Solid-Phase Cycling of Sulfur Cathodes using Coulombic Charging under Convective Flow

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The lithium-sulfur battery presents itself as a promising chemistry with the potential to replace current lithium-ion based chemistries due to its higher specific capacity, 1672 mAh/g-sulfur vs. 170 mAh/g-LiFePO4, and high power and energy densities. In addition to near tenfold higher theoretical capacity to current materials, sulfur is an abundant and cheap. However, compared to lithium-ion chemistries, lithium-sulfur electrodes have poor cyclability due in part to the soluble nature of long-chain polysulfide intermediates.

Characteristic plateaus are observed on the voltage profile during discharge, which are generally attributed to the reduction of long-chain polysulfides, at the upper voltage plateau, and the reduction of short-chain polysulfides at the lower plateau. Similarly, this separation can be observed by the occurrence of two distinct current peaks during cyclic voltammetry, however, the voltage plateaus during charge overlap, similarly the forward-scan current peaks in cyclic voltammetry also overlap to form a single combined peak, making an intermediate potential cutoff difficult to distinguish and employ for the prevention of soluble intermediate formation.

An alternative to utilizing potential cutoff, a Coulombic cutoff is effective at separating the intermediate reactions during charge while still using up to 75% of the cell’s capacity. Coulombic cutoff can be achieved through current integration or timed experiments at constant current charge/discharge.

Additionally, the avoidance of soluble intermediates during Coulombic charging is verified when cells are subjected to a convective flow. During voltage-cutoff charging while under convective flow, sulfur cathodes lose soluble intermediates to the bulk to both diffusion and convection, dramatically increasing the active material loss leading to rapid failure. However, Coulombic charging the cell exhibits the same capacities regardless of convective flow or diffusion alone, indicating that no soluble intermediates are formed.

Convection can also be used to rapidly screen and compare electrode materials by accelerating the loss of active material, helping determine the effectiveness of a material at preventing intermediate losses. The expedited fade created by the flow of electrolyte is consistent with the mechanism where capacity loss is a direct result of soluble intermediate loss generated at higher voltages.
**Speaker Biography:**

Dr. Donald A Dornbusch received his PhD in Chemical Engineering from the University of Missouri in May 2015. His research focused on lithium-sulfur, metallic anodes, and convective transport in electrochemical cells.

He is currently the director of research for Homeland Technologies, LLC- a National Science Foundation funded start-up focused on commercialization of academic lithium-sulfur research.