Supporting Information

N and S co-doped porous carbon spheres prepared using L-cysteine as a dual function agent for high-performance lithium-sulfur batteries

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2. Experimental Section

2.1 Materials preparation

Nitrogen and sulfur co-doped porous carbon spheres (NS-PCSs) were synthesized by a hydrothermal method along with KOH activation. In a typical experiment, 3.6g of D-glucose and 0.72g of L-Cysteine were dissolved in 40 mL of double distilled water. Then the mixed solution was filled into a Teflon lined, stainless steel autoclave (45 ml) and placed in a pre-heated furnace at 180 ^{o}C for 24 h. The obtained solid product was collected by filtration and washed three times with deionized water and alcohol before drying in vacuum oven at 70 ^{o}C overnight. Then activation was performed by impregnating the products with KOH with a weight ratio of 1:4 in a crucible under N_2 flow at a heating rate of 5 ^{o}C/min to 800 ^{o}C for 1h. The prepared NS-PCS (40 mg) was mixed with 60 mg of sulfur (Dk Nano technology, Beijing) and then this mixture was heated at 155 ^{o}C for 12h in a sealed vessel filled with argon protection to obtain NS-PCS/S hybrid. Porous carbon sphere (PCS) without adding L-Cysteine was also prepared as a reference sample. The porous carbon sphere-sulfur (PCS/S) hybrid with the same sulfur content compared with NS-PCS/S hybrid was also prepared for comparison.

Nitrogen doped porous carbon spheres (N-PCS) were prepared as follows: 200 μL pyrrole was added into 200 mL deioned water and this mixture was treated with ultrosonication for 1 hour to form a uniform solution. Then 18 mL concentrated hydrochloric acid was added, stirred in an ice bath for 2h, then freshly prepared 0.2g ammonium persulfate in 10 mL deioned water was dropped in, and it was stirred in the ice bath for 24h. The polypyrrole was obtained by filtration and washed with deioned water and ethanol three times and then dried at 60 ^{o}C for 24h. The polypyrrole was chemically activated by heating a PPy-KOH mixture (KOH/ PPy at a weight ratio of 4) under N_2 protection in 800 ^{o}C for 1h. The activated sample was then washed several times with HCl (0.1M) to remove any inorganic salts and then washed with deioned water unter natural pH. Finally, the N-PCS was obtained after dried and the N-PCS/S hybrid was prepared applied the same method compared with NS-PCS/S hybrid.

2.2 Materials characterization

All as-prepared materials were characterized by wide angle X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS, PHI5802), scanning electron microscopy (SEM, HITACHI-4800), transmission electron microscopy (TEM, FEI TECNAIG2 F30)) and thermogravimetric analysis (TG-DSC, Netzsch, STA449). Elemental analysis was carried out on an elementary Vario EL cube elemental analyzer. Raman spectra was recorded with a Lab RAM HR800 (Horiba) using 632 nm incident radiation. The surface area and pore volume of the samples were determined by nitrogen gas adsorption at 77 K with an automated adsorption apparatus (Micrometrics, ASAP 2020). The surface area was determined from the Brunauer-Emmett-Teller (BET) equation and pore size distribution (PSD) of NS-PCS was calculated based on adsorption-desorption using the density functional theory (DFT).

2.3 Electrochemical measurements

The carbon/sulfur electrode was prepared by mixing the carbon/sulfur hybrids, carbon black, and polyvinylidene difluoride (PVDF) with a weight ratio of 80:10:10 in NMP solvent to form a slurry. Then the slurry was coated onto a carbon-coated aluminum foil using a doctor blade and dried at 60 ^{o}C under vacuum for 24h. The coin cell (CR2032) with a metallic lithium anode was assembled in glove-box (Mbraun) to evaluate the electrochemical performance of the as-obtained sample. The mass loading of sulfur in the obtained cathode is ~1.2 mg cm^{-2} and microporous polypropylene sheet (Celgard 2500) was used as the separator. The electrolyte was 1M bis(trifluoromethane) sulfonamide lithium salt (LiTFSI) dissolved in a mixture of 1,2-dioxolane(DOL) and dimethoxymethane(DME) (1:1 by volume) with adding 1 wt% $LiNO_3$. Cyclic voltammetry (CV) experiment was carried out over the potential range of 1.5-3V (versus Li metal) at a scanning rate of 0.1 mV s^{-1} using VMP3 electrochemical workstation (Bio Logic Science Instruments). The galvanostatic charge/discharge performance test and the rate capability at different C-rates were performed using a Land 2001A cell test system (Wuhan, China) at 298K.

