

## High Energy Lithium-Sulfur Batteries for EV Applications

### Introduction

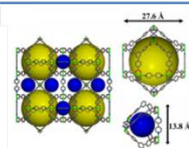
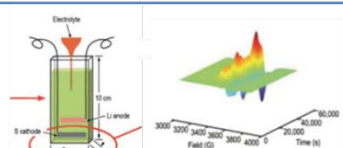

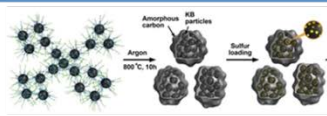
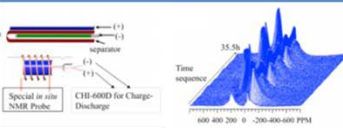
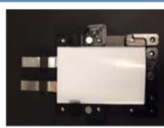
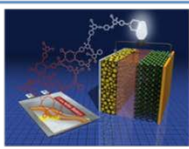
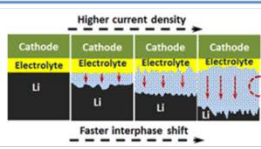

Advanced rechargeable batteries with high energy and power density and reduced cost have attracted significant attention due to the urgent demand of energy storage technologies for green transportation and large-scale energy storage applications. The Lithium-Sulfur (Li-S) battery has the potential to meet the requirements for those systems because of its high theoretical capacity, low cost, and environmentally benign attributes. However, the market penetration of Li-S batteries has been plagued by the poor cycling stability, low round-trip efficiency, and severe self-discharge rooted in the dissolution of long-chain polysulfide species and their reactions with Li metal anode. To overcome these hurdles, focused material research and mechanism understanding are urgently needed to understand the performance of thick sulfur cathode, develop effective electrolyte/additive and stabilized anode.

### Project Objective

The objective of this project is to develop high-energy, low-cost lithium sulfur (Li-S) batteries with long lifespan. All proposed work will employ high-areal-capacity sulfur cathode at a relevant scale for practical applications. High sulfur utilization rate will be achieved for high loading sulfur cathode with reduced electrode thickness and electrolyte amount. The diffusion process of soluble polysulfide out of the thick cathode and fundamental reaction mechanism of polysulfide under the electrical field will be explored by applying advanced characterization techniques to accelerate the development of Li-S battery technology. Alternative anode will be explored to address the lithium anode issue.

### Main Achievements

- Evaluated effects of carbon morphology and electrolyte/sulfur ratio on the performance of Li-S batteries.
- Identified the key challenges in preparation and application of thick sulfur electrodes in Li-S batteries.
- Discovered sulfur radicals and their interactions with electrolytes during cycling.
- Developed new Li-ion sulfur batteries to decouple the lithium metal anode issue from the system to enable focused study on sulfur cathode.

Materials development	Fundamental understanding	Practical application
 <p>▪ Ni metal organic framework (Ni-MOF) was developed to immobilize polysulfides.</p>	 <p>▪ Direct observation of sulfur radicals by in situ electron paramagnetic resonance (EPR).</p>	 <p>▪ Large area coating of high loading sulfur cathode.</p>
 <p>▪ Integrated Ketjen black (IKB) was developed for high loading sulfur cathode.</p>	 <p>▪ Electrochemical reaction pathways of sulfur were studied by in situ NMR technique.</p>	 <p>▪ Assembly of sulfur cell at Advanced Battery Facility (PNNL)</p>
 <p>▪ New electrolyte and non-Li anode were developed for Li-ion sulfur batteries.</p>	 <p>▪ Failure mechanism of Li metal anode in liquid electrolyte was studied.</p>	 <p>▪ New prototype of Li-ion sulfur battery.</p>

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### Publications & Patents

- Lu D, Zheng J, Li Q, Xie X, Ferrara S, Nie Z, Layla M, Nigel B, Zhang J, Graff G, Liu J and Xiao J. High Energy Density Lithium–Sulfur Batteries: Challenges of Thick Sulfur Cathodes. *Adv. Energy. Mater.* 2015, DOI: 10.1002/aenm.201402290.
- Lu D, Yan P, Shao Y, Li Q, Ferrara S, Pan H, Graff G, Polzin B, Wang C, Zhang J, Liu J and Xiao J. High performance Li-ion sulfur batteries enabled by intercalation chemistry. *Chem. Commun. (Cambridge, U K)* 2015, 51(70): 13454-13457.
- Zheng J, Lu D, Gu M, Wang C, Zhang J-G, Liu J, and Xiao J. How to Obtain Reproducible Results for Lithium Sulfur Batteries? *J Electrochem. Soc.* 2013, 160(11): A2288-A2292.
- “Electrolyte for lithium-sulfur batteries employing graphite as the anode”, Patent application #: 14/530,562
- “Electrodes for Li-S batteries”, Patent application #: 61/521,191