding)
- Additional studies show that using Process A again on the separated material can further improve separation efficiency
- Decreased separation efficiency of large pieces, 1-1.5”, is due to the size of the lab-scale equipment

~1/4” Electrodes



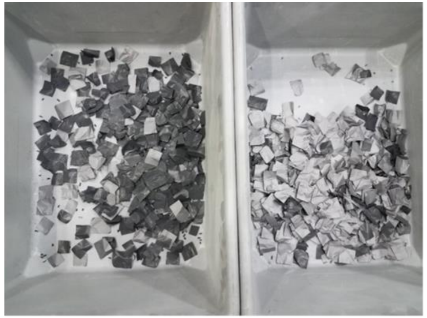
Fraction 1 Cathode/Al Foil | Fraction 2 Anode/Cu Foil

0.5-1” Electrodes



Fraction 1 Cathode/Al Foil | Fraction 2 Anode/Cu Foil

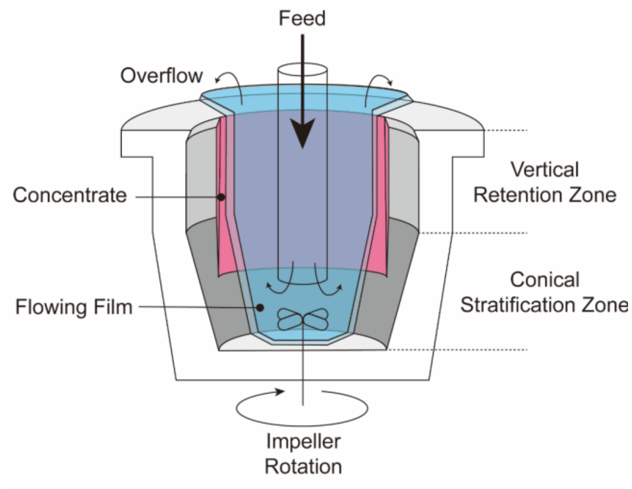
1-1.5” Electrodes

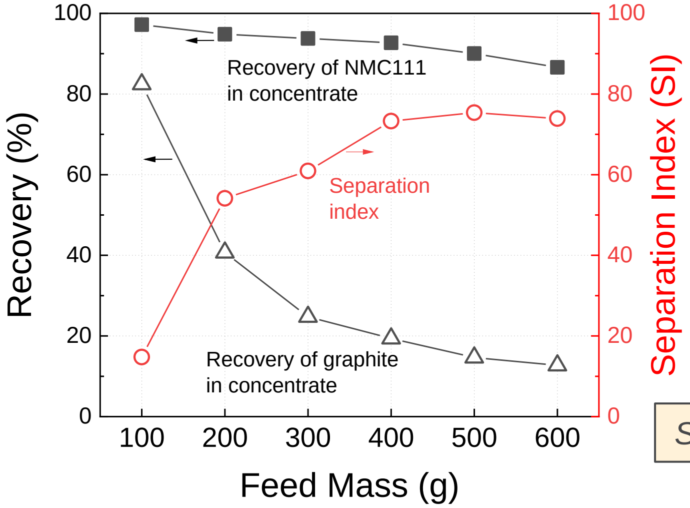


Fraction 1 Cathode/Al Foil | Fraction 2 Anode/Cu Foil

ReCell ADVANCED BATTERY RECYCLING

SOLVENT-BASED GRAVITY SEPARATION





Multistage Separation

Product	NMC111 (%)	Graphite (%)
Tail	0.60%	99.40%
Conc	99.03%	0.97%

SI = R(NMC111) – R(Graphite)

- Alternative to the previously demonstrated froth flotation process for anode/cathode separation
- Centrifugal gravity separation was used to separate a mixture of the pristine graphite and NMC111 powders.
- Over 90% of NMC111 is collected within the concentrator, and the recovery of graphite within the concentrator varies significantly with feed mass.
- After multistage separation processes, the concentrate product consisted of 99% of NMC111