II. Supplementary Figures:

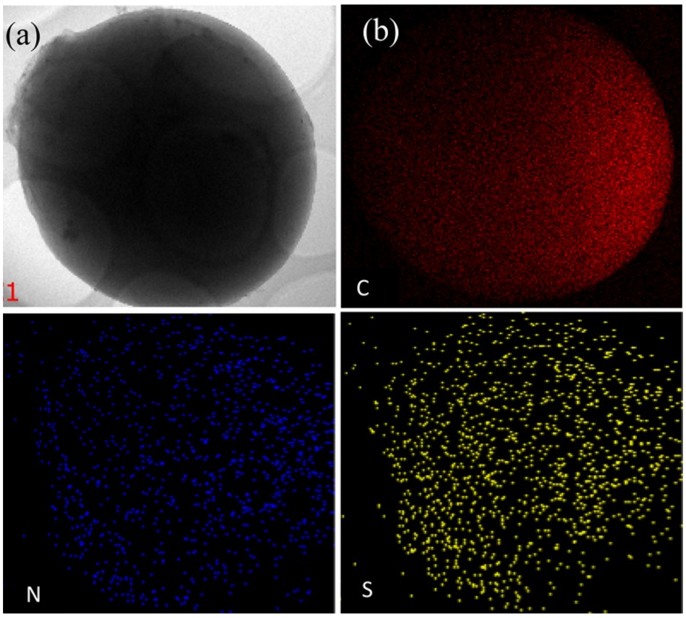

Fig. S1 STEM image of NS-PCS (a) and the corresponding elemental mapping of (b) carbon, (c) nitrogen and (d) sulfur.

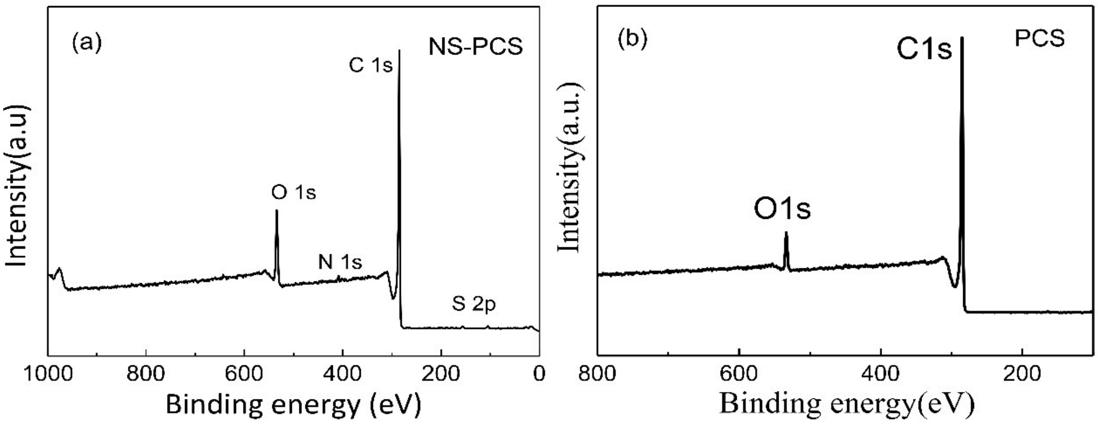

Fig. S2 XPS survey spectrum of the (a) NS-PCS and (b) PCS.

