Supporting Information

The Electrochemical Active Separator with Excellent Catalytic Ability toward High-Performance Li-S Batteries

Maoxu Wang,[a,b] Lishuang Fan,[a,b] Yue Qiu,[a,b] Dandan Chen,[a,b] Xian Wu,[a,b] Chenyang Zhao,[a,b] Junhan Cheng, [a,b] Yan Wang,[*c] Naiqing Zhang,[*a,c] and Kening Sun[*a,c]

a. State Key Laboratory of Urban Water Resource and Environment, Harbin Institute of Technology, Harbin 150001, China

b. School of Chemistry and Chemical Engineering, Harbin Institute of Technology, Harbin 150001, China

c. Academy of Fundamental and Interdisciplinary Sciences, Harbin Institute of Technology, Harbin 150001, China

*Corresponding Author

Naiqing Zhang (znqmww@163.com)

Academy of Fundamental and Interdisciplinary Sciences, Harbin Institute of Technology, Harbin, China

Tel: +451 86412153 Fax: +86-451-86412153

Experiment

Synthesis of VS_4/G: Thioacetamide (360 mg) and 120 mg $NaVO_4$ were dissolved into 100 mL beaker which contain 45 mL deionized water, and then add 15 mL graphene oxide dispersion (8 mg mL^{-1}) into the above solution. The mixed solution was stirred for 30 min and then transferred into 100 mL Teflon–lined stainless steel autoclave. Then the autoclave was kept at 160 °C for 12 h, and waiting for the temperature cool to room temperature naturally. The products was collected by centrifugation and washed with absolute ethanol and deionized water for several times. The final product was collected after freeze-drying.

Synthesis of VS_4/G-Separator: The VS_4/G was dispersed into absolute ethanol and sonicated for 4 h, and then filtrated the mixture of VS_4/G and ethanol onto a conventional separator (The Celgard 3501 separator). Afterwards, the VS_4/G-Separator was dried in the oven overnight and cut into circular pieces. The different thickness of VS_4/G-Separator was fabricated by adjusting the quality of VS_4/G. The G-Separator was prepared with the same method.

Material Characterization: The morphology and microstructure characterization for VS_4/G and was conducted using a SEM (Hitachi, SU8010) and HRTEM (G2 F20FEI Tecnai G2 F20 microscope at 200 kV). Elemental mapping was performed with SEM. The crystal structure were characterized by X-ray diffraction (PANalytical X'Pert PRO, monochromated Cu Kα radiation 40 mA, 40 kV). The method used in the paper is flat sample stage. Step size is 0.013°, scan range from 5° to 90° with a scan rate 1°/min. The sulfur content in the composite was tested by TG thermogravimetric analyzer system. The specific surface area, pore volume and N_2 adsorption/desorption isotherms were measured by using an ASAP 2020 (Micromeritics). X-ray

photoelectron spectroscopy (XPS) was performed using a Thermo Scientific K-Alpha XPS (Fisher Scientific Ltd, Nepean, ON). The energy resolution is 0.5 eV and the step size is 0.1 eV.

Electrochemical Measurements: 75 mg sulfur and 25 mg ketjen black were mixed uniformly and placed in an autoclave and heated at 155 °C for 12 h. Sulfur electrodes were fabricated by pasting sulfur slurry (80 wt% sulfur-ketjen black as active material, 10 wt% super P and 10 wt% PVDF binder) onto aluminum foil current collector. The slurry pasted Al foils were vacuum dried at 60ºC for 12 h. The sulfur electrodes were then cut into circular pellets and were used as working electrodes. The 2025 coin cells were assembled with Li metal disc as anode in a glovebox filled with Ar. The Celgard 3501 sheet was as the commercial separator. The electrolyte was composed of 1mol/L lithium bis (trifluoromethanesulfonyl) imide (LiTFSI) in a solvent of 1, 3-dioxolane (DOL) and dimethoxymethane (DME) (1:1 ratio by volume) with 2% $LiNO_3$ addition. CV test was recorded on a CHI 660D electrochemical workstation between 1.8 and 2.8 V. The charge transfer kinetics was investigated by EIS measurements using a PARSTAT 2273 advanced electrochemical system, the frequency range was set between 1 MHz and 1Hz and the amplitude is 10 mV with an ac signal. A Neware battery test system is used toperform charge/discharge measurements, the voltage window is 1.8–2.8 V for various current rates (1 C is equivalent to 1675 mA/g).

