Supporting Information

A New Layered Titanate \( \text{Na}_2\text{Li}_2\text{Ti}_5\text{O}_{12} \) as High Performance Intercalation Anode for Sodium-Ion Batteries

Yangyang Huang\(^a\), Jinsong Wang\(^b\), Lin Miao\(^b\), Yu Jin\(^a\), Jian Peng\(^a\), Qing Li\(^a\), Chun Fang\(^a\), Jiantao Han\(^a,*\) and Yunhui Huang\(^a,*\)

\(^a\) School of Materials Science and Engineering, Huazhong University of Science and Technology, Wuhan, Hubei 430074, China

\(^b\) Wuhan National Laboratory for Optoelectronics, and School of Optical and Electronic Information, Huazhong University of Science and Technology, Wuhan 430074, China

*E-mail: jthan@hust.edu.cn, huangyh@hust.edu.cn
Figure S1. X-ray diffraction pattern of as-prepared KLT sample.

Table S1. Structural parameters of KLT.

<table>
<thead>
<tr>
<th>FORMULA $K_2Li_2Ti_5O_{12}$, SPACE GROUP $CMCM$</th>
<th>$A_{OR} = 3.8123$ (Å), $B_{OR} = 15.5912$ (Å), $C_{OR} = 2.9734$ (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ATOM</td>
<td>site</td>
</tr>
<tr>
<td>TI</td>
<td>4c</td>
</tr>
<tr>
<td>K</td>
<td>4b</td>
</tr>
<tr>
<td>LI</td>
<td>4b</td>
</tr>
<tr>
<td>O1</td>
<td>4c</td>
</tr>
<tr>
<td>O2</td>
<td>4c</td>
</tr>
</tbody>
</table>
Figure S2. HR-TEM image of NLT (inset: corresponding intensity profile for the line scan across the lattice fringes and the FFT).

Figure S3. TG curves of as-obtained KLT and NLT sample heated in air at a heating rate of 10 °C min⁻¹.
Figure S4. (a) The charge/discharge profiles of NLT in different cycles at current density of 50 mAg$^{-1}$ in the voltage range of 0.01 – 2 V. (b) Rate performance of KLT in different current densities. (c) Long-term cycling performance at a current density of 50 mAg$^{-1}$.

Figure S5. The first discharge/charge curves of NLT.
Figure S6. The charge-discharge profiles with different rate of NLT.

Figure S7. (a) Cycling stability and (b) Coulombic efficiency for sodium cells of NLT electrode prepared with NaCMC or PVDF binders, discharged/charged at 0.1 Ag⁻¹ in the voltage range of 0.01 – 2 V.
**Figure S8.** (a, b) GITT profiles of NLT for 1st and 4th at a current density of 20 mA g$^{-1}$, respectively. Diffusion coefficient calculated from the GITT profiles for the electrode at (c) first and (d) fourth cycle, respectively.

**Figure S9.** Current step diagram of desodiation process of the NLT electrode for SIBs.

Figure S8a and b show the GITT curves of NLT electrode at the first and fourth cycles in a voltage range of 0.01 – 2.0 V. During the GITT measurements, the cell is
working with a pulse current at 20 mA g$^{-1}$ for an interval of 10 min followed by an open circuit relaxation of 40 min to obtain the steady-state voltage ($E_s$). A single titration profile is illustrated in Figure S9c with schematic labelling parameters, $\Delta E_t$, $\Delta E_s$, etc. The diffusion efficient of an electrode can be estimated by Fick’s second law of diffusion with the following equation:

$$D = \frac{4}{\pi} \left( \frac{m_B V_M}{M_B S} \right)^2 \left( \frac{\Delta E_s}{\tau \left( \frac{dE_s}{d\sqrt{\tau}} \right)} \right)^2 \quad (\tau \ll l^2 / D) \quad (1)$$

Where $m_B$ is electrode active mass, $M_B$ is the molar mass of material, $V_M$ is the molar volume of the material, $S$ is the overall contract area of the electrode and electrolyte, $L$ is the electrode thickness and $\tau$ is the pulse duration. Linear behavior of the potential vs. $t^{1/2}$ relationship shown in Figure S9a and b makes it possible for us to simplify equation (1) to the following form:

$$D = \frac{4L^2}{\pi \tau} \left( \frac{\Delta E_s}{\Delta E_s} \right)^2 \quad (\tau \ll l^2 / D) \quad (2)$$

Based on equation (2), the diffusion coefficients of Na$^+$ calculated from the GITT curves as a function of the cell voltage in the first and fourth sodiation/desodiation processes are shown in Figure S8c and d.
Figure S10. The CV curves of NLT at a scan rate of 0.1 mV s$^{-1}$. 