Stanford University

Advanced Electrolytes for Extreme Fast Charging

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Annual Merit Review

DOE Vehicle Technologies Program

Washington, DC

13 June, 2019

BAT401

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Overview

Timeline

- Start Date: Q1 FY19
- End Date: Q4 FY20
- Percent complete: 25%

Barriers

- Barriers addressed
  - Enable extreme fast charging at 10 minutes
  - Develop high conductivity electrolyte and additive package
  - Develop advanced diagnostics to validate advanced electrolytes

Budget

- Total project funding: 100% DOE
- FY19 Funding: $ 750K

Partners

- PIs: Chueh, Toney, Cui, Kaschmitter, Weker, Kee
- Collaborators:
  - Victor Koch, Covalent Associates
  - Konstatin Tikhonnov

Objectives & Relevance

Objectives: Develop an advanced electrolyte and four-step charging protocols that enable extreme fast charging on thick graphite-based anodes. Together with full-cell modeling, state-of-the-art X-ray and cryogenic electron microscopy (cryo-EM) characterization, we will:

(1) Tailor an advanced electrolyte paired with commercial graphite-based anodes to enable extreme fast charging in 10 minutes to 80% state-of-charge.
(2) Optimize the charging protocol to achieve 500 cycles with less than 20% capacity fade.
(3) Understand the impact of extreme fast charging on the battery components, and understand
and control Li plating/dead Li formation in full cells throughout extended cycling.
(4) Exhaustively validate results using not only through battery cycling but also in-situ X-ray
and ex-situ cryo-EM characterizations of full cells.

Objectives & Relevance

Relevance:

- Tomorrow's electric vehicles will require superior extreme fast-charging capability without compromising energy density and cost.
- Withcurrent state-of-the-art Li-ion battery technology, the general trend to reach energy densities >200 Wh/kg has been to design thicker, more dense electrodes. This poses a significant barrier to extreme fast charging since it is difficult to charge these thick electrodes at high rates without significant degradation. High rate charging on these anodes results in Li plating/dead Li, which can lead to graphite delamination, unacceptably high temperatures, and other irreversible side reactions such as the uncontrolled growth of solid electrolyte interphase.

Objectives & Relevance

Impact:

- Demonstrate extreme fast charging in 2Ah cells capable of achieving 500 cycles with less than 20% capacity fade.
- Provide new knowledge of lithium plating in full cells throughout extended cycling, a key failure mechanism in fast charging protocols.
- Enable development of EVs and PHEVs capable of roadside quick charging or rapid charging for cross country trips.

Milestones

Tasks	MS #	Task or subtask	Quarters 1	2	3	4	5	6	7	8
1.0		**Electrolyte development and coin cell testing**								
1.1		Prepare baseline electrolyte								
	1.1.1	Deliver baseline electrolyte	▲							
1.2		Adjust electrolyte composition								
	1.2.1	Report results of half cell testing and changes in electrolyte composition	▲	▲						
	1.2.2	Prepare modified electrolyte for use in nine 2Ah cells				▲				
	1.2.3	Report results of half cell testing and changes in electrolyte composition					▲	▲		
	1.2.4	Prepare optimized electrolyte for use in 18 2Ah cells								▲
2.0		**Physically based modeling**								
2.1		Quasi-1D submodels of cell assembly								
	2.1.1	Update cell architecture and materials definition in submodels	▲		▲		▲		▲	
2.2		3D cell-scale models								
	2.2.1	Update 3D models with refined quasi-1D models	▲		▲		▲		▲	
2.3		Develop empirical Li-plating correlations Li plating predictions								
	2.3.1	Update correlations based on advanced characterization			▲			▲		
2.4		Predict cell performance using continuously updated models								
	2.4.1	Report results from performance predictions			▲			▲		
3.0		**Full cell optimization**								
	3.1	Identify promising electrode materials and formulations								
	3.1.1	Deliver 5 multi-layer cells with baseline electrolyte to SLAC	▲							
	3.1.2	Report results from fast charging in half cells with baseline electrolyte		▲						
	3.2	Finalized materials section for full cells								
	3.2.1	Report fast charging in full single-layer cells with high-rate electrolyte			▲					
	3.2.2	Deliver 206 full single-layer cells with high-rate electrolyte to SLAC				▲				
	3.2.3	Deliver 200 full single-layer cells with high-rate electrolyte to SLAC				▲				
3.3		Optimize cell design in 2Ah cells								
	3.3.1	Fast charging in full multi-layer cells with high-rate electrolyte				▲				

