

Supporting Informations

Novel Sodium/Lithium-ion Anode Material Based on Ultrathin $\text{Na}_2\text{Ti}_2\text{O}_4(\text{OH})_2$ Nanosheet

*Yuping Zhang,^a Lin Guo,^{a *} Shihe Yang^{a,b}*

*^{a *} School of Chemistry and Environment, Beihang University. Beijing, 100191, P. R.*

China. E-mail: guolin@buaa.edu.cn

^b Department of Chemistry, The Hong Kong University of Science and Technology,

Clear Water Bay, Kowloon Hong Kong (P.R. China). E-mail : chsyang@ust.hk

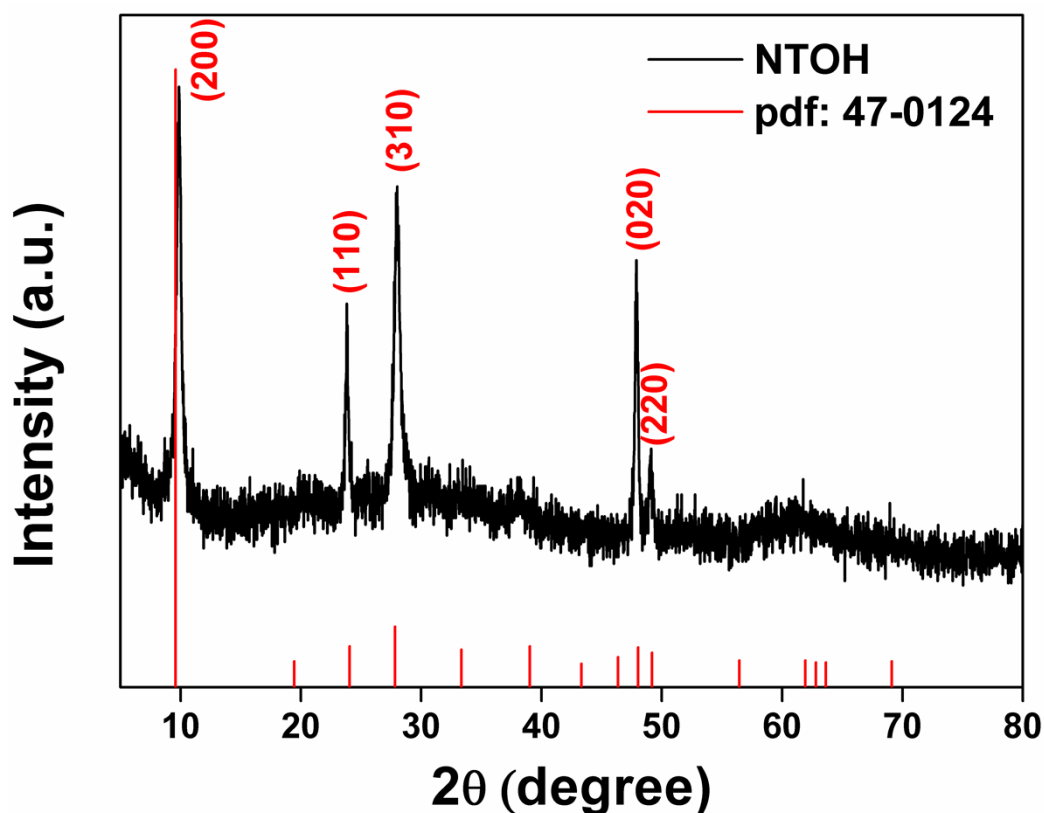


Figure S1. XRD pattern of the as prepared ultrathin NTOH nanosheets. All diffraction peaks are similar to the diffraction pattern of body-centered orthorhombic crystal structure of $\text{H}_2\text{Ti}_2\text{O}_4(\text{OH})_2$ (denoted as HTOH), the most obvious difference lie in that the (200) diffraction peak of NTOH is a bit higher shift than that of HTOH, as the bond distance of Na-O is larger than that of H-O.

