Porous honeycomb-like C_3N_4 /rGO composite as host for high performance Li-S batteries

Xiaomeng Bai¹, Chunsheng Wang¹, Caifu Dong^{1,2}, Chuanchuan Li^{1,2}, Yanjun Zhai³, Weiwei Si¹ and Liqiang Xu^{1*}

ABSTRACT Lithium-sulfur (Li-S) batteries have attracted extensive attention along with the urgent increasing demand for energy storage owing to the high theoretical specific capacity and energy density, abundant reserves and low cost of sulfur. However, the practical application of Li-S batteries is still impeded due to the low utilization of sulfur and serious shuttle-effect of lithium polysulfides (LiPSs). Here, we fabricated the porous honeycomb-like C_3N_4 (PHCN) through a hard template method. As a polar material, graphitic C_3N_4 has abundant nitrogen content (~58%), which can provide enough active sites to mitigate shuttle-effect, and then conductive reduced graphene oxide (rGO) was introduced to combine with PHCN to form PHCN/rGO composite in order to improve the utilization efficiency of sulfur. After sulfur loading, the PHCN/rGO/S cathode exhibited an initial discharge capacity of 1,061.1 mA h g⁻¹ at 0.2 C and outstanding rate performance at high current density of 5 C (495.1 mA h g⁻¹), and also retained 519 mA h g⁻¹ after 400 cycles at 1 C. Even at high sulfur loading (4.3 mg cm⁻²), the capacity fade rate was only 0.16% per cycle at 0.5 C for 200 cycles. The above results demonstrate that the special design of PHCN/rGO composite as sulfur host has high potential application for Li-S rechargeable batteries.

Keywords: porous honeycomb-like, graphitic C_3N_4 , long cycle performance, lithium-sulfur batteries

INTRODUCTION

Lithium-sulfur (Li-S) batteries have been considered as promising candidates for next-generation electric energy storage devices owing to the high theoretical specific capacity (1,675 mA h g⁻¹), high natural abundance, low

cost, and environmental friendliness [1–3]. Although Li-S batteries have many considerable advantages, there are still several problems for practical applications. First, the active sulfur and related Li_2S_2 / Li_2S are insulating, leading to low utilization efficiency of the active materials. Second, lithium polysulfides (LiPSs, Li_2S_n , $n > 2$) dissolved in the organic electrolyte would penetrate through the separator to Li anode, resulting in the serious “shuttle-effect” [4–7] and rapid capacity fading. Third, the large volume expansion (~80%) during cycling causes the destruction of host material structure.

In order to solve these problems, a series of materials as host has been studied. Among various materials, both non-polar materials such as highly conductive carbon-based materials, and polar materials including metal oxides [8,9], metal sulfides [10,11], metal nitrides [12] and metal carbides [13,14] have shown promising applications as sulfur hosts for Li-S batteries. However, the rational design of new sulfur host materials with special structure and high conductivity is still anticipated.

Graphitic C_3N_4 is a polar non-metal material [15], which includes three types of nitrogen in its intrinsic structure: pyridinic nitrogen, pyrrolic nitrogen and graphitic nitrogen [16]. It has been theoretically and experimentally testified [17,18] that the primary active sites are pyridinic nitrogen, which can interact with LiPSs to accelerate the electrochemical conversion processes and lessen polarization. Pyridinic nitrogen is sp^2 hybridized with lone pair electrons, so it can generate chemical interaction with LiPSs *via* Li–N to suppress shuttle-effect and improve the performance of Li-S batteries [19,20]. Recently, graphitic C_3N_4 with layered structure becomes

¹ Key Laboratory of Colloid & Interface Chemistry (Shandong University), School of Chemistry and Chemical Engineering, Ministry of Education, Shandong University, Jinan 250100, China

² Shenzhen Research Institute of Shandong University, Shenzhen 518057, China

³ Shandong Provincial Key Laboratory/Collaborative Innovation Center of Chemical Energy Storage & Novel Cell Technology, Liaocheng University, Liaocheng 252059, China