Milestones

	3.3.2	Deliver 25 full multi-layer cells with high-rate electrolyte to SLAC					▲			
Go/No Go		Deliver 9 2Ah cells with high-rate electrolyte to DOE					■			
	3.3.3	Deliver 6 full multi-layer cells with high-rate electrolyte to SLAC						▲		
3.4		Build deliverable 2Ah cells with final extreme fast charging technology								
	3.4.1	Build 36 2Ah cells ready for test and ship								▲
Go/No Go		Deliver 18 2Ah cells with high-rate electrolyte to DOE								■
4.0		**Cryo-EM characterization**								
4.1		Optimize anode thin slice preparation								
	4.1.1	Report optimized thin sectioning protocol	▲							
4.2		Characterize full cells with baseline electrolyte								
	4.2.1	Report cryo-EM characterization on cycled anodes				▲				
4.3		Characterize full single-layer cells with high-rate electrolyte								
	4.3.1	Report cryo-EM characterization on cycled anodes						▲		
4.4		Characterize full multi-layer cells with high-rate electrolyte								
	4.4.1	Report cryo-EM validation on cycled anodes								▲
5.0		**X-ray characterization**								
5.1		Characterize full cells with baseline electrolyte								
	5.1.1	Report X-ray characterization on cycled anodes				▲				
5.2		Characterize full single-layer cells with high-rate electrolyte								
	5.2.1	Report X-ray characterization on cycled anodes						▲		
5.3		Characterize full multi-layer cells with high-rate electrolyte								
	5.3.1	Report X-ray validation on cycled anodes								▲
6.0		**Charging profile optimization**								
6.1		Develop early predications using full cells with baseline electrolyte								
	6.1.1	Report early estimations of failure methods		▲						
6.2		Identify promising charging policy candidates								
	6.2.1	Report preliminary fast charging protocol from full cell testing				▲				
	6.2.2	Report promising policies from full cell testing with high-rate electrolyte					▲			
6.3		Identify optimized charging profile								
	6.3.1	Report optimized 3-step fast charging protocol for 2Ah cells							▲	

Approach: design a new electrolyte

10-minute XFC on LFP/graphite cells already possible with an anode thickness of 35 microns → **doubling thickness requires quadrupling in diffusivity → cannot be achieved solely by increasing electrolyte conductivity**

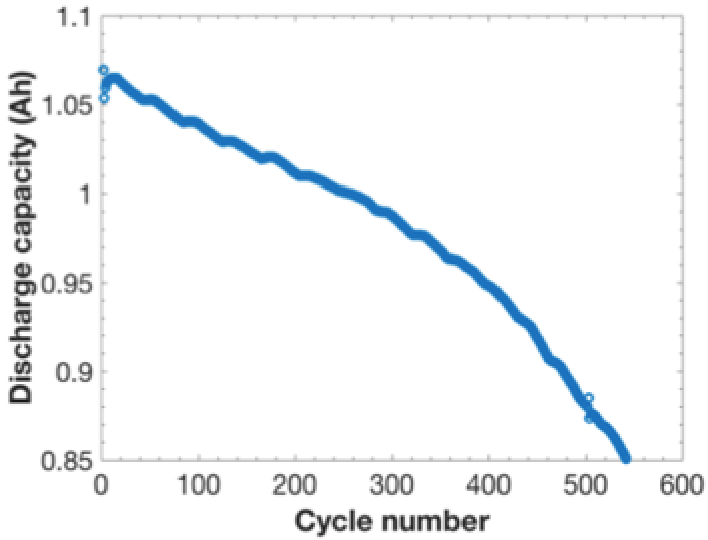


Figure 1. *Cycle lifetime for an A123 LFP/graphite (35-μm thick) 18650 cell under 6C CC-CV fast charging.*

