Highly Loaded Sulfur Cathode, Coated Separator, and Gel Electrolyte for High-Rate Lithium-Sulfur Battery

PI: Yong L. Joo, School of Chemical and Biomolecular Engineering, Cornell University

Project ID: bat512

Cornell University

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Overview

Project Timeline
Start: October, 2017
End: September, 2021
Percent Complete: ~90%

Budget
Total: $1.11M/3.5 years
Phase I: $360k/1.5 years
Phase II: $750k/2 years

Partners
- Jin Suntivich, Materials Science and Engineering, Cornell University
- Trung Nguyen, EIC Labs

Barriers
- Energy density for current battery electric vehicles (BEVs) is low (< 250 Wh/kg).
- Current Li-S batteries exhibit low sulfur utilization, poor cycling life and rate capability.
  - Higher sulfur loading w/o sacrificing cycle life and rate capability is needed.
  - Mitigation of polysulfides shuttling to increase capacity and improve cycle life of Li-S batteries
- Safety of BEVs are yet to be improved.

Relevance

Impact and Project's Relevance:

Lithium sulfur cells promise to increase the energy density and decrease the cost of batteries compared to the state of art Li-ion batteries (LIB). If the performance and cycling challenges can be alleviated, these systems hold the promise for meeting DOE EV targets.

The key features of the project's approach are the development of i) layered cathode with high sulfur loading ii) a polymer/ceramic separator coated with graphene, and iii) gel electrolyte for Lithium-Sulfur (Li-S) battery systems.

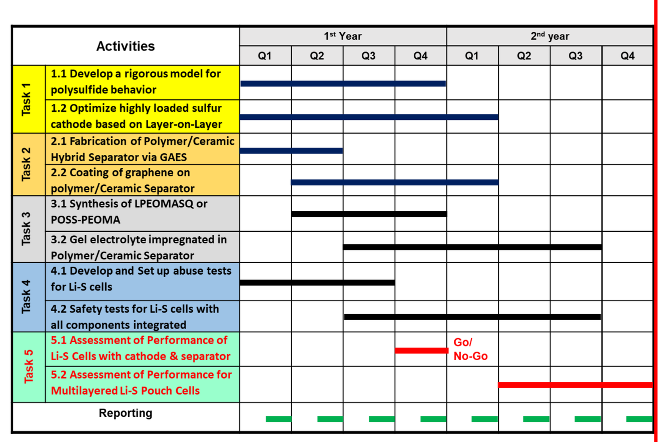
The developed highly loaded sulfur cathodes, conductive carbon coated separators, and gel electrolyte will mitigate the low rate capability, shuttling effect and limited cycle life in high performance Li-S batteries with improved safety. Rigorous abuse tests of Li-S batteries provide insight into the safety of Li-S batteries.

Objectives:

to research, develop, and demonstrate a 1 Ah Li-S battery prototype capable of achieving
- **energy density ≥500 Wh/Kg**
- **cycle life of ≥ 300 cycles**
- **at 0.5C discharge rate**

