High Energy Rechargeable Lithium-Metal Cells: Fabrication and Integration

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JUNE 25TH, 2021

2021 DOE VEHICLE TECHNOLOGIES PROGRAM
ANNUAL MERIT REVIEW

Project ID #: Bat369

Overview

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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 (For the entire Consortium
  - DOE share: 100%
- Funding for FY 2020: $10M (for Consortium)
- Funding for FY 2021: $10M (for Consortium)

Partners

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

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Relevance/Objectives

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- **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 450 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

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

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Milestones and Go/No-Go Decisions	Date	Status
Bench mark Li anode architecture with 50 micron Li anode using protocols for 350 Wh/kg cells and achieve over 200 cycles in coins	12/31/2020	Completed
Optimizing pressure effect for Li-S	3/31/2021	Completed
Provide new electrolyte formulation for Li-S	6/30/2021	On track
1) Fabricate and test a pouch cell capable of 350 Wh/kg and 450 cycles 2) Fabricate and test a pouch cell capable of 400 Wh/kg and 100 cycles 3) Demonstrate >450 Wh/kg pouch cell 4) Demonstrate good cycling of Li-S pouch cell 300 Wh/kg	9/30/2021	On track

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Approach

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- For Li-S cells: build a dense S/C electrode with minimum parasitic weight and reduced porosity for high-energy Li-S pouch cells
- For Li/NMC cells: balance interfacial reactions to achieve long cycle life
- Develop ML-assisted software to accelerate Li metal pouch cell design and research

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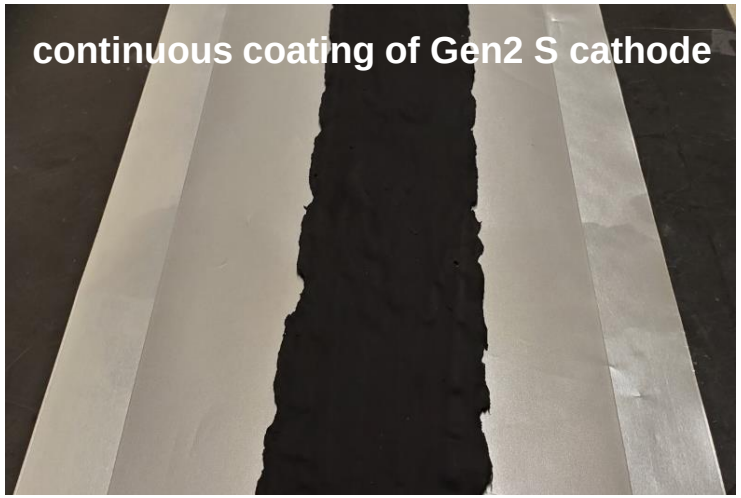
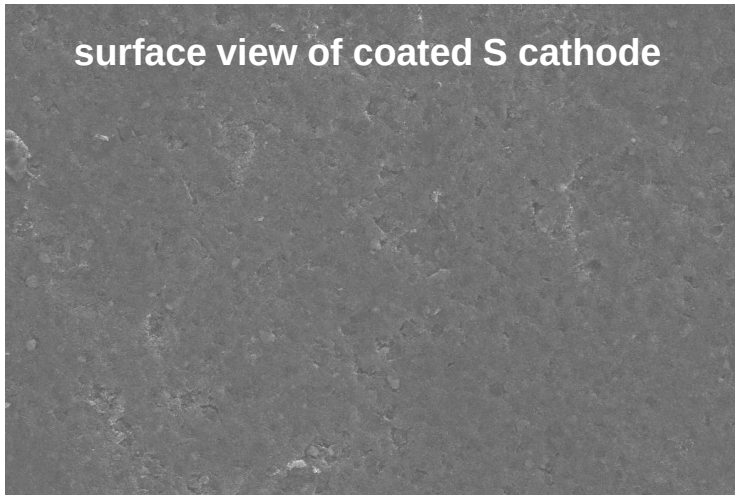
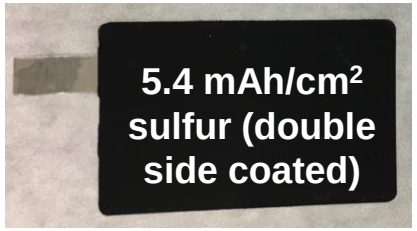
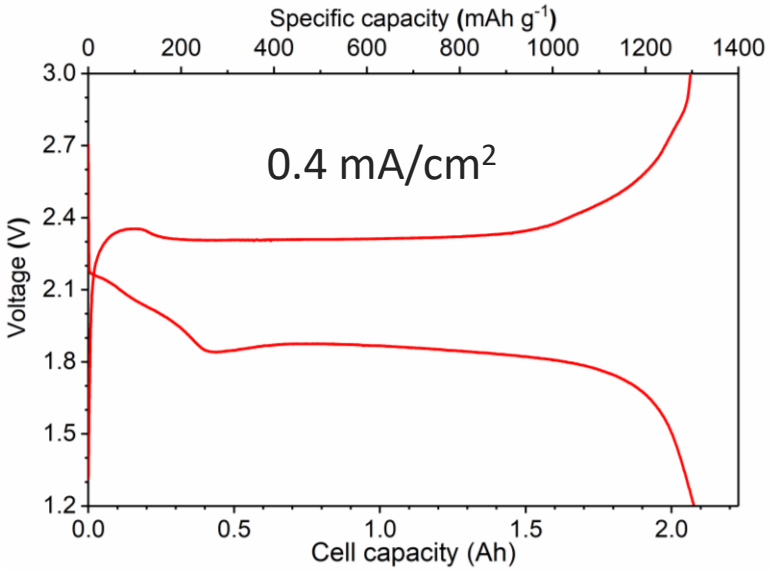
Technical Accomplishments: Gen2 Sulfur Cathode with Reduced Porosity and High Sulfur Content