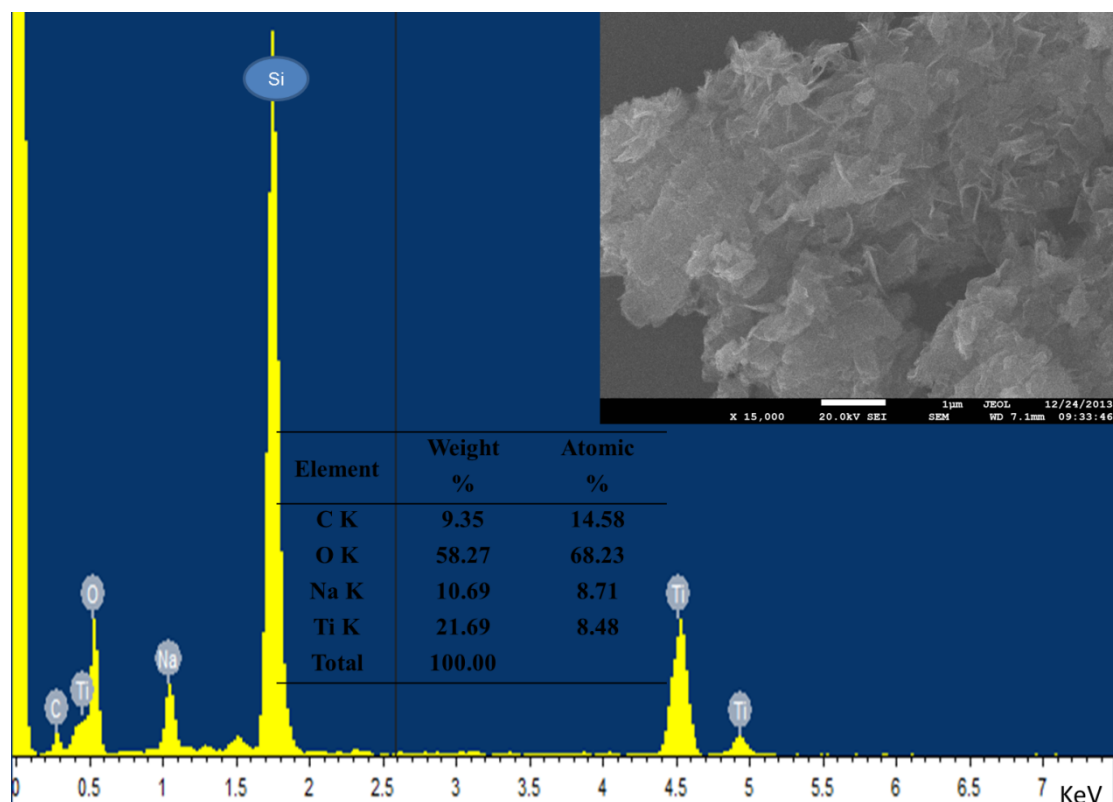


Figure S2. EDX spectrum of the ultrathin NTOH nanosheets (inset is an SEM image and inset table of element ratio distribution) As shown in EDX spectrum, the elements of Ti, Na, O are clearly observed, and as shown inset table of ultrathin NTOH nanosheets, Ti/Na atomic ratio is 1.0, which is equal to that of in $\text{Na}_2\text{Ti}_2\text{O}_4(\text{OH})_2$. In addition, the C and Si contents mainly derived from the pollution of electrically conductive adhesive and Si substrate.

