High Energy Rechargeable Lithium-Metal Cells: Fabrication and Integration

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PACIFIC NORTHWEST NATIONAL LABORATORY
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2020 DOE VEHICLE TECHNOLOGIES PROGRAM
ANNUAL MERIT REVIEW

Project ID #: Bat369

Overview

Timeline

- Project start date: December 2016
- Project end date: October 2021
- Percent complete: 60%

Barriers

- Low energy: Li metal anode will boost cell energy
- Short battery Life: mitigating side reactions will extend the cycling stability

Budget

- Total project funding: $50M
  - DOE share: 100%
- Funding for FY 2019: $10M
- Funding for FY 2020: $10M

Partners

- Battery 500 PIs
- 3 national labs
- 10 universities
- GM, Albemarle, Umicore

Relevance/Objectives

- **Overall Objectives**
  - Overcome the fundamental issues in building high-energy rechargeable Li metal batteries
  - Demonstration of long-term cycling of 500 Wh/kg Li metal cells
- **Objectives of this period**
  - Identify the cell-level scientific challenges in high-energy rechargeable Li metal batteries: Li-S and Li/NMC
  - Demonstrate 350 Wh/kg Li metal pouch cells for at least 350 stable cycles
  - Demonstrate 400 Wh/kg Li metal pouch cells for 100 stable cycles
- **Impacts**
  - Accelerate the development of high-energy rechargeable Li metal batteries for future vehicle electrification

Milestones: Keystone Project 3 for Cell Fabrication, Testing and Diagnosis

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Milestones and Go/No-Go Decisions	Date	Status
Understand the mechanism of high S loading and cycling stability	12/31/2019	Completed (Jie Xiao)
Quantify the 3D morphology of lithium anode under various pressure	3/31/2020	Completed (Shirley Meng)
Quantify the impact of cycling conditions using a standard single layer pouch cell design (pressure, charge/discharge rates and T)	6/30/2020	On track (Eric Dufek)
1) Fabricate and test a pouch cell capable of 350 Wh/kg and 350 cycles 2) Fabricate and test a pouch cell capable of 400 Wh/kg and 100 cycles	9/30/2020	On track (Jie Xiao)

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Approach

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- For Li-S cells: Identify the key factors that limit the reversible cycling at pouch cell level
- For Li/NMC cells: Push the limit of cycling by tuning Li/electrolyte interface
- Investigate the impacts of pressure on Li metal cells
- Develop a safety protocol for studying high-energy Li metal cells from electrode preparation, cell assembly, testing and disposal

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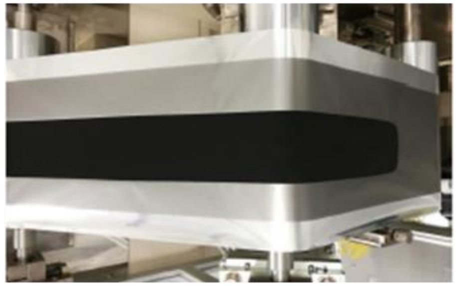
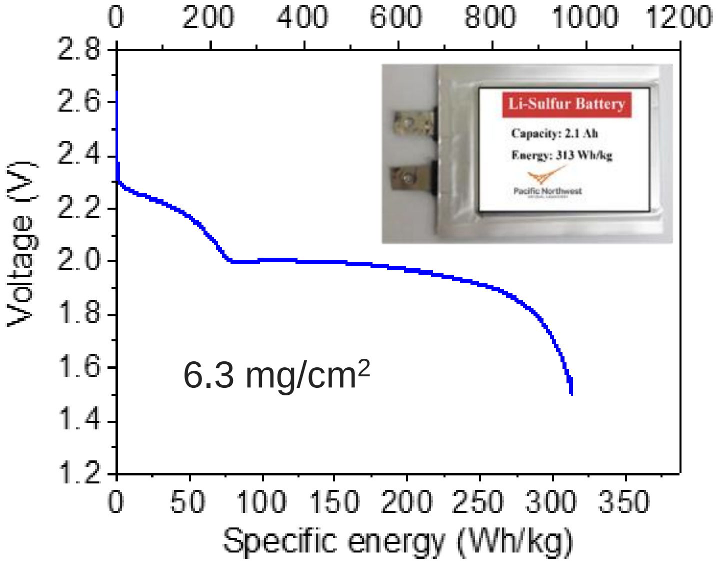
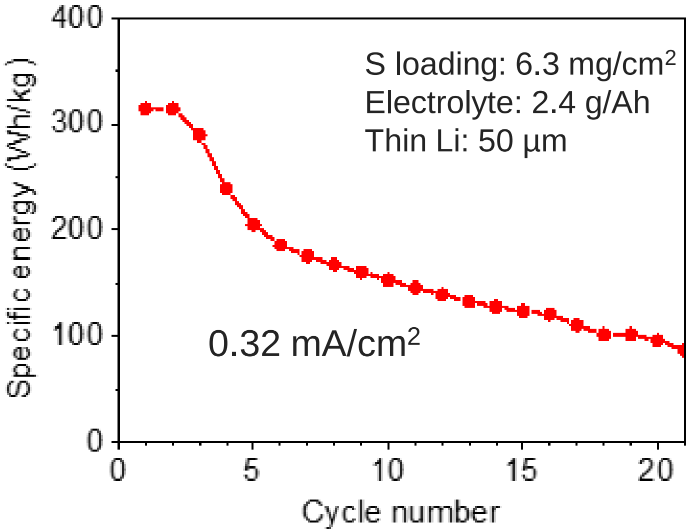
Technical Accomplishments:
Demonstration of High-energy Li-S

Large-scale
uniform S/C coating



Nano-structured micron-sized S/C composite with porosity control





- All S/C materials and electrodes are synthesized at PNNL and supplied to the entire consortium as baseline cathodes.
- Demonstration of 313 Wh/kg Li-S pouch cell: high S loading (6.3 mg/cm^2), lean electrolyte (2.4 g/Ah=2.5 m/g sulfur) and thin Li foil (50 µm).
- Cycling stability is poor: need to identify the dominant reason for limited cycling

Technical Accomplishments: The Amount of Electrolyte Determines Li-S Cycling Stability

