Electronic Supplementary Information (ESI)

Formation and Operating Mechanisms of Single-Crystalline Perovskite $NaNbO_3$ Nanocubes/Few-Layered Nb_2CT_x MXene Hybrids towards Li-Ion Capacitors

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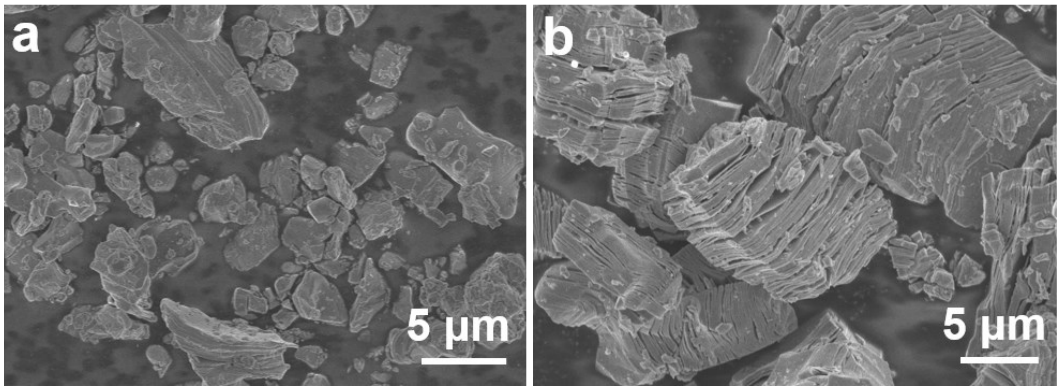

Fig. S1. FESEM images of (a) Nb_2AlC MAX and (b) m-Nb_2CT_x MXene.

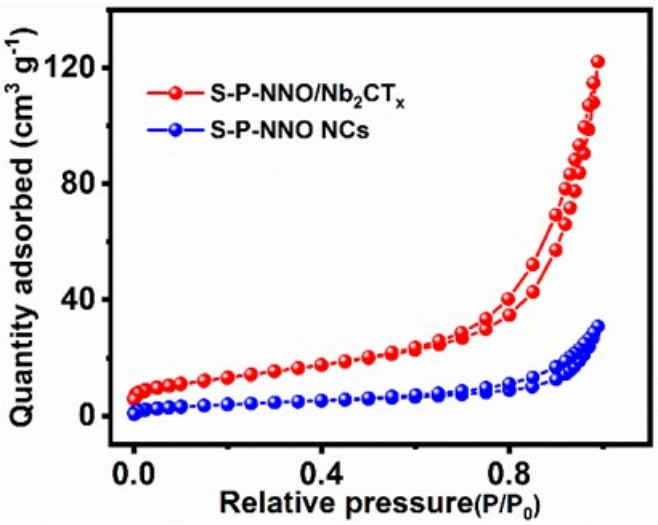

Fig. S2. Nitrogen adsorption/desorption isotherms of S-P-NNO NCs and S-P-NNO/f-Nb_2CT_x hybrid as indicated.

