Supporting Information

Diatomite-derived N-doped carbon aerogel loading 98% sulfur for enhancement of Li-S battery performance

Zhiyi Jiang [a, b, c], Shilin Zhang [d], Xiongbo Dong [a, b, c], Zonglin Xu [d], Muhammad Tariq Sarwar [a, b, c], Chenyu Yan [a, b, c], Caihong Yang [a, b, c], Aidong Tang [a, b, c,], Huming Yang [a, b, c]**

[a] *Engineering Research Center of Nano-Geomaterials of Ministry of Education, China University of Geosciences, Wuhan 430074, China*

[b] *Faculty of Materials Science and Chemistry, China University of Geosciences, Wuhan 430074, China*

[c] *Key Laboratory of Functional Geomaterials in China Nonmetallic Minerals Industry, China University of Geosciences, Wuhan 430074, China*

[d] *College of Chemistry and Chemical Engineering, Central South University, Changsha 410083, China*

[e] *Hunan Key Lab of Mineral Materials and Application, School of Minerals Processing and Bioengineering, Central South University, Changsha 410083, China*

[*] Correspondence: hmyang@csu.edu.cn (H. Yang), and adtang@csu.edu.cn; tangaidong@cug.edu.cn (A. Tang)

Experimental section

Materials

The purified diatomite was prepared via the scrubbing method as reported in a previous study[1]. The elemental composition of purified diatomite was listed in Table S10, Supporting Information. Dopamine hydrochloride ($C_8H_{11}NO_2 \cdot HCl$, 98%), Cobalt acetate tetrahydrate ($C_4H_6CoO_4 \cdot 4H_2O$, 99.5%), sodium chloride (NaCl), sodium nitrate ($NaNO_3$), sodium sulphate (Na_2SO_4), sodium bicarbonate ($NaHCO_3$), sodium dihydrogen phosphate (NaH_2PO_4) , hydrochloric acid (HCl, 37%), hydrofluoric acid (HF, 40 wt%), and other chemicals were purchased from China National Pharmaceutical Group Co. Ltd. (Sinopharm). All chemicals were used as received without further purification. Deionized water (18 MΩ · cm) was used throughout the experiments.

Prepare the C/DT

The precursor of diatomite is treated by the reported typical flotation method, and the flotation agent is sodium hexametaphosphate. Use a typical coating method reported: 4 g diatomite is added into 500ml deionized water, dispersed by ultrasound for 30 min, 0.05 g cobalt acetate tetrahydrate is added and dissolved in the suspension to accelerate PDA to form a uniform membrane on the surface of diatomite template. Then, 0.6g dopamine hydrochloride was added, 5 ml Tris buffer (1mm, pH = 8.5) was added to the suspension, stirred overnight at 0 ℃, dopamine was polymerized and coated on the surface of diatomite. The suspension is filtered, fully cleaned with ethanol and deionized water, and then dried overnight at 60 ° C under vacuum. The resulting black powder was annealed in N_2 atmosphere at 900 ° C for 2h to obtain T-C_{PDA}/DT. Then, T-C_{PDA}/DT was immersed in HF (20%) solution for 24 hours, the diatomite template was etched, and the residual metal was cleaned with hydrochloric acid. The suspension is centrifuged and washed with deionized water and ethanol for many times until the pH of the supernatant is about 7.Then T-C_{PDA}/DT was dried overnight under vacuum at 60 ℃ to obtain C_{PDA}/DT.

Compose the C_{PDA}/DT@S

C_{PDA}/DT@S composites were prepared by melt diffusion method, C_{PDA}/DT and sulfur powder were ground at the mass ratio of 1:49, **C_{PDA} and sulfur powder were ground and mixed evenly at the mass ratio of 3:7 and 1:49**, and heated at 155 ℃ in a closed container for 12 hours. Then the pure sulfur cathode is used as a comparison.

Synthesis of cathode plate

According to the ratio of 9:1, C_{PDA}/DT@S, PVDF, C_{PDA}/DT@S, PVDF respectively and grind and mix evenly. Appropriate amount of NMP was added and stirred at room temperature for 12 h to get uniform positive slurry. Then, the cathode slurry was coated on aluminum foil (coated carbon) by an auto-coating machine and dried at 90°C for 12h, and then it was cut into discs with a 12 mm diameter for assembling the cell battery. Adjust different coating thickness, and the sulfur mass of the cathode plate is about 1.5 mg.

Synthesis Solution phase Li_2S_6

Synthesis of Li_2S_6 solution and visualized adsorption measurements: Typically, the Li_2S_6 solution (3 mmol L^{-1}) for visualized adsorption measurements was prepared by mixing sulfur and Li_2S at a molar ratio of 5:1 in 1.0 M LiTFSI in DOL and DME (1:1 by volume) at 80 °C for 24 h.