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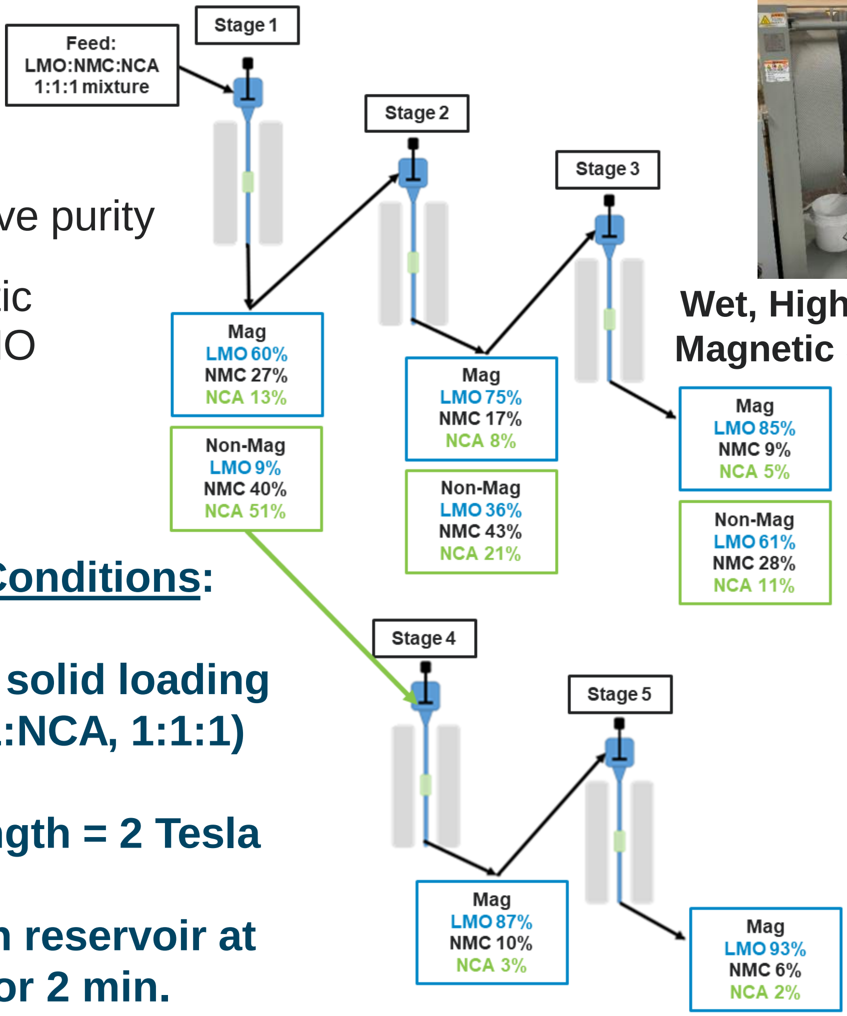
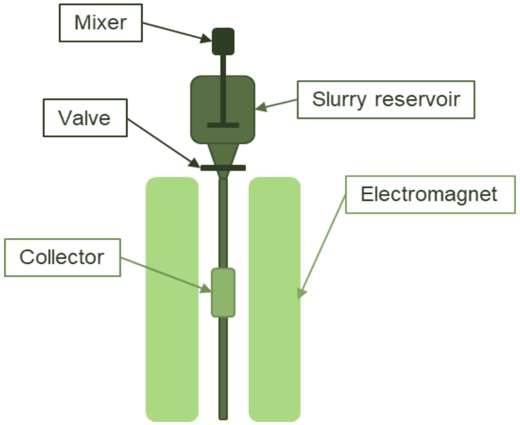
ELECTROMAGNET

Separation of Cathode Mixtures

- Multistage magnetic separation can improve purity
- After the 3rd stage, the yield of the magnetic fraction is 18% and contains 85% pure LMO
- Potential to be used as a cleanup step



Wet, High-Intensity Magnetic Separator



Separation Conditions:

Slurry = 5 wt.% solid loading (LMO:NMC111:NCA, 1:1:1)

Magnetic Strength = 2 Tesla

Slurry mixed in reservoir at 200 RPM for 2 min.

