

## **Supporting Information**

### **A novel bifunctional separator with self-assembled FeOOH/coated g-C<sub>3</sub>N<sub>4</sub>/KB bilayer in lithium sulfur battery**

*Yang Wang,<sup>a</sup> Liwen Yang,<sup>a</sup> Yanxiao Chen,<sup>a\*</sup> Qian Li,<sup>a</sup> Changtao Chen,<sup>a</sup>*

*Benhe Zhong,<sup>a</sup> Xiaodong Guo,<sup>ab</sup> Zhenguo Wu,<sup>a</sup> Gongke Wang<sup>c</sup>*

<sup>a</sup> College of Chemical Engineering, Sichuan University, Chengdu, 610065, PR China

<sup>b</sup> Institute for Superconducting and Electronic Materials, University of Wollongong,  
Wollongong, NSW 2522, Australia

<sup>c</sup> School of Materials Science and Engineering, Henan Normal University, Xinxiang,  
453007, China

Corresponding Author: Yanxiao Chen

E-mail: yxchen888@163.com

## **Measurement**

### **Fabrication of S/KB composite and cathode**

Ketjen black and elemental sulfur (weight ratio =1:4) were mixed and grounded for 45min. In argon atmosphere, the S/KB composite was successfully fabricated by heating at 155 °C for 12h. The weight ratio of S/KB, acetylene black and PVDF in the S/KB cathode is 8:1:1. The slurry, with NMP, was covered onto the aluminum foil by blade coating and dried for 12h with a temperature of 60 °C in vacuum. Then all the

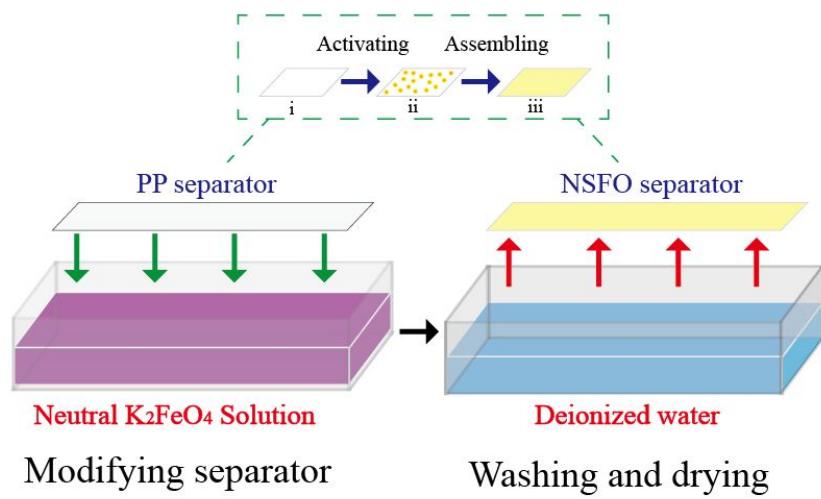
foils were cut into the same disks. Finally, the sulfur loading of the prepared S/KB cathode is about 1.2-1.5 mg/cm<sup>2</sup>.

## Assembly of Li-S battery and measurements

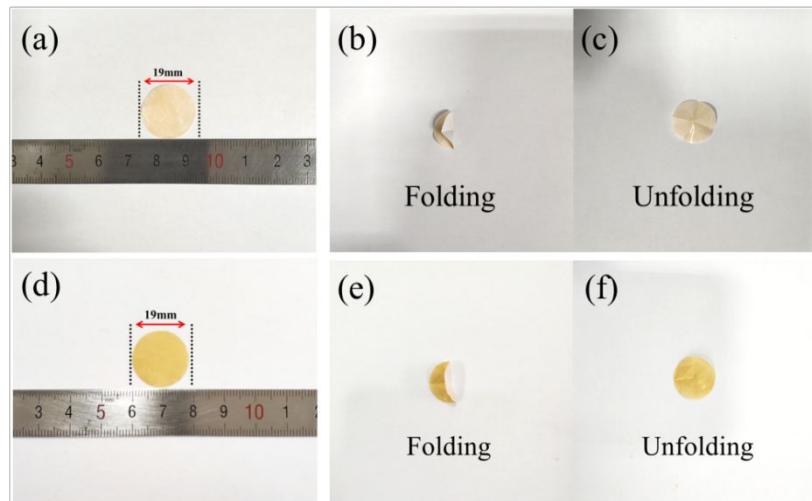
Li-S batteries were assembled in Ar atmosphere. All the cells consist of S/KB cathode, metallic lithium, modified separator and commercial electrolytes. Every cell needs 30μL electrolytes contained 1.0M of LiTFISI and 0.1M of LiNO<sub>3</sub> in DOL and DME (v:v=50%:50%). And the corresponding E/S ratio is approximately 15:1 (μL/mg). ZenniumIM6 workstation was employed to obtain the electrical impedance spectroscope (EIS). Electrochemical work station (LK 9805) was used to test cyclic voltammetry curves (1.7V-2.8V) at 0.1mV/s. LAND-CT2001A was used for collecting the galvanostatic charging and discharging data (1.7V-2.8V).

