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

ReCell
ADVANCED BATTERY RECYCLING

RECYCLING OF NON-CATHODE BASED BATTERY MATERIALS

RECELL CENTER FOR ADVANCED BATTERY RECYCLING

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Argonne National Laboratory

bat466
Virtual Poster
June 1-4, 2020

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%

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

Budget

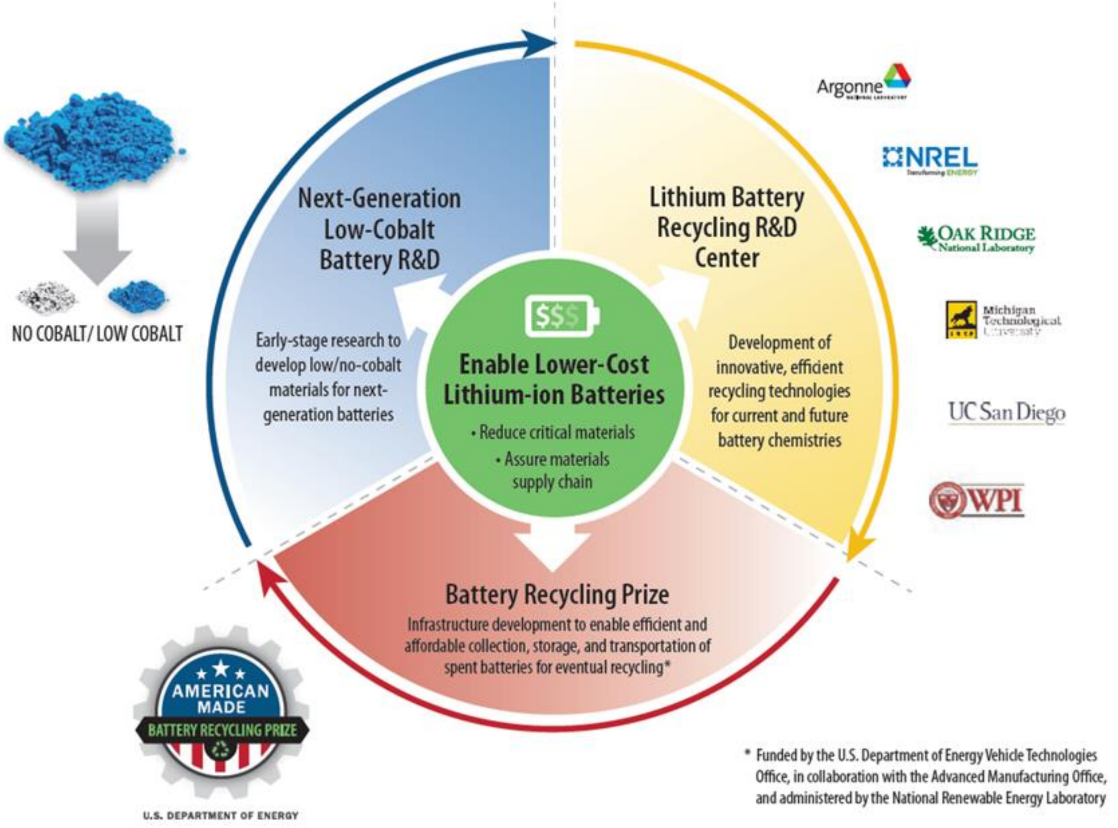
FY19	$4,615k
FY20	$5,150k

Partners

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

RELEVANCE

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



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

ReCell 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 that are not listed here.

COVID-19 has reduced lab time and may cause delays in completing FY20 milestones

RECYCLING OF NON-CATHODE BASED BATTERY MATERIALS

GOALS AND APPROACH

- To maximize the potential of the recycling process, all cell materials that can be recovered and reused must be looked at.
- All projects will be evaluated for compatibility and economic feasibility, and ultimately integrated into a coherent process with maximum material recovery and recycling.
- This focus area covers the following projects:
  - Cell pre-processing (size reduction) – Jessica Durham (ANL)
  - Electrolyte Components Recovery – Albert Lipson (ANL)
  - Solvent-based Delamination of Electrodes, Ilias Belharouak (ORNL)
  - Anode/Cathode Separation – Jessica Durham (ANL)

