Supporting Information

Sodium polyacrylate-derived porous carbon nanosheets for high-

performance lithium-sulfur batteries

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Experimental Part

Synthesis of HPCNS. HPCNS was synthesized following a simple solid phase calcination process. Firstly, 10 g PAAS powders (Mw = 5000,000~7000,000, Aladdin Reagent, Co. Ltd, Shanghai) was carbonized at 450 °C for 2 h in a tube furnace under N₂ atmosphere. Then the product was mixed with 1.2 g potassium hydroxide by ball-milling for 0.5 h, and calcined at 750 °C for 2 h with a temperature-increasing rate of 2 °C min⁻¹. The final product was washed with hydrochloric acid and distilled water three times and then dried.

Synthesis of HPCNS/S. The prepared HPCNS and elemental sulfur with a mass ratio of 3:7 were dispersed into 4 ml carbon bisulfide and magnetically stirred at 60 °C until completely dried. The resulting mixtures were then ground into fine powders, and latterly transferred to the tube furnace and heated at 155 °C for 12 h in N₂ atmosphere. Afterward, the HPCNS/S hybrid containing 70 wt% sulfur was obtained.

Synthesis of HPCNS/S@RGO. The graphene oxide (GO) was synthesized by oxidizing graphite powder using a modified Hummers' method. The synthesis process of RGO coated carbon-sulfur composite is as follow. First, 100 mg HPCNS/S was dispersed in 20 ml distilled water and 4 ml ethyl alcohol. After sonicating for 0.5 h, 100 mg sodium thiosulfate was added to the solution with sonication and magnetic stirring for 1 h. Then, 10 mg GO was dispersed in 40 ml water by sonication for 1 h. The HPCNS/S and sodium thiosulfate mixture was subsequently added dropwise to the above GO solution. Afterward, 2 ml diluted hydrochloric acid (1 M) was added to the mixture accompanied by sonication and stirring. Finally, the HPCNS/S@RGO hybrid was obtained through centrifugation, washing and drying treatments.

Materials characterization. The samples were characterized by X-ray diffraction (XRD) on a diffractometer (X'Pert PRO MPD 1.6 kV, Cu K α radiation), and the diffraction patterns were recorded in a 2 θ range of 10–80°. The morphology and structure characterization were measured with a Hitachi SU8020 field-emission scanning electron microscopy (SEM) and transmission electron microscopy at a voltage of 200 kV with Oxford Inca (TEM, JEM-2100F). The elemental mapping of a selective area was tested by energy-dispersive X-ray spectroscopy (EDS) method. HORIBA Jobin Yvon HR Evolution Raman spectrometer was used to record the Raman spectra of samples with a 633 nm laser excitation. N₂ adsorption and desorption isotherm curves were carried out via a Quantachrome Instruments (ASIQ). Surface bonding information was measured by ESCALAB250Xi X-ray photoelectron spectroscopy (XPS). Thermogravimetric analysis (TGA) was conducted with a STA449F3 thermal analyzer.

Electrochemical measurement. The working electrodes were assembled via standard 2032-type coin cells in an Ar-filled glove box. The cathode material's slurry was obtained by the mixture of as-prepared samples with Super P and 5 wt% polyvinylidene difluoride (PVDF) (weight ratio of 8:1:1). The slurry was coated on an aluminum foil and dried at 60 °C overnight in vacuum. The electrode was further cut into discs of Φ 12 mm with the active materials of 0.4–0.8 mg cm⁻². A Celgard 2400 membrane was used as the separator. The electrolyte consisting of 1.0 M lithium bis-(trifluoromethanesulfonyl)imide (LiTFSI) in the mixed solvents of 1,2-dimethoxyethane and 1,3-DOL (with 1 wt% LiNO₃) was employed. The cyclic performance of cells was measured on a Newar CT-3008 battery tested station in

galvanostatic strategy within the voltage range of 1.6-2.8 V at 0.5 C (837.5 mAh/g). Cyclic voltammetry (CV) curves were recorded between 1.6 and 2.8 V at 0.2 mV s⁻¹. The electrochemical impedance spectroscopy (EIS) measurements were carried out in the frequency range of 10^5 to 0.01 Hz with an amplitude of 5 mV.



Fig. S1 TG/DSC curves of PAAS under N_2 atmosphere with the heating rate of 10 °C/min. The inset shows the digital photo of PAAS powder. It can be seen that the PAAS has three stages during the calcination process including the early stage of residual water and small molecule loss, the decomposition of the precursor in the medium term, and the later stage of carbon graphitization start from ~450 °C. Thus, in the present study, the carbonization temperature of PAAS is set at 450 °C. The carbonization yield of PAAS at 450 °C for 2 h is ~65%. The yield from the carbonized char to porous carbon (HPCNS) is 10%.



Fig. S2 SEM images of HPCNS at different magnifications.



Fig. S3 An SEM image showing the morphology of HPCNS/S.



Fig. S4 TEM image of HPCNS/S@RGO.



Fig. S5 A TEM image of HPCNS.