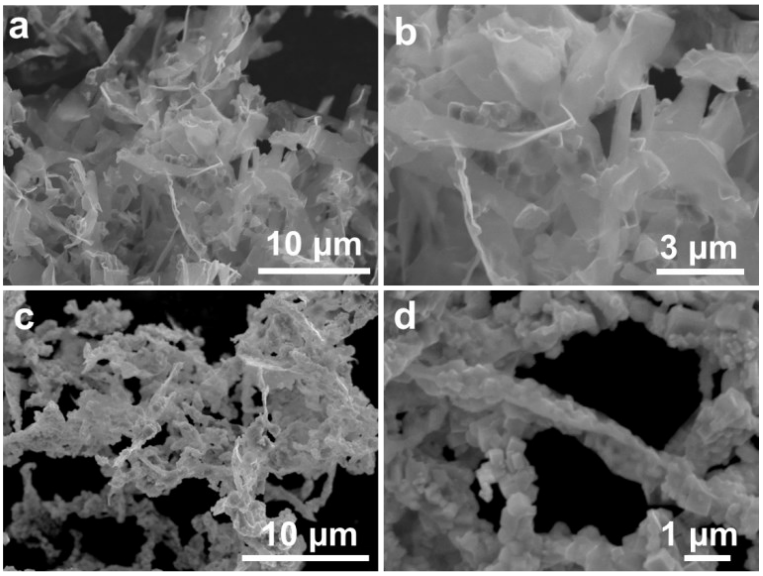

Fig. S3. SEM images of the S-P-NNO NCs and f-Nb_2CT_x NSs after freeze-drying with different mass ratios: (a, b) 1 : 1; (c, d) 3 : 1.

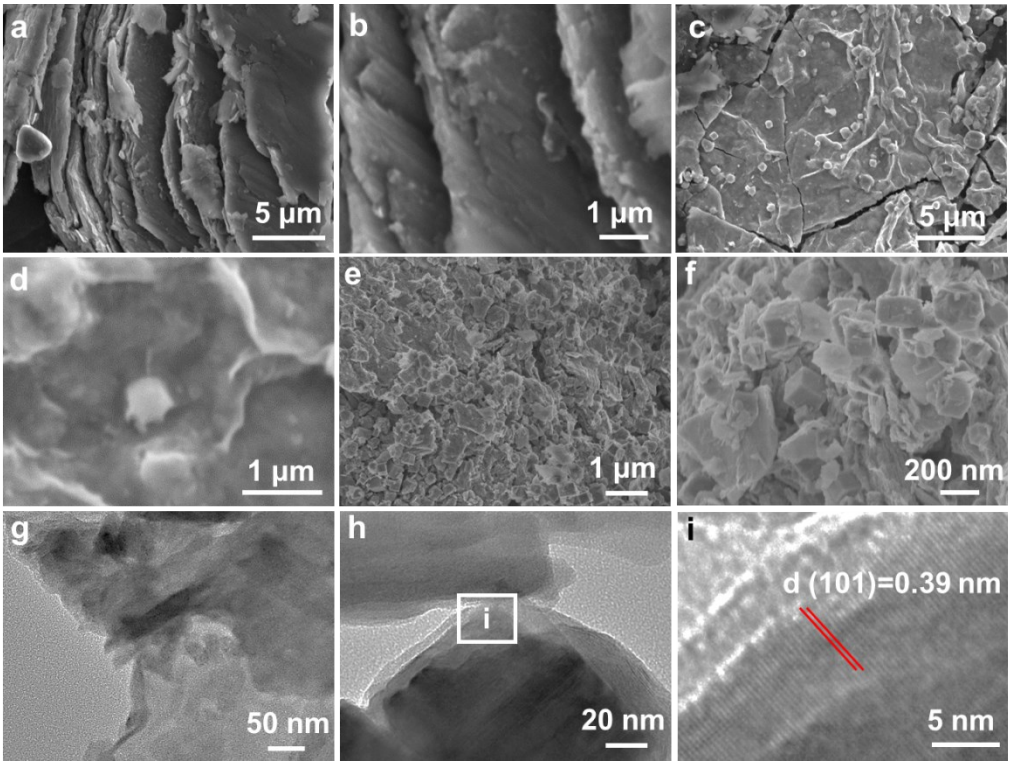

Fig. S4. SEM images of (a, b) f-1M-0.5h and (c, d) f-1M-1h; (e, f) SEM, (g, h) TEM and (i) HRTEM images of f-1M-2h.