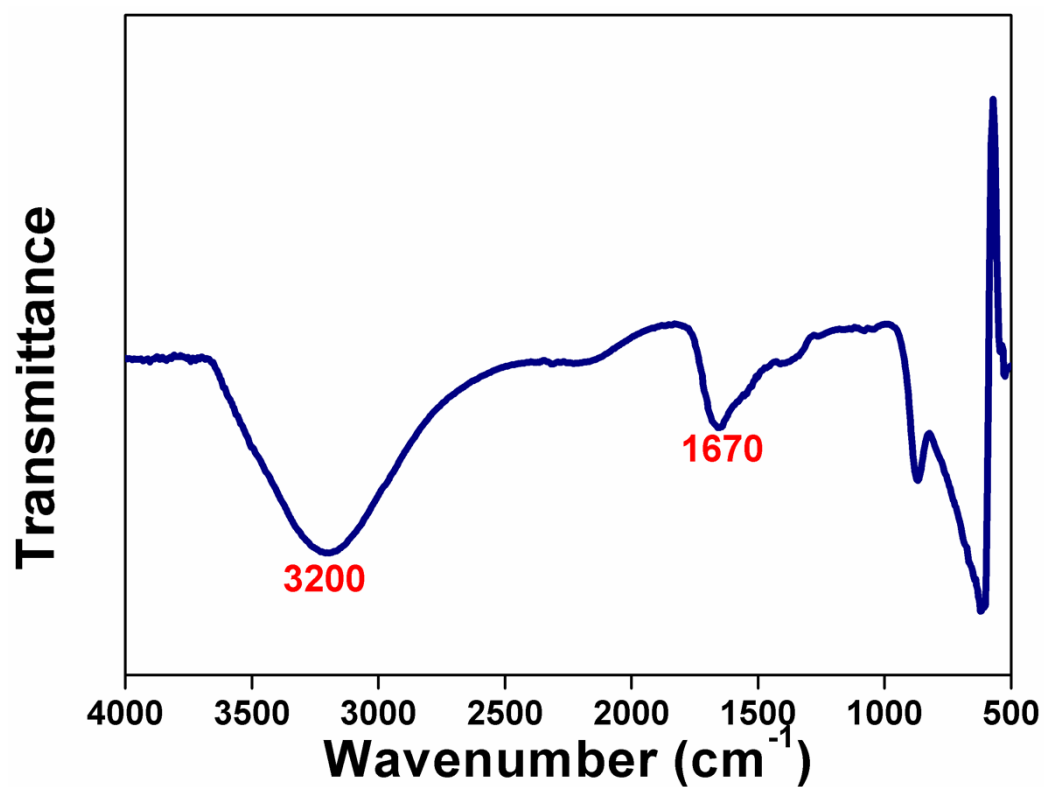


Figure S 3. IR spectra of as prepared NTOH nanosheets. A band at 3200 cm⁻¹ is due to stretching modes of bonded -OH groups and of water molecules coordinated to surface Ti⁴⁺ cations, the bending modes of which also produce a band at 1670 cm⁻¹ .^[1]

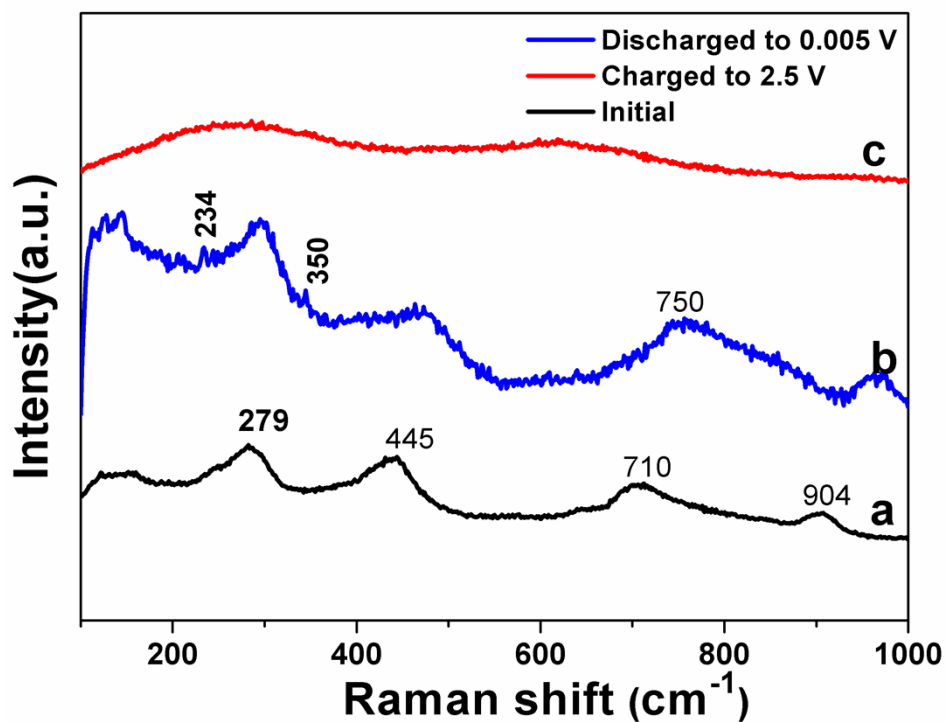


Figure S4. Raman spectra of initial NTOH electrode, and the electrode charged to 2.5 V and full discharged to 0.005 V over the coin cell of Lithium ion batteries. The a curve in Figure S4 is the Raman spectra of initial ultrathin $\text{Na}_2\text{Ti}_2\text{O}_4(\text{OH})_2$ electrode, the bands at 279, 445 cm^{-1} are ascribed to Na-O-Ti stretching. The band at 710 cm^{-1} and 750 cm^{-1} is respectively assigned to Ti-O-Ti and Ti-O stretching in TiO_6 , while the band at 904 cm^{-1} is indicated to symmetric stretch of short Ti-O bonds involving nonbridging oxygen coordinated with sodium ions. However, when the Li^+ inserted into the frame of NTOH, the lattice structure of NTOH was changed. As shown in b curve in Figure 4S, the intensity of Raman bands representing NTOH is weakening, indicating that Li^+ almost replaced the Na^+ . The bands at 350 cm^{-1} can be assigned to stretching vibration of Li-O bond, further proofing that Li^+ inserted into NTOH frame. In addition, when fully discharged NTOH experienced the process of charging, the Raman bands of fully charged NTOH was almost disappeared, indicating that Li^+

were extracted during the charge process, left only an Li-free frame as shown in the c curve in Figure 4S. [2-4]