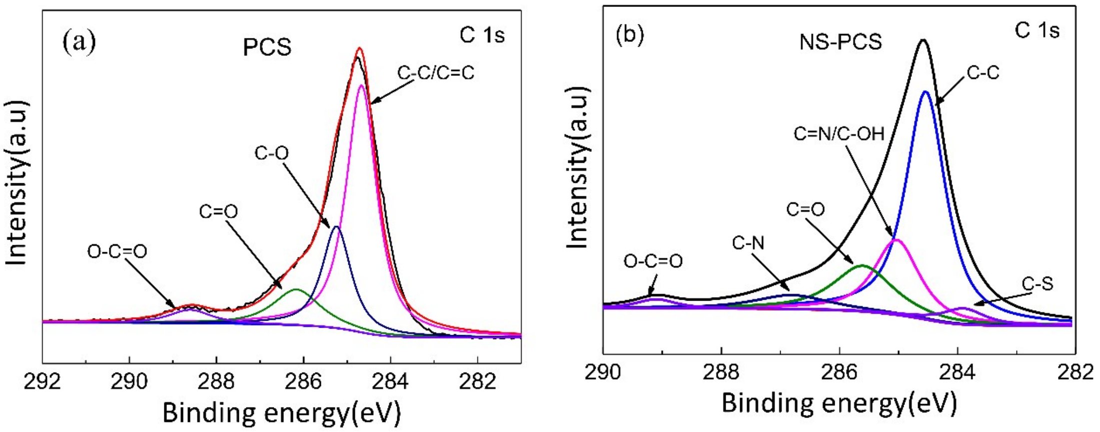

Fig. S3 The C 1s XPS spectrum of the (a) PCS and (b) NS-PCS.

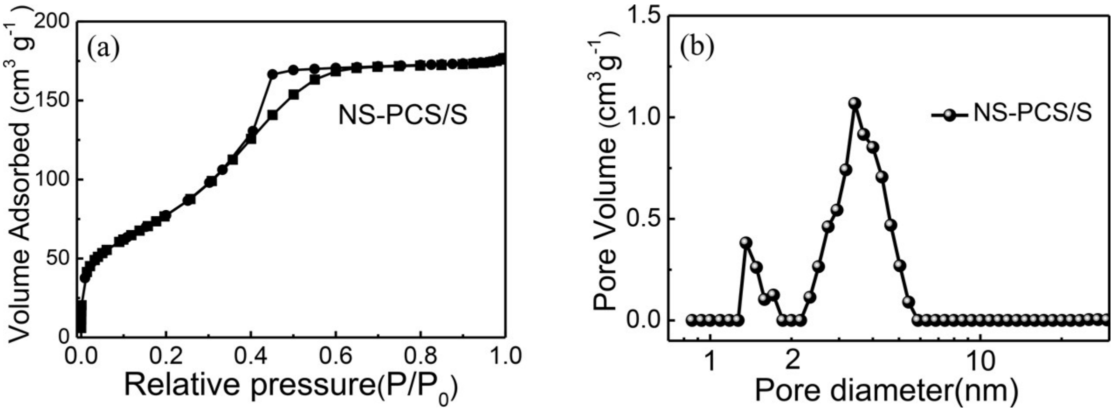

Fig. S4 Nitrogen adsorption-desorption isotherms (a) and pore size distribution (b) of NS-PCS/S hybrid.

