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A polydopamine coating ultralight graphene matrix as a highly effective polysulfide absorbent for high-energy Li–S batteries

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ABSTRACT

The soft polydopmine (PDA) layer with polar functional group was coated onto the ultralight graphene sheets (HRG) using a simple approach in a mixture of ethanol and water solution. Sulfur was further dispersed onto the surface of PDA-HRG sheets by S/CS_2 solution impregnation method to obtain PDA-HRG/S composite as the cathode material for lithium sulfur batteries. The resulting composites are characterized by SEM, TEM, XRD and so on. The ultralight graphene matrix could improve the conductivity of the electrode and offer a large surface for deposition of sulfur, while the coating of hydrophilic soft PDA layer can adsorb polar polysulfides on-site and accommodate the volume change of S during the discharging processes, resulting in an excellent electrochemical performance.

1. Introduction

In view of the wide spread application of portable electronic devices including mobile phones and laptops, rechargeable lithium-ion (Li-ion) batteries as the major energy storage in off-grid renewable energy is becoming the dominant power source in modern society. However, the progress in Li-ion batteries needs to be carried further to satisfy the ever-rising energy requirements for clean and efficient high-density and high-capacity energy storage devices [1–8]. Lithium/sulfur batteries with sulfur as a cathode and Li as an anode have attracted more and more attention because of its theoretical capacity of 1675 mAh g^{-1} and theoretical specific energy of 2600 Wh kg^{-1}. Sulfur also possesses other advantages including abundant, low cost, and nontoxic. Therefore, lithium sulfur batteries have been considered as one of the most potential next-generation high energy lithium batteries [9–12]. Nevertheless, several inherent problems, including low electrical conductivity of sulfur, volumetric expansion of the sulfur during discharge, the dissolution of intermediate polysulfide in electrolyte and shuttling effect of polysulfide between the positive and negative electrodes, result in low utilization of active sulfur, low specific capacity, low coulombic efficiency and poor cycle life, and thus greatly limitings practical application of Li–S batteries [13–17].

To solve these issues, various conducting matrix, such as mesoporous and microporous carbon [18–21], carbon nanotubes/fibres [22–26], hollow porous carbon spheres [27,28], graphene [29,30], and polymer [31–33], have been widely applied to confine the active sulfur. Among these carbon materials, graphene is promising and widely investigated because of its high electrical conductivity and large surface area. As reported in other literature [34], the S3pz2 electrons of sulfur have intimate interaction with the antibonding conjugated p* states of graphene plane, therefore, the introduction of graphene has several advantages including a large contact area with sulfur, a short transport path way for both electrons and Li^+, a confinement ability for polysulfides and sufficient space to alleviate sulfur volumetric change. However, the polysulfide species that are produced during discharge and charge processes are highly soluble and hard to be completely prevented by the weak interactions between nonpolar carbon and polar polysulfides. To mitigate this problem, in previous reports on graphene–sulfur composites, the so-called graphene was prepared by solution exfoliation of graphite oxide, in which a large number of

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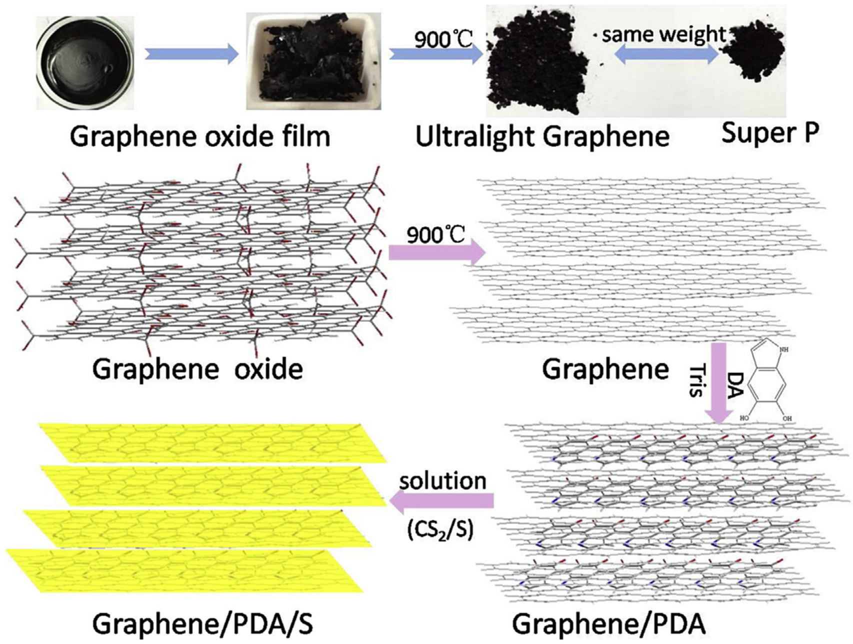

Fig. 1. Schematic illustration of the fabrication of PDA-coated ultralight graphene/sulfur composite.

oxygen-containing functional groups are introduced [35,36]. This approach is effective for confining the polysulfides with oxygen-containing functional groups but could greatly reduce the overall electronic conductivity due to it lacks an extended π-conjugated orbital system, which would compromise the batteries performance [37].

Our work is devoted to looking for the cathode material with better electrochemical performance for Li–S batteries to satisfy the ever-growing demands for higher energy/power densities. Herein, we use a facile approach to prepare PDA-HRG/S composite as a cathode for high-performance lithium sulfur batteries. A soft polar polydopamine layer was introduced to help trap the highly polar polysulfide from dissolving in electrolyte and to accommodate the volume expansion of S during the discharging processes. On one hand, the use of graphene could improve the conductivity of the entire electrode and offer a large surface for deposition of sulfur. On the other hand, dopamine can spontaneously polymerize on the surface or interspace of graphene sheets to form polydopamine

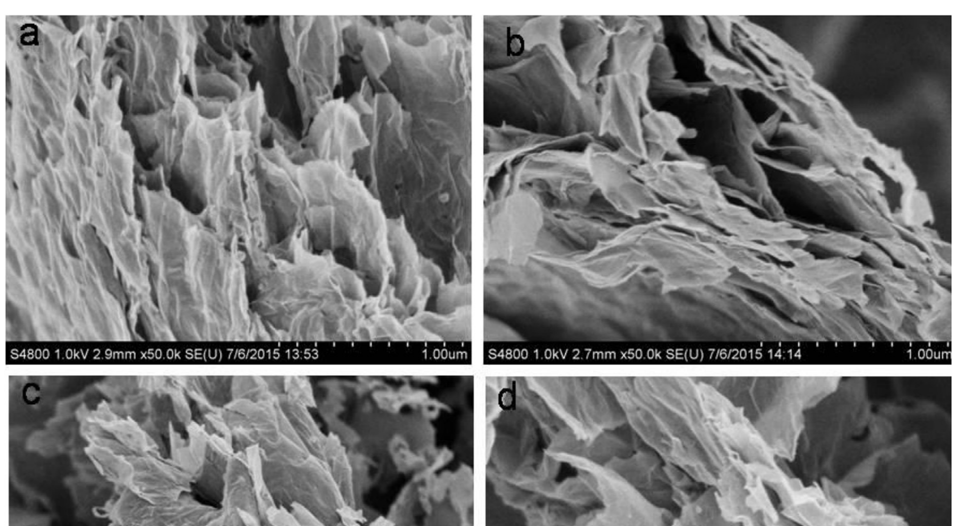

Fig. 2. FESEM images of HRG (a, b) and HRG/S (c, d).