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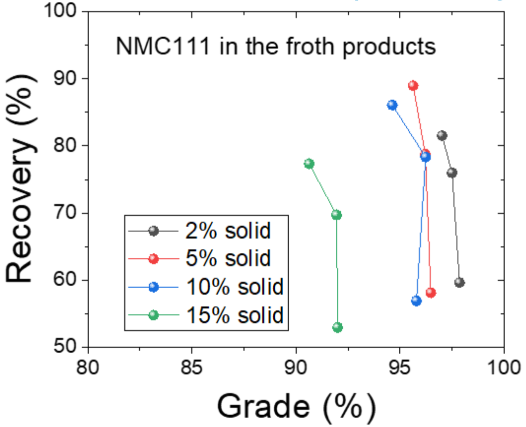
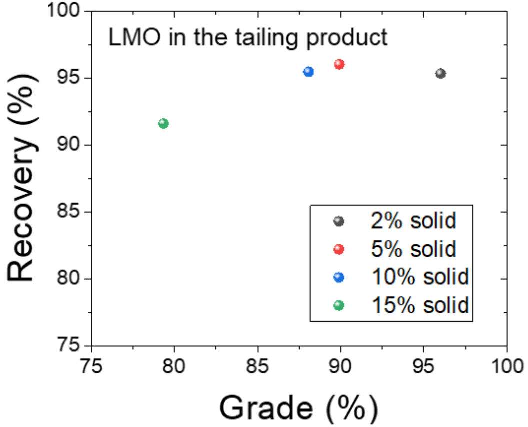
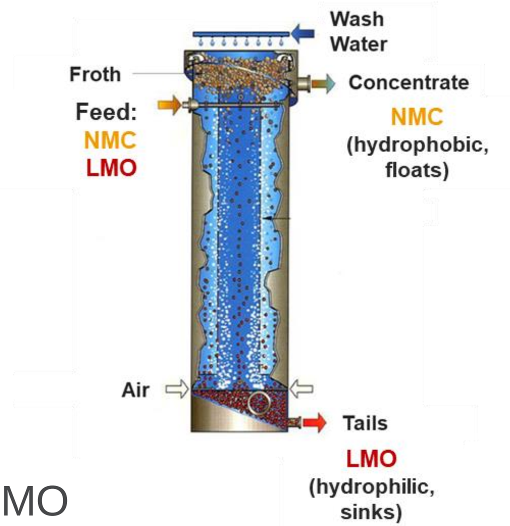
CATHODE/CATHODE SEPARATION BY FROTH FLOTATION

Initial processing in a bench scale Denver cell





Scale up in a froth column



- Froth flotation experiments were conducted using a rougher-cleaner circuit to separate a mixture of pristine NMC111 and LMO materials
- The grade of the NMC111 in the froth product reached 95-97% at a recovery rate of 80-90% after two flotation steps
- The tailing consisted of 90-98% of LMO by weight at a recovery rate of 95% or above
- Works well with other binary mixtures, such as NCA/LMO, LCO/LMO, and NMC111/NMC622

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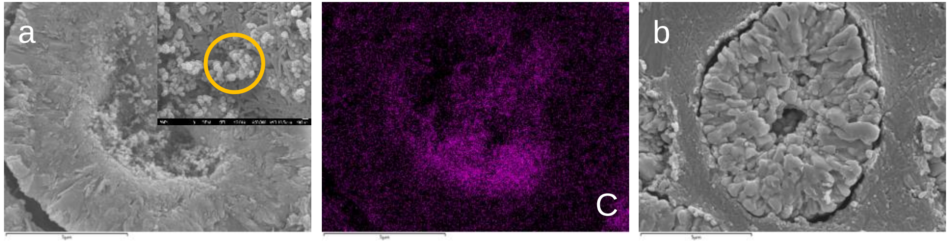
CARBON IMPURITY AFFECT DURING COPRECIPITATION

Tap density (g/ml) of precursors at different times

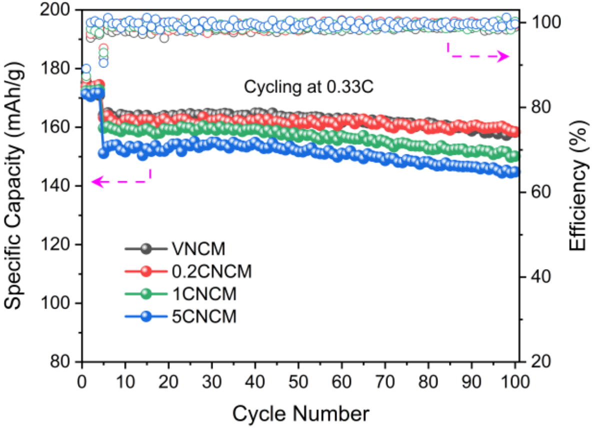
Sample	3hrs	6hrs	9hrs	12hrs
VNCM	0.65	1.01	1.45	1.85
0.2CNCM	0.86	1.51	1.70	1.92
1CNCM	0.89	1.51	1.75	1.86
5CNCM	0.95	1.47	1.76	1.83

- Carbon impurity accelerates the precipitation increasing initial tap density



SEM cross-sectional images of (a) CNCM precursor with EDS carbon signal and (b) CNCM cathode

