Supporting Information for

Comprehensive Design of the High Sulfur-Loading Li-S Battery

Based on MXene Nanosheets

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Supplementary Figures and Tables



Fig. S1 TGA curves of the KB/S and KB/S@Ti₃C₂T_x



Fig. S2 SEM image and corresponding elemental mapping analysis of the KB/S@Ti_3C_2T_x $\,$



Fig. S3 The TEM image and X-ray diffraction pattern of the $Ti_3C_2T_x$



Fig. S4 XRD patterns of the KB/S composite and the KB/S@Ti₃C₂T_x



Fig. S5 Charge/discharge curves of of KB/S at various rates ranging from 0.2 to 2 C



Fig. S6 Rate capability of KB/S electrode measured at different current densities from 0.2 to 2 C



Fig. S7 Zeta potential of Ti₃C₂T_x nanosheets, KB-PEI and KB@Ti₃C₂T_x



Fig. S8 N₂ adsorption-desorption isotherm of KB@Ti₃C₂T_x



Fig. S9 Photograph of Li_2S_6 adsorption by KB@Ti_3C_2T_x and KB in DME solution after 12 h $\,$

 Table S1 A comparison with the literatures about the thickness and areal mass of the interlayer in Li-S batteries

Interlayer	Areal mass (mg cm ⁻²)	Thickness (µm)	Performance	Refs.
PVDF-PEO/CB	0.6-0.7	10	751 mAh g^{-1} for 350 cycles, 0.3A g $^{-1}$.	[1]
NC@SA-Co and CNT-CNF hybrid network	0.45	10	787 mAh g ⁻¹ for 100 cycles, 0.5 C.	[2]
MoS ₂ /graphene	0.5	60	around 840 mAh g ⁻¹ in the initial 5 cycles, 1 A g ⁻¹ . 620 mAh g ⁻¹ for 200 cycles, 1 A g ⁻¹ .	[3]
RuO ₂ /mesoporous carbon	0.3	16	1276 mAh g ⁻¹ for the 1st cycle, 0.1 C. 665 mAh g ⁻¹ for the 300 cycles, 0.5 C.	[4]
CNT@ZIF	0.9	15	870 mAh g ⁻¹ for 100 cycles, 0.2 C.	[5]
KB@Ti ₃ C ₂ T _x	0.28	3	1281 mAh g^{-1} for the 1st cycle, 0.2 C. 1014 mAh g^{-1} for the 80 cycles, 0.2 C. 629 mAh g^{-1} for 400 cycles, 1 C.	This work



Fig. S10 The Li metal morphologies after cycling in different cells: (a) regular separator, (b) with KB@Ti₃C₂T_x coated separator.



Fig. S11 Nyquist plots of KB/S, KB/S@Ti₃C₂T_x electrodes with the pristine separators, and the KB/S@Ti₃C₂T_x electrode with the KB@Ti₃C₂T_x separator



Fig. S12 EIS spectra of the Li–S cell (a) KB/S electrode, (b)KB/S@Ti₃C₂T_x electrode and (c) KB/S@Ti₃C₂T_x electrode with KB@Ti₃C₂T_x coated separator after charging to 2.8 V, (d) and (e) Equivalent circuit for simulating the experimental data. The corresponding fitted resistances are listed in Table S2.

Sample	Cycle	$R_{sei}\left(\Omega ight)$	$R_{ct}\left(\Omega ight)$
	0	NO	38.3
	1st	97.7	54.6
KB/S electrode	5th	215.7	42.1
	10th	370.1	26.7
	20th	660.9	36.4
	0	NO	22.78
KB/S@TiaCaTy	1st	23.1	3.2
electrode	5th	16.0	3.8
	10th	14.4	0.9
	20th	13.67	1.9
KD/S@TieCeT	0	NO	21.62
electrode with	1st	36.1	2.8
$KB@Ti_3C_2T_x$ coated	5th	16.7	0.9
	10th	17.5	1.0
	20th	15.5	1.1

 Table S2 Corresponding simulated resistances from the EIS fitting

Table S3 Comparison of cycling performance with different cathode materi	als and
coating layers in Li-S batteries	