## Materials characterization

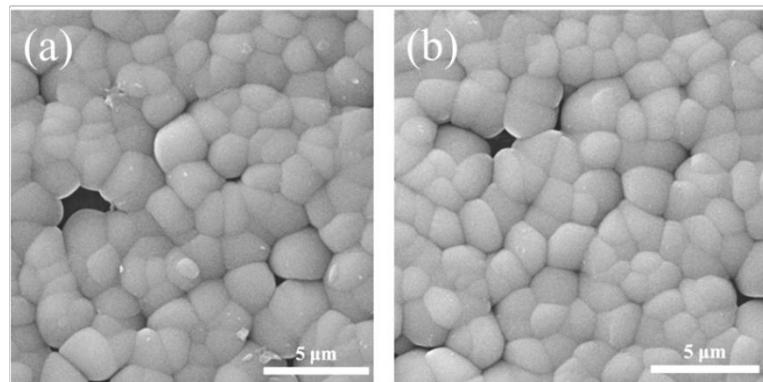
D/Max 2500 PC was used and analyzed to obtain the XRD patterns of modified separators (NSFO, ACSFO, SFO and G-SFO). SPA400 Seiko Instruments was used for checking the morphologies of modified separators (NSFO, PP, SFO and G-SFO). The EDS mapping of separators were all obtained by SEM. TA Instruments, Q600, was used to ensure the sulfur content of S/KB composite. The X-ray photoelectron spectroscopy of SFO and G-SFO separators were analyzed by Kratos Analytical Ltd.



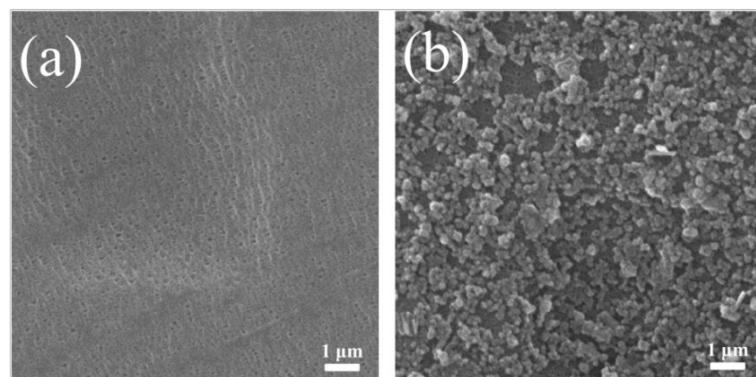
**Fig.S1** Schematic illustration of the fabrication process of NSFO modified separator.



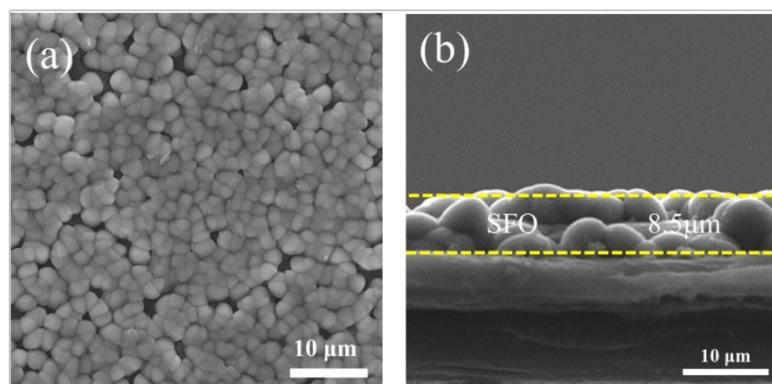
**Fig.S2** (a) Digital image of NSFO, (b)-(c) Folding and unfolding test of NSFO, (d) Digital image of SFO, (e)-(f) Folding and unfolding test of SFO.



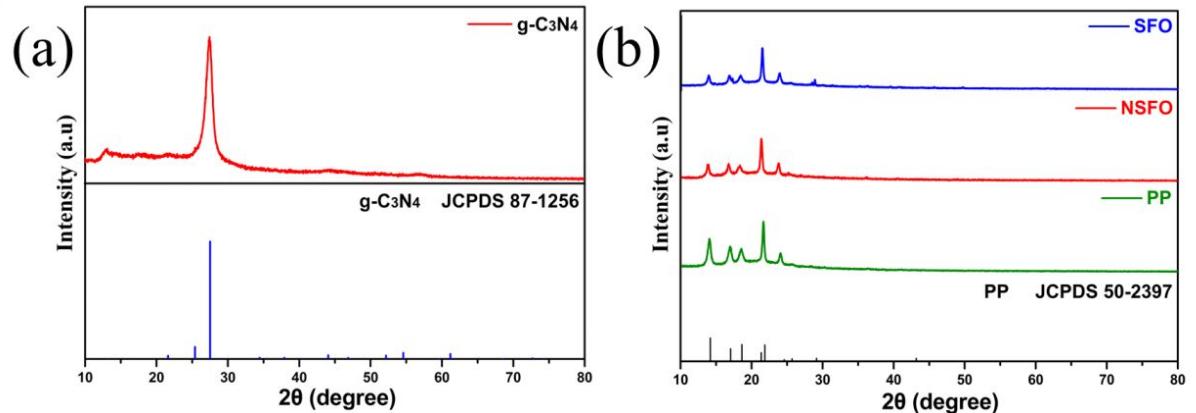
**Fig.S3** SEM images of SFO separator after sonication for (a) 5 min and (b) 10 min.