CELL PRE-PROCESSING

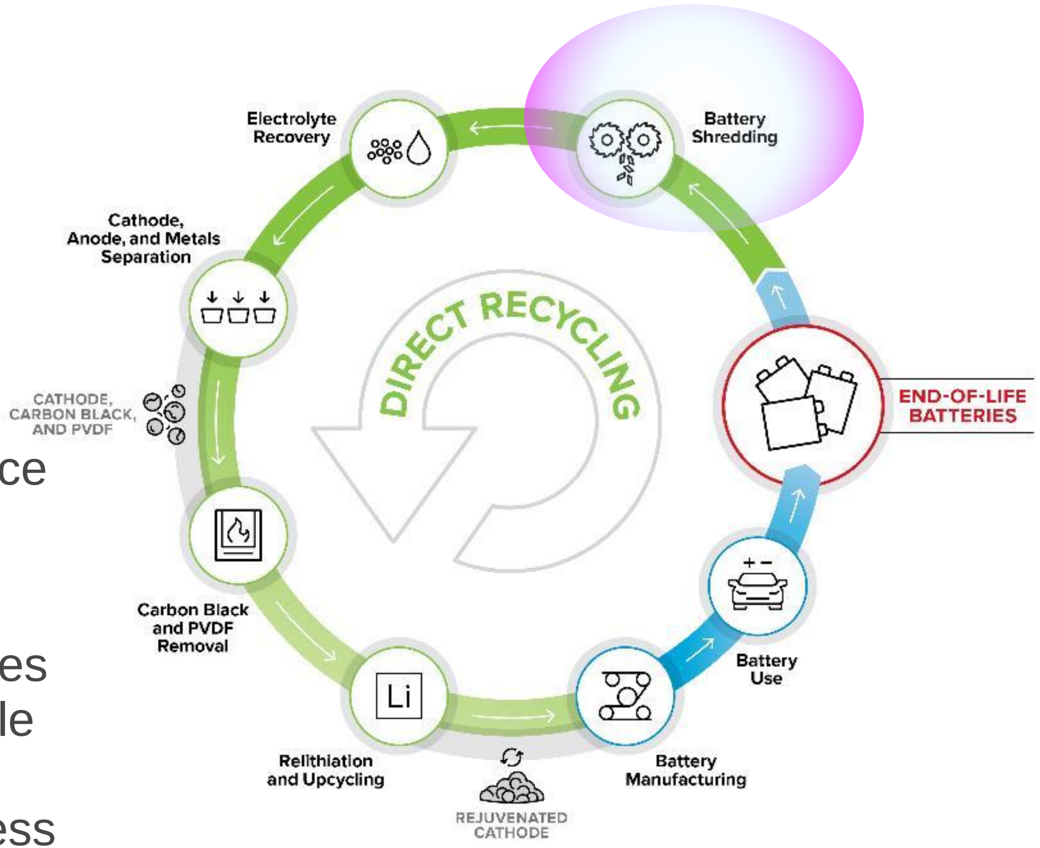
Project Description

Disassembly/ Dismantling

↓

Shredding/Crushing/ Milling/Punching

- Safely and cost-effectively size reduce batteries or manufacturing scrap to produce a useable black mass
- Commercial size reduction of batteries creates a large amount of fine particle contamination which can be carried throughout the direct recycling process



