











Project ID: bat381

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2019 DOE Vehicle Technologies Office Annual Merit Review



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OVERVIEW

Timeline

- Project start: October 2018
- Project end: September 2021
- Percent complete: ~15%

Budget

| Year 1 | \$4,615k |
|---------|----------|
| Argonne | \$2650k |
| NREL | \$965k |
| ORNL | \$550k |
| UCSD | \$150k |
| WPI | \$150k |
| MTU | \$150k |

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



RELEVANCE - RECELL CENTER

Objective:

Foster the development of cost-effective and environmentally sound processes to recycle lithium-ion batteries

Bring together battery recycling experts to bridge technical and economic gaps to enable industry adoption

Impact:

Reduced cost of ownership and helping to drive battery costs to DOE's \$80/kWh goal

Reduce primary material production to avoid material shortages and reliance upon foreign sources, increasing our nation's energy security

Minimize environmental impacts of the battery life cycle



MILESTONES

- Q1 (Center) Establish the battery recycling center's mission and include its targets and goals
 - ✓ <u>COMPLETED 12/21/18:</u>

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

- Q2 (NREL) Provide an initial progress report on roll-to-roll relithiation
 - ✓ <u>COMPLETED 3/29/19</u>: Roll-to-roll relithiation work is progressing and the concept is currently being tested using coin cells
- Q3 (ORNL) Provide an initial progress report on design for recycle initiative In progress
- Q4 (Argonne) Establish the ReCell Center's Battery Recycling Laboratory and Scale-up Facility In progress



APPROACH – DESIGN FOR RECYCLING

Current batteries are designed with performance in mind, without much thought towards end-of-life. Designing new batteries with consideration of end-of-life can improve recyclability. But to keep the batteries marketable, the center will develop new designs that trade minimal loss in energydensity performance for the ability to use lower-cost, new recycling processes.

- Cell Design
 - Pouch cell, J. Li (ORNL)
 - Cylindrical cell, A. Jansen (Argonne)





Fabricated NMC622 (3 mAh/cm²) Cathode and Capacity-matched Graphite Anode

Anode: LN3174-62-4 & 63-1 (double-sided) 91.83 wt% Superior Graphite SLC1520P 2 wt% Timcal C-45 6 wt% Kureha 9300 PVDF Binder 0.17wt% Oxalic Acid ReCell, 18650 cell build with cathode [LN3174-64-3&65-3] Prod: SLC1520P, Lot#: Lot#: 022626-376-551 "SS" = single sided, "DS" = double sided -> CALENDERED Cu Foil Thickness: 10 um Total Electrode Thickness: 180 µm (DS) SS Coating Thickness: 85 µm (SS) Porosity: 32.2 % Total SS Coating Loading: 12.49 mg/cm² Total SS Coating Density: 1.47 g/cm³ Estimated SS Areal Capacity: 3.78 mAh/cm² [Based on rev. C/10 of 330 mAh/g for 0.005 to 1.5 V vs. Li] Made by CAMP Facility

Cathode: LN3174-64-3 & 65-3 (double-sided) 90 wt% Targray NMC622 5 wt% Timcal C-45 5 wt% Solvay 5130 PVDF Binder ReCell. 18650 cell build with anode [LN3174-62-4&63-1] Prod: Targray SNMC03006, Lot#: TBD "SS" = single sided, "DS" = double sided -> CALENDERED Al Foil Thickness: 20 µm Total Electrode Thickness: 165 µm (DS) SS Coating Thickness: 73 µm (SS) Porosity: 31.2 % Total SS Coating Loading: 20.43 mg/cm² Total SS Coating Density: 2.82 g/cm³ Estimated SS Areal Capacity: 3.11(3.27) mAh/cm² [Based on rev. C/10 of 169(178) mAh/g for 3.0 to 4.2(4.3) V vs. Li] Made by CAMP Facility

Electrodes fabricated at CAMP Facility (Argonne) and BMF (ORNL)



Technical Accomplishments and Progress

CELL DESIGN

Designed and Fabricated Cylindrical and Prismatic Cells

Cylindrical cell design at Argonne Prismatic cell design at ORNL







- Cylindrical and prismatic cells with ports were designed and fabricated for cell rejuvenation
- Significant efforts were paid to seal the assemblies and ports



Characterized Flow-Pressure Relation in Prismatic Cells



- A test fixture was built to measure cell pressure vs flow rate.
- Cell pressure increased with increasing flow rate and initial compression.
- High initial compression may force part of solvent to flow between the jelly roll and pouch material.



Velocity dependence on inlet size and applied pressure

- Ports are placed on the opposite edges of the cell
- Flow rate: 1ccm flow



- Simplified model using average properties from cell components
- Extensive simulation required to incorporate individual layer properties



- Cell dimension: 58 mmx 90 mm x5.5 mm
- Cell thickness: 5.5 mm
- Inlet radii: 0.9 mm, 1.5 mm, 2.5 mm



- Slower velocity at the center of the cell with large inlet size due to more liquid flows towards the sides
- Velocity independent of applied pressure

Velocity distribution at the inlet (1 psi outer pressure)



Simulate flow patterns and pressure gradient

- Flow rate:1 ccm
- Port size: Ø=0.9 mm
- No external compression





- Due to unavailable property of in-plane permeability, through-plane permeability of separator was used in simulation.
- Diethyl ether was used due to known viscosity as a function of pressure.
- Darcy's law $\kappa = \alpha \frac{\varepsilon^3 d^2}{(1-\varepsilon)^2}$
- High permeability separator provides "highways" for liquid flow.



RESPONSE TO REVIEWERS

New Project FY19



REMAINING CHALLENGES AND BARRIERS

- Unavailable materials properties to accurately simulate flow patterns and cell pressure distribution
 - Permeability of liquids in cell components under compression
 - Viscosity of liquid under compression
- Solvent/electrolyte flow bypass of electrode assembly
- Non-uniform solvent/electrolyte flow distribution through cell assembly



FUTURE WORK

- Characterize flow-pressure in as-assembled cylindrical cells
- Characterize flow-pressure in cycled cylindrical and prismatic cells
- Estimate cell energy and power density in the new cell designs
- Screen solvents to effectively remove undesired compounds
- Rejuvenate harvested cycled electrodes and cycle in cells
- Evaluate cycle life benefit from electrolyte rejuvenation
- Assemble cylindrical and prismatic cells in proposed cell designs and cycle them

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



SUMMARY

- Designed and fabricated cylindrical and pouch cells that allow for electrolyte rejuvenation
- Fabricated state of the art electrodes (NMC622 and graphite) for this work
- Began characterizing the flow-pressure relationship during removal of undesirable compounds and injecting fresh electrolyte
- Initiated evaluation of cell energy and power density in new cell designs



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