Progress and Status of Battery500 Consortium Phase II

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Project ID bat317

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Overview

Timeline

- Project start date: 10/01/2021
- Project end date: 9/30/2026
- Percent complete: 33 percent

Barriers

- Barriers addressed
  - Increasing the energy density of advanced lithium (Li) batteries beyond what can be achieved in today's Li-ion batteries is a grand scientific and technological challenge.

Budget

- Total project funding: DOE share $75M
- Funding received in FY 2022: $15M
- Funding for FY 2023: $15M

Partners

- Project lead: PNNL
- Team: Binghamton Univ., BNL, INL, GM, Penn State Univ., Stanford Univ./SLAC, Texas A&M, UC San Diego, Univ. of Maryland, Univ. of Pittsburgh, Univ. of Texas, Austin, Univ. of Washington
- Industry Advisory Board

Relevancy:

Project Objectives

- Develop next generation high-energy, low-cost batteries for electric vehicles
- Design, fabricate and validate high energy pouch cells up to 500 Wh kg^{-1}
- Scale up pouch cell capacity up to 5-10 Ah
- Demonstrate long cycle life of up to 1,000 deep charge-discharge cycles
- Achieve total control of battery chemistries for robust, scalable and commercially viable technologies

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Critical Assumptions and Issues

- Li metal anode coupled with high-capacity high-Ni-NMC or S cathode can achieve a specific energy of up to 500 Wh/kg through cell level design and optimization of materials and architectures.

- Li metal anode problems can be addressed with a multidisciplinary approach combining new electrolytes, new metal protection methods and novel anode architectures.

- The failure modes involved in real cells involve high cathode loading and lean electrolyte and are much more complex than those investigated in diluted systems. A robust and system approach must be developed to quantify such processes and validate materials and cell performances.

- The high-energy cell design pushes the limits of all components and requires a multidisciplinary approach, the improvement, integration and deployment of the best materials and concepts accessible to the Battery500 team.

Approaches

- Fundamental breakthroughs in controlling the electrochemical reactions in high energy electrode materials and cells for next generation high-energy, low-cost batteries;

- Integrating development and discoveries from materials to cell level, and rapidly validating and incorporating latest results in realistic cells;

- Leveraging materials developed under other DOE programs, and state-of-the-art DOE facilities to understand and prevent degradation.

- Developing and deploying multi-disciplinary approaches and enhancing collaborations between national laboratories, universities and industry.

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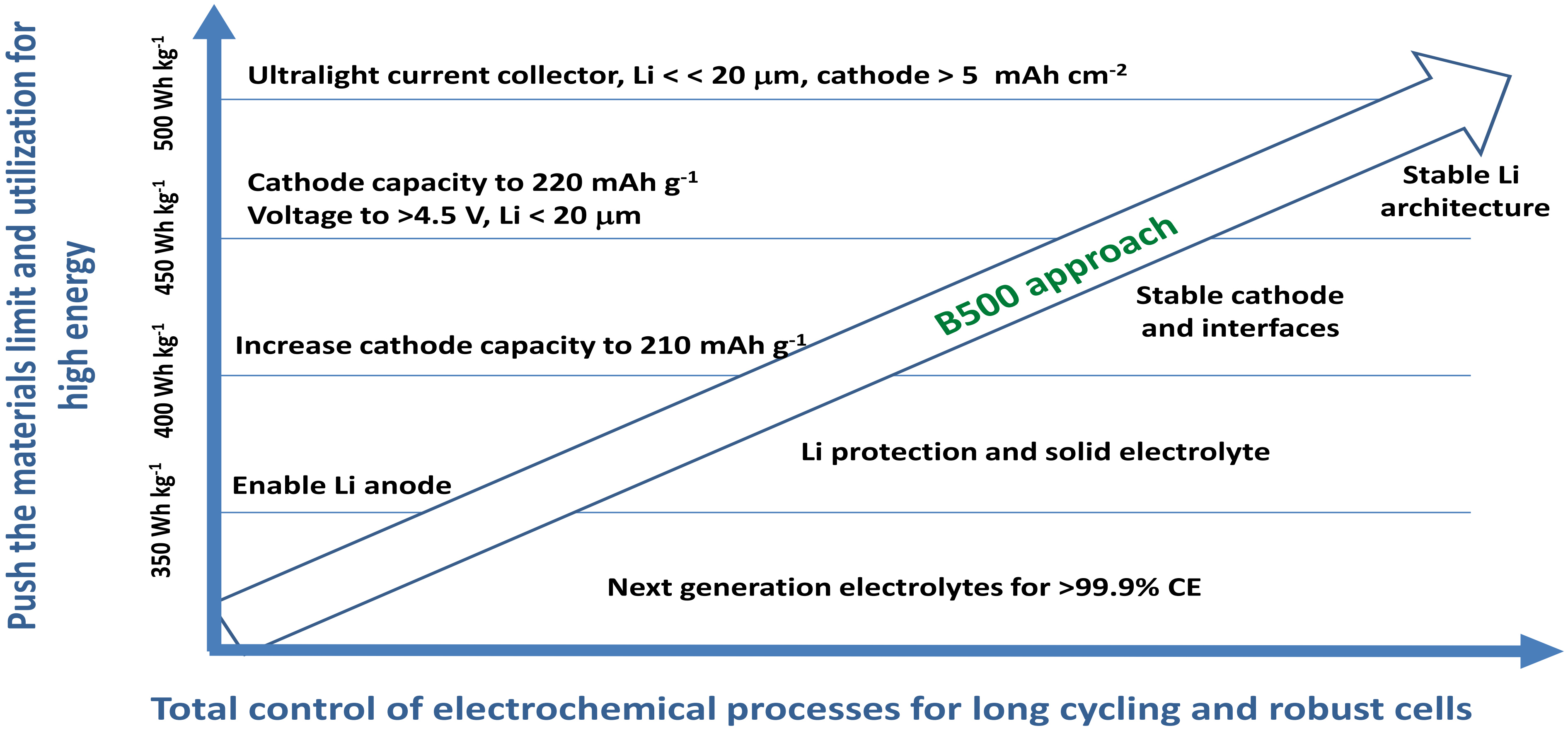
Technical Approaches

- Integrate and optimize the best materials at the cell level, and push the limit of materials performance and utilization to achieve high energy density and low cost;
- Understand and control component and system level electrochemical reactions and degradation mechanisms to realize long cycle, safe and reliable performance.



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Program Level Milestones

Milestones and Go/No-Go Decisions	Date	Status
Q1: Develop single layer pouch cell for SPAN chemistry	12/31/2022	Completed
Q2: Define processing, characterization, and QC/QA protocols for 20 g/batch scale SPAN materials for B500 team's use in investigations.	3/31/2023	Completed
Q3: Demonstrate the feasibility of 350Wh/kg Li/NMC pouch cell with up to 800 cycles	6/30/2023	On track
Q4: Demonstration of 450Wh/kg Li/NMC pouch cells with 250 cycles	9/30/2023	On track

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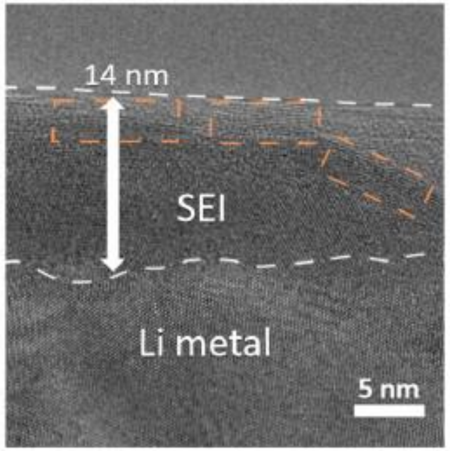
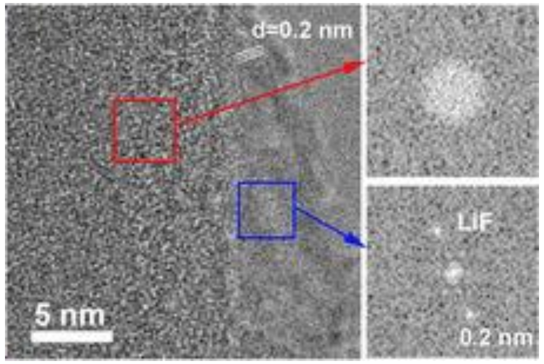
Technical accomplishment: pushing the frontier of science and understanding SEI reactions in Li metal systems