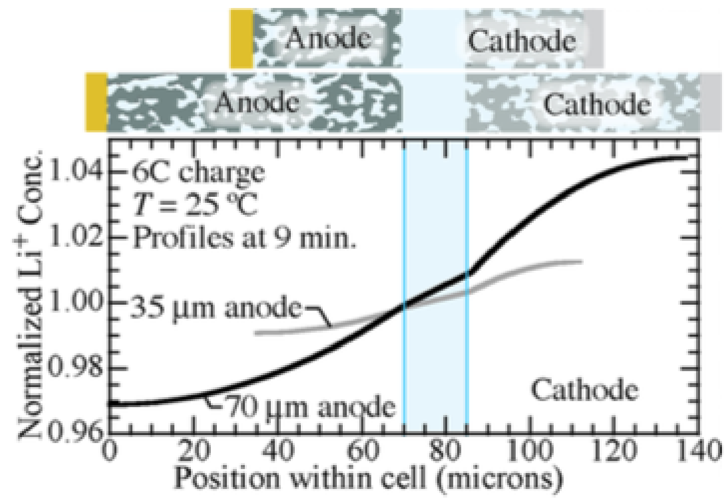


Figure 2. *Predicted effects of electrode thickness using a quasi-1D model.*

Need to (1) improve mass transport in electrolyte, (2) speed up desolvation kinetics, and (3) modify SEI to improve transport.

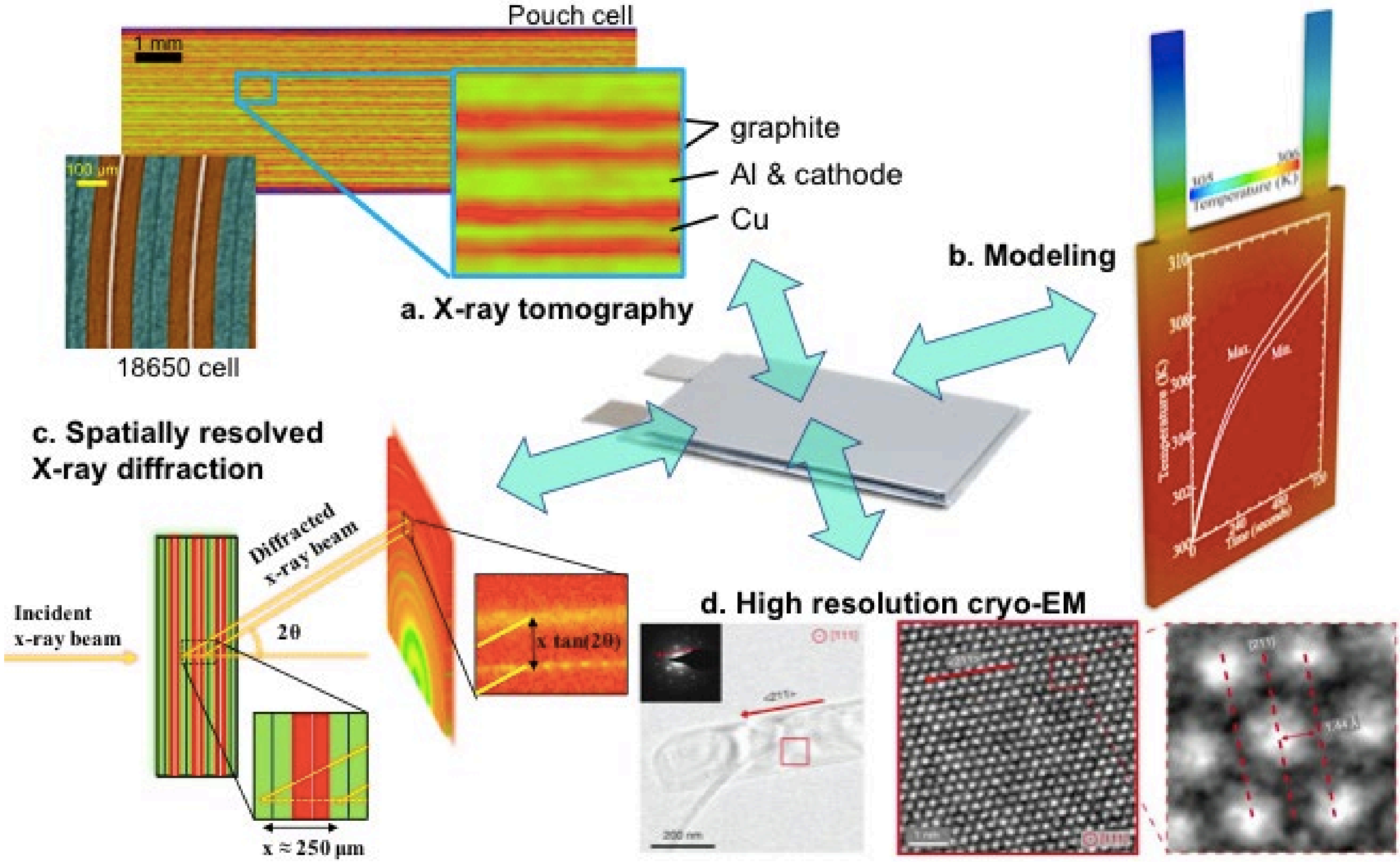
Approach: Validate via characterization & simulation

