Supporting Information

Guar Gum as Novel Aqueous Binder for Sulfur Composite Cathodes in Rechargeable Lithium Batteries

Qinyu Li[a], Huijun Yang [a],Lisheng Xie[a,b], Jun Yang [a], Yanna Nuli [a], Jiulin Wang[a*]

[a.] *Shanghai Electrochemical Energy Devices Research Center, Department of Chemical Engineering, Shanghai Jiao Tong University, Shanghai 200240, China, email: wangjiulin@sjtu.edu.cn*

[b.] *College of Environmental and Chemical Engineering, Yanshan University, Hebei 066004, China*

Experimental section

Sample preparation

The sulfur composite materials used in this work was mainly synthesized by two steps, including the ball milling and succedent heat treating at 300 °C in nitrogen for 450 min of elemental sulfur and polyacrylonitrile (PAN, Aldrich) at the mass ratio of 8:1.

Electrode fabrication

Preparation process of electrodes: S@pPAN with 42.3%wt S was mixed with Super P and GG binder in deionized water in the ratio of 8:1:1 to get slurry. For comparison, sulfur-based composite, carbon black and other binders, like CMC and PVDF, with a weight ratio of 8:1:1, were mixed in deionized water or N-methyl-2-pyrrolidone (NMP) solvent to get slurry. Then the slurry was coated onto a carbon coated aluminum foil and dried in vacuum for 12 hours at 60°C. After the dispersant was evaporated, the electrode film was cut to sheets with 12 mm in diameter and then dried at 70 ^{o}C under vacuum for 12 h. The electrode thickness is ca. 100μm and the cathode load is about 1 mg cm^{-2}.

Coin-cell assembly

Assembly of batteries: The CR2016-type coin cells were assembled in an argon-filled glove box using the above electrode as cathode, pure lithium foil as anode, and Celgard2300 film as separator. The electrolyte solution was 1 M $LiPF_6$ in ethylene carbonate (EC) / dimethyl carbonate (DMC) (1:1 by volume).

Physical characterizations

Fourier transform infrared (FTIR) spectra were collected on a Paragon 1000 spectropho-tometer (Perkin-Elmer, Inc., USA). The morphologies of the S@pPAN cathodes with various binders before and after charge-discharge tests were observed using scanning electron microscopy (Nova Nano SEM 450, FEI company, USA).

Electrochemical tests

The discharge/charge tests were carried out galvanostatically at 0.2C with the cell testing machine LAND-CT 2001A Cell Test System (Wuhan, China) between 1.0~3.0 V (vs. Li/Li^{+}) under room temperature. The rate performance of cells was evaluated using the same equipment at 1C, 3C, 5C, 7C, 9C, 10C and 1C, respectively. Electrochemical impedance spectroscopy (EIS) was measured with a CHI604D Electrochemical Workstation at the full-charged state over a frequency range from 100 kHz to 0.01 Hz with an AC amplitude of 5 mV. All experiments were conducted

under room temperature.
Cyclic voltammetry (CV) measurement was performed on CHI604A Electrochemical Workstation (Shanghai, China) at a scanning rate of 0. 5 mVs^{-1}.

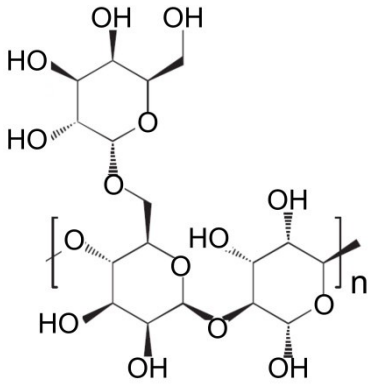

Fig. S1 Chemical structures of Guar Gum (GG)