Revealing detailed SEI structures

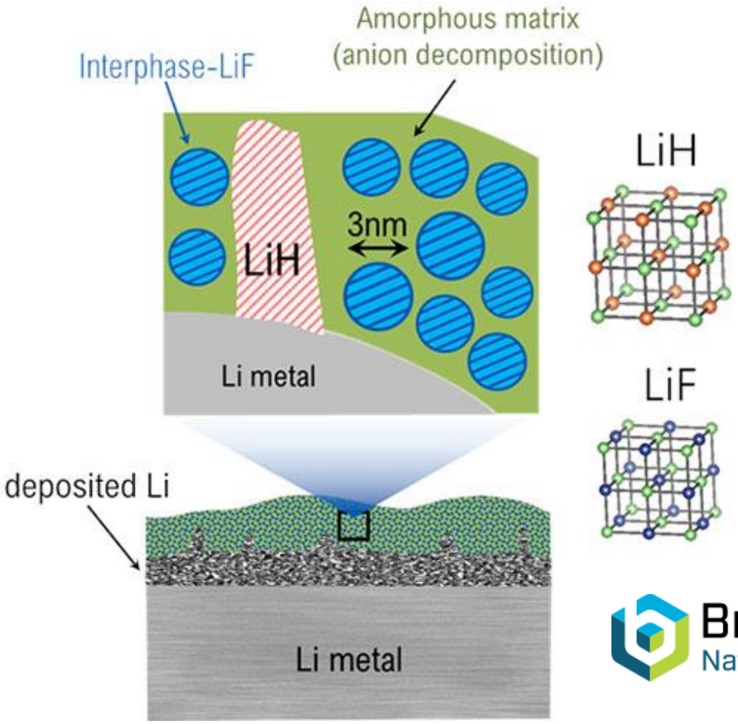




SLAC NATIONAL ACCELERATOR LABORATORY

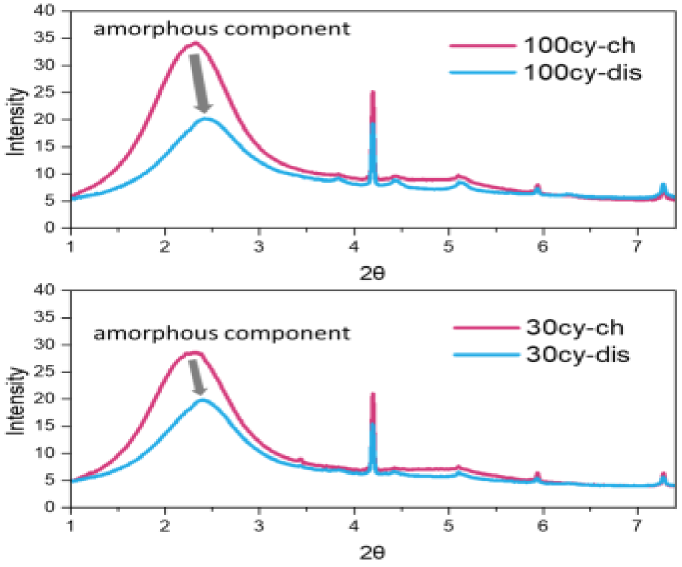
UC San Diego

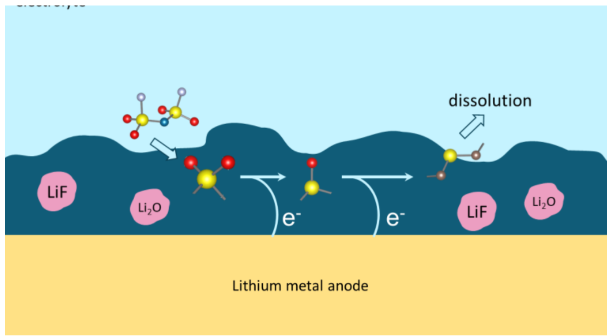
Identifying new phases (LiH)



Brookhaven National Laboratory

Time resolved processes





1. Validated SEI model from the literature
2. Detailed information on composition and structures of SEI
3. Time resolved information on SEI
4. Opportunity to quantify species and reactions
5. New lessons on cell failure

1. Li, Y.Z. et al. Atomic structure of sensitive battery materials and Interfaces revealed by cryo-electron microscopy. Science 358, 506-510 (2017).
2. Wang, X.F. et al. New Insights on the Structure of Electrochemically Deposited Lithium Metal and Its Solid Electrolyte Interphases via Cryogenic TEM. Nano Letters 17, 7606-7612 (2017).
3. Shadike, Z. et al. Identification of LiH and nanocrystalline LiF in the solid-electrolyte interphase of lithium metal anodes. Nature Nanotechnology 16, 549-554 (2021).

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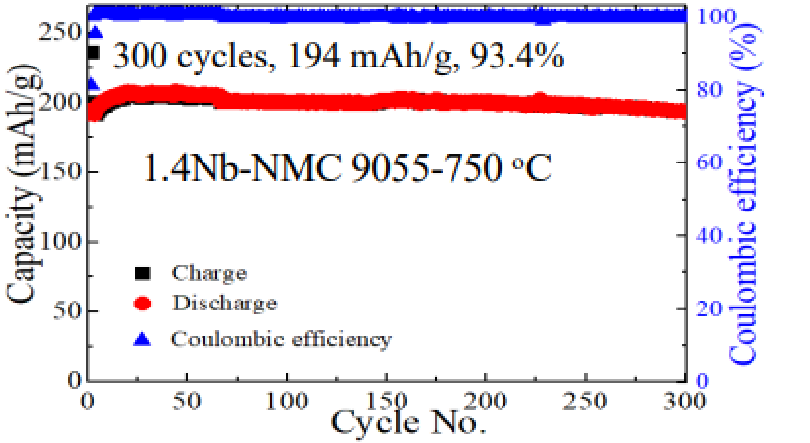
Technical Accomplishment: Pushing the limit of materials performance

90%Ni: Nb modification with M47 gives good electrochemistry

90% Ni NMC

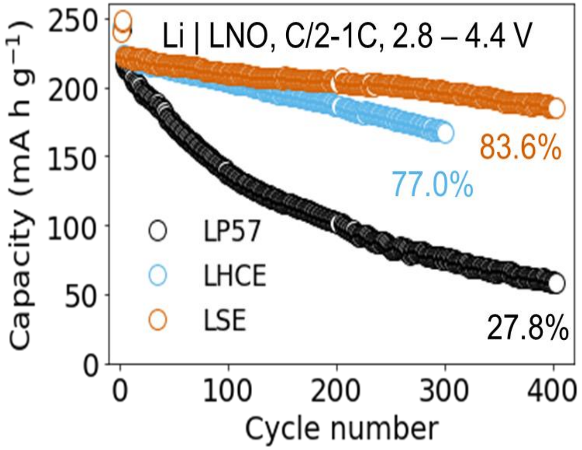
- Nb introduced at lithiating stage
- Reduces cracking
- Reduces impedance growth
- M47 improves capacity retention over carbonate

After 200 deep charge/discharge cycles

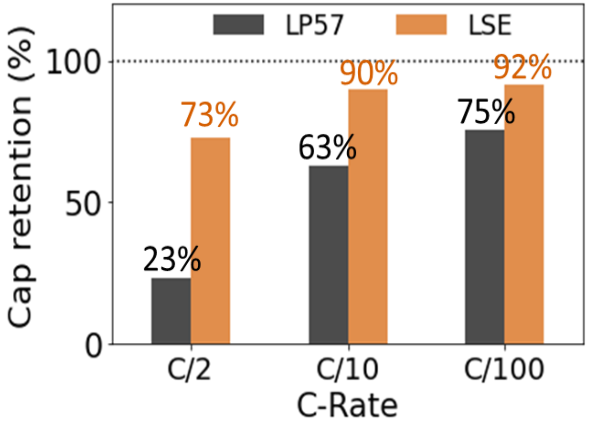


2.8 to 4.4 V; C/10 charge and C/3 discharge; PNNL (75 uL) ;13-15 mg/cm^2; Li foil

