Battery Safety Science Webinar Series

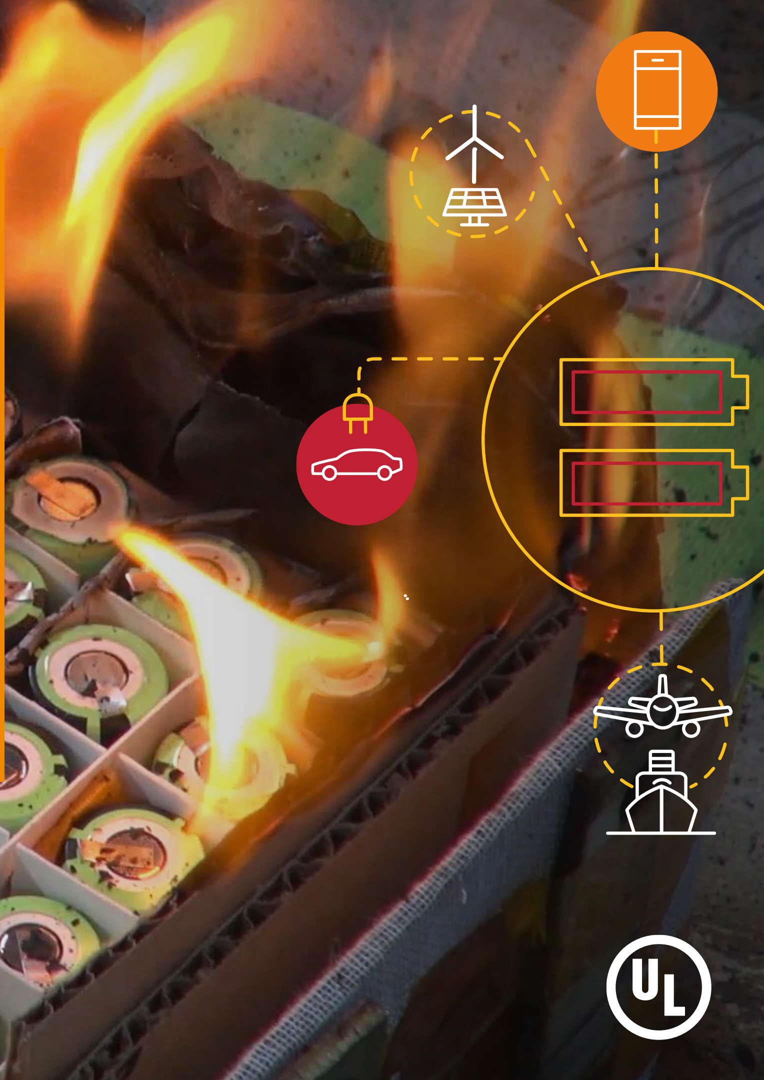
Advancing safer energy storage through science

July 19, 2021

How Analysis Helps Guide Battery Recycling R&D at the ReCell Center

Host **Dr. Tapesh Joshi**

Presenter **Dr. Linda L. Gaines**
Chief Scientist, ReCell Center for Advanced Battery Recycling
Transportation Systems Analyst, Argonne National Laboratory



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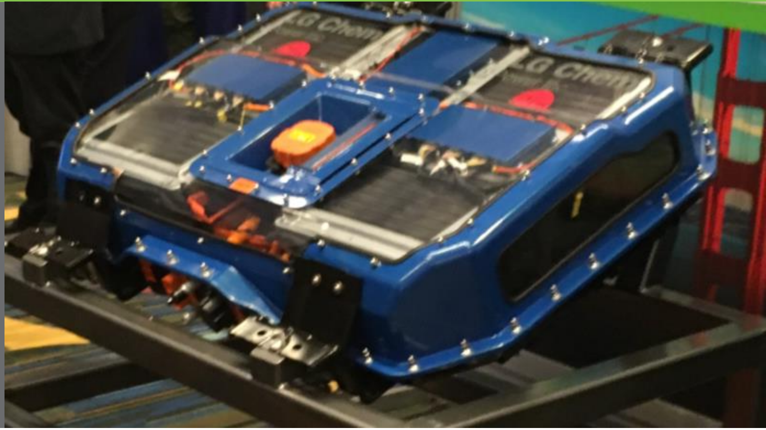
ReCell
ADVANCED BATTERY RECYCLING

HOW ANALYSIS HELPS GUIDE BATTERY RECYCLING R&D AT THE RECELL CENTER



LINDA GAINES
Transportation Systems Analyst
Argonne National Laboratory
lgaines@anl.gov

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WHAT IS THE RECELL CENTER?

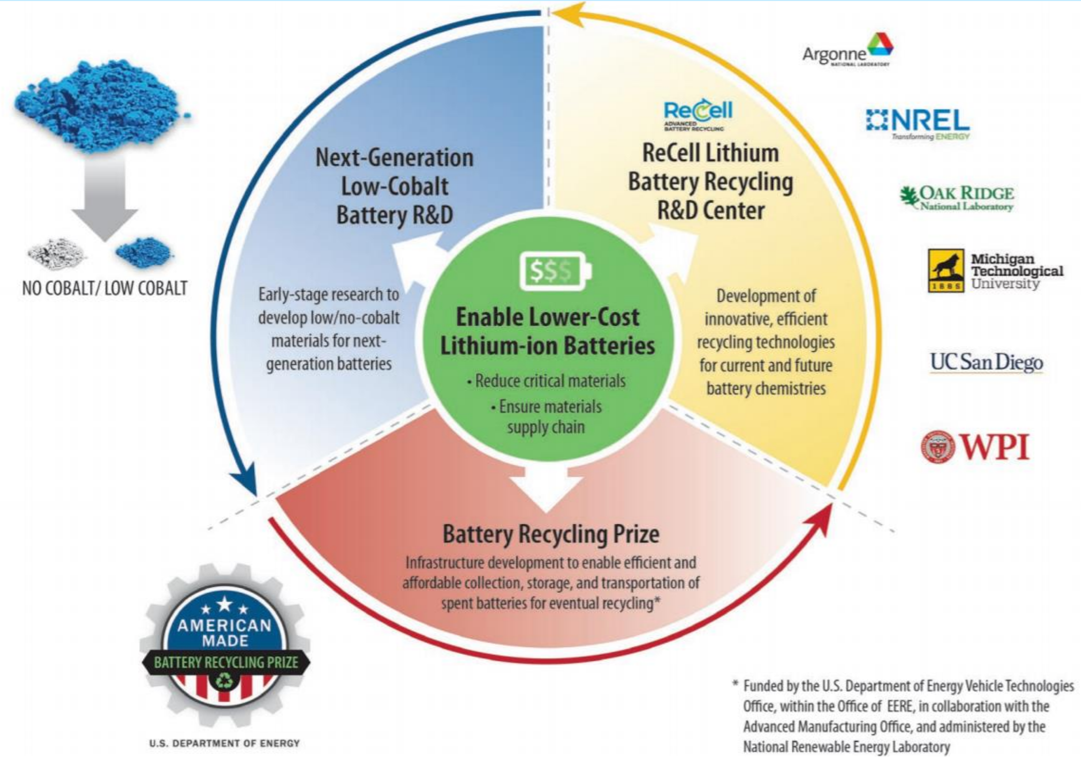
The center is a $15 million collaboration of three national laboratories and three universities, led by Argonne, advised by industry, and funded by the DOE Vehicle Technologies Office. It is tasked to develop a cost-effective process for recycling lithium-ion batteries within three years.

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

RECELL IS PART OF DOE’S CRITICAL MATERIALS PLAN

to reduce the cost of EV battery packs to <$150/kWh with technologies that significantly reduce or eliminate the dependency on critical materials (such as Co) and utilize recycled material feedstocks.



* Funded by the U.S. Department of Energy Vehicle Technologies Office, within the Office of EERE, in collaboration with the Advanced Manufacturing Office, and administered by the National Renewable Energy Laboratory

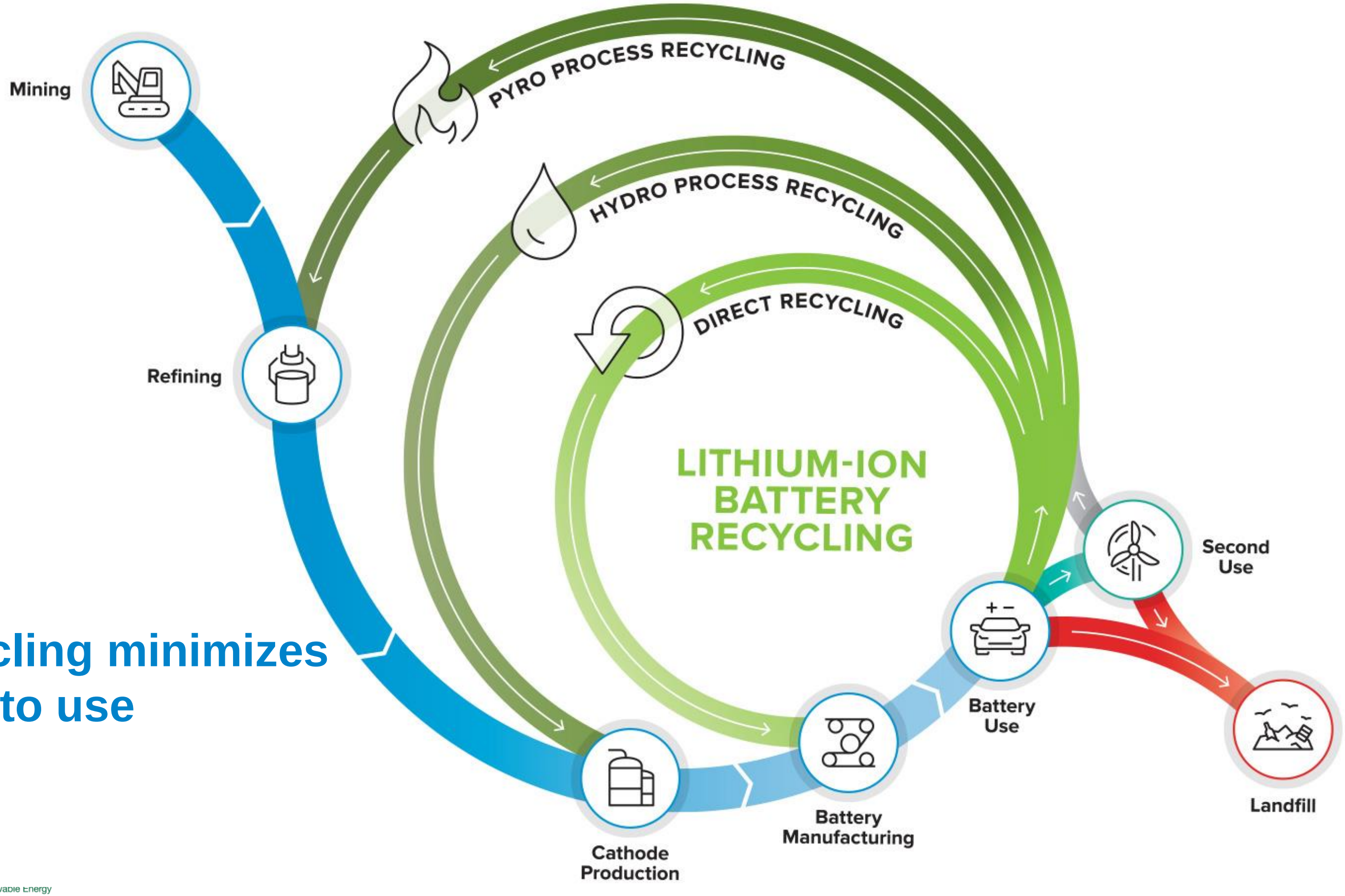
WHERE DO LI-ION BATTERIES END UP NOW?

