

Supporting information

Biao Chen,^{a,b†} Simi Sui,^{a,c†} Fang He,^{a,*} Chunnian He,^{a,b,d} Hui-Ming Cheng,^{e,f} Shi-Zhang Qiao,^{g,*} Wenbin Hu,^{a,b,d,*} and Naiqin Zhao^{a,b}

^a School of Materials Science and Engineering, Tianjin Key Laboratory of Composite and Functional Materials, Key Laboratory of Advanced Ceramics and Machining Technology (Ministry of Education), Tianjin University, Tianjin, 300350, People's Republic of China

^b National Industry-Education Platform of Energy Storage, Tianjin University, 135 Yaguan Road, Tianjin 300350, People's Republic of China

^c Tianjin Key Laboratory of Materials Laminating Fabrication and Interface Control Technology, School of Materials Science and Engineering, Hebei University of Technology, Tianjin, 300401, People's Republic of China

^d Joint School of National University of Singapore and Tianjin University, International Campus of Tianjin University, Binhai New City, Fuzhou, 350207, People's Republic of China

^e Faculty of Materials Science and Energy Engineering/Institute of Technology for Carbon Neutrality, Shenzhen Institutes of Advanced Technology, Chinese Academy of Sciences, Shenzhen, 518055, People's Republic of China

^f Shenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, Shenyang, 110016, People's Republic of China

^g School of Chemical Engineering and Advanced Materials, The University of Adelaide, Adelaide, South Australia, 5005, Australia

† These authors are equal major contributors

* Corresponding authors

Table S1. Summary of the electrochemical performance of TMDC/G heterostructures applied in SCs modified by different interfacial strategies.
N.A. indicates not available.

Samples	Specific Capacitance	Capacitance Retention	Energy density (Power density)	Ref.
Interfacial doping model				
3D MoS ₂ /NG aerogel	532 F g ⁻¹ (1 A g ⁻¹)	93.6% (10000 cycles at 10 A g ⁻¹)	17.24 Wh kg ⁻¹ (500 W kg ⁻¹)	1
MoS ₂ /NG	146 F g ⁻¹ (20 A g ⁻¹)	91.3% (1000 cycles at 2 A g ⁻¹)	N.A.	2
MoS ₂ /NSG	549.4 F g ⁻¹ (10 mV s ⁻¹)	93.2% (10000 cycles at 20 A g ⁻¹)	65.2 Wh kg ⁻¹ (900 W kg ⁻¹)	3
MoS ₂ /NPG	588 F g ⁻¹ (1 A g ⁻¹)	91.67% (5000 cycles at 3 A g ⁻¹)	24.34 Wh kg ⁻¹ (300 W kg ⁻¹)	4
MoS ₂ -O-G	265 F g ⁻¹ (10 mV s ⁻¹)	92% (1000 cycles)	63 Wh kg ⁻¹	5
Interfacial stacking model				
MoS ₂ /G superlattice	2483 F g ⁻¹ (10 A g ⁻¹)	92% (8000 cycles at 10 A g ⁻¹)	N.A.	6
MoS ₂ /G superlattice	290 F cm ⁻³ (0.5 A g ⁻¹)	90% (10000 cycles at 0.5 A g ⁻¹)	N.A.	7
Interfacial orientation model				
V-MoS ₂ /rGO fiber	330 F g ⁻¹ (0.1 A g ⁻¹)	95% (10000 cycles, 10 A g ⁻¹)	69.44 μWh cm ⁻² (0.5 mW cm ⁻²)	8
V-MoS ₂ /G	2182.33 mF cm ⁻² (1 mA cm ⁻²)	116.83% (5000 cycles at 10 mA cm ⁻²)	130.34 Wh m ⁻² (4000 W m ⁻²)	9
V-MoS ₂ /G	428 F g ⁻¹ (1 A g ⁻¹)	88% (5000 cycles at 6 A g ⁻¹)	36.5 Wh kg ⁻¹ (9 W kg ⁻¹)	10

Table S2. Summary of the electrochemical performance of TMDC/G heterostructures applied in ZIBs modified by different interfacial strategies. N.A. indicates not available.