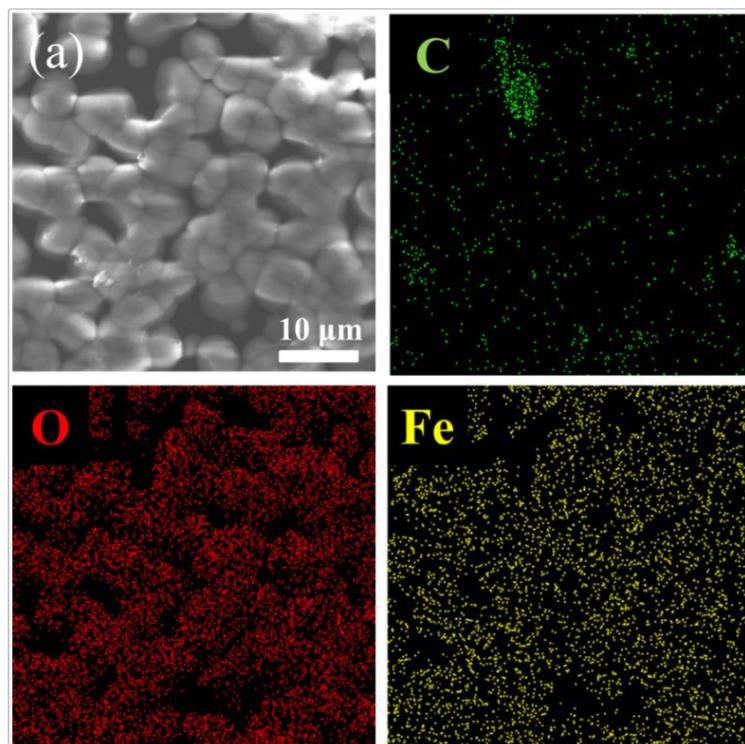
**Fig.S4** (a) SEM of pristine PP separator, (b) SEM of NSFO separator



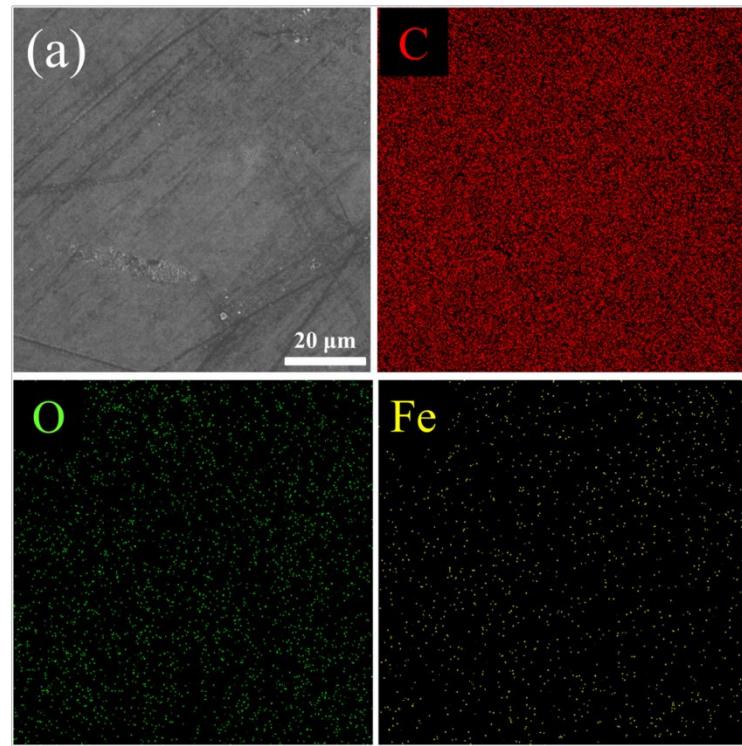
**Fig.S5** (a) SEM of SFO separator, (b) Cross-section SEM of SFO separator



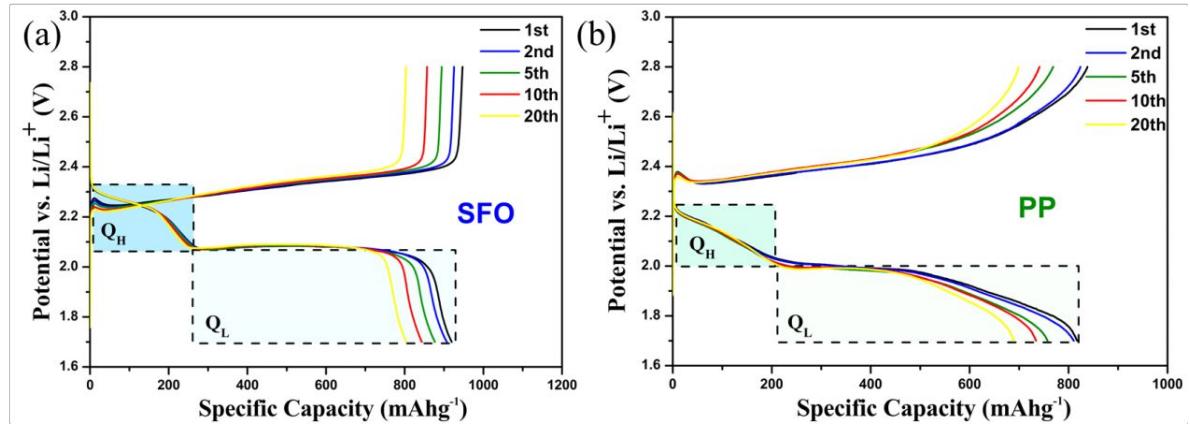
**Fig.S6** (a) XRD patterns of  $\text{g-C}_3\text{N}_4$ , (b) XRD patterns of NSFO and SFO separators.



