

Polypyrrole as Bifunctional Materials for Advanced Li-S Batteries

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Abstract : The practical application of Li-S batteries is hampered due to poor cycling stability caused by electrolyte-dissolved lithium polysulfides. Dual functionalities such as strong chemical adsorption stability and high conductivity are highly desired for an ideal host material for a sulfur-based cathode. Polypyrrole (PPy), as a conductive polymer, was widely studied as matrixes for sulfur cathode due to its high conductivity and strong chemical interaction with soluble polysulfides. Thus, a novel cathode structure consisting of a free-standing sulfur-polypyrrole cathode and a polypyrrole coated separator was designed for flexible Li-S batteries. The PPy materials show strong interaction with dissoluble polysulfides, which could suppress the shuttle effect and improve the cycling stability. In addition, the synthesized PPy film with a rough surface acts as a current collector, which improves the adhesion of sulfur materials and restrain the volume expansion, enhancing the structural stability during the cycling process. For further enhancing the cycling stability, a PPy coated separator was also applied, which could make polysulfides into the cathode side to alleviate the shuttle effect. Moreover, the PPy layer coated on commercial separator is much lighter than other reported interlayers. A soft-packaged flexible Li-S battery has been designed and fabricated for testing the practical application of the designed cathode and separator, which could power a device consisting of 24 light-emitting diode (LED) lights. Moreover, the soft-packaged flexible battery can still show relatively stable cycling performance after repeated bending, indicating the potential application in flexible batteries. A novel vapor phase deposition method was also applied to prepare uniform polypyrrole layer coated sulfur/graphene aerogel composite. The polypyrrole layer simultaneously acts as host and adsorbent for efficient suppression of polysulfides dissolution through strong chemical interaction. The density functional theory (DFT) calculations reveal that the polypyrrole could trap lithium polysulfides through stronger bonding energy. In addition, the deflation of sulfur/graphene hydrogel during the vapor phase deposition process enhances the contact of sulfur with matrixes, resulting in high sulfur utilization and good rate capability. As a result, the synthesized polypyrrole coated sulfur/graphene aerogel composite delivers a specific discharge capacity of 1167 mAh g⁻¹ and 409.1 mAh g⁻¹ at 0.2 C and 5 C respectively. The capacity can maintain at 698 mAh g⁻¹ at 0.5 C after 500 cycles, showing an ultra-slow decay rate of 0.03% per cycle.

Keywords : polypyrrole, strong chemical interaction, long-term stability, Li-S batteries

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