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Sulfur-Impregnated, Sandwich-Type, Hybrid Carbon Nanosheets with Hierarchical Porous Structure for High-Performance Lithium-Sulfur Batteries

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Sandwich-type hybrid carbon nanosheets (SCNMM) consisting of graphene and micro/mesoporous carbon layer are fabricated via a double template method using graphene oxide as the shape-directing agent and SiO₂ nanoparticles as the mesoporous guide. The polypyrrole synthesized in situ on the graphene oxide sheets is used as a carbon precursor. The micro/ mesoporous strcutures of the SCNMM are created by a carbonization process followed by HF solution etching and KOH treatment. Sulfur is impregnated into the hybrid carbon nanosheets to generate S@SCNMM composites for the cathode materials in Li-S secondary batteries. The microstructures and electrochemical performance of the as-prepared samples are investigated in detail. The hybrid carbon nanosheets, which have a thickness of about 10–25 nm, high surface area of 1588 m² g⁻¹, and broad pore size distribution of 0.8-6.0 nm, are highly interconnected to form a 3D hierarchical structure. The S@SCNMM sample with the sulfur content of 74 wt% exhibits excellent electrochemical performance, including large reversible capacity, good cycling stability and coulombic efficiency, and good rate capability, which is believed to be due to the structure of hybrid carbon materials with hierarchical porous structure, which have large specific surface area and pore volume.

1. Introduction

The various emerging electric vehicles and advanced portable electronics demand the improvement of rechargeable battery technology to achieve higher energy density.^[1] Despite the numerous advantages, the energy density of lithium ion batteries is limited by the low capacity of cathode materials. In fact,

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it is difficult to improve the electrochemical capacity of transition metal intercalation compounds such as LiCoO₂ LiMn₂O₄ due to their capacity being close to the theoretical value. Therefore, rechargeable batteries beyond just those using lithium ions have been extensively investigated as alternatives. Among these, a lithium-sulfur (Li-S) battery is an attractive technology because it has a number of desirable properties. The Li-S battery has high theoretical energy density of 2500 W h kg⁻¹, calculated on the basis of the Li anode $(3860 \text{ mA h g}^{-1})$ and the S cathode (1675 mA h g^{-1}). In combination with the natural abundance, low cost, and environmental friendliness of elemental sulfur, the Li-S battery has become a great potential candidate for the next generation of high-energy rechargeable batteries.^[2]

However, the performance of the Li-S battery is limited by the insulating problem of sulfur and the dissolution and shuttling problem with polysulfides

in liquid electrolyte.^[3] To overcome this, considerable research effort have been placed on the optimization of the organic electrolyte,^[4] preparation of conductive polymer-sulfur composites,^[5] and fabrication of carbon-sulfur composites.^[6] In recent years, various types of carbon materials including carbon nanotubes,^[7] micro/mesoporous carbon,^[8] graphene or graphene oxide,^[9] carbon hollow sphere^[10] and carbon nanofibers^[11] have

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been proven to be effective and facile candidates for improving the conductivity of sulfur and minimizing the diffusion of lithium polysulfides due to their excellent conductivity, large specific surface area, and strong adsorption properties. The corresponding carbon-sulfur composites have been fabricated and achieved a specific capacity exceeding 1000 mA h g⁻¹ at 0.1C for the mass of sulfur. From the application point of view, a practical Li-S battery requires high reversible capacity, good rate capability, and excellent cycling stability. Although the Wan group has reported that smaller sulfur impregnated in CNT@MPC achieved excellent stability, relatively low sulfur loading leads to low capacity calculated based on the mass of the whole composite.^[12] A yolk-shell nanoarchitecture S-TiO₂ has been fabricated for long cycle Li-S cell, whereas microsized sulfur particles and TiO₂ with poor conductivity affect the rate capability to some extent.^[13] Very recently, monoclinic sulfur nanowires (NWs) encapsulated in aligned carbon nanotubes, followed by sputtering a Pt layer exhibit excellent performance, however, the complicated process for preparation and the high cost of noble metal Pt limit its practical application potential.^[14] Despite this progress, the design of novel carbon nanomaterials for enveloping a relatively high content of sulfur to improve its overall performance, including reversible capacity, rate capability, and cycling stability, is still a challenge in this field.

