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Supplementary Information

Novel Na_2TiSiO_5 anode material for lithium ion batteries

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Experiments

Material synthesis: NTSO is synthesized by a hydrothermal method. In a typical process: urea, SiO_2 and TiO_2 with a molar ratio of 0.5:1:1 were dispersed in 15 ml, 10 mol/L NaOH aqueous, then stirred and sonicated for 1 h. The mixed solution was transferred to a Teflon stainless reactor and held at 160 ℃ for 6 h. The white product was thoroughly washed by water and dried at 80 ℃.

Electrode preparation: NTSO, acetylene carbon, polyvinylidene fluoride (PVDF, dispersed in N-methyl-2-pyrrolidone solution) are mixed as a weight percent of 7:2:1. Then the slurry is coated on Cu foil by a blade and dried at 80 ℃ in a vacuum oven. The electrodes are punched into 12 mm disks for battery assembling, the loading mass is about 1 mg/cm^2. For a typical assembly process: electrode, separator (Celgard 2500) and lithium metal (as a counter electrode, diameter: 10 mm) are encapsulated into a CR2032-type coin cell in a glovebox filled with argon. The electrolyte is made up from 1 M $LiPF_6$ in EC/DEC (1:1 vol.%). The working potential ranges from 0.1 to 3 V.

Characterization: X-Ray diffraction (XRD) patterns for Rietveld analysis are recorded by D8 Advance (Bruker) setting at 40 kV/40 mA with a Cu target (Kα1 radiation, λ = 0.1541 nm, 0.8 s/step), the 2θ degree ranges from 10-120°. The Rietveld analysis is based on a Topas software for cell parameters simulation. Cyclic voltammetry (CV) curves is measured by Solartron Metrology. Battery test is performed on Neware battery test equipment. N_2 isotherm adsorption-desorption measurement is performed by an ASAP 2020 at 77 K.

Density functional theory (DFT) method[1,2] are implemented in the Vienna ab initio package (VASP) with pseudopotentials established by the projector-augmented wave (PAW)[3] method and the Perdew−Burke−Ernzerh (PBE).[4] For total energy calculations, we used a cutoff energy of 450 eV to ensure good convergence. The Brillouin zone was adopted with a 2×3×3 Γ-centered k-mesh for 1a×1b×2c supercell (Containing four Na_2TiSiO_5 molecular units). All structures were relaxed until the energy and force was less than 10−5 eV and 0.01 eV/Å, respectively. In the GGA +U schemes, U eff (U-J) is fixed to 2.5 eV for Ti-3d state. [5,6] The formation energy of insertion lithium atom is given by

$$\Delta E = \frac{E(Na_2Li_xTiSiO_5) - E(Na_2TiSiO_5) - xE(Li)}{x} \quad \text{(S1)}$$

Where E is the total energy. The E(Li) was obtained by optimization of Li Metal (Imm).

Reference

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Table S1. Refined structure of Na_2TiSiO_5 obtained from XRD data

Na_2TiSiO_5 (P4/nmm space group), $a = b = 6.504(2)$ Å, $c = 5.068(1)$ Å, $V = 214.4(1)$ Å^3						
Atoms	Site	x	y	z	Occupancy	B
Ti	2c	0.5	0	0.926(3)	1	0.3379
Na	4e	0.25	0.25	0.5	1	0.5796
Si	2a	0	0	0	1	0.2521
O	8i	0	0.127(2)	0.273(4)	1	0.6646
O	2c	0.5	0	-0.576(9)	1	0.504