Little or no actual recycling is consummated in the US

- Essentially all EOL batteries now are from consumer electronics
  - A few EV batteries are sold on sites like e-Bay to the DIY market
- Some are turned into Best Buy and get recycled in Canada (>2000 t/y)
- Some are in your house
- A few get thrown in the garbage
- Many are still inside a device
- Phones and laptops are reused in the US or **exported**; vehicles, too
- Some devices to be reused get new batteries and the old ones recycled
- Only anecdotal and partial information is available

ReCell analysts will work with Prize winners to verify benefits

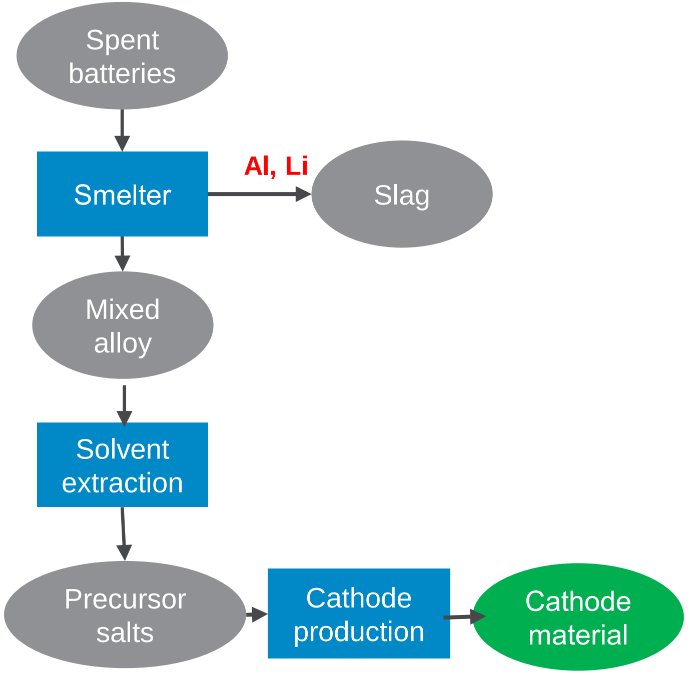
LI-ION BATTERY LIFECYCLE



Direct recycling minimizes steps back to use

PYROMETALLURGICAL PROCESSING IS HIGH TEMPERATURE AND LARGE SCALE

Umicore pilot plant is designed to process 7,000 tonnes per year



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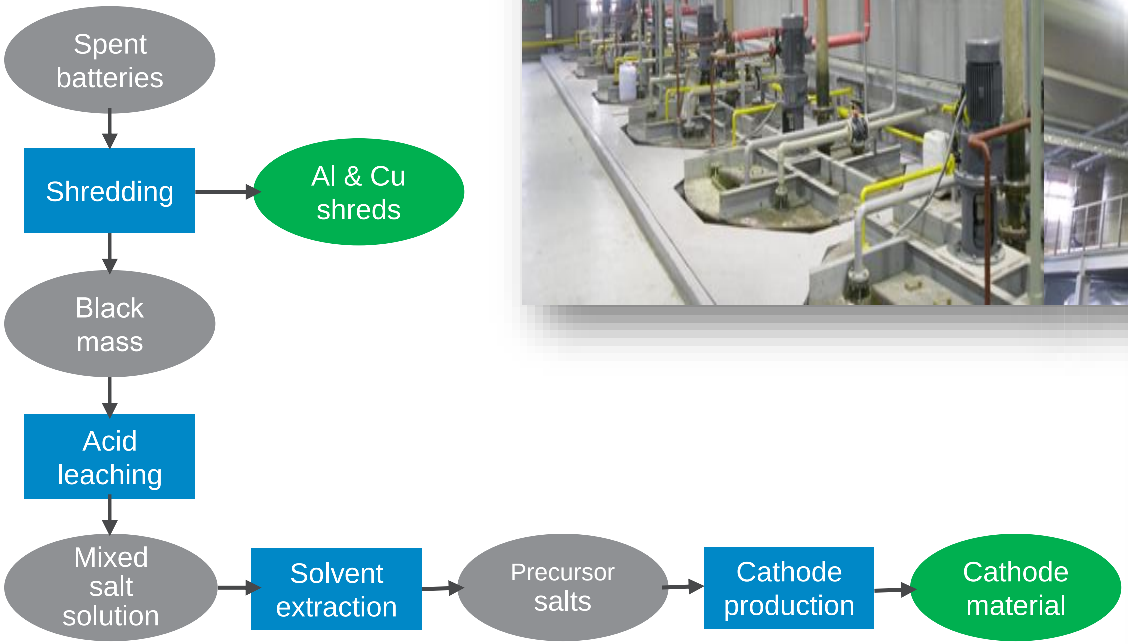
HYDROMETALLURGY

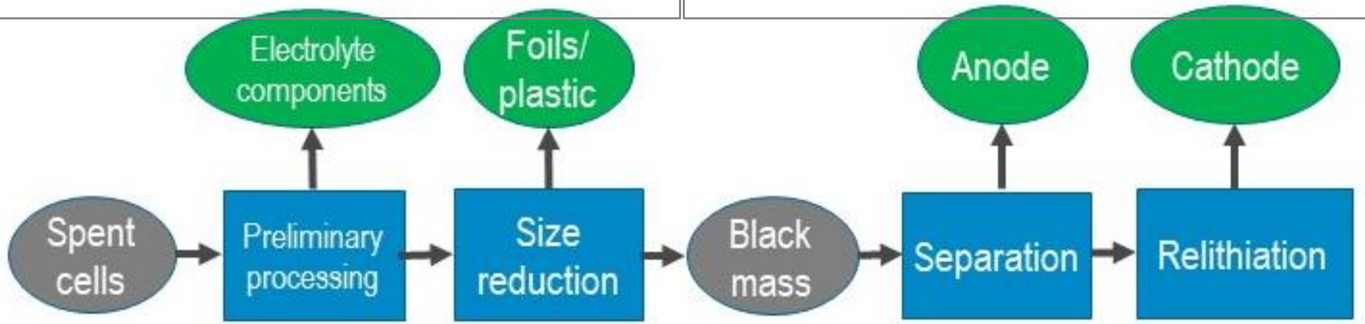
Materials are dissolved in acid and components are separated



Courtesy of SMCC

DIRECT RECYCLING is the recovery, regeneration, and reuse of battery components directly without breaking down the chemical structure.

BENEFITS	CHALLENGES
▪ **Retains valuable chemical structure** ▪ Enables economic recovery of more materials ▪ Could be used now for manufacturing scrap ▪ Low temperature, low energy ▪ Avoids most impacts of virgin material production	▪ Separating multiple cathode chemistry particles ▪ Product may be obsolete formulation ▪ Degradation may limit repeats ▪ Buyer must be assured of quality ▪ Not demonstrated on industrial scale



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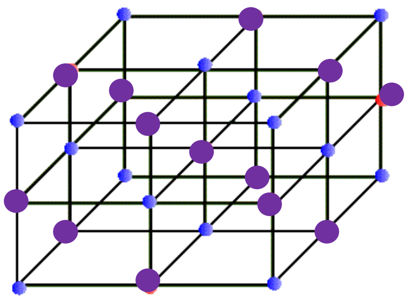
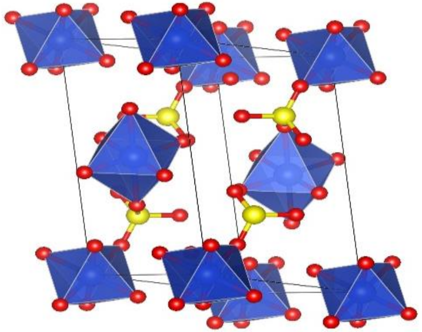
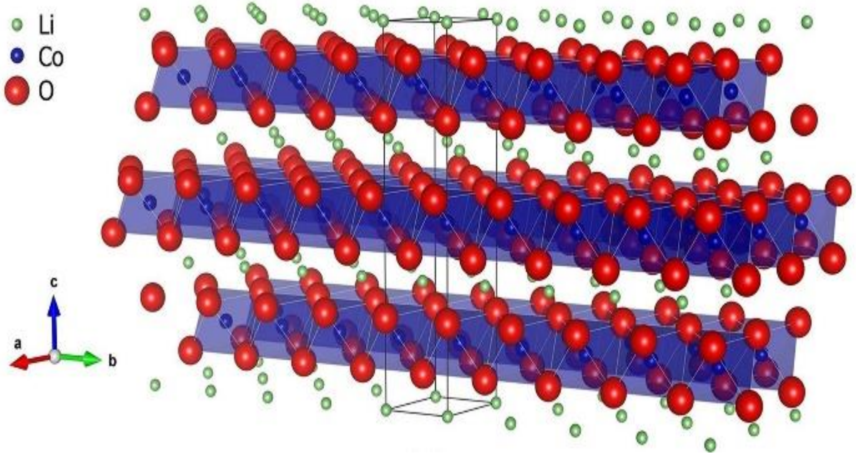
DIRECT RECYCLING RECOVERS HIGHLY STRUCTURED MATERIAL







