

Electrolyte Structure-Function Relationship and Solvent-Dictated Reaction Mechanisms in Li-S and Li-O₂ Batteries

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Lithium-sulfur (Li-S) and lithium-Oxygen (Li-O₂) batteries have attracted intensive research attentions owing to their potential to provide higher gravimetric energy density than conventional Li-ion batteries. However, Li-S and Li-O₂ batteries have been suffering from poor cycle life, low round-trip efficiency and low practical energy density compared to conventional Li-ion batteries. Different from intercalation-based charge storage, Li-S and Li-O₂ both operate based on solution-mediated redox processes. The development of stable and reversible Li-S and Li-O₂ chemistries requires fundamental understanding on how solvent and salt affect the reaction mechanism and mediation processes. In this presentation, we combine various in situ/operando spectroscopy/analytical techniques to probe the electrolyte structure-function relationship and solvent dictated reaction mechanisms in Li-S and Li-O₂ batteries. We exploit online electrochemical mass spectrometry, operando UV-Vis spectroscopy, rotating ring disk electrode and synchrotron-based X-ray absorption near edge structure spectroscopy to investigate the role of solvent and salt (e.g., donicity, electron-accepting ability, dielectric constant etc) on the reaction mechanism and solution mediation processes in sulfur and oxygen redox chemistries. We will discuss the interplay between solution mediation and solid phase precipitation and the synergistic effect of employing redox mediators and novel cell design in improving the rate and cycle life of Li-S and Li-O₂ batteries.