* Corresponding author (email: xulq@sdu.edu.cn)

the focus of research and has been applied as sulfur host due to the advantages of high nitrogen content and intrinsic polarity. Graphitic C_3N_4 as host with a long cycle life and high areal sulfur loading has been reported. For example, graphitic C_3N_4 as host showed an ultralow capacity fade rate of 0.04% per cycle for 1,500 cycles owing to the strong polysulfides trapping capability of the material [17]. Graphitic C_3N_4 and graphene hybrid as host delivered an excellent cycling performance with only 0.087% capacity fading per cycle over 600 cycles because the graphitic C_3N_4 can effectively prevent the shuttle-effect by catalyzing the fast conversion of soluble LiPSs [21]. The interactions of layer structured graphitic C_3N_4 with S_8 and LiPSs have also been studied using first-principles calculations. The results showed that graphitic C_3N_4 had a physical and strong binding with S_8 (which may result in improved sulfur loading) and it also had a chemical and stronger interaction with LiPSs [18].

Layered graphitic C_3N_4 as host material has been proved with many advantages. However, porous honeycomb-like C_3N_4 (PHCN) as sulfur host has not been reported up to date. As the semiconducting nature of graphitic C_3N_4 limits the electron transport and the utilization of sulfur to some extent [22], highly conductive reduced graphene oxide (rGO) was selected to incorporate the PHCN to overcome the problem. The composite has three advantages as sulfur host: (1) the PHCN can accelerate the redox reaction kinetics and supply strong chemical adsorption for LiPSs to mitigate the shuttle-effect. (2) The specific structure design is beneficial to the infiltration and storage of electrolyte, alleviating volume expansion during cycling, and the mesoporous structure could improve physical adsorption of LiPSs. (3) rGO could provide fast ion/electron pathways effectively. The obtained electrode displays excellent performances. Therefore, the hybrid electrode is proved to be an effective way to deliver an excellent electrochemical performance for Li-S batteries.

EXPERIMENTAL SECTION

Preparation of silica spheres

The uniform silica (SiO_2) spheres were synthesized according to the previously reported literature [23]. At first, 15 mL of deionized water, 40 mL of ethanol, and 2.5 mL of ammonia were mixed under stirring. Then, 2.0 mL of tetraethyl orthosilicate (TEOS) was quickly added to the above solution. The white precipitate was obtained after stirring for 12 h. The product was washed with deionized water and ethanol several times and collected by cen-

trifugation (6,800 rpm, 15 min). Finally, the product was dried in vacuum oven at 60°C for 4 h.

Preparation of porous honeycomb-like C_3N_4

SiO_2 spheres (0.9 g) were dispersed in 25 mL of deionized water by sonication for 10 min to form a solution A; in the meantime, 3.1 g of polyimine (PEI) was dissolved in 500 mL of deionized water and stirred for 5 min to form a solution B. Then solution A was poured into solution B and the mixture was transferred into an oil bath heated from room temperature to 90°C and then maintained at 90°C for 3 h. After the solution was cooled to room temperature, the product was centrifuged for three times with water, and dried in vacuum oven at 60°C for 6 h. Then, the product was calcined in argon at 550°C for 2 h [24]. The obtained powder was added to a cyanamide solution (CH_2N_2 , 12 mL, 50 wt.%) and stirred for 20 min, and the product was centrifuged and dried in vacuum at 60°C for 24 h. The obtained compound was further annealed in argon at 550°C for 4 h. After the product was soaked into HF solution (20 mL, 5 wt.%) for 8 h to remove SiO_2 , pure PHCN was obtained finally.

Preparation of PHCN/rGO

Firstly, 60 mg of PHCN was dispersed in 10 mL of ethanol and sonicated for 5 min to obtain a homogeneous suspension C. In the meantime, 30 mg of rGO was also dispersed in 30 mL of ethanol with sonication for 30 min to obtain a homogeneous suspension D. Then, suspension C was added into D and stirred for 12 h. Finally, the PHCN/rGO composite was obtained through evaporation and freeze-drying processes.

Preparation of the PHCN/rGO/S composite

The PHCN/rGO/S composite was prepared through the melt-diffusion method. Typically, the PHCN/rGO powder and elemental sulfur with a weight ratio of 3:7 were mixed uniformly in a mortar and then transferred into a sealed glass vessel. The mixture was heated from room temperature to 155°C and kept at 155°C for 12 h. rGO/S composite was prepared using the same method mentioned above.