Long cycling with improved electrolytes

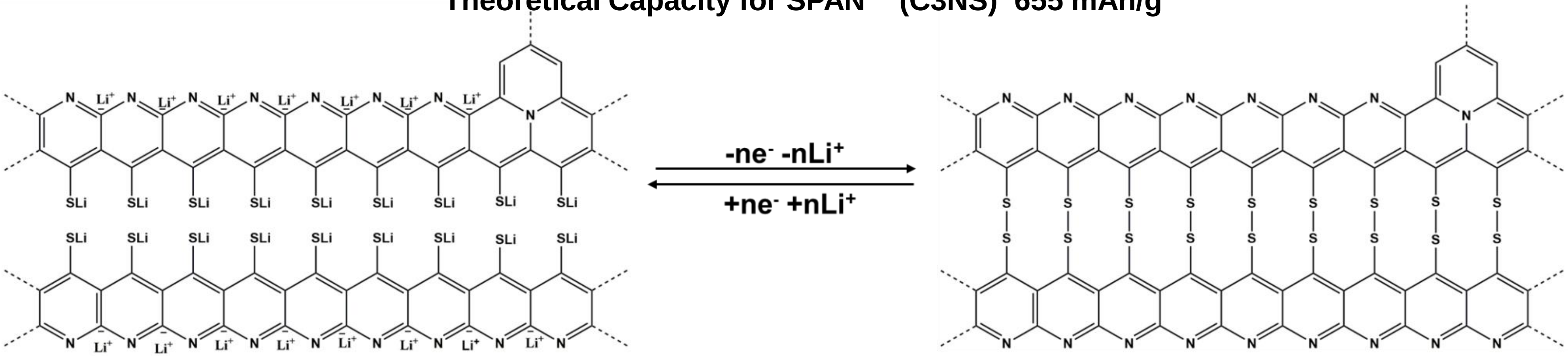


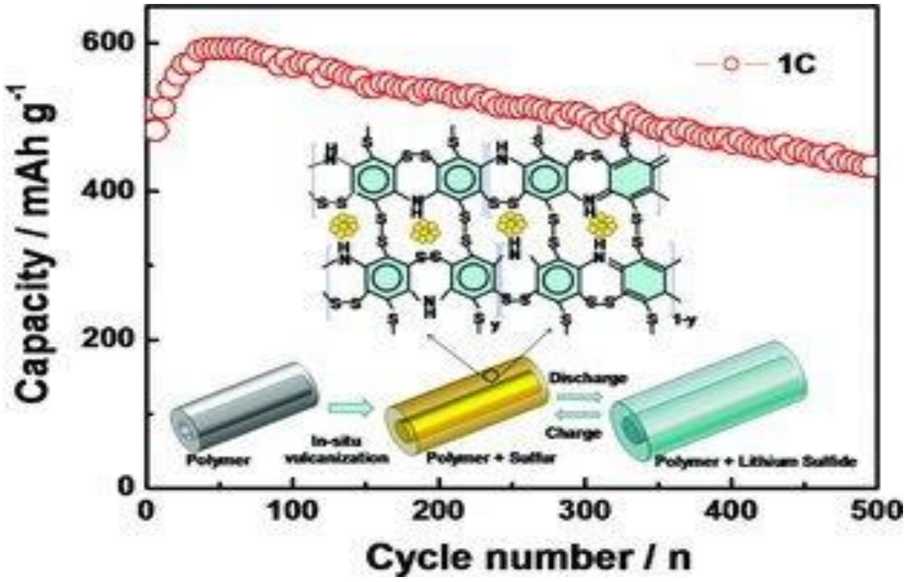
After 400 cycles

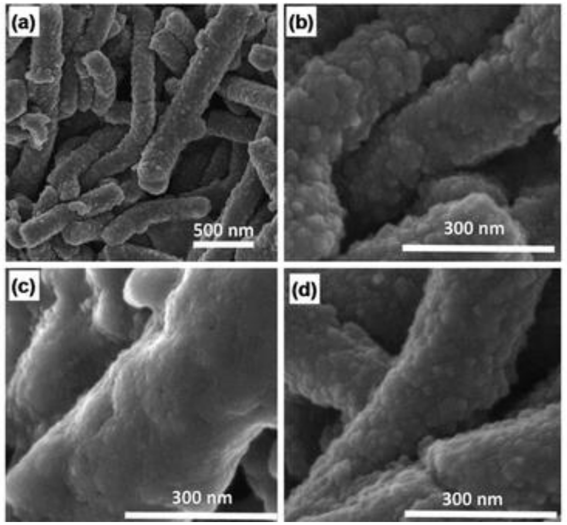


Technical Accomplishment: Li-Span for ultra-low cost batteries

Theoretical Capacity for SPAN (C3NS) 655 mAh/g

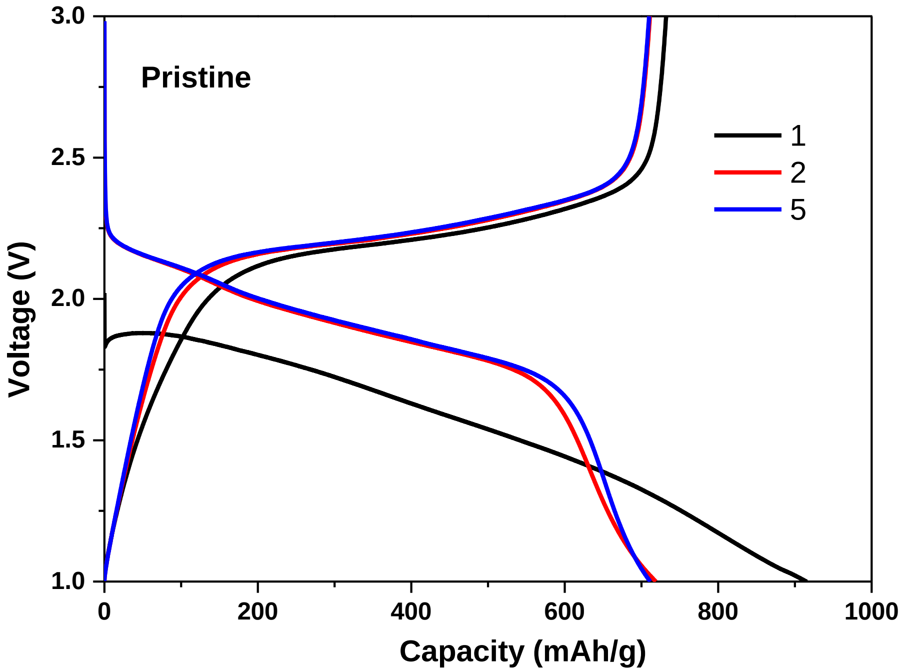




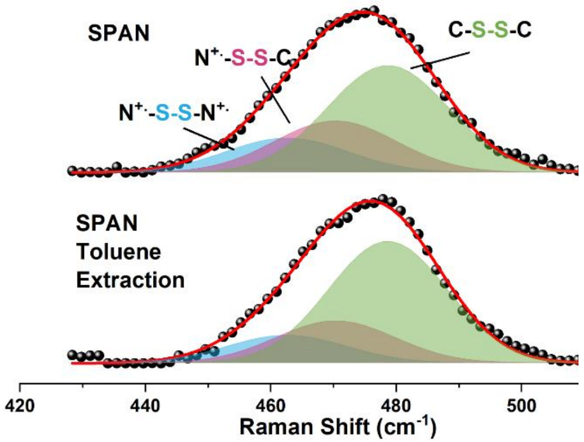


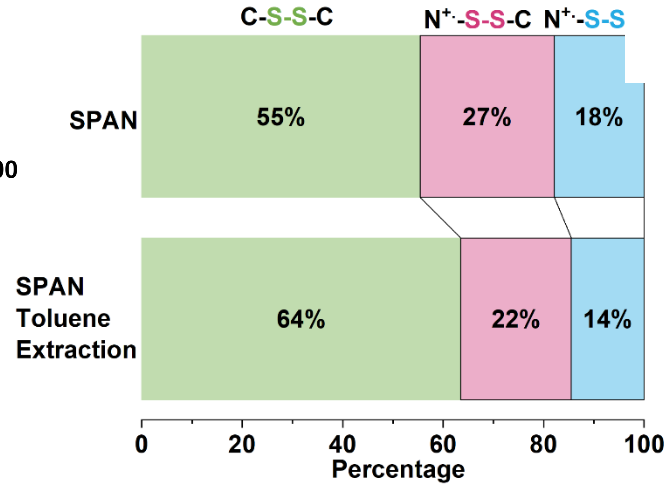
Lifen Xiao, Yuliang Cao, Jie Xiao, Birgit Schwenzer, Mark H. Engelhard, Laxmikant V. Saraf, Zimin Nie, Gregory J. Exarhos, Jun Liu “A Soft Approach to Encapsulate Sulfur: Polyaniline Nanotubes for Lithium-Sulfur Batteries with Long Cycle Life,” Advanced Materials, January 2012 https://doi.org/10.1002/adma.201103392Citations: 917