Results

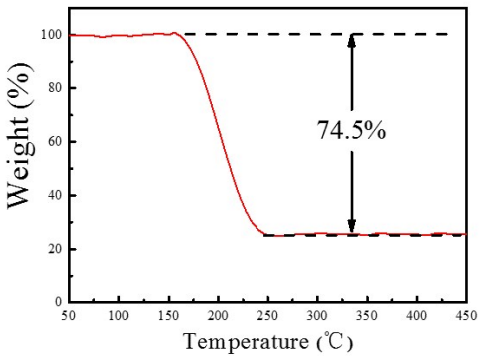

Figure S1. Thermogravimetric (TG) curves of sulfur cathodes with ketjen black.

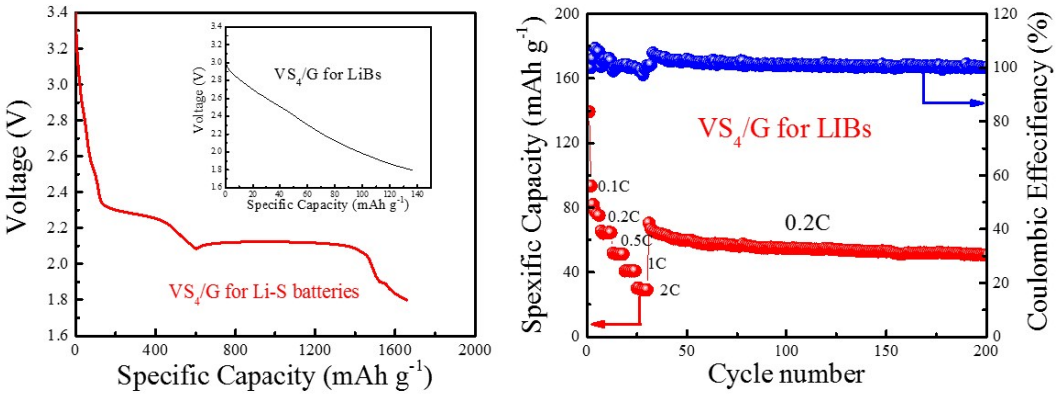

Figure S2. a) The discharge profiles of VS_4/G-Separator for Li-S batteries at 0.1 C couple with VS_4/G for LiBs. b) The rate performance and cycle performance of VS_4/G in Li-S batteries system.

Table S1. A brief summary of the reported new separator with modified materials for Li–S batteries

Separator	Sulfur wt%	Rate	Initial Capacity (mAh/g)	Cycled Number	Decay Per Cycle	Ref.
VS_4/G	74.5	0.5 C	1050	500	0.06%	This work
		2 C	810	2500	0.021%	
Capsule	63	0.5C	893	300	0.25%	1
Al_2O_3/G	55	1 C	1333	450	0.10%	2
Mesoporous Carbon	60	2 C	800	500	0.062%	3
MoS_2	65	0.5 C	800	600	0.083%	4
NbC	66.6	0.5 C	1082	150	0.29%	5
TiO_2-CB	60	0.5 C	1200	500	0.12%	6
CNF	50	0.2 C	1000	60	6%	7
SWCNT	70	0.2 C	718	150	0.11%	8
Functionalized CNT	70	0.5 C	1050	400	0.11%	9

Reference

1. H.-J. P. Jin Xie, Jia-Qi Huang, Wen-Tao Xu, Xiang Chen, and Qiang Zhang, Angew.Chem. Int.Ed. 2017, 56,16223 –16227
2. J.-Y. Hwang, H. M. Kim, S. Shin and Y.-K. Sun, *Advanced Functional Materials*, 2018, 28, 1704294
3. S. H. Chung and A. Manthiram, *Advanced materials*, 2014, **26**, 7352-7357.
4. Z. A. Ghazi, X. He, A. M. Khattak, N. A. Khan, B. Liang, A. Iqbal, J. Wang, H. Sin, L. Li and Z. Tang, *Advanced materials*, 2017, 29, 1606817.
5. G. L. W.L Cai, K.L Zhang, G.N Xiao, C Wang, K.F Ye, Y. Z. Z.W Chen, and Yitai Qian, *Advanced Functional Materials,* 2018, 28, 1704865
6. G. Xu, Q.-b. Yan, S. Wang, A. Kushima, P. Bai, K. Liu, X. Zhang, Z. Tang and J. Li, *Chemical Science*, 2017, **8**, 6619-6625.
7. X. Yu, J. Joseph and A. Manthiram, *Materials Horizons*, 2016, **3**, 314-319.
8. C. H. Chang, S. H. Chung and A. Manthiram, *Small*, 2016, **12**, 174-179.
9. R. Ponraj, A. G. Kannan, J. H. Ahn, J. H. Lee, J. Kang, B. Han and D. W. Kim, *ACS applied materials & interfaces*, 2017, **9**, 38445-38454.