Samples	Interlayer of TMDC (nm)	Specific Capacitance	Specific Capacitance	Capacitance Retention	Energy density	Ref.
Interfacial doping model						
V-MoS ₂ /C	1.02	385 mAh g ⁻¹ (0.1 A g ⁻¹)	230 mAh g ⁻¹ (0.8 A g ⁻¹)	88% (150 cycles at 0.1 A g ⁻¹)	N.A.	11
Interfacial stacking model						
G/ML-MoS ₂ /G	0.7	202.6 mAh g ⁻¹ (0.1 A g ⁻¹)	104.5 mAh g ⁻¹ (4 A g ⁻¹)	98.6% (600 cycles at 1A g ⁻¹)	148.2Wh kg ⁻¹	12
hollow MoS ₂ /NC superlattice	0.96	247.8 mAh g ⁻¹ (0.1 A g ⁻¹)	100.9 mAh g ⁻¹ (8 A g ⁻¹)	85.6% (3200 cycles at 1A g ⁻¹)	72.6 Wh kg ⁻¹	13
MoS ₂ /G superlattice	1.16	285.4 mAh g ⁻¹ (0.05 A g ⁻¹)	141.6 mAh g ⁻¹ (5 A g ⁻¹)	88.2% (1800 cycles at 1A g ⁻¹)	157.5Wh kg ⁻¹	14
MoS ₂ /PEDOT superlattice	1.29	312.5 mAh g ⁻¹ (0.1 A g ⁻¹)	83.6 mAh g ⁻¹ (15 A g ⁻¹)	90.1% (4000 cycles at 5A g ⁻¹)	184.7Wh kg ⁻¹	15
MoS ₂ /CTAB superlattice	1	181.8 mAh g ⁻¹ (0.1 A g ⁻¹)	78.5 mAh g ⁻¹ (10 A g ⁻¹)	92.8% (2100 cycles at 10A g ⁻¹)	N.A.	16
Interfacial orientation model						
V-MoSSe/G	0.64	253.8 mAh g ⁻¹ (0.2 A g ⁻¹)	124.2 mAh g ⁻¹ (5 A g ⁻¹)	83% (1200 cycles at 2A g ⁻¹)	N.A.	17
V-VS ₂ /G	0.97	238 mAh g ⁻¹ (0.1 A g ⁻¹)	190 mAh g ⁻¹ (5 A g ⁻¹)	93% (1000 cycles at 5A g ⁻¹)	N.A.	18

Table S3. Summary of the electrochemical performance of TMDC/G heterostructures applied in AMSBs modified by different interfacial strategies. N.A. indicates not available.