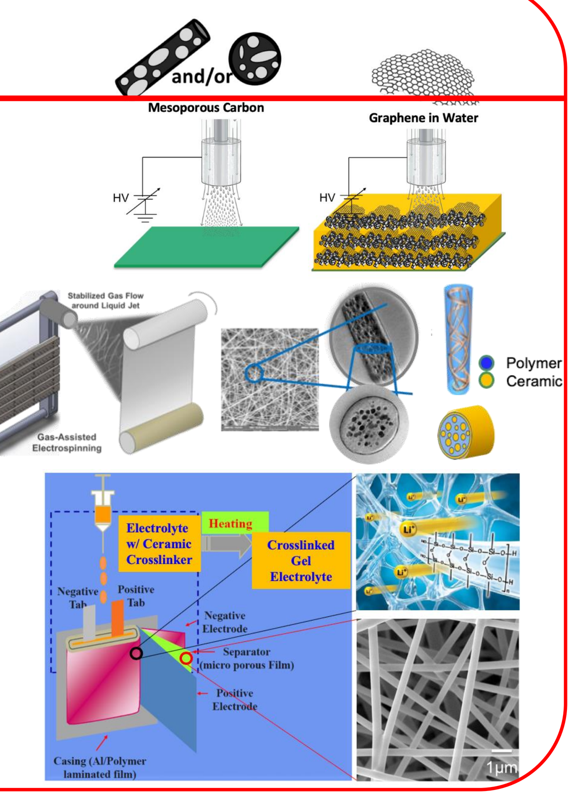
Milestones

- **Develop a rigorous model for polysulfide behavior (Q1 Y1 – Q4 Y1) (completed)**
- **Optimize highly loaded sulfur cathode based on Layer-on-Layer (Q1 Y1 – Q2 Y2) (in progress)**
- **Fabrication of Polymer/Ceramic Hybrid Separator via GAES (Q1 Y1 – Q2 Y1) (completed)**
- **Develop Polymer/Ceramic Hybrid Separator with Graphene Coating (Q2 Y1 – Q1 Y2) (completed)**
- **Synthesis of LPEOMASQ or POSS-PEOMA (Q2 Y1 – Q4 Y1) (completed)**
- **Gel electrolyte impregnated in Polymer/Ceramic Separator (Q3 Y1 – Q3 Y2) (in progress)**
- **Set up abuse tests for Li-S cells (Q1 Y1 – Q3 Y1) (completed)**
- **Safety tests for Li-S cells with all components integrated (Q3 Y1 – Q3 Y2) (in progress)**
- **Assessment of Performance of Li-S Cells with cathode/separator (Q4 Y1) (in progress)**
- **Assessment of Performance for Multilayered Li-S Pouch Cells (Q2 Y2 – Q4 Y2) (in progress)**



Approach

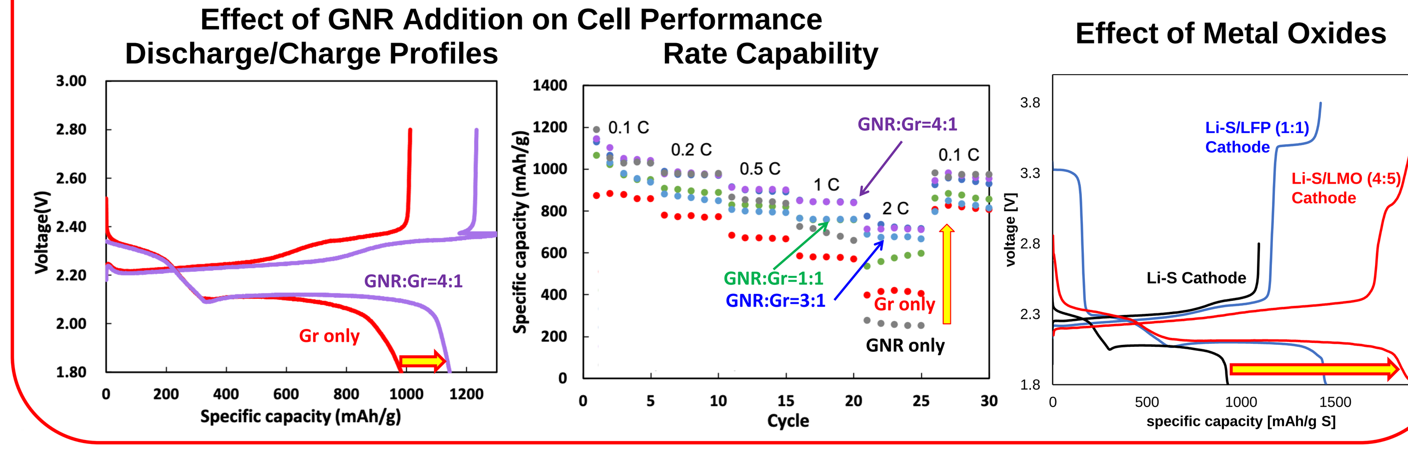
1. Highly Loaded Sulfur Cathode for High Rate Li-S Batteries
   - Utilization of Layer-on-Layer Approach based on Air-Controlled Electrospray
   - Optimization of Layering Structures via Experiments and Simulation
2. Graphene Coated Polymer/Ceramic Separator for Mitigation of Polysulfide Shuttling
   - Development of High Rate, Thermally Stable, Non-flammable Polymer/Ceramic Separator via Gas-Assisted Electrospinning
   - Graphene Coating on Polymer/Ceramic Hybrid Separators
3. Gel Electrolyte for Enhanced Safety
   - Development of Gel Electrolyte based on Ceramic Crosslinker
   - Incorporation of Gel Electrolyte into Polymer/Ceramic Hybrid Separator
4. Fabricate and evaluate 1 to 3 Ah Li-S battery Pouch Cell Prototypes
5. Assessment of interfaces and stability of Li-S cells



