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Renewable Energy

Overview of the Battery Materials Research Program

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June 23rd, 2021

Project ID #
BAT108

Battery Materials Research (BMR)

- Charter
  - Research new, high-capacity materials that are affordable and promote a safe, high-energy cell design. Present emphasis on sulfur, solid electrolytes and lithium.
- Issues
  - Li Metal: High reactivity and dendrite growth
  - Sulfur: Polysulfide shuttle and poor utilization
  - Solid-State Electrolytes: Low ionic conductivity and high interfacial resistance
- Approaches
  - Engineer a host for lithium and/or an artificial SEI layer to protect lithium surface
  - Design novel structures to encapsulate polysulfides
  - Investigate new, low-cost and conformal solid-state electrolytes
  - Develop advanced modeling and characterization techniques to investigate and mitigate the reactivity at the interphases/interfaces.

- **Participants:** National Labs (7), Industry (3), Academia (15)
- **7 Topic Areas, 51 research projects**

Topic Area	Number of Projects
Modeling	12
Diagnostics	8
Polymer and Solid-state Electrolytes	13
Metallic Lithium	6
Sulfur Electrodes	5
Air Electrode/Electrolyte	3
Sodium-ion Batteries	4
Total	**51**

Cost/benefits of Solid-State Electrolytes over Liquid

	Polymer and Composites	Oxides	Sulfide Based
Representative	PEO, PEO + Garnet	Garnets (LLZO)	xLi_2S-$(100-x)P_2S_5$, Li_6PS_5X (X = Cl, Br, I), LGPS
Material Phase	Amorphous	Crystalline	Crystalline or Glass
Ionic Conductivity	Poor	Fair	Good
Air Stability	Good	Good	Poor
Stability Against Li Anode	Good	Good	Poor
Stability Against High V Cathode	Fair	Good	Poor
Ease of Manufacturing/ Processing Technique	Good/ Roll-to-roll	Fair/ Tape casting then sintering	Good/ Roll-to-roll
Stack Pressure Required	✓	✓	✓
Companies	Hydro Quebec, Bollore, Seeo	Ion Storage System, Quantumscape	Toyota, Samsung, Solid Power, PolyPlus

Cost/benefits of Solid-State Electrolytes over Liquid (2)

Case	Anode	Cathode	Cost Impact ($/KWh)	Technology Advantage
1	Lithium Metal	Conventional (NMC)	↓	Non-combustible electrolyte reduces risk of thermal runaway, allowing for tighter cell packaging and improved volumetric energy density.
			↓	Improved cell safety resulting in simpler battery thermal management
			↓	Ability for bipolar cell construction resulting in higher voltage and energy
			↓	Ability to thin anode resulting in higher volumetric energy
			↓	Reduced cell formation time
2	Metal Free	Conventional (NMC)	↓	Same as Case 1 + No anode resulting in even higher volumetric energy
3	Lithium Metal	Next Generation	↓	Same as Case 1 + Abundant, low-cost cathode such as sulfur

Polymer Electrolytes

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- Two well-known lithium metal polymer battery technologies were developed with DOE/VTO support.
  - Both operate at elevated temperatures (60°C– 80°C)
  - Suitable for applications that require continuous operation (e.g., public transportation)
- Bolloré/Avestor/Hydro Quebec-3M/ANL PEO based technology
  - Blue Cars in Paris, France (car-sharing)
    - 30 kWh, 160-mile range
  - City Buses in Wiesbaden, Germany
    - 441 kWh, 125-mile range even in winter