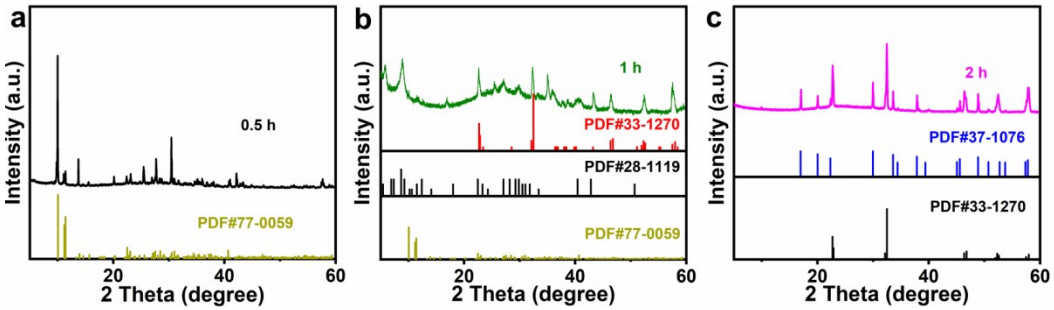

Fig. S5. XRD patterns of (a) f-1M-0.5h, (b) f-1M-1h, and (c) f-1M-2h.

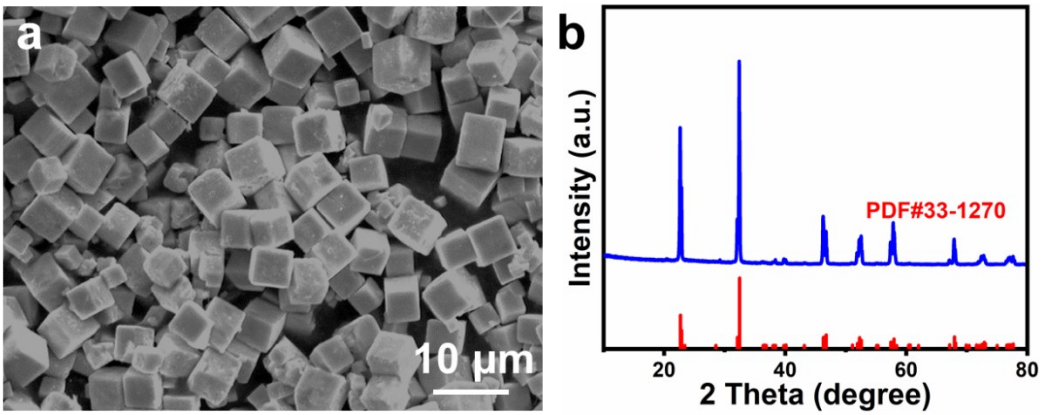

Fig. S6. (a) SEM image and (b) XRD pattern of f-1M-12h.

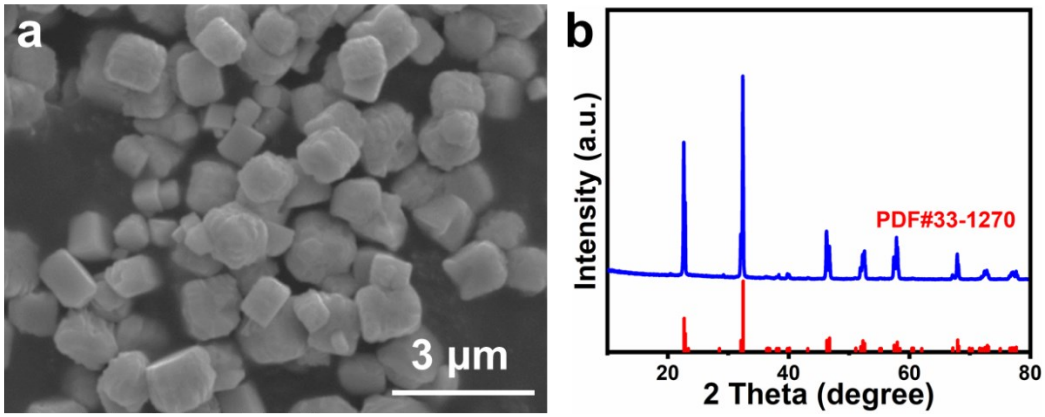

Fig. S7. (a) SEM image and (b) XRD pattern of the f-3M-3h.