Much research has revealed that providing the rapid pathways for ionic and electronic transport can effectively improve the performance of energy storage and conversion systems such as Li ion batteries and supercapactors.^[15] As examples, S@graphene/single-walled nanotube hybrids exhibiting high rate capability with 860 mA h g⁻¹ at 1C and 650 mA h g⁻¹ at 5C, which is due to the fact that 3D graphene/SWCNTs conductive network can accelerate ionic and electronic transport, have been fabricated.^[16] Very recent report has also described carbon nanosheets derived from poly(benzoxazine-co-resol) for high-rate supercapacitors, attributed to the thin nanosheet facilitating the diffusion of electrolyte ions and transport of electrons.^[17] Recently, graphene with the 2D one-atom-thick structure has received extensive attention due to its unique properties.^[18] Since the reduction of graphene oxide (GO) has

been recognized as one of the most efficient methods for the large-scale production of graphene,^[19] GO with good solubility in solvents due to its abundant oxygen-containing functional groups, such as hydroxyl, epoxide, carbonyl, carboxyl, etc., provides many opportunities for the fabrication of graphene-based hybrid nanocomposites.^[20] To date, these graphene-based nanomaterials have been widely applied in electrochemical energy storage because of the electrochemical nature being greatly enhanced with their high specific surface area and highly conducting matrix.^[21]

Here, we present a rational design and fabrication of sandwich-type hybrid carbon nanosheets with abundant accessible micropores and mesopores (SCNMM) through a double template method using graphene oxides as the shape-directing agent and SiO₂ nanoparticles (NPs) as the



mesopore guides. The polypyrrole synthesized in situ on the graphene oxide sheets was used as carbon precursor. The micro/mesoporous strcutures were created by a carbonization process followed by HF solution etching and KOH treatment. Then S@SCNMM was prepared by sulfur infiltration via capillary force at 155°C, as shown in Scheme 1. Such hybrid carbon materials with hierarchical pores (micro and mesopores), large surface and pore volume, thin nanosheets, and sandwich-type structure were used to encapsulate sulfur with several specific goals in mind: 1) to minimize lithium polysulfide dissolution and shuttling in the electrolyte; 2) to preserve fast transport of lithium ions to the coated sulfur; 3) to facilitate good transport of electrons from the poorly conducting sulfur; and 4) to maximize the amount of sulfur loading. The as-prepared SCNMM with the sulfur content of 74 wt% used as the cathode materials in Li-S cell exhibits excellent electrochemical performance including high reversible capacity (1370 mA h g⁻¹ at 0.5 C after 1st cycle), good cycling stability (860 mA h g⁻¹ at 1 C after 100 cycles), and good rate capability (510 mA h g^{-1} at 10 C).

2. Results and Discussion

The nanostructure of the typical as-prepared hybrid carbon nanosheets (SCNMM) was first investigated using scanning electron microscopy (SEM) and transmission electron microscopy (TEM). As shown in Figure 1a, the hybrid carbon nanosheets with embedded SiO₂ nanoparticles are easily observed. After removal of SiO₂ nanoparticles and treatment with KOH, the nanosheets with abundant micropores and mesopores were generated and these hybrid carbon nanosheets with thickness of 10-25 nm are highly interconnected to form 3D hierarchical structure (Figure 1b,c). Compared with SCNMM, fewer pores of the corresponding carbon nanosheets without using the SiO₂ nanoparticles as templates (APG) or KOH treatment (SPG) can be observed clearly (Supporting Information Figure S1,S2). The results from these SEM observations are further confirmed by the analysis of the pore structure. From the nitrogen adsorption/desorption isotherms (Supporting Information Figure S3),



Scheme 1. Schematic illustration of the procedure for preparing S@SCNMM.

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Figure 1. a) SEM image of SiO₂ NPs inserted-hybrid carbon nanosheet; b,c) SEM images at different magnification of SCNMM. d) Cross-sectional high-resolution TEM (HRTEM) image of SCNMM. The cross-sectional HRTEM specimen was prepared by slicing nanosheets embedded in epoxy with an ultramicrotome.

