LI-ION BATTERY RECYCLING R&D AT THE RECELL CENTER

LINDA GAINES

Chief Scientist, ReCell Center

lgaines@anl.gov

ACS Annual Meeting
March 23, 2020

WHY RECYCLE?

- 8 million tons of battery scrap could require disposition by 2040
- Objective is to provide batteries with minimum lifecycle impacts
 - Energy use/emissions
 - Resource depletion
 - Cost
 - Waste generation

ANL projection (based on IEA global PEV projection)

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

COBALT SUPPLY COULD BE A CONSTRAINT

- Co is a key component of the cathode
 - Battery usage being reduced
- Half of the world's Co is in Congo
 - Political issues
 - Human rights issues
- Co price is volatile
- Current recycling efforts focus on Co
- Li is less of an issue

Element	Cumulative Demand to 2025 (1000 T)	Reserves (1000 T)	% of Reserves
Lithium	370	14,000	1.6
Cobalt	1,000	6,900	14.5
Nickel	1,200	89,000	1.3

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

RIBBON CUTTING FEBRUARY 15, 2019

NOBEL PRIZE WINNER SAYS BATTERY RECYCLING KEY TO MEETING ELECTRIC CAR DEMAND

The Nobel Prize in Chemistry 2019

IL Niklas Elmehed, © Nobel Media,
John B. Goodenough
Prize share: 1/3

IL Niklas Elmehed, © Nobel Media,
M. Stanley Whittingham
Prize share: 1/3

IL Niklas Elmehed, © Nobel Media,
Akira Yoshino
Prize share: 1/3

- The 2019 Nobel Prize in Chemistry was awarded to John Goodenough, M. Stanley Whittingham, and Akira Yoshino for the development of lithium-ion batteries.
- **“The point is whether EV batteries can be recycled,” said Akira Yoshino. The cost should pay off if all of waste car batteries in Japan are collected and processed.**
- “The world’s transition to battery power... is set to boost demand for commodities from copper to nickel and cobalt. But there’s also concerns that miners won’t be able to expand raw material supply fast enough, and any shortfall will offer bigger opportunities for recycling.”

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 tones per year

PYROMETALLURGICAL PROCESSING AVOIDS SOME ORE PROCESSING ISSUES

PROS

- Flexible process input
- No sorting or size reduction required
- Valuable metals are recovered
 - Co, Ni, Cu
 - Economics depends on them
- SO_x emissions avoided
- Commercially viable now

CONS

- Li, Al go to slag*
- Organics burned
- Additional processing needed to separate metals
- Extensive and expensive gas treatment
- High temperature
- Capital intensive
- Requires high volume
- Not useful for LFP

* Li recovered if price high enough

HYDROMETALLURGY

Materials are dissolved in acid and components are separated

Courtesy of SMCC

HYDROMETALLURGY RECOVER ALL METALS

PROS

- Substrate foils recovered directly
- Low temperature, low energy
- Li recovered by precipitation
- Output can be converted to cathode precursors
- Can be used for any mix of cells
- Large-scale commercial operations in China and Korea

CONS

- Requires size reduction
- Acid breaks down cathode structure
- No valuable product from LFP
- Solvent extraction needed to separate Co and Ni*

*Or recover Co/Ni salt mixture

DIRECT RECYCLING RECOVERS MOST MATERIALS

Wastes are avoided and revenue generated

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

BENEFITS

- Retains valuable chemical structure
- Enables economic recovery of more materials
- Could be used now for manufacturing scrap at low volumes
- Low temperature, low energy
- Avoids most impacts of virgin material production

CHALLENGES

- 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

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

Cathode, production energy, and aluminum are notable contributors

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

RESEARCH AT CENTER HAS FOUR FOCUS AREAS

**DIRECT
CATHODE
RECYCLING**

**OTHER
MATERIAL
RECOVERY**

**DESIGN
FOR
RECYCLING**

**MODELING
AND
ANALYSIS**

PROCESS R&D IS SUPPORTED BY PRODUCTION OF STANDARD MATERIALS AND CELLS

Results are verified by standard testing and analysis

DIRECT CATHODE RECYCLING

CHALLENGE

- Contaminants from processing
- Mixtures of cathode chemistries
- Old chemistry

SOLUTION

- Binder removal
- Cathode separation
- Relithiation
- Compositional change

IMPACT

- Increased product value (even w/out Co)
- Decreased processing and waste
- Decreased dependence on imported raw materials