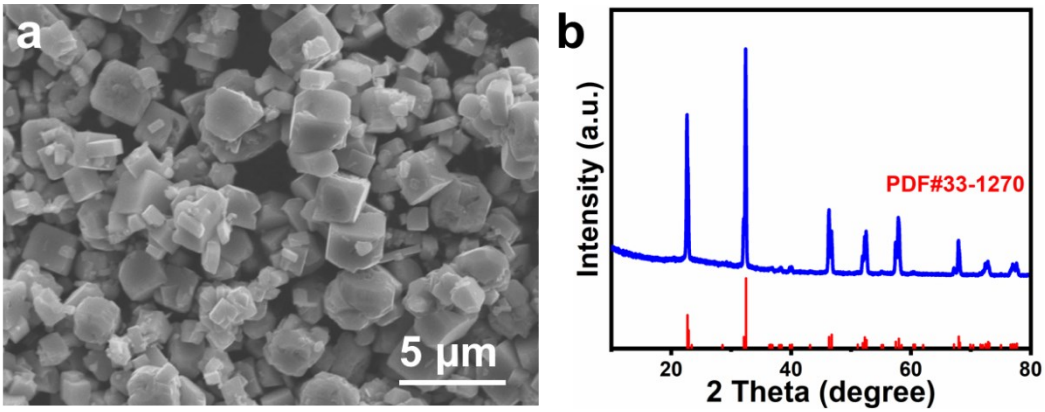

Fig. S8. (a) SEM image and (b) XRD pattern of the m-1M-3h.

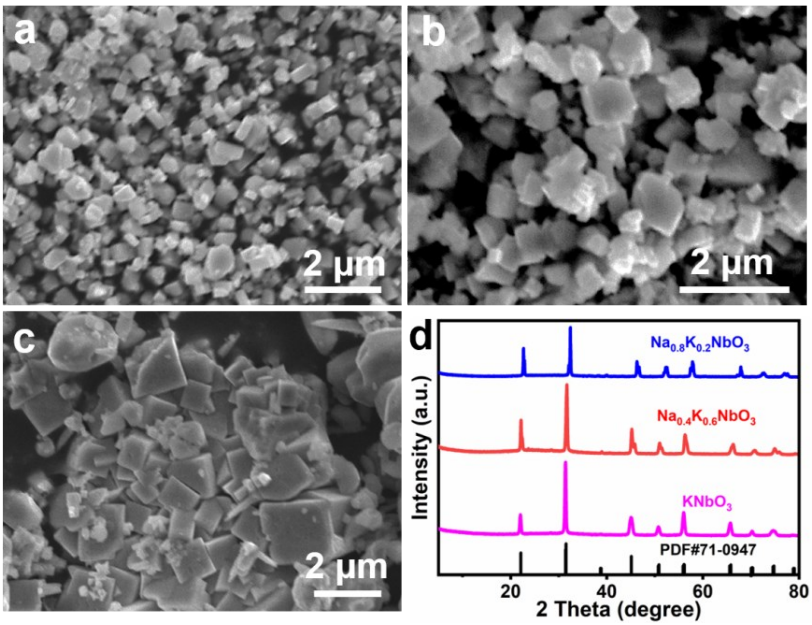

Fig. S9. SEM images of (a) $KNbO_3$, (b) $Na_{0.4}K_{0.6}NbO_3$ and (c) $Na_{0.8}K_{0.2}NbO_3$, respectively, and XRD patterns of the three as indicated.

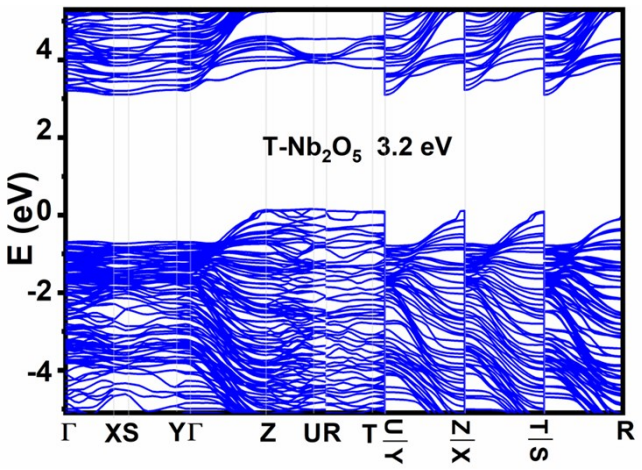

Fig. S10. Band structure of the T-Nb_2O_5.