Are we really suppressing irreversible lithium plating?

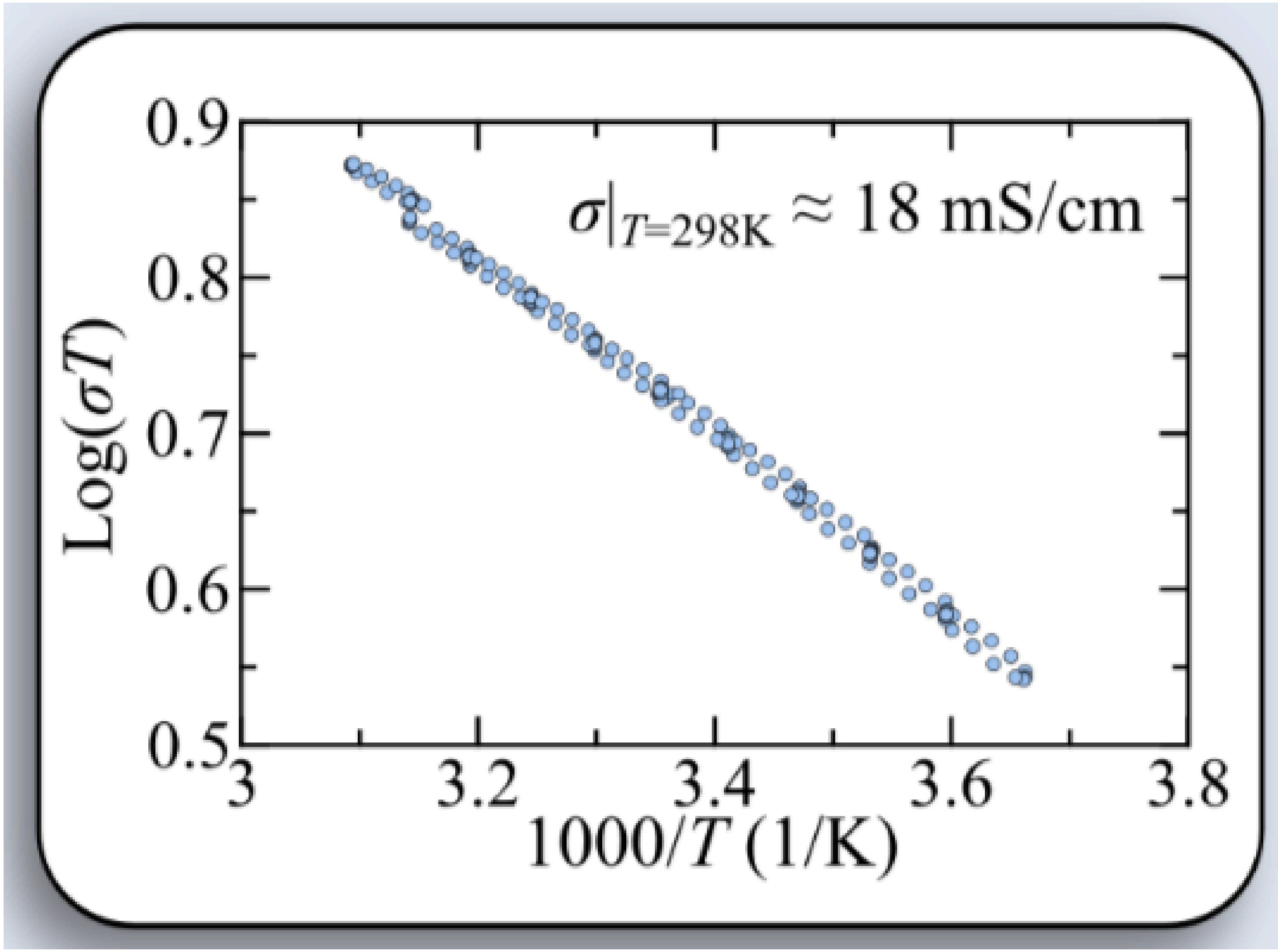
(1) X-ray microscopy & diffraction: detecting Li plating for full SLPC & MLPC cells