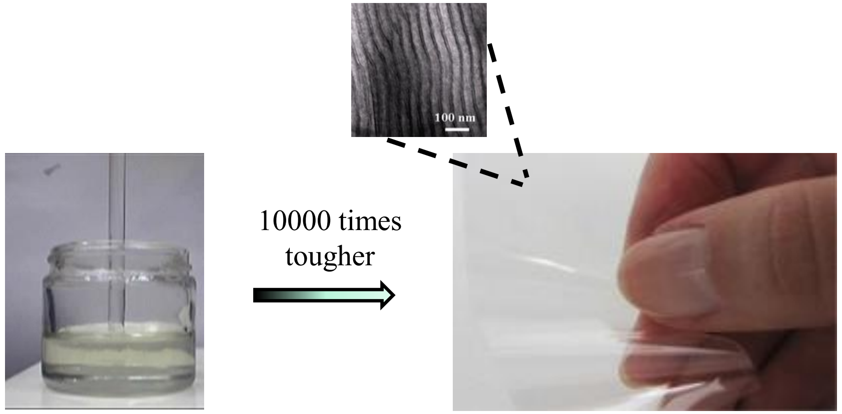
Bolloré Lithium Metal Polymer Battery Module

Bolloré Blue Cars

Solid-state Battery Powered Bus

Polymer Electrolytes (2)

- Bosch/SEEO/LBL PEO Block Copolymer
  - Demonstrated as a viable technology under the DOE/USABC Technology Assessment Program.
    - 11 Ah cells achieved more than 500 DST cycles with < 7% capacity loss at ANL.
    - Thermal ramp and overcharge tests were conducted on a module (165 V,11 Ah) at SNL.
    - Technology successfully demonstrated with a high voltage cathode when SEEO was acquired by Bosch in 2015. (Required addition of a second, more stable polymer)



Block Copolymer Approach Provides Ionic Conductor with Mechanical Stability.

SEEO Lithium Metal Cell and Module

Polymer Electrolytes (3)

Shortcomings

- Requires high-temperature operation
- Existing commercial technology uses $LiFePO_4$ and displays a low specific energy.
- PEO-based polymer is not stable against high-voltage cathodes such as LiNMC.
  - May require a separate, more stable polymer for the composite cathode.

Oxide-based Solid-State Electrolytes (LLZO)

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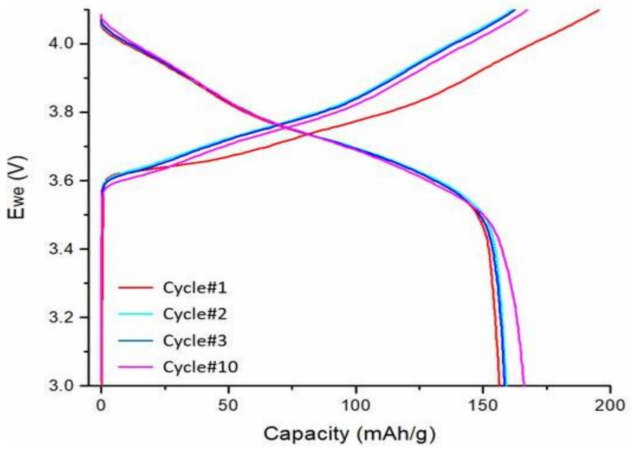
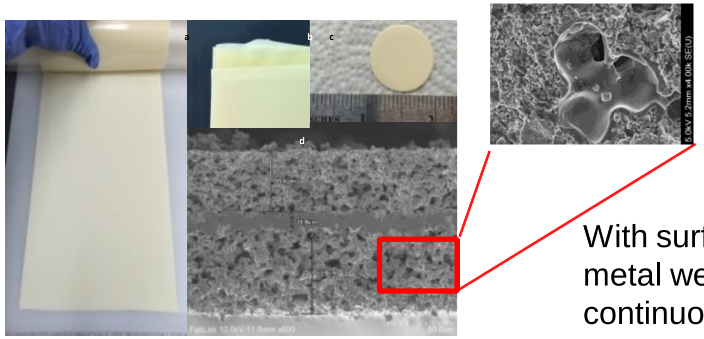
- Very attractive due to its stability against both lithium metal and high-voltage cathodes.
- No practical-size cells are available to date. Only small lab-cells are being built for evaluation.