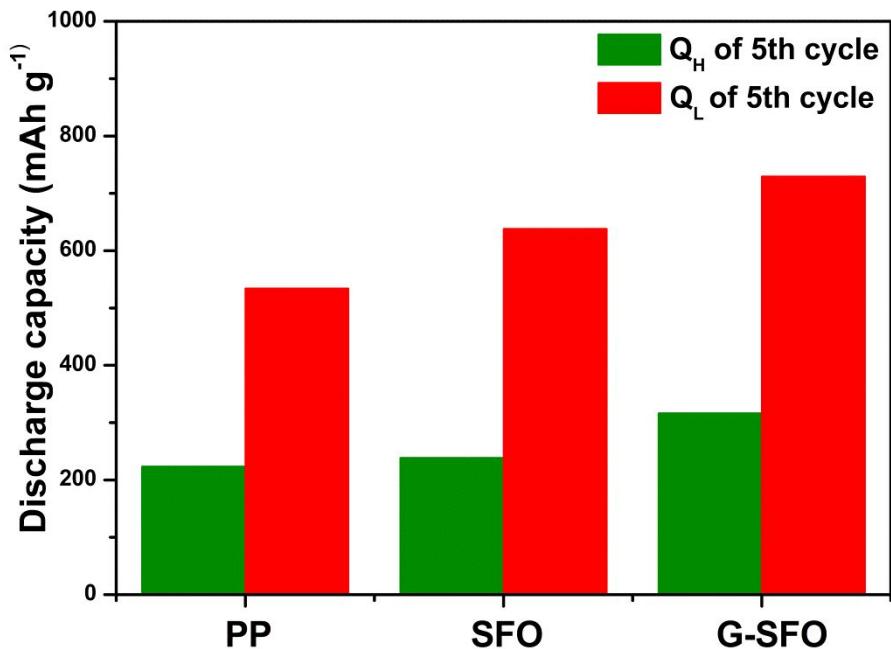
**Fig.S7** (a) SEM and the corresponding EDS mapping of SFO separator.



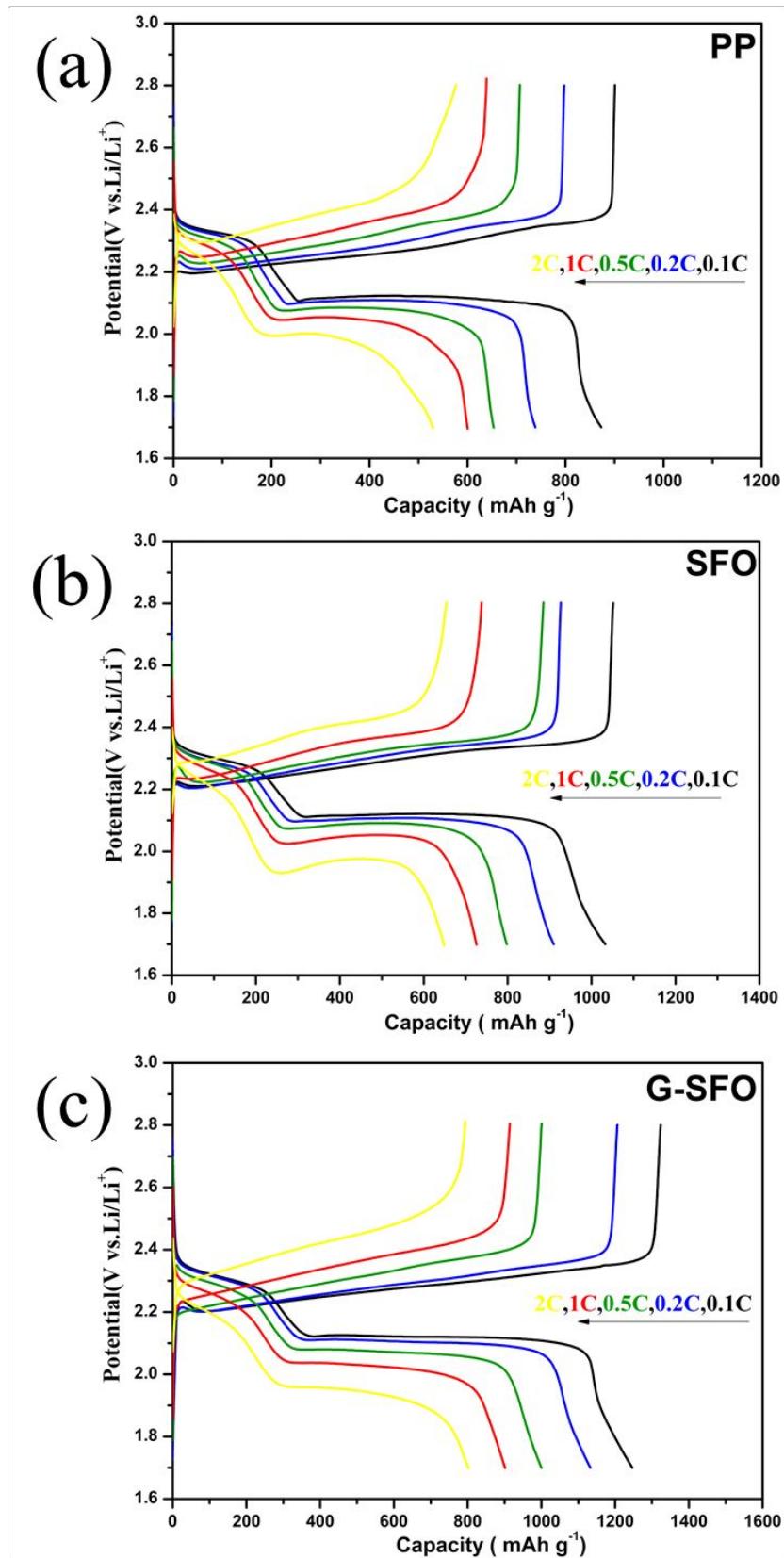
**Fig.S8** (a) SEM and the corresponding EDS mapping of SFO separator after etching.