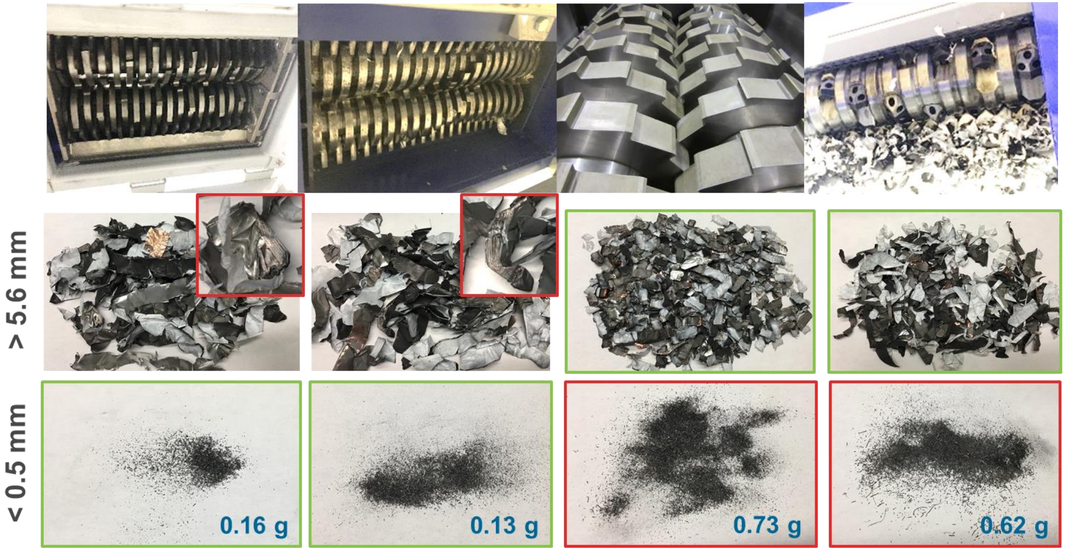
CELL PRE-PROCESSING

Comparison of Shredders for Size Reduction

- Project goals:
  - Determine scalable size reduction technologies for batteries or manufacturing scrap
  - Explore effects of size and size distribution of broken down battery materials with respect to separation efficiency
- Collaborated with a shredder to shred 40 Ah pristine, dry pouch cells in four different shredder systems

Shredder Systems

ST-15	ST-25	SSD	Camec
Dual Shaft	Dual Shaft	Dual Shaft	Single Shaft

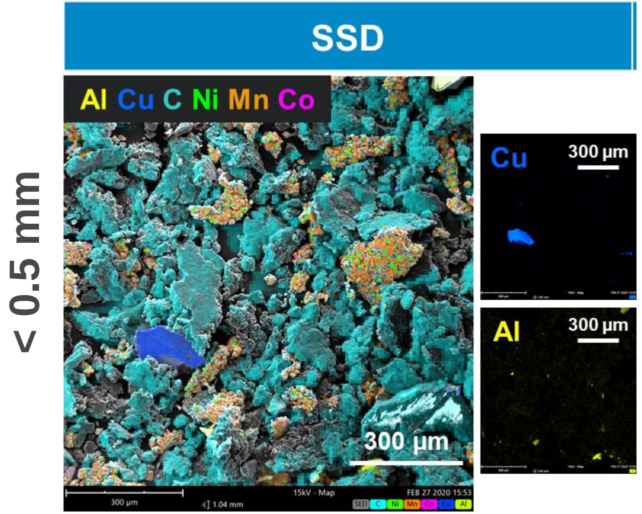


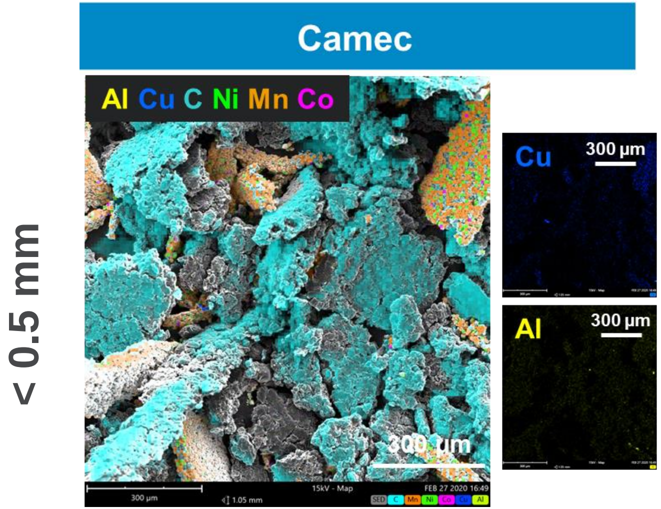
ST-15	ST-25	SSD	Camec
POOR MATERIAL LIBERATION LOW FINE PARTICLE CONTENT	POOR MATERIAL LIBERATION LOW FINE PARTICLE CONTENT	GOOD MATERIAL LIBERATION CONSISTENT SIZE DISTRIBUTION GREATER FINE PARTICLE CONTENT	GOOD MATERIAL LIBERATION POOR SIZE DISTRIBUTION GREATER FINE PARTICLE CONTENT