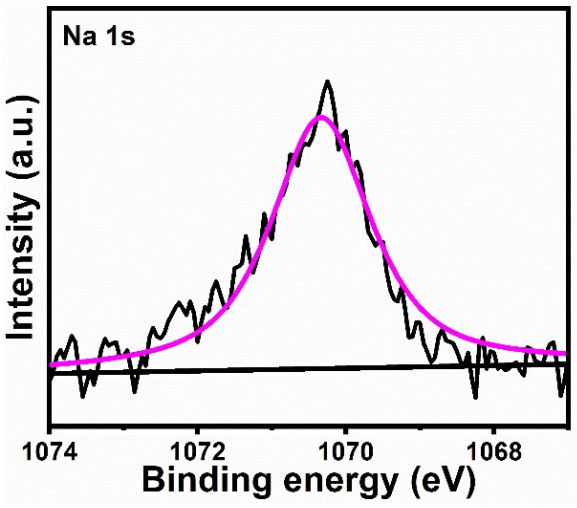

Fig. S11. Na 1s spectrum of the S-P-NNO/f-Nb_2CT_x.

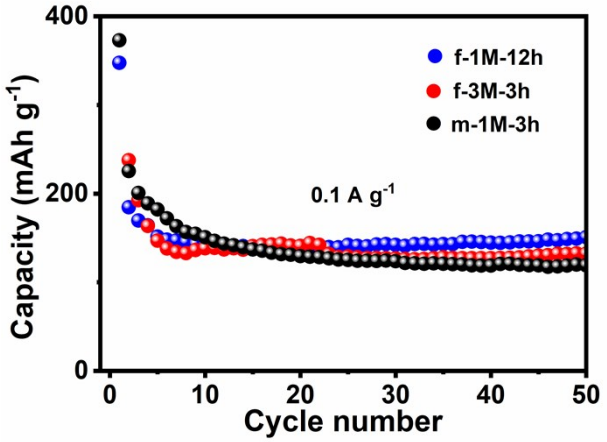

Fig. S12. Cycle performance of f-1M-12h, f-3M-3h and m-1M-3h at 0.1 A g^{-1}.

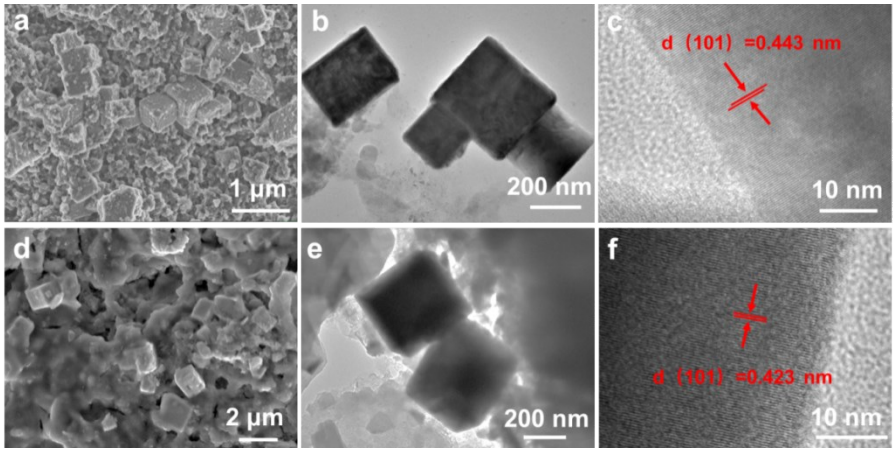

Fig. S13. (a) SEM, (b) TEM and (c) HRTEM images of the S-P-NNO after 1500 cycles at 1.0 A g^{-1}. (d) SEM, (e) TEM, and (f) HRTEM images of the S-P-NNO/f-Nb_2CT_x after 1500 cycles at 1.0 A g^{-1}.