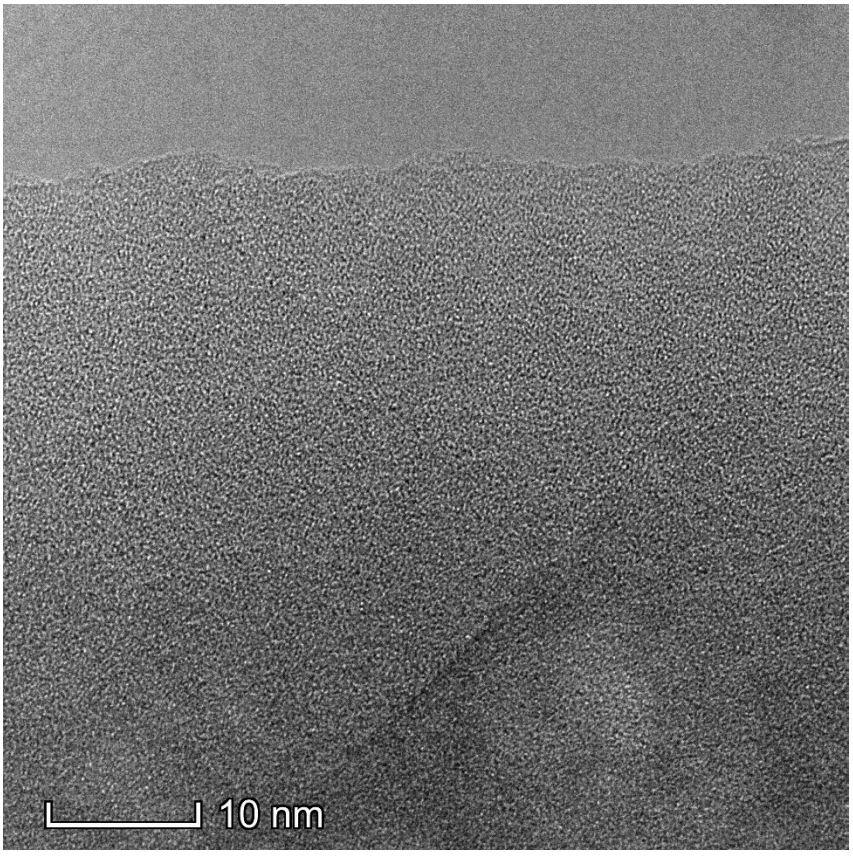

Fig. S1 TEM image of an amorphous region in NTSO.

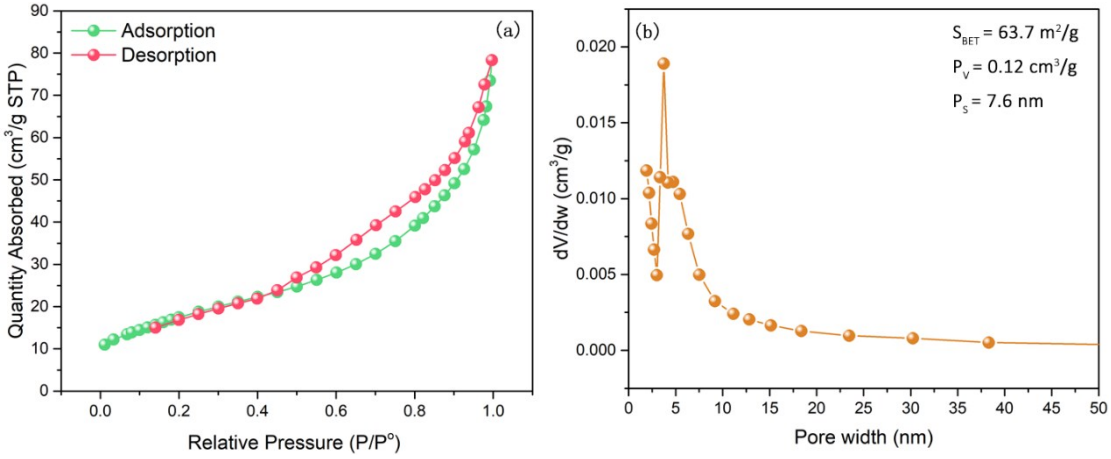

Fig. S2 (a) N_2 isothermal adsorption-desorption curves and (b) pore size distribution plot. (P_V: pore volume, P_S: pore size.)