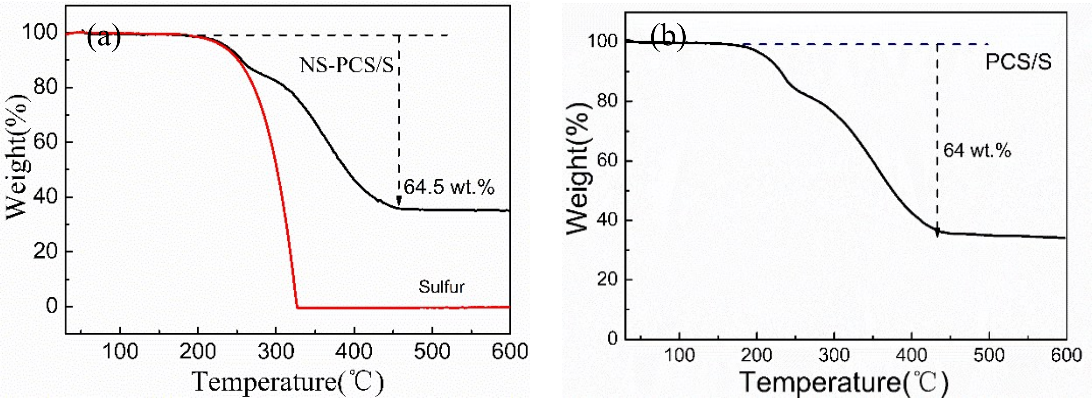

Fig. S5 TG curves of the (a) NS-PCS/S and (b) PCS/S hybrid.

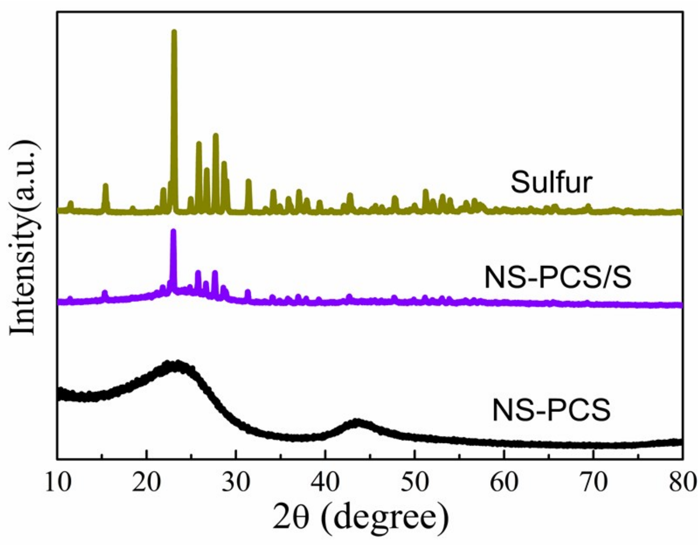

Fig. S6 XRD patterns of the sulfur, NS-PCS and NS-PCS/S hybrid.

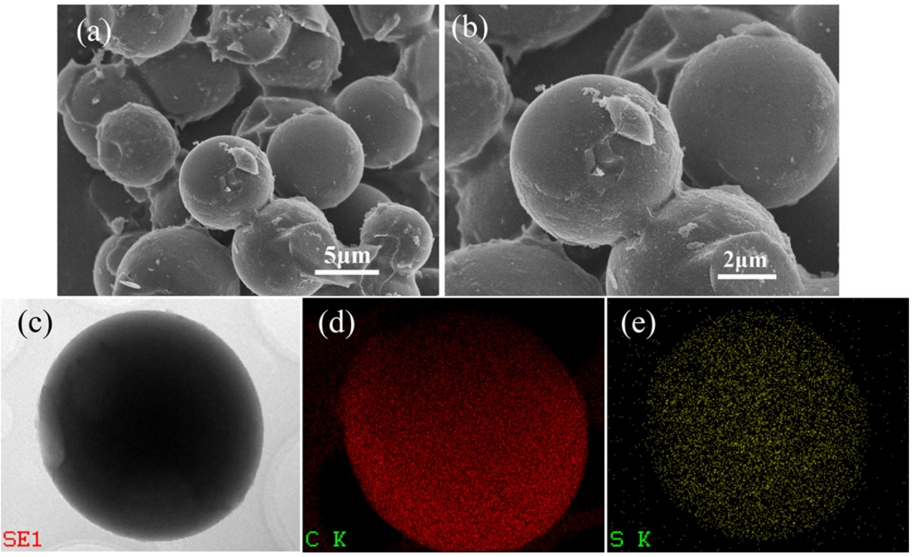

Fig. S7 SEM (a and b) and STEM images (c) of the NS-PCS/S hybrid and corresponding elemental mappings of (d) carbon and (e) sulfur.

