Simulations and X-ray Spectroscopy of Li-S Chemistry

U.S. DEPARTMENT OF ENERGY Energy Efficiency & Renewable Energy

PI/Co-PI: Nitash Balsara / David Prendergast (LBNL)

Technical Approach:

- *In situ* X-ray absorption spectroscopy (XAS) of Li-S charge-discharge reaction mechanisms
- Theoretical calculation of X-ray absorption spectra of polysulfide species dissolved in solvent electrolytes
- Molecular dynamic (MD) free energy calculations to determine equilibrium distribution of polysulfide species in solvent electrolyte
- Coupled experimental/theoretical approach to determine speciation of polysulfide solutions and battery cathode

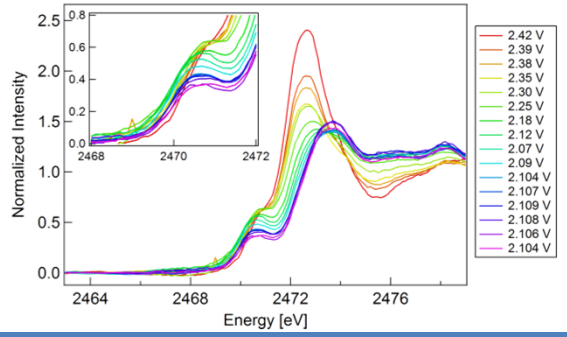
Status:

- Established sulfur K-edge XAS of lithium polysulfides in solvent electrolyte from first principles calculations
- Used theoretical and experimental XAS to identify polysulfide radical anions formed during discharge of an Li-S battery
- Coupled experimental/theoretical approach used to examine polysulfide radical anion formation and stability in ether-based solvents and dimethylformamide (DMF)

Technology:

In situ XAS/electrochemical experiments coupled to theoretical MD and XAS calculations.

Figure: *In situ* XAS of discharging Li-S battery



Objectives:

- Perform *in situ* XAS studies of Li-S chemistry to determine the charge/discharge reaction pathways of Li-S batteries
- Determine equilibrium distribution of polysulfide species in solvent electrolytes using MD free energy calculations
- Investigate solution phase structure and thermodynamics of lithium sulfur polysulfide agglomerates

Deliverables: Lithium sulfur battery discharge and charge reaction mechanisms determined by in situ XAS/electrochemical experiments and supported by theoretical MD and XAS calculations.

Funding:

Duration: 4 yrs (Yr 4)
FY16 Budget: $578K (DOE)

Milestones:

- **Q1:** Calculate thermodynamics of polysulfide disproportionation and radical formation in poly(ethylene oxide) (PEO) and DMF
- **Q2:** Determine polysulfide distribution in PEO and DMF by coupling theoretical calculations to experimental spectroscopy of polysulfide solutions
- **Q3:** Perform in situ XAS tests of Li-S charge/discharge
- **Q4:** Vary charge/discharge rate for in situ XAS testing; compare resulting reaction pathways.