

Development of High Energy Density Li-Sulfur Cells

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May 13

Project ID
#ES125

OVERVIEW

Timeline

44% completed

Start: Sep. 30, 2011

End: Jan. 15, 2015

Barriers Addressed

- Power and energy density
- Cycle and calendar life
- Abuse tolerance

Budget

- FY 2012: \$2,166,675
 - DOE share: \$1,524,373
 - Cos share: \$642,302
- FY 2013: \$2,405,452
 - DOE Share: \$1,642,452
 - Cost Share: \$763,000

Partners

- EC Power
- Johnson Controls
- Argonne National Lab

RELEVANCE - OBJECTIVES

Develop a **full lithium-sulfur battery system** for high energy density, efficiency, and good cycle life, and safe operation.

Project scope

Design of full lithium-sulfur cell:

- Nanocomposite Sulfur Cathode – high energy/power, stable
- Advanced Lithium- or Silicon-Based Anode – high energy/power, stable
- Electrolyte – nonflammable, stable, low self-discharge
- Optimized cell design

Performance targets

4Ah cells

600 Wh/L (cell level)

Cycle life 500+ cycles

Excellent safety characteristics

MILESTONES

- 1. Advanced material development and characterization (10/2011-09/2012)**
 - Evaluate baseline cells
 - Develop and test 1st-generation cathode, anode, electrolyte
 - Thermal and failure mechanism studies
- 2. Material scale up and 1Ah pouch cell development (10/2012-09/2013)**
 - Cathode and anode scale-up, continued improvement, and 1 Ah pouch cell design and testing
 - Continued investigation of electrolytes and failure mechanisms
- 3. Large format prismatic cell design (10/2013-09/2014)**
 - Continued scale-up, failure mechanism analysis, and 4 Ah prismatic cell design and testing
 - Cell modeling and optimization

KEY DELIVERABLES

- 1st set (NCM Baseline Cells): Cell size: ~ 1Ah pouch cells. [Completed]
- 2nd set (Li-S Cells): Cell size: ~ 1Ah pouch cells. [Sep. 2013]
- 3rd set (Li-S Cells): Cell size: ~ 4Ah pouch cells. [Sep. 2014]

APPROACH - OVERVIEW

Anode

- Investigating/optimizing Li and Si composite anodes
- Exploring polymer electrolytes

Cathode

- Investigating sulfur-carbon nanocomposites
- Pursuing methods for increased sulfur loading

Electrolyte

- Determining new electrolytes and additives
- Assessing and developing novel solvents
- Optimizing electrolytes for high capacity and low self-discharge

Cell design

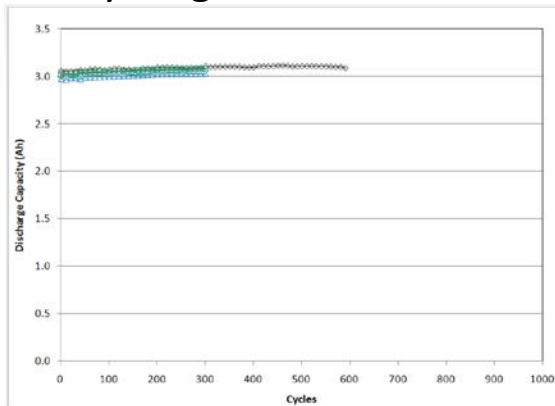
- Optimizing cells design for chemistry requirements
- Conducting modeling and experimental testing