Adsorption tests

The Li_2S_6 solution was prepared by dissolving sulfur and Li_2S in a mixed solvent of 1,3-dioxolane/1,2-dimethoxyethane (DOL/DME) with a volume ratio of 1:1. Then, the mixture was stirred under Ar protection at 60°C for 48 h to obtain the Li_2S_6 solution with brown color. The Li_2S_6 solution was added into three vials (6 mol L^{-1} , 3 mL), and

two of them were added with 15 mg PHCN/rGO and rGO, respectively. The whole testing process was operated in an argon-filled glovebox.

Electrochemical measurements

The tests for electrochemical performance were carried out by using CR2032 coin cells with a lithium foil as the counter and reference electrode at room temperature. The slurry was prepared by using PHCN/rGO/S, ketjen black, and polyvinylidene difluoride (PVDF) with a mass ratio of 8:1:1 (wt.%) in the solvent of *N*-methyl-pyrrolidinone (NMP). The homogeneous slurry was pasted onto an aluminum foil. Then the electrode with a mass loading of sulfur 1.1–1.3 mg cm⁻² (with round disk-like shape and a diameter of 12 mm) was dried in a vacuum oven for 4 h. The electrolyte was 1 mol L⁻¹ lithium bis-trifluoromethane sulfonimide (LiTFSI) in DOL/DME (*v/v*=1:1) containing 2 wt.% LiNO₃, the separator was Celgard 2500 membranes. The cells were assembled in an argon-filled glovebox. The cycling performance was measured by using a battery test station (LANDCT-2001A, Wuhan, China) at room temperature with a voltage window from 1.7 to 2.8 V. The cyclic voltammetry (CV) was carried out at a scanning rate of 0.1 mV s⁻¹ on an electrochemical workstation (Shanghai CHI 760E).

Materials characterization

The morphologies and structures of the samples were examined by transmission electron microscopy (TEM, JEM-1011, Japan) and field emission scanning electron microscopy (FESEM, JSM-7600F, Japan). The characterization of the structure of the samples was performed using an X-ray diffraction (XRD, Bruker D8-Advanced X-ray) equipped with Cu K α radiation. Energy dispersive spectrometry (EDS) was used to investigate the elements of materials. The contents of sulfur in the composites were measured by thermal gravimetric analysis (TGA, SDTA851) from room temperature to 700°C with a heating rate of 5°C min⁻¹. The Brunauer-Emmett-Teller (BET) specific surface area and the pore size distribution were determined by the nitrogen adsorption-desorption analysis. X-ray photoelectron spectra (XPS) measurements were carried out using an ESCALAB 250 spectrometer with Mg K α irradiation.

RESULTS AND DISCUSSION

The schematic step for synthesizing the PHCN is illustrated in Scheme 1. SiO₂ spheres were used as the hard templates and modified by PEI, then 12 mL of CH₂N₂ solution (50 wt.%) was added and stirred for 20 min and

Scheme 1 Illustration of the fabrication process of porous honeycomb-like C₃N₄.

then dried for 24 h. The sample was further annealed at argon atmosphere at 550°C for 4 h. Finally, pure PHCN was obtained by etching SiO₂ using HF solution [24]. Fig. S1 shows the morphology of the template SiO₂, in which the spheres have an average diameter of ~100 nm. Fig. S2 displays the morphology of SiO₂@CH₂N₂ spheres, which have a core-shell structure and larger diameters compared with the original SiO₂ spheres, indicating that the surface of SiO₂ spheres were uniformly coated with CH₂N₂, and the thickness of CH₂N₂ shell is ~10 nm.

The typical TEM image (Fig. 1a) and SEM images with different magnifications (Fig. 1b, c) of PHCN obviously demonstrate that PHCN possesses mesoporous structure. The TEM image (Fig. 1d) and SEM images (Fig. 1e, f) of PHCN/rGO indicate that both PHCN and rGO still maintain their original morphology and structure, and composites form successfully. The TEM (Fig. 1g) and SEM (Fig. 1h) images of PHCN/rGO/S indicate that sulfur has been loaded successfully. The SEM image and corresponding elemental mappings of the PHCN/rGO/S composite in Fig. 1i indicate that C, N and S are all uniformly distributed in the composite.

