

Supporting Information

Electrochemical Oscillation during Galvanostatic Charging and Discharging of Zr-modified $\text{Li}_4\text{Ti}_5\text{O}_{12}$ in Li-Ion Batteries

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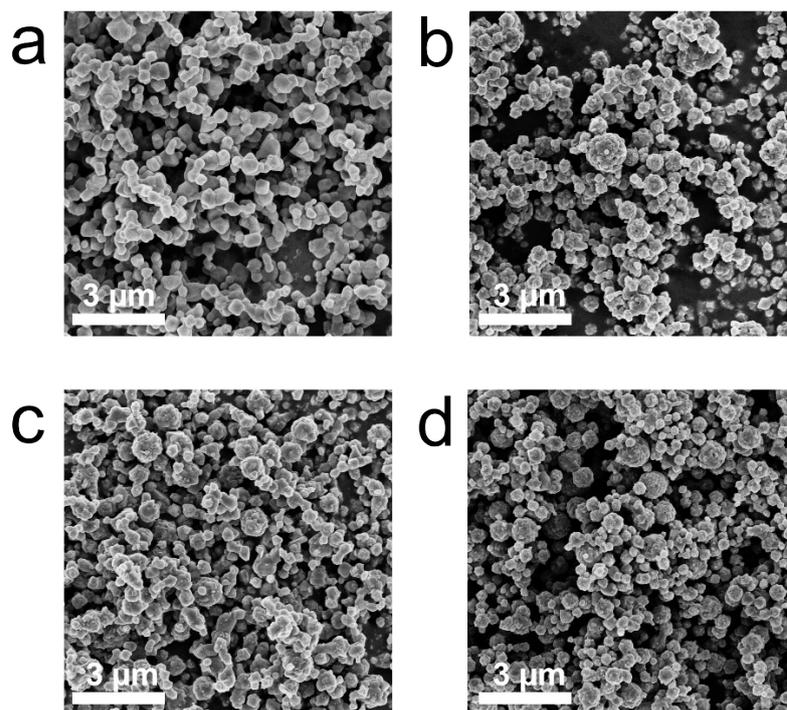


Figure S1. The SEM images of $\text{Li}_{4.4}\text{Ti}_5\text{O}_{12}$ (a), $\text{Li}_{4.4}\text{Ti}_{4.75}\text{Zr}_{0.25}\text{O}_{12}$ (b), $\text{Li}_{4.4}\text{Ti}_{4.5}\text{Zr}_{0.5}\text{O}_{12}$ (c) and $\text{Li}_{4.4}\text{Ti}_{4.25}\text{Zr}_{0.75}\text{O}_{12}$ (d) with an element ratio of Li:Ti:Zr=4.4:5:0, 4.4:4.75:0.25, 4.4:4.5:0.5 and 4.4:4.25:0.75 in the raw materials, respectively, which are sintered at 800 °C in air for 8 h.