SCNMM displays a Brunauer-Emmett-Teller (BET) surface area of 1558 m² g⁻¹ (micropore area 1058 m² g⁻¹), and a total pore volume of 1.1 cm³ g⁻¹ (a relative pressure $p/p_0 = 0.99$) with a broad porous size distribution of 0.8-6 nm by density functional theory (DFT) method, while SPG and APG show the much smaller specific surface area and pore volume. Carbon spheres with inserted SiO₂ nanoparticles from polyprrole spheres prepared under identical conditions in the absence of GO can be obtained (Supporting Information Figure S4), implying the shape and structure directing effect of GO during the formation of the SCNMM. This also indirectly verifies that graphene was planted in the hybrid carbon nanosheets to form a sandwichtype structure. To provide a direct proof for the sandwich-type structure, the sample was observed using TEM after slicing the SCNMM embedded in epoxide resin with an ultramicrotome. As can be seen from Figure 1d and Supporting Information Figure S5, the cross sections of hybrid carbon nanosheet display straight or bent shapes of the nanosheets with the thickness between 10 nm and 25 nm, which is agreement with SEM observations. More closely, the interplanar spacing is calculated to be about 0.36 nm and matches well with the (002) crystalline planes of graphene, confirming that the graphene percolates in the central position of hybrid carbon nanosheets. X-ray photoelectron spectroscopy (XPS) reveals that the SCNMM is only composed of C, N, and O elements (Supporting Information Figure S6). Scanning transmission electron microscopy with energy dispersive X-ray spectroscopy (STEM-EDS) elemental mapping also indicates the homogeneous spreading of N element throughout these carbon nanosheets (Supporting Information Figure S7).

From the SEM and TEM observations of S@SCNMM (Figure 2a-c), no agglomerated sulfur particles or crystalline sulfur were found on the surface of carbon nanosheets. However, X-ray photoelectron spectroscopy (XPS) reveals the presence of element sulfur in the nanosheet (Supporting Information Figure S8), and STEM-EDS elemental mapping verifies the homogeneous spreading of sulfur throughout the hybrid carbon nanosheets (Figure 2d). The possible reason for this is that the sulfur is well dispersed on the hierarchical pores or the surface of carbon nanosheets because of the excellent wettability between carbon and sulfur, which was also confirmed by powder X-ray diffraction (XRD) results. The powder XRD patterns of a physical mixture of SCNMM and S, S@SCNMM-83%, and S@SCNMM-74% samples accordingly with 83, 74 wt% sulfur loading proved by thermogravimetric analysis (TGA; Supporting Information Figure S9) are presented in Figure 3. Elemental sulfur generally exists in a very stable orthorhombic crystalline structure (JCPDS: 08-0247), which can be easily found in the physical mixture of SCNMM and S. After heating at 155 °C for 8 h, the characteristic diffraction peaks attrib-

uting to orthorhombic crystalline sulfur and the obvious broad peak of carbon around at 23° were observed in S@SCNMM-83% sample. After further heating for 8 h at 160 °C under Ar flow rate of 10 mL min⁻¹, no evident peak for crystalline sulfur was observed, suggesting that the sulfur is amorphous, that the sulfur is highly dispersed on the hierarchical pores, or that the surface of the hybrid carbon substrates are unable to crystallize.^[12] This result agrees with the TEM observations.

These as-prepared samples were assembled into Li-S batteries as cathode materials and their electrochemical Li storage capabilities were evaluated. The cyclic voltammogram (CV) measurements were first obtained for S@SCNMM-74% and are shown in Figure 4a. During the first cathodic scan, the peak at around 2.36 V can be assigned to the opening ring reduction of cyclic S₈ to higher-order Li polysulfides (Li₂S_n, 4 < n < 8).^[3-11,13,14] The peak at about 2.1 V likely corresponds to the further reduction of these polysulfides to lower-order Li polysulfides (such as Li₂S₂, Li₂S).^[3-14] In the subsequent anodic scan, the two expected oxidation peaks overlap and form one broad peak at about 2.4 V, which may be due to high over-potential for conversion of Li2S to lithium polysulfide.[8h] As cycling proceeds, this broad anodic peak splits into two peaks at 2.37 and 2.30 V. The oxidation peak at 2.37 V becomes less smaller, while another new peak at 2.30 V, which is related to the formation of Li_2S_n (n > 2), increases in intensity. In addition, the cathodic peak current increases slightly in intensity. After





Figure 2. a) SEM image of S@SCNMM; b,c) TEM images at different magnifications of S@ SCNMM. d) STEM-EDS elemental maps of S@SCNMM.

the second cycle, both the peak currents and CV peak positions undergo very small changes, suggesting relatively good capacity retention. These CV results indicate that these carbon nanosheets can help to prevent sulfur from dissolving into the electrolyte due to its hierarchical pores and large surface.