(2) Cryo-EM: probe location of lithium plating and quantify SEI

(3) Simulation: resolve local rate of lithium plating

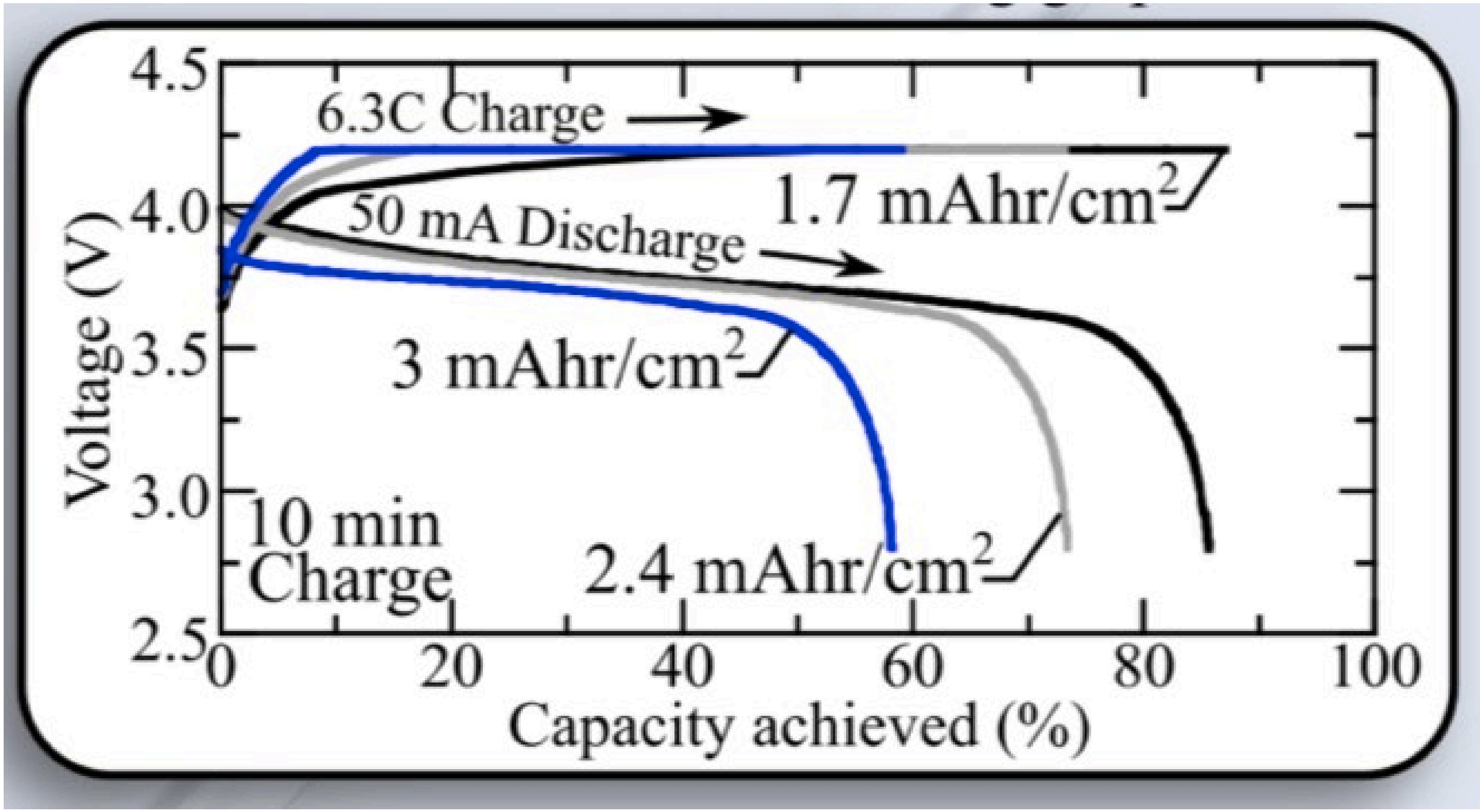


Technical Progress: high conductivity electrolyte