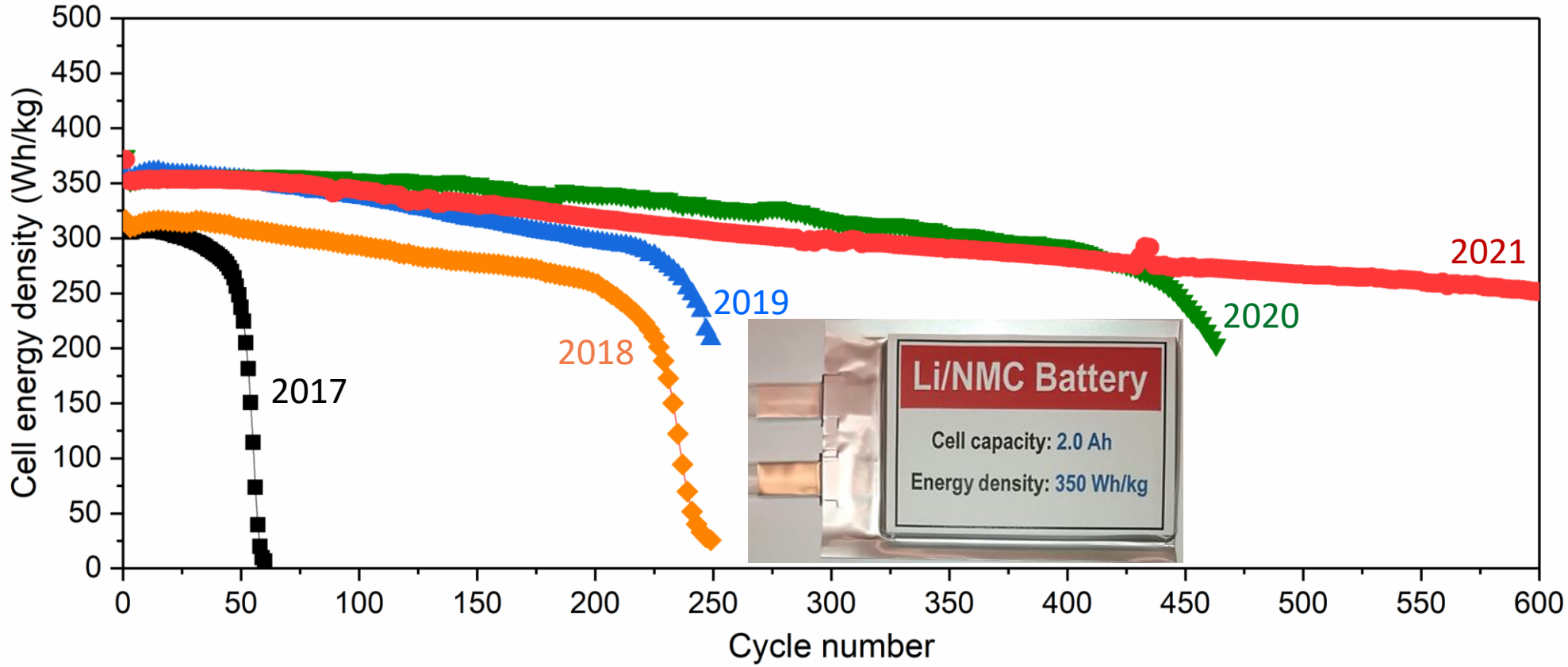






- Gen2 S cathode has >70% S in the entire cathode and reduced porosity to 50-62% to minimize electrolyte intake within cathode.
- Dense Gen2 S cathode still delivers ~1300 mAh/g capacity during the first discharge, building a good foundation for high-energy Li-S pouch cells.
- Patents on S electrode and electrolyte additives are licensed for evaluation by large electronics company.

Technical Accomplishments: 350 Wh/kg Li/NMC622 Pouch Cell Achieved 600 Stable Cycles with 76% Energy Retention