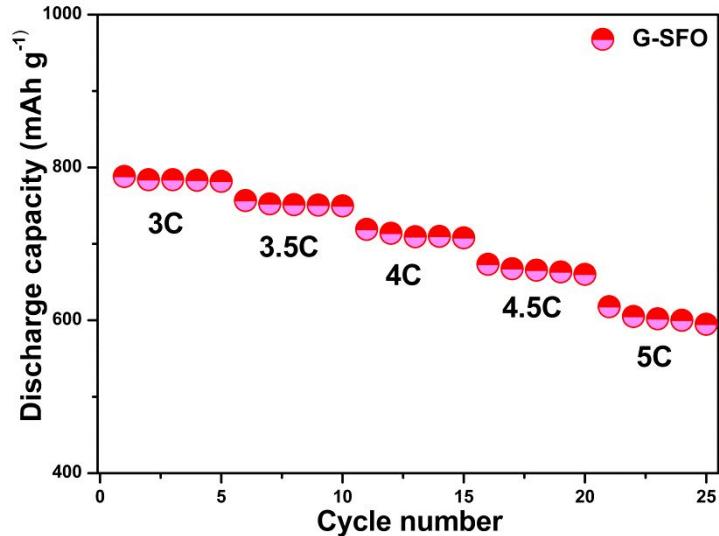
**Fig.S9** (a) Charge–discharge curves of the cell with SFO separator at 0.2C, (b) Charge–discharge curves of the cell with PP separator at 0.2C.



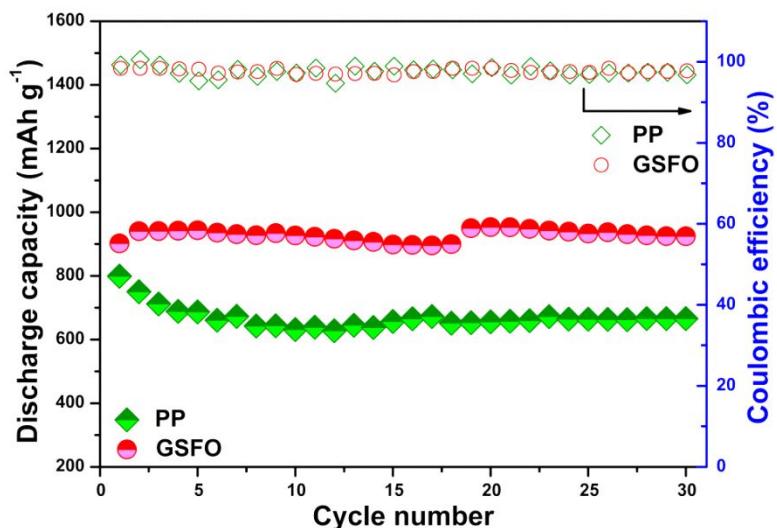
**Fig.S10** The  $Q_H$  and  $Q_L$  values of cells with PP, SFO, G-SFO separators at the 5<sup>th</sup> cycle at 0.2C.



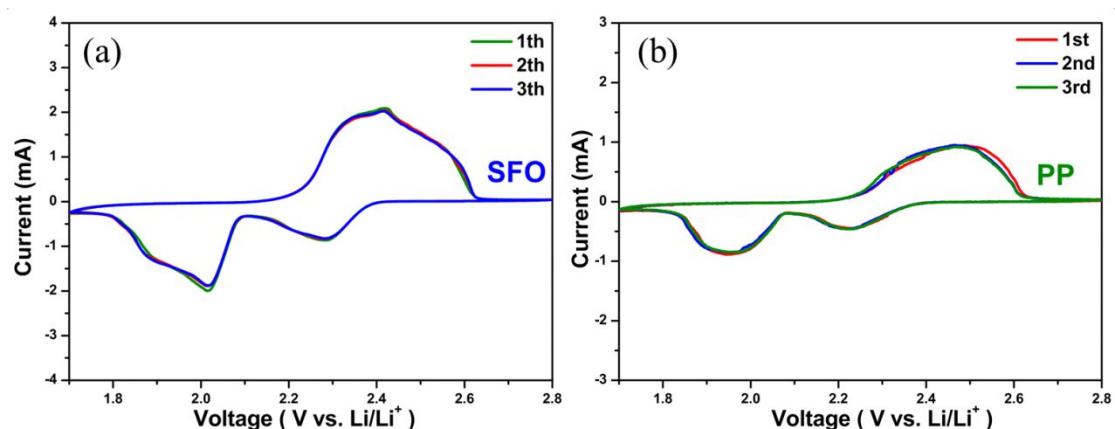
**Fig.S11** Charge–discharge curves of the cells with (a) PP separator, (b) SFO separator and (c) G-SFO separator from 0.1C to 2C.



