Adsorption Mechanisms of Lithium Polysulfides on Graphene-Based Interlayers in Lithium Sulfur Batteries

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One of the most critical problems in lithium-sulfur (Li-S) batteries is the shuttle effect. The transfer of soluble lithium polysulfides (LiPSs) from the sulfur cathode to the lithium anode leads to a degradation in Li-S battery capacity and life cycles. Recent studies reveal that the carbon-based interlayer materials introduced between the cathode and anode can effectively improve the shuttle effect problem and increase the battery life cycles. In this work, different types of the N-doped, S-doped, and N, S co-doped graphene surfaces are investigated by theoretical calculations. We find that a strong interaction may exist between some of the heteroatom-doped graphene surfaces and lithium ions, and that the adsorption of LiPSs may proceed via one of the three mechanisms, the dissociative, the destructive, and the intact adsorptions. Detailed structural and electronic analyses indicate that the Li-trapped N, S co-doped graphene interlayers (NSG1 and NSG2) could efficiently reduce the shuttle effect through the intact adsorption mechanism. Our results provide a plausible explanation on the observed better performance of the N, S co-doped graphene interlayers in Li-S batteries.

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