Single-walled carbon nanohorns with a unique horn-shaped structure as a scaffold for lithium-sulfur batteries

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1. Experimental section

Preparation of the C-S composite: The C-S composite was prepared via a facile melt-diffusion strategy. SWCNHs used here were produced by a simple arc-discharge method.[1] Typically, carbon material (SWCNHs or MWCNTs) and sulfur with weight ratios of 1:4 were dissolved in 5 ml CS_2 to form a homogeneous solution under ultrasonic radiation. Subsequently, CS_2 was allowed to completely evaporate from the solution under magnetic stir to obtain the C-S composite. The final C-S product was placed in a crucible, then heated to 155 ℃ and kept at that temperature for 24 h in an argon environment. The composite was denoted as SWCNHs-S or MWCNTs-S.

2. Material characterization

The structure and morphology of composites were characterized by X-ray diffraction (XRD, RIGAKU SCXmini), scanning electron microscope (SEM, JSM-6700F), and transmission electron microscope (TEM, Tecnai G2 F20). Thermal gravimetric analysis (TGA, NETZSCH STA449 C) tests were measured from 30 to 600 °C at a heating rate of 10 K min^{-1} in a N_2 environment to evaluate the weight content of sulfur in the composites. The nitrogen adsorption/desorption isotherms and the pore size distribution were determined by Brunauer-Emmett-Teller (BET) measurements using an ASAP-2020 surface area analyzer. The pore size distribution was calculated from the desorption data using the Barret-Joyner-Halenda (BJH) modal.

3. Electrochemical test

The electrochemical tests were performed via CR2025 coin-type test cells which were fabricated in an argon-filled glove box using lithium metal as the counter electrode and a Celgard 2300 membrane. The cathode slurry was prepared by mixing 80 wt% C-S composite, 10 wt% conducting carbon (ketjen black, KB), and 10 wt% polyvinylidene fluoride (PVDF) in N-methyl-2-prrolidone (NMP) solvent dispersant. Positive electrodes were produced by pasting the slurry on Ni foam and then drying at 60 °C for 12 h. The electrolyte solution was 1 M lithium bis(trifluoromethylsulfonyl)imide (LiTFSI) in a mixed solvent of 1,2-dimethoxy ethane (DME) and 1,3-dioxolane (DOL) (a volume ratio of 1 : 1) with 0.25 M lithium nitrate addictive ($LiNO_3$). The charge-discharge performance of the cells was tested with LAND CT-2001A instrument. The cut-off potentials were 1.9 and 2.7 V at room temperature. Cyclic voltammetry (CV) tests were performed on a CHI660C Electrochemical Workstation. The cut-off potentials were 1.8 and 2.8 V at room temperature. The specific capacity of the electrodes was calculated on the mass of elemental sulfur.

Table 1 Textural characteristic of samples

Samples	BET surface area (m^2 g^{-1})	Total pore volume (cm^3 g^{-1})
SWCNHs	203	0.48
SWCNHs-S	0.94	0.01

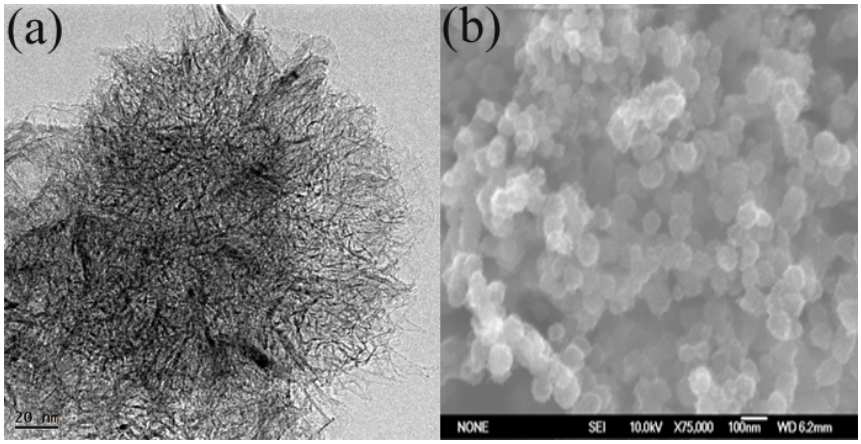

Fig. S1 (a) HRTEM and (b) SEM images of the SWCNHs.

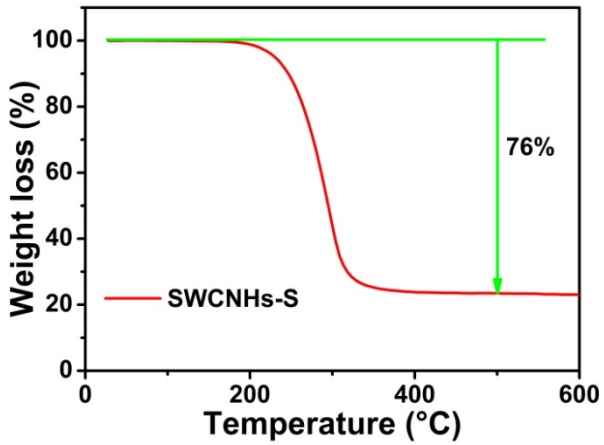

Fig. S2 Thermal analysis curve of the SWCNHs-S.