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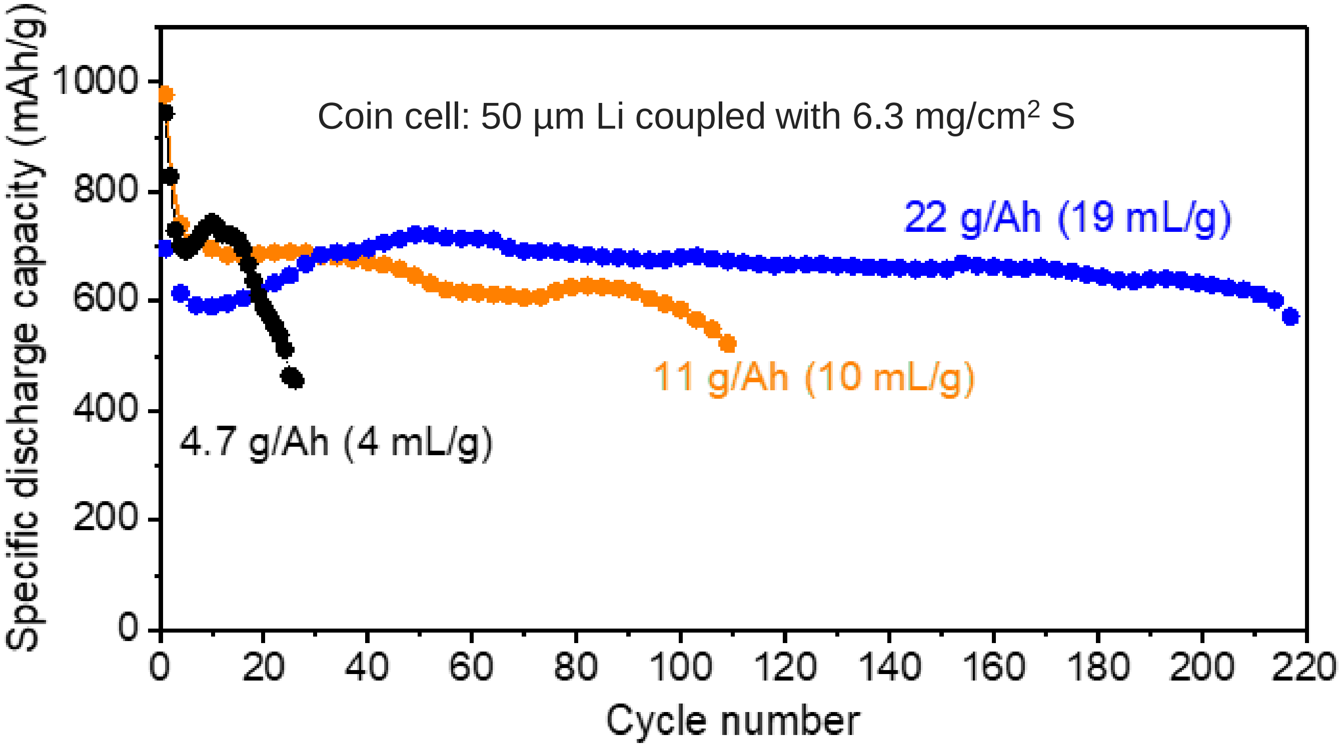

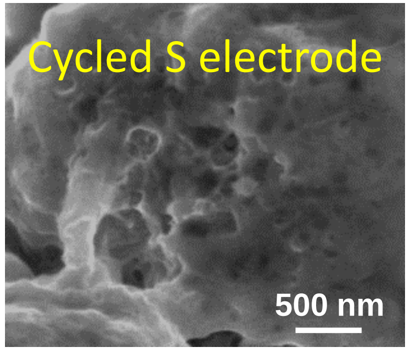

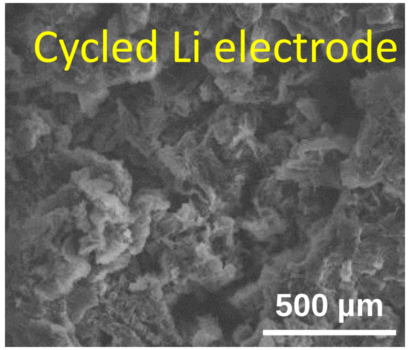

- Electrolyte depletion is faster in Li-S cells than in Li-NMC cells
  - Electrolyte reacts with BOTH polysulfide cathode and Li anode.
  - Electrolyte distribution in DENSE and high S loading cathode is inhomogeneous.
- 50 µm Li is sufficient for the first few hundreds of Li-S cycling: Electrolyte content determines how long the stable cycling lasts
- The main reason for poor cycling in pouch cells: very lean electrolyte (only 2.5 mL g^{-1})

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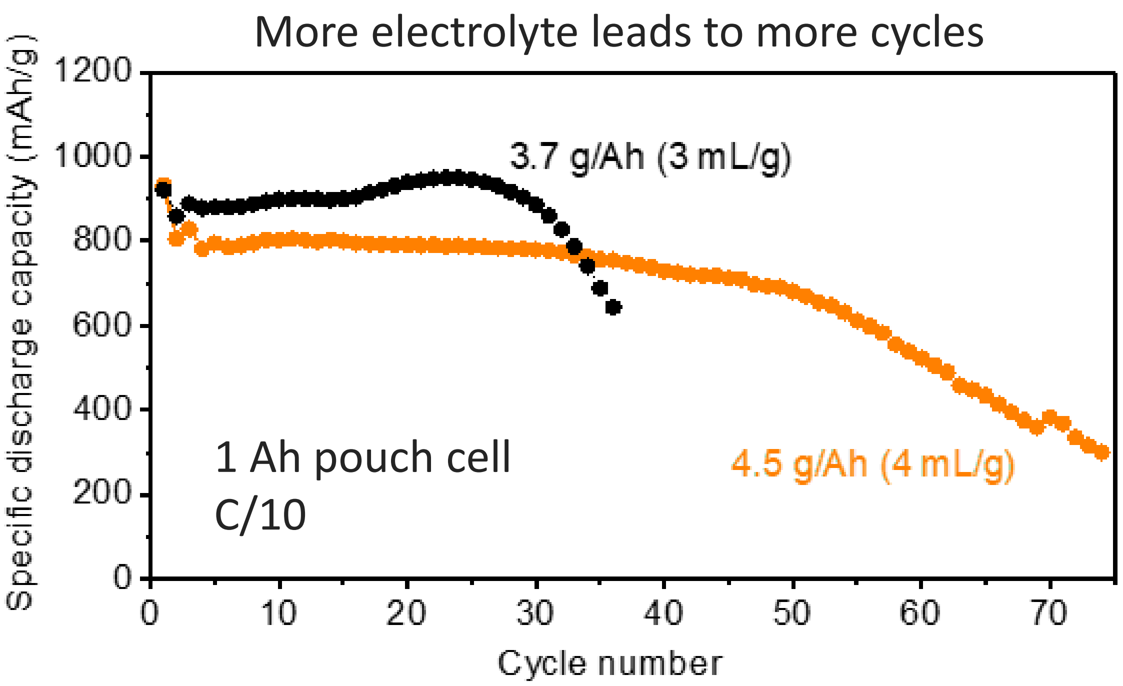
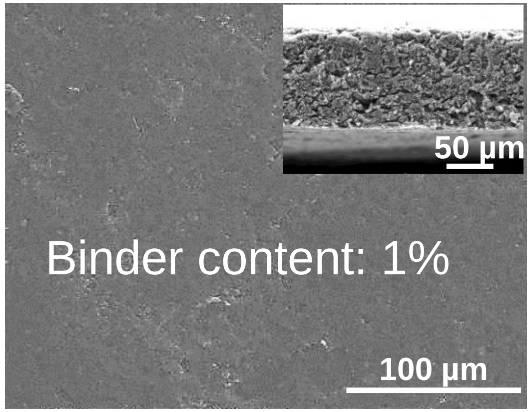
Technical Accomplishments: Maximizing Electrolyte Amount in Pouch Cell is the First Step to Extend Li-S Cycling