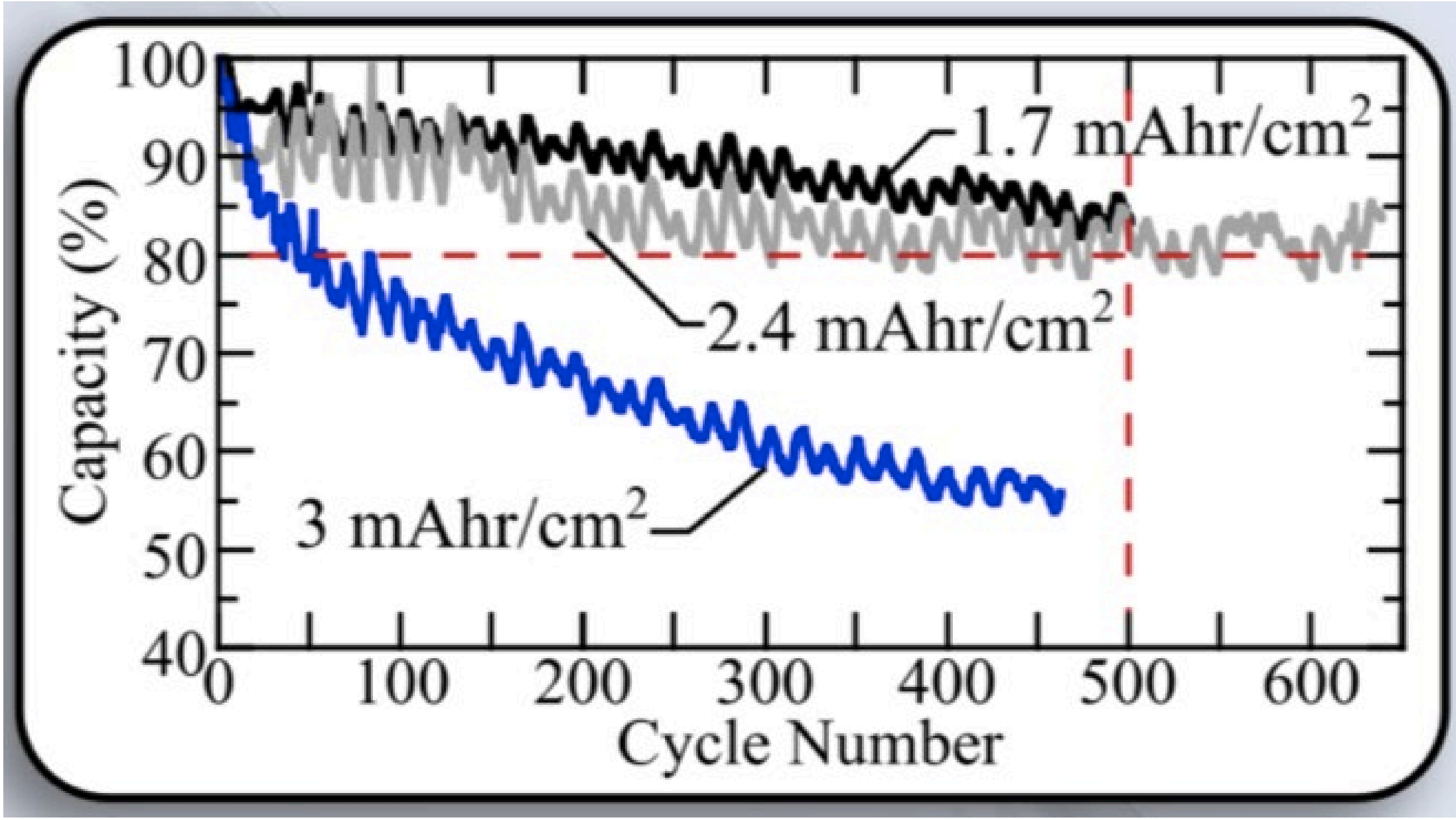
A novel low viscosity, high Li salt electrolyte has been synthesized, achieving 18 mS/cm conductivity at room temperature.