Assemble the CR2025 battery

In the glove box in Ar atmosphere, the CR2025 button half battery was assembled with 1M LiTFSI DME:DOL=1:1V%, 1% $LiNO_3$ solution as electrolyte, lithium sheet as anode and polypropylene microporous Celgard 2400 as separator. The ratio of electrolyte to sulfur is 10 μL: 1 mg.

In addition place two pieces of cathode materials on both sides of the separator, drip the Li_2S_6 electrolyte. Then, obtain a CR2025 symmetrical battery for catalytic performance test

Characterization

X-ray diffraction: To characterize the sample phase structure and ingredients of prepared products, X-ray diffraction (XRD) (Bruker D8 Advance) was performed with Cu Kα (λ=0.15406 nm) radiation at a scanning rate of 5°/s.

Scanning electron microscope: The field-emission scanning electron microscope (FESEM, JEOL JSM 6700F) was used to characterize the morphology of the synthesized material in Zhongkebaice China.

AC-HADDF-STEM: The scanning transmission electron microscopy ： HADDF-STEM (FEI Titan3 G2 60-300) was used to characterize the morphology of the synthesized material in Tsinghua University.

XPS test：The chemical and electronic states were recorded by X-ray photoelectron spectroscopy (XPS, Thermo Scientific K-Alpha$^+$) by Shiyanjia Lab (www.shiyanjia.com)

Electrochemical measurements ：Cells were tested for charge and discharge on the LAND CT2001A battery test system, the test voltage range was 1.7 V - 2.8 V. Cyclic voltammetry (CV) and electrochemical

impedance spectroscopy (EIS) tests were performed on the Interface1000T-10140 electrochemical working station form Gmary. The CV voltage window of the scan was 1.7 - 2.8 V, and the scan rate was 0.1 mV s^{-1}. The EIS was performed by applying an ac amplitude of 5 mV in the frequency range from 0.01 - 10^5.

Measurement of mass loss of cathode containing sulfur: The METTLER TOLEDO X56 PPM thermogravimetric microbalance was used to carry out sulfur loss on the positive electrode material heated at 155℃, and the average value was obtained after 10 groups were measured on average.

