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## **Mechanism Panel Abstract & Speaker Biography**

### **Operando Spectromicroscopy of Lithium-Sulfur Batteries**

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Li-S batteries have a unique chemistry that achieves high capacity via chemical transformation rather than Li intercalation. Elemental sulfur  $S_8$  is reduced through a series of soluble Li polysulfides ( $Li_2S_x$ ,  $2 \leq x \leq 8$ ) to a final solid discharge product,  $Li_2S$ . However, Li-S suffers from unrealized theoretical capacity and rapid capacity fade due to loss processes that are not well-understood. Batteries are spatially and chemically heterogeneous. Deciphering the speciation and distribution of dissolved polysulfides in the anode, cathode, and especially the electrolyte during cycling is critical for realizing the potential of Li-S. We present *operando* characterization using sulfur K-edge X-ray absorption spectroscopy (XAS) performed at SSRL. Synchrotron radiation provides high energy resolution for distinguishing the complicated polysulfide spectral features. From this information, we can observe a spatially heterogeneous and chemically diverse system to gain insight into how sulfur speciation and spatial distribution contribute to capacity fade.

Typically, *operando* X-ray characterization is conducted perpendicular to battery electrodes, providing an average of the chemistry through the entire stack. The cross-sectional pouch cell geometry demonstrated here provides a spatially resolved picture of sulfur diffusion between the cathode and anode, which cannot be achieved by typical characterization methods. Cells consisting of a sulfur/Super P carbon cathode, Li anode, and 1 M  $LiClO_4$  + 0.5  $LiNO_3$  in 1,3-dioxolane/1,2-dimethoxyethane were discharged, and maps were collected at energies sensitive to the sulfur and Li polysulfide species. Examples of these maps demonstrating changes in  $S_8$  and  $Li_2S$  concentrations during discharge are shown in Figure 1. Tuning the incident X-ray energy allows for targeted observations of species such as polysulfides, sulfates, elemental sulfur, and lithium sulfide during cycling.

A novel electrochemical cell with well-defined electrode geometry, electrolyte volume, and current collection has been designed at SSRL to map the battery cross-section in fluorescence mode. This cell will mitigate the challenges of using pouch cells for X-ray experiments such as water and oxygen contamination and sample-to-sample variation. It will also provide more robust, longer-term cycling that better replicates the offline electrochemistry of a typical coin or pouch cell. Because the new cell will increase reproducibility, we can alter electrode architecture and electrolyte composition to directly compare polysulfide and sulfur distributions, illuminating the direct influence that these parameters have on battery chemistry. Using this new design, changes in sulfur and polysulfide chemistry are systematically correlated to structure and performance.

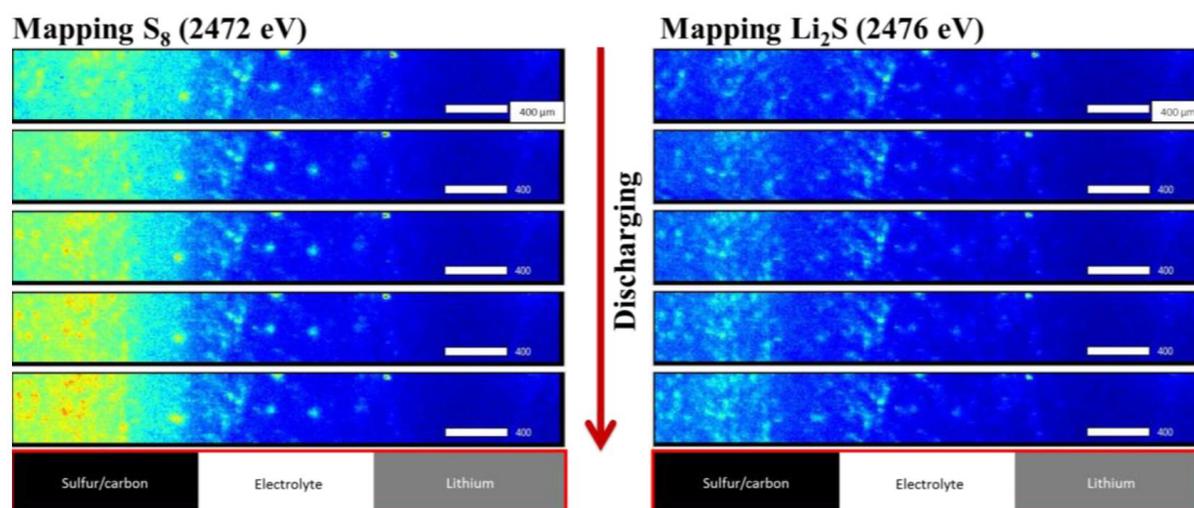


Fig 1. Multi-energy maps of  $S_8$  (left) and  $Li_2S$  (right) during discharging. The cross-sectional cell orientation is shown below for reference. The color map is such that red/yellow indicates higher intensity and blue is lower intensity.

### Speaker Biography:

Elizabeth (Beth) Miller is a postdoctoral scholar in the Toney Group at SLAC National Accelerator Laboratory's Stanford Synchrotron Radiation Lightsource. Her research focuses on characterization of lithium-sulfur batteries using advanced X-ray imaging and spectroscopy methods as part of the Joint Center for Energy Storage Research, the Department of Energy's Energy Innovation Hub for transportation and grid battery technology, and Battery500, a multi-research institution consortium to develop batteries for electric vehicles.



Beth grew up in the suburbs of Philadelphia, Pennsylvania and earned a B.S. with honors in Materials Science and Engineering with a minor in Chemistry from The Pennsylvania State University in 2010. She earned her Ph.D. from Northwestern University in Materials Science and Engineering in 2015 as a fellow in the Department of Energy National Nuclear Security Administration's Stewardship Science Graduate Fellowship. Her graduate research in the Barnett Group focused on developing new anode chemistries for intermediate temperature solid oxide fuel cells. During her time in graduate school, she collaborated with both domestic and international labs and universities to pursue her passion for energy research including Sandia National Laboratories as part of the Sunshine to Petrol project on solar thermochemical processes and at the Technical University of Denmark – Risø's Department of Energy Conversion and Storage, furthering international collaborations on fuel cell technologies.

Teaching and mentoring were integral parts of Beth's motivation in graduate school, which she has continued as a postdoc by mentoring undergraduate and graduate students. She enjoys sharing her enthusiasm for materials science and clean energy with younger students, especially women and girls, via outreach programs at SLAC and Stanford to spread excitement about STEM fields. In her spare time, Beth enjoys baking delicious desserts and exploring California's beautiful landscapes.