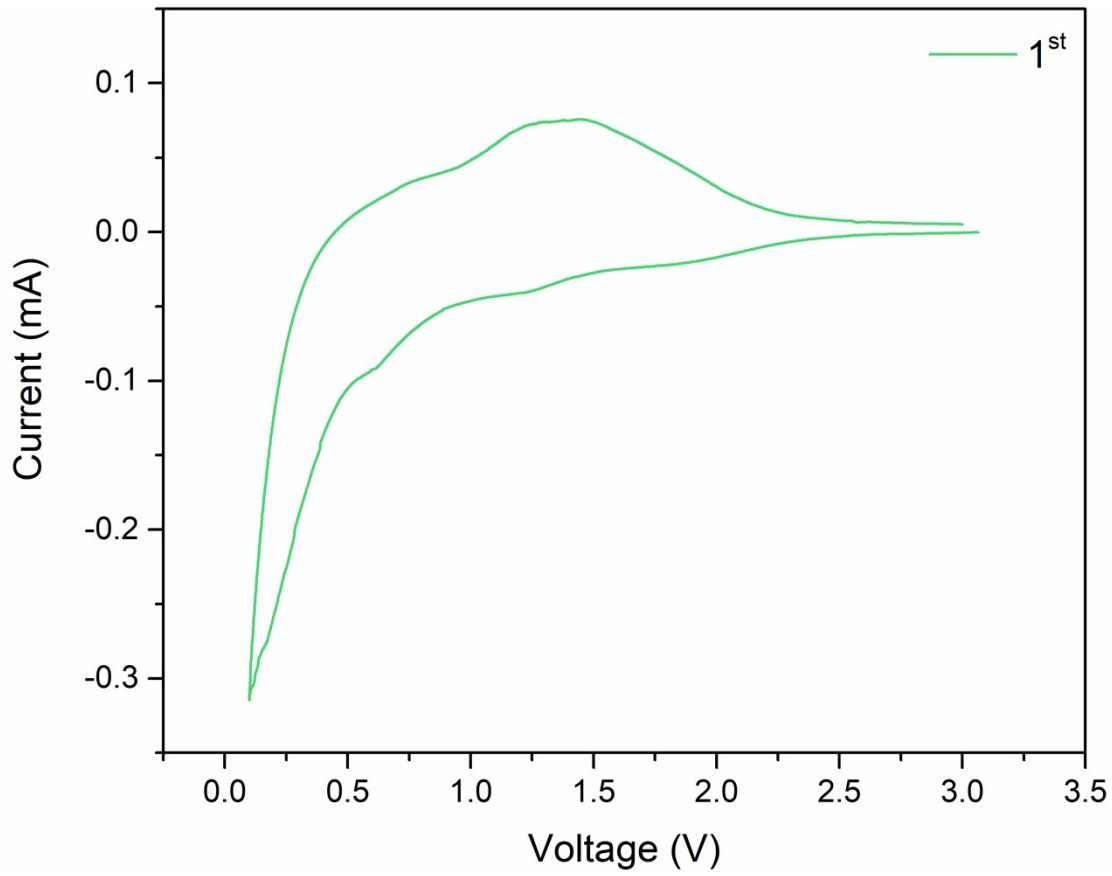

Fig. S3 first cycle of CV curves at a scan rate of 0.1 mV/s.