Cobalt has a simple cubic structure; nickel impurities can substitute

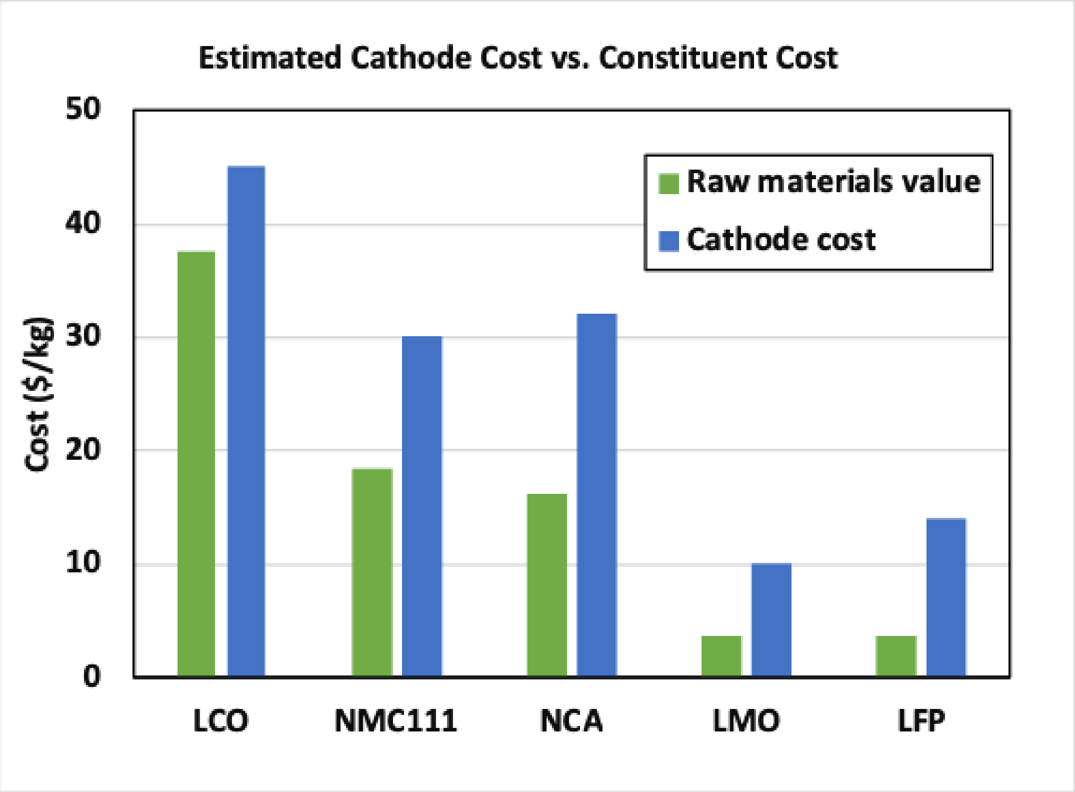
Cobalt sulfate is octahedral

LCO has an ordered layered structure

CATHODE VIABILITY IS KEY TO ECONOMICS

FOR CATHODES WITH REDUCED COBALT CONTENT

Cathode materials are valuable, even if constituent elements aren't



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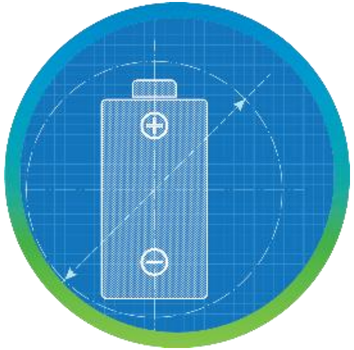
RESEARCH AT CENTER HAS FOUR FOCUS AREAS

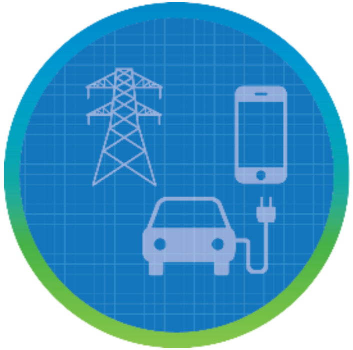
DIRECT CATHODE RECYCLING

OTHER MATERIAL RECOVERY



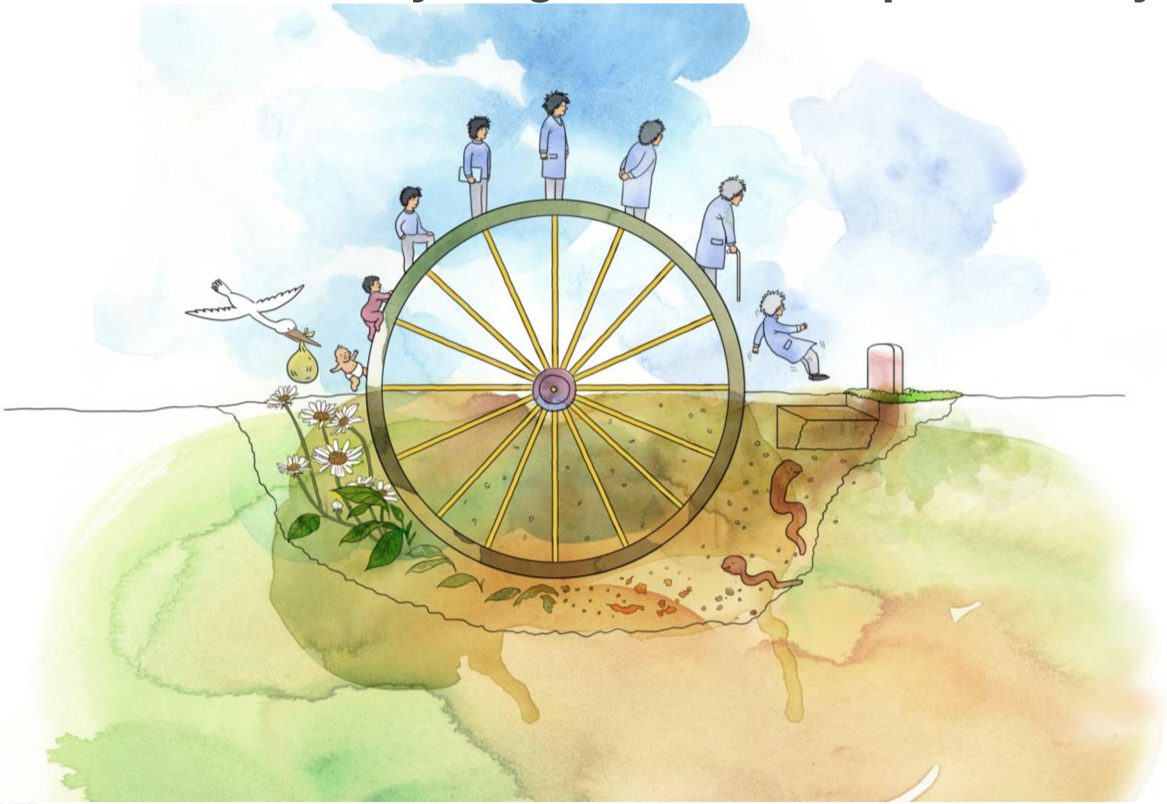
DESIGN FOR SUSTAINABILITY

MODELING AND ANALYSIS



LIFECYCLE ANALYSIS EVALUATES PROCESS IMPACTS

of a product's life cycle, from raw material acquisition through production, use, end-of-life treatment, recycling, and final disposal if any.



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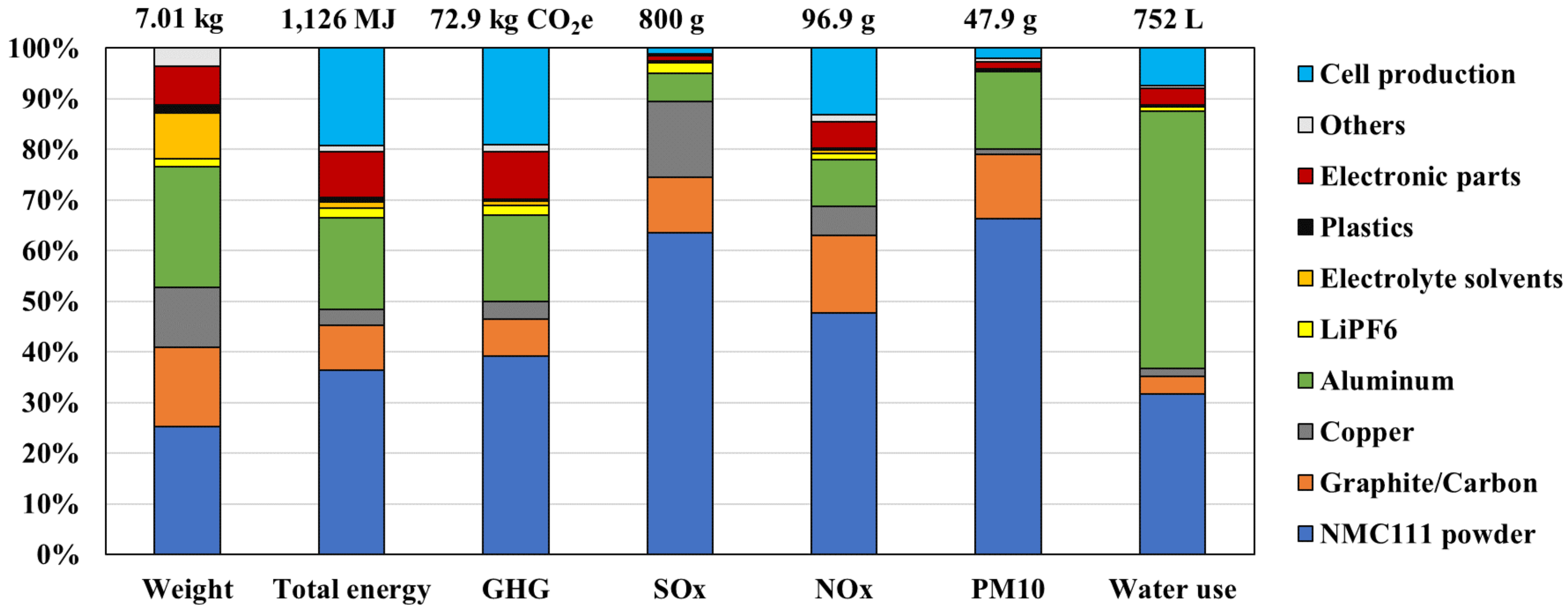
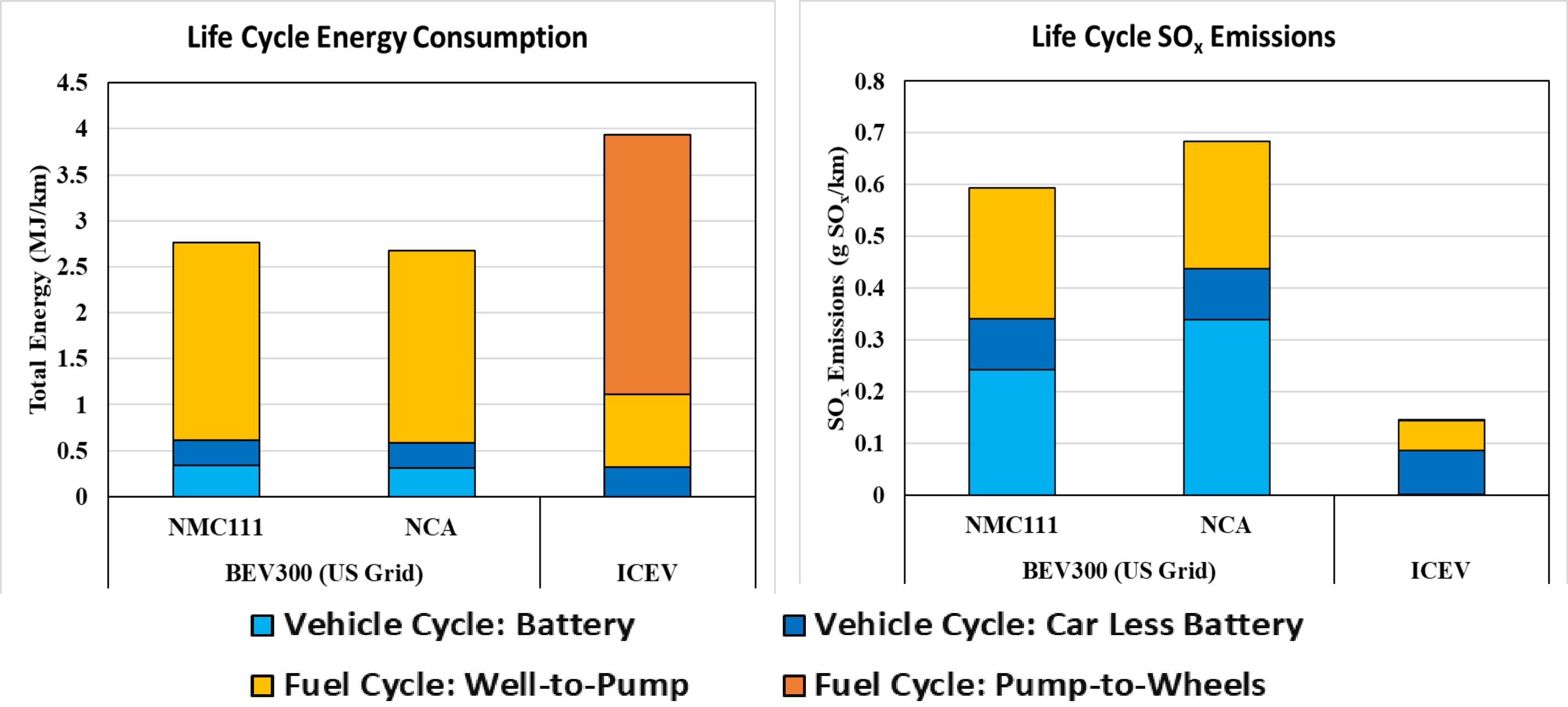
CRADLE-TO-GATE ENVIRONMENTAL IMPACTS: 1KWH NMC111 CELLS

