

## **Electronic Supporting Information**

# Binding Low Crystalline MoS<sub>2</sub> Nanoflakes on Nitrogen-Doping Carbon Nanotube: Towards High-Rate Lithium and Sodium Storage

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### **Methods Section**

#### **Material Characterizations**

The X-ray diffractometer (XRD, Rigaku D/max 2550 VB<sup>+</sup>) with Cu Kα radiation was utilized to investigate the phase structure of materials at a scanning rate of 5° min<sup>-1</sup>. Scanning electron microscopy (SEM, JSM-6510LV) was further used to study the morphology of the samples. And the morphology character and atomic arrangement were explored via transmission electron microscopy (TEM, JEM-2100F) and high-resolution transmission electron microscopy (HRTEM, JEM-2100F). The elemental analysis and surface chemical composition of samples were carried out through energy-dispersive X-ray spectroscopy (EDS) and X-ray Photoelectron Spectroscopy (XPS, ESCALab250) with the C1s photoelectron peak at 284.6 eV as the reference. The content analysis of composite was measured via thermogravimetric analysis (TGA, NETZSCH STA449F3) from 30 to 700 °C with a heating rate of 5 °C min<sup>-1</sup> in air. Moreover, the specific surface area of materials was determined using nitrogen adsorption isotherms at 77K with Brunauer-Emmett-Teller (BET, Micromeritics, ASAP 2020). Raman spectrometer (HORIBA Labram HR Evolution) was used to collect raman spectra of as-prepared products.

#### **Electrochemical Measurements**

The electrode can be fabricated as follows process. The active material (70 mg) and super P (15 mg) were firstly blended well, and then this mixture was added into the polyvinylidene fluoride (PVDF, 15 mg) binder with N-methyl-2-pyrrolidone (NMP) as solvent under whisking for 12 h. The as-formed homogeneous slurry was coated onto Cu foil and air-dried at 50 °C. Finally, the working electrodes with the mass loading of 1.0~1.3 mg were obtained after further drying in a vacuum at 80 °C for 12 h. The electrochemical performances of samples were measured in a CR2016 coin cell, that was assembled in an argon filled glovebox (H<sub>2</sub>O < 0.5 ppm, O<sub>2</sub> < 0.5 ppm). For LIBs, Li foil was as both counter electrode and reference

electrode. The electrolyte was a solution of 1 M LiPF<sub>6</sub> in ethylene carbonate/dimethyl carbonate (volume ratio of 1:1). While, Na foil was as both counter electrode and reference electrode for SIBs. And the electrolyte was a 1 M NaClO<sub>4</sub> solution in ethylene carbonate and propylene carbonate system (volume ratio of 1:1). Galvanostatic discharge/charge files of electrodes were performed on Land CT2001A battery cycler at various current densities. Cyclic voltammetry (CV) curves at various scanning rates and Electrochemical impedance measurements (EIS) at the open–circuit voltages were further investigated on MULTI AUTOLAB M204 (MAC90086).



Fig. S1. (a) XRD pattern and (b) EDS of the product after alternating current treatment.



Fig. S2. The SEM (a) and elemental mapping images (b-e) of  $MoS_2/N$ -CNT.



Fig. S3. The pore size distribution results of  $MoS_2$  and  $MoS_2/N$ -CNT based on their adsorption curves.

Moreover, the pore size distribution results of  $MoS_2$  and  $MoS_2/N$ -CNT have been further analyzed by the Barrett-Joyner-Halenda method, as is shown in Fig. S3. The  $MoS_2/N$ -CNT composite exhibits some mesopores and macropores originating from the introduction of CNT, which is beneficial to ion transport.



Fig. S4. TG curve of  $MoS_2/N$ -CNT-Raw composite from room temperature to 700 °C under air.



Fig. S5. (a) The survey spectrum of  $MoS_2/N$ -CNT composite. (b) C 1s XPS spectrum of  $MoS_2/N$ -CNT composite.



Fig. S6. SEM of  $MoS_2/N$ -CNT electrode after 800 cycles at a current density of 2 A g<sup>-1</sup> for LIBs.

The SEM result of  $MoS_2/N$ -CNT electrode has been measured after 800 cycles at a current density of 2 A g<sup>-1</sup> for LIBs, which is shown in Fig. S6. It can be seen that  $MoS_2$  displays volume expansion. While the N-CNT can still well anchor the  $MoS_2$  as an integrated electrode after cycles. Herein, the introduction of N-CNT can be helpful to maintain the stable structure and improve the cycling stability.