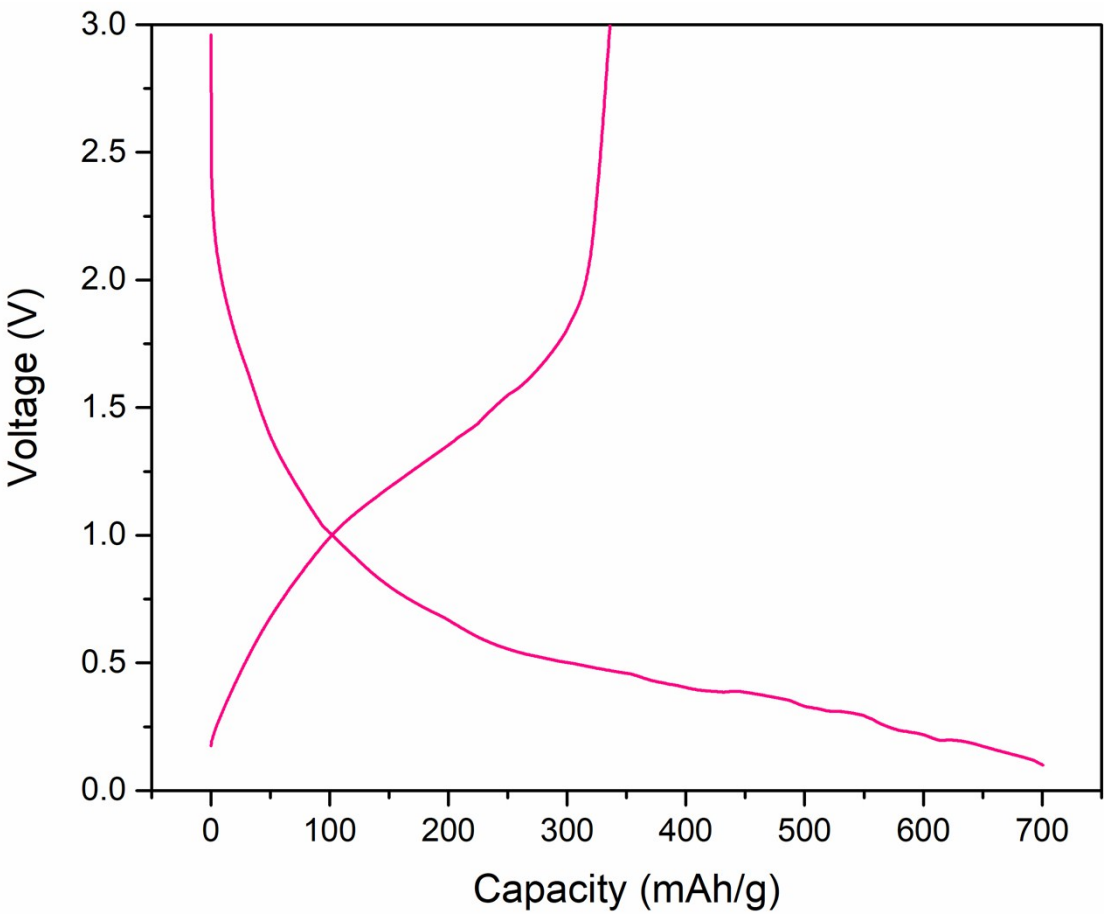

Fig. S4 the initial charge/discharge curve of NTSO at a current density of 100 mA/g.