Cathode, production energy, and aluminum are notable contributors



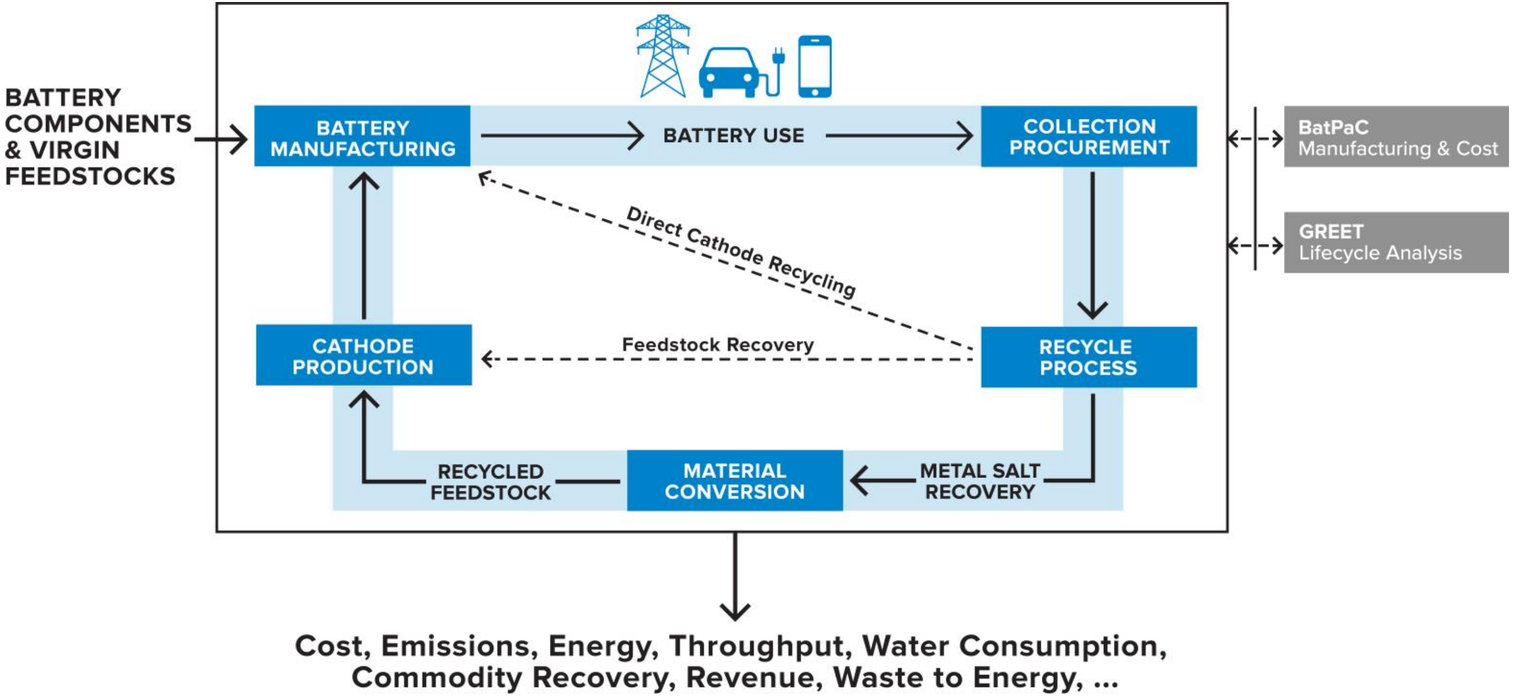
BATTERY CONTRIBUTES LITTLE TO EV LIFECYCLE ENERGY

but significantly to SO_x emissions



EVERBATT MODEL IDENTIFIES THE MOST EFFICIENT AND ECONOMIC PROCESSES

There are many potential recycling pathways for batteries. Modeling and analysis can guide process development without the need to actually try all options.



EverBatt received a 2019 R&D100 award.

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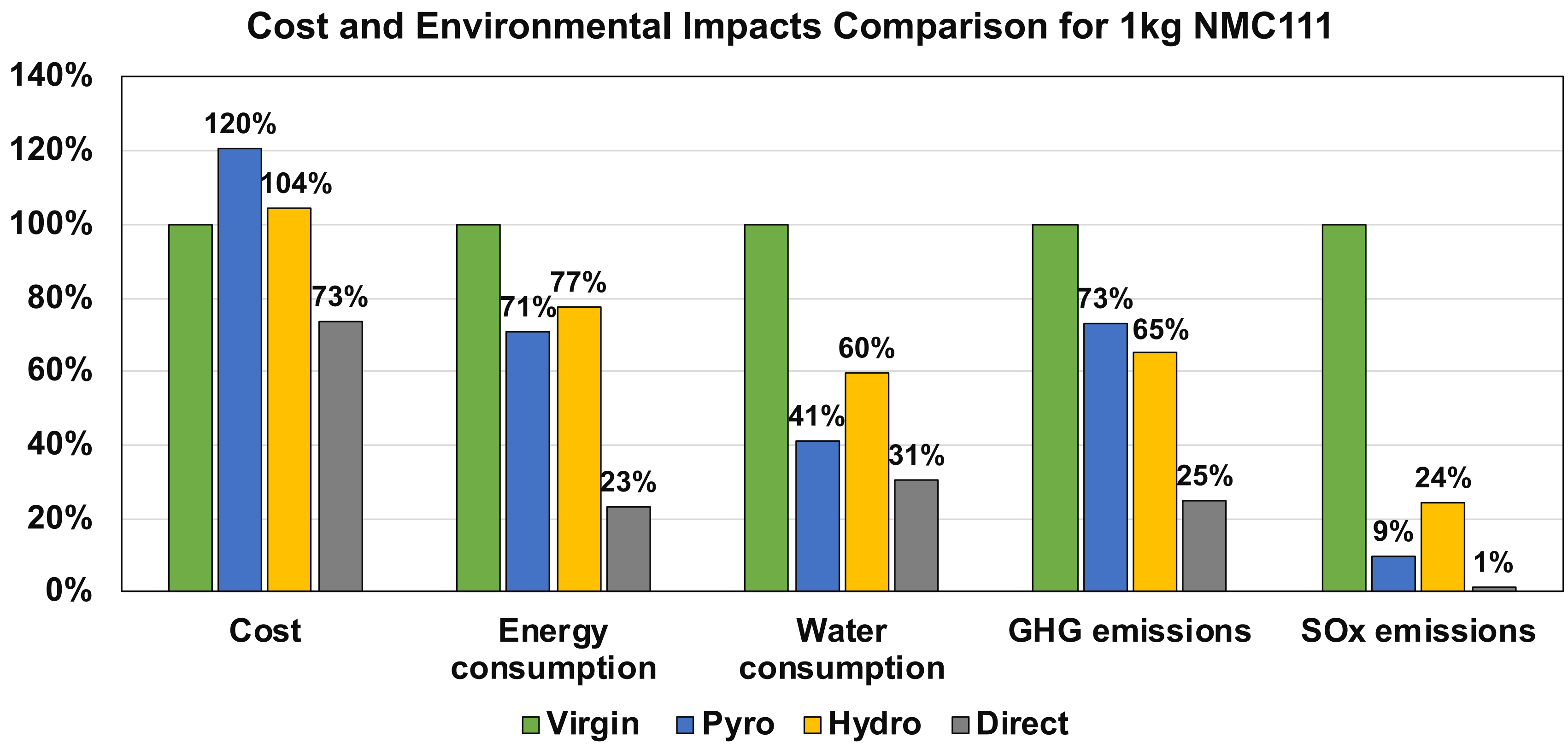
DIRECT RECYCLING HAS LOWEST IMPACTS



"HUB AND SPOKE PROCESSING" COULD REDUCE TOTAL RECYCLING COST

Transportation of hazardous material is costly

- Collected batteries are sent to distributed pretreatment facilities to be neutralized.
- Black mass from pretreatment is transported to one or a few recycling facilities to recover valuable materials.
- Pretreatment plants need to be spread to reduce transport cost but large enough to enjoy economies of scale

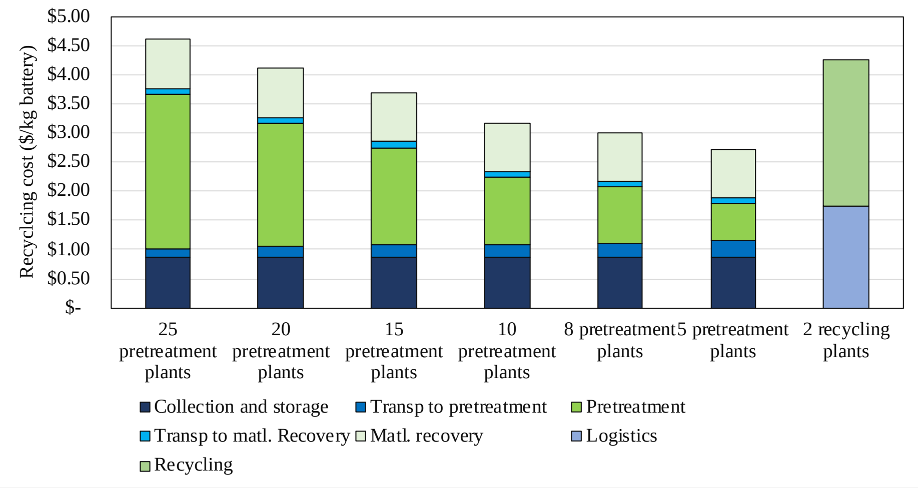


Figure 1 shows the total costs of deploying multiple distributed pretreatment facilities with a combined capacity of 50,000 T/y, plus 2 hydrometallurgical facilities with a combined capacity of 30,000T/y, compared to that for 2 hydrometallurgical facilities with a combined capacity of 50,000 T/y without pretreatment.

