Understanding the Role of Graphene Oxide in Li/S Cells from Quantum Mechanics Calculations

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Li/S electrochemical cells are considered promising candidates for rechargeable batteries with high specific energy[1-4]. A strategy[5] was proposed to immobilize S on quasi-two-dimensional graphene oxides (GO) to prepare GO-S nanocomposite positive electrodes for Li/S cells.

We report results of quantum mechanics (QM) studies of GO and Li/S cells. Experiments show that Li/S cells are limited by the dissolution of Li polysulfides into the electrolyte during the discharge process. Electrochemical studies find that incorporating GO into the sulfur positive electrode greatly improves the battery performance[5]. This is due to the ability of GO to adsorb polysulfides[6]. Further investigations are needed to better understand chemical reactions and interactions at the GO-S positive electrode.

The cathodic reaction involves the reaction to convert S_8 to Li_2S. Using density functional theory (DFT) calculations, we find the intermediates of the reactions include LiS_8, S_4, LiS_4, Li_2S_4, LiS_2, and Li_2S_2. Our further calculations show the adsorption process between Li polysulfides and GO functional groups (=O, -OH, C-O-C). We also perform calculations to simulate the interaction between Li polysulfides and tetraglyme. We find that the interaction between the Li cation and the GO functional groups plays an important role in the adsorption process.

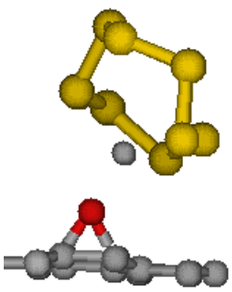

Figure 1: LiS_8 on GO.

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