- In an appropriated designed pouch cell structure, three major factors impact Li metal cycle life:
  - Li thickness
  - electrolyte recipe and amount
  - continuous cell impedance increase due to SEI accumulation: wet SEI vs. dry SEI
- Slowing down the side reactions to retard SEI accumulation: balancing the relative amounts of Li and electrolyte

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

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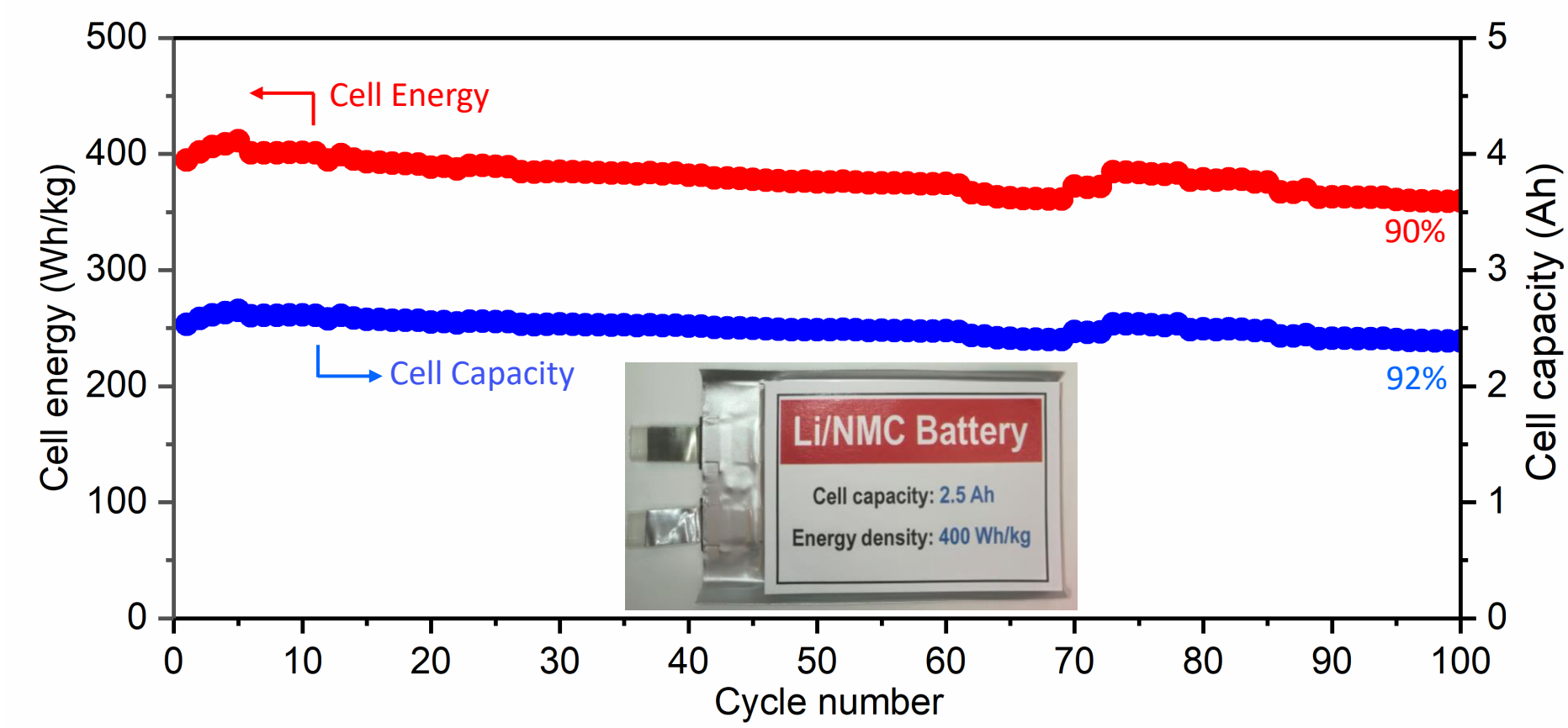