Figure 4b depicts the second cycle discharge/charge typical voltage profiles of as-prepared electrodes at the 0.5C rate ($1C = 1675 \text{ mA h g}^{-1}$). The voltage profile of the S@SCNMM-74% electrode demonstrates a high reversible capacity around 1370 mA h g⁻¹ sulfur, which is 81% of the theoretical capacity



Figure 3. XRD patterns of S@SCNMM-74%, S@SCNMM-83%, a physical mixture of S and SCNMM.



of 1675 mA h g^{-1} (full reduction of S to Li_2S). To the best of our knowledge, this value is higher than most previous reports,[5-13,16] impling that the current electrode design facilitates electronic and ionic transport so that most of sulfur in a reversible reaction with Li ions. The discharge curve of S@ SCNMM electrode shows two voltage plateaus at 2.2-2.35 V and 2-2.1 V, which is in agreement with the peaks in the CV and also well documented in the literature.^[4–11] Compared to S@SCNMM, S@APG and S@SPG show the lower reversible capacity, relating to the smaller specific area surface and pore volume of these hybrid carbon materials. This result indicates that the high reversible capacity of S@SCNMM enabled synergistic effect from two strategies including the treatment with KOH and using SiO₂ NPs as mesoporous templates. Furthermore, the lower value of voltage differences between the charging and discharging plateaus also suggests the battery based on the S@ SCNMM has better reversibility than that of S@APG and S@SPG. In addition, a physical mixture of sulfur and SCNMM (S/CNMM) used as cathode material exhibits a capacity of 804 mA h g⁻¹, which is much lower than the S@SCNMM. The result implies that the sulfur has been inserted into the SCNMM in the composite S@SCNMM.

The rate capability, an important challenging and key aspect in Li-S battery operations, was also tested under the galvanostatic mode at various discharge-charge rates. The corresponding capacities of S@SCNMM-74% electrode calculated display in Figure 4c. A reversible capacity of ≈ 1200 mA h g⁻¹ was achieved at a current rate of 1C. The capacity decreased gradually with the increased current rate. It is noticeable that a high discharge capacity of ca.860 mA h g⁻¹ was obtained at 5C, and a retention capacity of about 70% can still be preserved when the C-rate is increased 10-fold from 0.5 C to 5C. In addition, even at a rate of 10C, a substantial capacity of 510 mA h g^{-1} was retained during the discharge time of ≈ 110 s. These results suggest that the S@SCNMM nanocomposite has an excellent rate capability, which is clearly superior to those of most other Li-S batteries reported.^[5-11,13,16] This is believed to be attributed to the nanostructured electrode design, including the thin nanosheet with abundant hierarchical pores and sandwich-type with graphene inserted into hybrid carbon, which facilitate electronic transport and ionic diffusion and thus overcome the electronically insulating nature of sulfur.

Because the cycling performance is also critical for Li-S battery applications, an endurance test of S@SCNMM-74% electrode was carried out under the galvanostatic mode at 1C (Figure 4d). After a 100 cycle stability test, the capacity of sulfur is preserved and is as high as 860 mA h g⁻¹ at 1C. In addition, after the 5th cycle discharge, S@SCNMM-74% achieved a capacity of 1155 mA h g⁻¹ and maintained a capacity retention of 75% at the end of 100 cycles, corresponding to a small



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Figure 4. a) Cyclic voltammograms of S@SCNMM/Li cell at a scan rate of 0.02 mV s⁻¹. b) Second charge/discharge potential profiles of S@SCNMM/Li cell, S@APG/Li cell, S@SPG/Li and S/SCNMM/Li cell at 0.5C. c) Reversible capacity vs. current density (rate capability) of the S@SCNMM/Li cell. d) Cycle performance at a constant current rate of 1C and the corresponding coulombic efficiency of S@SCNMM/Li cell.

capacity decay of about 0.25% per cycle. During the repeated charge/discharge of the electrode, the outward migration of dissolved polysulfides and the loss of active material were greatly suppressed by SCNMM with this structural design, as evident from the high coulombic efficiency (Figure 4d).

To investigate the effect of the sulfur loading, discharge charge measurements on three samples, S@SCNMM-74%, S@SCNMM-83%, and S@SCNMM-89%, were conducted at 0.5C, and the corresponding second cycle discharge/charge typical voltage profiles are shown in **Figure 5**. A reversible capacity decrease is observed from around 1370 mA h g^{-1} to 1120 mA h g^{-1} with increasing the sulfur loading from 74 to 83%. This sug-



Figure 5. The second charge/discharge potential profiles of S@SCNMM/ Li cell with different sulfur contents of 74%, 83% and 89% at 0.5C.

gests that a large sulfur loading on the same carbon nanosheets is unfavorable for an improvement on the active-sulfur utilization due to the highly insulating nature of sulfur, which is in agreement with previous reports.^[8c,8g],Because it is important to calculate the energy density of a battery based on the overall mass of the composite for considering its practical applications,^[8h] it is impressive that the S@SCNMM-89% exhibits a reversible capacity of 965 mA h g⁻¹. Even based on the mass of the entire S@SCNMM composite, the capacity still reaches as high as 858 mA h g⁻¹ at a current rate of 0.72 A g⁻¹ (0.43C).