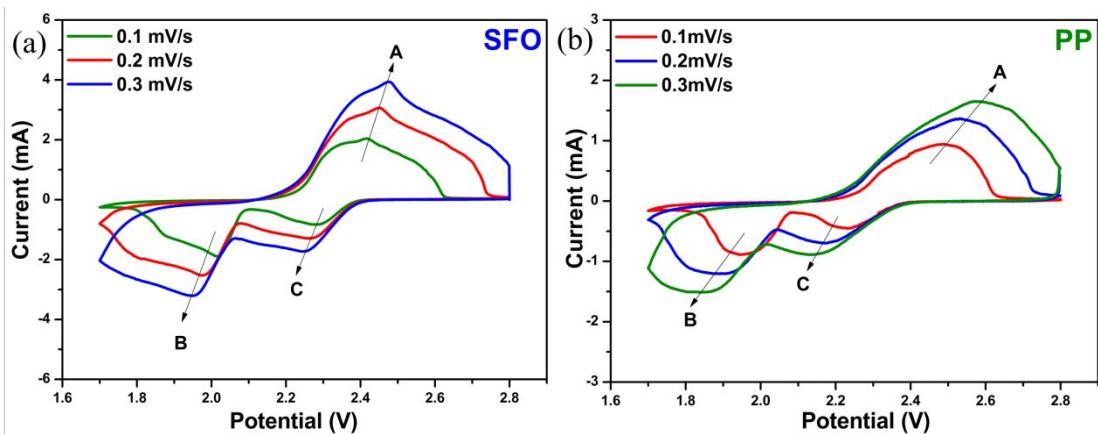
**Fig.S12** Rate capacity of the battery with G-SFO separator from 3C to 5C.



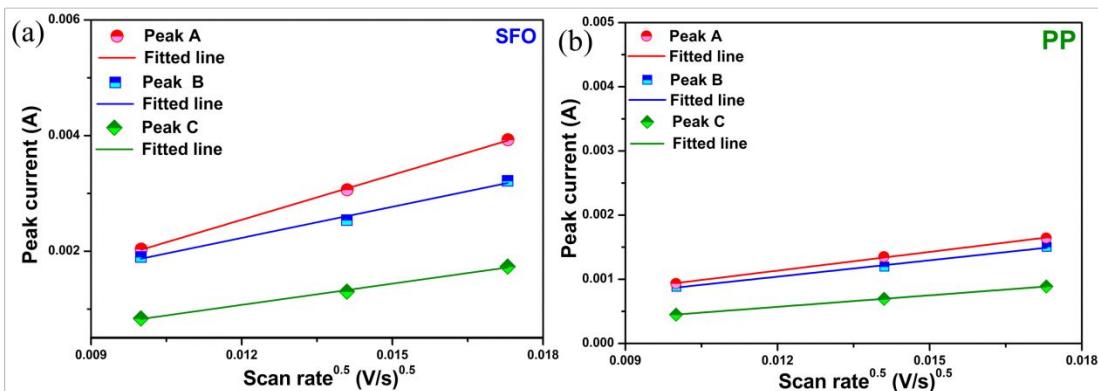
**Fig.S13** Cycling performance of the cells with PP and G-SFO separators with high sulfur loading at 0.2 C.



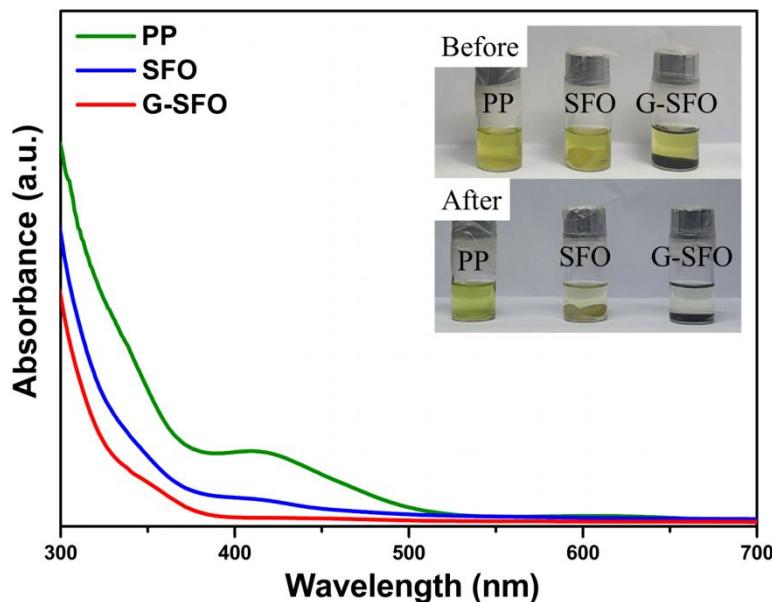
**Fig.S14** (a) CV curves of cells with SFO separator, (b) CV curves of cells with PP separator at 0.1mV/s for 3 cycles.