CELL PRE-PROCESSING

Examination of Fine Particle Contamination

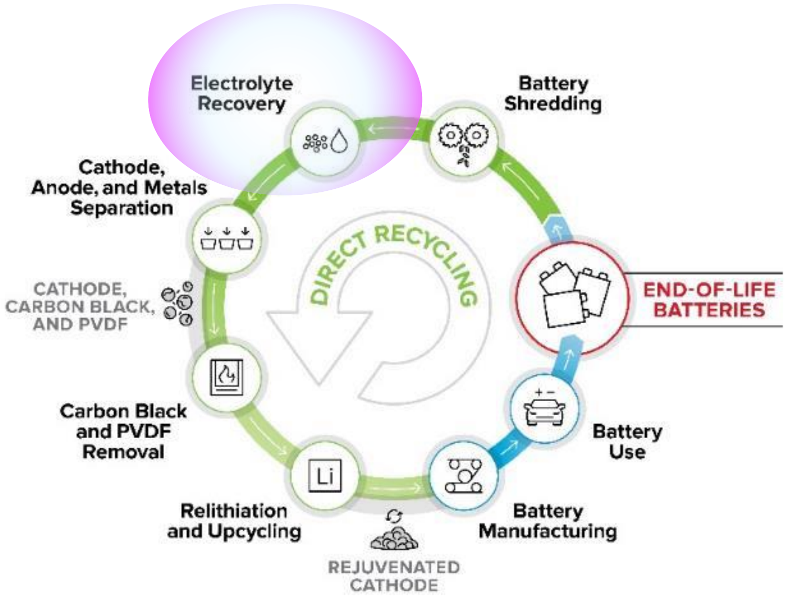
- Down selected shredder systems to those that provided the best material liberation, which will expedite component separation and material recovery in the direct recycling process
  - SSD (dual shaft)
  - Camec (single shaft)
- Cathode, anode, and metal foils are observed in the SEM
  - Electrodes are delaminated from foil, likely because of stress during shredding
  - Foils and material as small as 100 nm to a few micron are present (difficult to remove)





ELECTROLYTE RECOVERY

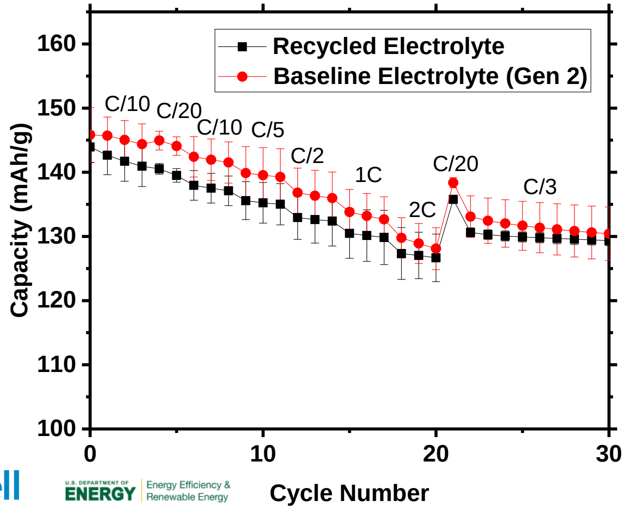
- **Project Description:**
  - Utilize and determine efficiency of various methods to extract electrolyte
  - Determine the cost effectiveness of these methods
  - Demonstrate the ability to recover viable $LiPF_6$ and use it to reconstitute an electrolyte
  - Find low-cost purification methods to make the $LiPF_6$ product easier to reuse or sell

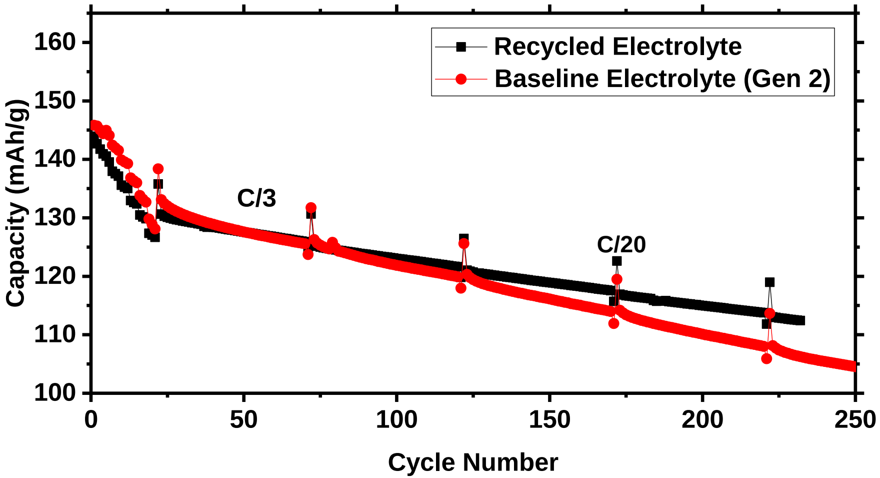


- **Project Goals:**
  - Find a cost-effective method to remove electrolyte
  - Recover as much of the used electrolyte components as is economically feasible
  - Find a cost-effective purification method to separate components

ELECTROCHEMICAL PERFORMANCE OF RECOVERED ELECTROLYTE IN FULL CELLS

- Electrolyte was extracted from cathode pieces using DEC, and evaporated at 90ºC under argon gas. The residue was diluted with 1:1 EC:DEC to make approximately 1 M $LiPF_6$ solution, which was then used as an electrolyte to assess electrochemical performance in a full cell (coin cell).
  - Initial capacity is similar to Gen 2, although the initial capacity fade is more rapid, but then stabilizes at a lower slope than Gen 2. This indicates faster SEI growth.