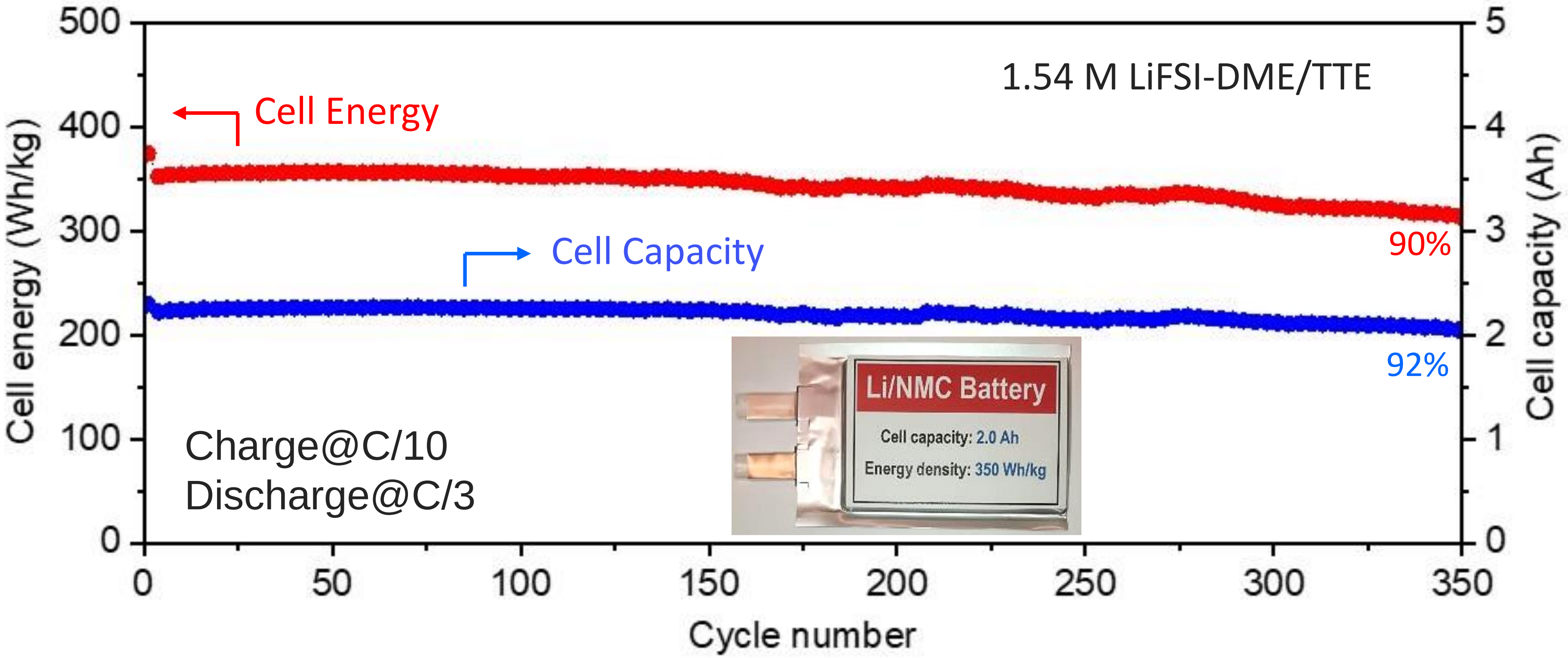


example to reduce parasitic weight



- **Save weight for electrolyte to extend Li-S cycling**
  - Increase S utilization so less S is needed to meet the same areal capacity
  - Reduce the “dead” weight e.g., binder content in the cathode
  - Reduce cathode porosity (<50%): minimize electrolyte intake on cathode side without sacrificing performance
- Reducing polysulfide crossover and Li metal stabilization are being investigated concurrently by other teams.

Technical Accomplishments: Stable Cycling of 350 Wh/kg Li/NMC622 Pouch Cell



- Prototyping Li metal pouch cells demonstrate stable cycling: >350 cycles with 90% capacity retention (still under testing).
- A great platform to accelerate Batt500 innovation: electrode architecture, electrolyte, cell design, cell balance etc.
- Prototyping pouch cells were also shipped out for independent validation.