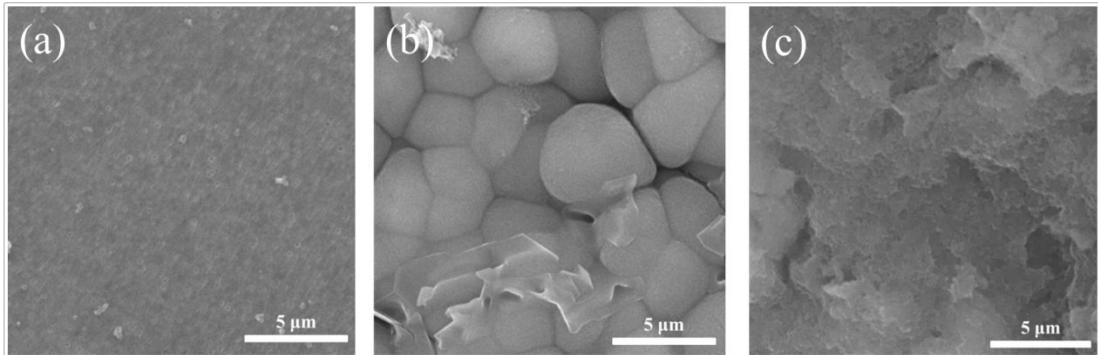
**Fig.S15** CV curves of cells with (a) SFO separator and (b) PP separator from 0.1mV/s to 0.3mV/s.



**Fig.S16** The linear fits of the peak currents for cells with (a) SFO separator and (b) PP separator.



**Fig.S17** The adsorption-UV tests of PP, SFO and G-SFO separators.



**Fig.S18** SEM image of the (a) PP separator, (b) SFO separator and (c) G-SFO separator after cycling.

**Table S1.** Impedance parameters of different separators

Cell	Impedance	
	$R_s(\Omega)$	$R_{ct}(\Omega)$
<b>PP</b>	3.46	98.09
<b>SFO</b>	3.12	84.35
<b>G-SFO</b>	2.08	50.67

**Table S2.** Apparent Li<sup>+</sup> diffusion coefficients ( $D_{Li}^{app}$ ) of cells with different separators

Apparent Li <sup>+</sup> diffusion coefficients			
$D_{Li}^{app}$	Peak A	Peak B	Peak C
<b>PP</b>	$6.92 \times 10^{-9}$	$5.26 \times 10^{-9}$	$2.62 \times 10^{-9}$
<b>SFO</b>	$4.88 \times 10^{-8}$	$2.35 \times 10^{-8}$	$1.09 \times 10^{-8}$
<b>G-SFO</b>	$9.78 \times 10^{-8}$	$4.32 \times 10^{-8}$	$2.33 \times 10^{-8}$

**Table S3.** Summary of electrochemical performance of Li-S batteries with different modified separators and interlayers

Materials	Sulfur loading (mg cm <sup>-2</sup> )	Thickness (μm)	Rate Capability (mAh/g)			Cycling Stability			Reference
			0.5C	1C	2C	Rate	Cycles	Decay (per cycle)	
<b>FeOOH/KB/g-C<sub>3</sub>N<sub>4</sub></b>	<b>1.2-1.5</b>	<b>12.1</b>	<b>1000</b>	<b>901</b>	<b>802</b>	0.5C	<b>500</b>	<b>0.080%</b>	<i>N/A</i>
						1C	<b>900</b>	<b>0.055%</b>	<i>N/A</i>
<b>Y<sub>2</sub>O<sub>3</sub>/KB<sup>1</sup></b>	1.3	12	997	908	846	1C	200	<b>0.1135%</b>	Ref S1
<b>ZnO/PPy<sup>2</sup></b>	0.8	20	718	609	404	0.2C	100	<b>0.517%</b>	Ref S2
<b>Cr<sub>2</sub>O<sub>3</sub>/CNF<sup>3</sup></b>	1.3-1.6	-	990	960	890	0.2C	260	<b>0.090%</b>	Ref S3
<b>Fe<sub>3</sub>O<sub>4</sub>/PG<sup>4</sup></b>	0.6-0.9	15	789	673	589	1C	2000	<b>0.020%</b>	Ref S4
<b>TiO<sub>2</sub>/CNFs<sup>5</sup></b>	0.8	35	940	740	620	1C	500	<b>0.051%</b>	Ref S5
<b>ZnO/Graphene<sup>6</sup></b>	1.1-1.5	-	1131	895	807	1C	500	<b>0.093%</b>	Ref S6
<b>BaTiO<sub>3</sub>/Rgo/CNFs<sup>7</sup></b>	1	30	963	852	758	0.3C	200	<b>0.200%</b>	Ref S7
<b>LaLiO<sub>2</sub><sup>8</sup></b>	1.1-1.5	-	820	700	600	1C	250	<b>0.052%</b>	Ref S8
<b>KB/IR<sup>9</sup></b>	0.8	20	886	783	653	1C	500	<b>0.105%</b>	Ref S9
<b>Ca(OH)<sub>2</sub>/C<sup>10</sup></b>	1.2-1.5	4	950	910	830	0.5C	250	<b>0.112%</b>	Ref S10