- 400 Wh/kg pouch cell demonstrates stable cycling (still under testing).
- The weights of all cell components are included.

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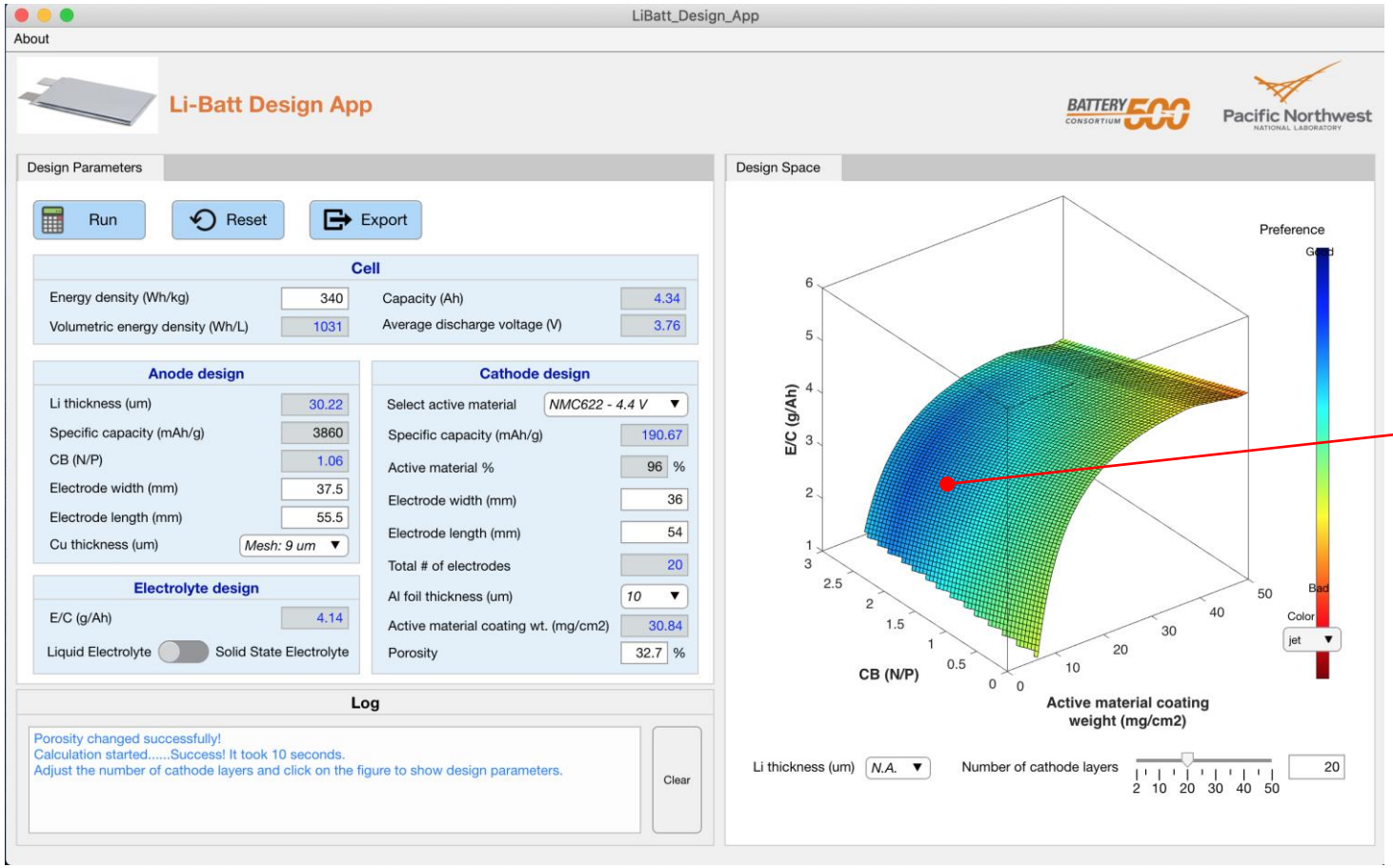
Technical Accomplishments: ML-Assisted Li-Batt App Facilitates Li Metal Battery Research at Industry Relevant Scales