Technical Accomplishments:
400 Wh/kg Pouch Cell Based on Li/NMC811

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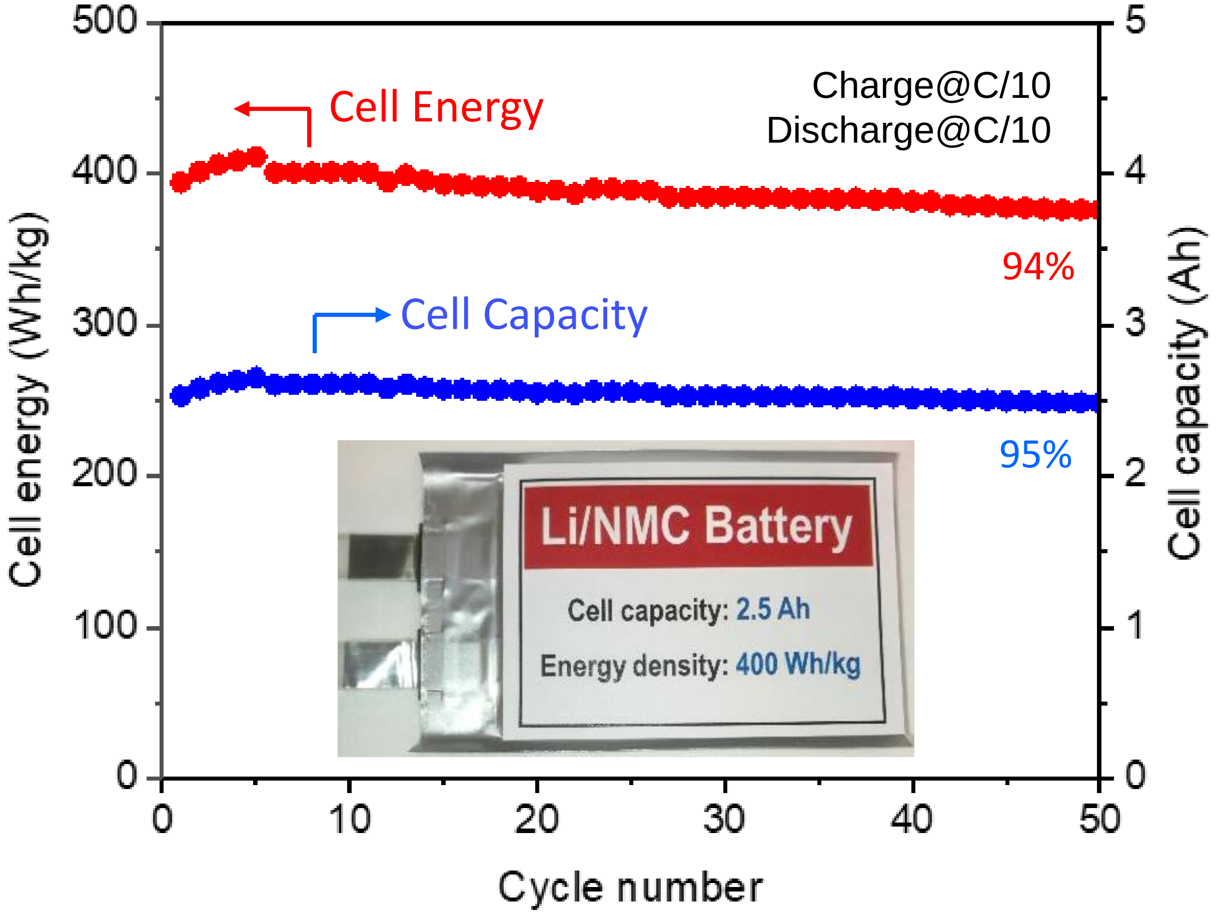

- 400 Wh/kg pouch cell demonstrates stable cycling (still under testing)