TECHNICAL ACCOMPLISHMENT AND PROGRESS

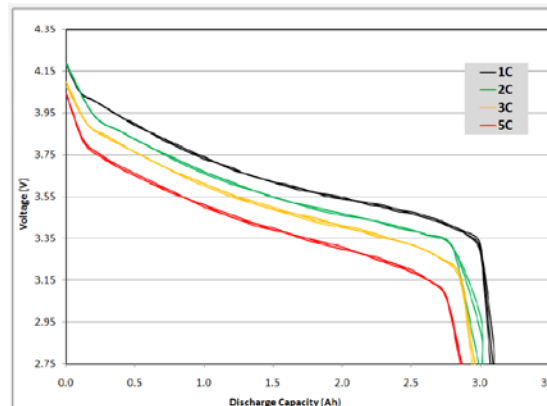
- Scaled up cathode material production to 50g level, with plans for further scale-up to 500g level
- Developed cathode composites for improve polysulfide adsorption, high sulfur loading, and improved rate performance
- Developed electrolytes capable of stable cycling with 1200 mAh/g capacity
- Developed improved Li powder-based and Si-based anodes
- Characterized thermal safety of many Li-S cell components
- Delivered baseline cells for independent testing

SUBMITTED BASELINE CELLS FOR INDEPENDENT TESTING

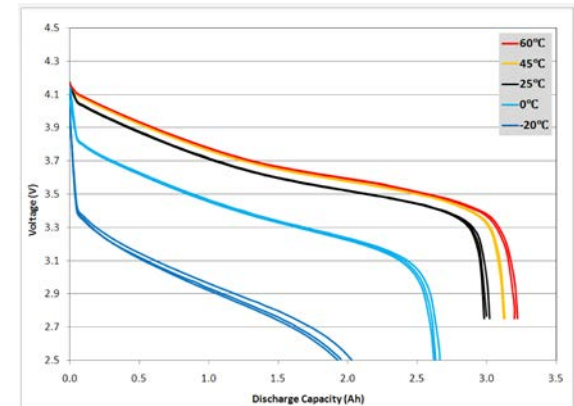
Cycling Performance



Rate Performance

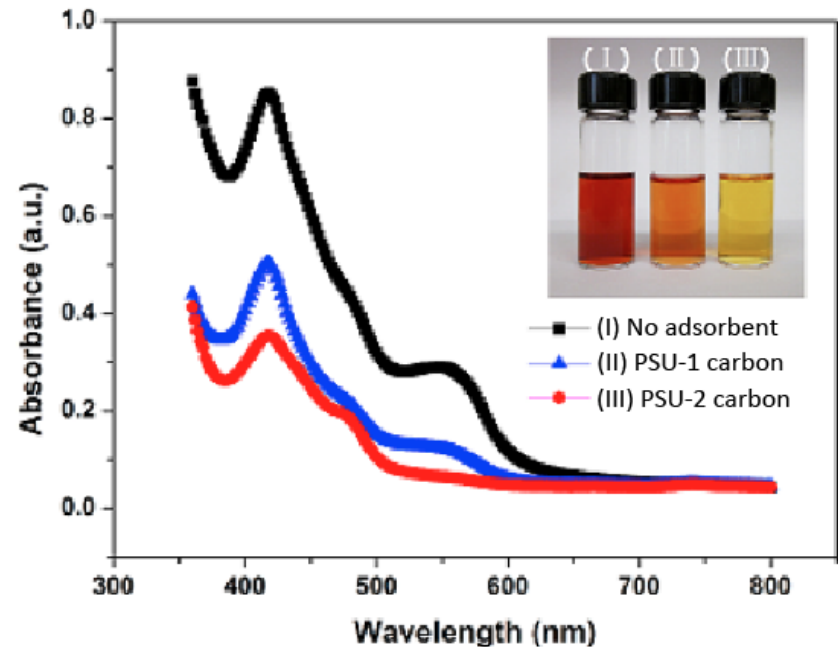
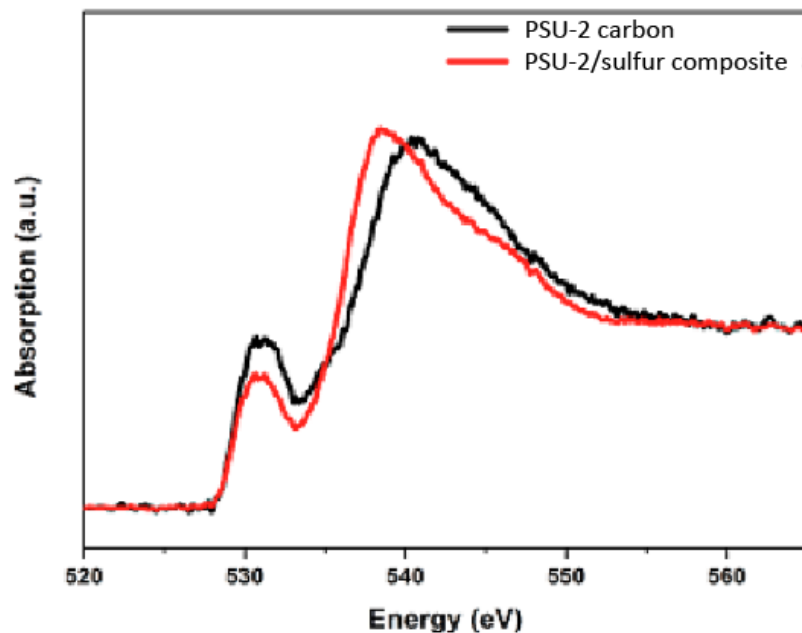