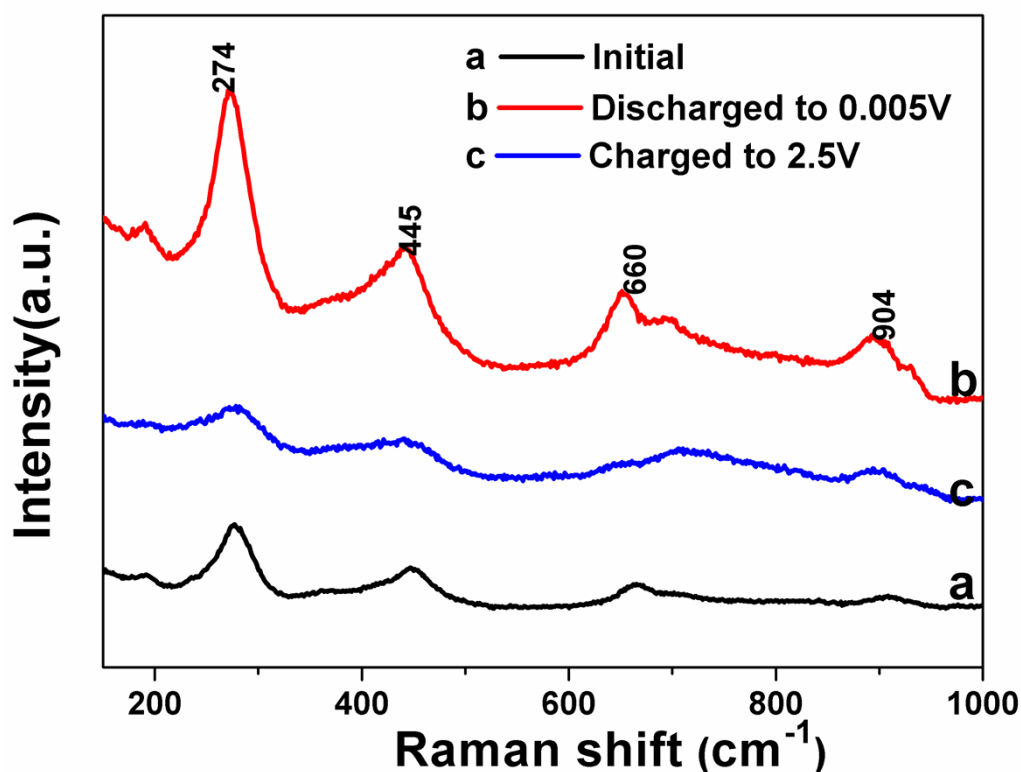


Figure S5. Raman spectra of initial NTOH electrode, and the electrode charged to 2.5 V and full discharged to 0.005 V over the coin cell of sodium ion batteries. As shown in Figure 5S, the bands at 274, 445 and 660 cm⁻¹ are indexed to Na-O-Ti stretching. The band at 905 cm⁻¹ is indicated to symmetric stretch of short Ti-O bonds involving nonbridging oxygen coordinated with sodium ions. The a) curve is Raman spectra of initial NTOH , the curve b) represents the fully discharged NTOH and the curve c is Raman spectra of fully charged NTOH. There is no obvious position difference of Raman bands between curve a, b and c, only difference is lying in the intensity of the Raman band. The Raman intensity signal of b is much higher than that of a, indicating that more Na⁺ inserted in to frame of NTOH, strengthening the Raman signal. However, when the fully Na-inserted NTOH experiences the charge process, the

Raman intensity is decreasing comparing with the initial NTOH, indicating that Na^+ extracted during the process.

