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# **Supplementary Materials**

#### A High Strength, Free-Standing Cathode Constructed by Regulating

## Graphitization and Pore Structure in Nitrogen-Doped Carbon Nanofibers for

### Flexible Lithium-Sulfur Battery

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Fig. S1 SEM images of SiO<sub>2</sub>/G/NPCFs with SiO<sub>2</sub> concentration of 30 mg mL<sup>-1</sup>.



**Fig. S2** XRD diffraction pattern of S/G/NPCFs (inset: partial enlarge pattern of S/G/NPCFs).



Fig. S3 Elemental mapping of S/G/NPCFs.



**Fig. S4** Mechanical strength comparisons of this work with some other flexible S/C cathodes.



Fig. S5 a) Nitrogen adsorption-desorption isotherms of NCFs and b)  $SiO_2/G/NPCFs$  (insets are the corresponding pore size distribution curves).



Fig. S6 Mechanical properties of sulfur composite film without adding  $SiO_2$ . a) Photographs of S/NCFs composite film and b) S/G/NCFs composite film.



**Fig. S7** a) TG curves of S/NCFs (in nitrogen) and S/G/NCFs (in nitrogen); b) TG curves of SiO<sub>2</sub>/NPCFs (in air) and S/NPCFs (in nitrogen); c) TG curves of SiO<sub>2</sub>/G/NPCFs (in air) and S/G/NPCFs (in nitrogen).



Fig. S8 TEM images of a) SiO<sub>2</sub>/G/NPCFs, b) G/NPCFs;



Fig. S9 HRTEM of G/NPCFs (inset is the SAED pattern).



Fig. S10 TEM images of NPCFs.



Fig. S11 HRTEM image of NPCFs (inset is the SAED pattern).



**Fig. S12** Energy efficiency and Coulombic efficiency of S/G/NPCFs electrode at different current densities.



**Fig. S13** a) Rate capabilities of S/NPCFs and S/G/NPCFs electrode; b) Typical discharge-charge curves of S/NPCFs electrodes recorded at current rates of 0.1 C, 0.5 C, 1 C, 2 C, and 5 C.



**Fig. S14** The open-circuit voltage of a flexible battery using S/G/NCFs electrode as cathode at different bending angles.

	Precursor	Carbonization	Etch	Sulfurization
With adding	SiO <sub>2</sub> /GO/PA	SiO./C/NPCEs		S/C/NPCFs
GO and SiO <sub>2</sub>	Ν	5102/0/111 CF5	Unit Cry	5/0/111 CF5
With adding			NDCEs	S/NDCEs
SiO <sub>2</sub>			MICIS	5/INF CF 5
Without adding			NCFs	S/NCFs
GO and SiO <sub>2</sub>			ners	SACTS
Without adding				S/C/NCEs
SiO <sub>2</sub>			Griters	5/0/11015

**Table S1.** The detail information of the as-prepared samples' abbreviations.

Sample	BET surface area(m <sup>2</sup> g <sup>-1</sup> )	Total pore volume(cm <sup>3</sup> g <sup>-1</sup> )	Micropore volume(cm <sup>3</sup> g <sup>-1</sup> )
NCFs	577	0.277	0.227
SiO <sub>2</sub> /G/NPCFs	232	0.371	0.093
G/NPCFs	429	0.522	0.173
S/G/NPCFs	14	0.095	0.005

Table S2. Textural parameters of NCFs, SiO<sub>2</sub>/G/NPCFs, G/NPCFs, S/G/NPCFs.

**Table S3.** Comparison of rate capabilities of representative S cathode materials in the literature.

Matorial	Sulfur	Rate capability	
Material	Loading		
		815 mAh g <sup>-1</sup> at 0.5 C	
S/C/NDCE		735 mAh g <sup>-1</sup> at 1 C	
S/G/NPCFS	53 wt.% (flexible)	670 mAh g <sup>-1</sup> at 2 C	
This work		540 mAh g <sup>-1</sup> at 5 C	
		$(1 \text{ C} = 1675 \text{ mA g}^{-1})$	
		1500 mAh g <sup>-1</sup> at ~0.03 C	
graphene/sultur	66.7 Wt%	750 mAh g <sup>-1</sup> at ~0.5 C	
Rej. SI	(flexible)	500 mAh g <sup>-1</sup> at ~1 C	
CNF/sulfur	40 wt%	645 mAh g <sup>-1</sup> at ~0.06 C	
Ref. S2	(N/A)	437 mAh g <sup>-1</sup> at ~0.6 C	
Cu-CNF/sulfur	52 wt%	590 mAh g <sup>-1</sup> at ~0.06 C	
Ref. S3	(N/A)	419 mAh g <sup>-1</sup> at ~0.6 C	
Graphene/N-doped	~55 wt%	800 mAh g <sup>-1</sup> at 1C	
Hollow Carbon	(flexible)	600 mAh g <sup>-1</sup> at 2C	