Temperature Performance



- 3Ah baseline NMC/graphite cells were tested and showed 100% capacity retention after 600 cycles at 1C rate and 92.7% retention after 300 cycles at 5C rate.
- Cells were submitted for independent testing by Idaho National Lab.

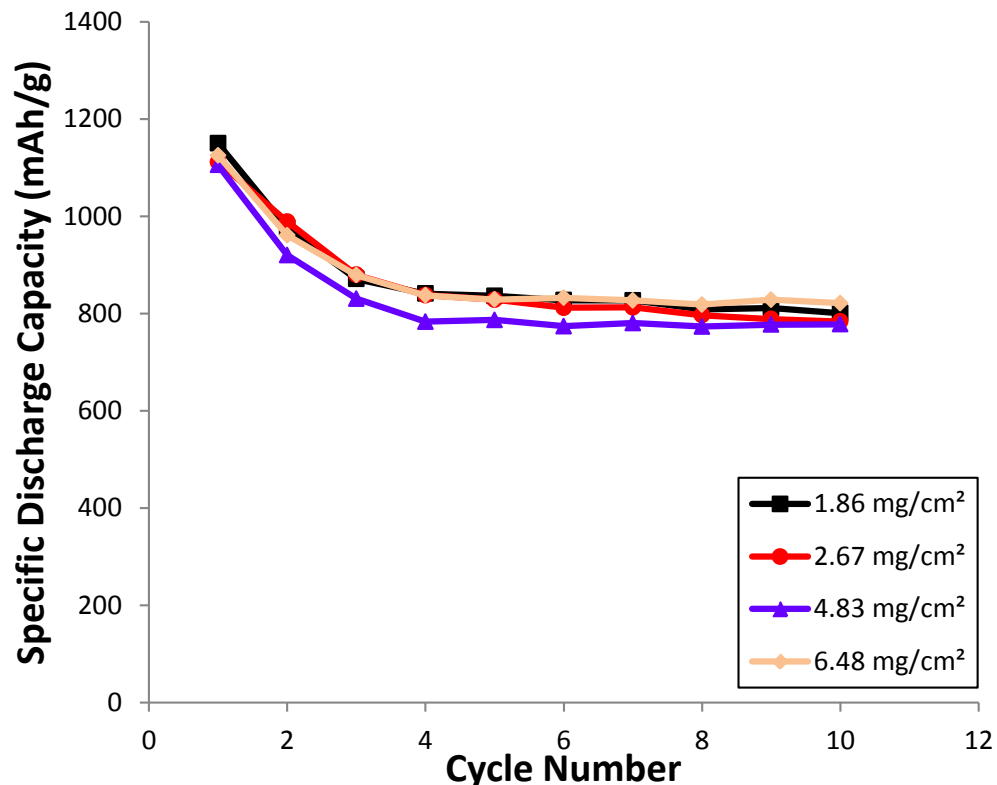
PSU-2 SULFUR CATHODES WITH IMPROVED SULFUR ADSORPTION



- UV-vis spectroscopy (right) shows that PSU-2 cathode material traps sulfur better than the baseline PSU-1 cathode material.
- XANES (left) shows interaction between oxygen and sulfur in PSU-2 carbon.