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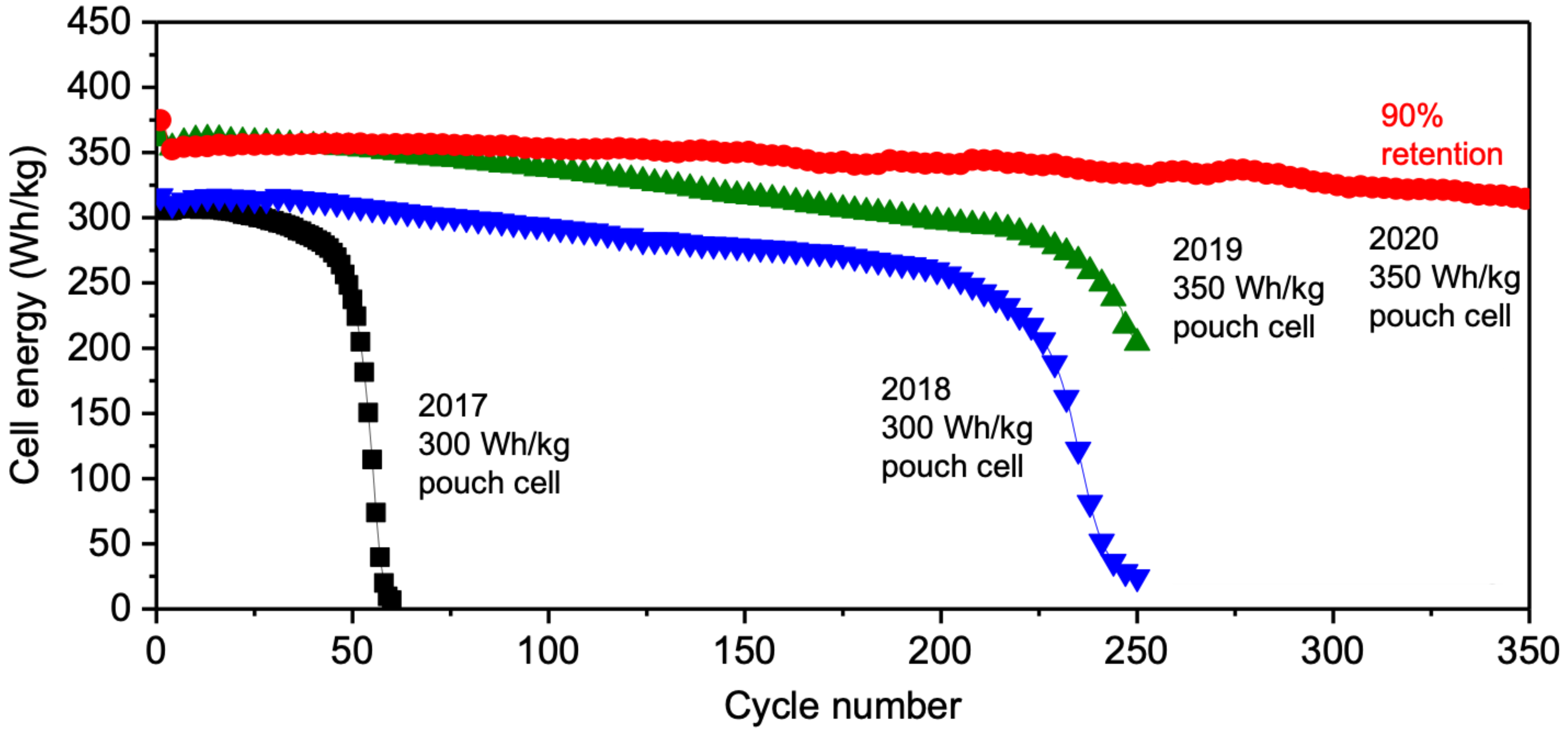
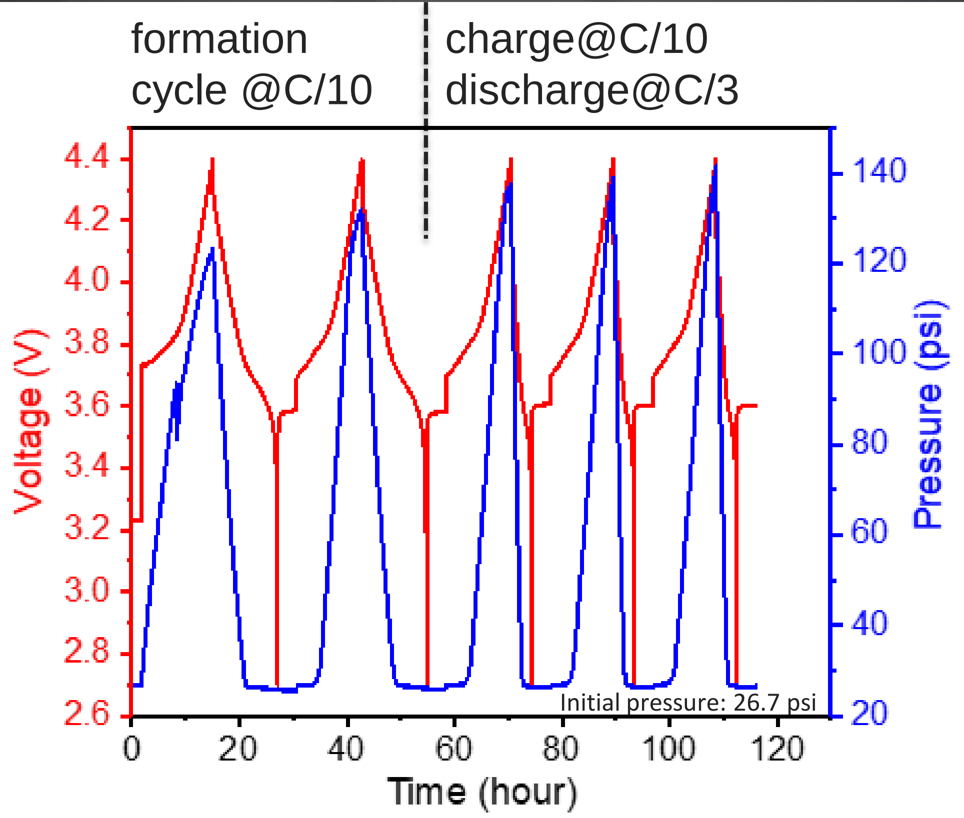
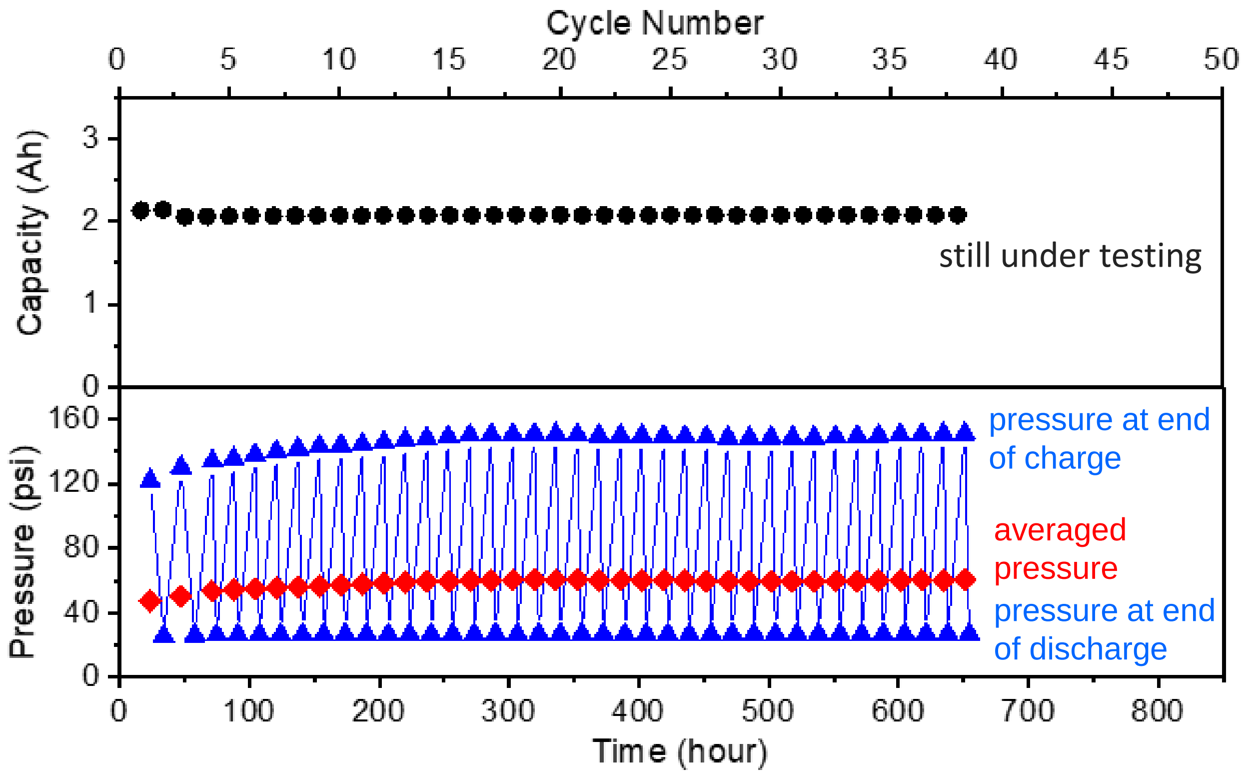
Technical Accomplishments:
Increasing Cycling and Reducing Cell Swelling



- Steady improvement of pouch cell cycling through integrated modification of electrode architecture, electrolyte recipe and cell design.
- Pouch cell swelling was significantly reduced from 110% (after only 10 cycles) to 35% (after stable 300 cycles).

Technical Accomplishments: *In Operando* Pressure Measurement during Pouch Cell Cycling





- Cell pressure change is consistent with reversible use of Li metal anode.
- Cell pressure slightly increases at the end of each charge and almost goes back to initial pressure (26.7 psi) at the end of each discharge for the first 37 cycles.
- Correlation between cell pressure change and “dead” Li buildup is underway.