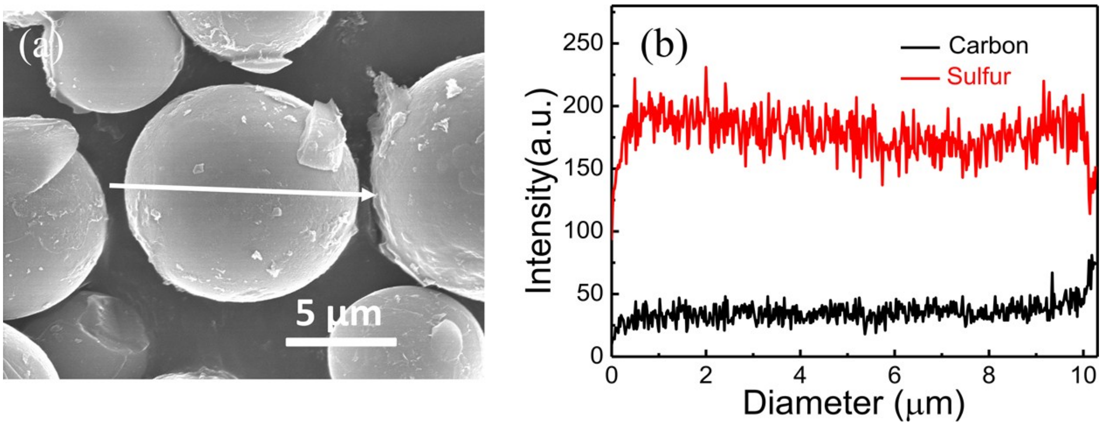

Fig. S8 (a) SEM image of NS-PCS/S hybrid; (b) the linear element distribution of sulfur and carbon along the arrow line of (a).

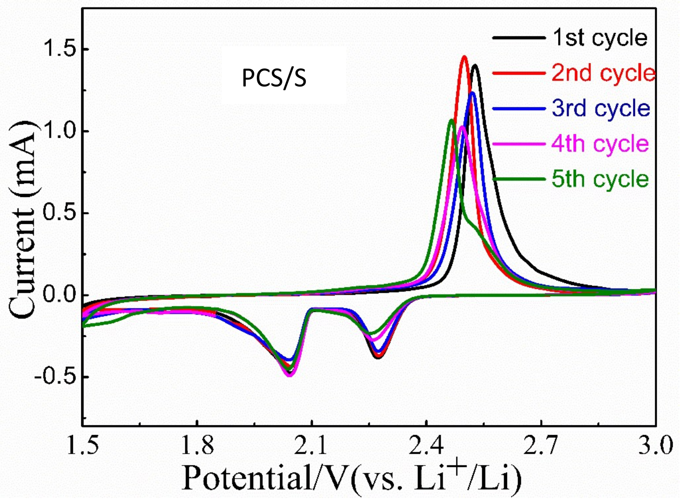

Fig. S9 CV curves of PCS/S hybrid at a sweep rate of 0.1 mV s^{-1}.