The electrochemical impedance spectroscopy (EIS) measurements were performed to investigate the internal resistance of the Li-S battery and the corresponding Nyquist plots are shown in Figure 6. All the impedance plots for the fully discharged states are composed of a semicircle in the high-frequency region relating to the charge transfer resistance of the cathode and a sloping, straight line in the low-frequency region corresponding to the Li-ion diffusion within the cathode. The intersection of the semicircle on the real axis at high frequency demonstrates that the electrode/electrolyte interfacial resistance remains almost unchanged, which indicates that the electrode processes do not induce the increase in the resistance of interfacial passivation films. For the charge-transfer resistance (R_{ct}) , it is apparent that the value after the third cycle discharge is the smallest. With the cycle processes, the R_{ct} values gradually increase from 35Ω to 45Ω . This may be due to the relatively poor electrical contact caused by the slow accumulation of Li₂S in the sulfur cathode, which is consistent with the slight decrease in the capacity with cycle processing. After 100th cycles, the R_{ct} value is still preserved at a low value of 45 Ω ,

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Figure 6. EIS spectra of the S@SCNMM/Li cell at different cycles after full discharging to 1.0 V at 1C. The inset is the enlarged plots of the high frequency regions.

also further confirming that the hybrid carbon nanosheets with hierarchical pores and sandwich-type structure facilitate electronic and charge transfer.

3. Conclusion

In summary, we have demonstrated the rational design and fabrication of thin hybrid carbon nanosheets with a sandwich structure, high surface area of 1558 m² g⁻¹, and abundant of micropores and mesopores with sizes ranging from 0.8 to 6.0 nm. Such hybrid carbon materials with unique structure are used as a conducting matrix to encapsulate sulfur as Li-S batteries and have substantial influence on the electrochemical performance of the batteries. Thin nanosheets combined with nanosized micropores and mesopores are favorable for the accessibility of the rapid diffusion of organic electrolyte ions and the effective penetration of sulfur. The nanosized microporous and mesoporous hybrid carbon with large pore volumes and specific surface areas can minimize lithium polysulfide dissolution and shuttling in the electrolyte, in addition to maximizing the amount of sulfur loading. Meanwhile, graphene percolated in the central position of thin hybrid carbon nanosheets can facilitate the transport of electrons during the processes of charging and discharging. The synergy effects from such a design of carbon material leads to its high capacity (1370 mA h g⁻¹ at 0.5 C after 1st cycle), good cycle stability (860 mA h g⁻¹ at 1 C after 100 cycles), and good rate capability (510 mA h g⁻¹ at 10 C). This comprehensive performance is superior to that of most other Li-S batteries that have been reported (Supporting Information Table S1). In addition to Li-S battery applications, thin hybrid carbon nanosheets with such a design should have a large spectrum of applications, including other lithium batteries, supercapacitors, or electrochemical sensors, in which efficient ionic and electronic transport is critical.

4. Experimental Section

Synthesis of SiO₂ Nanoparticles (NPs) and Graphene Oxide: SiO₂ NPs were synthesized by a modified process according to previous reports.^[22]



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L-Lysine (0.3 g) was dissolved in 300 mL deionized water. Tetraethyl orthosilicate (TEOS, 20 g) was then added. This mixture was stirred at 60 °C for 20 h to generate the SiO₂ NPs colloidal solution. Graphene oxide (GO) was prepared from graphite flakes by the modified Hummers method.^[23] A homogeneous graphene oxide aqueous dispersion (1.0 mg mL⁻¹) was sonicated for 2 h before use.