Fig. 2a shows the XRD patterns of PHCN and PHCN/rGO, in which two main diffraction peaks of PHCN located at around 13.4° and 27.1° are observed. The obvious peak at 27.1° could be attributed to the interlayer stacking (002) of PHCN, and the weak peak at 13.4° corresponds to the in-plane (100) repeating motifs of the tri-s-triazine network [15–17]. Compared with the PHCN, a new broad peak at 43.5° appears in the PHCN/rGO pattern, and the shape of the peak centered at 27.1° becomes wider owing

Figure 1 (a) A typical TEM image of PHCN. (b, c) SEM images of PHCN. (d) A typical TEM image of PHCN/rGO. (e, f) SEM images of PHCN/rGO. (g) A typical TEM image of PHCN/rGO/S. (h) SEM image of PHCN/rGO/S. (i) A typical SEM image and the corresponding elemental mapping of PHCN/rGO/S.

to the addition of rGO. Fig. 2b displays the TGA curve of PHCN/rGO/S under a nitrogen atmosphere, and sulfur content in the composite is determined to be 72 wt%. Fig. S3 shows the TGA curve of rGO/S. The nitrogen adsorption-desorption isotherms are shown in Fig. 2c, which belong to a typical IV hysteresis. According to the BET model, the specific surface areas of PHCN/rGO and PHCN/rGO/S composite are 112.7 and $25.7 \text{ m}^2 \text{ g}^{-1}$, respectively. And the total pore volume is 0.363 and $0.201 \text{ cm}^3 \text{ g}^{-1}$, respectively. The obviously decrease of the specific surface area and pore volume after sulfur impregnation indicates that part of pores are occupied by sulfur [4]. Fig. S4 shows the nitrogen adsorption-desorption isotherms of rGO and rGO/S, in which the specific surface area of rGO and rGO/S composite are 358.5 and $8.9 \text{ m}^2 \text{ g}^{-1}$, and the corresponding total pore volume is 0.566 and $0.065 \text{ cm}^3 \text{ g}^{-1}$, respectively, also indicating that the most of pores are occupied by sulfur after sulfur loading. The pore size distribution curves of PHCN (Fig. S4c) and PHCN/rGO (Fig. 2d) indicate that the pore size is basically distributed

within 10 nm, further verifying that PHCN possesses mesoporous structure, and the incorporation of rGO does not affect the porous structure.

The XPS survey spectrum of the PHCN/rGO in Fig. 3a indicates that the atomic percentage of C, N and O is 56.06%, 32.06% and 11.88%, respectively. The atomic contents of C, N and O detected from the EDS spectrum (Fig. S5) are 60.39%, 30.88% and 7.85%, respectively. Both results indicate that the nitrogen content is abundant. The C 1s spectrum of PHCN/rGO in Fig. 3b is deconvoluted into three peaks, in which the typical peak at 284.7 eV is identified as the reference from C-C to calibrate the binding energy of elements [19]. The peaks at 285.7 and 288.7 eV could be attributed to C-O and C-N-C, respectively. The N 1s spectrum in Fig. 3c can be deconvoluted into three peaks, which are ascribed to pyridinic N (399.4 eV), pyrrolic N (400.7 eV), and graphitic N (401.6 eV). It is known that pyridinic N is approved to be one of the primary available adsorption sites for LiPSs in N-containing non-metal materials [25]. The abundant

Figure 2 (a) XRD patterns of the PHCN and PHCN/rGO. (b) A typical TGA curve of PHCN/rGO/S. (c) N_2 adsorption-desorption isotherm loop of PHCN/rGO and PHCN/rGO/S composites. (d) Pore size distribution of the PHCN/rGO.