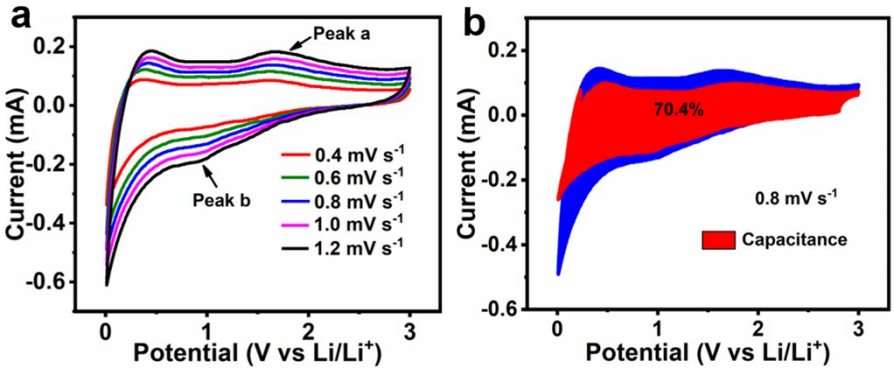

Fig. S14. Electrochemical kinetics analysis of the S-P-NNO electrode. (a) CV curves at various scan rates ranging from 0.4 to 1.2 mV s^{-1}, (b) CV curve with pseudocapacitive (red) and battery-type (blue) contributions at 0.8 mV s^{-1}.

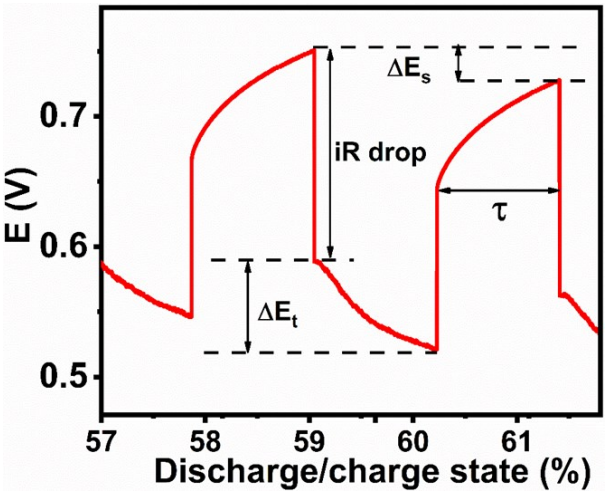

Fig. S15. GITT curves of the S-P-NNO/f-Nb_2CT_x anode with the ΔE_τ, ΔE_s and τ during discharge and charge processes.