New chemical bonds for high specific energy revealed by advanced characterization techniques

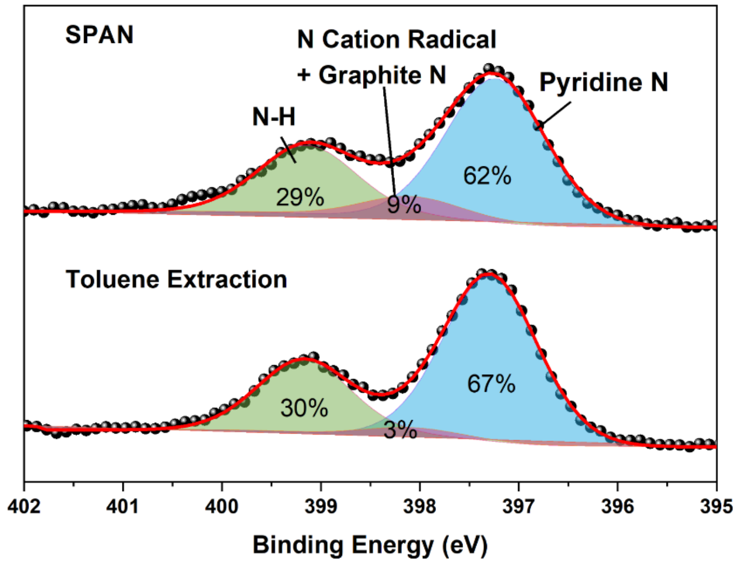


Revisiting Raman

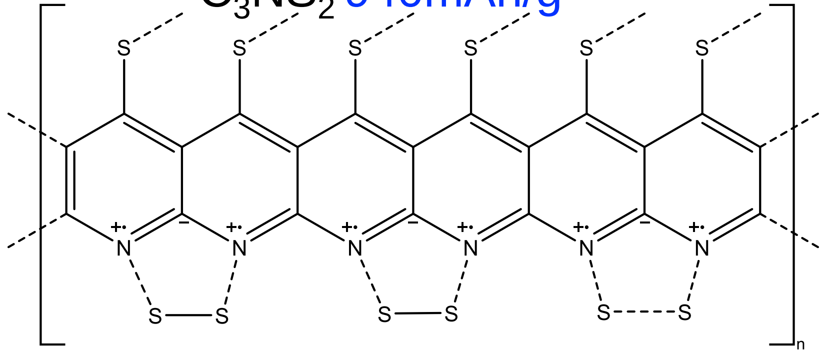




Revisiting N XPS



C_3NS_2 940mAh/g



"Structural Transformation in Sulfurized Polymer Cathode to Enable Long Life Rechargeable Lithium Sulfur Batteries"
S. Wang, B. Lu, B. Liaw, Y. S. Meng, P. Liu, under revision

TEXAS A&M UNIVERSITY

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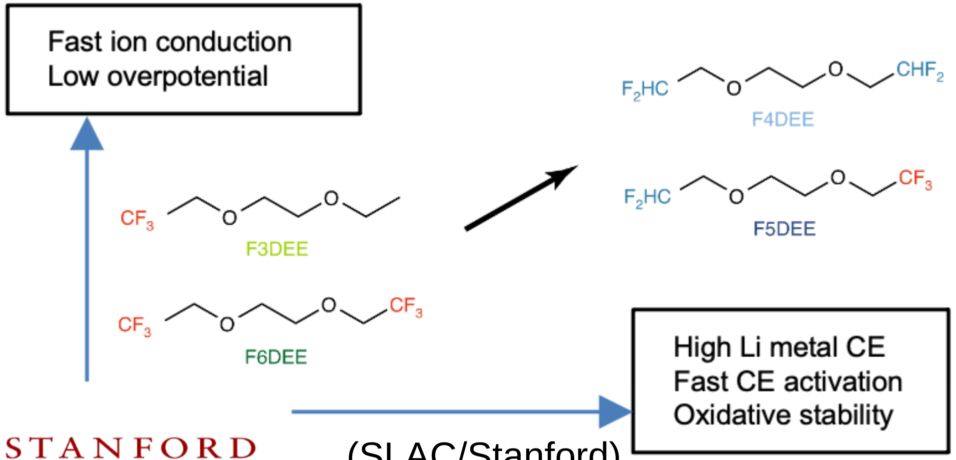
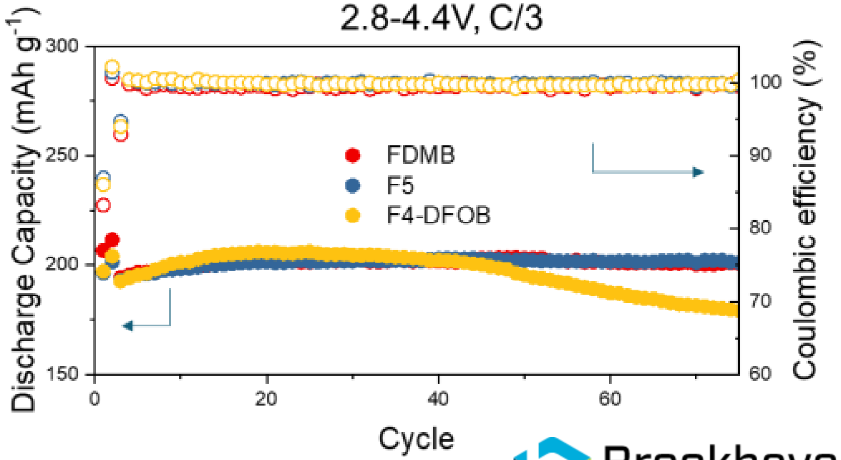
Integrated approach for new electrolytes

Electrolyte design and development



STANFORD UNIVERSITY (SLAC/Stanford)

Independent evaluation

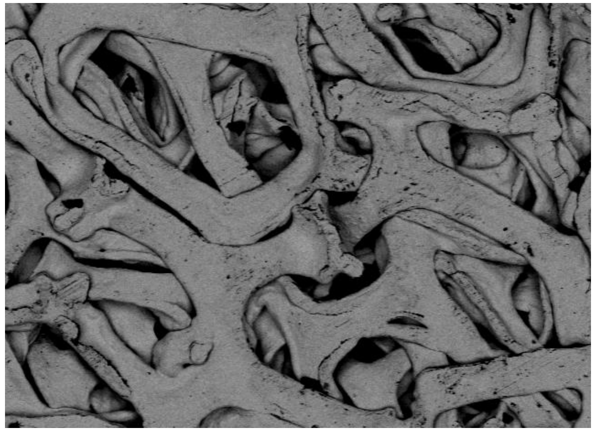


(BNL)

Brookhaven National Laboratory

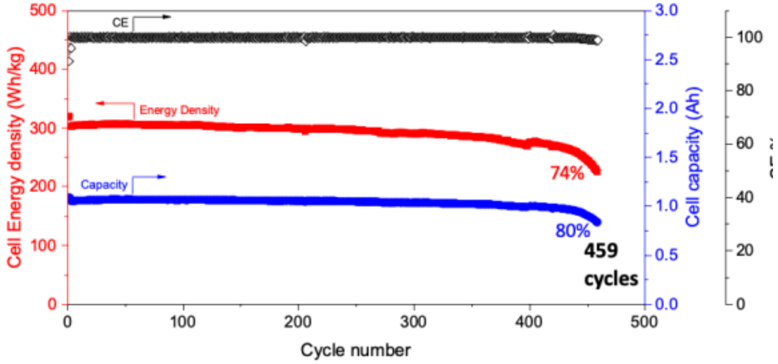
Stability against substrate

UNIVERSITY of WASHINGTON



(UW)