- Carbon nano-particles are observed inside precursor, and after sintering, cathode particles with central pore are formed



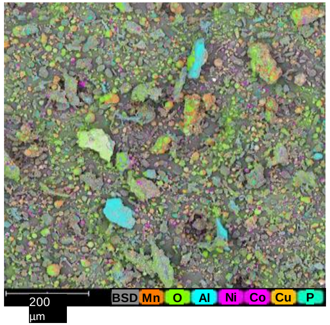
Rietveld refinement results of NCM622 cathodes

Sample	a-axis (Å)	c-axis (Å)	volume (Å^3)	Ni_{Li} (%)	χ^2
VNCM	2.865	14.214	101.04	3.84	2.78
0.2CNCM	2.867	14.220	101.25	3.93	2.75
1CNCM	2.867	14.219	101.24	4.01	2.87
5CNCM	2.867	14.224	101.31	4.18	2.90

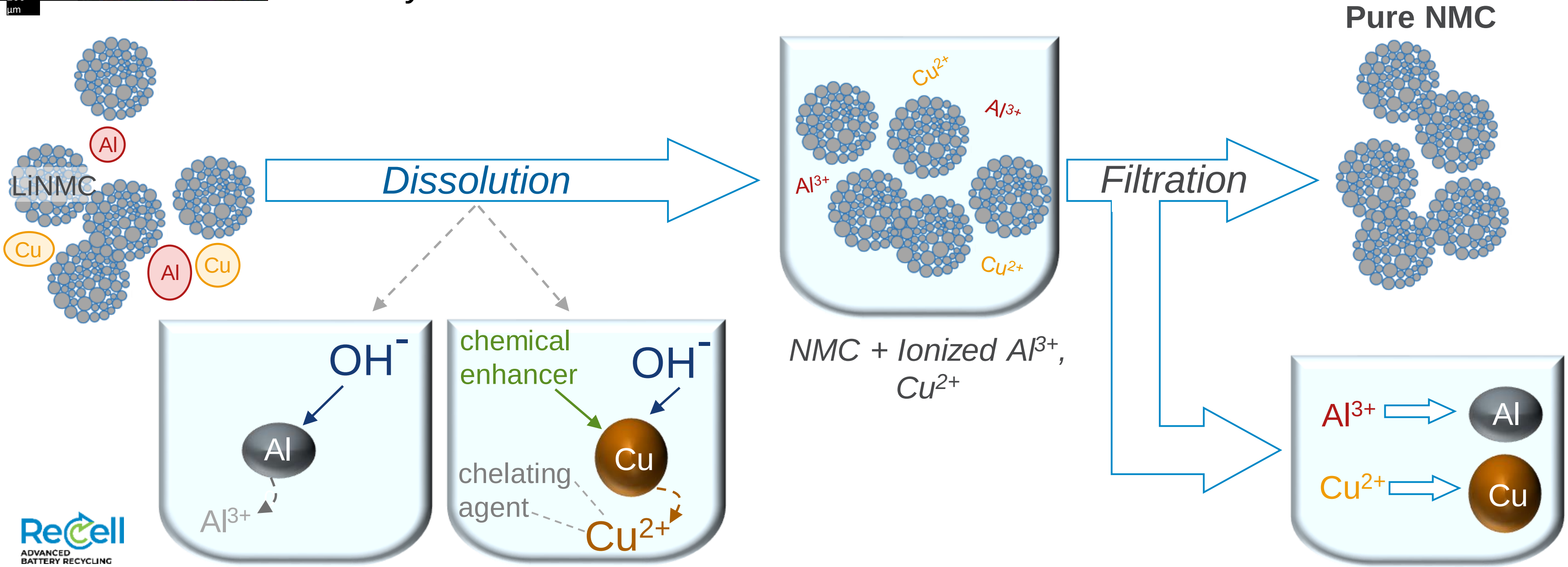
- No impurity peaks in XRD pattern
- Cation mixing degree increases with C content, which hinders electrochemical performance.