Reference

1. T. T. Reddy and S. Tammishetti, *Polym. Degrad. Stabil.*, 2004, **86**, 455-459

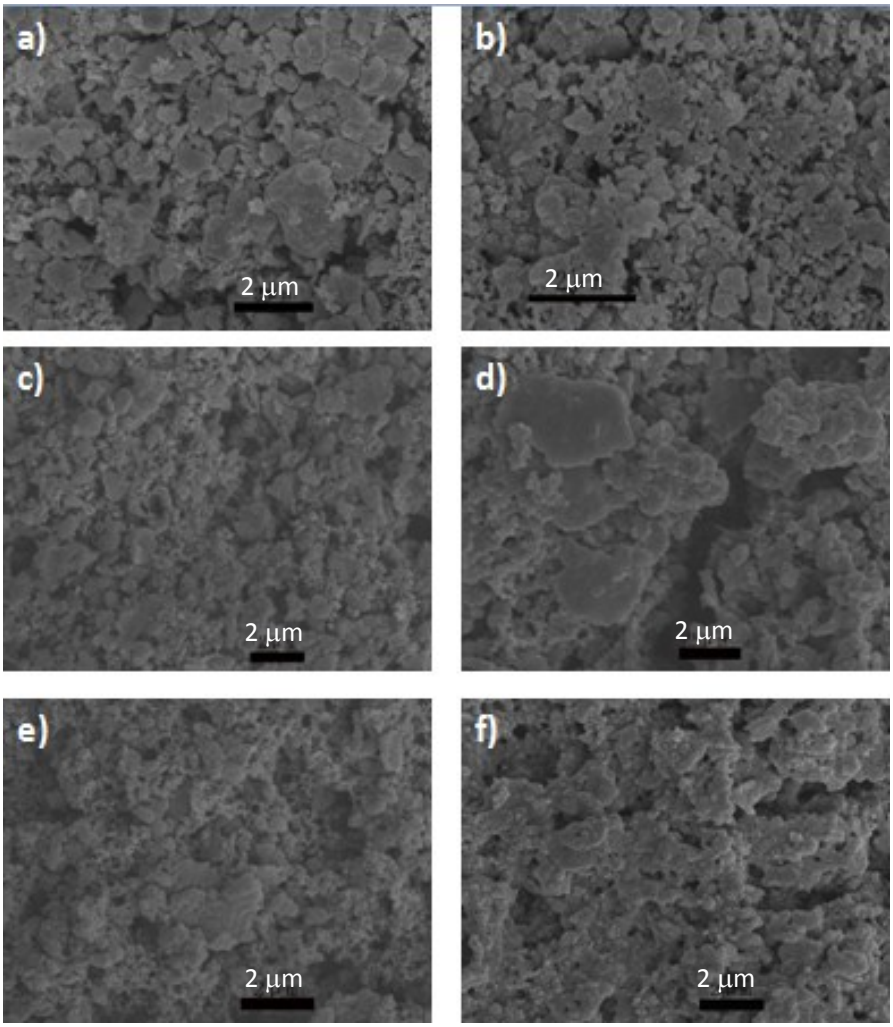

Fig. S2 Morphologies of the S@pPAN cathodes with GG binder as prepared (a) and after100 cycles (b), S@pPAN/PVDF as prepared (c) and after 100 cycles (d), S@pPAN/CMC as prepared (e) and after 100 cycles (f).

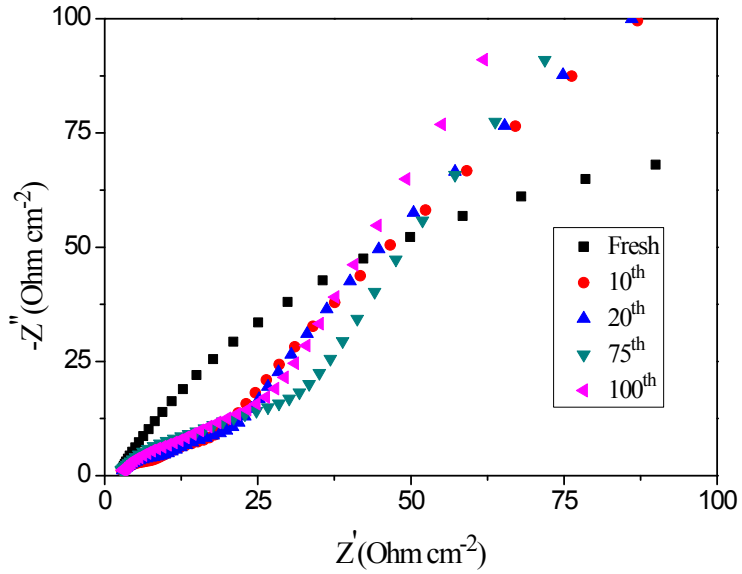

Fig. S3 Nyquist plots for the S@pPAN cathodes with PVDF binder.