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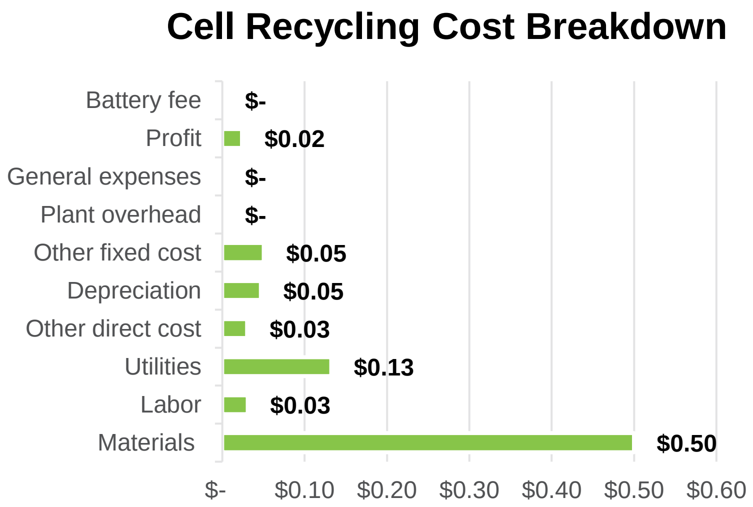
SCALE UP OF ELECTROLYTE RECOVERY

- Scaled up electrolyte extraction with DEC to about 300 mL of solvent and 500 mL of packed electrodes
  - Yield was 1.8 g of crystals (mostly $LiPF_6$ solvate).
  - Losses in transfers and drying process.
  - Drying to solids is challenging at larger scales.
    - Possibility for concentration only.
  - Recovered DEC from the process and will be determining its purity for reuse.

ELECTROLYTE RECOVERY - COST MODELING

- Primary cost drivers are electricity and cost of DEC solvent
  - Highest energy user is the evaporator
    - Current model is only an estimate of the energy usage and capital costs for the evaporator



Revenue **$1.41/kg** battery recycled

Total Cost **$0.81/kg** battery recycled

Profit **$0.60/kg** battery recycled

SUPERCRITICAL CO_2 EXTRACTION

- CRADA with industrial partner was established
- Supercritical CO_2 may react with the electrolyte materials in a unique way -
  - Past literature reports have indicated that supercritical CO_2 is very inefficient for extracting $LiPF_6$ on its own
  - Adding co-solvent can substantially improve efficiency
- Potentially supercritical CO_2 can be used to simply purify the $LiPF_6$ concentrate
  - sc-CO_2 will remove impurities, leaving pure salts
  - This would reduce the volume of material treated, improving the costs for extraction compared to extracting all the materials with sc-CO_2

Extraction vessel

Separator vessel 1

Separator vessel 2

SOLVENT-BASED ELECTRODE RECOVERY

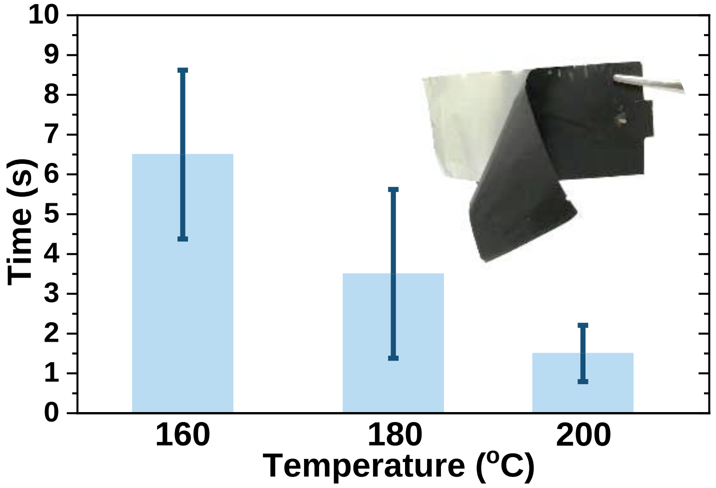
Project Description:

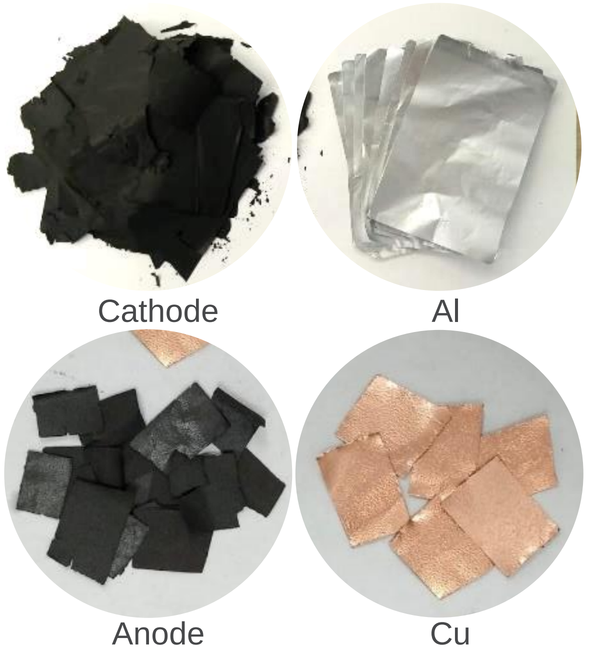
- Demonstrate wet-chemical approaches for separating the black mass from metal foils by either solvating the PVDF binder (SolveY process) or weakening its binding with laminates (SolveX process)
- Use green solvents that are inexpensive, nontoxic, do not cause water and/or air pollution, and do not damage active materials and current collectors

Project Goal:

- Develop efficient solvent-based recovery processes for the separation of black mass from current collectors
- Recover active materials without damaging their structure, morphology, and performance
- Reclaim current collectors without any corrosion to them

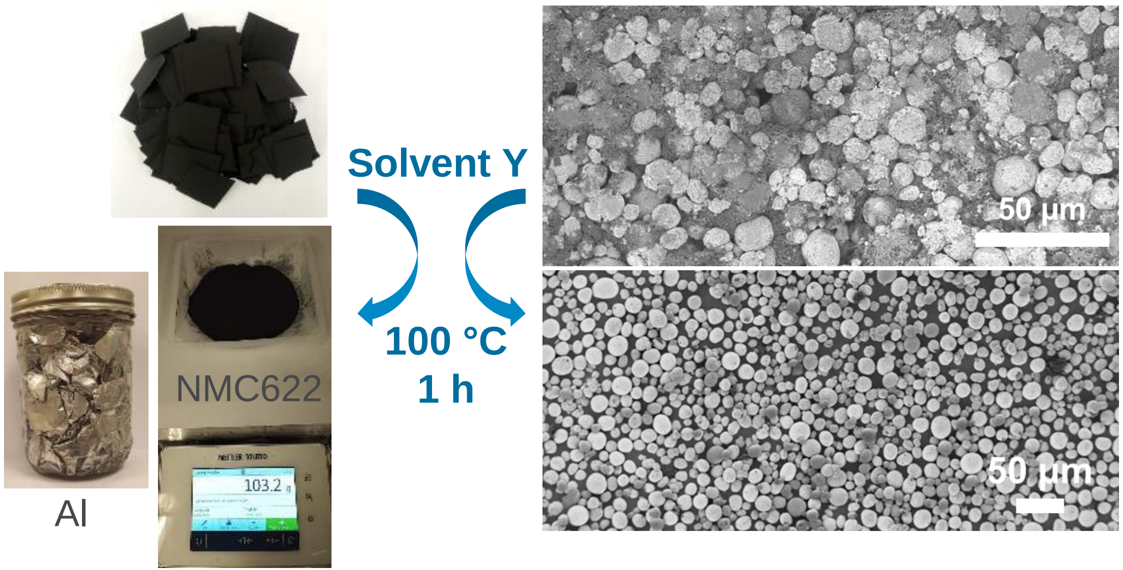
SOLVENT-BASED ELECTRODE RECOVERY

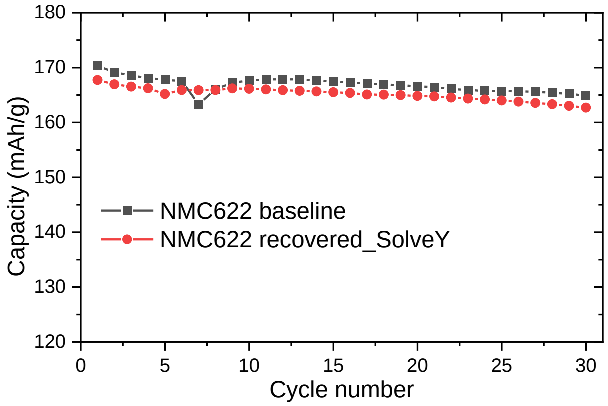




- Developed a SolveX process to delaminate electrode materials including both cathodes and anodes from current collectors
- Demonstrated highly efficient delamination in the green solvent X with 100% separation efficiency in seconds
- Proved that SolveX process does no damage to the active materials, does not corrode the current collectors, and has negligible influence on electrochemical performance

