## **Supporting Information**

## Targeted synthesis and reaction mechanism discussion of Mo<sub>2</sub>C based insertion-

## type electrodes for advanced pseudocapacitors

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Fig. S1 TGA curve of the as-prepared N-Mo<sub>2</sub>C/C sample.

To determine the carbon content of the N-Mo<sub>2</sub>C/C composite, thermogravimetric analysis (TGA) was performed in air flow from 30 to 700 °C. Two weight loss districts can be observed in Fig. S1, the weight loss (m<sub>1</sub>, 98 wt%) originate from the evaporation of adsorbed water below 200 °C; the following stage (m2, 126 wt%) from 300 to 700 °C corresponds to the combustion of surface carbon and the phase transition of MoO<sub>2</sub> MoO<sub>3</sub>. Thus, the content for Mo<sub>2</sub>C (m<sub>2</sub> to is  $(wt\%)/M(MoO_3)/2 \times M(Mo_2C))/m_1(wt\%) \times 100 wt\% = 91 wt\%$ . Therefore, the carbon content in the N-Mo<sub>2</sub>C/C composite was 9%.



Fig. S2 XPS survey spectrum of N-Mo<sub>2</sub>C/C sample consists of Mo 3d, C 1s, N 1s,

Mo  $3p_{1/2}$ , and O 1s species.



Fig. S3 CV and GCD curves after long-term cycling: (a) and (b) CV curves at different scan rates from 1 to 2000 mV s<sup>-1</sup>; (c) and (d) GCD curves at different current densities from 0.5 to 200 A  $g^{-1}$ .



**Fig. S4** (a,b) SEM morphology of N-Mo<sub>2</sub>C/C nanobelts electrodes after long-term cycling.



Fig. S5 (a-d) TEM images of N-Mo<sub>2</sub>C/C nanobelts electrode after long-term cycling.



**Fig. S6** *Operando* synchrotron XRD patterns of N-Mo<sub>2</sub>C/C nanobelts electrode under open circuit voltage, full discharge and charge.

Electrodes	Electrolyte	Capacitance	Current Density	Cycling	Ref.
			/ Scan rate	performance	
R-MoO <sub>3-x</sub>	LiClO <sub>4</sub>	550 C g <sup>-1</sup>	100 mV s <sup>-1</sup>	10000	1
				(~76%)	
GC/MoO <sub>3-x</sub>	$Na_2SO_4$	307 C g <sup>-1</sup>	1 A g <sup>-1</sup>	-	2
MoO <sub>2</sub> /GO	LiClO <sub>4</sub>	1097 C g <sup>-1</sup>	2 mV s <sup>-1</sup>	10000	3
		390 C g <sup>-1</sup>	1000 mV s <sup>-1</sup>	(~80%)	
MoO <sub>2</sub>	LiClO <sub>4</sub>	70 F g <sup>-1</sup>	4 A g <sup>-1</sup>	5000	4
		(63 C g <sup>-1</sup> )		(~72%)	
MoS <sub>2</sub> @BPC	NaClO <sub>4</sub>	179.8 mAh g <sup>-1</sup>	15 A g <sup>-1</sup>	5000	5
		(647 C g <sup>-1</sup> )		(73%)	
Mo <sub>2</sub> C (52.6%)/GR	LiPF <sub>6</sub>	310 mAh g <sup>-1</sup>	1.6 A g <sup>-1</sup>	100	6
		(1116 C g <sup>-1</sup> )		(~89%)	
Mo <sub>0.654</sub> C@CNS	LiPF <sub>6</sub>	495 mAh g <sup>-1</sup>	5 A g <sup>-1</sup>	680	7
		(1782 C g <sup>-1</sup> )		(<100%)	
Mo <sub>2</sub> C nanosheets	Na PF <sub>6</sub>	85.2 mAh g <sup>-1</sup>	2 A g <sup>-1</sup>	1200	8
		(306.7 C g <sup>-1</sup> )		(<100%)	
MnO <sub>2</sub> -Mo <sub>2</sub> C NFs	Na <sub>2</sub> SO <sub>4</sub>	302 F g <sup>-1</sup>	1 A g <sup>-1</sup>	5000	9
		(302 C g <sup>-1</sup> )		(92.6%)	
Mo <sub>2</sub> C/NCF	КОН	1250 F g <sup>-1</sup>	1 A g <sup>-1</sup>	5000	10
		(750 C g <sup>-1</sup> )		(<100%)	
MoSe <sub>2</sub> -Mo <sub>2</sub> C	КОН	285 F g <sup>-1</sup>	15 A g <sup>-1</sup>	10000	11
		(285 C g <sup>-1</sup> )		(98%)	
N-Mo <sub>2</sub> C/C	LiClO <sub>4</sub>	1139 C g <sup>-1</sup>	$1 \text{ mV s}^{-1}$	15000 (>100%)	This work
		151 C g <sup>-1</sup>	2000mV s <sup>-1</sup>		
		1166 C g <sup>-1</sup>	1 A g <sup>-1</sup>		

**Table S1.** Comparison of the electrochemical performances of N-Mo<sub>2</sub>C/C with previously reported Mo-based electrodes.

## References

- H. S. Kim, J. B. Cook, H. Lin, J. S. Ko, S. H. Tolbert, V. Ozolins and B. Dunn, *Nat. Mater.*, 2017, 16, 454-460.
- J. Yang, X. Xiao, P. Chen, K. Zhu, K. Cheng, K. Ye, G. Wang, D. Cao and J. Yan, *Nano Energy*, 2019, 58, 455-465.
- Y. Zhu, X. Ji, S. Cheng, Z. Y. Chern, J. Jia, L. Yang, H. Luo, J. Yu, X. Peng, J. Wang, W. Zhou and M. Liu, *ACS Nano*, 2019, 13, 9091-9099.
- 4. D. V. Pham, R. A. Patil, C.-C. Yang, W.-C. Yeh, Y. Liou and Y.-R. Ma, *Nano Energy*, 2018, **47**, 105-114.
- Y. Li, H. Wang, B. Huang, L. Wang, R. Wang, B. He, Y. Gong and X. Hu, J. Mater. Chem. A, 2018, 6, 14742-14751.
- 6. B. Wang, G. Wang and H. Wang, J. Mater. Chem. A, 2015, **3**, 17403-17411.
- J. Zhu, K. Sakaushi, G. Clavel, M. Shalom, M. Antonietti and T. P. Fellinger, J. Am. Chem. Soc., 2015, 137, 5480-5485.
- J. Li, Q.-Q. Yang, Y.-X. Hu, M.-C. Liu, C. Lu, H. Zhang, L.-B. Kong, W.-W. Liu, W.-J. Niu, K. Zhao, Y.-C. Wang, F. Cheng, Z. M. Wang and Y.-L. Chueh, *ACS Sustainable Chem. Eng.*, 2019, 7, 18375-18383.
- M. Shi, L. Zhao, X. Song, J. Liu, P. Zhang and L. Gao, ACS Appl. Mater. Interfaces, 2016, 8, 32460-32467.
- 10. K. J. Samdani, D. W. Joh and K. T. Lee, J. Alloy. Compo., 2018, 748, 134-144.
- D. Vikraman, S. Hussain, K. Karuppasamy, A. Feroze, A. Kathalingam, A. Sanmugam, S.-H. Chun, J. Jung and H.-S. Kim, *Appl. Catal. B-Environ.*, 2020, 264, 118531.