Supplementary Results

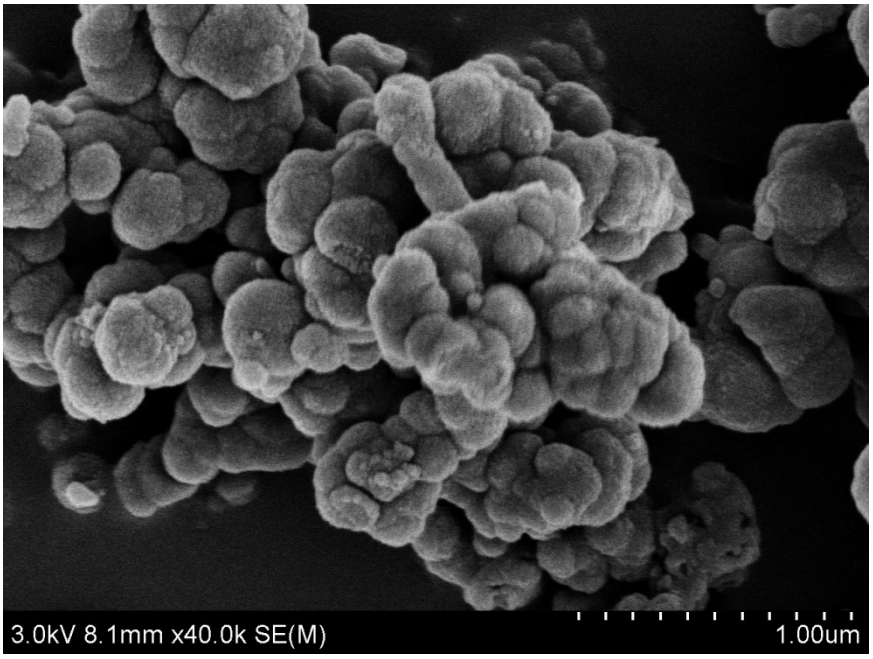

Fig. S1 SEM of the sample of only PDA calcined

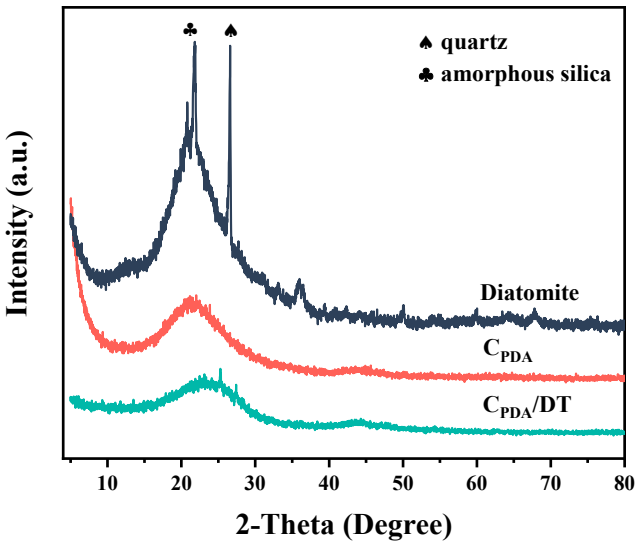

Fig. S2 XRD patterns of diatomite, C_{PDA}/DT and C_{PDA}.

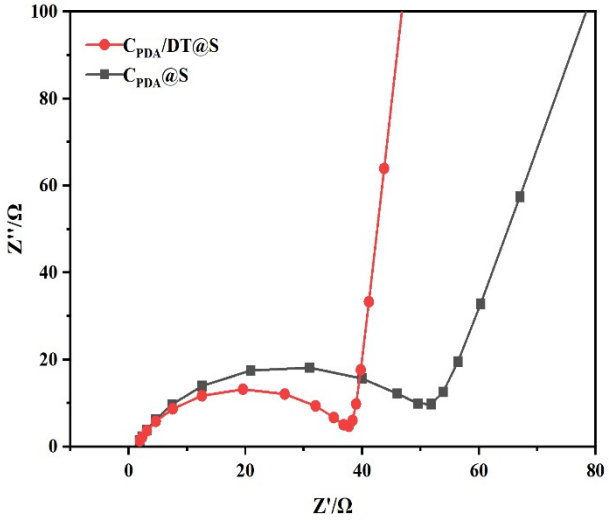

Fig. S3 EIS diagram after fitting of C_{PDA}/DT and C_{PDA}.

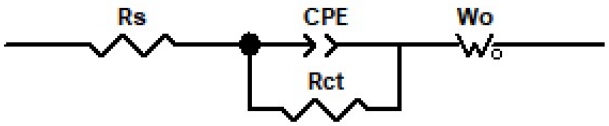

Table. S1 Detailed value of EIS fitting resistance

Sample	*Rs（Ω）*	*Rct（Ω）*	*Wo-R*	*Wo-T*	*Wo-P*
C_{PDA}/DT@S cathode	1.431	35.22	741.3	3.014	0.84
C_{PDA}@S cathode	1.328	49.47	1140	3.37	0.79

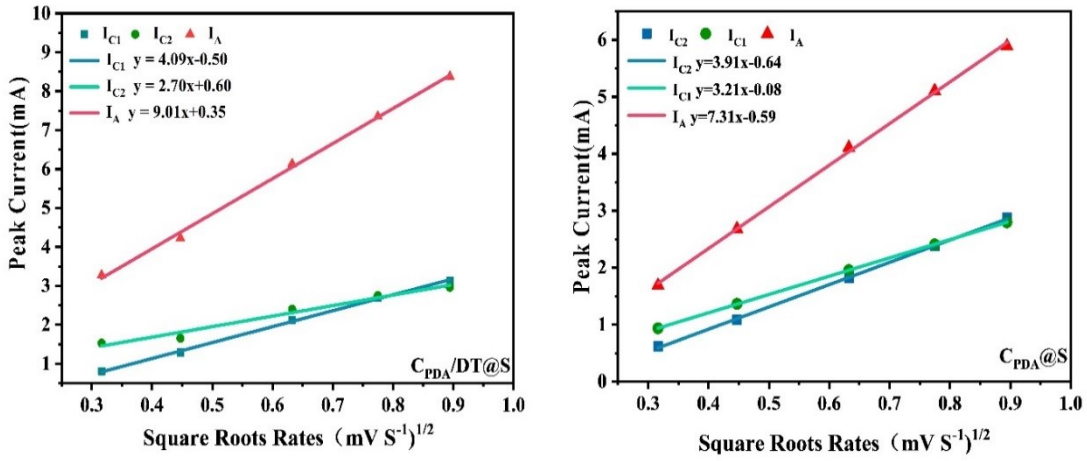

Fig. S4 CV curves peak current of I_{C2} (cathodic reduction process, $S_8 \rightarrow Li_2S_x$, $3 \leqslant x \leqslant 8$), I_{C1} (cathodic reduction process, $Li_2S_x \rightarrow Li_2S_2/Li_2S$) for three cathodes with multiple effects and I_A (anodic oxidation process, $Li_2S_2/Li_2S \rightarrow S_8+Li$)

$$I_p = (2.69 \times 10^5) n^{1.5} \cdot A \cdot D_{Li^+}^{0.5} \cdot C_{Li^+} \cdot v^{0.5}$$

