Electronic Supplementary Information

Unraveling Na-ion storage performance of vertically-aligned interlayer-expanded two-dimensional MoS₂@C@MoS₂ heterostructure

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Fig. S1. (a) XRD pattern and (b, c) FE-SEM images at different magnification of MoO₃ NRs.



Fig. S2. XRD patterns of various raming rate tuned samples (pristine MoO₂ NRs, 5 °C min⁻¹, 2 °C min⁻¹, 1 °C min⁻¹, pristine MoS₂ NRs).



Fig. S3. (a, b) FE-SEM images of MoO_2 NRs at different magnification.



Fig. S4. (a, b) FE-SEM images at different magnification of MoS_2 NRs at ramping rate of 5 °C min⁻¹.



Fig. S5. (a, b) FE-SEM images at different magnification of MoS_2 NRs at ramping rate of 2 °C min⁻¹.



Fig. S6. (a, b) FE-SEM images at different magnification of MoS_2 NRs at ramping rate of 1 °C min⁻¹.



Fig. S7. EDS spectra of various sulfurized samples obtained at ramping rate of (a) 5 °C min⁻¹, (b) 2 °C min⁻¹, (c) 1 °C min⁻¹ and (d) full MoS₂ NRs.



Fig. S8. (a) Cycling stability and (b) rate capability of MoO₃ NRs.



Fig. S9. (a) Cycling stability and (b) rate capability of MoS_2 NRs at ramping rate of 5 °C min⁻¹.



Fig. S10. (a) Cycling stability and (b) rate capability of MoS_2 NRs at ramping rate of 2 °C min⁻¹.



Fig. S11. (a) Cycling stability and (b) rate capability of MoS_2 NRs at ramping rate of 1 °C min⁻¹.



Fig. S12. (a) Cycling stability and (b) rate capability of MoO₂ NRs.

Electrode Material	Initial Coulombic efficiency (%)	Cycling stability	Rate capability	References
HMF-MoS ₂	60.2	384 mAh g ⁻¹ at 100 mA g ⁻¹ (After 100 cycles)	226 mAh g ⁻¹ at 5 A g ⁻¹	[1]
BD-MoS ₂	<75	354 mAh g ⁻¹ at 500 mA g ⁻¹ (After 1000 cycles)	262 mAh g ⁻¹ at 5 A g ⁻¹	[2]
Expanded MoS ₂ @ Carbon fiber	65.1	241 mAh g ⁻¹ at 1000 mA g ⁻¹ (After 700 cycles)	109 mAh g ⁻¹ at 10 A g ⁻¹	[3]
MoS ₂ @C paper	79.5	268 mAh g ⁻¹ at 0.08 A g ⁻¹ (after 100 cycles)	205 mAh g ⁻¹ at 1 A g ⁻¹	[4]
1T-MoS ₂	64	313 mAh g ⁻¹ at 50 mA g ⁻¹ (After 200 cycles)	175 mAh g ⁻¹ at 2 A g ⁻¹	[5]
MoS ₂ Nanoflowers	-	350 mAh g ⁻¹ at 50 mA g ⁻¹	300 mAh g ⁻¹ at 1 A g ⁻¹	[6]
MoS _{2-x} Se _x /GF	55	208 mAh g ⁻¹ at 0.2 A g ⁻¹ (78.6% after 500 cycles)	~115 mAh g ⁻¹ at 5 A g ⁻¹	[7]
MoS ₂ /RGO Sponge	67.4	372 mAh g ⁻¹ at 0.1 A g ⁻¹ (after 50 cycles)	192 mAh g ⁻¹ at 1 A g ⁻¹	[8]
MoS ₂ /RGO Sponge	64	420 mAh g ⁻¹ at 0.1 A g ⁻¹ (after 160 cycles)	284 mAh g ⁻¹ at 1 A g ⁻¹	[9]
S/MoS ₂	78.6	413.2 mAh g ⁻¹ at 0.1 A g ⁻¹ (after 100 cycles)	358.8 mAh g ⁻¹ at 0.5 A g ⁻¹	[10]
MoS ₂ @C@MoS ₂	86.3	434 mAh g ⁻¹ at 100 mA g ⁻¹ (After 100 cycles)	365 mAh g ⁻¹ at 4000 mA g ⁻¹	This work

Table S1. Comparison of $MoS_2@C@MoS_2$ performance with other MoS_2 based anodes.

Table S2. Comparison of $MoS_2@C@MoS_2$ performance with other metal sulfides/selenides based anodes.