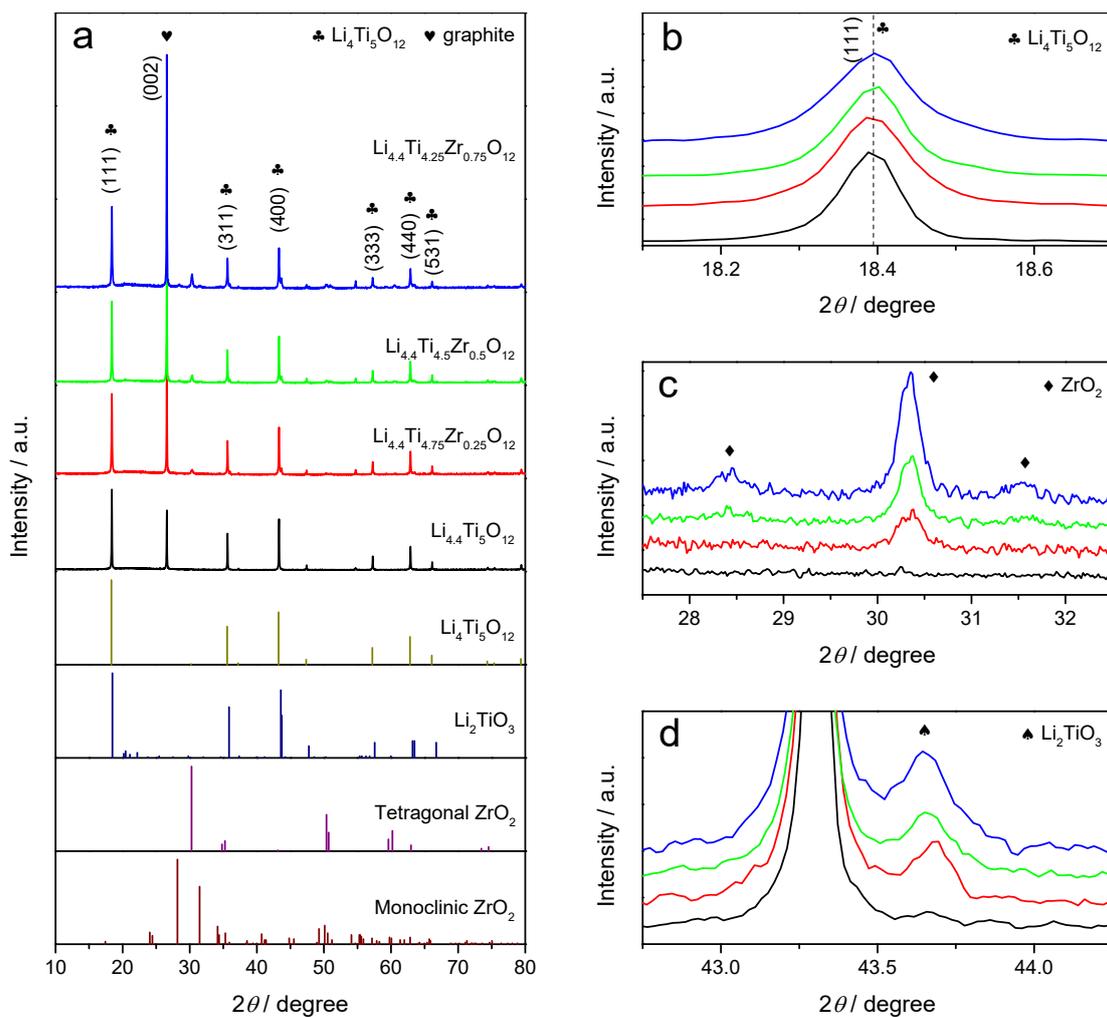


Figure S2. (a) The XRD patterns of $\text{Li}_{4.4}\text{Ti}_5\text{O}_{12}$ (black), $\text{Li}_{4.4}\text{Ti}_{4.75}\text{Zr}_{0.25}\text{O}_{12}$ (red), $\text{Li}_{4.4}\text{Ti}_{4.5}\text{Zr}_{0.5}\text{O}_{12}$ (green) and $\text{Li}_{4.4}\text{Ti}_{4.25}\text{Zr}_{0.75}\text{O}_{12}$ (blue), and the enlarged XRD peaks for $\text{Li}_4\text{Ti}_5\text{O}_{12}$ (b), ZrO_2 (c) and Li_2TiO_3 (d) phases. Here, all XRD patterns are calibrated by the (002) peak of graphite.

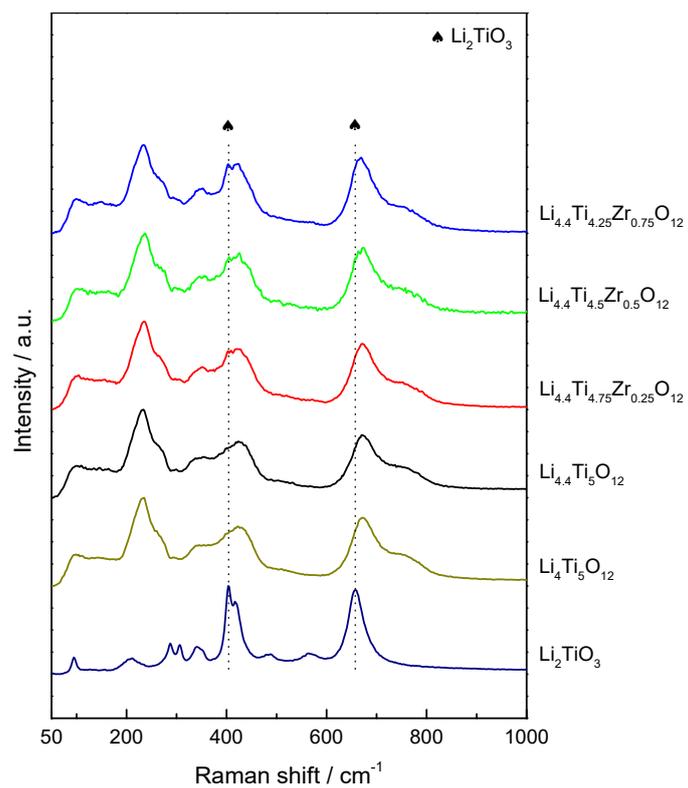


Figure S3. The Raman spectra of $\text{Li}_{4.4}\text{Ti}_5\text{O}_{12}$ (black), $\text{Li}_{4.4}\text{Ti}_{4.75}\text{Zr}_{0.25}\text{O}_{12}$ (red), $\text{Li}_{4.4}\text{Ti}_{4.5}\text{Zr}_{0.5}\text{O}_{12}$ (green) and $\text{Li}_{4.4}\text{Ti}_{4.25}\text{Zr}_{0.75}\text{O}_{12}$ (blue), as well as the Raman spectra of $\text{Li}_4\text{Ti}_5\text{O}_{12}$ (dark yellow) and Li_2TiO_3 (navy) phases for comparison.