Samples	Sulfur contents & sulfur loading mass	Electrochemical performance (initial capacity and degradation rate)	Rate Performance	Highest S loading	Voltage window (vs. Li/Li ⁺)	Ref.
Interfacial doping model						
Ni-MoS ₂ /rGO	68.4 wt% & 1 mg cm ⁻²	937.8 mAh g ⁻¹ at 0.5 C and 0.077% for 140 cycles	757.2 mAh g ⁻¹ at 2C	5.89 mg cm ⁻²	1.8–2.8 V	19
MoS ₂ /NG	~80% & 5.2 mg cm ⁻²	772 mAh g ⁻¹ at 0.2 C and 0.185% for 100 cycles	512 mAh g ⁻¹ at 3C	5.2 mg cm ⁻²	1.7–2.8 V	20
MoSe ₂ /NG	~62% & 1.1 mg cm ⁻²	1118 mAh g ⁻¹ at 0.2 C and 0.136% for 100 cycles	632 mAh g ⁻¹ at 1C	/	1.8–2.8 V	21
WSe ₂ /NG	1.2 mg cm ⁻²	1063 mAh g ⁻¹ at 0.2 C and 0.063% for 300 cycles (1C)	743.2 mAh g ⁻¹ at 6C	10.5 mg cm ⁻²	1.7–2.8 V	22
Co-MoS ₂ /G	1 mg cm ⁻²	~1100 mAh g ⁻¹ at 1 C and 0.029% for 1000 cycles	941 mAh g ⁻¹ at 2C	5.27 mg cm ⁻²	1.8–2.8 V	23
Interfacial stacking model						
NG/ReS ₂ /NG	63% & 1.4 ~ 1.6 mg cm ⁻²	854 mAh g ⁻¹ at 2 C and 0.064% for 800 cycles	810 mAh g ⁻¹ at 2C	6.4 mg cm ⁻²	1.7–2.8 V	24
WSe ₂ /G superlattice	73.8 wt%	923 mAh g ⁻¹ at 1 C and 0.0374% for 500 cycles	569.5 mAh g ⁻¹ at 5C	5.2 mg cm ⁻²	1.7–2.8 V	25
MoS ₂ /G superlattice	1.02 mg cm ⁻²	1232 mAh g ⁻¹ at 0.2 C and 0.08% for 200 cycles	593 mAh g ⁻¹ at 5C	3.8 mg cm ⁻²	1.7–2.8 V	26

Table S3. Summary of the electrochemical performance of TMDC/G heterostructures applied in AMSBs modified by different interfacial strategies. N.A. indicates not available (continued).

Samples	Sulfur contents & sulfur loading mass	Electrochemical performance (initial capacity and degradation rate)	Rate Performance	Highest S loading	Voltage window (vs. Li/Li ⁺)	Ref.
Interfacial orientation model						
V-MoS ₂ /G	79.1% & 5.6 mg cm ⁻²	1384 mAh g ⁻¹ at 0.1 C and 0.028% for 500 cycles	530 mAh g ⁻¹ at 5C	5.6 mg cm ⁻²	1.7–2.8 V	27
V-ReS ₂ /G	1 mg cm ⁻²	600 mAh g ⁻¹ at 1 C and 0.0825% for 400 cycles	577 mAh g ⁻¹ at 2C	2.2 mg cm ⁻²	1.7–2.8 V	28
V-MoSSe/G	4.2 mg cm ⁻²	1138 mAh g ⁻¹ at 1C and 0.044% for 1000 cycles	593 mAh g ⁻¹ at 5C	6.5 mg cm ⁻²	1.7–2.8 V	29

Table S4. Summary of the electrochemical performance of TMDC/G heterostructures applied as electrocatalysts modified by different interfacial strategies. N.A. indicates not available.

Samples	Type	Electrolytes	Overpotential at specific current density	Current density at specific overpotential	Tafel slope	Durability	Ref.
Interfacial doping model							
MoS ₂ -Fe-C	OER	6M KOH	155 mV at 10 mA cm ⁻²	N.A.	43 mV dec ⁻¹	120 h	30
MoS ₂ /NSG	OER	1M KOH	360 mV at 10 mA cm ⁻²	N.A.	98.2 mV dec ⁻¹	10000 cycles	31
MoS ₂ /NG	ORR	0.1M KOH	onset potential +0.95 V	peak potential +0.82 V	N.A.	10000s	32
MoS ₂ /NG	ORR	0.1M KOH	onset potential -0.12 V	peak potential -0.23 V	N.A.	N.A.	33
Interfacial orientation model							
V-ReS ₂ /G	ORR	0.1M KOH	onset potential 0.82 V vs RHE	half wave potential 0.635 V	76 mV dec ⁻¹	3000 s	34
V-ReS ₂ /NG	ORR	0.1M KOH	N.A.	diffusion-limited current density 5.58 mA cm ⁻²	76 mV dec ⁻¹	3000 s	35
V-MoS ₂ /NC	CO ₂ RR	0.5 M H ₂ SO ₄	onset potential ~40 mV	34.31 mA cm ⁻² at 590 mV	N.A.	24 h	36