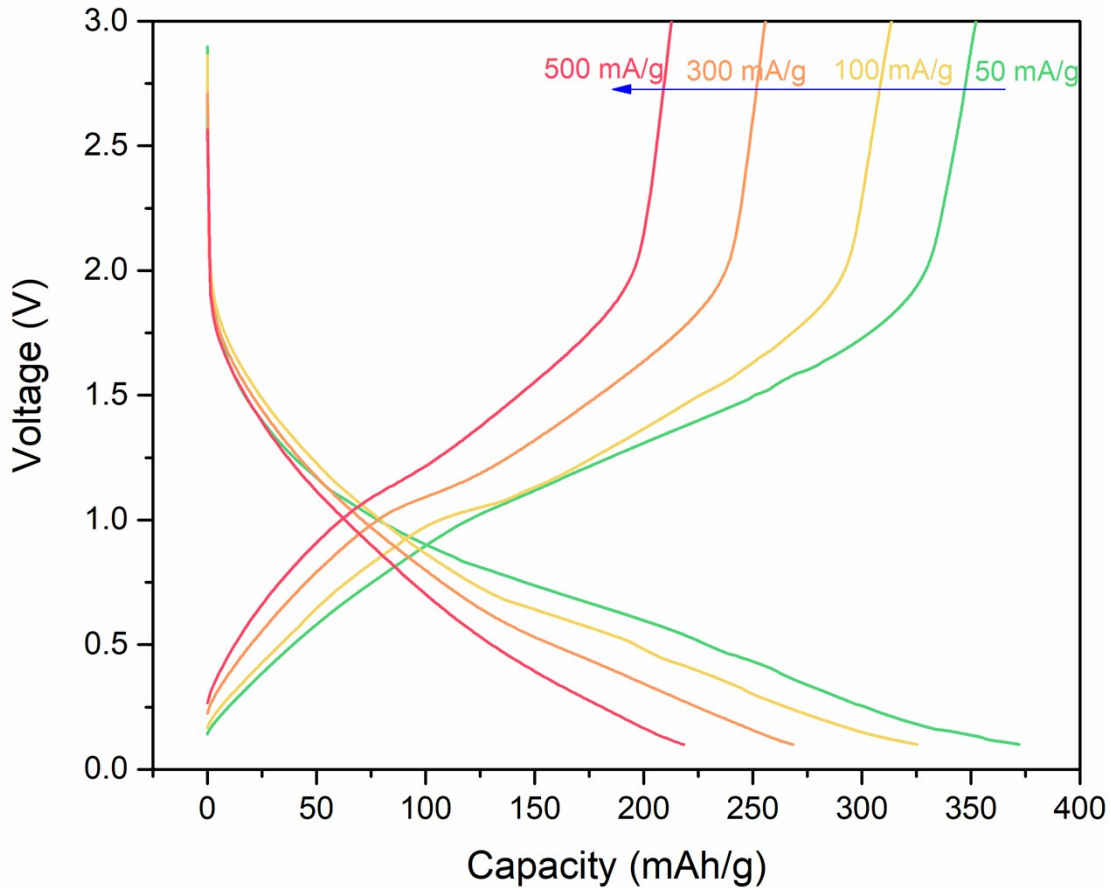

Fig. S5 the charge/discharge curves of NTSO at different current densities. The second cycle is used for the curve at 50 mA/g,