Technical Accomplishments: Good Practices for Rechargeable Li Metal Batteries

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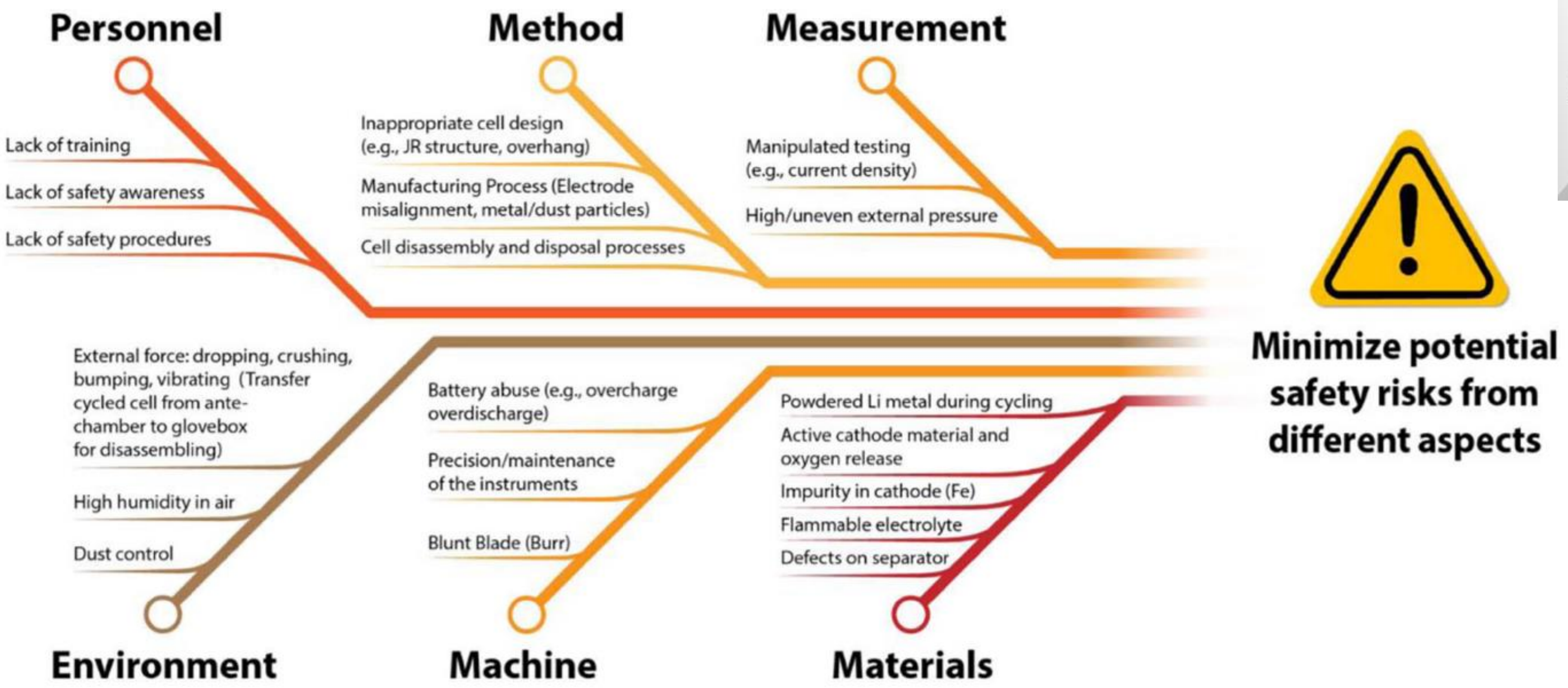

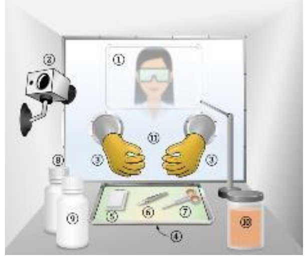

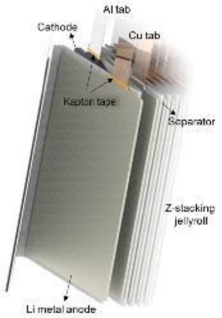

- Developed and shared a safety protocol to minimize safety risks of high-energy batteries (not just Li metal cells): slurry mixing, electrode design, cell fabrication, cell screening, testing, disassembly and disposal

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Responses to Previous Years Reviewers' Comments

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- This project was not reviewed last year.

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Collaboration and Coordination with Other Institutions

- Industry:
  - General Motors
  - Albemarle
  - Umicore

- University:
  - SUNY Binghamton: materials selection
  - Univ. Washington: separator coating
  - UC San Diego: testing on PNNL electrolytes
  - Univ. Pittsburg: supplied S/C composite for electrode coating
  - Penn State Univ.: testing of thick NMC and S electrodes made at PNNL
  - Univ. Houston: testing of PNNL new electrolyte
  - Stanford/SLAC: electrodes and electrolyte testing
  - UT Austin: Supplied high-Ni NMC to PNNL for evaluation
  - Univ. Maryland/Army research Lab: electrolyte development

- National Laboratory
  - Idaho National Lab: independent testing of PNNL-made pouch cell; co-develop safety protocol
  - Brookhaven Nation Lab: characterization of PNNL fabricated electrodes/electrolytes
  - SLAC: new electrolyte characterization

Remaining Challenges and Barriers

- Push the cell energy towards 500 Wh/kg with stable cycling by integrating new concepts from Battery500 Consortium
  - Coin cell protocol developed in FY18 helps much to accelerate coin cell-to-pouch cell transition
- Balance of high energy and cycle life of Li metal cells
- Dendrite-induced cell shorting
  - C/10 charging is used for now to decouple cell shorting and cell failure caused by Li/electrolyte depletion.

