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Polysulfide reduction accelerator-modified sulfurized polyacrylonitrile as a high performance cathode material for lithium-sulfur battery

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Table S1 Characteristics of various SPAN composites reported in literature

Material [Reference]	S content of material	Cycle performance,1C=1.675A/g calculated based on sulfur mass / composite	capacity retention
pPAN–S/MWCNTs[1]	35.1%	0.5C，100 cycles 1398mAh/g 491mAh/g	96.5%
PAN–S–VA[2]	36.89%	0.25C，200 cycles 1292mAh/g 477mAh/g	96.5%
S/PAN/$Mg_{0.6}Ni_{0.4}O$[3]	38.5%	0.1C，100 cycles 1223mAh/g 470mAh/g	~100%
S–PAN[4]	42%	0.2C，100 cycles 1050mAh/g 441mAh/g	88%
SPAN/RGO[5]	44 %	0.1C，100 cycles 1245mAh/g 548mAh/g	85%
pPAN–S@GNS[6]	47%	0.1C，100 cycles 1276mAh/g 600mAh/g	~86%
pPAN-S/GNS[7]	47%	0.1C，100 cycles 1200mAh/g 564mAh/g	80%
S/PAN/Graphene[8]	47.3%	0.1C，100 cycles 996mAh/g 471mAh/g	77%
NiS_2-SPAN	**46%**	**0.2A/g，100 cycles** **1533mAh/g 705mAh/g**	**89%**

Experimental section

Preparation of the SPAN and NiS_2-SPAN materials

The mixture of $NiCO_3$ (0.1g), sulfur (6g) and PAN (2g) was carried out by ball-milling at 500rpm for 6h in ethanol medium. Before further processing, the mixture was dried at 60℃ in a vacuum for 24h. Then the NiS_2-SPAN composite was prepared by heating the above mixture at 350℃ for 5h at 5℃/min. The synthesis method for SPAN composite is similar to the preparation of NiS_2-SPAN composite, but without the addition of $NiCO_3$ in the ball-milling process.

Materials characterization

The elemental contents of the NiS_2-SPAN and SPAN materials were determined by means of elemental analysis (CHNS, Vario EL Cube, Elementar). The content of Ni in the NiS_2-SPAN composite was determined quantitatively by inductively coupled plasma atomic emission spectrometer (ICP-AES, HORIBA Jobin Yvon ULTIMA-C). The microstructures of the samples were studied by scanning electron microscopy (SEM, JEOL, JSM-7401F) transmission electron microscopy (TEM) (JEM-2010F, JEOL, Tokyo, Japan). The phase compositions of the NiS_2-SPAN and SPAN materials were obtained by a Rigaku D/max 2500PC (Rigaku, Corp, Japan) using Cu Kα radiation in the 2θ range of 5^o to 70^o. X-ray photoelectron spectra (XPS) were recorded on an AXIS Ultra DLD spectrometer (Kratos) to analyze the surface composition.

Electrochemical measurements

The electrochemical experiments were carried out with CR2025 type coin cells assembled in an argon-filled glove box. The working electrode was prepared by casting the slurry of 80wt% active material, 10wt% acetylene black and 10wt% LA132 (as the binder) on an Al foil as a current collector. The electrodes were dried at 60℃ under vacuum for 24h and then were cut into discs with 14mm in diameter. Sample electrodes with cathode composite loadings of *ca.* 2.5mg/cm^2, corresponding to sulfur load of 1.15mg/cm^2. 1M $LiPF_6$ solution in ethylene carbonate/dimethyl carbonate/diethyl carbonate (EC: DMC: DEC, volume ratio=1:1:1) as the liquid electrolyte. A Celgard2325 membrane was used as the separator. A lithium-foil was used as counter electrode. The cells were charged/discharged on a LAND CT2001A multi-channel battery test system at room temperature in a voltage range of 1-3V (vs. Li/Li$^+$). Electrochemical impedance spectroscopy (EIS) was carried out on a CHI660D electrochemical workstation in the frequency range from 10^5 to 0.01Hz and the amplitude of the used perturbation was 5mV. Cyclic voltammetry measurements were carried out on the CHI660D electrochemical workstation over the potential range 1-3V vs. Li$^+$/Li at a scan rate of 0.1mV/s. The current density was calculated based on the sulfur mass of the active material.

Table S2 Elemental analysis of SPAN and NiS_2-SPAN

Sample	C (wt. %)	Ni (wt. %)	S (wt. %)
SPAN	37.38	—	45.95
NiS_2-SPAN	36.61	1.3	46.01