Accomplishments: Cathodes w/ enhance S Utilization

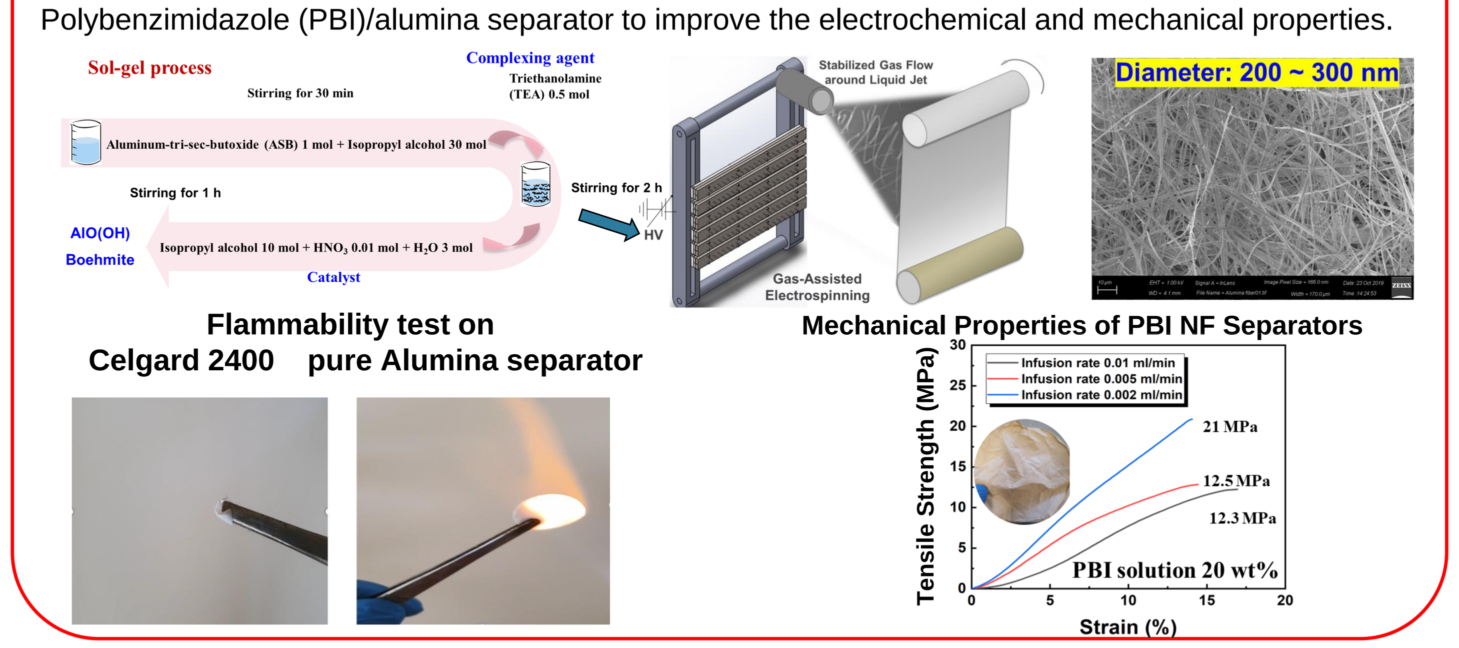
Replacing a significant fraction of graphene (Gr) with Graphene Nanoribbon (GNR) that is obtained from unzipping CNT in the sulfur incorporated mesoporous carbon/graphene cathode further increases the capacity and rate capability due to high conductivity and additional ion transport along the unzipped edge planes of GNR.