Electrical Conductivity	Magnetic Susceptibility	Crystal Density
LCO	LFP	LCO
NMC	NMC	NMC
NCA	NCA	NCA
LMO	LCO	LCO
LFP	LMO	LMO

Separation can exploit several material properties. *Courtesy of Argonne*

CATHODE RECOVERY IS KEY FOCUS AREA

- Chemical relithiation (ANL)
- Electrochemical relithiation (NREL)
- Ionothermal relithiation (ORNL)
- Hydrothermal relithiation (UCSD)
- Binder removal (ANL)
- Cathode-cathode separation (ANL)
- Cathode-cathode separation via froth flotation (MTU)
- Role of impurities in recycled cathodes (WPI)
- Cathode composition change (ANL)

Recycled cathode powder ready to be used in a new battery.
Courtesy of University of California San Diego

BINDER REMOVAL VIA THERMAL PROCESSING

FOCUS AREA : DIRECT CATHODE RECYCLING

Binder must be removed before relithiating separated cathode materials.

Project Description: Thermal decomposition at temperatures as low as 500°C can be used to eliminate the binder. As opposed to solvent-based methods, no liquid waste is produced. Furnace type and loading affect process results (see figure at right).

Accomplishments: We demonstrated processes to remove binder from pristine materials and delithiated materials. This process also enables simultaneous relithiation of delithiated cathode materials by addition of lithium hydroxide

Future Work: In FY20, this project will focus on the difficulties in process scale-up, as well as testing with real materials.

PI: A. Lipson

Institution: Argonne

OTHER MATERIALS RECOVERY

CHALLENGE

- Low-value materials
- Cost-effective processes

SOLUTION

- Cost-effective recovery of electrolyte components (ANL)
- Direct recycling of anode
 - Hydrothermal recovery of black mass (ORNL)
 - Anode/cathode separation and purification (ANL)

IMPACT

- Increased revenue potential
- Decreased waste treatment

A laboratory scale froth flotation unit is used to separated anode powders from cathode powders.

Courtesy of Michigan Technological University

DESIGN FOR RECYCLING

CHALLENGE

- Minimizing cost and performance impacts

SOLUTION

- New cell designs (ANL and ORNL)
- New cell component design
- Enabling cell flushing for rejuvenation

IMPACT

- Reduced cost of recycling
- Overall cost reduction
- Reduced number of cells reaching end of life
- Extended cell life for primary- or second-use applications

Initial design that will be used to determine pressures and flows needed to “rinse” cells

MODELING AND ANALYSIS

CHALLENGE

- Myriad of recycling pathways
- Need for preliminary review of new technologies to streamline work
- Need for research validation

SOLUTION

- Supply chain modeling (NREL)
- EverBatt technical/economic/lifecycle model (ANL)

IMPACT

- Efficient work plan to achieve Center's objectives

EverBatt compares process costs and impacts for 1 Kg NMC111

TRANSPORTATION COST

Classification as hazardous can balloon shipping cost

Transportation and Recycle Cost Breakdown

Transportation cost (\$/kg battery)

SUPPLY FROM RECYCLING DEPENDS ON DEMAND GROWTH CURVE

Fraction rises rapidly after demand growth slows

FIRST YEAR ACCOMPLISHMENTS

- Removed binder by thermal treatment without significantly altering electrochemical performance.
- Achieved hydrothermal delamination of graphite anodes; optimizing cathode delamination.
- Designed, fabricated, and tested prismatic cells with ports. Data were used to model flow patterns.
- Developed updated version of EverBatt and made available for download.
- Determined grain orientation from cathode samples via electron back-scattering diffraction (EBSD).
- Hosted industry collaboration meeting with >130 attendees from as far away as Australia

Grain orientation via EBSD

ReCell

ADVANCED
BATTERY RECYCLING

Thanks to:
Samm Gillard and Dave Howell

U.S. DEPARTMENT OF
ENERGY

Energy Efficiency &
Renewable Energy

VEHICLE TECHNOLOGIES OFFICE

This presentation has been created by Argonne National Laboratory, a U.S. Department of Energy laboratory managed by UChicago Argonne, LLC, under Contract No. DE-AC02-06CH11357. The U.S. Government retains for itself, and others acting on its behalf, a paid-up, nonexclusive, irrevocable worldwide license in said article to reproduce, prepare derivative works, distribute copies to the public, and perform publicly and display publicly, by or on behalf of the Government.