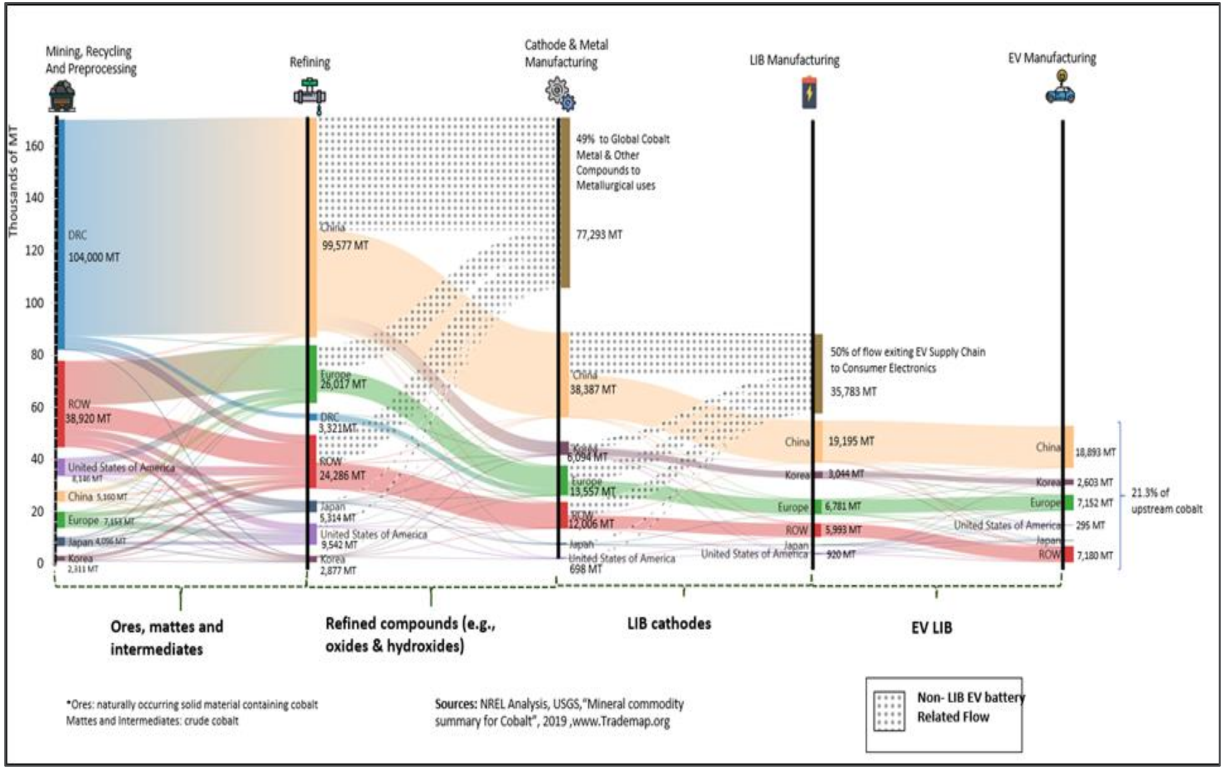
SUPPLY CHAIN ANALYSIS TRACKS FLOWS OF KEY MATERIALS

The majority of battery materials are refined in China

NREL is analyzing the material and component supply chain for Li-ion manufacturing and recycling to determine the dynamic factors driving the economic viability of this nascent industry.

Material availability, supply shocks, and technology adoption all impact the success of battery recycling and the electrification of the transportation sector.

Global Flow of Cobalt



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HOW MUCH MATERIAL IS THERE?

How many NMC811 car batteries could you make using reserves?

Element	kg/kWh*	kg per car @85 kwh/car	US reserves (kT)**	World reserves (KT)**	number for US (millions)	global number (billions)
Cobalt	0.08	6.8	53	7100	7.8	1.0
Nickel	0.6	51	100	94,000	2.0	1.8
Lithium	0.1	8.5	750	21,000	89	2.5
Manganese	0.07	5.95	230,000	1,300,000	38,656	219

* from Shabbir Ahmed 2/8/21;
NMC811-Graphite System from BatPaC 4.0 1Oct2020.

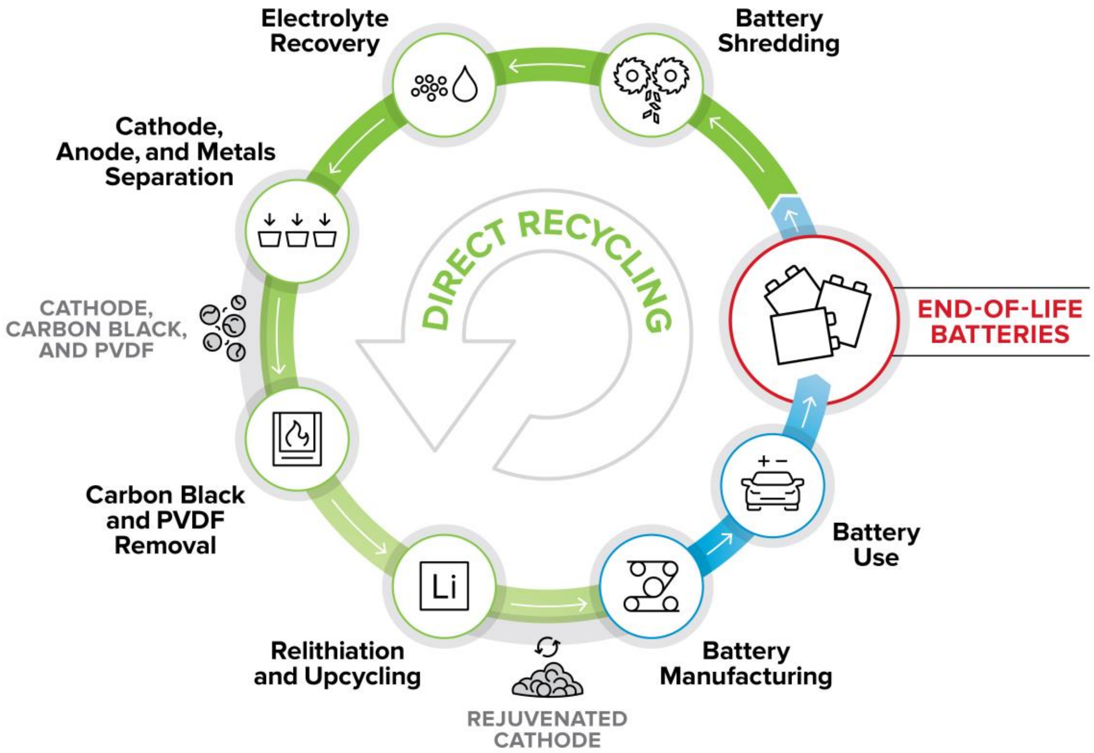
** USGS Mineral Commodity Summaries 2021

Not enough, but recycling can help… eventually.

UNIT PROCESS ORDER WILL BE OPTIMIZED

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

Typical Direct Recycling Process Flow



CELL PRE-PROCESSING IS THE FIRST STEP

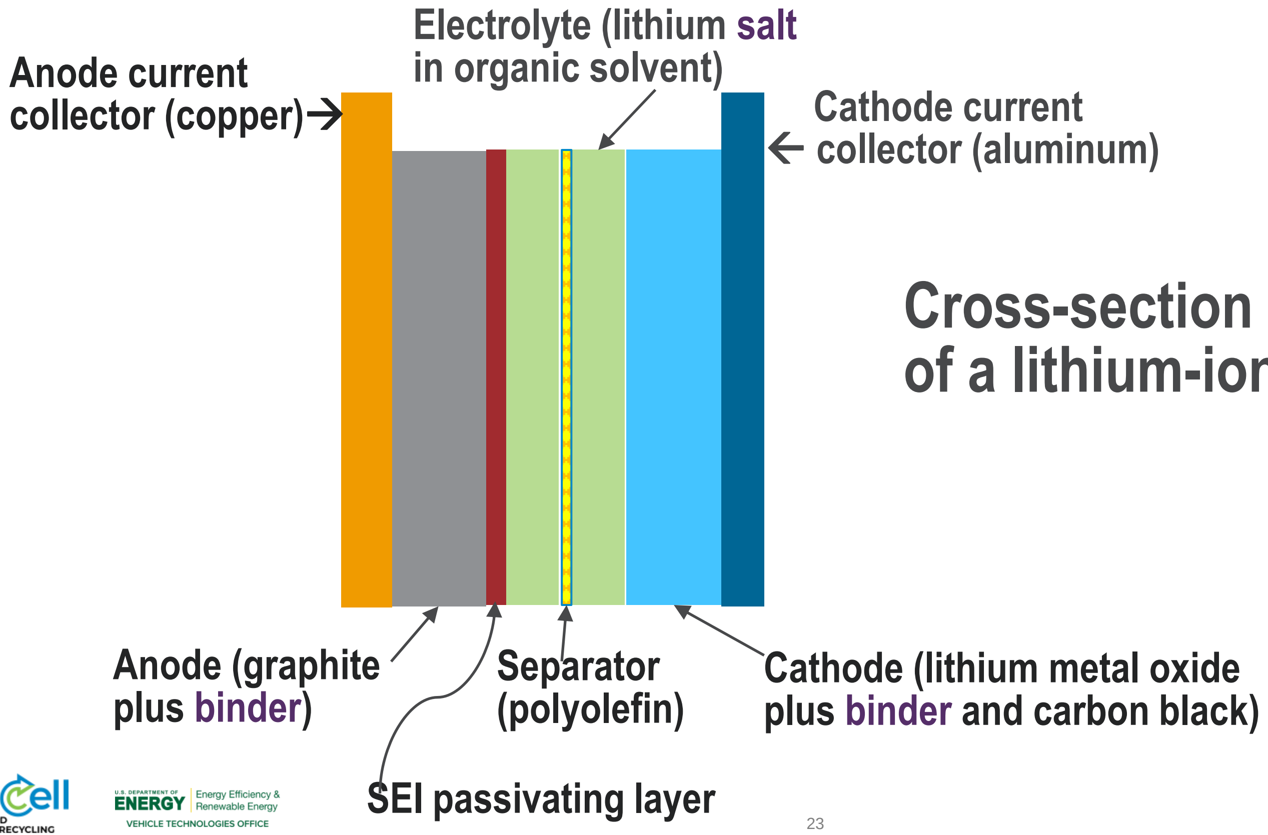
Disassembly/ Dismantling

Shredding/Crushing/ Milling/Punching

- Safely and cost-effectively size reduce cells or scrap to produce black mass
- Minimize creation of fine particle contamination to be carried forward into the direct recycling process