Also, incorporation of metal oxide additives such as LFP or LMO that are commonly used as LIB cathodes into the Li-S cathode enhances sulfur utilization



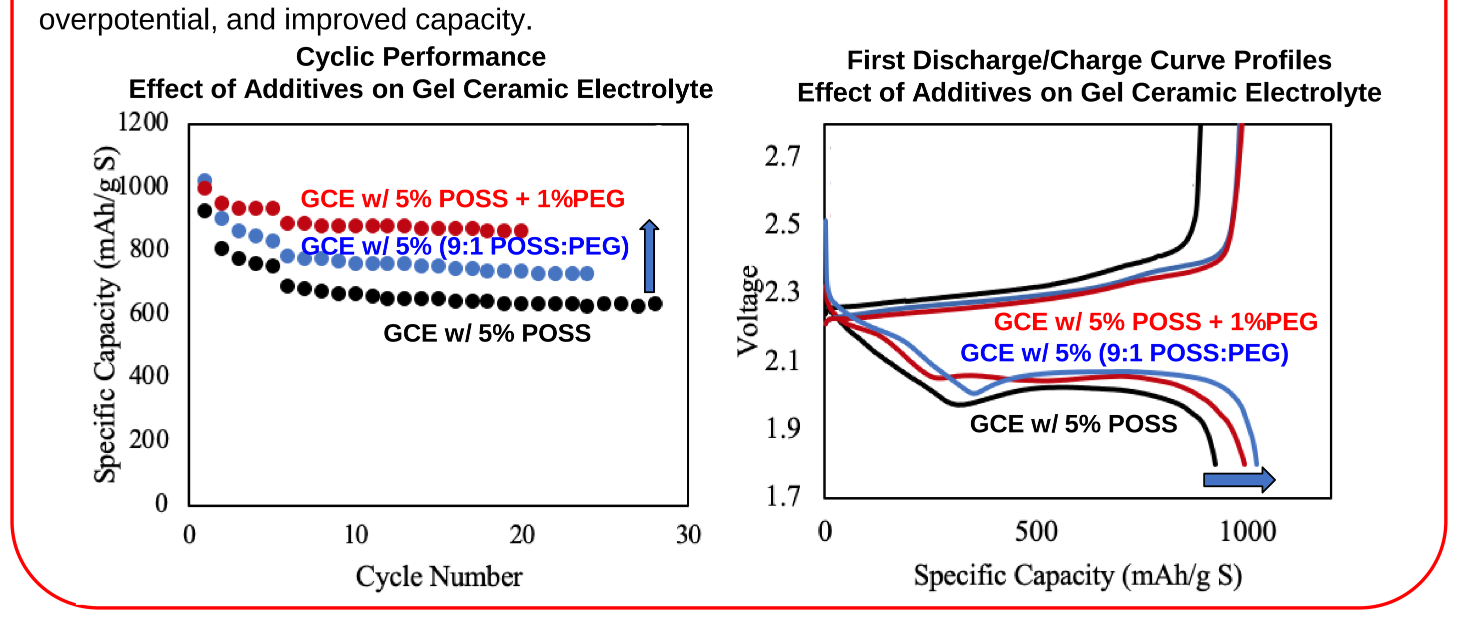
Accomplishments: Ceramic/Polymer Separator

Hybrid separators based on Polyimide(PI)/Polysilsesquioxane (PSSQ) have been upgraded with Polybenzimidazole (PBI)/alumina separator to improve the electrochemical and mechanical properties.



Accomplishments: Safe Gel Ceramic Electrolyte

Our **gel ceramic electrolyte (GE)** demonstrates that gel network created by the ceramic cross-linker can **effectively trap soluble polysulfides.** Inclusion of ionically conductive PEG results in reduced overpotential, and improved capacity.