Technical Progress: single layer pouch cells



Good charge rate capability achieved for 2.4 mAh/cm2 electrode loading. Single-layer graphite/LCO pouch cells with capacities 80, 110 and 140 mAh.

Technical Progress: single layer pouch cells

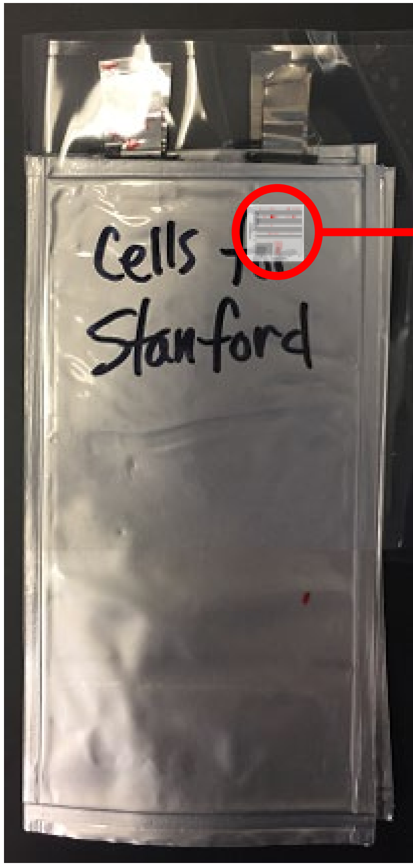


Promising cycle life achieved for 1.7 and 2.4 mAh/cm2 electrode loading. Improvement needed for 3 mAh/cm2 electrode loading.

1.7mAh/cm2 graphite loading (80 mAh): 500mA (6.2C), 4.2V max V, 10 minute cutoff, 50 mA discharge