Proposed Future Work

- Li-S pouch cell design to balance energy and cycling
- Further improve cycling of 400/350 Wh/kg Li pouch cells
- Stabilize interfacial reaction between Li and electrolyte
- Study pressure impacts in pouch cell cycling

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

Summary

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- Identified the key parameters that limits the cycling stability of high-energy Li-S cells.
- Successful demonstration of 350 Wh/kg and 400 Wh/kg Li pouch cell with stable cycling
- *In operando* cell pressure measurement has been developed to monitor and understand pouch cell pressure changes upon cycling and its correlation with Li microstructure evolution.
- Developed safety protocol of rechargeable Li metal batteries being shared with research community and industry

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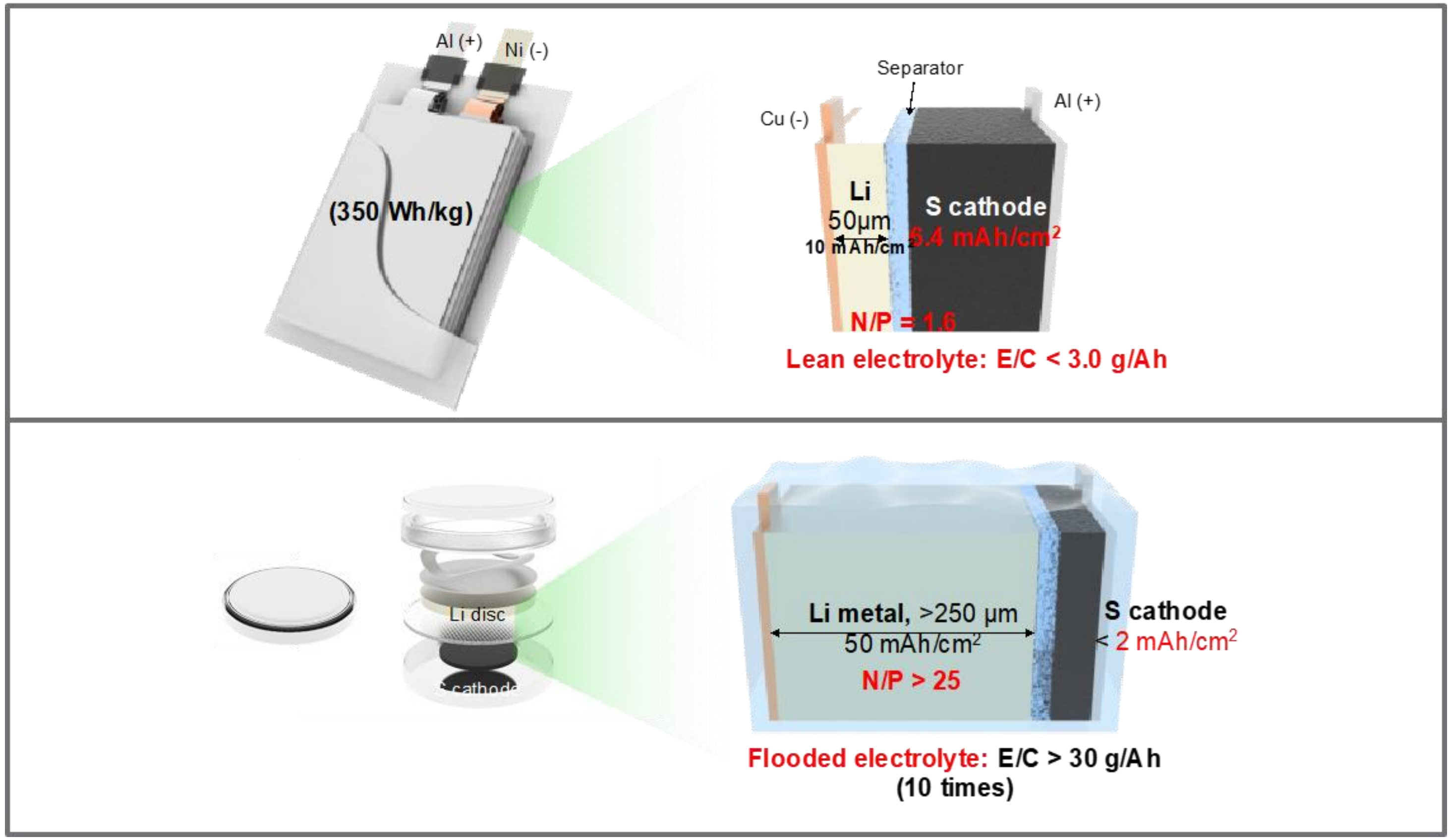
Acknowledgement

- DOE/EERE/VTO: Battery500
- Key contributors: C. Niu, D. Liu, D. Lv, L.Shi, A. Baranovskiy, B.Wu, W. Xu, J. Zhang, J. Liu
- Battery500 PIs and their teams

Technical Backup Slides

Technical Backup:
Li-S Cell Cycling is "Manageable"



- Three key parameters are drastically different: cathode areal capacity, N/P ratio and electrolyte amount
- A standard coin cell testing protocol has been developed for Batt500 consortium.

Technical Backup:
Discharge Curves S cathode with 1% Binder

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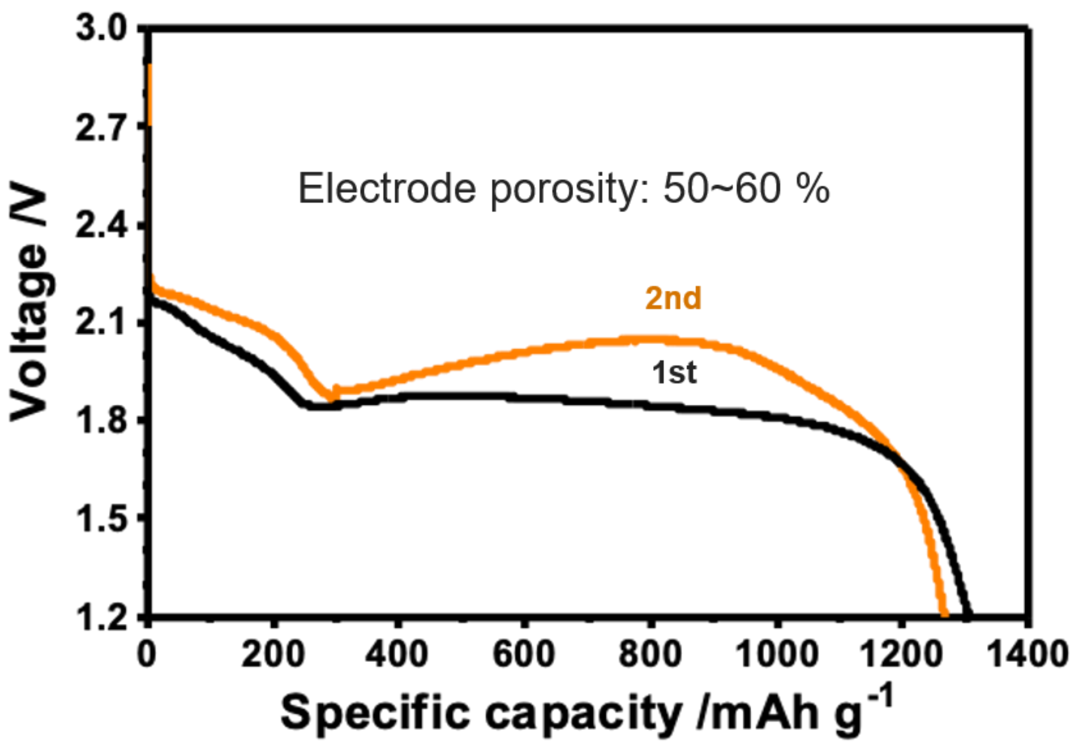

- With only 1% binder and reduced porosity, the utilization of S is still quite high: 1300 mAh/g.
- An "activation" process is observed presumably assigned to the improved wetting during the 2nd cycling.