Responses to Previous Year Reviewers' Comments

The below is the response to comments on poster last year by reviewers
- **Sulfur content of the overall cathode: 60%**
- **Electrolyte to sulfur (E/S) ratio: 10 (µl/mg).**
- **Information on Electrolyte: Liquid electrolyte of 1M LiTFSI, 0.15M of $LiNO_3$ in a 1:1 volume ratio of 1,2-dimethoxyethane (DME) and 1,3-Dioxolane (DOL). This year, we started combining our work on polymer/ceramic hybrid separator and gel electrolyte to investigate the synergy between these two.**
- **The protocol for testing Sulfur system: We started using the test protocol published by Battery 500.**
- **Cycling Performance and Rate Capability of Gel Ceramic Electrolyte System: They have been provided.**
- **Higher Sulfur Loading: We started increasing sulfur loading to 4 to 6 mg/cm².**
- **Cell Polarization for gel electrode: It appears that some fraction of liquid electrolyte with crosslinker soaks into the sulfur cathode and is gelled upon heating, creating additional resistance. We will look into minimize electrolyte amount to reduce this negative effect.**

Collaboration/Partnership

PI: Yong Joo (Chemical & Biomolecular Engineering)

Fabrication of Highly Loaded Sulfur Cathodes

Development of Polymer-ceramic Hybrid Separator and Graphene coated Separators

Incorporation of Gel Electrolyte

Co-PI: Jin Suntivichi (Materials Science & Engineering)

Quantification of Surface-Polysulfide Interactions

Measuring Transport through Separators

Collaborator: Trung Nguyen (EIC Labs)

Fabrication and Evaluation of Li-S Pouch Cells

Rigorous Safety Tests.

Proposed Future Research

For the 6 month extension, we will optimize and create synergy among Highly Loaded Cathode, Coated Separator and Gel Electrolyte

Perform Integrated Assessment of Performance of Li-S Cells with Cathode/Separator

Incorporate Gel Electrolyte into Polymer/Ceramic Separator w/ and w/o Graphene Coating

Perform Rigorous Safety Tests.

Develop rigorous abuse test system for Li-S batteries.

Perform Safety tests of Li-S cells with all components integrated.

Fabricate and evaluate 1 - 3 Ah Li-S Battery Pouch Cell Prototype.

Milestones for 6 Month Extension

Milestone	Type	Description
Optimize highly loaded sulfur cathode based on Layer-on-Layer	Technical	Optimize Layer Structure to Achieve Areal capacity > 5 mAh/cm² for 300 cycles
Develop Polymer/Ceramic Hybrid Separator w/ Gr Coating	Technical	Achieve Coating thickness < 10 µm and Gr loading < 0.5 mg/cm²
Gel electrolyte impregnated in Polymer/Ceramic Separator	Technical	Achieve 80% of capacity with liquid electrolyte at 2C
Assessment of Performance for Multilayered Li-S Pouch Cells	Technical	Validate that multi-layered Li-S pouch cells (two layered 2" x 6" or four layered 2" x 3") exhibit ≥ 1 Ah, ≥ 500Wh/kg and cycle life of ≥ 500 cycles at approximately 0.5C

Summary of Phase II – 2nd Year

During the 2nd year, we focused on developing highly loaded sulfur cathode, polymer/ceramic separator and gel electrolyte for high rate capable Li-S batteries. Quantifying surface-polysulfide interactions as well as polysulfide transport is also currently underway.

1. Highly Loaded Cathodes: To further enhance the sulfur utilization, we incorporated i) graphene nanoribbon (GNR) in sulfur incorporated mesoporous carbon/graphene cathodes, which resulted in increase in capacity and rate capability, yielding 700 mAh/g at 2C. Incorporation of metal oxides such as LFP and LMO also enhances sulfur utilization. We will carry out ex-situ post-cycling characterization to understand the morphology of the oxide-added Li-S cells to build a design rule for future Li-S cell design.

2. Polymer/Ceramic Hybrid Separators: Hybrid separators based on Polyimide(PI)/Polysilsesquioxane (PSSQ) have been upgraded with Polybenzimidazole (PBI)/alumina separator to improve the electrochemical and mechanical properties.

3. Gel Electrolyte: Our gel ceramic electrolyte (GE) demonstrates that gel network created by the ceramic cross-linker can effectively trap soluble polysulfides. Inclusion of ionically conductive PEG results in reduced overpotential, and improved capacity.

4. Creating the Synergy among Components: During the 6 month extension period, coin cells with layered cathode and graphene coated polymer/ceramic hybrid separator with and without gel electrolyte will be fabricated to create the synergy among the components and the cell performance for the cells with all the developed components integrated in coin cell as well as pouch cells will be evaluated.