Tab. S1 The fitted EIS data of cells with various binders in different states

Cycle Number		Fresh	10^{th}	20^{th}	75^{th}	100^{th}
$R_e(\Omega)$	Guar gum	4.266	5.024	3.455	3.414	3.583
	PVDF	3.959	3.979	4.084	3.647	3.893
$R_f(\Omega)$	Guar gum	34.74	15.33	15.72	12.73	12.77
	PVDF	168.13	26.56	24.62	32.69	26.73

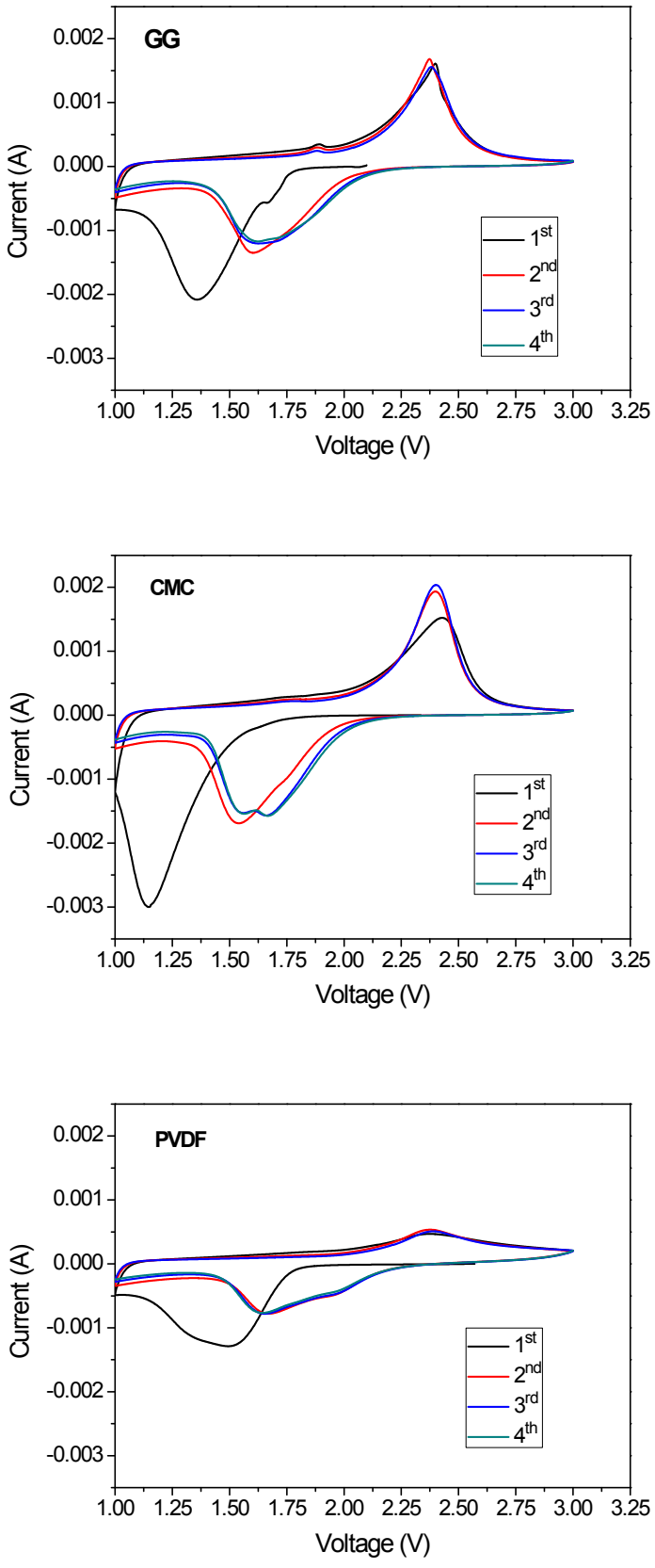

Fig. S4 CV spectra of the cathodes with various binders.

