Lithiation and Delithiation Processes in Lithium–Sulfur Batteries from Ab Initio Molecular Dynamics Simulations

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1. INTRODUCTION

Lithium–sulfur (LiS) batteries are widely used as the energy storage device in portable consumer electronics, although their limited energy density is still a major challenge. From a practical viewpoint, it is crucial to deliver a specific capacity of about 300 Wh/kg, which leads to applications in a low-power needle such as in-rental-range electric vehicles. The cathode materials used in LiS batteries have a limited structure, and sulfur is an excited or generated form, making it an essential metal in all electrochemical reactions. Such an essential cathode is essential for the stability and cyclability that are required for practical batteries. The underlying reasons responsible for the limited capacity.

In LiS, one of the most significant factors is the electron transfer, which is characterized by a reduction potential of 2.3 eV, which is the highest among all lithium-based cathode materials. Unlike other cathode compounds, to which the electrode structure remains stable, a series of complex electrochemical reactions take place in sulfur and the structural stability of the cathode material is maintained, leading to a much improved capacity. However, the high capacity is often reduced by the structural instability and the decrease in the electrical conductivity. The sulfur cathode is also seen to be an ideal candidate for a high specific capacity battery. For these reasons, sulfur is among the most promising cathode materials for LiS batteries due to the subject of certain research efforts. In this context, the concept of anode cathode as a cathode material has emerged as a unique approach to designing high-performance cathode materials.

However, the actual reaction sequence is extremely complex, and many intermediate molecular species that initiate these processes are not yet apparent. Two major platforms have been suggested:

- Lithium-sulfur batteries are a promising alternative to the Li-ion technology due to their high theoretical capacity and low cost. However, sulfur cathode undergoes a series of complex electrochemical reactions with substantial structural changes, resulting in poor stability and cyclability. Using ab initio molecular dynamics simulations, the atomistic processes that take place at the cathode/electrolyte interface during charge and discharge were revealed. Novel atomistic insights were proposed, including the formation of various intermediate species and the importance of local lithium concentration, that could help to improve our fundamental understanding of the electrochemical processes observed in lithium-sulfur batteries.