Figure 3 XPS spectra of PHCN/rGO. (a) Survey spectrum, (b) C 1s spectrum, (c) N 1s spectrum, and (d) O 1s spectrum.

electronegative pyridinic N in PHCN/rGO could provide lots of active sites to anchor the discharge intermediates by forming Li–N bond. The O 1s spectrum in Fig. 3d indicates that PHCN/rGO also contains C=O and C–O groups that belong to rGO and small fraction of PHCN. The oxygen groups are beneficial to the adsorption of LiPSs to the PHCN/rGO composite that could restrain LiPSs intermediates [17]. Fig. S6 shows the XPS survey spectrum of PHCN/rGO/S, in which the peak of N 1s spectrum shifts to a higher binding energy, while the peak of C 1s spectrum is almost unchanged. The above results indicate that there are strong interactions between sulfur and the host material, and N plays an important role for chemisorption.

Fig. 4a shows the typical CV curves of the PHCN/rGO/S electrode at a scan rate of 0.1 mV s^{-1} with a voltage range of 1.7–2.8 V, in which two obvious reduction peaks are observed in the cathodic scan. The first reduction peak originates from the conversion of S_8 to soluble long-chain Li_2S_n ($4 \leq n \leq 8$), while the second reduction peak is attributed to long-chain Li_2S_n further reduced to insoluble short-chain $\text{Li}_2\text{S}_2/\text{Li}_2\text{S}$. In the anodic scan, two oxidation peaks are related to the reversible conversion from $\text{Li}_2\text{S}_2/\text{Li}_2\text{S}$ to Li_2S_n ($4 \leq n \leq 8$) and ultimately converted to S_8 . The CV curves of the succedent cycles almost overlap and become sharp after the activation of the first cycle. This phenomenon illustrates the decreased polarization and the good reversibility of reaction of the

Figure 4 Electrochemical performances. (a) CV profiles of PHCN/rGO/S at a scan rate of 0.1 mV s^{-1} . (b) Rate capability of PHCN/rGO/S. (c) Charge-discharge profiles of PHCN/rGO/S at 0.2 C. (d) Cycle performance of PHCN/rGO/S and rGO/S at 0.2 C. (e) Long cycle performance of PHCN/rGO/S at 0.5 C.

PHCN/rGO/S cathode [26,27]. Fig. S7 shows the CV profiles of rGO/S electrode at a scan rate of 0.1 mV s^{-1} . The rate performance of the PHCN/rGO/S cathode is shown in Fig. 4b, in which high reversible capacities of 930.9, 811.3, 723.3, 667.6, 620, and $495.1 \text{ mA h g}^{-1}$ are obtained at the different current densities of 0.1, 0.2, 0.5, 1, 2 and 5 C, respectively. After the initial 10 cycles at 0.1 C, the specific capacity of the PHCN/rGO/S cathode fades from 1,321.4 to $930.9 \text{ mA h g}^{-1}$, which might be caused by the partial dissolution of LiPSs that inevitably occurs at initial few cycles. There is only slight capacity degradation in the subsequent cycles. Furthermore, the Coulombic efficiency (about 100%) is stable at different current rates. When the current density is set back to 0.2 C, the specific capacity is similar to the original capacity of 0.2 C, indicating the good stability and reversibility of PHCN/rGO/S electrode [28]. Compared with PHCN/rGO/S cathode, rGO/S presents an inferior rate performance as shown in Fig. S8. As the current density increases, the capacity becomes low and unstable, which demonstrates that the addition of PHCN can improve the rate performance even at fast charge transfer condition. Fig. S9 shows the charge-discharge voltage profiles of PHCN/rGO/S and rGO/S cathodes at different current densities from 0.1 to 5 C with voltage ranging from 1.7 to 2.8 V. When the current densities are 0.1, 0.2, 0.5 and 1 C, the discharge voltage profiles of PHCN/rGO/S have two longer and flat plateaus compared with rGO/S, and when the current density increases to 2 and 5 C, the PHCN/rGO/S discharge voltage profiles still have two plateaus, while the rGO/S discharge voltage profiles have no obvious plateaus, suggesting a lower polarization and less kinetic barrier of PHCN/rGO/S cathode. In addition, the discharge-charge profiles of PHCN/rGO/S are well-maintained at different current densities, especially with a good performance at 5 C (495 mA h g^{-1}). Fig. 4c shows the corresponding charge-discharge profiles of PHCN/rGO/S electrode at 0.2 C. The typical high plateau could be attributed to the conversion of S_8 into long-chain Li_2S_n ($4 \leq n \leq 8$), and the low plateau is ascribed to the further reduction from Li_2S_n ($4 \leq n \leq 8$) to solid $\text{Li}_2\text{S}_2/\text{Li}_2\text{S}$ [29]. In the charge voltage profile, the plateau implies the reversible conversion of $\text{Li}_2\text{S}_2/\text{Li}_2\text{S}$ into sulfur. The voltage plateaus of 1st, 10th, 20th, 50th and 80th cycles remain similar and stable, demonstrating that PHCN can alleviate the shuttle-effect of LiPSs. Fig. 4d shows the cycling performances of PHCN/rGO/S and rGO/S cathodes at a rate of 0.2 C. The PHCN/rGO/S cathode delivers a high initial capacity of $1,061.1 \text{ mA h g}^{-1}$ and retains at 694 mA h g^{-1} after cycling with good capacity retention of