The rapid lithium ions diffusion is favorable for sulfur conversion kinetics, which can be obtained by testing the CV curve at different scan rates. According to Randles-Sevcik equation. The $\boldsymbol{I_P}$ is the peak current and $\boldsymbol{D_{Li}}^+$ is the lithium-ion diffusion coefficient. $\boldsymbol{V}$ is the voltage scanning rate. In the lithium-sulfur battery system, because $\boldsymbol{n}$ **(**charge transfer number), $\boldsymbol{A}$ **(**active electrode area), and $\boldsymbol{C_{Li}}^+$ (lithium-ion concentration) are certain values, the ratio of $\boldsymbol{I_P}$ to $\boldsymbol{V^{1/2}}$ can be used to approximately reflect the size of $\boldsymbol{D_{Li}}^+$.

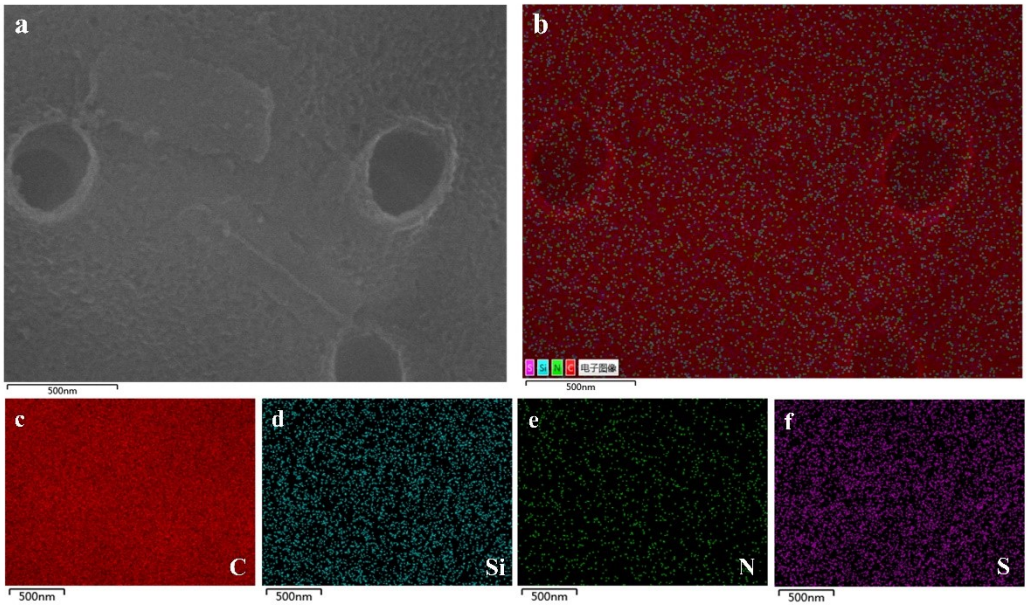

Fig S5 (a) SEM images of CPAD/DT@S, and Elemental mapping images of (b) CPAD/DT@S for elements (c) C, (d) Si, (e) N,and (f) S respectively

Fig. S6 Digital photos of C_{PDA}/DT and SuperP respectively

With the same mass of carbon materials, C_{PDA}/DT has a larger and more objective volume, which also proves its lightweight characteristics. It can effectively reduce the carbon mass bi in the cathode material of lithium sulfur battery.

Table S2 Comparison of carbon content and cycling performance of cathode

Sulfur content in the composite (wt%)	*Specific capacity (mAh/g-sulfur)*	*Ref.*
25	1520@ 0.5C	Ref. 1
37	1224@ 0.5C	Ref. 1
42	1058@ 0.5C	Ref. 1
42	1227@ 0.1C	Ref. 1
47	1150@ 0.5C	Ref. 2
47	950@ 3C	Ref. 2
51	1050@ 0.5C	Ref. 3
51	652@ 3C	Ref. 3
53	770@5C	Ref. 4
53	1152@1C	Ref. 4
55	1068@ 0.1C	Ref. 1
60	1418@0.5C	Ref. 5
60	630@ 4C	Ref. 5
64	1200@ 0.5C	Ref. 6
64	713@ 10C	Ref. 6
80	1440@ 0.2C	Ref. 7
80	800@ 6C	Ref. 7
98	**846@0.1C**	**★This Work**
98	**702@0.2C**	**★This Work**
98	**703@0.5C**	**★This Work**
98	**601@1C**	**★This Work**
98	**535@2C**	**★This Work**
98	**452@4C**	**★This Work**

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