REFERENCES

1. Y. Yuan, H. Lv, Q. Xu, H. Liu and Y. Wang, *Nanoscale*, 2019, **11**, 4318-4327.
2. B. Xie, Y. Chen, M. Yu, T. Sun, L. Lu, T. Xie, Y. Zhang and Y. Wu, *Carbon*, 2016, **99**, 35-42.
3. Z. Li, J. Zhao, Z. Yin, X. Wang, Z. Wu, D. Liu, M. Zhu and X. Wang, *Energy & Fuels*, 2021, **35**, 2692-2703.
4. S. Zhao, W. Xu, Z. Yang, X. Zhang and Q. Zhang, *Electrochimica Acta*, 2020, **331**, 135265.
5. E. G. da Silveira Firmiano, A. C. Rabelo, C. J. Dalmaschio, A. N. Pinheiro, E. C. Pereira, W. H. Schreiner and E. R. Leite, *Advanced Energy Materials*, 2014, **4**, 1301380.
6. N. Feng, R. Meng, L. Zu, Y. Feng, C. Peng, J. Huang, G. Liu, B. Chen and J. Yang, *Nature Communications*, 2019, **10**, 1372.
7. Y. Zhuo, E. Prestat, I. A. Kinloch and M. A. Bissett, *ACS Applied Energy Materials*, 2022, **5**, 61-70.
8. T. Guan, Z. Cheng, Z. Li, L. Gao, K. Yan, L. Shen and N. Bao, *Industrial & Engineering Chemistry Research*, 2022, **61**, 3840-3849.
9. C. Han, Z. Tian, H. Dou, X. Wang and X. Yang, *Chinese Chemical Letters*, 2018, **29**, 606-611.
10. H. Sun, H. Liu, Z. Hou, R. Zhou, X. Liu and J.-G. Wang, *Chemical Engineering Journal*, 2020, **387**, 124204.
11. J. Xu, Z. Dong, K. Huang, L. Wang, Z. Wei, L. Yu and X. Wu, *Scripta Materialia*, 2022, **209**, 114368.
12. H. Li, Q. Yang, F. Mo, G. Liang, Z. Liu, Z. Tang, L. Ma, J. Liu, Z. Shi and C. Zhi, *Energy Storage Materials*, 2019, **19**, 94-101.
13. C. Li, C. Liu, Y. Wang, Y. Lu, L. Zhu and T. Sun, *Energy Storage Materials*, 2022, **49**, 144-152.
14. S. Li, Y. Liu, X. Zhao, Q. Shen, W. Zhao, Q. Tan, N. Zhang, P. Li, L. Jiao and X. Qu, *Advanced Materials*, 2021, **33**, 2007480.
15. S. Li, C. Huang, L. Gao, Q. Shen, P. Li, X. Qu, L. Jiao and Y. Liu, *Angewandte Chemie International Edition*, 2022, **n/a**, e202211478.
16. Z. Yao, W. Zhang, X. Ren, Y. Yin, Y. Zhao, Z. Ren, Y. Sun, Q. Lei, J. Wang, L. Wang, T. Ji, P. Huai, W. Wen, X. Li, D. Zhu and R. Tai, *ACS Nano*, 2022, **16**, 12095-12106.
17. H. Li, B. Chen, R. Gao, F. Xu, X. Wen, X. Zhong, C. Li, Z. Piao, N. Hu, X. Xiao, F. Shao, G. Zhou and J. Yang, *Nano Research*, 2022, DOI: 10.1007/s12274-022-5108-6.
18. T. Chen, X. Zhu, X. Chen, Q. Zhang, Y. Li, W. Peng, F. Zhang and X. Fan, *Journal of Power Sources*, 2020, **477**, 228652.
19. R. Zhang, Y. Dong, M. A. Al-Tahan, Y. Zhang, R. Wei, Y. Ma, C. Yang and J. Zhang, *Journal of Energy Chemistry*, 2021, **60**, 85-94.
20. B. Yu, Y. Chen, Z. Wang, D. Chen, X. Wang, W. Zhang, J. He and W. He, *Journal of Power Sources*, 2020, **447**, 227364.
21. H. Wong, X. Ou, M. Zhuang, Z. Liu, M. D. Hossain, Y. Cai, H. Liu, H. Lee, C.-Z. Wang and Z. Luo, *ACS Applied Materials & Interfaces*, 2019, **11**, 19986-19993.
22. P. Wang, F. Sun, S. Xiong, Z. Zhang, B. Duan, C. Zhang, J. Feng and B. Xi, *Angewandte Chemie International Edition*, 2022, **61**, e202116048.