Integration with pouch cell



(Stanford electrolyte tested at PNNL)

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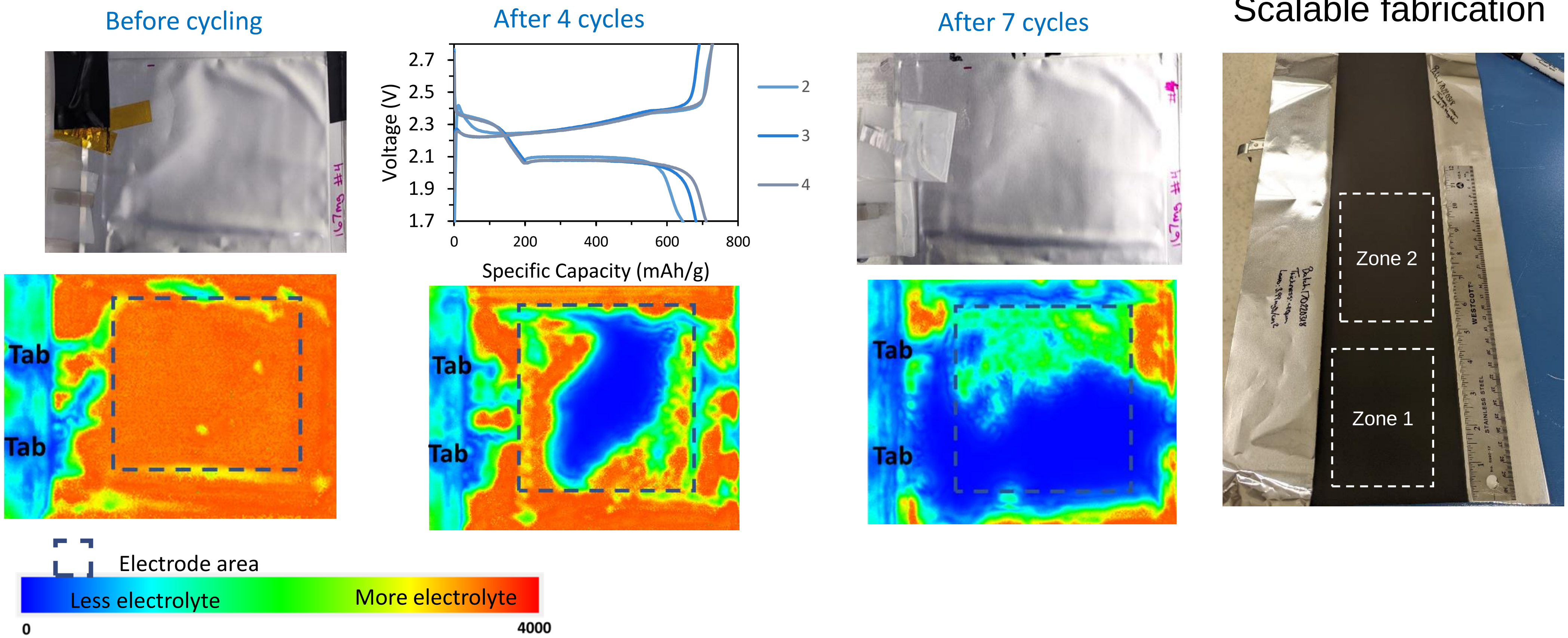
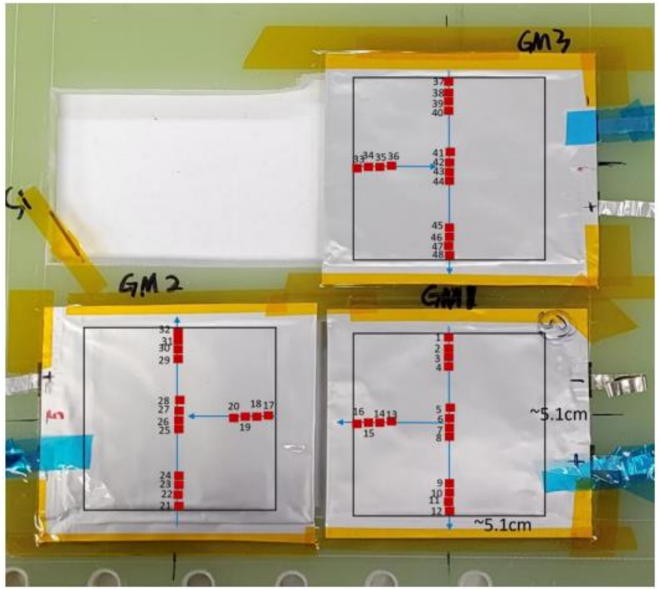
Ultrasonic mapping characterization of Li-S pouch cell

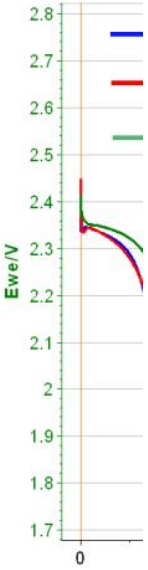
- ➢ One SLP cell with shelf-life for 4 months (electrolyte filled with no cycling)
- ➢ Periodically Ultrasonic checking after cycling (no pressure)



- ❑ Slight puffiness was observed on cycled cells.
- ❑ It's evident that electrolyte has been consumed and gases generated with electrochemical cycles.
- ❑ The results show close correlation between electrolyte distribution and cell performance.