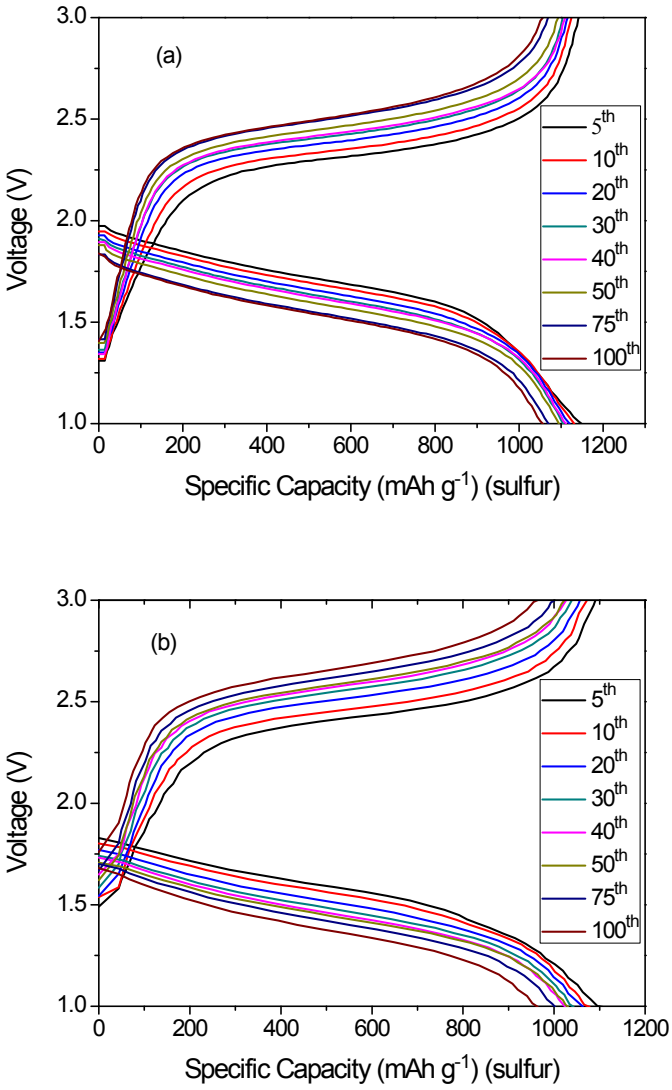

Fig. S5 The charge–discharge profiles of the S@pPAN with GG binder at 3C (a) and 7C (b)

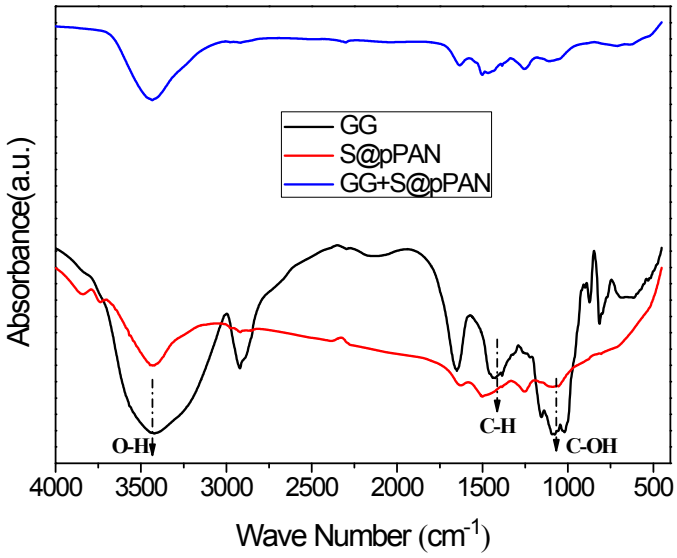

Fig. S6 FTIR images of GG, S@pPAN and S@pPAN/GG electrode.

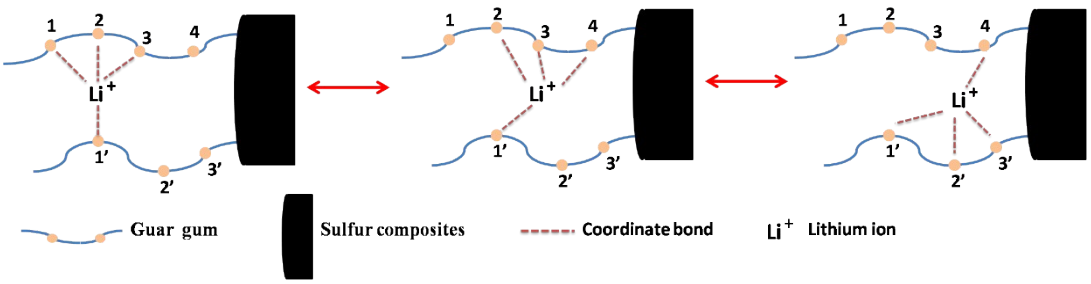

Fig.S7 Schematic lithium-ion transfer in the Guar gum binder.