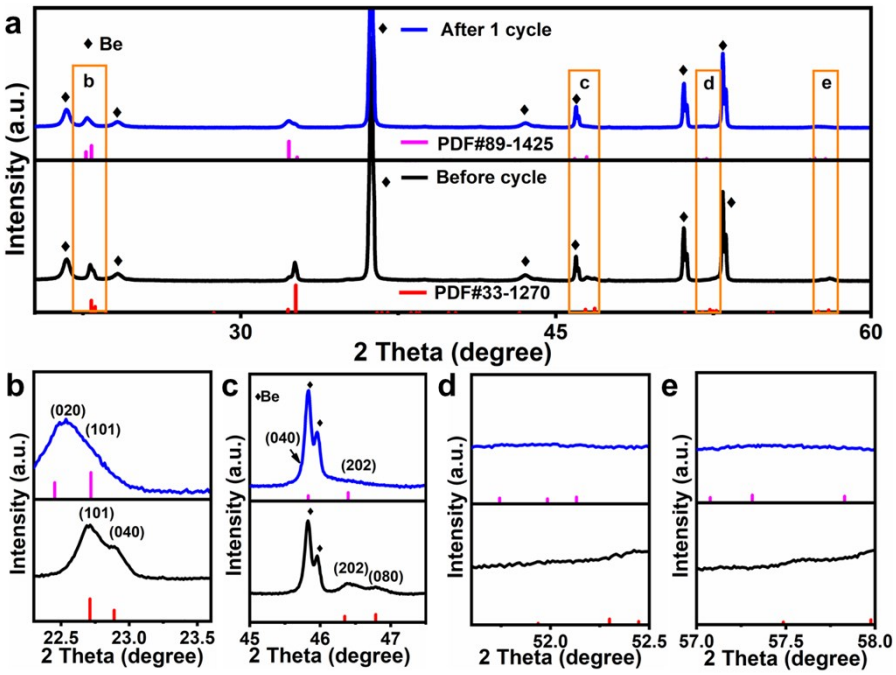

Fig. S16. XRD patterns of (a) S-P-NNO and (b – e) corresponding partial enlarged images before and after the 1st cycle.

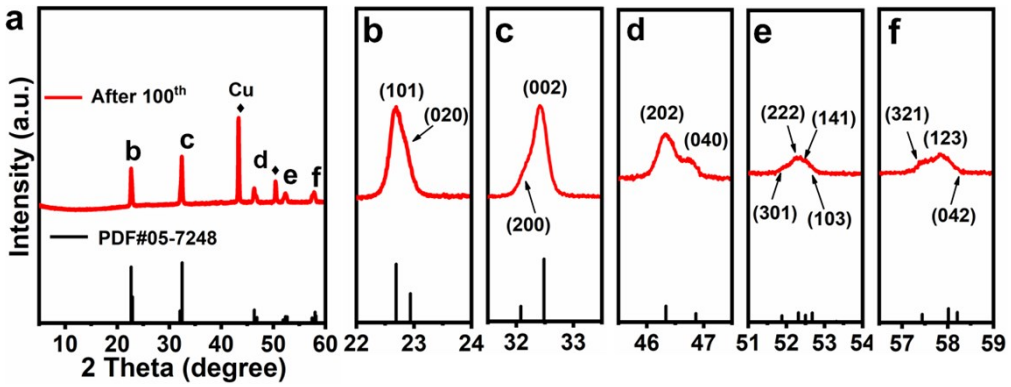

Fig. S17. XRD patterns of (a) S-P-NNO and (b – f) corresponding partial enlarged images after the 100th cycle.

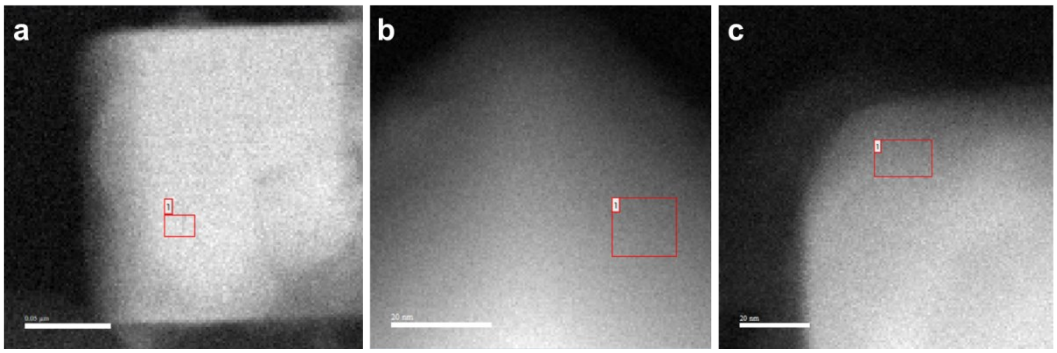

Fig. S18. STEM images of the S-P-NNO electrode after (a) the 1st, (b) 100th, and (c) 500th cycles, respectively. The red rectangular regions in panels for EELS tests.