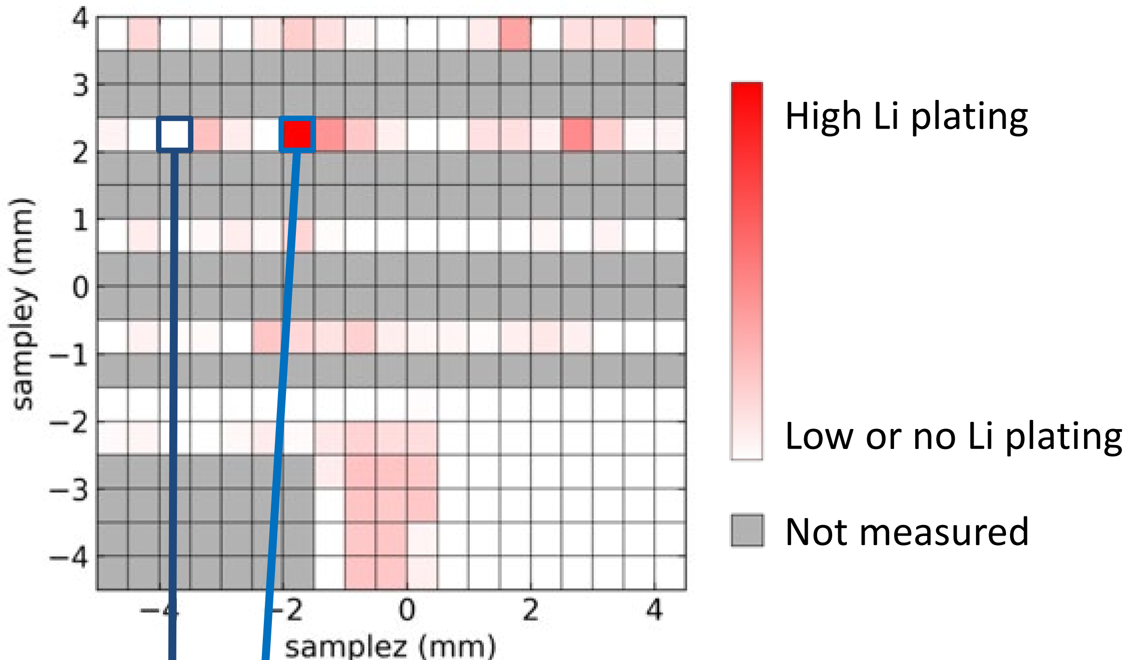
2.4mAh/cm2 graphite loading (110 mAh): 700mA (6.4C), 4.2V max V, 10 minute cutoff, 50 mA discharge3.0mAh/cm2 graphite loading (140 mAh): 900mA (6.3C), 4.2V max V, 10 minute cutoff, 50 mA discharge

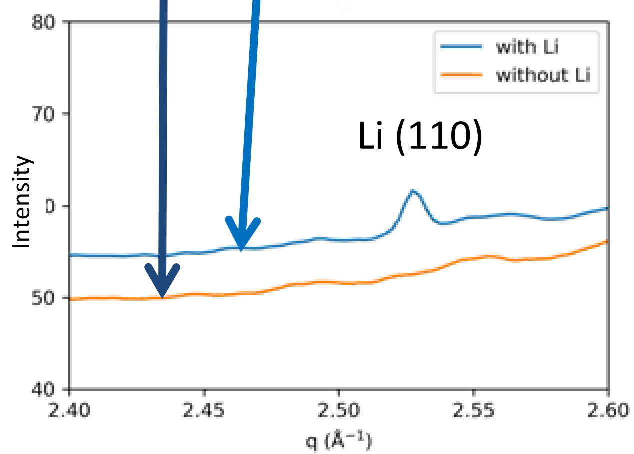
Detection of Li plating in full cells via X-ray diffraction



Initial capacity: 60 mAh

Final capacity: 20 mAh





Regions of low and high Li plating detected by 2D XRD

Response to Previous Year Reviewer’s Comments

None

Collaboration & Coordination with Other Institutions

Stanford Synchrotron Radiation Lightsource

Advanced Photon Source

Covalent Associates

Remaining Challenges & Barriers

- Achieve 80% charge acceptance within 10 minutes.
- Develop additive package that enables 500 cycles.

Proposed Future Research

- Continue to optimize salt concentration, co-solvent and additive package to achieve > 0.020 mS/cm conductivity at RT.
- Optimize charging protocol to increase charging capacity at 10 minutes to 80%.
- Perform X-ray and electron characterizations to assess the effect of electrolyte on SEI thickness and lithium plating during extreme fast charging.

Summary

- A low viscosity, high conductivity electrolyte has been developed.
- X-ray diffraction microscopy & cryogenic electron microscopy reveal Li plating and SEI formation.

Acknowledgement

This work was by the Assistant Secretary for Energy Efficiency and Renewable Energy, Office of Vehicle Technologies, Battery Materials Research Program, U.S. Department of Energy. We grateful acknowledge the guidance from Brian Cunningham.