Reference

1. J. Liu, Q. Zhang, T. Zhang, J.-T.Li, L. Huang and S.-G. Sun, *Adv. Funct. Mater.*, 2015, **25**, 3599-3605

Tab. S2 Comparison of the electrochemical performances for sulfur electrodes fabricated by recently published novel binders with S/C materials.

Sulfur material	Electrode composition (active materials: conductive additives: binder)	Type of binder	Cycliability ($mAhg^{-1}$)		Rate performance ($mAhg^{-1}$)	Reference
			1^{rt}	50^{th}		
S-KB (80wt% S)	8:1:1	LA132	1169	885	—	Ref.1
S-CNF (75.7wt%S)	7:2:1	CMC+SBR	1313	610	—	Ref.2
S-MPC (70wt% S)	7:2:1	SiO_2-impregnated polymer blend	1380	1100	1200（0.2C）→ 420（5C）	Ref.3
S-C (70wt%S)	7:1:2	P(VDF-TPFE)	1200	895	—	Ref.4
S-C (58wt%S)	8:1:1	PS(DCP-PEG)/Li^+	1000	905	1150（0.2C）→ 690（3C）	Ref.5
S-C (55wt%S)	8:0:2	Gum arabic (GA)	1386	1090	1250（0.2C）→ 800（5C）	Ref.6
S@pPAN (45wt%S)	8:1:1	Carbonyl-β -Cyclodextrin	1543	1456	—	Ref.7
Elemental Sulfur	6:3:1	CMC+SBR	870	660	—	Ref.8
Elemental Sulfur	5:4:1	PEO+PVP	1380	1050	—	Ref.9
Elemental Sulfur	63:7:30	Geltin	1132	408	—	Ref.10
Elemental Sulfur	55:35:10	Na-alginate	776	508	—	Ref.11
Elemental Sulfur	6:3:1	PAA	758	325	—	Ref.12

Reference:

1. X. Hong, J. Jin, Z. Wen, S. Zhang, Q. Wang, Chen Shen and K. Rui, *J. Power Sources*, 2016, **324**, 455-461.
2. M. Rao, X. Song, H. Liao and E.J. Cairns, *Electrochim. Acta,* 2012,**65**, 228-233.
3. G. Li, W. Cai, B. Liu, Z. Li, *J. Power Sources*, 2015, **294**, 187-192.

4. H. Wang, V. Sencadas , G. Gao, H. Gao, A. Du, H. Liu and Z.Guo, *Nano Eng*,2016,**26**, 722-728
5. Y.-J. Zhong,Z. Liu,X. Zheng, S.-L. Luo, N. -Yi Yuan,*Solid State Ionics,* 2016, **289**, 23-27.
6. G. Li, M. Ling, Y. Ye, Z. Li, J. Guo, Y. Yao, J. Zhu, Z. Lin and S. Zhang, *Adv. Energy Mater,*2015, **5** ,1500878
7. J. Wang, Z. Yao, C. W. Monroe, J. Yang and Y. Nuli, *Adv. Funct. Mater.*, 2013, **23**, 1194-1201.
8. M. He, L.-X. Yuan, W.-X. Zhang, X.-L. Hu and Y.-H. Huang, *J. Phy. Chem. C*, 2011, **115**, 15703-15709.
9. M. -J. Lacey, F. Jeschull, K. Edström and D. Brandell, *J. Power Sources*, 2014, **264**, 8-14.
10. J. Sun, Y. Huang,W. Wang, Z. Yu, A. Wang and K. Yuan, *Electrochim. Acta,* 2008, **53**, 7084-7088.
11. W. Bao, Z. Zhang, Y.Gan, X.Wang and J.Li, *J. Energ.Chem*,2013, **22**, 790-794.
12. Z. Zhang, W, Bao, H.Lu, M.Jia, K. Xie, Y. Lai and J.Li, *J.Electrochem. Soc.*, 2012, **1**, A34-A37.