Investigation of electrochemical reactions of Li-S pouch cells

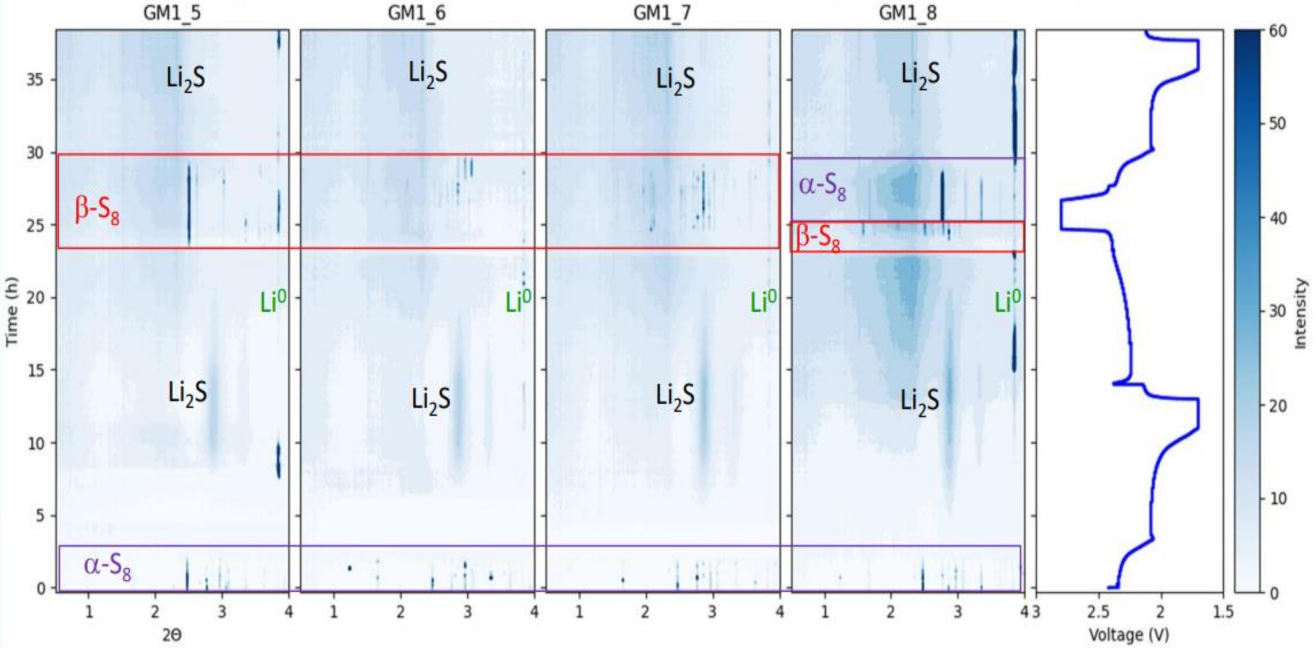




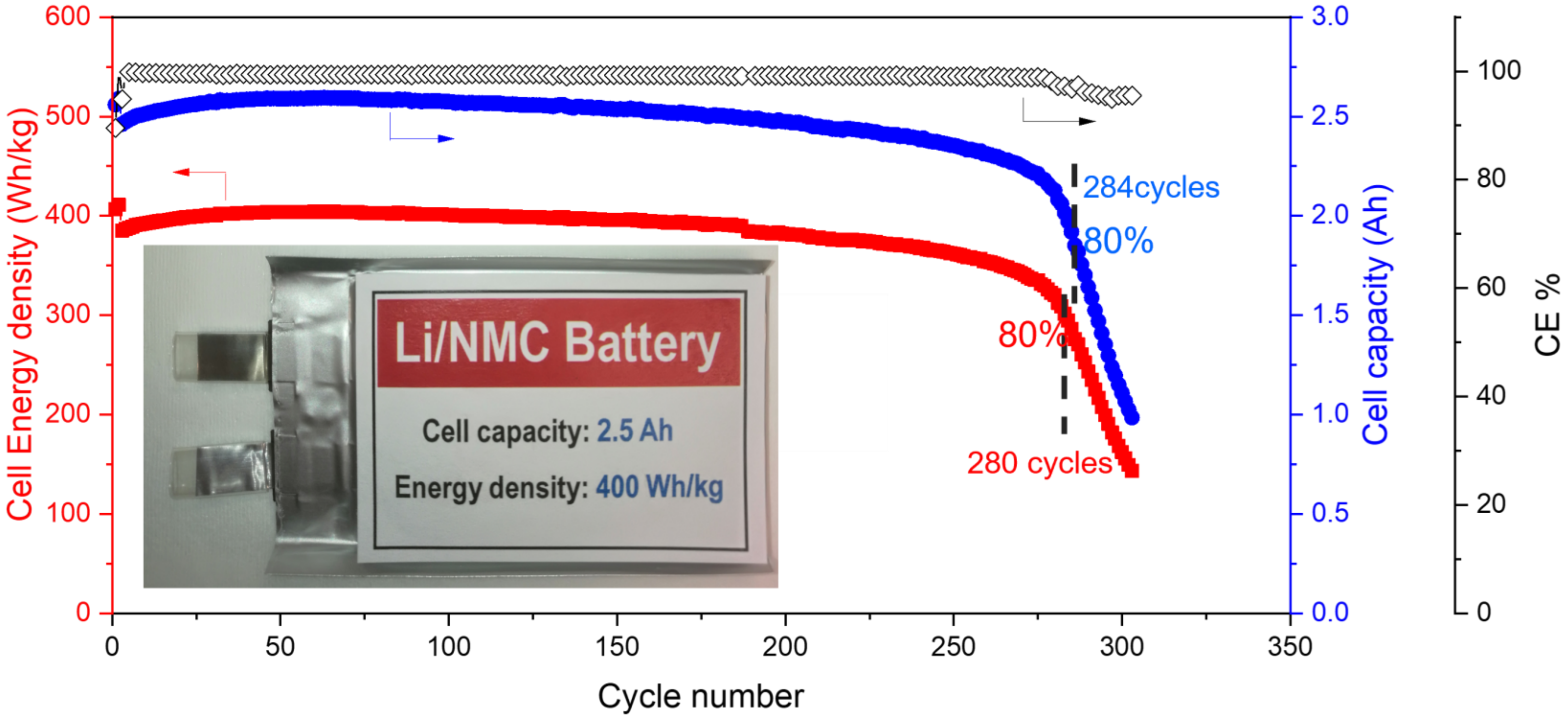
- 10 minute loop (240x repeats)
- 48 total points / 16 points per cell
  - 4 groups (Horiz; Upper / Center / Lower)
  - 2 mm spacing of points in a group

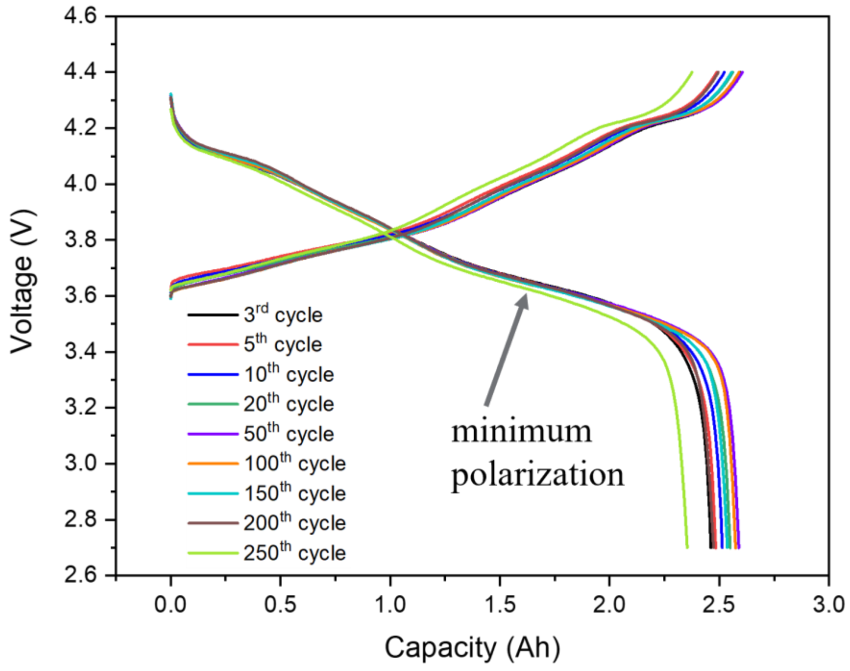
Quantification – opportunities and limitations

1. Quantified 3 crystalline phases: α-S_8, β-S_8, Li_2S (PO uncertainty for S_8 of ~25%)
2. Know relative amounts of any 1 phase at all 48 points on all 3 cells
3. Cannot yet compare 3 phases to each other (total S amount)



Technical Accomplishments and Progress:
400 Wh/kg Li Metal Pouch Cell with >250 Cycling