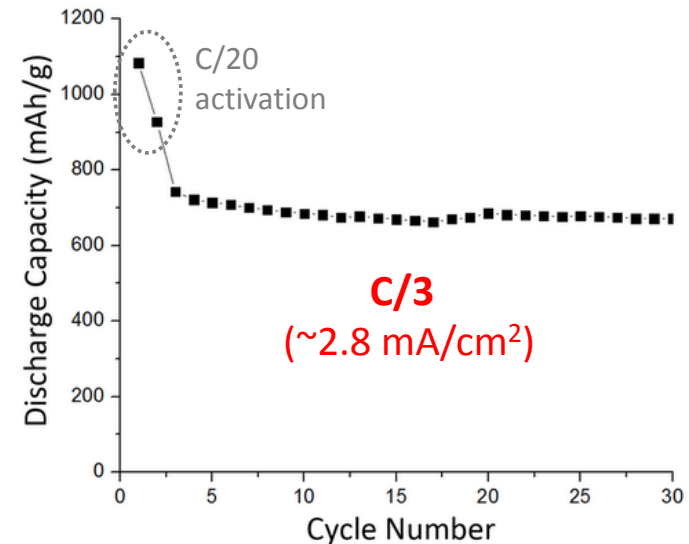
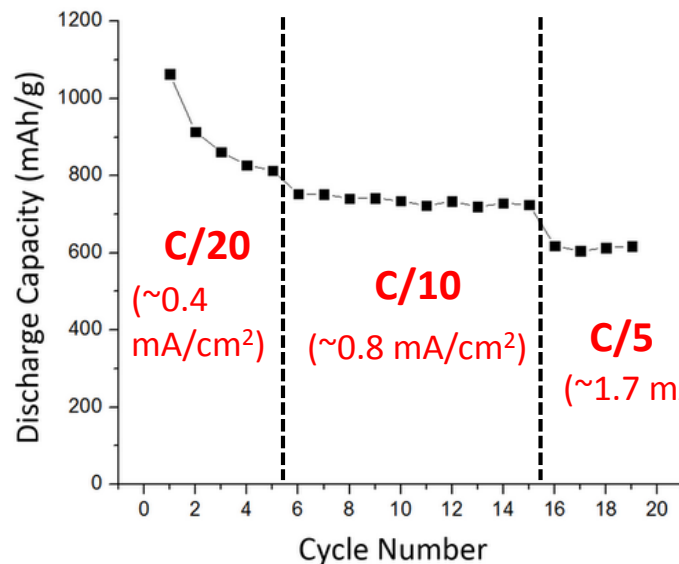
PSU-3 SULFUR CATHODES WITH HIGH SULFUR LOADING

Cycling @ C/10 rate (first 2 cycles, C/20)



- PSU-3 cathodes are able to achieve much higher loading than the baseline PSU-1 cathodes while maintaining good performance.

HIGH-LOADING PSU-4 SULFUR CATHODES WITH IMPROVED RATE PERFORMANCE

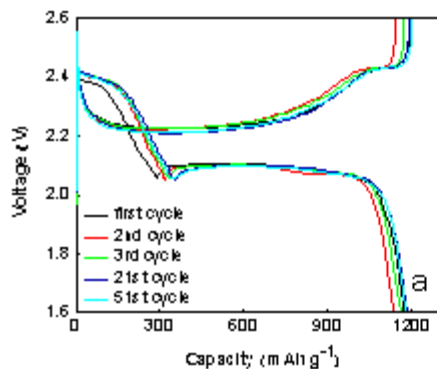


- PSU-4 cathodes have a high sulfur loading of 5 mg/cm² and greatly improved rate performance over previous cathode designs.

