

Lithium-Sulfur Energy Storage Development for Space Applications

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Introduction

Objectives:

To transition lithium-sulfur battery from prototype large format pouch cells as developed under Navitas' Phase II SBIR's with NAVAIR and NASA to space-qualified cylindrical 18650 format, with optimizing electrode formulation, separator coating and electrolyte design intended to increase energy density and cycle life. This is a multi-phase program, leading to development and qualification of 18650 format LSB cells that meet S-144 qualification and can deliver > 250 Wh/kg with cycle life > 1000.

Approaches:

Address Material Limitations:

- Ceramic host active material that entraps polysulfides and improves cycle life
- Ceramic-coated separator to eliminate polysulfide shuttling and improve sulfur utilization for long cycle life (1000+)
- Partially Fluorinated Ether (PFE) and Glyme (PFG) electrolyte to reduce polysulfide dissolution, protect lithium anode, and improve cell cycle life

Address Process Challenges:

- Wet-slurry coating for high sulfur loading cathode (> 3.5 mg S/cm²)
- Electrode/electrolyte design for high C-rate performance
- Multifunctional coating on separator at a pilot scale
- Electrolyte synthesis scaleup and low E/S ratio for high energy density
- Li anode protection for long cycle life

Navitas' Sulfur Cathode, Coated Separator, and PFE/PFG Electrolyte

- Color of Li₂S₂ solution after contact with different hosts.
- Proprietary ceramics play important role in strong absorption and fast strong polysulfide absorption.
- Fast electron transfer
- Proprietary ceramics play important role in strong absorption and fast redox reaction of polysulfide.
- Double-side coated sulfur cathode with S loading > 3.6 mg/cm².
- SEM of uncalendered sulfur electrode showed relative uniformity.

- PFE and PFG as co-solvents can reduce polysulfide dissolution in electrolyte.
- SEM of uncalendered sulfur electrode showed relative uniformity.

- Multifunctional coating is around 2-4 μm thick.
- Porosity (Gurley) reduction is less than 10%.
- Separator pilot coating was conducted with reverse gravure coating technic.
- Achieved uniformly-coated separator roll of > 500 m long.

- Typical first charge-discharge curve of coin cell or single-layer-pouch cell with optimized Navitas cathode.
- Sulfur loading: ~ 3.6 mg S/cm²; Porosity of electrode: 55 - 60%; E/S ratio: 5 μl/mg S.
- Discharge curve showed obvious two voltage plateaus. At C/20 rate, specific capacity is ~1100 mAh/g.

- PFE and PFG as co-solvents can reduce flammability of electrolyte. Electrolyte after ignition is non-flammable with high percentage of PFE and PFG.
- PFE/PFG could change viscosity, ionic conductivity of electrolyte.
- PFE/PFG could change the conversion kinetics between high-order and low-order polysulfides.
- Could help SEI layer formation on Li anode.

Coated Separator

- Uniform coating with multifunctional materials on plain separator mitigated polysulfide shuttling.
- EIS results demonstrated that the cells assembled with coated separator have smaller solution resistance (R_s) and charge-transfer resistance (R_c).
- The cells with coated separator showed higher sulfur specific capacity and better cycle life due to mitigation of polysulfide shuttling, increase in electrode conductivity with an extra conductive layer against cathode, and prevention of side reactions on lithium anode.

Electrolyte Development

- The voltage profile collects the last cycle in each C-rate (0.05C, 0.1C, 0.2C, 0.3C, 0.5C, 0.67C, 1C, and then 0.1C).
- Beyond C/2 rate, the low-order polysulfide nucleation process of baseline electrolyte hits the cutoff voltage limit of 1.6 V due to the slow kinetic reaction.
- Modified electrolyte promotes low-order LIPS conversion reaction and lowers conversion overpotential.
- Modified electrolyte improved high-C rate performance; S capacity is ~600 mAh/g at 1C, satisfies S-144 application.

Li-Sulfur 18650 Cylindrical Cells

- Navitas' first cylindrical 18650 Li-S cell
- Weight: 29.5 g
- OCV: 3.2 V
- Capacity: 2200 mAh
- EIS of 18650 cylindrical cell
- Low R_s value: < 0.1 Ohm
- Low Charge Transfer resistance: R_c < 0.075 Ohm.
- The Li-S 18650 cylindrical cells were built with base line electrolyte, prime uncoated separator.
- At C/20 rate, the specific capacity of sulfur is relatively stable.
- The optimized electrolyte and coated separator future will be applied in the future cell builds to improve cycle life.
- Temperature effect on cell performance
- Room T., distinguished second voltage plateau, ~900 mAh/g.
- 0 °C, lower voltage for second plateau, 730 mAh/g.
- 20 °C, no second voltage plateau, 150 mAh/g.

Collaboration/Partnership

- John Zhang (Argonne National Laboratory)**
 - Development of innovative electrolyte with different PFE/PFG solvents, lithium salts and concentrations.
 - Evaluation of physical and chemical properties of PFE/PFG electrolyte, ionic conductivity, flammability, polysulfide dissolution reduction, SEI formation.
 - Investigation of effect of PFE/PFG electrolyte on high-C-rate and cycle life performance.
 - Scale-up and incorporation of down selected PFE/PFG electrolyte into 18650 cells with low E/S ratio to achieve high energy density.
- Kevin Sinclair (Lockheed Martin)**
 - Consulting service
 - Providing cell testing and feedback on prototype samples
 - Evaluating 18650 cells to meet S-144 qualification (C/2.25 charge, C/1.5 discharge).
- Tyson Craig (Battery Innovation Center)**
 - Building Li-S 18650 cylindrical cells.
 - Optimizing jelly-roll design and 18650 cell assembly.

Navitas Products

Custom Cell (40 Ah) Heavy Duty Off-Road 6T Battery Forklift Batteries Navitas' Round Cells for Navy & LMS

Navitas' Capabilities for Cell Builds and Future Collaboration with Potential Partners

Jellyroll Winding Tab Welding Electrolyte Filling Cell Crimping Automatic Electrode Stacking Machine 1 million cells/Year Automated Round Cell Assembly Line

Conclusions

- High sulfur loading cathode with great physical properties and electrochemical performance were successfully achieved. Ceramic as sulfur host has a strong polysulfide absorption.
- Multifunctional coatings on separator reduced polysulfide shuttling, which improved the utilization of sulfur and improved Li-S cell cycle life.
- Optimized electrolyte improved Li-S high- rate performance due to the reduction of overpotential, therefore improved the slow kinetic lower order LIPS conversion.
- 18650 cylindrical Li-S cells were successfully built. The solution resistance (R_s) and charge transfer resistance (R_c) are less than 0.1 Ω. The specific capacity of sulfur reaches ~ 900 mAh/g. The cells can perform at 0 °C with sulfur specific capacity of ~ 700 mAh/g.

Future Work

- Formulation modification of sulfur cathode (carbon host, additive, binder).
- Electrolyte selection (solvent, concentration, additives, lithium salts), and Li anode protection to improve cell cycle life.
- Improve high-C rate performance and extend cycle life of Li-S 18650 cells to meet S-144 qualification (C/2.25 charge, C/1.5 discharge) by combining cathode modification, separator coating, innovative electrolyte, and Li anode protection.

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