Preparation of SCNMM: The sample SCNMM was synthesized using a double template method with graphene oxide as the shapedirecting agent and SiO₂ NPs as the mesopores guides. The polypyrrole synthesized in situ on the graphene oxide sheets was used as carbon precursor. The micro/mesoporous strcuture was created by carbonization process followed by HF solutions etching and KOH treatment. Briefly, 100 mL GO aqueous dispersion, 100 mL SiO₂ NPs colloidal solution and 4.0 mL pyrrole were added into 150 mL deionized water sequentially. After being treated with ultrasound for 0.5 h and stirred in the ice bath for another 2 h, ammonium peroxydisulfate (12.0 g) in 50 mL deionized water was rapidly poured into the above solution. The mixture was allowed to stir in the ice bath for 12 h. Finally, the precipitate was washed with distilled water and ethanol, and dried in a vacuum oven at 60 °C for 12 h to yield a black powder. Then, the powder was heated to 850 °C for 2 h under N₂ atmosphere, followed by the treatment with 20% HF solution to generate SPG. The mixture of SPG (0.4 g) and KOH (1.0 g) in ethanol/water co-solvent was stirred under 70 °C. After evaporation of the solvents, the mixture was transfered to the nickel crucible and was heated untiled to 800 °C and kept for 2 h, with a heating rate of 5 °C min⁻¹ and a Ar flow rate of 10 mL min⁻¹, to generate the SCNMM.

A Typical Procedure to Prepare S@SCNMM: Sulfur (Aldrich, with a purity of >99.995%) and SCNMM were mixed according to a mass ratio of 1:10 to yield a black mixture. The mixture was then sealed in a glass container and heated at 155 °C for 10 h. In order to further decrease the S content or removal of agglomerated sulfur particles, some of the as-synthesized samples (S@SCNMM) were heat treated under N₂ flow rate of 10 mL min⁻¹ at 160 °C for different periods. In addition, S@SPG and S@APG were also prepared with the same method and the sulfur contents were determined by TGA (Supporting Information Figure S10). The sulfur contents in the as-prepared composites were also calculated by the mass change before and after the formation of the composite, which almost agrees with TG analysis.

Characterization: The content of sulfur was confirmed using a TG/ DTA thermogravimetric analyzer (Diamond PE) under an N₂ atmosphere at a heating rate of 10 °C min⁻¹ from room temperature to 500 °C, with a flow rate of 80 mL min⁻¹. SEM images were obtained with a Nova NanoSEM 200 scanning electron microscope (FEI, Inc.). TEM, HRTEM images, and STEM-EDS elemental mapping were recorded with a JEOL-2100 instrument. Powder XRD was conducted on a Bruker D8 Advance X-ray diffractometer using CuK α radiation (λ = 0.15418 nm) at a scanning rate of 4° min⁻¹ in the 2 θ range from 10° to 80°. XPS measurements were carried out with an ultrahigh vacuum setup, equipped with a monochromatic Al K α X-ray source and a high resolution Thermo ESCALAB 250 analyzer. Specific surface area, pore volume and pore size distribution were determined by the BET method on a Micromeritics ASAP 2020 instrument.

Electrochemical Measurements: To prepare the working electrodes, as-prepared composite was mixed with acetylene black and poly(vinyl difluoride) (PVDF) in a weight ratio of 80:10:10 with *N*-methyl pyrrolidone (NMP) as a dispersant. The paste was compressed into a thin piece with a roller, followed by cutting into a film disk of 14 mm in diameter and approximately 1.5–2.0 mg in weight. The as-obtained film disk was dried in a vacuum oven at 50 °C for 12 h. CR2025-type coin cells were fabricated by sandwiching a porous polypropylene separator between the film disk of S@SCNMM and a lithium metal foil in a high-purity argonfilled glove box. 1 wt% anhydrous lithium nitrate (analytical grade) and 1 M LiN(CF₃SO₂)₂ (LiTFSI) in a mixed solvent of 1.3-dioxolane (DOL) and dimethyl ether (DME) at a volume ratio of 1:1 were used as the electrolyte, purchased from Fosai New Material Co., Ltd (Suzhou).

Galvanostatic charge/discharge tests were conducted to evaluate the electrochemical capacity and cycle stability of the electrodes on the



basis of the active sulfur at current densities of 0.5C, 1C, 2C, 3C, 5C, 7C, 10C ($1C = 1675 \text{ mA h g}^{-1}$) from 1.0 to 3.0V using a Neware-CT3008W instrument (Shenzhen NEWARE China). CV data were recorded on a CHI660e electrochemical workstation (Shanghai Chenhua) between 1.7 and 2.8 V to characterize the redox behavior and the kinetic reversibility of the cell. The ac impedance was measured with fresh cells at the open circuit potential. This was also carried out using a CHI 660e electrochemical workstation. The ac amplitude was 5 mV and the frequency ranged from 100 kHz to 0.01 Hz.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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