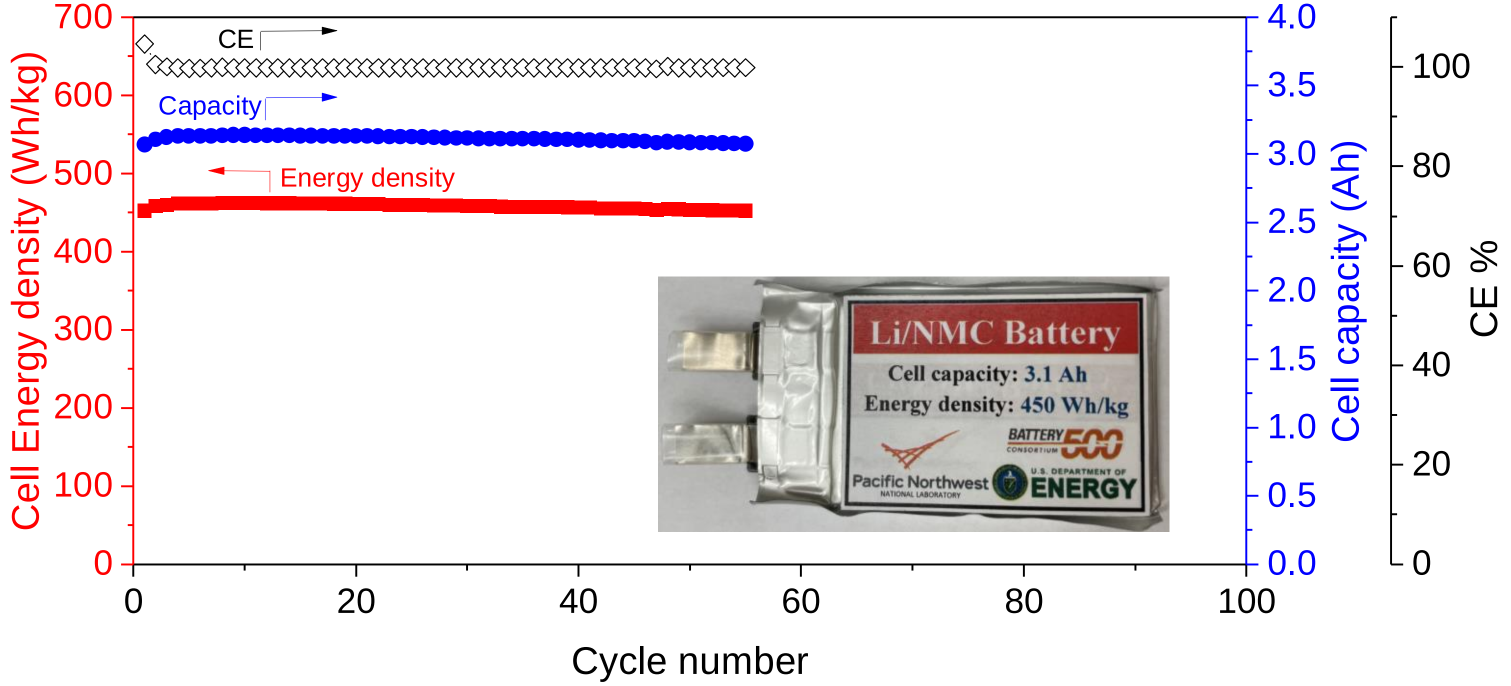


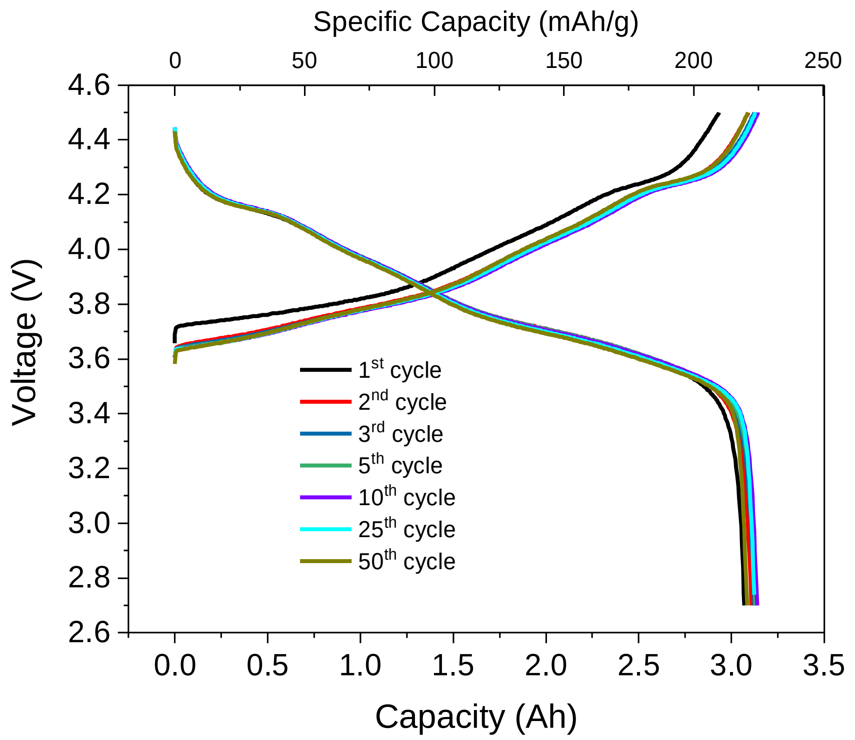


Formation: C/10 charge and discharge for 2 cycles
Subsequent cycling: C/10 Charge, 0.3C Discharge 2.7-4.4V @25° C

- Demonstration of 2.5 Ah Li metal pouch cell with 400 Wh/kg cell-level energy and >250 stable cycles.
- Minimum polarization for the first 250 cycles: a significant improvement

Technical Accomplishments and Progress:
Demonstration of 450Wh/kg Li/NMC Pouch Cell





- The most recent pouch cell achieves 460Wh/kg with minimum polarization.
- Cycling is in progress.

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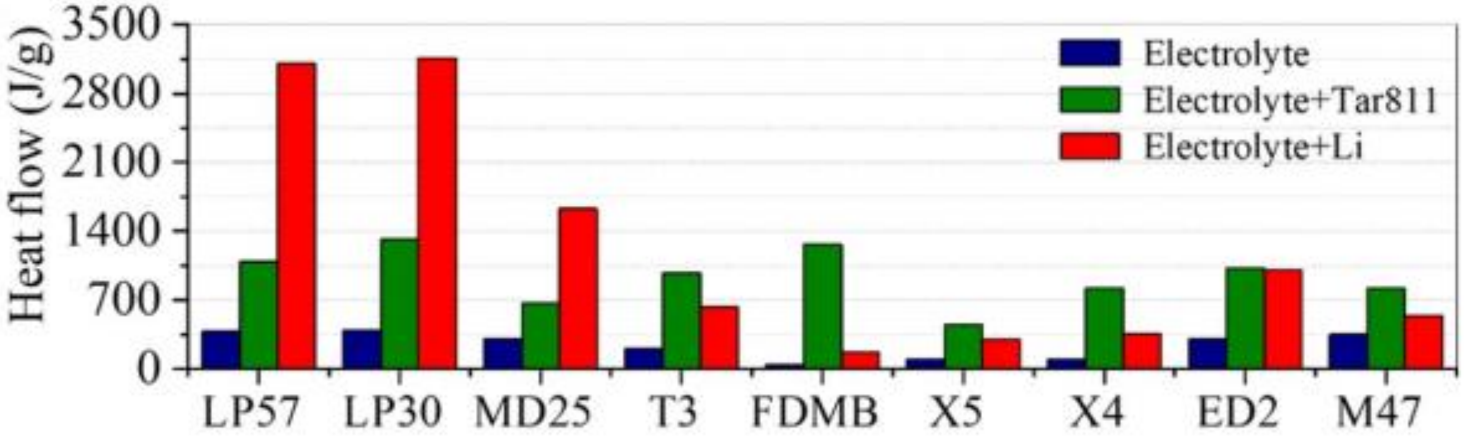
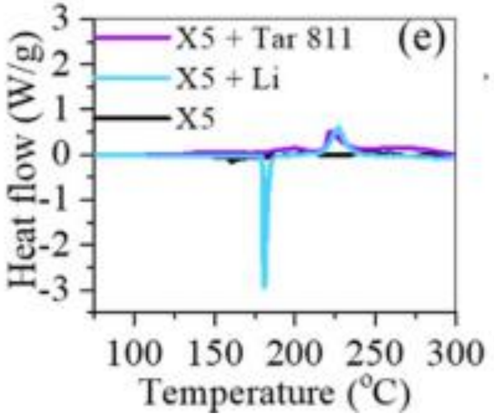
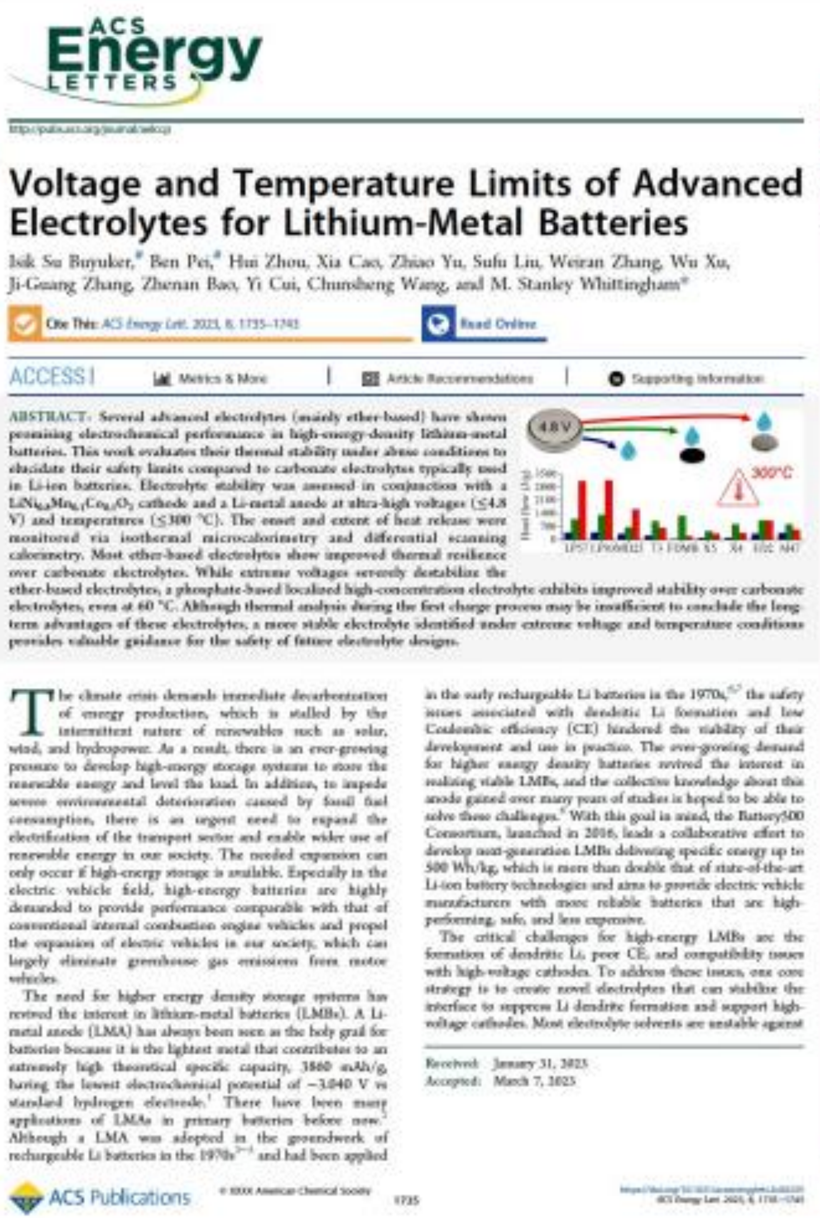
NMC811: Initial Stability Studies of Electrolytes Documented