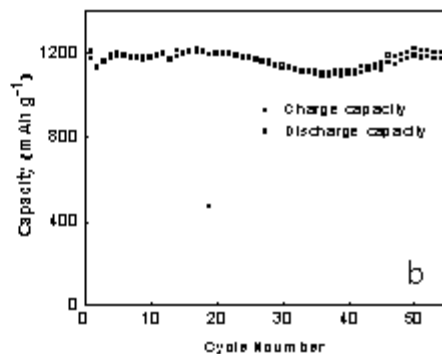
HIGH-PERFORMANCE ELECTROLYTES

ANL-E-1

Charge/Discharge Curves

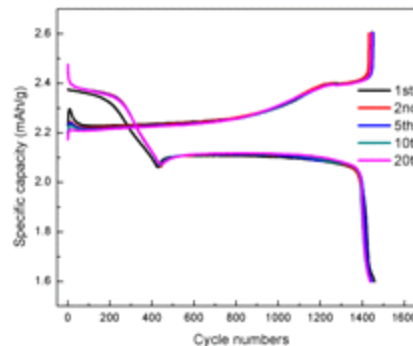


Cycling Performance

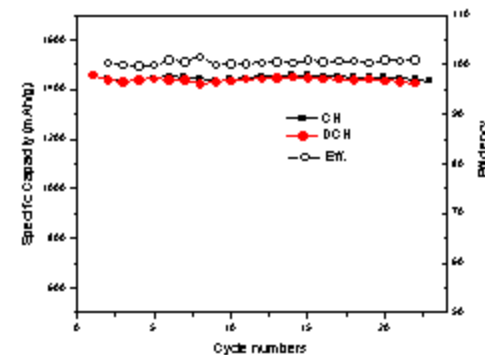


PSU-E-1

Charge/Discharge Curves

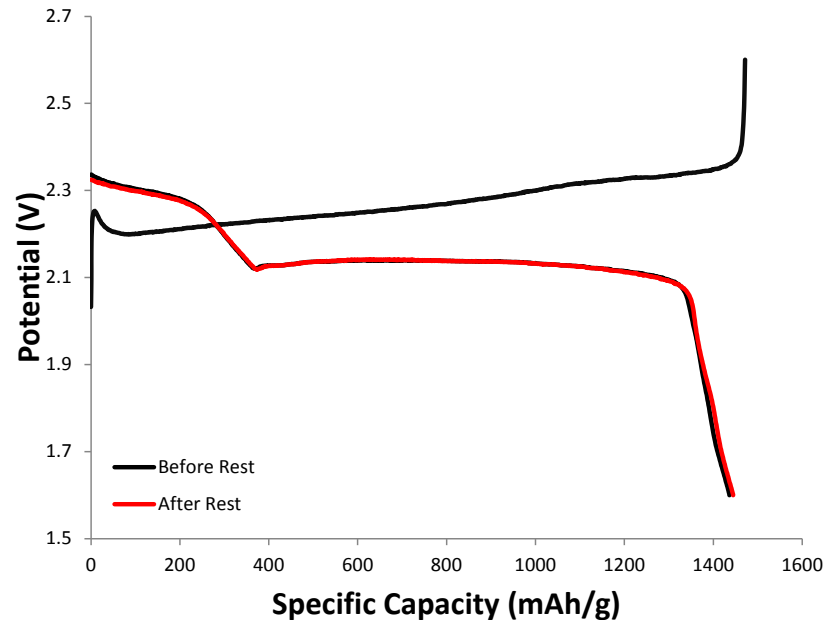


Cycling Performance



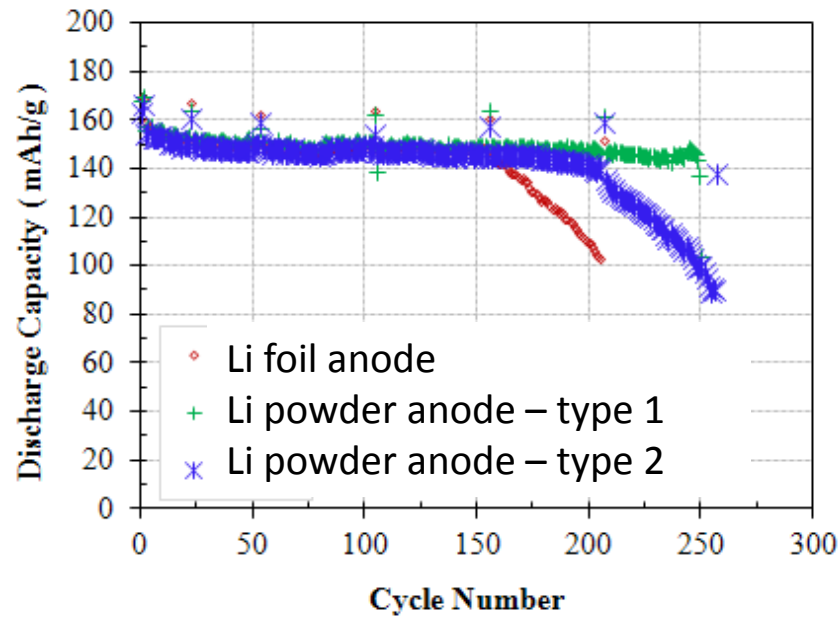
- New ANL-E-1 and PSU-E-1 electrolytes showed excellent cycling performance and coulombic efficiency with both PSU-1 and simple ball-milled sulfur cathodes.

SELF-DISCHARGE OF LI-S BATTERIES



- Self-discharge of Li-S cells with baseline electrolyte was found to be as high as 60% in 10 h at 40°C.
- With new PSU-E-2 electrolyte, self-discharge of cells with simple ball-milled sulfur cathodes with loading ~ 1 mg S/cm² was found to be less than 1% (shown above).