Electrode Material	Initial Coulombic efficiency (%)	Cycling stability	Rate capability	Reference
NiS ₂ -rGO	65	313 mAh g ⁻¹ at 100 mA g ⁻¹ (after 200 cycles)	168 mAh g ⁻¹ at 2C rate	[11]
VS ₄ -rGO	75.1	240.8 mAh g ⁻¹ at 100 mA g ⁻¹ (after 50 cycles)	192.2 mAh g ⁻¹ at 800 mA g ⁻¹	[12]
SnS-C	79	260 mAh g ⁻¹ at 100 mA g ⁻¹ (after 300 cycles)	145 mAh g ⁻¹ at 1000 mA g ⁻¹	[13]
SnS-MWCNT	73.3	391 mAh g ⁻¹ at 100 mA g ⁻¹ (after 50 cycles)	410 mAh g ⁻¹ at 500 mA g ⁻¹	[14]
CoMoS ₃	77.7	411 mAh g ⁻¹ at 2 A g ⁻¹ (after 300 cycles)	349 mAh g ⁻¹ at 10 A g ⁻¹	[15]
CoSe ₂ @N- PGC/CNTs	75.6	424 mAh g ⁻¹ at 0.1 A g ⁻¹ (after 100 cycles)	368 mAh g ⁻¹ at 5 A g ⁻¹	[16]
Fe ₇ Se ₈ @NC	78.7	367 mAh g ⁻¹ at 0.5 A g ⁻¹ (after 100 cycles)	251 mAh g ⁻¹ at 2 A g ⁻¹	[17]
FeS/N-C	51.5	511 mAh g ⁻¹ at 0.2 A g ⁻¹ (after 100 cycles)	260 mAh g ⁻¹ at 4 A g ⁻¹	[18]
CoS ₂ @N,S- doped carbon	70	510 mAh g ⁻¹ at 0.1 A g ⁻¹ (after 100 cycles)	288 mAh g ⁻¹ at 2 A g ⁻¹	[19]
MoSe ₂ /N,P- doped rGO	69.6	337 mAh g ⁻¹ at 0.1 A g ⁻¹ (after 100 cycles)	244.4 mAh g ⁻¹ at 1 A g ⁻¹	[20]
MoS ₂ @C@MoS ₂	86.3	434 mAh g ⁻¹ at 100 mA g ⁻¹ (after 100 cycles)	365 mAh g ⁻¹ at 4000 mA g ⁻¹	This work



Fig. S13. Linear fit curve for peak current vs square root of scan rates for $MoS_2@C@MoS_2$ electrode.



Fig. S14. Contribution of capacitive and diffsuion controlled reaction for pristine $MoS_2 NRs$.



Fig. S15. (a) XRD pattern, (b) FE-SEM image, (c) cycling stability and (d) rate capability of NVPF cathode.

References

- [1] Y. Li, R. Zhang, W. Zhou, X. Wu, H. Zhang and J. Zhang, ACS Nano 2019, 15, 5533.
- [2] K. Yao, J. Huang, M. Ma, L. Fu, X. Shen, J. Li and M. Fu, Small 2019, 15, 1805405
- [3] C. Zhao, C. Yu, M. Zhang, Q. Sun, S. Li, M.N. Banis, X. Han, Q. Dong, J. Yang, G. Wang,
 X. Sun and J. Qiu, *Nano Energy* 2017, 41, 66.
- [4] X. Xie, T. Makaryan, M. Zhao, K.L. Van Aken, Y. Gogotsi and G. Wang, *Adv Energy Mater*. 2016, 6, 1502161.
- [5] X. Geng, Y. Jiao, Y. Han, A. Mukhopadhyay, L. Yang and H. Zhu, *Adv. Funct. Mater.* 2017, 27, 1702998.
- [6] Z. He, L. Wang, K. Zhang, J. Wang, F. Cheng, Z. Tao and J. Chen, *Angew. Chem.* 2014, 126, 130088.
- [7] G. Jia, D. Chao, N.H. Tiep, Z. Zhang and H.J. Fan, *Energy Storage Materials* 2018, 14, 136.
- [8] J. Li, W. Qin, J. Xie, R. Lin, Z. Wang, L. Pan and W. Mai, Chem. Eng. J. 2018, 332, 260.
- [9] Z. Che, Y. Li, K. Chen and M. Wei, J. Power Sources 2016, 331, 50.
- [10] Z. Xu, K. Yao, Z. Li, L. Fu, H. Fu, J. Li, L. Cao and J. Huang, J. Mater. Chem. A 2018, 6, 10535
- [11] T. Wang, P. Hu, C. Zhang, H. Du, Z. Zhang, X. Wang, S. Chen, J. Xiong and G. Cui, ACS Appl. Mater. Interfaces 2016, 8, 7811.
- [12] R. Sun, Q. Wei, Q. Li, W. Luo, Q. An, J. Sheng, D. Wang, W. Chen and L. Mai, ACS Appl. Mater. Interfaces 2015, 7, 20902.
- [13] C. Zhu, P. Kopold, W. Li, P.A. Aken, J. Maier and Y. Yu, Adv. Sci. 2015, 2, 1500200.
- [14] W. Wang, L. Shi, D. Lan and Q. Li, J. Power Sources 2018, 377, 1.
- [15] S.H. Yang, S.-K. Park, J.K. Kim and Y.C. Kang, J. Mater. Chem. A 2019, 7, 13751.

- [16] S.K. Park, J.K. Kim and Y.C. Kang, Chem. Eng. J. 2017, 328, 546.
- [17] M. Wan, R. Zeng, K. Chen, G. Liu, W. Chen, L. Wang, N. Zhang, L. Xue, W. Zhang and Y. Huang, *Energy Storage Materials* 2018, 10, 114.
- [18] Y. Liu, W. Zhong, C. Yang, Q. Pan, Y. Li, G. Wang, F. Zheng, X. Xiong, M. Liu and Q. Zhang, J. Mater. Chem. A 2018, 6, 24702.
- [19] F. Han, T. Lv, B. Sun, W. Tang, C. Zhang and X. Li, RSC Adv. 2017, 7, 30699.
- [20] A. Roy, A. Ghosh, A. Kumar and S. Mitra., Inorg. Chem. Front. 2018, 5, 2189.