Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A. This journal is © The Royal Society of Chemistry 2022

Supporting Information

Composite gel-polymer electrolyte for high-loading polysulfide cathodes

Li-Ling Chiu<sup>a</sup> and Sheng-Heng Chung<sup>a,b,\*</sup>

<sup>a</sup> Department of Materials Science and Engineering, National Cheng Kung University,

No. 1, University Road, Tainan City 701

<sup>b</sup> Hierarchical Green-Energy Materials Research Center, National Cheng Kung

University, No. 1, University Road, Tainan City 701

\*Sheng-Heng Chung: <u>SHChung@gs.ncku.edu.tw</u>

To whom correspondence should be addressed

## **Supporting Figures**



**Fig. S1.** Electrolyte affinity analysis: the contact angle of (a,c) polypropylene substrate and (b,d) PMMA-based GPE.



**Fig. S2.** Fourier-transform infrared (FTIR) spectroscopy of polymethyl methacrylate (PMMA)-based gel-polymer electrolyte (GPE) with different PMMA:LiTFSI ratios of 100:0, 95:5, 90:10, 85:15, and 80:20 wt% and the reference polymeric separator.



Fig. S3. Morphology analysis of PMMA-based GPE with different PMMA:LiTFSI ratios: (a) 100:0, (b) 95:5, (c) 90:10, (d) 85:15, and (e) 80:20 wt%.



**Fig. S4.** Solid-state NMR of PMMA-based GPE with different PMMA:LiTFSI ratios of 100:0, 95:5, 90:10, 85:15, and 80:20 wt%: (a) <sup>1</sup>H NMR and (b) <sup>7</sup>Li NMR.



**Fig. S5.** Discharge/charge curves of high-sulfur-loading polysulfide cathodes with PMMA-based GPE with different PMMA:LiTFSI ratios: (a) 100:0, (b) 95:5, (c) 90:10, (d) 85:15, and (e) 80:20 wt%.



**Fig. S6.** Microstructural inspection from the anode side of PMMA-based GPE (a) before and (b) after cycling.



Fig. S7. Discharge/charge curves of high-sulfur-loading polysulfide cathodes with PMMA-based GPE (PMMA:LiTFSI = 90:10 wt%): (a) 4 mg cm<sup>-2</sup> sulfur, (b) 8 mg cm<sup>-2</sup> sulfur, and (c) 10 mg cm<sup>-2</sup> sulfur; and (d) rate capacity of sulfur loading of 10 mg cm<sup>-2</sup>.



Fig. S8. Ionic conductivities of PMMA-based GPE before and after soaking electrolyte.



**Fig. S9.** Cyclic voltammetry (CV) analysis of high-sulfur-loading polysulfide cathodes with 4 mg cm<sup>-2</sup> sulfur and with PMMA-based GPE (PMMA:LiTFSI = 90:10 wt%) at different scan rates: (a) 0.010 mV s<sup>-1</sup>, (b) 0.015 mV s<sup>-1</sup>, (c) 0.020 mV s<sup>-1</sup>, and (d) 0.025 mV s<sup>-1</sup> and (e) lithium-ion diffusion coefficient analysis.



**Fig. S10.** CV analysis of high-sulfur-loading polysulfide cathodes with 8 mg cm<sup>-2</sup> sulfur and with PMMA-based GPE (PMMA:LiTFSI = 90:10 wt%) at different scan rates: (a)  $0.010 \text{ mV s}^{-1}$ , (b)  $0.015 \text{ mV s}^{-1}$ , (c)  $0.020 \text{ mV s}^{-1}$ , and (d)  $0.025 \text{ mV s}^{-1}$  and (e) lithiumion diffusion coefficient analysis.



**Fig. S11.** CV analysis of high-sulfur-loading polysulfide cathodes with 10 mg cm<sup>-2</sup> sulfur and with PMMA-based GPE (PMMA:LiTFSI = 90:10 wt%) at different scan rates: (a) 0.010 mV s<sup>-1</sup>, (b) 0.015 mV s<sup>-1</sup>, (c) 0.020 mV s<sup>-1</sup>, and (d) 0.025 mV s<sup>-1</sup> and (e) lithium-ion diffusion coefficient analysis.

## Supporting Table

Table S1. Comparative of battery performance and electrochemical abilities with gel-

A	В	C	D	E	F	G	Н
2016	60	1.8	PAN/PMMA-	1.43	475.2	3.0	[S1]
			electrospun/PETEA				
2016	56	1.5	PETEA	0.79	296.6	1.7	[S2]
2016	34	1	PVDF/PMMA/PVDF	1.17	399	2.5	[S3]
2017	82	5.2	PVDF/PEO/ZrO <sub>2</sub>	4.57	720.8	9.6	[S4]
2017	48	2	PVDF-HFP/LATP	1.8	440.6	3.9	[S5]
2018	70	1	PDA-modified PVDF-	1.22	850.8	2.6	[S6]
			based GPE				
2018	49	1.2	poly(acrylic acid)	0.67	275.4	1.4	[S7]
			(PAA)				
2019	48	1	PMMA/separator/AB	0.99	477.4	2.1	[S8]
2019	49	0.55	PP(Celgard	0.61	539.1	1.3	[S9]
			2400)/PMMA				
2019	38	1	PVDF/PMMA	0.55	210.4	1.2	[S10]
2019	54	1	PAN/PEO/LATP	0.90	487.9	1.9	[S11]
2019	48	1.2	PVDF-HFP/PETT	0.72	288.5	1.5	[S12]
2020	48	2	PETT-DA, PVDF-HFP	1.41	338.2	3.0	[S13]
2020	64	2	PEGDA/LLZTO	2.40	768.6	5.0	[S14]
2020	65	2	PVDF-HFP/PSD	2.46	798.9	5.2	[S15]

polymer electrolyte in lithium-sulfur batteries.