more details here: https://www.pnnl.gov/news-media/building-better-battery-faster

Export Li Pouch Cell Design Table

	Parameters	
Cathode	Active material	NMC622-4.4 V
	Specific Capacity (mAh/g)	190.7
	Active material %	96%
	Electrode width (mm)	36
	Electrode length (mm)	54
	The total number of Cathode Electrodes	20
	The total number of Al foil	17
	Al foil thickness (um)	10
	Al density (g/cc)	2.7
	Active Material Coating weight (mg/cm2)	30.8
	Porosity (%)	33%
Electrolyte	Electrolyte/capacity (g/Ah) (E/C)	4.1
Anode	Active Material	Li
	Specific Capacity (mAh/g)	3860
	Li thickness (um)	30.2
	CB (N/P)	1.1
	Electrode width (mm)	37.5
	Electrode length (mm)	55.5
	Cu thickness (um)	9
	Cu areal density (mg/cm2)	4.8
	Inactive components(Separator, packaging, Tab)	2.2
Cell	Average discharge voltage (V)	3.8
	Capacity (Ah)	4.3
	Energy density (Wh/kg)	340
	Volumetric energy density (Wh/L)	1031.1

- Provide a fast understanding of realistic Li metal pouch cells without having to access pouch cell facility
- Accelerate innovation at relevant scales and help battery prototyping and manufacturing
- Licensed to many institutions and companies

Responses to Previous Years Reviewers' Comments

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1. "Why the team is focusing on gravimetric energy density when volumetric energy density is more important for vehicles."
   - *We consider both but only mentioned gravimetric energy density in the presentation. For 350Wh/kg reported in this presentation, the volumetric energy density is 780 Wh/L.*
2. "..there is mention of the final result of its work but not the approach or the details of how the accomplishment was achieved."
   - *Per the guidelines from export control office, for cells >350Wh/kg energy, we can only report performances without disclosing any detail.*
3. How many cycles at what C levels could be achieved at 300 Wh/kg?
   - *At the same C/3 discharge rate, more cycling is expected in 300 Wh/kg cells because of the additional electrolyte.*
4. The reviewer's response was that there is no theory put forth as to why more electrolyte results in better cycle life but not higher capacity per gram of S.
   - *It will help both but the intention is to save more electrolyte to extend Li metal cycling in Li-S cells.*
5. The reviewer observed that the team also wants to reduce the porosity, which seems to go against a higher electrolyte concentration.
   - *There is no plan to use concentrated electrolyte for Sulfur cathode with high mass loading.*

Collaboration and Coordination with Other Institutions

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- 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

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Remaining Challenges and Barriers

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- Push the cell energy towards 500 Wh/kg with stable cycling by integrating innovation and engineering
  - Baseline performance will accelerate the innovation
- 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.