PROCESSING REQUIRES MANY SEPARATIONS

Commercial technologies lose some of the materials



Cross-section of one layer of a lithium-ion cell

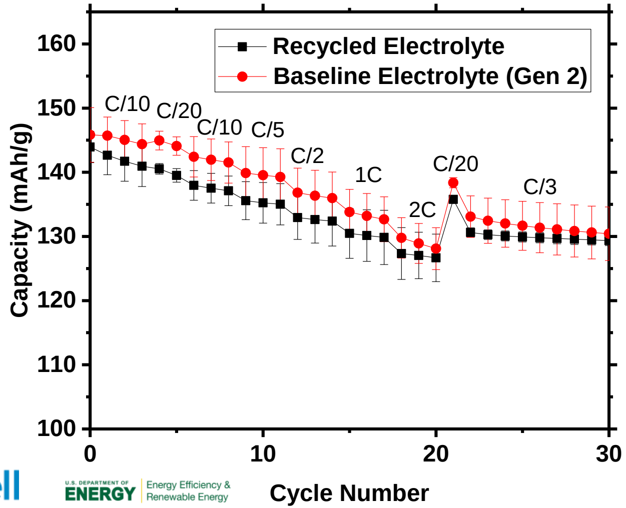
REVISED DESIGNS COULD ENABLE RECYCLING

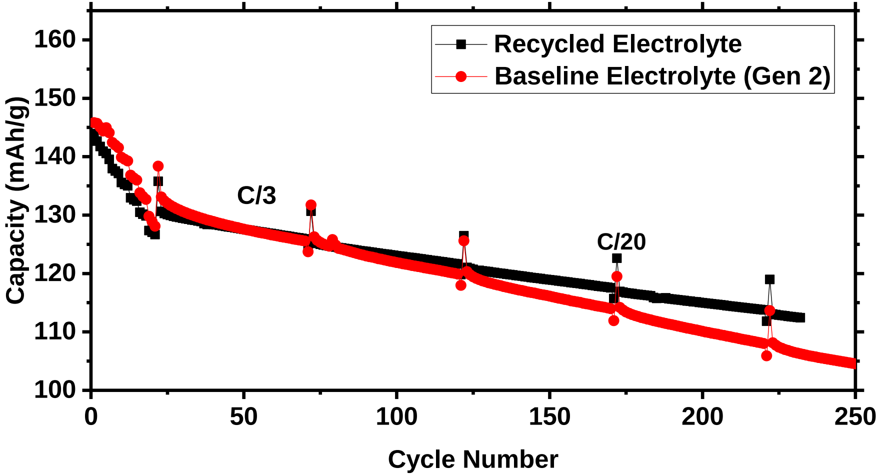
Is there anything you can do at the basic research level?

- Uniformity and standardization would enable robotic disassembly
  - Make varied packs from standard modules
  - Limit cell form factors; use larger cells
- Limiting material choices would reduce separation steps needed
- Reversible joining may raise manufacturing cost
  - Adhesives and binders you can dissolve
  - Thermoplastics not thermosets
  - Nuts and bolts instead of welds
- Simpler structure is easier to take apart
  - Integrate pack with vehicle structure: a help or hindrance?
- These changes would also facilitate repurposing and repair

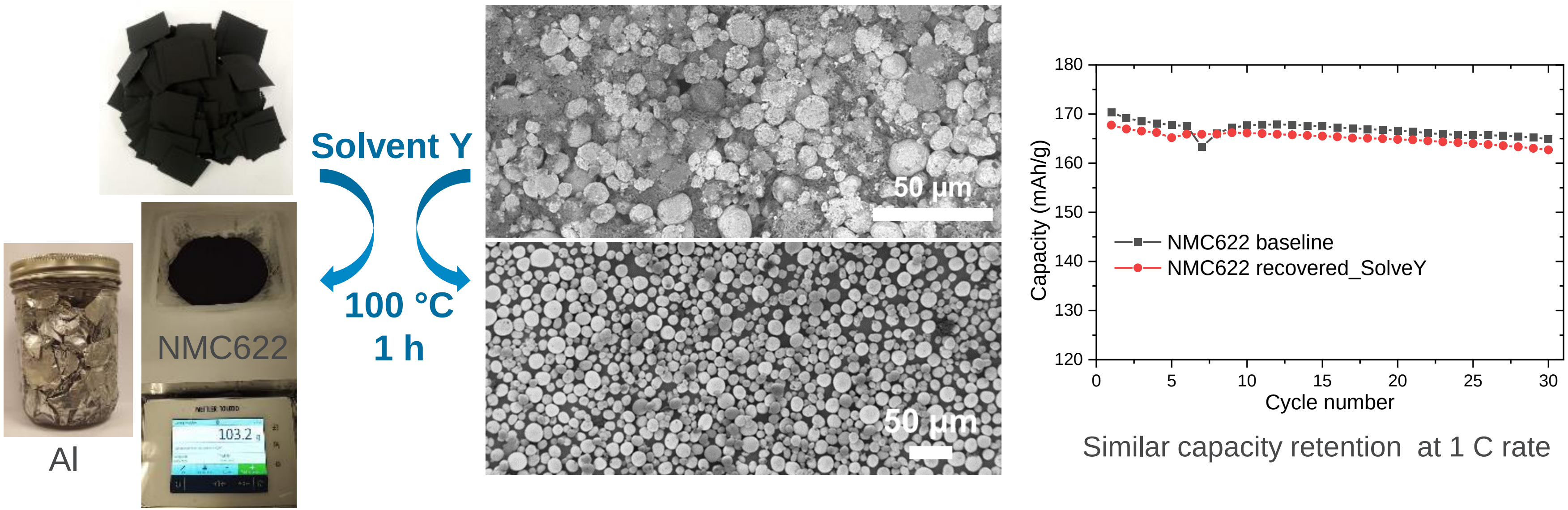
RECOVERED ELECTROLYTE SALT PERFORMS WELL IN FULL CELLS

- 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.





SOLVENT RECOVERS ELECTRODES AND BINDER



Similar capacity retention at 1 C rate

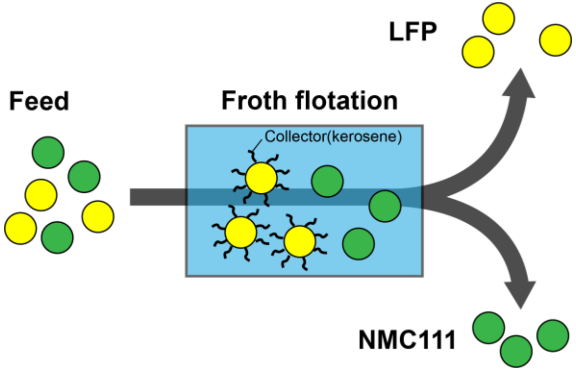
- Developed a SolveY process to fully separate active electrode materials from current collectors and PVDF binder in a green solvent.
- Proved that the process does not damage the active materials or corrode the current collectors. Slight reduction in electrochemical performance.

POWDERS ARE SEPARATED BY FROTH FLOTATION

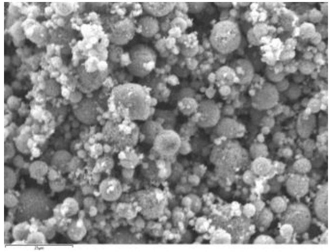
Can separate cathode from anode or one cathode from another

- LFP separated from NMC111 (1:1 by weight) by froth flotation using kerosene as collector
- Collector effectively increases the hydrophobicity of LFP particles, and thus increases the separation efficiency to ~99%

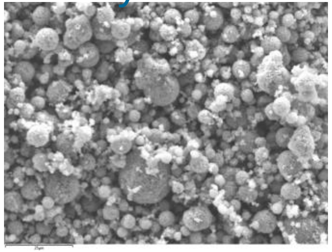
Product	Weight (g)	Composition (wt. %)	
		%LFP	%NMC11
Froth	9.25	99.18%	0.82%
Tailing	9.04	1.08%	98.92%



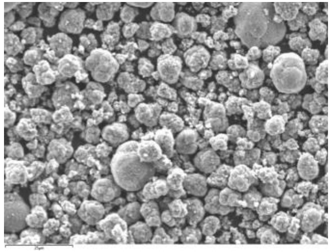
Pristine LFP



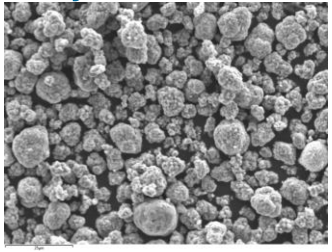
Recycled LFP



Pristine NMC111



Recycled NMC111

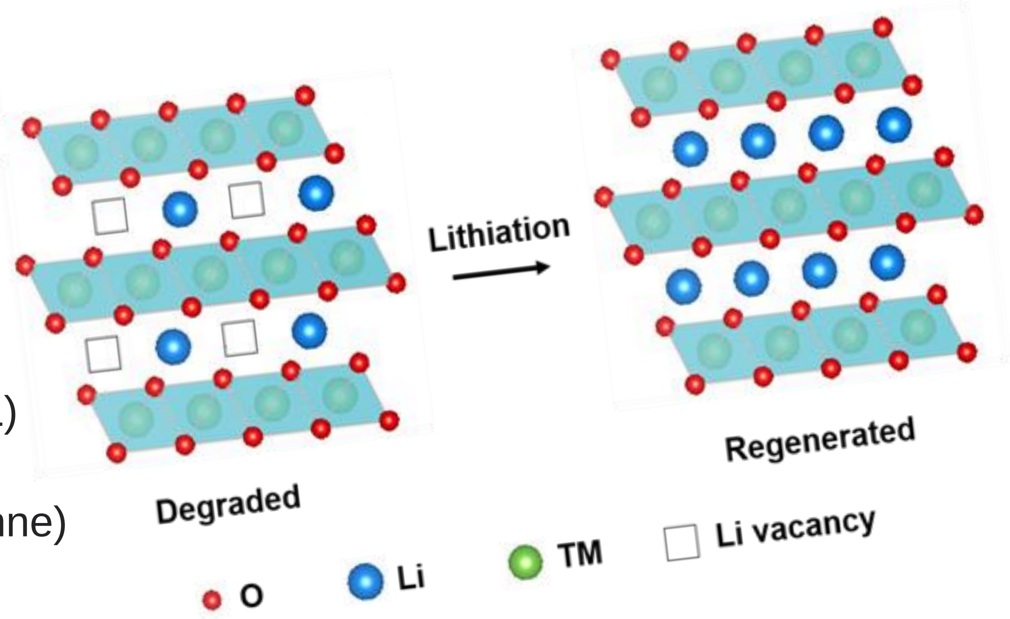


DIRECT RECYCLING UPGRADES CATHODE

Product must be as good as new… or better

Several phenomena contribute to the gradual drop in lithium-ion battery performance, including surface degradation, cathode instability, reactivity with organic electrolyte components, and surface films. These phenomena need to be reversed and performance restored.