Cathode materials	Materials on Separators	Sulfur loading (mg cm ⁻²)	Electrochemical performance	Refs.
3D alkalized Ti ₃ C ₂ MXene nanoribbon/S	Ti ₃ C ₂	0.7-1	899 mAh g ⁻¹ for the 1st cycle, 0.5 C, 611 mAh g ⁻¹ for 50 cycles, 0.5 C.	[S6]
Carbon Black/S	Ti ₃ C ₂	1.2	743.7 mAh g^{-1} for the 1st cycle, 1 C, 495 mAh g^{-1} for 500 cycles, 1 C.	[S7]
CNTs/S	Ti ₃ C ₂ T _x /CNTs 10%	1.2	760 mAh g^{-1} for the 1st cycle, 1 C, 630 mAh g^{-1} for 200 cycles, 1 C.	[S8]
super P/S	Ti ₃ C ₂ nanosheet /glass fiber	1.9	820 mAh g ⁻¹ for the 1st cycle, 0.5 A/g, 721 mAh g ⁻¹ for 100 cycles, 0.5 A/g.	[S9]
KB/S	Ti ₃ C ₂ T _x eggshell	2.07	862 mAh g^{-1} for the 1st cycle, 1 C, 672 mAh g^{-1} for 150 cycles, 1 C.	[S10]
KB/S@Ti ₃ C ₂ T _x	KB@Ti ₃ C ₂ T _x	1.5	880 mAh g^{-1} for the 1st cycle,1 C, 629 mAh g^{-1} for 400 cycles, 1 C.	This work



Fig. S13 Long-term cycling performance at 1 C of the KB/S electrode with the KB@Ti₃C₂T_x separator

Table S4 Comparisons of the high-loading Li-S batteries with MXenes in the
electrodes or in the separators

Cathode	Separator	Sulfur loading (mg cm ⁻²)	Performance	Refs.
TiO ₂ QDs@ MXene/S	blank	5.5	533 mAh g ⁻¹ for 100 cycles, 0.2 C.	[S11]
KB/S	Ti ₃ C ₂ T _x eggshell	2.07	862 mAh g^{-1} for the 1st cycle, 1 C, 672 mAh g^{-1} for 150 cycles, 1 C.	[S12]
S/Carbon Black	Ti ₃ C ₂	2.8	850.9 mAh g ⁻¹ for 30 cycles, 0.2 C.	[S13]
Ti ₃ C ₂ /C-S	blank	2.5	475 mAh g^{-1} for the 200 cycles, 1 C.	[S14]
MNSs@d- Ti ₃ C ₂ /S	blank	3.7	474 mAh g ⁻¹ for 500 cycles, 1 C.	[S15]
KB/S@Ti ₃ C ₂ T _x	KB@ Ti ₃ C ₂ T _x	5.6	810 mAh g ⁻¹ (4.5 mAh cm ⁻¹)for the 1st cycle, 0.2 C, 600 mAh g ⁻¹ (3.4 mAh cm ⁻¹) for 100 cycles, 0.2 C.	This work

Supplementary References

- [S1] X. W. Fu, Y. Wang, L. S, W. H. Zhong, A polymeric nanocomposite interlayer as ion-transport-regulator for trapping polysulfides and stabilizing lithium metal. Energy Stor. Mater. 15, 447-457 (2018). https://doi.org/10.1016/j.ensm.2018.06.025
- [S2] Y. J. Li, P. Zhou, H. Li, T. T Gao, L. Zhou et al., A freestanding flexible singleatom cobalt-based multifunctional interlayer toward reversible and durable

lithium-sulfur batteries. Small Methods 4(3), 1900701 (2020). https://doi.org/10.1002/smtd.201900701