65.4%, while the rGO/S cathode delivers a higher initial capacity of $1,227.4 \text{ mA h g}^{-1}$ and a reversible specific discharge capacity of $572.2 \text{ mA h g}^{-1}$ after the cycles, corresponding to 46.6% capacity retention. All the capacities mentioned here are discharge capacities. The cycling stability of PHCN/rGO/S cathode is obviously better than that of rGO/S, because the nitrogen in PHCN could offer proper chemical adsorption ability of LiPSs [30–34]. The cathode at 0.5 C in Fig. 4e achieves a high capacity of $833.2 \text{ mA h g}^{-1}$, and $566.8 \text{ mA h g}^{-1}$ could be retained after 400 cycles with a good capacity retention (68%). The long cycle performance of rGO/S at 0.5 C displays in Fig. S10, where the cathode achieves the highest capacity of $851.4 \text{ mA h g}^{-1}$ and retains 341 mA h g^{-1} after 400 cycles with capacity retention (40.1%). The different electrochemical behaviors further prove that PHCN plays a key role in adsorbing LiPSs and reducing the side reactions. The introduction of PHCN can efficiently depress the dissolution and diffusion of LiPSs [35,36]. Therefore, PHCN is a promising sulfur host material for Li-S batteries to achieve long cycle performance.

Fig. 5a shows the long cycle performance of PHCN/rGO/S and rGO/S at 1 C. The PHCN/rGO/S cathode still delivers a specific capacity of 519 mA h g^{-1} with a good capacity retention rate (70.1%) after 400 cycles, while the rGO/S cathode only delivers a specific capacity of 363 mA h g^{-1} with a capacity retention of 44.1% after 400 cycles. The advantages of lower polarization and superior specific capacity of PHCN/rGO/S electrode further indicate that PHCN can adsorb the LiPSs and enhance the utilization efficiency of sulfur in electrodes. Fig. 5b shows the PHCN/rGO/S electrode with an areal sulfur loading of 3.1 mg cm^{-2} delivers a high initial capacity of $900.1 \text{ mA h g}^{-1}$ and 657 mA h g^{-1} could be retained after 110 cycles at 0.2 C (with good capacity retention of 73%). Fig. 5c shows the performance of the cathode with an areal sulfur loading of 4.3 mg cm^{-2} at 0.5 C, where the electrode still has good capacity retention of 69.9% after 200 cycles. The results of Fig. 5b and c demonstrate that the electrode has excellent cycling stability at high sulfur loading and PHCN is a promising host material for Li-S batteries to achieve excellent cycle performance. The lithium ion diffusion of electrodes, which is related to the redox kinetics behavior, was further studied. Fig. S11 shows the CV curves of the electrode detected at different rates, and the results of electrochemical impedance spectroscopy (EIS) analyses are shown in Fig. S12.