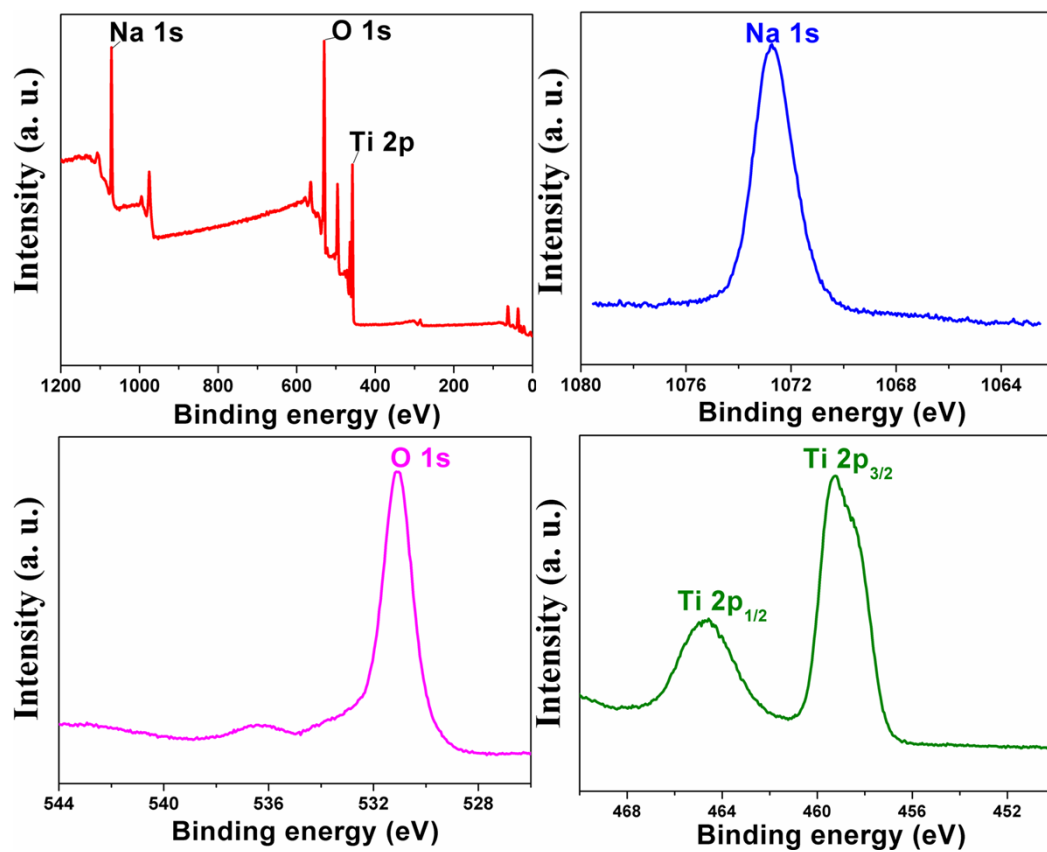


Figure S6. XPS spectra of as prepared $\text{Na}_2\text{Ti}_2\text{O}_4(\text{OH})_2$ nanosheet.

Table1S. Impedance parameters calculated from equivalent circuits from fresh cell of NIB and LIB

Ion-storage	Re (Ω)	Rct (Ω)	Zw (Ω)
SIBs	20	150	45
LIBs	4	60	59

Table S2. Comparison of Na₂Ti₂O₄(OH)₂ (this work) and various reported material for Li-ion batteries

Material	Reversible Capacity (mAh/g)	Rate	Ref. (manuscript)
Na₂Ti₂O₄(OH)₂	120/500cycles/ SIBs	10C	This work
nanosheet	150/500cycles/LIBs	1C	This work
SnO₂	420/100cycles/SIBs	20 mA/g (current density)	27
	163/50cycles/LIBs	100 mA/g (current density)	26
V₂O₅	160/100cycles/LIBs	300 mA/g (current density)	30
	135/100cycle/SIBs	320 mA/g (current density)	31
TiO₂	125/200cycle/LIBs	1/3 C rate	32
	140/60cycle/SIBs	50 mA/g (current density)	33

Reference

- [1] V. Augugliaro, S. Coluccia, V. Loddo, L. Marchese, G. Martra, L. Palmisano, M. Schiavello, *Applied Catalysis B: Environmental*, **1999**, 20, 15-27.
- [2] D. K. Lee, H.-W. Shim, J. S. An, C. M. Cho, I.-S. Cho, K. S. Hong, D.-W. Kim, *Nanoscale Res Lett*, **2010**, 5, 1585-1589.
- [3] T.-F. Yi, L. Jiang, J. Liu, M. Ye, H. Fang, A. Zhou, J. Shu, *Ionics*, **2011**, 17, 799-803.
- [4] T. Yi, Y. Xie, L. Jiang, J. Shu, C. Yue, A. Zhou, M. Ye, *RSC Advances*, **2012**, 2, 3541-3547.