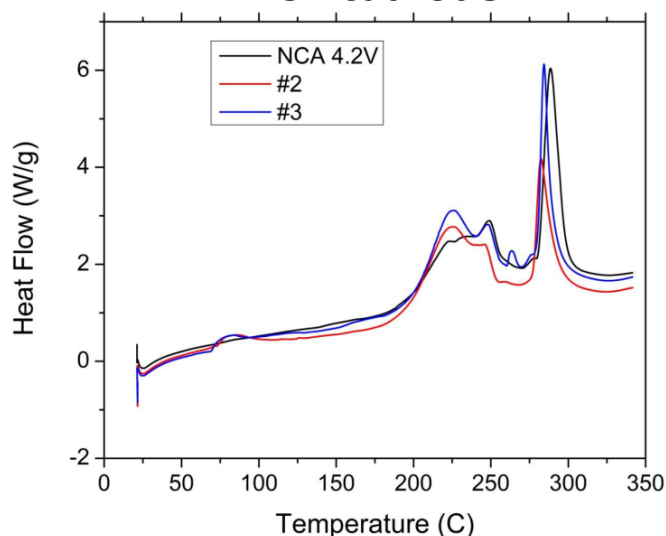
LI POWDER-BASED ANODE



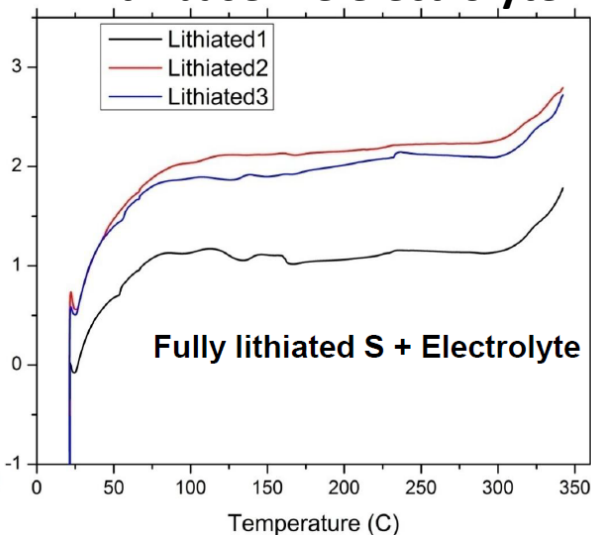
- Li powder-based anode shows cycling stability superior to that of Li foil.

