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Role of Polysulfides in Self-Healing Lithium-Sulfur Batteries

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The presentation will review works on the room temperature lithium sulfur batteries since the year 2000. Namely, progresses in capacity, cycle life and efficiency of the cells will be discussed from the science and engineering standpoints. In this context, we will discuss our most recent results based on the development of a novel electrolyte that was designed to solve the cycling instability and inefficiency that are inherent to the Li-S battery. The new electrolyte consisted of lithium polysulfide species (Li_2S_x) dissolved in dimethoxyethane solvent.

The sulfur cathode in the Li-S battery offers superior theoretical capacity (1672 mAh.g^{-1}) compared to all Li-ion battery cathodes. Li-S batteries have the advantages of using an abundant, nontoxic and low-cost cathode material. However, several performance-related issues prevent the development of practical Li-S batteries. These issues are rapid capacity fading and low coulombic efficiency, which are believed to be linked to the dissolution of lithium polysulfides from the sulfur electrode into the electrolyte. To overcome these shortcomings, we developed a conceptually new approach based on mitigating the sulfur loss by leveling the concentration gradient of the polysulfide species at the cathode/electrolyte interface. Because of this concentration gradient, when these species are produced at the cathode, they do not readily migrate into the electrolyte; instead, they are retained at the cathode (Figure 1). The concentration gradient was realized by utilizing novel electrolytes containing pre-dissolved lithium polysulfides (Li_2S_x) as lithium salt along with a redox shuttle inhibitor. With this new electrolyte, electrodes made from a simple mixture of carbon black and sulfur powders demonstrated a superior high capacity and almost 100% coulombic efficiency with excellent cyclability (Figure 2). Furthermore, this novel electrolyte does not need conventional lithium salts, such as LiTFSI, since lithium polysulfides (Li_2S_x) promote Li-ion conduction in the electrolyte; they also contribute to the overall capacity of the Li-S cell.

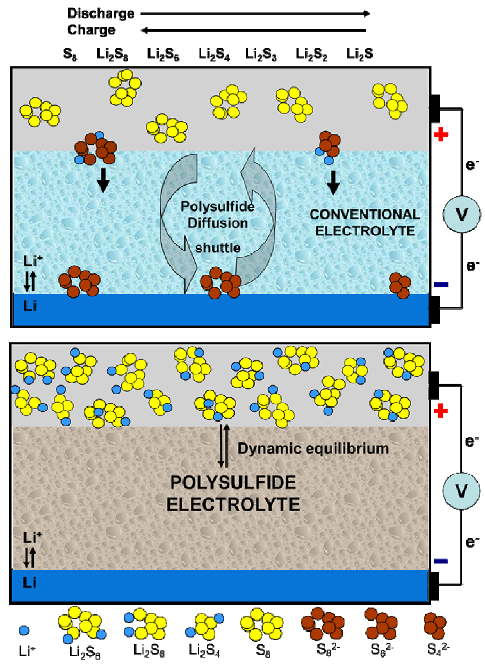

Figure 1. Schematic diagram of (up) Li-S battery using a conventional electrolyte, in which polysulfides produced at the cathode during discharge dissolve into the electrolyte; (down) Li-S battery using the polysulfide electrolyte, in which produced polysulfides are retained at the cathode.

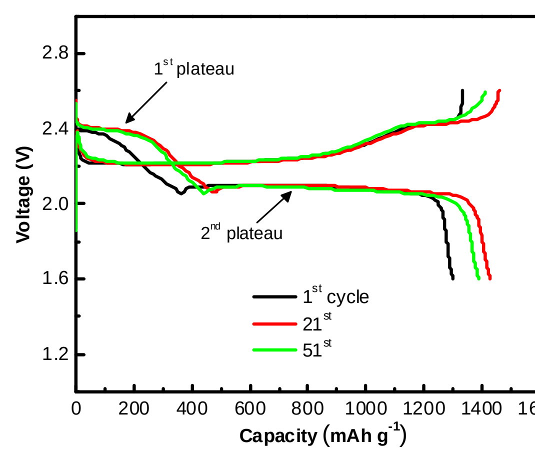

Figure 2. Charge-discharge voltage profiles of Li-S cell containing polysulfide electrolyte.

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