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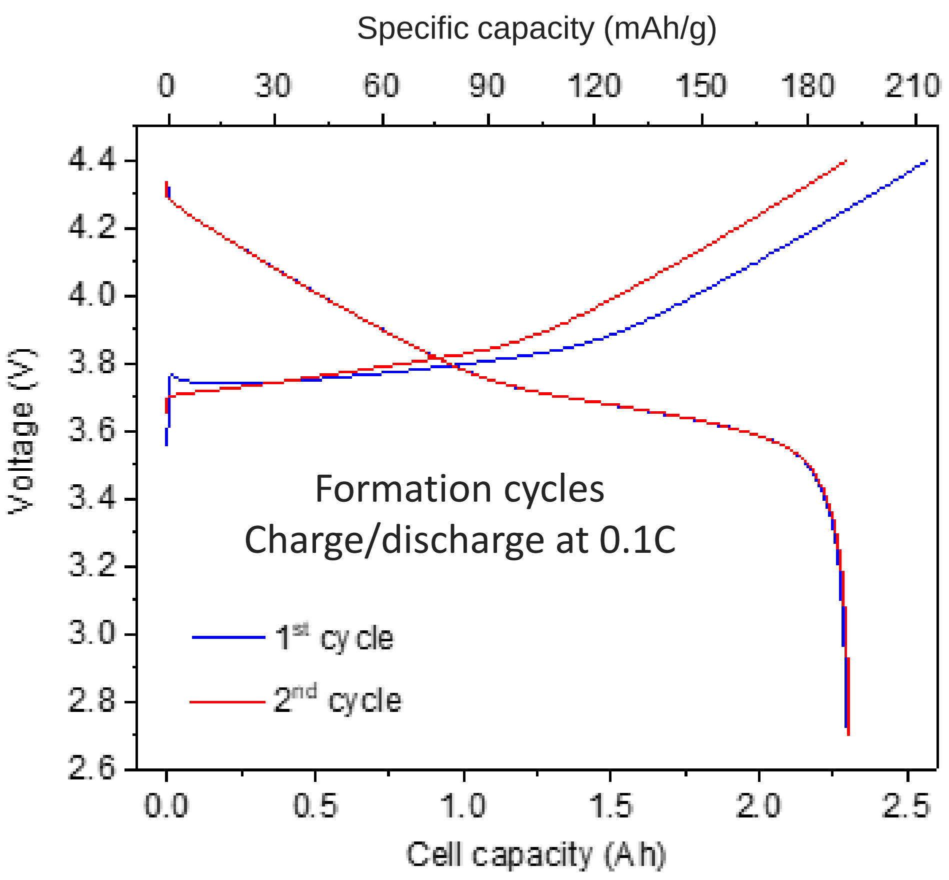
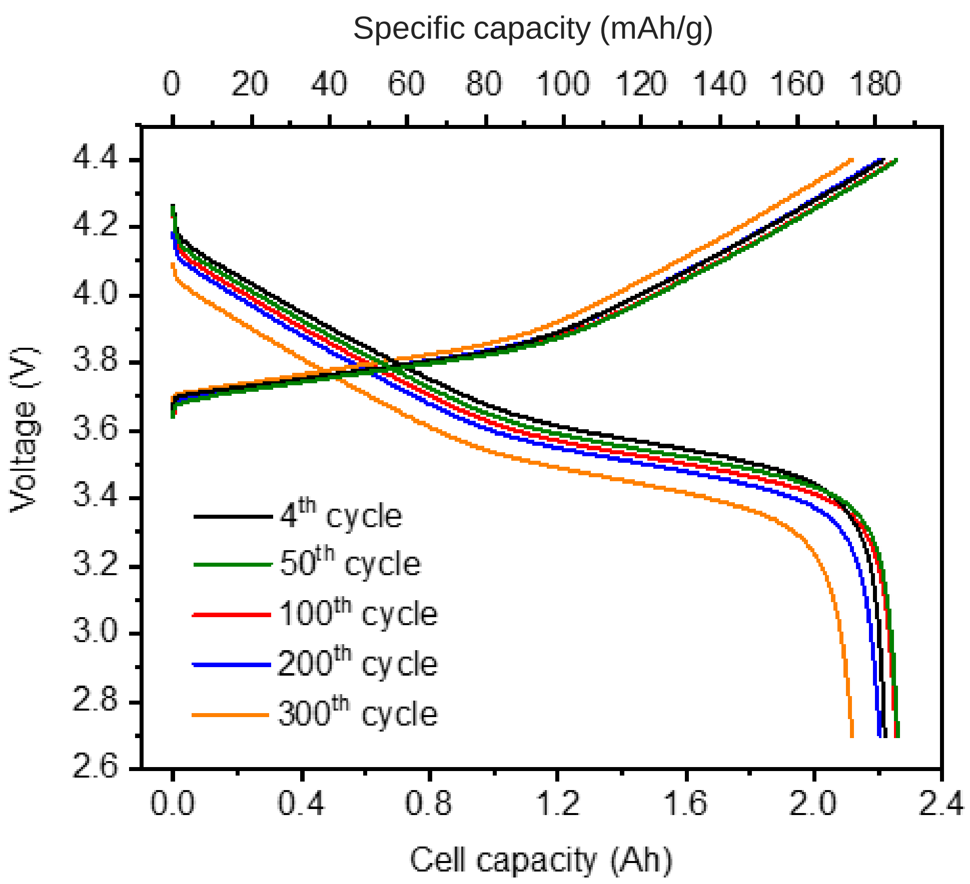
Technical Backup: Voltage Profile of 350 Wh/kg Li/NMC622 Pouch Cell





- Good utilization of NMC622 in thick and dense cathode (22.4 mg/cm^2 and 35% porosity)