THERMAL SAFETY OF LI-S BATTERIES

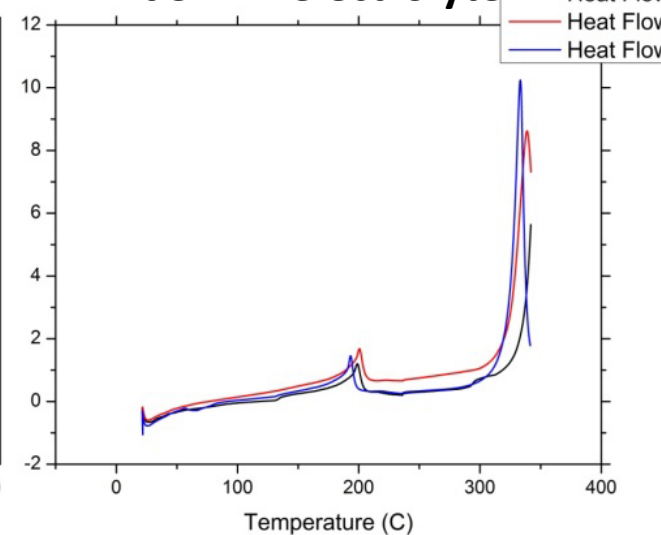
**Comparison:
NCA cathode**



**Lithiated PSU-1 cathode
with baseline electrolyte**

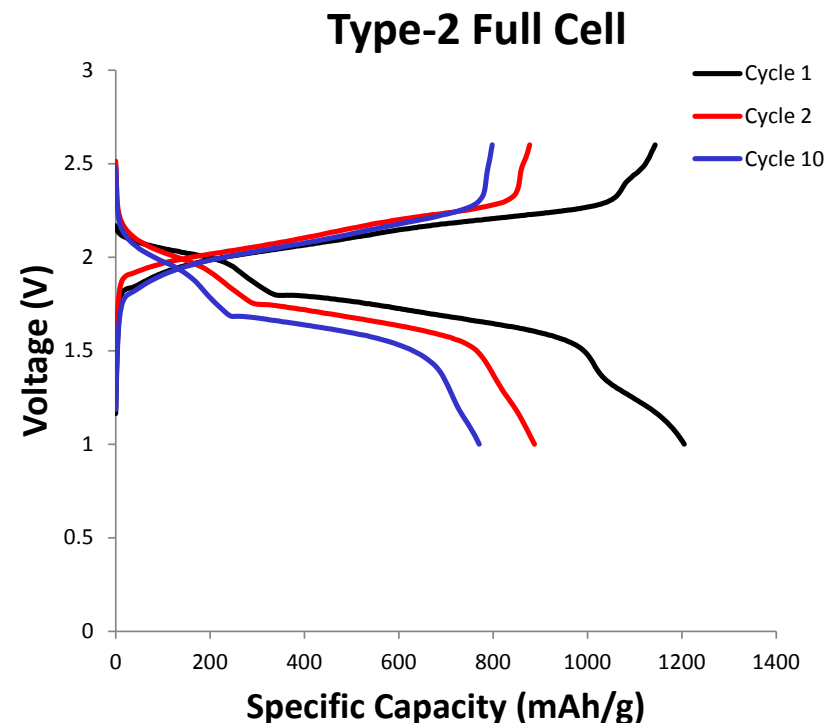
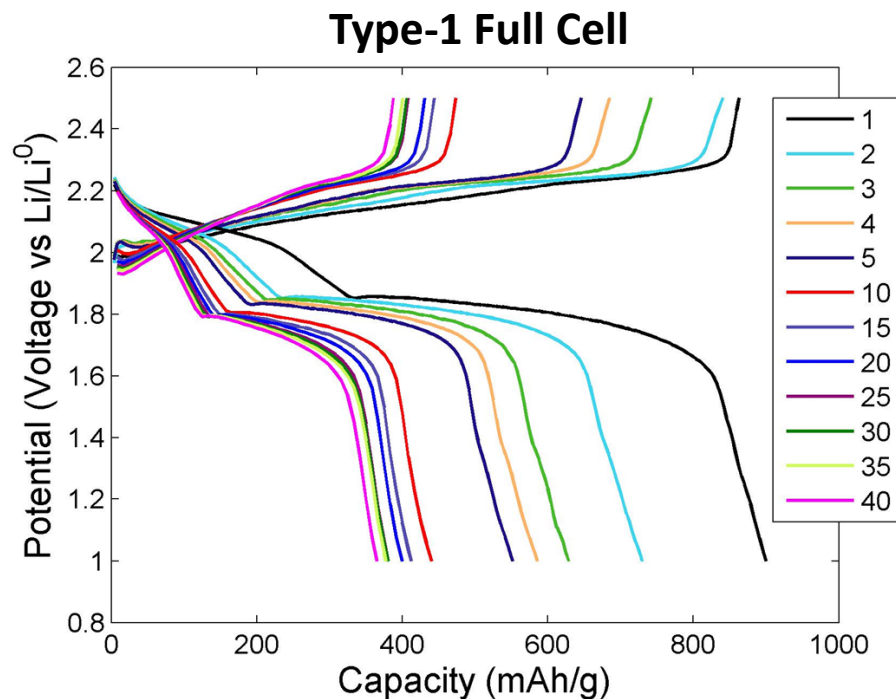


**Si-based anode with
JCI-E-1 electrolyte**



- The PSU-1 sulfur cathode has excellent thermal stability in electrolyte even after cycling - no major thermal events up to at least 350°C.
- The Si-based anode with JCI-E-1 electrolyte has very good thermal stability - no major thermal events up to 330°C.

SILICON/SULFUR FULL CELLS



- Silicon/sulfur full cells were successfully cycled.
- Cell disassembly and separate half-cell testing for type-1 cells showed that capacity loss was primarily related to sulfur loss from the cathode.

COLLABORATION

- Worked with Johnson Controls, Argonne National Lab, and EC Power to establish strong project baselines.
- Working with Argonne National Lab on concurrent electrolyte development and testing.
- Working with Johnson Controls on silicon-based anode development, self-discharge testing, and overall cell testing and design.
- Working with EC Power on lithium powder-based anode development and overall cell design and testing.
- Independent testing of baseline cells is being conducted by Idaho National Laboratory.

FUTURE WORK

- Continue development of electrolyte systems for better overall battery performance, particularly in terms of lowering self-discharge and improving full-cell performance
- Further scale up cathode material production and improve cathode rate performance
- Perform extensive safety tests (nail penetration, oven testing, etc)
- Optimize full cell design and determine key parameters
- Design and test full 1 Ah pouch cells

SUMMARY

- Scaled up cathode material production
- Developed cathode composites for high sulfur loading and improved rate performance
- Developed electrolytes capable of stable cycling with high capacity
- Characterized and addressed battery self-discharge
- Developed improved Li powder-based and Si-based anodes
- Characterized thermal safety of many Li-S cell components
- Delivered baseline cells for independent testing