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Proposed Future Work

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- Li-S pouch cell design to balance cell energy and cycling
- Move towards 500 Wh/kg Li metal pouch cells
- Improve the utilization of both lithium metal and electrolyte
- Complete the pressure study to further extend the cell cycling

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

Summary

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- Improved S% and reduced electrode porosity to minimize electrolyte intake within cathode side without sacrificing S utilization.
- Demonstrated 350 Wh/kg and 400 Wh/kg Li pouch cell with stable cycling
- Developed Li metal pouch cell design software to accelerate research innovation and support industry for Li metal cell prototyping and manufacturing

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Acknowledgement

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

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Technical Backup Slides

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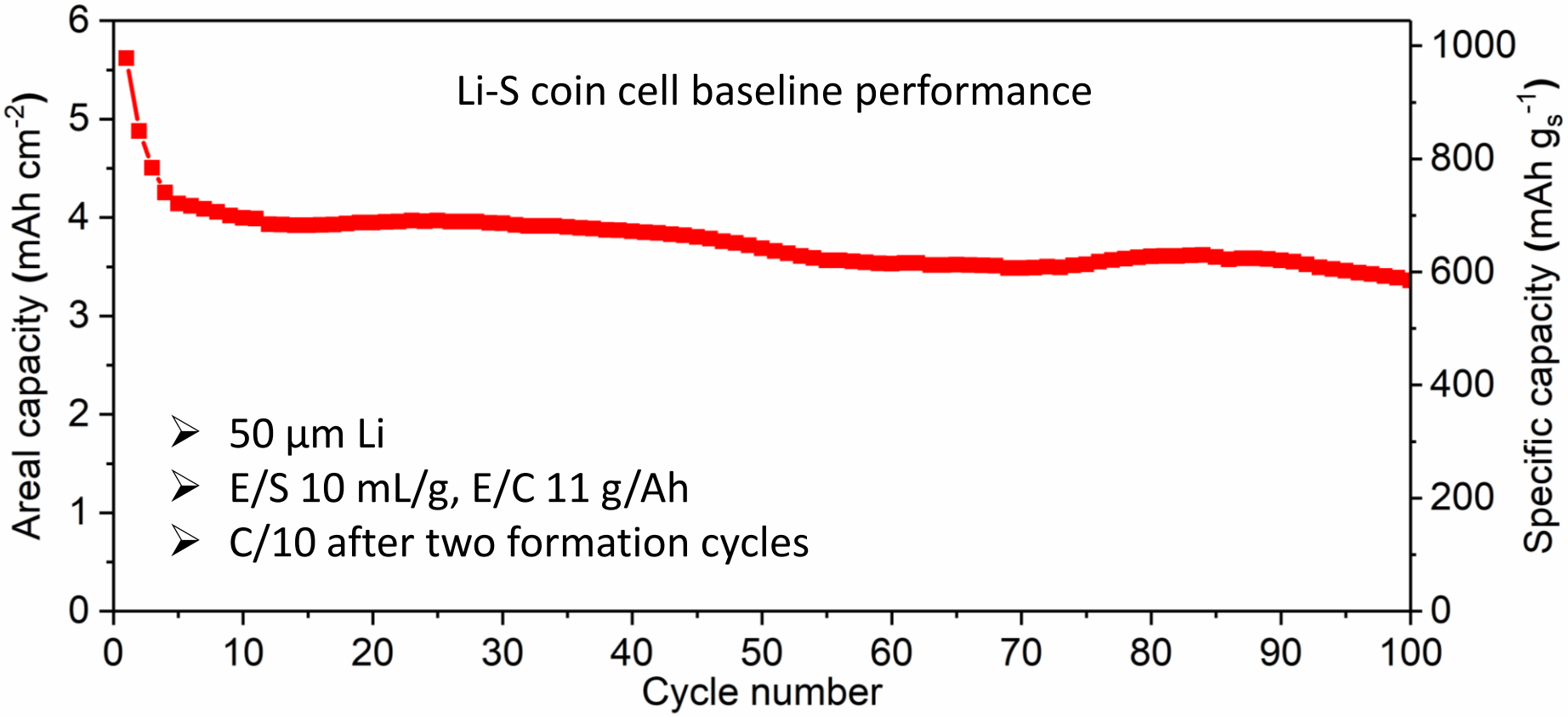
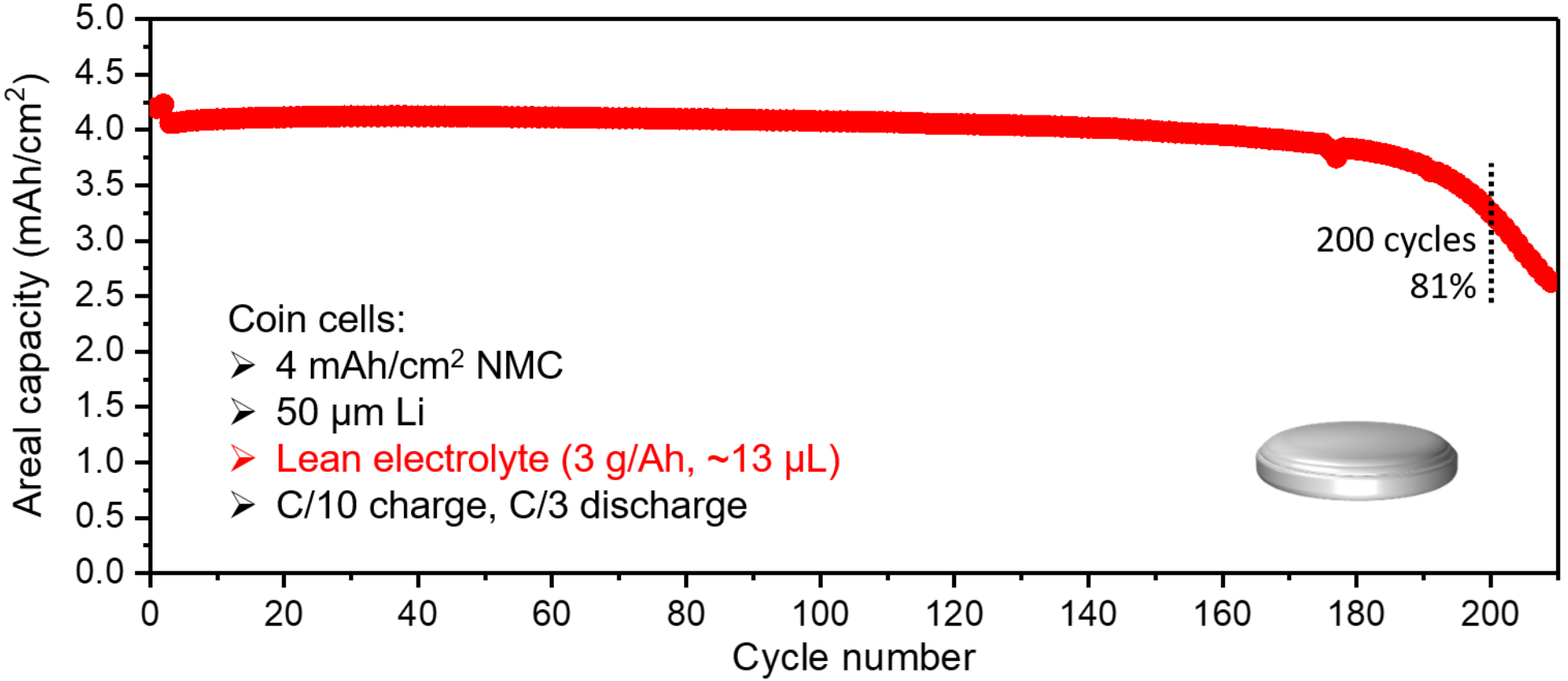
Technical Accomplishments: Established Li-S Coin Cell Testing Protocol and Baseline Performance for Benchmarking