Lawrence Berkeley National Laboratory (@C/10)



Freeze Casting Processing
Li/LLZO/NMC-622

Scalable and Reproducible Process to Fabricate Multilayer Garnet Structures



With surface treatment, Li metal wets garnet surface continuously inside porous support

University of Maryland: Trilayer LLZO Structure, Li/LLZO/Sulfur

Oxide-based Solid-State Electrolytes (LLZO) (2)

Shortcomings

- High-temperature sintering could induce an imperfect buried interface.
- No known scalable roll-to-roll manufacturing process exists for thin membranes.
- Difficult to achieve and sustain an intimate interface between the cathode and extremely-rigid electrolyte.

Sulfide-based Electrolytes

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- Attractive due to their high ionic conductivities and being easy-to-produce
  - Amorphous glass can be drawn from preforms.
  - Powder can be made through mechanical milling or solvent- based processes.
  - Scalable roll-to-roll manufacturing of membrane is being demonstrated.
  - Lamination to the positive electrode requires hot pressing.
  - Several companies can make 1 Ah – 5 Ah prototype cells for evaluation.

Preform of $LiPO_3$

Draw Tower

Source: Iowa State University

Sulfide-based Electrolytes (2)

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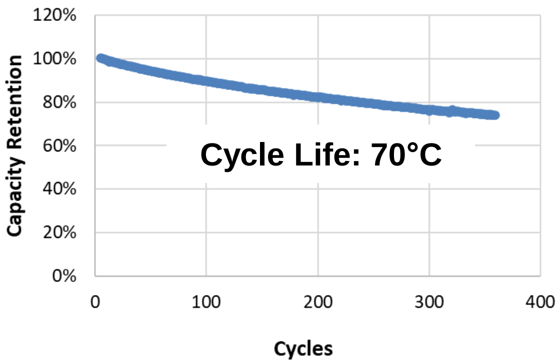
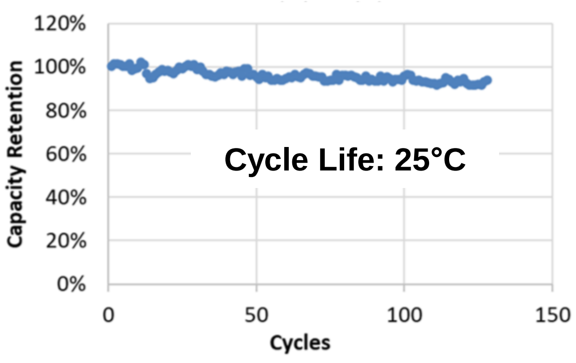
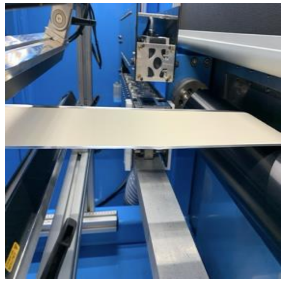
Shortcomings

- ❑ Component materials are not stable against lithium and high-voltage cathodes.
- ❑ Creating an intimate contact at electrode interfaces is very challenging.
- ❑ Most prototype cells available for benchmarking must operate at a high temperature to extend cycle life.

Solid Power ASSB Pouch Cell
(NMC622 cathode 3.0 mAh/cm^2, 2.8-4.2V, C/5-C/5)







Continuous Roll-to-Roll Cell Production

2-Ah Production-Line Cell

Summary

- Developing a thin, low-cost, conformal solid electrolyte that can stop lithium dendrites is very challenging.
- Major show-stoppers for current solid-state electrolytes are at the interfaces – both electrochemical and mechanical.
- Applying stack pressure helps improve cycle life but most reported pressures are simply not suitable for practical applications.
- Numerous scientific challenges need to be resolved before all-solid-state batteries become a reality.