

Role of the Electrolyte Solvent Effects on Polysulfides Adsorption Characteristics in Metal-sulfur Batteries

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The adsorption strengths of polysulfides and cathode-anchoring materials (CAMs) play a crucial role in mitigating the shuttle effect and enhancing the reaction kinetics of the sulfur reduction reaction in metal-sulfur batteries (MSBs). However, most theoretical studies overlook the intrinsic influence of electrolyte solvents. This work investigates the role of solvents in the adsorption characteristics of polysulfides and CAMs in sodium-sulfur (Na-S) batteries. Specifically, MoS₂, a two-dimensional semiconducting transition metal dichalcogenide, is designed as a CAM. Using density functional theory (DFT), we examine polysulfide adsorption on pristine and doped MoS₂ monolayers. Several key parameters (*i.e.*, the electronic density of states (DOS), Bader charge analysis, adsorption energies, and charge density differences of both bare surfaces and complexes) are evaluated under two conditions: (i) vacuum and (ii) various conventional solvents with low to high dielectric screening. Our findings reveal that solvent-induced charges can slightly modify the DOS and energy gap of MoS₂. Although these modifications are minor, the presence of solvents significantly weakens the adsorption energies. Higher solvent dielectric constants lead to weaker adsorption energies, corresponding to reduced charge transfer and lower DOS at the Fermi level. These results manifest the crucial role of solvation effects in accurately predicting interaction strengths between polysulfides and MoS₂ as a CAM in MSBs.