- **Thermal and Voltage Stability – ex-situ DSC and operando DSC**
  - Li metal can be melted in all electrolytes
  - Ethers more stable than carbonates; all stable to 4.4 V at 30°C
  - **X5** (Stanford) has lowest reactivity and thermal evolution
  - M47 gives higher capacity retention than carbonate







ACS Energy Letters

Voltage and Temperature Limits of Advanced Electrolytes for Lithium-Metal Batteries

Isik Su Buyuker,# Ben Pei,# Hui Zhou, Xia Cao, Zhiao Yu, Sufu Liu, Weiran Zhang, Wu Xu, Ji-Guang Zhang, Zhenan Bao, Yi Cui, Chunsheng Wang, and M. Stanley Whittingham*

Cite This: ACS Energy Lett. 2023, 8, 1735–1743

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ABSTRACT: Several advanced electrolytes (mainly ether-based) have shown promising electrochemical performance in high-energy-density lithium-metal batteries. This work evaluates their thermal stability under abuse conditions to elucidate their safety limits compared to carbonate electrolytes typically used in Li-ion batteries. Electrolyte stability was assessed in conjunction with a $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ cathode and a Li-metal anode at ultra-high voltages (≤4.8 V) and temperatures (≤300 °C). The onset and extent of heat release were monitored via isothermal microcalorimetry and differential scanning calorimetry. Most ether-based electrolytes show improved thermal resilience over carbonate electrolytes. While extreme voltages severely destabilize the ether-based electrolytes, a phosphate-based localized high-concentration electrolyte exhibits improved stability over carbonate electrolytes, even at 60 °C. Although thermal analysis during the first charge process may be insufficient to conclude the long-term advantages of these electrolytes, a more stable electrolyte identified under extreme voltage and temperature conditions provides valuable guidance for the safety of future electrolyte designs.

The climate crisis demands immediate decarbonization of energy production, which is stalled by the intermittent nature of renewables such as solar, wind, and hydropower. As a result, there is an ever-growing pressure to develop high-energy storage systems to store the renewable energy and level the load. In addition, to impede severe environmental deterioration caused by fossil fuel consumption, there is an urgent need to expand the electrification of the transport sector and enable wider use of renewable energy in our society. The needed expansion can only occur if high-energy storage is available. Especially in the electric vehicle field, high-energy batteries are highly demanded to provide performance comparable with that of conventional internal combustion engine vehicles and propel the expansion of electric vehicles in our society, which can largely eliminate greenhouse gas emissions from motor vehicles.

The need for higher energy density storage systems has revived the interest in lithium-metal batteries (LMBs). A Li-metal anode (LMA) has always been seen as the holy grail for batteries because it is the lightest metal that contributes to an extremely high theoretical specific capacity, 3860 mAh/g, having the lowest electrochemical potential of −3.040 V vs standard hydrogen electrode.[1] There have been many applications of LMAs in primary batteries before now.[2] Although a LMA was adopted in the groundwork of rechargeable Li batteries in the 1970s[3–4] and had been applied in the early rechargeable Li batteries in the 1970s,[6,7] the safety issues associated with dendritic Li formation and low Coulombic efficiency (CE) hindered the viability of their development and use in practice. The ever-growing demand for higher energy density batteries revived the interest in realizing viable LMBs, and the collective knowledge about this anode gained over many years of studies is hoped to be able to solve these challenges.[8] With this goal in mind, the Battery500 Consortium, launched in 2016, leads a collaborative effort to develop next-generation LMBs delivering specific energy up to 500 Wh/kg, which is more than double that of state-of-the-art Li-ion battery technologies and aims to provide electric vehicle manufacturers with more reliable batteries that are high-performing, safe, and less expensive.

The critical challenges for high-energy LMBs are the formation of dendritic Li, poor CE, and compatibility issues with high-voltage cathodes. To address these issues, one core strategy is to create novel electrolytes that can stabilize the interface to suppress Li dendrite formation and support high-voltage cathodes. Most electrolyte solvents are unstable against

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BINGHAMTON UNIVERSITY

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Proposed Future Work

- Develop next generation high-energy, low-cost batteries for electric vehicles
- Address Li metal anode challenges at full cell levels and achieve more than 99.9% Coulombic efficiency (CE) for Li deposition
- Design, fabricate and validate high energy pouch cells up to 500 Wh kg^{-1} based on Li||NMC and Li||S
- Scale up pouch cell capacity up to 5-10 Ah
- Demonstrate long cycle life of up to 1,000 deep charge-discharge cycles
- Fabricate alternate low-cost and resource-secured Li||S cells of ~300 Wh kg^{-1} with up to 1,000 cycles
- Achieve total control of battery chemistries for robust, scalable and commercially viable technologies
- Perform cost analysis and develop strategies to significantly less than \$100 $(kWh)^{-1}$
- Independently validate cell performances.

Summary

Keystone Projects 1:

- Understand and push the limit of materials
- Develop stable electrode and electrolyte materials for high efficiency Li deposition/stripping
- New high capacity storage mechanisms

Keystone Project 2:

- Develop and fabricate high capacity electrodes
- Optimize electrode architectures and manufacturing for high energy cells

Keystone Project 3:

- Design, fabrication, testing and validation of pouch cells
- Protocols and systematic approaches for testing, validation, and safety
- Achieve stable cycling for 300, 350, 400, 450 Wh kg^{-1} pouch cells

Cross-Cutting Thrust:

- Develop capabilities for understanding, quantifying and predicting degradation and failure from materials to cells
- Reveal and control reaction heterogeneity in real cells

Program Level:

- Meet and exceed all milestones and deliverables
- Deliver significant improvement of cycling life for high energy pouch cells
- Obtain deep understanding of the degradation mechanisms

Integration and Collaboration

Materials suppliers; ABR: NMC cathodes and electrolytes

Materials suppliers; BMR: Li-S, Li metal, solid electrolytes.

Cell manufacturers; end users .

Keystone 1
Materials and Interfaces

Pushing materials limits and getting the most capacity

Keystone 2
Electrode Architectures

Maximizing materials utilization and improving stability

Keystone 2
Cell Design, Integration and Testing

Fabricating pouch cell and optimizing performance

Cross-cutting: solving Li metal challenge and controlling reactivity on all levels

DOE facilities: materials synthesis and scale-up (MERF), cell design, cost analysis, battery prototyping (ABF), state-of-the-art characterization and user facilities (NSLS-II, SLAC, EMSL, etc.).