Supporting Information

MXene derived three-dimensional carbon nanotubes network encapsulate CoS₂ nanoparticles as anode material for solid-state sodium-ion batteries

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Electrochemical characterization

Electrodes were prepared by slurry coating method. In detail, the active material (CoS₂/CNTs/TiO_xN_y), conductive additive (acetylene black) and binder (polyvinylidene fluoride) were mixed at a weight ratio of 8:1:1 with N-methyl-2-pyrrolidone to get slurry. Then, the slurry was coated on the current collector (copper foil) and the electrodes were dried under vacuum at 120 °C for overnight. All CR2023-type coin cells SSIBs were assembled by electrode, sodium metal foil and PFSA-membranes in Argon-filled glove box. Furthermore, liquid sodium-ion batteries (LSIBs) were assembled and characterized as well. All CR2023-type coin cells about LSIBs were assembled by electrode, sodium foil, glass diaphragm (Whatman, GF/D) and electrolyte (1 M NaClO₄ dissolved in EC and DEC with a volume ratio of 1:1) in Argon-filled glove box. The cyclic voltammetry (CV) and galvanostatic charge/discharge profiles were investigated with CHI 660C and Land battery testing system with a potential range of 0.01-3 V. Zennium electrochemical workstation was used to measure Nyquist plots of the electrode.

Materials characterization

The morphologies of samples were observed by field emission scanning electron microscopy (FESEM, JSM-7800F, Japan) and transmission electron microscopy (TEM, JEM-2100, Japan). The elements distribution was observed by energy dispersive spectrometry (EDS). Powder X-ray diffraction (XRD, MAXima-X XRD-7000, Cu K α radiation) was used to detect the crystal structures of samples. The LabRAM HR Evolution (Horiba) was used to test *in-situ* Raman spectrum with laser ex-citation at 532nm. The X-ray photoelectron spectroscopy (XPS, Thermo Scientific ESCALAB 250Xi electron spectrometer) measurements were analyzed to chemical composition of the products and the in-depth analysis was carried out at beam energy of 4000 eV (contrast sample was Ta₂O₅). The pore structure and surface properties of CoS₂/CNTs/TiO_xN_y were further characterized by N₂ adsorption/desorption measurements (Quadrasorb evo 2QDS-MP-30).



Fig.S1 low-magnification FESEM image of Co/CNTs/TiOxNy.



Fig.S2 (a) FESEM image of CoS_/CNTs/TiO_xN_y; (b)-(d) TEM images of CoS_/CNTs/TiO_xN_y.



Fig.S3 EDS spectrum of Co, S, Ti, C and N elements in $CoS_2/CNTs/TiO_xN_y$.



Fig.S4 FESEM image of CoS₂/CNTs.



Fig.S5 Electrochemical performance of $CoS_2/CNTs/TiO_xN_y$ in LSIBs. (a) CV curves at 0.1 mV s⁻¹ scan rate; (b) The charge/discharge curves of cycling performance at 1 A g⁻¹; (c) Cycling performance at 1 A g⁻¹ and the coulombic efficiency; (d) Charge/discharge curves of rate performance; (e)Rate capability tests at various current densities; (f) Nyquist plots of before and after first cycle.



Fig.S6 Comparation of CV curves at 0.1 mV s⁻¹ scan rate



Fig.S7 Comparison of battery performance with previous literatures^{S1-8}. (a) Operating temperature. (b) Employed

current density and cycle number.



Fig.S8 Electrochemical performance in solid sate sodium-ion batteries (a) The charge/discharge curves of rate performance of $CoS_2/CNTs/TiO_xN_y$; (b) Rate capability tests at various current densities and (c) The charge/discharge curves of rate performance of $Co/CNTs/TiO_xN_y$.



Fig.S9 (a) CV curves at various scan rates; (b) Relationship between logarithm anodic peak current and logarithm

scan rates.



Fig.S10 (a) BET adsorption-desorption isotherm and (b) pore size distribution of $CoS_2/CNTs/TiO_xN_y$.

Reference

- S1 Y. W. Zheng, Q. W. Pan, C. M. Clites, B. W. Byles, E. Pomerantseva, C. Y. Li, *Adv. Energy Mater.* 2018, 8, 1801885.
- S2 F. Colò, F. Bella, J. R. Nair, M. Destro, C. Gerbaldi, *Electrochimica Acta* 2015, 174, 185-190.
- S3 Q. Ma, J. J. Liu, X. G. Qi, X. H. Rong, Y. J. Shao, W. F. Feng, J, Nie, Y, S. Hu, H. Li, X. J. Huang, L. Q. Chen, Z, B. Zhou, J. Mater. Chem. A 2017, 5, 7738-7743.
- S4 S. F. Song, M. Kotobuki, F. Zheng, C. H. Xu, S. V. Savilov, N. Hu, L. Lu, Y. Wang, W. D. Z. Li, J. Mater. Chem. A 2017, 5, 6424-6431.
- S5 Z. Z. Zhang, K. Q. Xu, X. H. Rong, Y. S. Hu, H. Li, X. J. Huang, L. Q. Chen, J. Power Sources 2017, 372, 270-275.
- S6 Y. L. Ni'mah, M. Y. Cheng, J. H. Cheng, J. Rick, B. J. Hwang, J. Power Source 2015, 278, 375-381.
- S7 H. C. Gao, B. K. Guo, J. Song, K. Park, J. B. Goodenough, Adv. Energy Mater. 2015, 5, 1402235.
- S8 H. S. Li, Y. Ding, H. Ha, Y. Shi, L. L. Peng, X. G. Zhang, Adv. Mater. 2017, 29, 1700898.