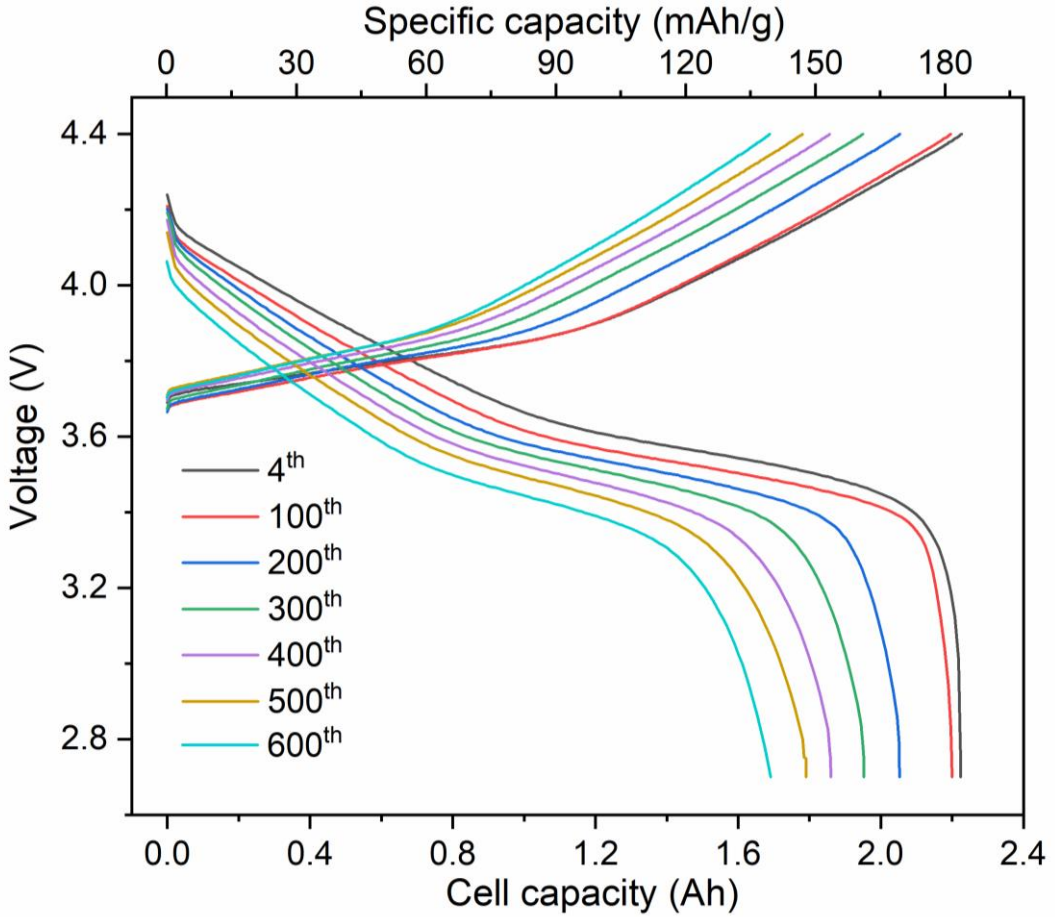
- Li-S coin cell protocol has been updated to ensure testing are conducted at similar conditions and results can be reproduced across different teams in the consortium.
- Effective new concepts need to surpass the baseline performance of Li-S coin cells.

Technical Backup:
Li/NMC Coin Cell Baseline Performance



- FY21 Li/NMC coin cell protocol has been updated
- Baseline performances of Li/NMC tested using the protocol has been provided to the consortium for benchmarking.

Technical Backup:
Pouch Cell Polarization Increases after Extensive



- Upon extensive cycling, cell polarization gradually increases in Li pouch cells due to the SEI buildup and impedance increase.