Novel Organosulfide electrodes for Lithium-Sulfur Battery

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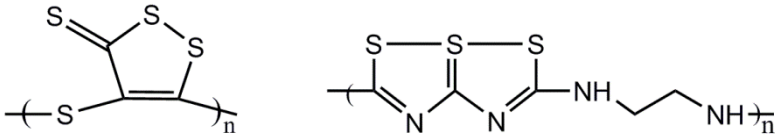
Recently, Organodisulfide compounds have attracted extensive research interests as cathode materials for Li-S batteries, due to their advantages of higher theoretical capacity, environmental friendness, lightweight and abundant resources. The key moiety of high specific capacity in organodisulfide compounds is the reversible two-electron redox reaction of the disulfide bond (S-S).

However, the major problem of organodisulfide compounds as electrode materials in Li-S is the large capacity fading during cycling caused by the high solubility of polysulfides in liquid electrolytes. In order to improve the cyclic stability of the electrodes, one of the important strategies is to polymerize the small molecules into insoluble polymer backbones.



PDTT PETDP

Figure 1. Molecular configuration of PDTT and PETDP

Based on this strategy, the electrochemical performance and energy storage mechanism of Poly [1, 2-dithiole-3-thion-4(5)-thio] (PDTT) and Poly [2-(1, 2-ethylenediamino)-1, 6, 6a, Δ^4-trithia-1, 6-diaza-pentalen-5-yl] (PETDP) were designed, synthesized, and investigated. Based on the two-electron reaction, PDTT can provide theoretical capacity of 326 mAh g^{-1}, and the theoretical capacity of PETDP is calculated to be as high as 371 mAh g^{-1}. Electrochemical performances of PDTT and PETDP were evaluated by cyclic voltammogram (CV) and galvanostatic discharge/charge measurements. CV results indicated that PDTT electrode has two pairs of redox peaks at 2.09/2.38 and 2.29/2.63 V. PETDP cathode also presents two pairs of well-defined redox peaks at 2.11/2.26 and 2.37/2.42 V, respectively. Cyclic performance of PDTT and PETDP was compared using two type of electrolytes, the ether based (1M LiTFSI in DME/DOL) and carbonate based (1M $LiPF_6$ in EC/DMC) electrolytes. Results show that both of polysulfides deliver better cyclic stability in ether based electrolyte than in carbonate based one. In addition, the discharge and charge profiles of PDTT electrode are consistent with its CV curves and having specific capacity of 321 mAh g^{-1} at a current density of 0.1 C (1C=33 mA g^{-1}), this value is as high as 98.4% of the theoretical capacity. Even at 0.5C rate, high initial capacity of 318 mAh g^{-1} (97.5% of theoretical value) was obtained, confirming the almost fully utilization of active materials. When cycled over 50 cycles, PDTT electrodes can still deliver high capacities of 291 mAh g^{-1} with capacity retentions of 90.6%, which is much higher than most of the Li-S batteries, showing a high potential to be used as organodisulfide cathode materials for high capacity Li-S batteries.

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