2020	60	1.1	PVDF-HFP	1.1	600	2.3	[S16]
2020	70	2	PVDF/PSPEG	1.89	660.1	4.0	[S17]
2021	70	1	PEI/PEGME	0.94	658	2.0	[S18]
2021	76	6	PEO/LiTFSI	3.82	484.1	8.0	[S19]
2021	60	1.5	PVA-CN or PDXL	1.59	637.8	3.3	[S20]
	73	10	PMMA/LiTFSI	7.10	509.8	15.1	This
							work

A: Year, B: sulfur content [wt%], C: sulfur loading [mg cm<sup>-2</sup>], D: polymer, E: areal capacity [mA·h cm<sup>-2</sup>], F: gravimetric capacity [mA·h g<sup>-1</sup>], G: energy density [mW·h cm<sup>-2</sup>], H: references.

## **Supporting References**

- S1. M. Liu, H. R. Jiang, Y. X. Ren, D. Zhou, F. Y. Kang and T. S. Zhao, *Electrochim. Acta*, 2016, **213**, 871-878.
- M. Liu, D. Zhou, Y.-B. He, Y. Fu, X. Qin, C. Miao, H. Du, B. Li, Q.-H. Yang, Z. Lin, T. S. Zhao and F. Kang, *Nano Energy*, 2016, 22, 278-289.
- W. Yang, W. Yang, J. Feng, Z. Ma and G. Shao, *Electrochim. Acta*, 2016, 210, 71-78.
- S4. S. Gao, K. Wang, R. Wang, M. Jiang, J. Han, T. Gu, S. Cheng and K. Jiang, J. Mater. Chem. A, 2017, 5, 17889-17895.
- S5. Y. Xia, X. Wang, X. Xia, R. Xu, S. Zhang, J. Wu, Y. Liang, C. Gu and J. Tu, *Chem. Eur. J.*, 2017, 23, 15203-15209.
- S6. D.-D. Han, S. Liu, Y.-T. Liu, Z. Zhang, G.-R. Li and X.-P. Gao, J. Mater. Chem. A, 2018, 6, 18627-18634.
- S7. S. Song, L. Shi, S. Lu, Y. Pang, Y. Wang, M. Zhu, D. Ding and S. Ding, *J. Membr. Sci.*, 2018, 563, 277-283.
- S8. D. Yang, L. He, Y. Liu, W. Yan, S. Liang, Y. Zhu, L. Fu, Y. Chen and Y. Wu, J. Mater. Chem. A, 2019, 7, 13679-13686.
- S9. C. Deng, Z. Wang, S. Wang, J. Yu, D. J. Martin, A. K. Nanjundan and Y. Yamauchi, ACS Appl. Mater. Interfaces, 2019, 11, 541-549.
- S10. Y. Liu, D. Yang, W. Yan, Q. Huang, Y. Zhu, L. Fu and Y. Wu, *iScience*, 2019, 19, 316-325.
- S11. X. Wang, X. Hao, Y. Xia, Y. Liang, X. Xia and J. Tu, J. Membr. Sci., 2019, 582, 37-47.
- S12. Y. Xia, Y. F. Liang, D. Xie, X. L. Wang, S. Z. Zhang, X. H. Xia, C. D. Gu and J.
  P. Tu, *Chem. Eng. J.*, 2019, **358**, 1047-1053.

- S13. X. Wang, X. Hao, D. Cai, S. Zhang, X. Xia and J. Tu, *Chem. Eng. J.*, 2020, **382**, 122714.
- S14. D. Shao, L. Yang, K. Luo, M. Chen, P. Zeng, H. Liu, L. Liu, B. Chang, Z. Luo and X. Wang, *Chem. Eng. J.*, 2020, **389**, 124300.
- S15. J.-H. Jiang, A.-B. Wang, W.-K. Wang, Z.-Q. Jin and L.-Z. Fan, J. Energy Chem., 2020, 46, 114-122.
- S16. J. Yu, S. Liu, G. Duan, H. Fang and H. Hou, Compos. Commun., 2020, 19, 239-245.
- S17. Y.-Q. Shen, F.-L. Zeng, X.-Y. Zhou, A.-b. Wang, W.-k. Wang, N.-Y. Yuan and J.-N. Ding, J. Energy Chem., 2020, 48, 267-276.
- S18. T. Zhang, J. Zhang, S. Yang, Y. Li, R. Dong, J. Yuan, Y. Liu, Z. Wu, Y. Song, Y. Zhong, W. Xiang, Y. Chen, B. Zhong and X. Guo, ACS Appl. Mater. Interfaces, 2021, 13, 44497-44508.
- S19. L.-L. Chiu and S.-H. Chung, Polymers, 2021, 13, 535.
- S20. Y.-J. Yang, R. Wang, J.-X. Xue, F.-Q. Liu, J. Yan, S.-X. Jia, T.-Q. Xiang, H. Huo, J.-J. Zhou and L. Li, *J. Mater. Chem. A*, 2021, 9, 27390-27397.