Fig. S6 TG analysis of HPCNS/S and HPSNS/S@RGO in air atmosphere from room temperature to 750 °C (10 °C /min). The weight losses of the composites below 260 °C and above 490 °C can be ascribed mainly to the evaporation of sulfur and decomposition of HPCNS, respectively. However, in the temperature range of 260 °C to 340 °C, there is a gentle slope for HPCNS/S@RGO that might be attributed to the loss of both sulfur and RGO. Therefore, the content of RGO is estimated to be 5.1 wt% (100%-64.6%-30.3%=5.1 wt%) in the final hybrid.



Fig. S7 The TG curve of HPCNS/S@RGO under N_2 atmosphere. It can be divided into three stages according to the different sulfur loss rates. Stage I, below 300 °C, the weight loss of sulfur is ~4 wt%; Stage II, 300-400 °C, the weight loss of sulfur is 40 wt%; and Stage III, above 400 °C, the weight loss of sulfur is 20.6 wt%.



Fig. S8 (a) The Nitrogen adsorption and desorption isotherms of the HPCNS/S, HPCNS/S@RGO hybrid and, (b) the pore size distribution curves. The corresponding surface area for HPCNS/S, HPCNS/S@RGO hybrids are 73 and 34 m² g⁻¹, respectively.



Fig. S9 The survey XPS spectrum of HPCNS/S@RGO composite.



Fig. S10 Cycling performance and Coulombic efficiency of HPCNS/S@RGO cathode at the rate of 1 C (the capacity is based on the mass of sulfur). the initial capacity is 1150 mAh/g, which reduces to 982 mAh/g at the third cycle. In the extended cycles, the capacity gradually reduces to 512 mAh/g at the 300th cycle, indicating a capacity decay of ca. 0.16% per cycle at the last 297 cycles.



Fig. S11 Cycling performance of HPCNS/S@RGO cathode at 1 C rate with enhanced sulfur loading of 1.34 mg/cm².



Fig. S12 Nyquist plots of HPCNS/S@RGO and HPCNS/S before (a) and after (b) 200 cycles.

Table S1 The electrochemical properties of HPCNS/S@RGO comparing with other reported S-carbon structures.

Active material	S content	Capacity (mAh/g) @1st cycle	Cycling stability	Ref.
HPCNS/S@RGO	64.6 wt%	1262 mAh/g @0.5C	690 mAh/g @200th cycle, 0.5C	This work
S-Bamboo carbon fibers	60 wt%	1014 mAh/g @0.2C	447 mAh/g @200th cycle, 0.2C	1
S-PAN derived carbon nanofiber mat	50-65 wt%	1042 mAh/g @0.1C	634 mAh/g @20th cycle, 0.1C	2
S-PVP/PAN/PMMA carbonized nanofibers	56 wt%	1175 mAh/g @0.2C	623 mAh/g @300th cycle, 0.2C	3
S-Porous organic polymer derived carbon matrix	60 wt%	~1340 mAh/g @1C	631 mAh/g @300th cycle, 1C	4
S-Coconut shell derived carbon	60 wt%	~1520 mAh/g @1C	854 mAh/g @200th cycle, 1C	5
S-PAN/PMMA carbonized nanofibers	57 wt%	1296 mAh/g @0.2C	635 mAh/g @300th cycle, 0.2C	6
S-Plane tree bark derived carbon	48 wt%	1159 mAh/g @0.2 A/g	410 mAh/g @60th cycle, 0.2 A/g	7
S-Corncob derived carbon	69 wt%	1317 mAh/g @0.1C	411 mAh/g @50th cycle, 0.1C	8
S-Gelatin pyrolyzed carbon	53.3 wt%	1209 mAh/g @1C	600 mAh/g @200th cycle, 1C	9

References:

- X. Zhang, Y. Zhong, X. Xia, Y. Xia, D. Wang, C. a. Zhou, W. Tang, X. Wang, J. B. Wu and J. Tu, ACS Appl. Mater. Interfaces, 2018, 10, 13598-13605.
- 2. C. Dillard, S.-H. Chung, A. Singh, A. Manthiram and V. Kalra, *Mater. Today Energy*, 2018, 9, 336-344.
- 3. J. S. Lee, J. Jun, J. Jang and A. Manthiram, Small, 2017, 13, 1602984.
- Z. Xiao, D. Kong, J. Liang, B. Wang, R. Iqbal, Q.-H. Yang and L. Zhi, *Carbon*, 2017, 116, 633-639.
- 5. Z.-H. Chen, X.-L. Du, J.-B. He, F. Li, Y. Wang, Y.-L. Li, B. Li and S. Xin, ACS Appl. Mater. Interfaces, 2017, 9, 33855-33862.
- J. S. Lee, W. Kim, J. Jang and A. Manthiram, *Adv. Energy Mater.*, 2017, 7, 1601943.
- 7. J. Xu, K. Zhou, F. Chen, W. Chen, X. Wei, X.-W. Liu and J. Liu, ACS Sustain

Chem. Eng., 2016, 4, 666-670.

- J. Guo, J. Zhang, F. Jiang, S. Zhao, Q. Su and G. Du, *Electrochim. Acta*, 2015, 176, 853-860.
- Y. Qu, Z. Zhang, X. Zhang, G. Ren, Y. Lai, Y. Liu and J. Li, *Carbon*, 2015, 84, 399-408.