23. W. Liu, C. Luo, S. Zhang, B. Zhang, J. Ma, X. Wang, W. Liu, Z. Li, Q.-H. Yang and W. Lv, *ACS Nano*, 2021, **15**, 7491-7499.
24. N. Wei, J. Cai, R. Wang, M. Wang, W. Lv, H. Ci, J. Sun and Z. Liu, *Nano Energy*, 2019, **66**, 104190.
25. C. Zhang, B. Fei, D. Yang, H. Zhan, J. Wang, J. Diao, J. Li, G. Henkelman, D. Cai, J. J. Biendicho, J. R. Morante and A. Cabot, *Advanced Functional Materials*, 2022, **32**, 2201322.
26. W. Li, D. Wang, Z. Song, Z. Gong, X. Guo, J. Liu, Z. Zhang and G. Li, *Nano Research*, 2019, **12**, 2908-2917.
27. B. Cui, X. Cai, W. Wang, P. Saha and G. Wang, *Journal of Energy Chemistry*, 2022, **66**, 91-99.
28. H. Liu, B. Chen, H. Qin, N. Wang, E. Liu, C. Shi and N. Zhao, *Applied Surface Science*, 2020, **505**, 144586.
29. H. Li, R. Gao, B. Chen, C. Zhou, F. Shao, H. Wei, Z. Han, N. Hu and G. Zhou, *Nano Letters*, 2022, DOI: 10.1021/acs.nanolett.2c01779.
30. F. Gong, M. Liu, L. Gong, S. Ye, Q. Jiang, G. Zeng, X. Zhang, Z. Peng, Y. Zhang, S. Fang and J. Liu, *Advanced Functional Materials*, 2022, **32**, 2202141.
31. Y. Yan, S. Liang, X. Wang, M. Zhang, S.-M. Hao, X. Cui, Z. Li and Z. Lin, *Proceedings of the National Academy of Sciences*, 2021, **118**, e21110036118.
32. C. Du, H. Huang, X. Feng, S. Wu and W. Song, *Journal of Materials Chemistry A*, 2015, **3**, 7616-7622.
33. K. Zhao, W. Gu, L. Zhao, C. Zhang, W. Peng and Y. Xian, *Electrochimica Acta*, 2015, **169**, 142-149.
34. Y. B. Kang, X. Han, S. Kim, H. Yuan, N. Ling, H. C. Ham, L. Dai and H. S. Park, *ACS Nano*, 2021, **15**, 5560-5566.
35. Y. Kang, X. Han, H. Yuan, B. Y. Xia and H. S. Park, *International Journal of Energy Research*, 2021, **45**, 19586-19596.
36. H. Li, X. Liu, S. Chen, D. Yang, Q. Zhang, L. Song, H. Xiao, Q. Zhang, L. Gu and X. Wang, *Advanced Energy Materials*, 2019, **9**, 1900072.