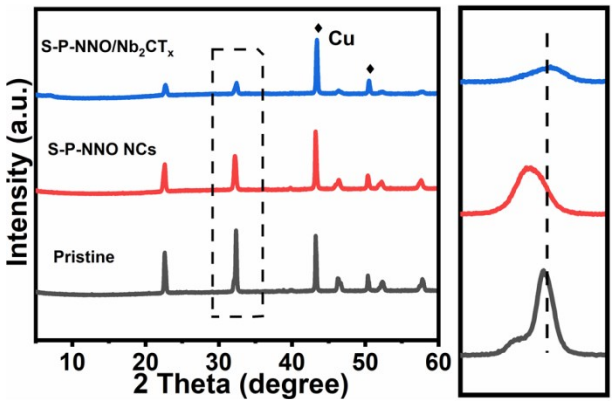

Fig.S19. *Ex-situ* XRD patterns of S-P-NNO and S-P-NNO/ f-Nb_2CT_x after 1st cycle.

Table S1. Approximate mass loading of each device

Electrodes	Mass loading (mg cm^{-2})	Reference
S-P-NNO/f-Nb_2CT_x//AC	4.0 – 4.8	This work
3S-Nb_2O_5-HoMSs//AC	3.0 – 5.0	[1]
N-NbOC//AC	~1.98	[2]
NRC-40//AC	4 – 4.8	[3]
Nb_2O_5//MSP-20	1.4 – 4.9	[4]
T-Nb_2O_5 NRs//AC	3.5 – 4.2	[5]

Experimental supplemental data

Synthesis of $KNbO_3$: Typically, 17.7 g of NaOH (Sinopharm Chemical ReagentCo., Ltd) was putted in 40 mL of the f-Nb_2CT_x solution. After stirred for 20 min, the mixture was transferred into a Taylor reactor (50 mL in volume) at 200 ºC for 3 h, and naturally cooled to room temperature (RT). The $KNbO_3$ power was obtained by centrifugation, and dried at 60 ºC for 12 h.

Synthesis of $Na_{0.4}K_{0.6}NbO_3$: 0.64g of NaOH and 10.6 g of KOH were putted in 40 mL of the f-Nb_2CT_x solution. After stirring for 20 min, the mixture was transferred into a Taylor reactor (50 mL in volume) at 200 ºC for 3 h, and naturally cooled to RT. The $Na_{0.4}K_{0.6}NbO_3$ power was obtained by centrifugation, and dried at 60 ºC for 12 h.

Synthesis of $Na_{0.8}K_{0.2}NbO_3$: 1.28 g of NaOH and 3.54 g of KOH were putted in 40 mL of the f-Nb_2CT_x solution. After stirring for 20 min, the mixture was transferred into a Taylor reactor (50 mL in volume) at 200 ºC for 3 h, and naturally cooled to RT. The $Na_{0.8}K_{0.2}NbO_3$ power was obtained by centrifugation, and dried at 60 ºC for 12 h.

References

1 R. Y. Bi, N. Xu, H. Ren, N. L. Yang, Y. G. Sun, A. Cao, R. B. Yu and D. Wang, *Angew. Chem. Int. Ed.* 2020, **59**, 4865.

2 S. Hemmati, G. Li, X. L. Wang, Y. L. Ding, Y. Pei, A. P. Yu and Z. W. Chen, *Nano Energy* 2019, **56**, 118.

3 C. H. Lai, D. Ashby, M. Moz, Y. Gogotsi, L. Pilon and B. Dunn, *Langmuir* 2017, **33**, 9407.

4 E. Lim, H. Kim, C. Jo, J. Chun, K. Ku, S. Kim, H. I. Lee, I. Nam, S. Yoon, K. Kang and J. Lee, *ACS Nano* 2014, ***8***, 8968.

5 L. Qin, Y. Liu, S. Y. Xu, S. C. Wang, X. Sun, S. H. Zhu, L. R. Hou and C. Z. Yuan, *Small Methods* 2020, **4**, 2000630.