Magnéli Phase Titanium Oxide as a Novel Anode Material for Potassium Ion Batteries

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Figure S1. (a) X-ray diffraction pattern, (b) scanning electron microscopy (SEM) images, (c) cross-sectional SEM images, and (d) high-resolution transmission electron microscopy images of TiO$_2$/CNT microspherical composites.

Figure S2. TGA curve of TiO$_2$/CNT microspherical composites.
Figure S3 shows the TEM images of the TiO$_2$-precursor/CNT composites obtained right after mixing the pristine CNTs and TiO$_2$-precursor solution. Note that 10~20 nm TiO$_2$ precursor were precipitated in contact with the pristine CNTs in the TiO$_2$-precursor/CNT composites. This could be attributable to the CH-π interactions between the alkyl groups of titanium ethoxide organometallic Ti source and the aromatic walls of pristine CNTs.$^1$
Figure S4. Electrochemical properties of CNTs electrode in 0.8 M KPF\textsubscript{6} 1:1 EC/DEC electrolyte in the potential window of 0.01–2.5 V. (a) First three charge-discharge curves, (b) dQ/dV curves based on the charge-discharge curves of (a), (c) charge/discharge curves at increasing current density from 0.05 to 3 A g\textsuperscript{-1}, (d) rate capability, and (e) cycling performance.
Figure S5. Electrochemical properties of bare TiO$_2$ electrode in 0.8 M KPF$_6$ 1:1 EC/DEC electrolyte in the potential window of 0.01–2.5 V. (a) First three charge-discharge curves, (b) dQ/dV curves based on the charge-discharge curves of (a), (c) charge/discharge curves at increasing current density from 0.05 to 1 A g$^{-1}$, (d) rate capability, and (e) cycling performance
Figure S6. HR-TEM image of fully charged Ti$_6$O$_{11}$/CNT composite.
**Figure S7.** Ex-situ XRD patterns of the bare TiO$_2$ electrode for selected states of charge in the (a) first and (b) second cycle in the potential window of 0.01 – 2.5 V vs K/K$^+$ at a current density of 0.05 A g$^{-1}$.

**Figure S8.** HR-TEM image of bare TiO$_2$. 
**Figure S9.** (a) Cycling performance and (b) ex-situ XRD patterns of the Ti$_6$O$_{11}$/CNT electrode for selected cycles in the potential window of 0.01 – 2.5 V vs K/K$^+$ at a current density of 0.05 A g$^{-1}$.

**Figure S10.** SEM images of the Ti$_6$O$_{11}$/CNT composite electrode (a) in pristine state (b) after 500 cycle.
Table S1 Comparison of potassium ion storage performance of Ti$_6$O$_{11}$/CNT composite with other oxide-based anodes$^{2-4}$

| Active materials | Reversible Capacity | Rate performance | Cycle performance | Reference |
|------------------|--------------------|------------------|------------------|-----------|
| Ti$_6$O$_{11}$/CNT composite | 148 mAh g$^{-1}$ at 50 mA g$^{-1}$ | 91 mAh g$^{-1}$ at 500 mA g$^{-1}$ | 74 mAh g$^{-1}$ at 200 mA g$^{-1}$  500 cycles (76%, 1$^{st}$ cycle) | This work |
| K$_2$Ti$_8$O$_{17}$ | 118 mAh g$^{-1}$ at 20 mA g$^{-1}$ | 44 mAh g$^{-1}$ at 500 mA g$^{-1}$ | 110 mAh g$^{-1}$ at 20 mA g$^{-1}$  50 cycles (63%, 2$^{nd}$ cycle) | [2] |
| K$_2$Ti$_4$O$_9$ | 74 mAh g$^{-1}$ at 30 mA g$^{-1}$ | 79 mAh g$^{-1}$ at 100 mA g$^{-1}$ | 37 mAh g$^{-1}$ at 100 mA g$^{-1}$  30 cycles (47%, 2$^{nd}$ cycle) | [3] |
| K$_2$Ti$_4$O$_9$ | 142 mAh g$^{-1}$ at 20 mA g$^{-1}$ | 81 mAh g$^{-1}$ at 300 mA g$^{-1}$ | 44 mAh g$^{-1}$ at 50 mA g$^{-1}$  100 cycles (61%, 2$^{nd}$ cycle)  47 mAh g$^{-1}$ at 200 mA g$^{-1}$  900 cycles (51%, 2$^{nd}$ cycle) | [4] |

Reference

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