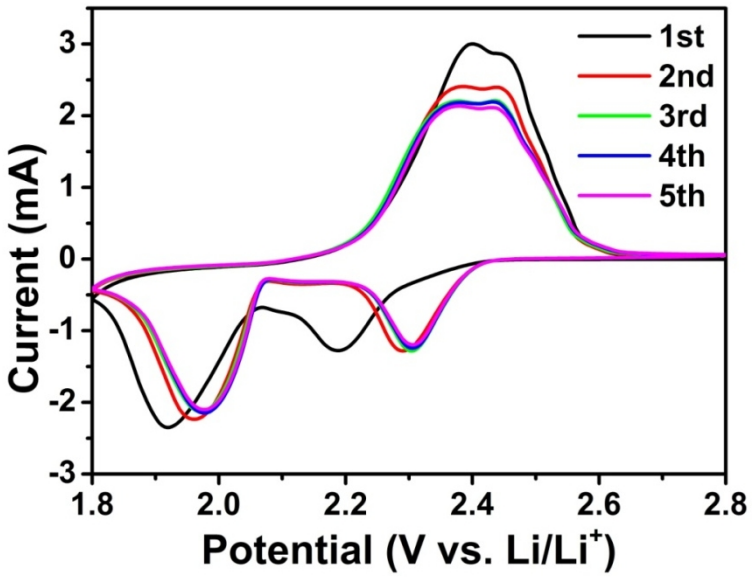

Fig. S3 Cyclic voltammogram of the SWCNHs-S electrode.

As seen in the Fig. S3, the reduction peak shifts to higher potentials and the oxidation peak shifts to lower potentials upon cycling, indicating improved reversibility of the cell. [2] Meanwhile, their peak intensity and integrated area mostly became coincidence in the successive cycles, validating the good cycling performance of the SWCNHs-S electrode.

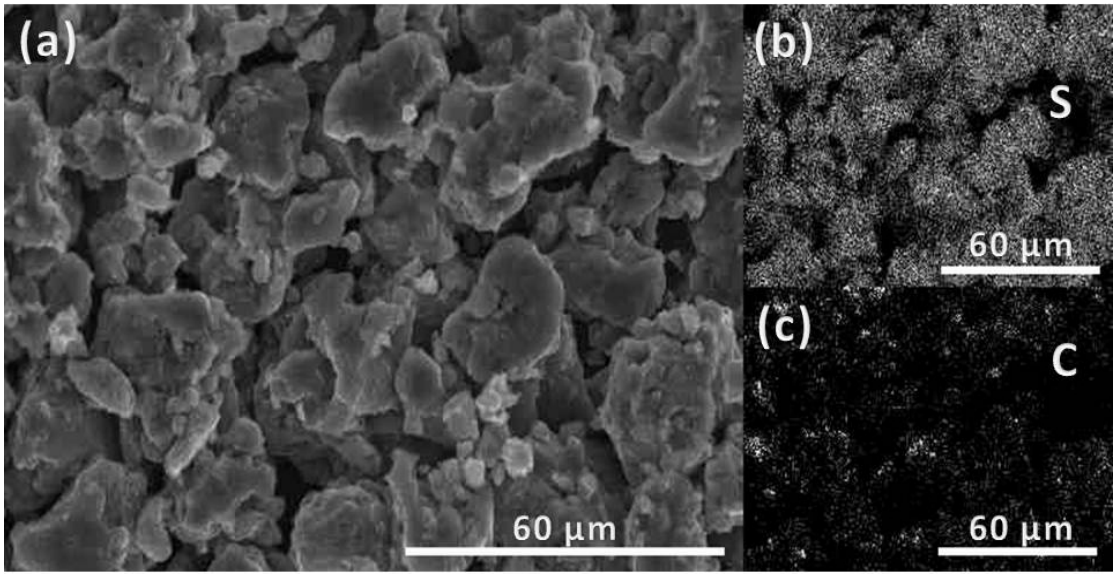

Fig. S4 (a) a low-magnification FESEM image of SWCNHs-S composite; (b) Elemental mapping of sulfur and (c) carbon.

Table 2 Comparison of the performances of the Li-S batteries based on the SWCNHs with those of other typically carbon matrix

Classification	BET surface area ($m^2 g^{-1}$)	Total pore volume ($m^3 g^{-1}$)	S content (wt %)	Cycle performance of the composites(1C=1675 mA g^{-1})	Ref.
SWCNHs	**203**	**0.48**	**76**	**693 mAh g^{-1}/1600 mA g^{-1}/100th cycle**	**Our work**
SWCNTs	**798**	**1.72**	**55.6**	**441 mAh g^{-1}/1 C/100th cycle**	**1**
Graphene	**598**	**2.61**	**67**	**424 mAh g^{-1}/0.2 C/100th cycle**	**2**
	--	**--**	**44.5**	**662 mAh g^{-1}/1 C/100th cycle**	**3**
	--	**--**	**73**	**615 mAh g^{-1}/1C/100th cycle**	**4**
GO	**--**	**--**	**66**	**954 mAh g^{-1}/0.1C/50th cycle**	**5**
Mesoporous carbon	**1472**	**2.28**	**57.2**	**878 mAh g^{-1}/0.05C/50th cycle**	**6**
	1175	**4.8**	**83.2**	**613 mAh g^{-1}/0.1C/50th cycle**	**7**

-- These values are not obtained in the literature.

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