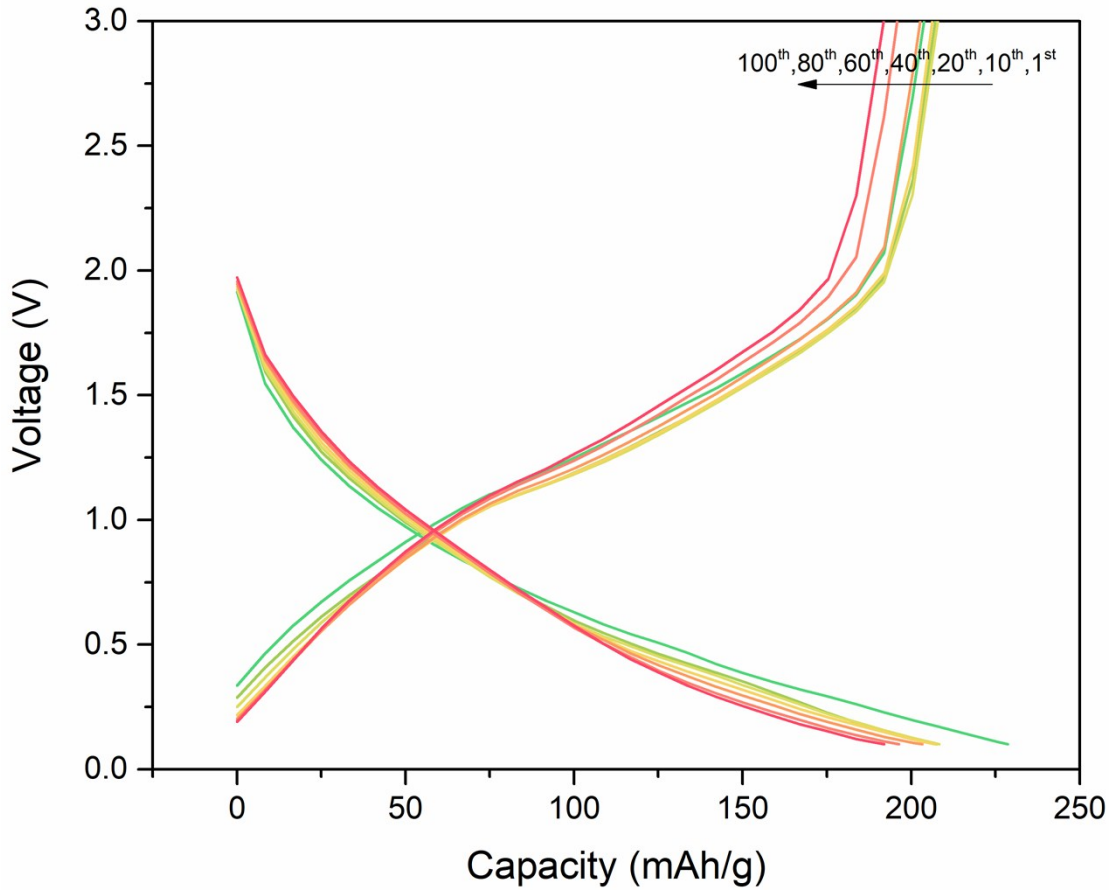

Fig. S6 the selected curves after different cycles at a current density of 500 mA/g, the interval time for date collection is bit of long and leads to the obvious incontinuity in the curves.

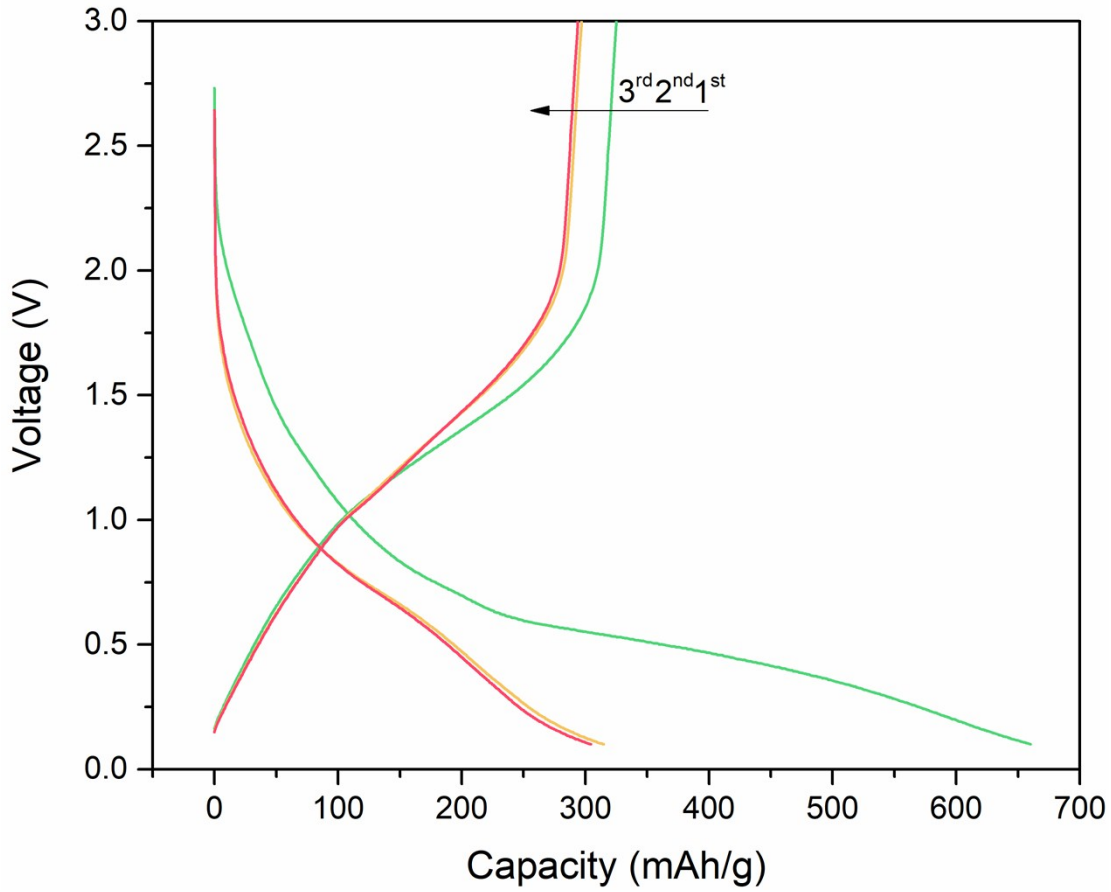

Fig. S7 first three cycles for battery activation under 20 mA/g and then tested at 500 mA/g for 100 cycles.