ReCell ADVANCED BATTERY RECYCLING

BLACK MASS PURIFICATION: MOTIVATION & OVERVIEW



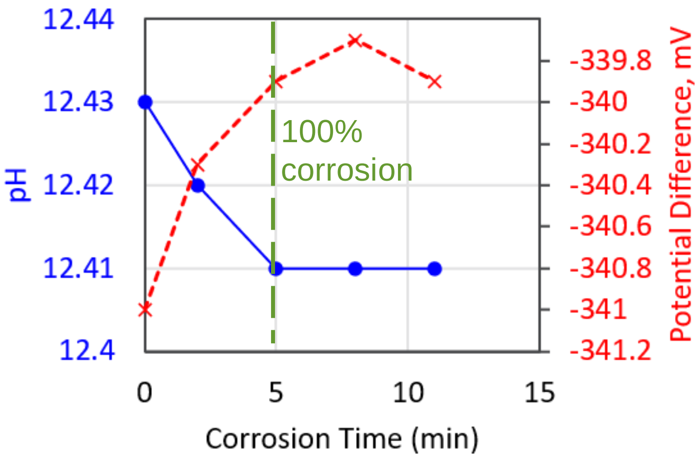
- Shredded black mass contains trace Al & Cu: potentially has safety impacts and reduces the performance of recycled material
- **Identify** and **optimize** BM purification process to enable **complete and rapid dissolution** of solid contaminants (Al, Cu) **without adversely impacting** structure or electrochemical performance of NMC



ReCell ADVANCED BATTERY RECYCLING

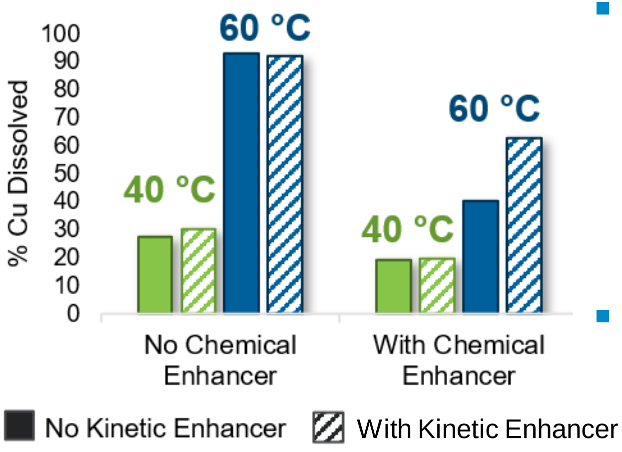
BLACK MASS PURIFICATION: HIGHLIGHTED RESULTS

Al Corrosion



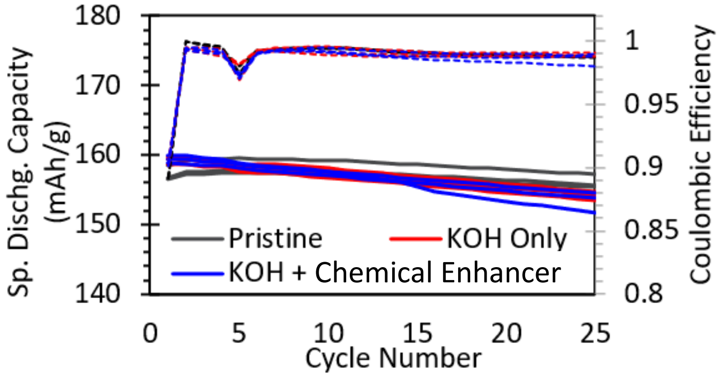
- Aluminum corrosion is accelerated via elevated temperature and kinetic enhancement in alkaline solution
- Full corrosion achieved in **5 min** (*40 °C w/ kinetic enhancement; pH 13*)

Cu Corrosion



- Exploring use of chemical enhancer; chelating agent; kinetic enhancer; and elevated temperature to improve kinetics and thermodynamics
- **>93%** corrosion demonstrated to date; improvements ongoing

Impacts on NMC



- Purification treatment **does not significantly impact** electrochemical performance of NMC

SUMMARY AND FUTURE WORK

- Processes have been developed to enable pure cathode to be recovered as a feedstock for relithiation and upcycling
- Minimizing contamination at every stage of the process is needed to retain the material performance
  - Continuing to evaluate the effects of contamination and find ways to eliminate it if it is unavoidable
- Many different separation modalities have been found that with a correctly designed process can give high purity with high yields
- We are working to scale up these processes to kg quantities and more
  - Shredder to be installed shortly
  - Froth column being optimized for anode/cathode and cathode/cathode separations
- We are applying these processes to manufacturing scrap, which can enable their implementation before significant quantities of end-of-life batteries are available for recycling
- Developing an overall recycling process that recovers as much material as possible, at the lowest processing cost, and with cathode that has comparable performance to the pristine

Any proposed future work is subject to change based on funding levels

RESPONSE TO REVIEWERS

Not reviewed last year

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