- [S3] P. Q. Guo, D. Q. Liu, Z. J. Liu, X. N. Shang, Q. M. Liu, D. Y. He, Dual functional MoS₂/graphene interlayer as an efficient polysulfide barrier for advanced lithium-sulfur batteries. Electrochim. Acta 256, 28-36 (2017). https://doi.org/10.1016/j.electacta.2017.10.003
- [S4] J. Balach, T. Jaumann, S. Mühlenhoff, J. Eckert, L. Giebeler, Enhanced polysulphide redox reaction using a RuO₂ nanoparticle-decorated mesoporous carbon as functional separator coating for advanced lithium–sulphur batteries. Chem. Comm. **52**(52), 8134-8137 (2016). https://doi.org/10.1039/C6CC03743G
- [S5] F. Wu, S. Y. Zhao, L. Chen, Y. Lu, Y. F. Su et al., Metal-organic frameworks composites threaded on the CNT knitted separator for suppressing the shuttle effect of lithium sulfur batteries. Energy Stor. Mater, 14, 383-391 (2018). https://doi.org/10.1016/j.ensm.2018.06.009
- [S6] Y. Dong, S. Zheng, J. Qin, X. Zhao, H. Shi, X. Wang, Z. S. Wu, All-MXenebased integrated electrode constructed by Ti₃C₂ nanoribbon framework host and nanosheet interlayer for high-energy-density Li–S batteries. ACS Nano 12(3), 2381-2388 (2018). https://doi.org/10.1021/acsnano.7b07672
- [S7] J. Song, D. Su, X. Xie, X. Guo, W. Bao, G. Shao, G. Wang, Immobilizing polysulfides with mxene-functionalized separators for stable lithium–sulfur batteries. ACS Appl. Mater. Interfaces 8(43), 29427-29433 (2016). https://doi.org/10.1021/acsami.6b09027
- [S8] N. Li, Y. Xie, S. Peng, X. Xiong, K. Han, Ultra-lightweight Ti₃C₂T_x MXene modified separator for Li–S batteries: Thickness regulation enabled polysulfide inhibition and lithium ion transportation. J. Energy Chem. 42, 116-125 (2020). https://doi.org/10.1016/j.jechem.2019.06.014
- [S9] C. Lin, W. Zhang, L. Wang, Z. Wang, W. Zhao, W. Duan, J. Jin, A few-layered Ti₃C₂ nanosheet/glass fiber composite separator as a lithium polysulphide reservoir for high-performance lithium–sulfur batteries. J. Mater. Chem. A 4(16), 5993-5998 (2016).https://doi.org/10.1039/C5TA10307J
- [S10] L. Yin, G. Xu, P. Nie, H. Dou, X. Zhang, MXene debris modified eggshell membrane as separator for high-performance lithium-sulfur batteries. Chem. Eng. J. 352, 695-703 (2018). https://doi.org/10.1016/j.cej.2018.07.063
- [S11] X. T. Gao, Y. Xie, X. D. Zhu, K. N. Sun, X. M. Xie, Y. T. Liu, B. Ding, Ultrathin MXene nanosheets decorated with TiO₂ quantum dots as an efficient sulfur host toward fast and stable Li–S batteries. Small 14(41), 1802443 (2018).https://doi.org/10.1002/smll.201802443
- [S12] L. Yin, G. Xu, P. Nie, H. Dou, X. Zhang, MXene debris modified eggshell

membrane as separator for high-performance lithium-sulfur batteries. Chem. Eng. J. **352**, 695-703 (2018).https://doi.org/10.1016/j.cej.2018.07.063

- [S13] J. Song, D. Su, X. Xie, X. Guo, W. Bao, G. Shao, G. Wang, Immobilizing polysulfides with mxene-functionalized separators for stable lithium–sulfur batteries. ACS Appl. Mater. Interfaces 8(43), 29427-29433 (2016). https://doi.org/10.1021/acsami.6b09027
- [S14] H. Y. Zhou, Z. Y. Sui, K. A, L. W. Lin, H. Y. Wang, B. H. Han, Investigating the electrocatalysis of Ti₃C₂/carbon hybrid in polysulfide conversion of lithium–sulfur batteries. ACS Appl. Mater. Interfaces 25, 547-554 (2020). https://doi.org/10.1021/acsami.9b23006
- [S15] H. Zhang, Q. Qi, P. G. Zhang, W. Zheng, J. Chen et al., Self-assembled 3D MnO₂ nanosheets@delaminated-Ti₃C₂ aerogel as sulfur host for lithium–sulfur battery cathodes. ACS Appl. Energy Mater. 2(1), 705-714 (2018).______ https://doi.org/10.1021/acsaem.8b01765