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Covalent Organic Frameworks as Sulfur Hosts for Lithium–Sulfur Batteries

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[Introduction]

Electrical energy storage is one of the most critical needs of 21st century society. Lithium-sulfur batteries (LSBs) have attracted considerable attention from both the academic and industrial communities for their potential capability of meeting practical applications in new electrical energy storage. LSBs have many conspicuous advantages, such as a high theoretical capacity of 1675 mAh/g, low cost, natural abundance and environmental friendliness, which making LSBs become one of the most promising next-generation batteries. The shuttling process involving lithium polysulfides is one of the major factors responsible for the degradation in capacity of LSBs. Two approaches have been employed to overcome this defect: one is to load sulfur into high porous materials, and another is to combine sulfur and to form a cross-linked polymer. We supposed that a combination of the two approaches using covalent organic framework-graft-polysulfide (COF-graft-PS) would be an efficient methodology. A COF-graft-PS was synthesized by graft co-polymerization of elemental sulfur onto the skeleton of a functionalized COF. The polysulfide can be robustly impregnated into the pores through the strong covalent bonds between COF and sulfur. The COF serves as the host as well as the cross-links of polysulfides.

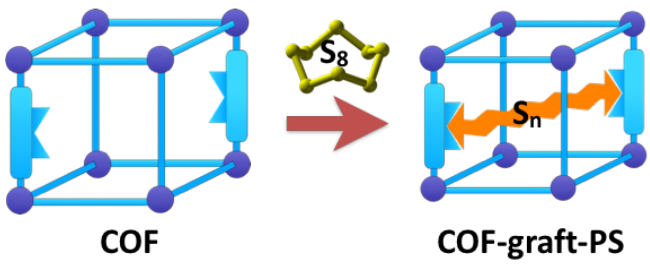

Figure 1. Synthetic strategy of COF-graft-PS in this research.

[Experimental]

Firstly, an alkinyl group functionalized COF was synthesized by a typical solvothermal method. Then, the COF and sulfur were mixed and heated. By copolymerization of sulfur and functional groups on the surface of the COF pores, the sulfur can be robustly impregnated into the pores through the strong covalent bonds between host and sulfur and the COF-graft-PS will be generated. The COF-graft-PS was utilized as the cathode for LSBs and the electrochemical properties were tested.

[Results and Discussion]

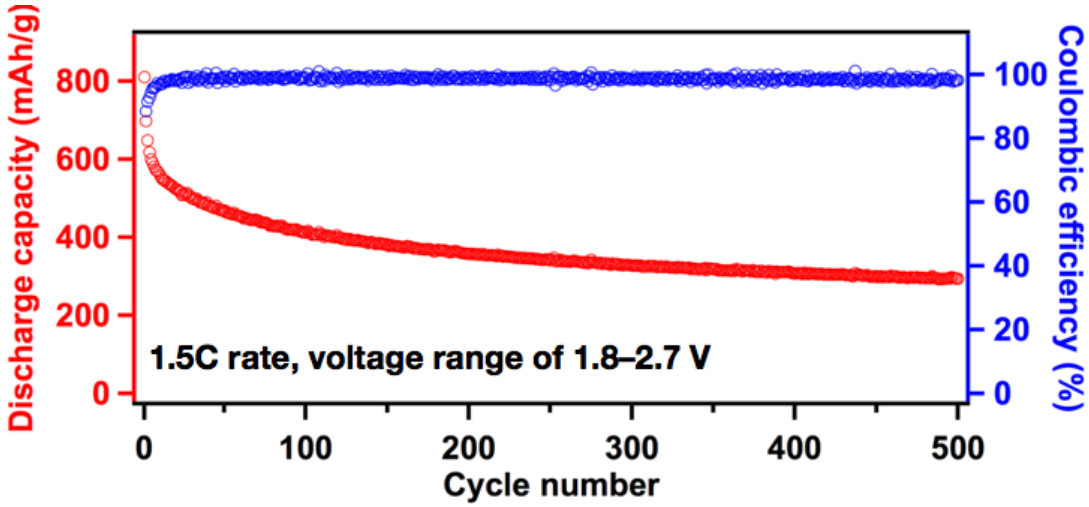

Figure 2. Cycling performance of COF-graft-PS over 500 cycles at a charge/discharge rate of 1.5 C rate.

The COF-graft-PS based LSBs show high capacity and great cycling performance. The stable cycling performance is due to the robust sulfur cathode structure in which sulfur is homogeneously distributed throughout the regular pores within the framework together with the C-S covalent links. The chemical sulfur impregnation within the pores of COF effectively suppresses the dissolution of polysulfides into the electrolyte.