SOLVENT-BASED ELECTRODE RECOVERY





Similar capacity retention at 1 C rate

- Developed a SolveY process to fully separate active electrode materials from current collectors and PVDF binder in a green solvent Y
- Proved that the SolveY process shows no damages to the active materials, no corrosion to the current collectors, and slight reduction in electrochemical performance

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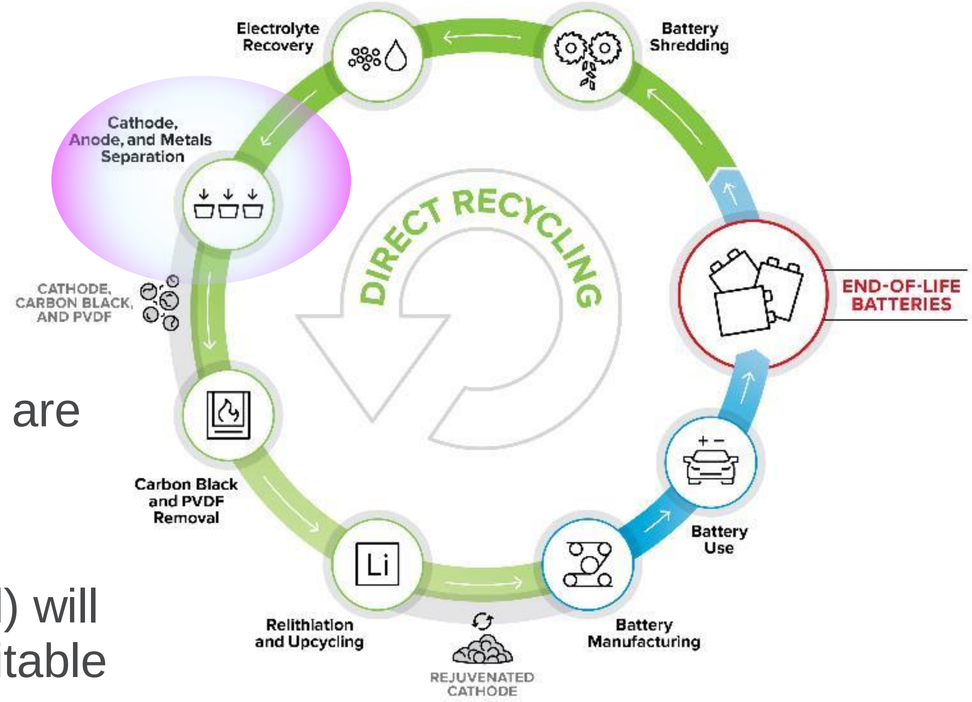
ANODE/CATHODE SEPARATION AND PURIFICATION

Project Description

Pouch Cell

Contents

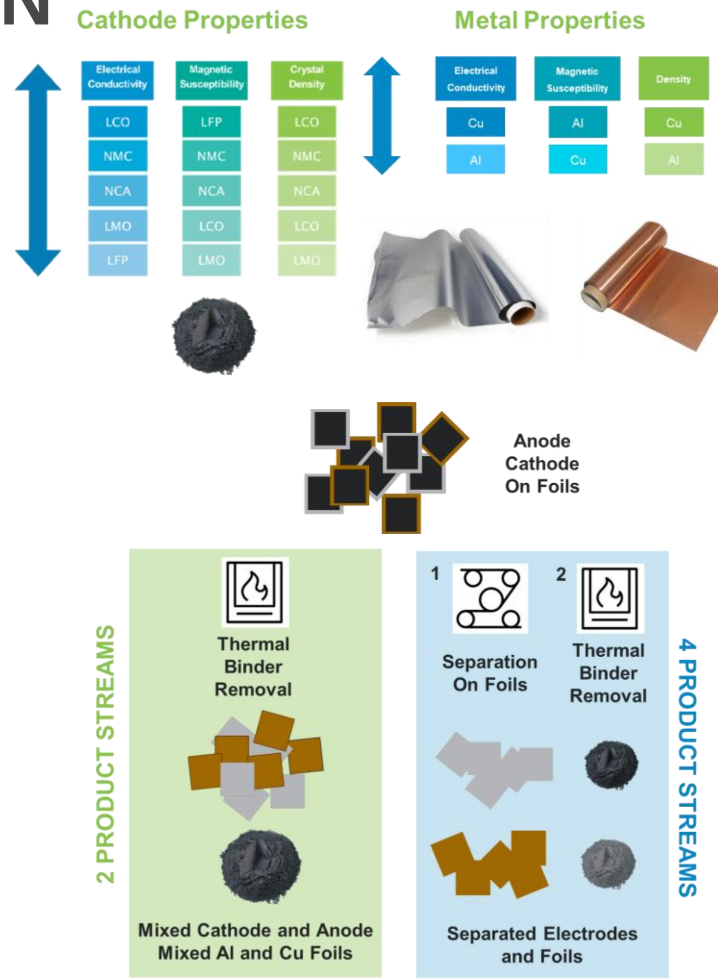
- After battery shredding, component separation, and electrolyte recovery we are left with electrodes
- Production of multiple clean streams of material (cathode, anode, and Al/Cu foil) will make direct battery recycling more profitable