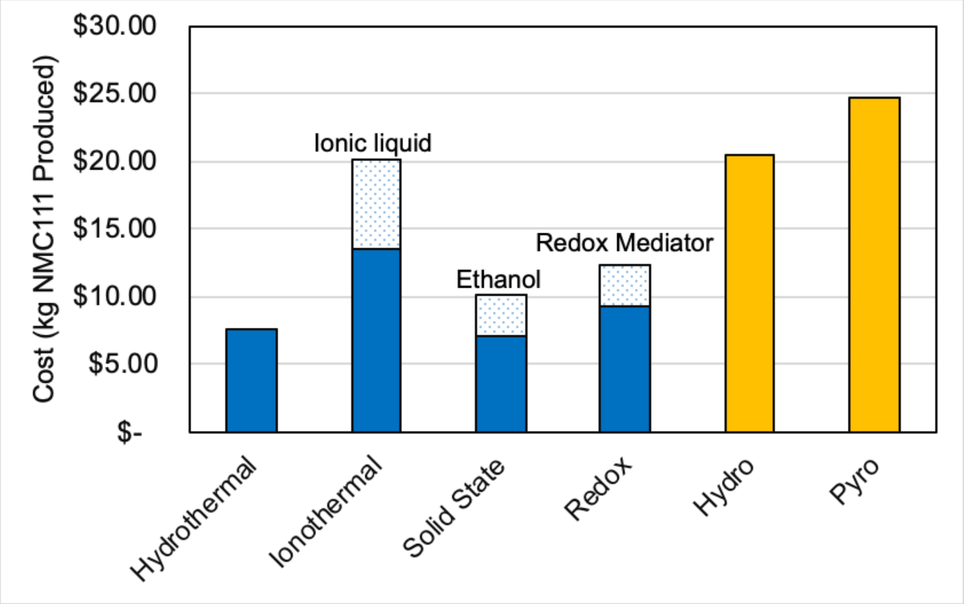
- **Relithiation**
  - Electrochemical (NREL)
  - Solid State (Argonne)
  - Hydrothermal (UCSD)
  - Ionothermal (ORNL)
  - Redox Mediated NREL)
  - Roll to Roll Processing (NREL)
- **Upcycling**
  - Compositional Change (Argonne)



MATERIALS ARE KEY TO RELITHIATION COSTS

Further research targeted at reducing high material costs

- Replace or reduce use:
  - Ionic liquid (ionothermal)
  - Redox mediator (redox)
  - Ethanol (solid state)
- Hydrothermal lowest cost
- Solid state could potentially be lower
- Results for commercial-scale plant; scaling will not change ordering



Dashed bars represent potential cost reductions by closed-loop recycling of key materials; blue bars represent other costs for 10,000 T/y direct recycling plants; yellow bars represent costs for 10,000 T/y pyrometallurgical (pyro)/hydrometallurgical (hydro) recycling plants plus costs to convert recovered materials into cathode powder.

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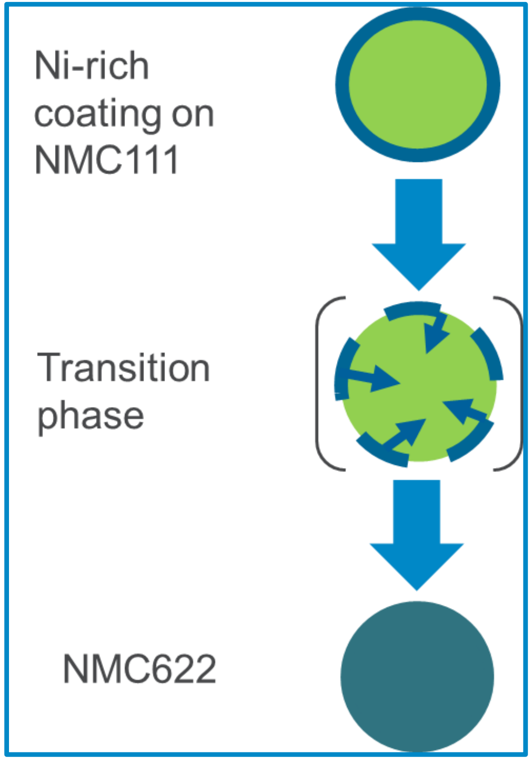
UPCYCLING WILL REVAMP OBSOLETE CATHODE

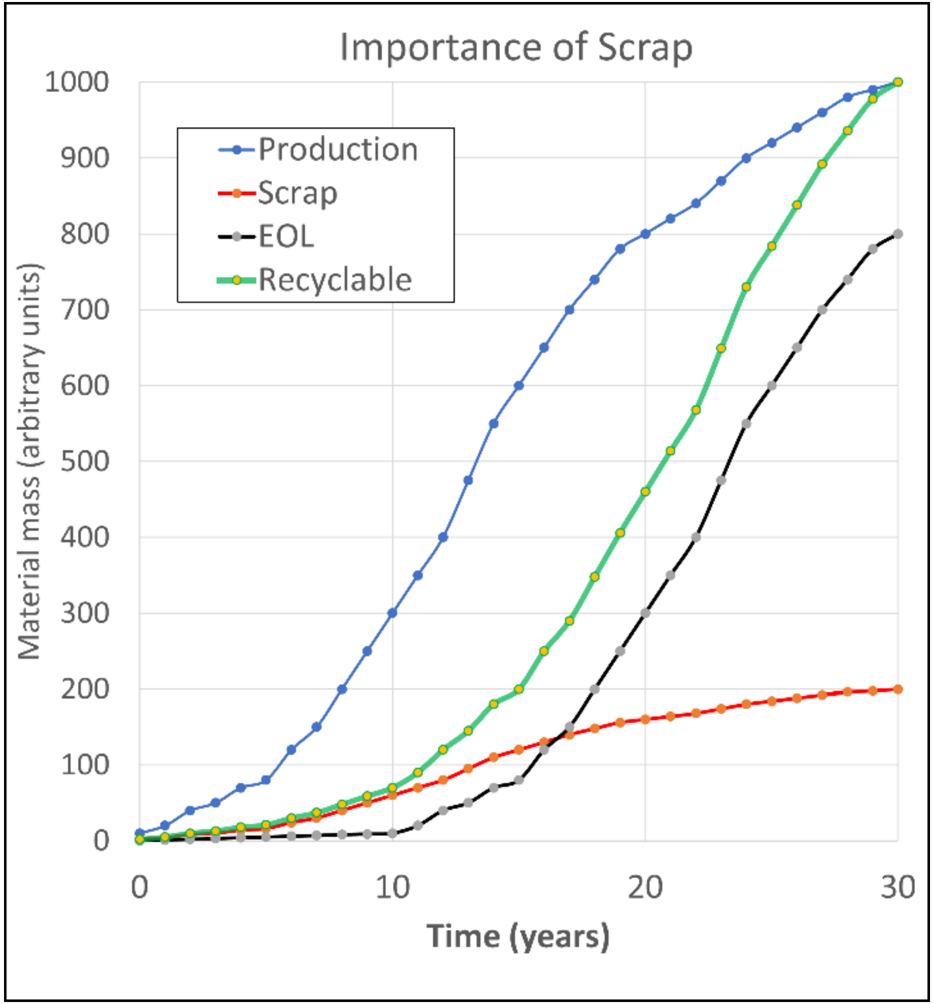
Processes are being developed to alter cathode stoichiometry

- Developing methods to convert NMC111 to NMC622
  - Intermediate phases may be formed
- Relithiation must precede upcycling
- Coating method affects electrochemical performance
  - Developed a low solvent volume method to uniformly coat Ni/O/OH on the particle surface
- Starting salts, reaction temperature, reaction time, and atmosphere are important variables
  - Ni(III) results in good lattice match and phase homogeneity; we're exploring use of NiOOH
- Using NMC mixture as input reveals differences in thermal stability and interparticle diffusion