## Reference

- (1) Wang, S. W.; Qian, X. Y.; Jin, L. N.; Rao, D. W.; Yao, S. S.; Shen, X. Q.; Xiao, K. S.; Qin, S. B. Separator Modified by  $\text{Y}_2\text{O}_3$  Nanoparticles-Ketjen Black Hybrid and Its Application in Lithium-Sulfur Battery. *Journal of Solid State Electrochemistry* **2017**, *21* (11), 3229-3236, DOI: 10.1007/s10008-017-3649-5.
- (2) Yin, F. X.; Ren, J.; Zhang, Y. G.; Tan, T. Z.; Chen, Z. H. A PPy/ZnO Functional Interlayer to Enhance Electrochemical Performance of Lithium/Sulfur Batteries. *Nanoscale Research Letters* **2018**, *13*, DOI: 10.1186/s11671-018-2724-x.
- (3) Guan, Y. P.; Liu, X. J.; Akhtar, N.; Wang, A. B.; Wang, W. K.; Zhang, H.; Suntivich, J.; Huang, Y. Q. Cr<sub>2</sub>O<sub>3</sub> Nanoparticle Decorated Carbon Nanofibers Derived from Solid LeatherWastes for High Performance Lithium-Sulfur Battery Separator Coating. *Journal of the Electrochemical Society* **2019**, *166* (8), A1671-A1676, DOI: 10.1149/2.1181908jes.
- (4) Xiang, Y.; Wang, Z.; Qiu, W.; Guo, Z.; Liu, D.; Qu, D.; Xie, Z.; Tang, H.; Li, J. Interfacing Soluble Polysulfides with a SnO<sub>2</sub> Functionalized Separator: An Efficient Approach for Improving Performance of Li-S Battery. *Journal of Membrane Science* **2018**, *563*, 380-387, DOI: 10.1016/j.memsci.2018.06.004.
- (5) Liang, G. M.; Wu, J. X.; Qin, X. Y.; Liu, M.; Li, Q.; He, Y. B.; Kim, J. K.; Li, B. H.; Kang, F. Y. Ultrafine TiO<sub>2</sub> Decorated Carbon Nanofibers as Multifunctional Interlayer for High-Performance Lithium-Sulfur Battery. *ACS Appl. Mater. Interfaces* **2016**, *8* (35), 23105-23113, DOI: 10.1021/acsami.6b07487.
- (6) Shi, N. X.; Xi, B. J.; Feng, Z. Y.; Wu, F. F.; Wei, D. H.; Liu, J.; Xiong, S. L. Insight into Different-Microstructured ZnO/Graphene-Functionalized Separators Affecting the Performance of Lithium-Sulfur Batteries. *Journal of Materials Chemistry A* **2019**, *7* (8), 4009-4018, DOI: 10.1039/c8ta12409d.
- (7) Zhang, S. Q.; Qin, X. Y.; Liu, Y. M.; Zhang, L. H.; Liu, D. Q.; Xia, Y.; Zhu, H.; Li, B. H.; Kang, F. Y. A Conductive/Ferroelectric Hybrid Interlayer for Highly Improved Trapping of Polysulfides in Lithium-Sulfur Batteries. *Advanced Materials Interfaces* **2019**, *6* (22), DOI: 10.1002/admi.201900984.

- (8) Bizuneh, G. G.; Fan, J. M.; Sun, C.; Yuan, X. F.; Xue, F.; Deng, D. R.; Lei, J.; Lin, X. D.; Jia, Y. J.; Yang, J. F.; Yan, H.; Wang, X. Y.; Zheng, M. S.; Dong, Q. F. LaLiO<sub>2</sub>-Based Multi-Functional Interlayer for Enhanced Performance of Li-S Batteries. *Journal of the Electrochemical Society* **2019**, *166* (2), A68-A73, DOI: 10.1149/2.0271902jes.
- (9) Zuo, P. J.; Hua, J. F.; He, M. X.; Zhang, H.; Qian, Z. Y.; Ma, Y. L.; Du, C. Y.; Cheng, X. Q.; Gao, Y. Z.; Yin, G. P. Facilitating the Redox Reaction of Polysulfides by an Electrocatalytic Layer-Modified Separator for Lithium-Sulfur Batteries. *Journal of Materials Chemistry A* **2017**, *5* (22), 10936-10945, DOI: 10.1039/c7ta02245j.
- (10) Shao, H. Y.; Huang, B. C.; Liu, N. Q.; Wang, W. K.; Zhang, H.; Wang, A. B.; Wang, F.; Huang, Y. Q. Modified Separators Coated with a Ca(OH)<sub>2</sub>-Carbon Framework Derived from Crab Shells for Lithium-Sulfur Batteries. *Journal of Materials Chemistry A* **2016**, *4* (42), 16627-16634, DOI: 10.1039/c6ta06828f.