ANODE/CATHODE SEPARATION AND PURIFICATION

Project Goals

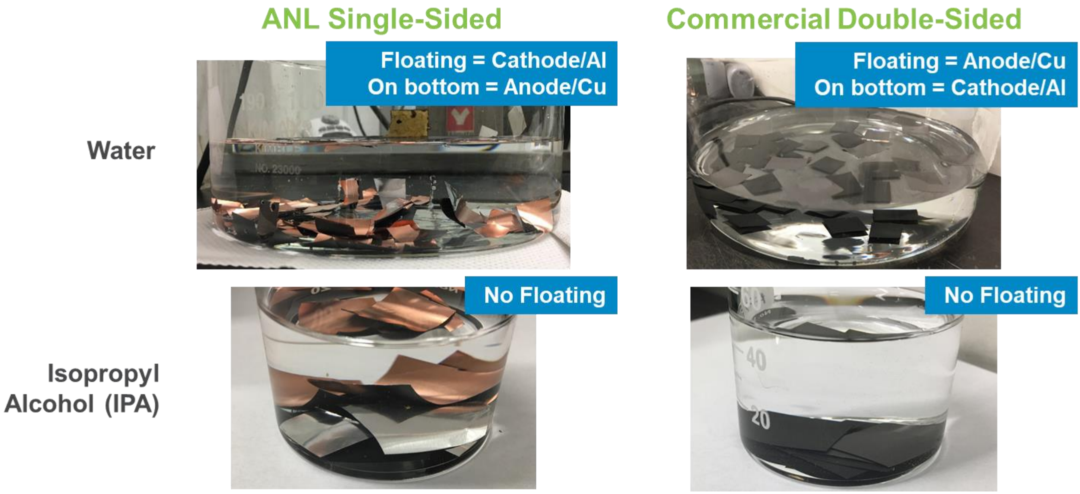
- Separation technologies will be explored for separation of anode, cathode, and metals where:
  - The anode and cathode are powders
  - The electrodes are still attached to the foils
- The properties of the powders or metals will be exploited to determine the most efficient mode of separation (e.g., density or gravity based, magnetic, triboelectric, eddy current, etc.)



ANODE/CATHODE SEPARATION AND PURIFICATION

Flotation as a Method of Electrode Separation

- IPA was chosen as a non-aqueous solvent that is less likely to leach lithium from the surface of Ni-rich cathode material
- Flotation properties differ for single (floating cathode) *vs*. double sided (floating anode) laminates which is due to tradeoff between hydrophobicity of the carbonaceous anode and density of the copper foil
- IPA had no flotation effect with laminates



FOCUS AREA SUMMARY SLIDE – ACCOMPLISHMENTS AND RESULTS

- Several options have been considered and investigated for safe shredding of cells.
- Four different commercial shredder systems were tested to size reduce dry, pristine 40 Ah pouch cells.
- Electrochemical performance of recovered electrolyte salt determined to be similar to that of pristine material.
- EverBatt analysis indicates potential profitability of electrolyte recovery and recycling.
- Robust, very efficient solvent-based delamination of electrodes has been developed.
- Flotation as a method of electrodes (anode/cathode) separation has been investigated.
- For more information please see www.recellcenter.org, where our Quarterly Reports are posted.

COLLABORATION AND ACKNOWLEDGEMENTS

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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

Take a picture of this slide

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Phone
(866) 226-4650
Meeting ID: 749 203 749

Slido Q/A website

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Event Code
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