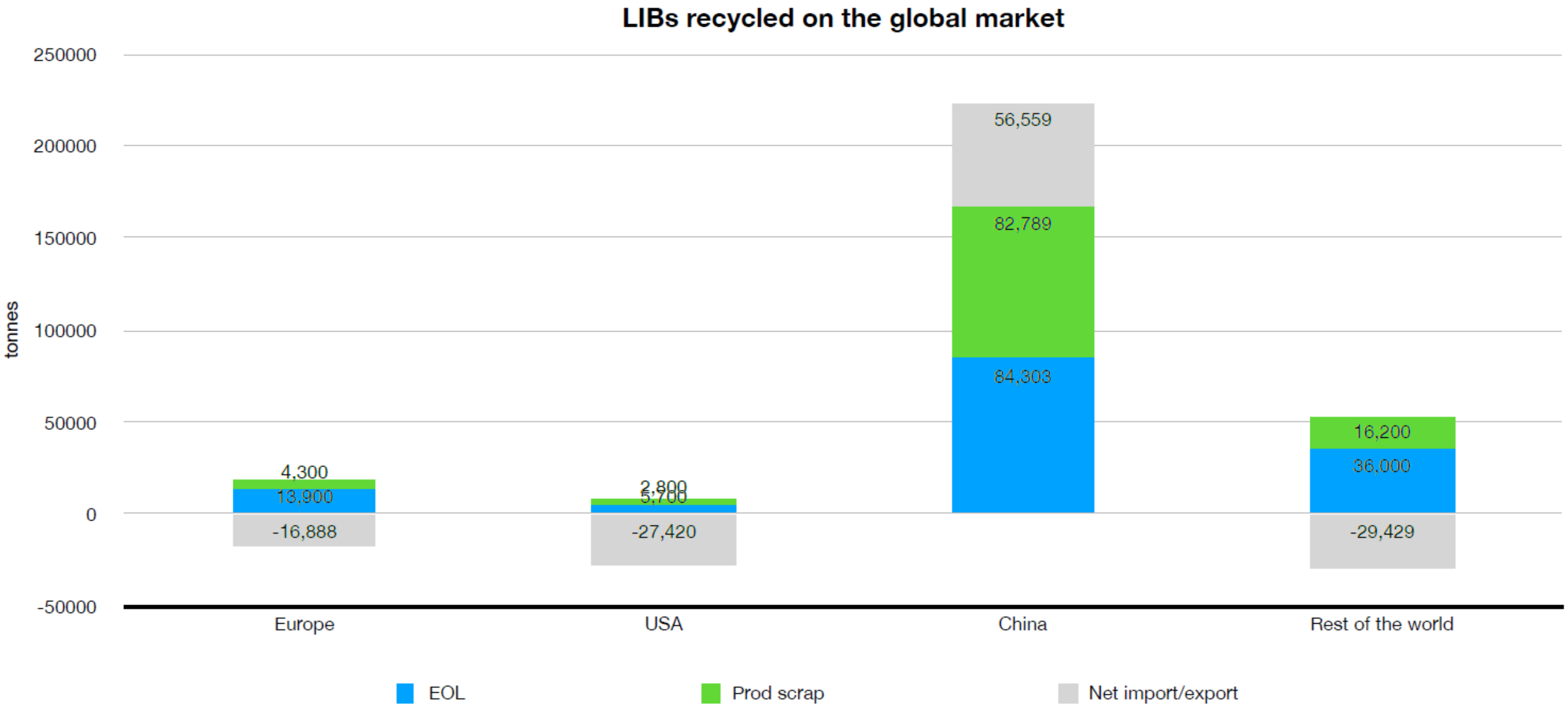
NEW RECYCLING PLANTS' MAIN FEED IS PRODUCTION SCRAP

Artifact of rapid growth



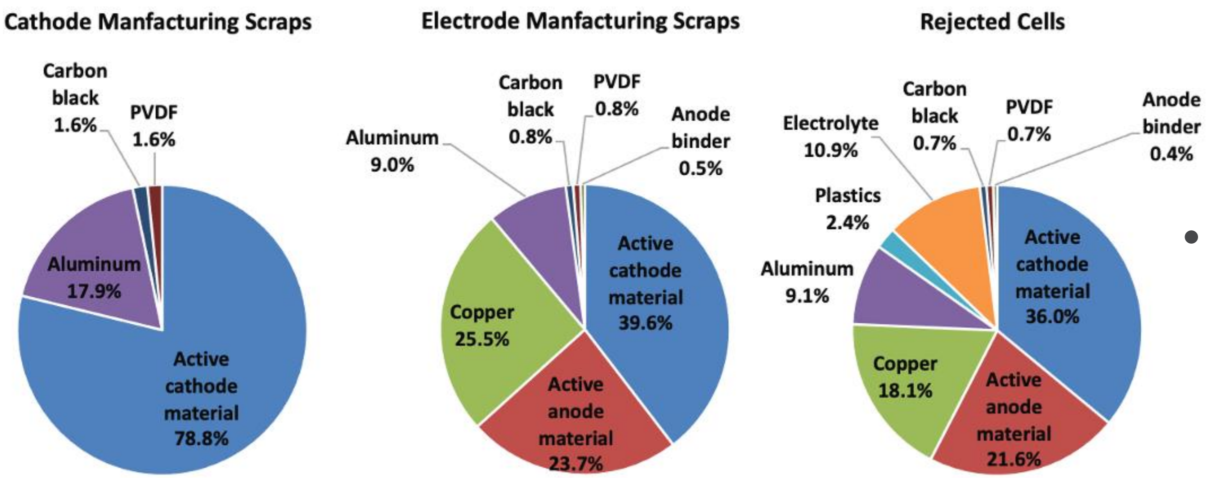
SCRAP IS KEY WORLDWIDE

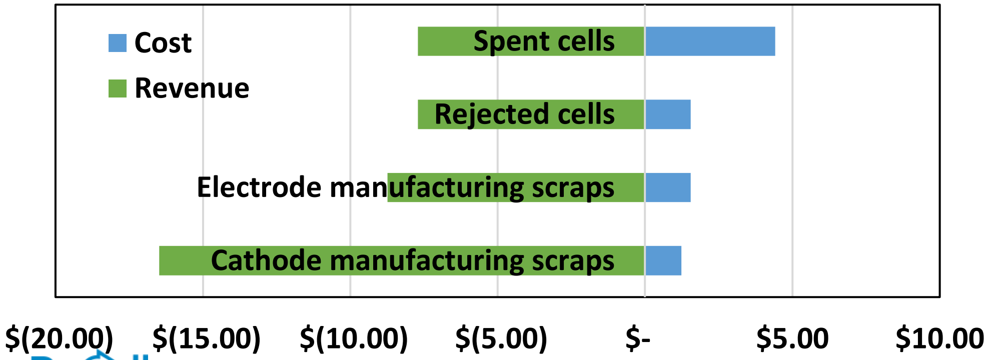
Exports and imports are also important



Source: The lithium-ion battery life cycle report 2021, Circular Energy Storage

EVERBATT SHOWS SCRAP RECOVERY IS PROMISING

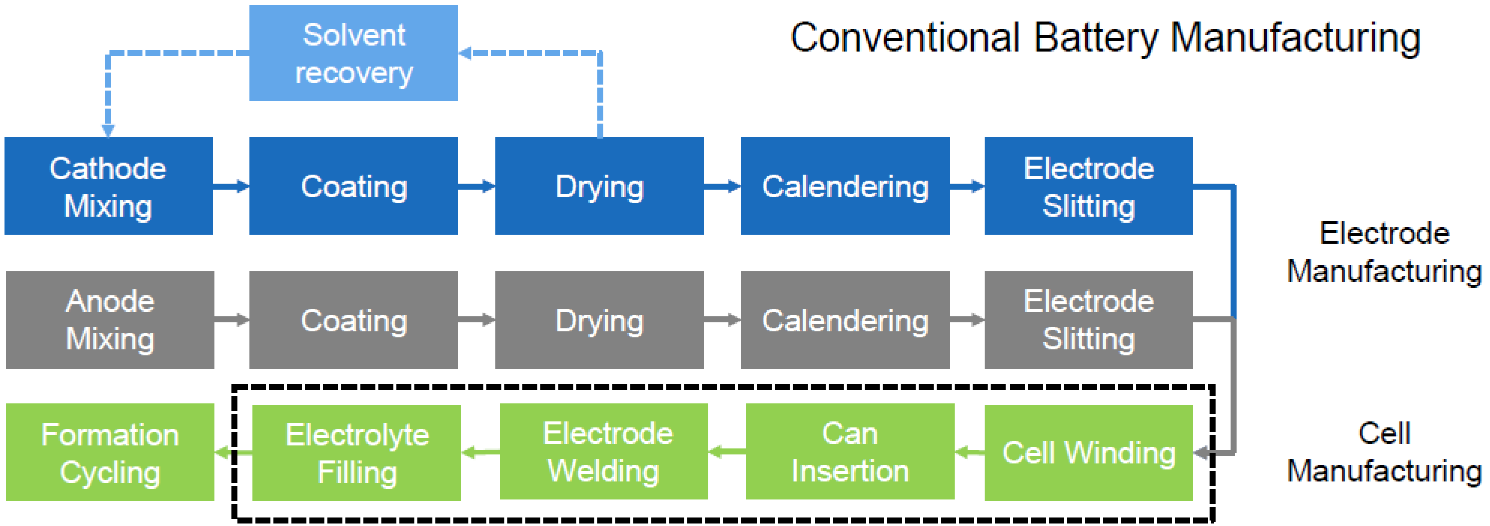




- Scrap contains fewer components in higher concentrations
- Composition is known and current
- Recovered material does not need upgrading

CAN FEED SCRAP INTO RECELL LOOP OR RIGHT BACK INTO MANUFACTURING

Need to compare costs and scales



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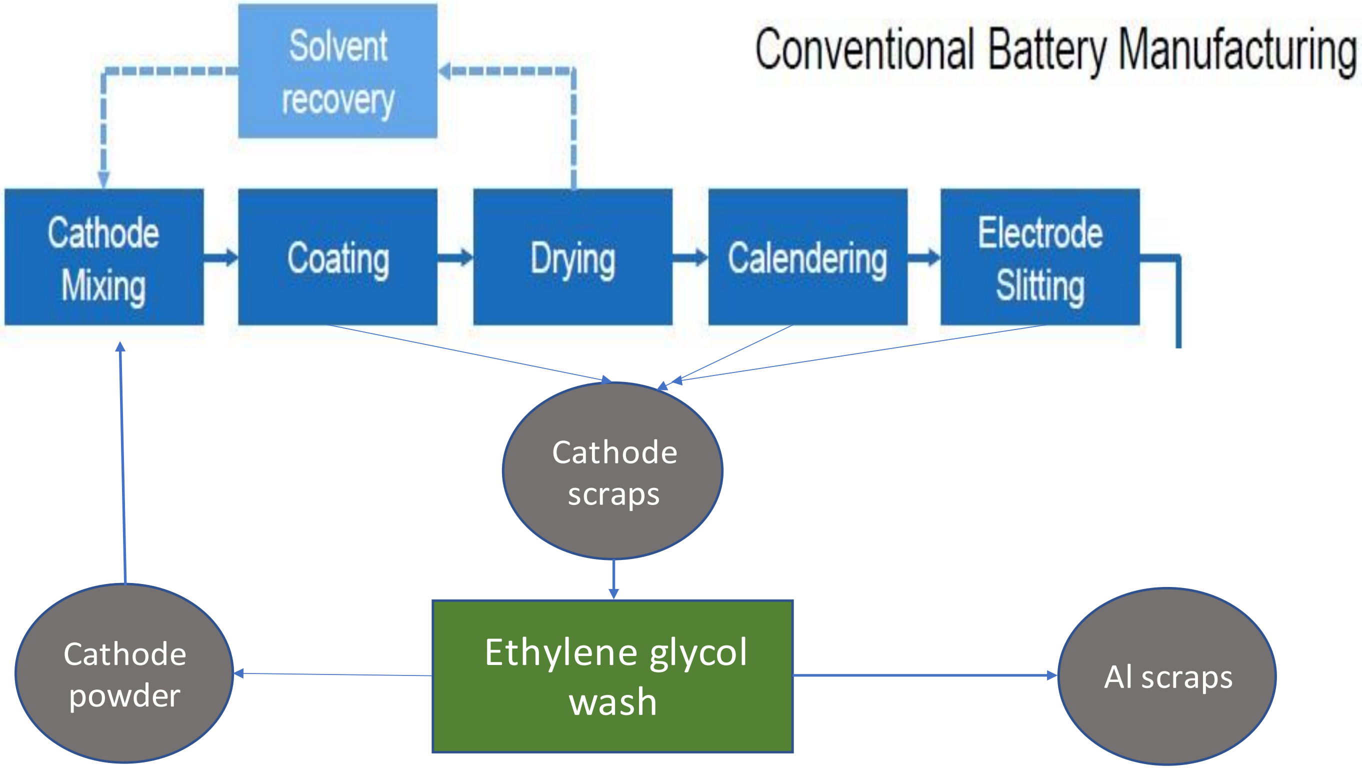
SCRAP FROM CATHODE PRODUCTION

Could go back into cathode production after separation from Al foil



CROSSCUTTING EFFORTS SUPPORT LAB WORK

- Standardize (and supply) materials, cell types, and test protocols across projects to compare recycling methods (ANL/CAMP & MERF)
- Provide technical rationale and diagnostic criteria to determine process suitability (ANL/POST-TEST & NREL)
- Check quality by quantifying chemical signatures at end of life and after regeneration (ANL/POST-TEST & NREL)
- Provide feedback to different recycling processes (*e.g.,* re-lithiation parameters) (ALL)
- Assess quality of regenerated material from different recycling methods (ALL)

Cross Cutting Effort interacts will all Focus Areas under ReCell

TECHNICAL ACCOMPLISHMENTS

ReCell Industry Collaboration Meeting; November 7-8, 2019

134 people
from 76 organizations

Provided an opportunity for ReCell and industry stakeholders to exchange challenges and ideas.

The meeting included stakeholders from every corner of the vehicle battery value chain

Feedback from participants led to planning a second meeting for the fall of 2020, now postponed.

TECHNICAL ACCOMPLISHMENTS

Center accomplishments – cont'd

- ReCell Laboratory Space
- Equipment
  - Screener
  - Magnet
  - Froth column
  - Calciners
  - Powders hood
  - Sink/float separation
  - Aspirator
  - Continuously stirred tank reactor

REMAINING CHALLENGES AND BARRIERS

- Recovering materials that perform as well as new ones
- Obtaining value from a 10-year-old battery chemistry
- Developing technical and economic data sufficient to enable down-selection
- Getting industry buy-in for commercialization
- Developing new recycling processes for future batteries
  - Sodium or magnesium-based cathodes
  - Lithium metal anodes and solid-state electrolytes

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Thank you for your time.

Session host **Dr. Tapesh Joshi**
tapesh.joshi@ul.org

Presenter **Dr. Linda L. Gaines**
lgaines@anl.gov

Learn more about our battery safety science research and initiatives at:

Web: ul.org/focus-areas/battery-safety
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