Nanosphere/sulfur		430 mAh g <sup>-1</sup> at 3C
Ref. S4		
		1045mAh g <sup>-1</sup> at 0.2 C
Polysulfides/CN1/AC	55 wt%	955 mAh g <sup>-1</sup> at 0.5 C
NF@MnO2	(flexible)	878 mAh g <sup>-1</sup> at 1 C
Kef. SS		773 mAh g <sup>-1</sup> at 2 C
		1078 mAh g <sup>-1</sup> at 0.2 C
C/Dilanar Carlan	< 40	964 mAh g <sup>-1</sup> at 0.5 C
S/Bilayer Carbon	< 40  Wt%	890 mAh g <sup>-1</sup> at 1 C
<i>Kej.</i> S0	(N/A)	720 mAh g <sup>-1</sup> at 2 C
		685 mAh g <sup>-1</sup> at 3 C
		656 mAh g <sup>-1</sup> at 0.4 C
C/CNIT Network	(0	571 mAh g <sup>-1</sup> at 0.8 C
S/CNT INELWOIK	60 Wl%	541 mAh g <sup>-1</sup> at 1 C
<i>Kej.</i> 57	$(\mathbf{N}/\mathbf{A})$	503 mAh g <sup>-1</sup> at 2 C
		452 mAh g <sup>-1</sup> at 5 C
Aming functionalized	70 x + 9/	~950 mAh g <sup>-1</sup> at 0.5 C
Carbon Nanatuka	/0 wt% (56 wt% in the	~890 mAh g <sup>-1</sup> at 1 C
		~650 mAh g <sup>-1</sup> at 2 C
<i>Kej.</i> 58	electrode)	$\sim$ 300 mAh g <sup>-1</sup> at 4 C
S/ hierarchical		
Microporous-	50.5	812  mAb  scl  st = 0.5  C
mesoporous	$50.5 \text{ W}^{10}$	$613 \text{ mAn g}^2 \text{ at } \sim 0.5 \text{ C}$
Carbonaceous	(42.9 wt% in the	$401 \text{ mAh } \text{g}^{-1} \text{ at } \sim 1 \text{ C}$
Nanotubes	electrode)	491 mAn g <sup>-1</sup> at ~2 C
Ref. S9		

#### **References:**

- (S1) P. Kumar, F.-Y. Wu, L.-H. Hu, S.A. Abbsa, J. Ming, C.-N. Lin, J. Fang, C.-W. Chu, L.-J. Li, *Nanoscale*, 2015, 7, 8093-8100.
- (S2) L. Zeng, F. Pan, W. Li, Y. Jiang, X. Zhong, Y. Yu, Nanoscale, 2014, 6, 9579-9587.
- (S3) L. Zeng, Y. Jiang, J. Xu, M. Wang, W. Li, Y. Yu, Nanoscale, 2015, 7, 10940-10949.
- (S4) G.-M. Zhou, Y.-B. Zhao, A. Manthiram, *Adv. Energy Mater.*, 2015, **5**, 1402263-1402272.
- (S5) H.-H. Xu, L. Qie, A. Manthiram, Nano Energy, 2016, 26, 224-232.
- (S6) H.-S. Kang, Y.-K. Sun, Adv. Funct. Mater., 2016, 26, 1225-1232.
- (S7) L. Sun, D.-T. Wang, Y.-F. Luo, K. Wang, W.-B. Kong, Y. Wu, L.-N. Zhang, K.-L. Jiang, Q.-Q. Li, Y.-H. Zhang, J.-P. Wang, S.-S. Fan, ACS Nano, 2016, 10, 1300-1308.
- (S8) L. Ma, H.-L. Zhuang, S.-Y. Wei, K. E. Hendrickson, M. S. Kim, G. Cohn, R. G. Henning, L. A. Archer, ACS Nano, 2016, 10, 1050-1059.
- (S9) K. Mi, Y. Jiang, J.-K. Feng, Y.-T. Qian, S.-L. Xiong, Adv. Funct. Mater., 2016, 26, 1571-1579.