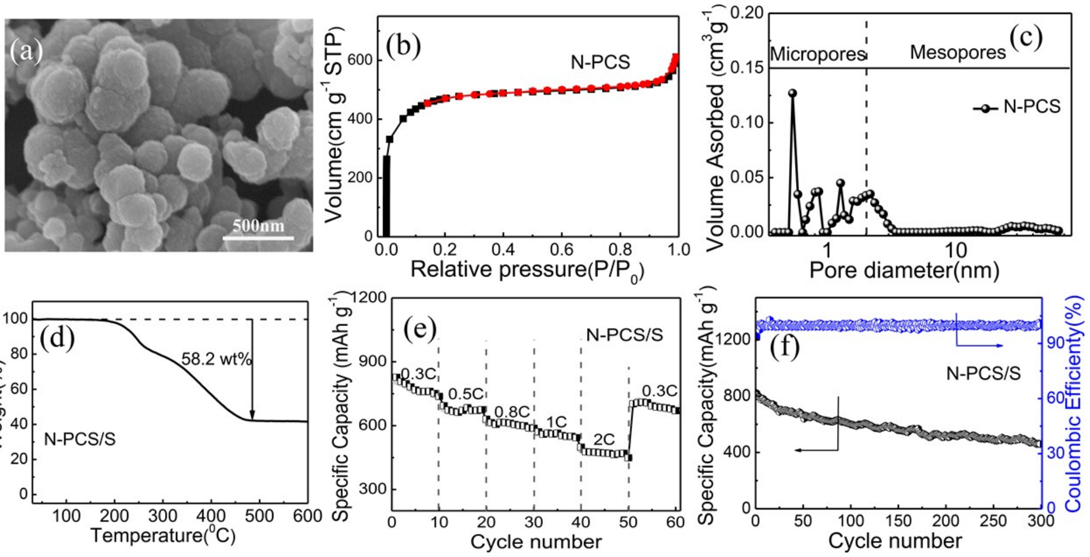

Fig. S10 (a) SEM image and (b) nitrogen adsorption-desorption isotherms and (c) pore-size distributions of the N-PCSs. (d) TG curve of the N-PCS/S hybrid, (e) the rate performance and (d) cycle performance (at 0.3C) of N-PCS/S hybrid.

The specific surface area and pore volume of N-PCSs is 1742.5 m^2 g^{-1} and 0.93 cm^3 g^{-1}, respectively. Moreover, the sulfur content in the N-PCS/S hybrid is about 58.2 wt%, according to the result of TG analysis.

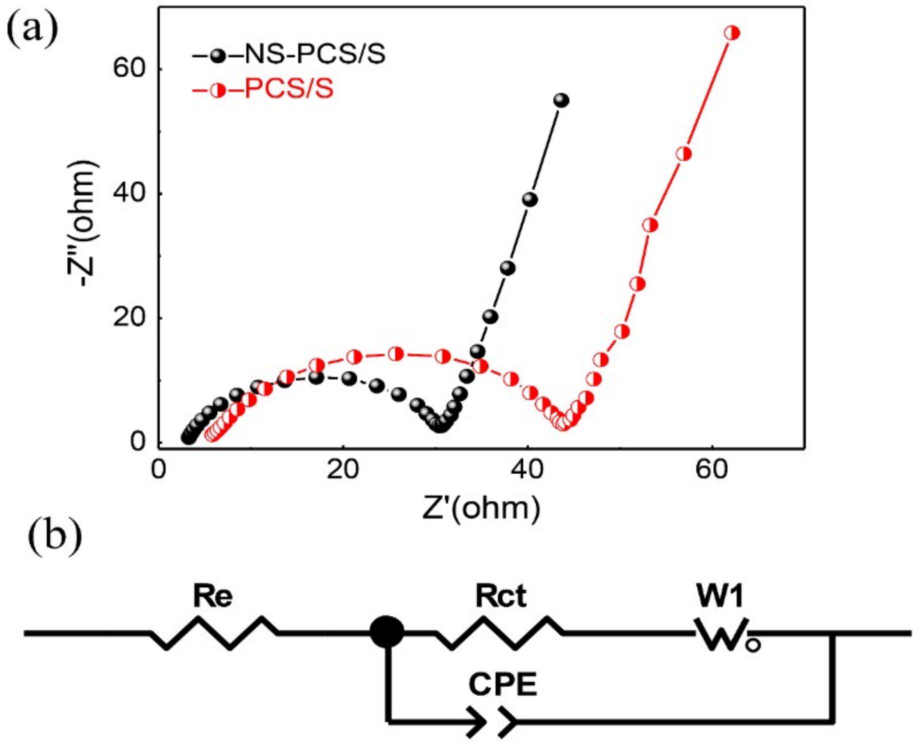

Fig. S11 (a) Nyquist plots and (b) the equivalent circuit used to fit the impedance data of the NS-PCS/S and PCS/S hybrids.