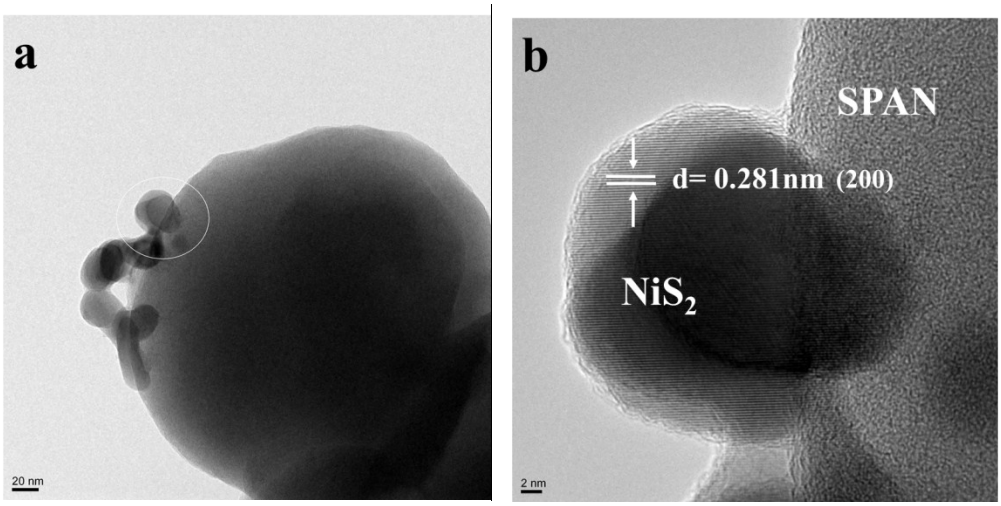

Fig.S1 (a) TEM image and (b) HR-TEM image for NiS_2-SPAN.

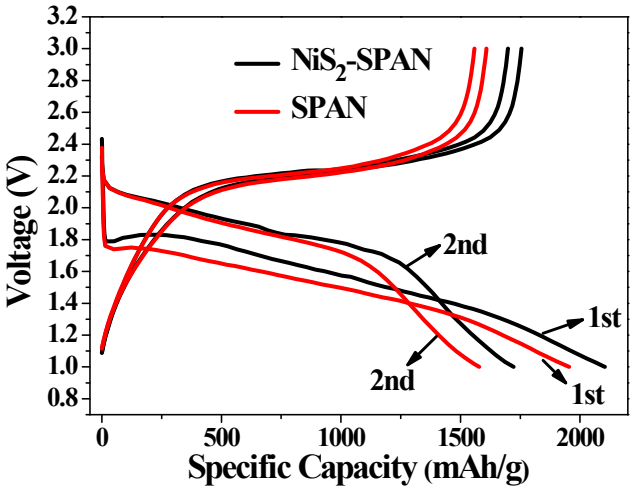

Fig.S2 Initial and second discharge/charge curves of SPAN and NiS_2-SPAN.

Table S3 The fitted results of EIS.

Electrode	**$R_1(\Omega)$**	**$R_2(\Omega)$**	**$R_3(\Omega)$**
SPAN	4.26	127	46.98
NiS_2-SPAN	5.19	105.80	21.12

Table S4 The cathodic and anodic peaks from the first CV scan.

Electrode	$E1_a$(V)	$E1_c$(V)	ΔE1	$E2_a$(V)	$E2_c$(V)	ΔE2	$E3_a$(V)	$E3_c$(V)	ΔE3
SPAN	2.38	1.22	**1.16**	2.44	1.7	**0.74**	2.5	1.63	**0.87**
NiS_2-SPAN	2.35	1.39	**0.96**	2.38	1.75	**0.63**	2.39	1.7	**0.69**

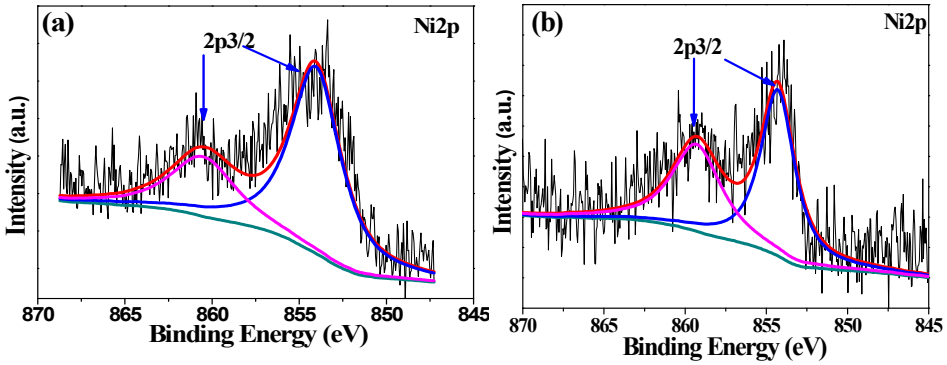

Fig.S3 Ni2p of NiS_2-SPAN in (a) discharged state and (b) charged state.

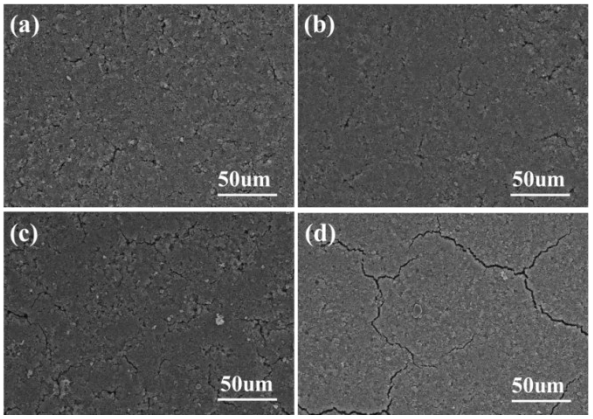

Fig.S4 SEM images of (a, c) NiS_2-SPAN and (d, f) SPAN electrodes before and after 100 cycles.

References

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