**Fig. S7.** Rate behavior of MoS<sub>2</sub>-based anodes for LIBs from previous literatures compared against the current work.



Fig. S8. CV curves of  $MoS_2/N$ -CNT electrode at various scan rates from 0.1 mV s<sup>-1</sup> to 0.7 mV s<sup>-1</sup>.



**Fig. S9.** (a) Nyquist plots of pure  $MoS_2$  and  $MoS_2/N$ -CNT composite after 100 cycles at 200 mA g<sup>-1</sup> for SIBs. (b) Z' vs w<sup>-1/2</sup> plots of  $MoS_2$  and  $MoS_2/N$ -CNT for Warburg impedance analysis.

The EIS analysis of pure MoS<sub>2</sub> and MoS<sub>2</sub>/N-CNT composite for SIBs has been added in Fig. S9. The Nyquist plots of pure MoS<sub>2</sub> and MoS<sub>2</sub>/N-CNT composite are abtained after 100 cycles at 200 mA g<sup>-1</sup> in Fig. S9a. It shows the resistance  $R_{ct}$  of MoS<sub>2</sub>/N-CNT is obvious smaller than pure MoS<sub>2</sub> electrode. Also, the Warburg impedance coefficient ( $\sigma$ ) relative to Z' have been further calculated, as is shown in Fig. S9b. The slop of MoS<sub>2</sub>/N-CNT (57.7) is much smaller than that of MoS<sub>2</sub> (222.4), illustrating the higher Na<sup>+</sup> diffusion rate of this composite electrode.



**Fig. S10.** Rate behavior of MoS<sub>2</sub>-based anodes for SIBs from previous literatures compared against the current work.



Sample	Electrical Conductivity (S cm <sup>-1</sup> )
MoS <sub>2</sub>	8.21 ×10 <sup>-5</sup>
MoS <sub>2</sub> /N-CNT	78.12

Table S1 The electrical conductivity data of pure  $MoS_2$  and  $MoS_2/N$ -CNT.

The electrical conductivity of pure  $MoS_2$  and  $MoS_2/N$ -CNT has been investigated by fourpoint probe tester (SDY-40, Guangdong, China). The data has been presented in Table S1 based on the average values of three samples for each electrode. The  $MoS_2/N$ -CNT shows significantly higher electrical conductivity than that of pure  $MoS_2$ .

### References

- S1 B. Chen, Y. Meng, F. He, E. Liu, C. Shi, C. He, L. Ma, Q. Li, J. Li, N. Zhao, *Nano Energy* 2017, 41, 154.
- S2 G. Wang, J. Zhang, S. Yang, F. Wang, X. Zhuang, K. Müllen, X. Feng, *Adv. Energy Mater.* **2018**, *8*, 1702254.
- S3 L. Ma, B. Zhao, X. Wang, J. Yang, X. Zhang, Y. Zhou, J. Chen, ACS Appl. Mater. Interfaces 2018, 10, 22067.
- S4 Y. Jiao, A. Mukhopadhyay, Y. Ma, L. Yang, A. M. Hafez, H. Zhu, *Adv. Energy Mater.* **2018**, *8*, 1702779.
- S5 L. Jiang, B. Lin, X. Li, X. Song, H. Xia, L. Li, H. Zeng, ACS Appl. Mater. Interfaces 2016, 8, 2680.
- S6 R. Wang, S. Gao, K. Wang, M. Zhou, S. Cheng, K. Jiang, Sci. Rep. 2017, 7, 7963.
- S7 Y. Teng, H. Zhao, Z. Zhang, L. Zhao, Y. Zhang, Z. Li, Q. Xia, Z. Du, K. Świerczek, *Carbon* 2017, 119, 91.
- S8 J. Wu, Z. Lu, K. Li, J. Cui, S. Yao, M. Ihsan-ul Haq, B. Li, Q.-H. Yang, F. Kang, F. Ciucci, J.-K. Kim, J. Mater. Chem. A 2018, 6, 5668.
- S9 B. Chen, H. Lu, J. Zhou, C. Ye, C. Shi, N. Zhao, S. Z. Qiao, Adv. Energy Mater. 2018, 1702909.
- S10 D. Xie, X. Xia, Y. Wang, D. Wang, Y. Zhong, W. Tang, X. Wang, J. Tu, *Chem. Eur. J.* 2016, 22, 11617.