Color fading contrast experiments were carried out to further verify the adsorption capacity of PHCN/rGO for LiPSs. Fig. S13 shows three small glass bottles with 3 mL

Figure 5 Electrochemical performances. (a) Long cycle performance of PHCN/rGO/S and rGO/S at 1 C. (b) The electrochemical performances at 0.2 C with high sulfur mass loading of 3.1 mg cm^{-2} . (c) The electrochemical performances at 0.5 C with high sulfur mass loading of 4.3 mg cm^{-2} .

of Li_2S_6 solution (6 mol L^{-1} in DME solvent). The same amount (15 mg) of the PHCN/rGO and rGO was respectively added into two small glass bottles, and the third one only with Li_2S_6 solution served as a reference [37–39]. After 10 h, the decolorization of the solution after the addition of PHCN/rGO was clearly seen, indicating PHCN/rGO possessed good chemisorption capability for LiPSs.

CONCLUSIONS

In summary, PHCN architecture with abundant nitrogen active sites has been successfully designed and synthesized. PHCN/rGO as sulfur host for Li-S batteries through interaction with LiPSs to suppress the shuttle-effect has a stable long cycle life. The introduction of rGO increases the electric conductivity of electrode and then improves the utilization efficiency of sulfur, and the porous honeycomb-like structure is in favor of the infiltration and storage of electrolyte in the electrodes, and thus accommodating the volume expansion. The composite material as host for Li-S batteries shows excellent charge-discharge performance even with high areal sulfur loading. The above results demonstrate that PHCN is a promising host material for Li-S batteries.

Received 18 January 2019; accepted 13 April 2019;
published online 4 June 2019

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Acknowledgements This work was supported by the Chinese Academy of Sciences Large Apparatus United Fund (U1832187), the National Natural Science Foundation of China (21471091), the Natural Science Foundation of Shandong Province (ZR2019MEM030), Guangdong Province Science and Technology Plan Project for Public Welfare Fund and Ability Construction Project (2017A010104003), the Fundamental Research Funds of Shandong University (2018JC022), and Taishan Scholar Project of Shandong Province (ts201511004).

Author contributions Xu L and Bai X conceived the idea. Bai X designed and performed the experiments, analyzed the results and wrote the manuscript. All authors discussed the results and commented on the manuscript.

Conflict of interest The authors declare no conflict of interest.

Supplementary information Supporting data are available in the online version of the paper.

Xiaomeng Bai got her Bachelor degree from Kunming University of Science and Technology. Now, she is a master student under the supervision of Prof. Liqiang Xu at the School of Chemistry and Chemical Engineering, Shandong University, China. Her research interests mainly focus on the design and preparation of host materials for lithium-sulfur batteries.

Liqiang Xu received his BSc degree in chemistry from Liaocheng University in 2000. In 2005, he received his PhD degree in inorganic chemistry from the University of Science and Technology of China. Then he worked at Shandong University, and from May 2012 to May 2013, he worked as a Research Fellow in Nanyang Technology University in Singapore. He is currently a professor at the School of Chemistry and Chemical Engineering, Shandong University, China. His research interests focus on energy related

inorganic functional materials.

多孔蜂窝状 C_3N_4 /石墨烯作为高性能锂硫二次电池正极载硫材料

白晓梦¹, 王春省¹, 董才富^{1,2}, 李川川^{1,2}, 翟艳军³, 司卫卫¹, 徐立强^{1*}

摘要 锂硫(Li-S)二次电池因其具有较高的理论比容量和能量密度, 以及硫资源丰富, 成本低廉等优点而备受关注. 然而, 由于硫的利用率低以及多硫化物(LiPSs)的穿梭效应严重, 锂硫二次电池的实际应用仍然受到限制. 本论文通过硬模板法合成了多孔蜂窝状 C_3N_4 (PHCN), 合成的 C_3N_4 经载硫后, 所得正极PHCN/rGO/S在0.2 C的电流密度下有较高的初始放电比容量($1061.1 \text{ mA h g}^{-1}$)和良好的倍率性能(如在大电流密度5 C下, 其放电比容量为 $495.1 \text{ mA h g}^{-1}$); 在1 C的电流密度下循环400圈, 其容量仍保持在 519 mA h g^{-1} ; 即使在高面载量(4.3 mg cm^{-2})、0.5 C的电流密度下循环200圈, 其每圈的容量衰减率仅0.16%. 上述结果表明, 多孔蜂窝状 C_3N_4 /rGO是一种有发展潜力的锂硫电池正极载体材料.