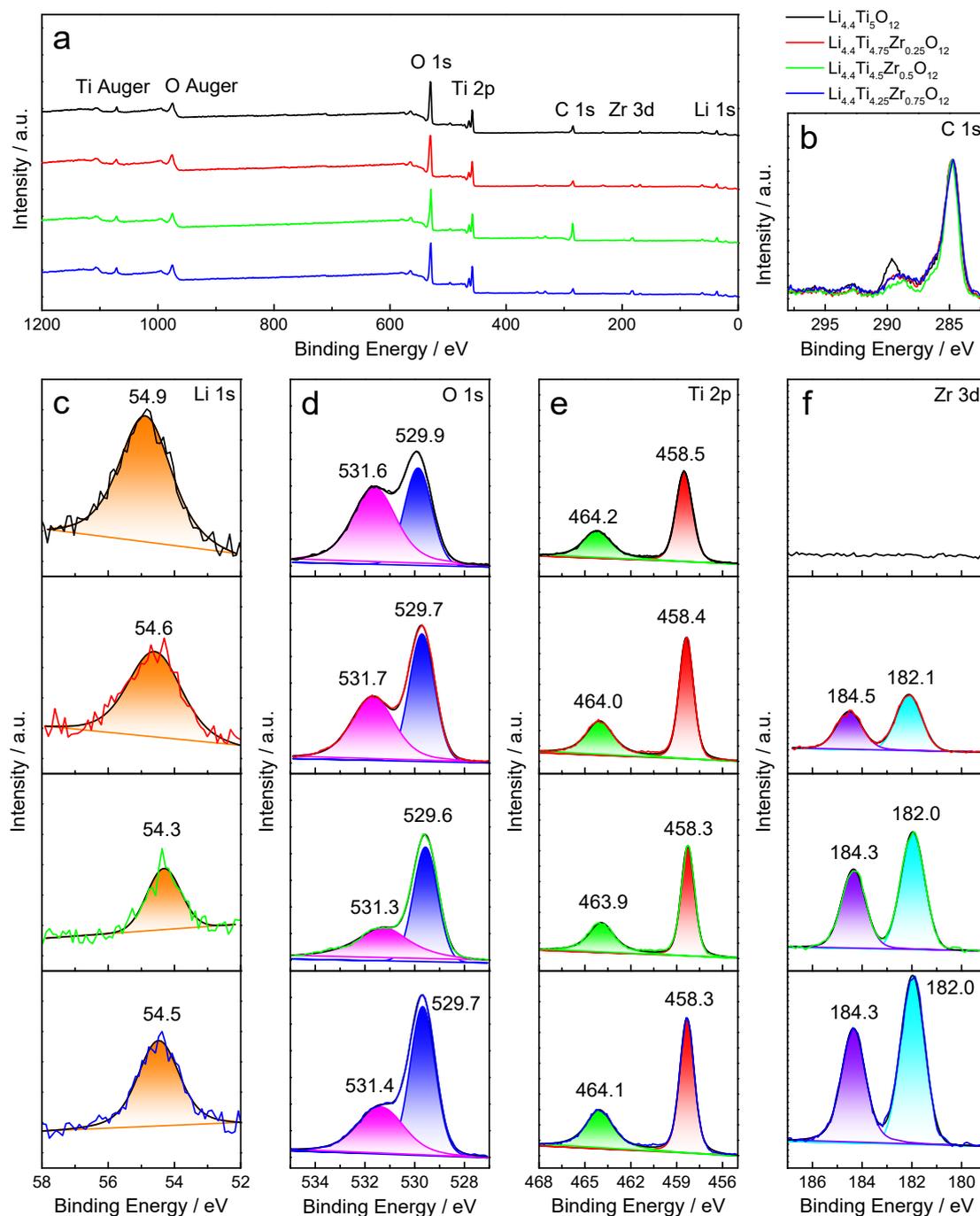


Figure S4. The measured and fitted XPS spectra of $\text{Li}_{4.4}\text{Ti}_5\text{O}_{12}$ (black), $\text{Li}_{4.4}\text{Ti}_{4.75}\text{Zr}_{0.25}\text{O}_{12}$ (red), $\text{Li}_{4.4}\text{Ti}_{4.5}\text{Zr}_{0.5}\text{O}_{12}$ (green) and $\text{Li}_{4.4}\text{Ti}_{4.25}\text{Zr}_{0.75}\text{O}_{12}$ (blue): (a) survey, (b) C 1s, (c) Li 1s, (d) O 1s, (e) Ti 2p and (f) Zr 3d.

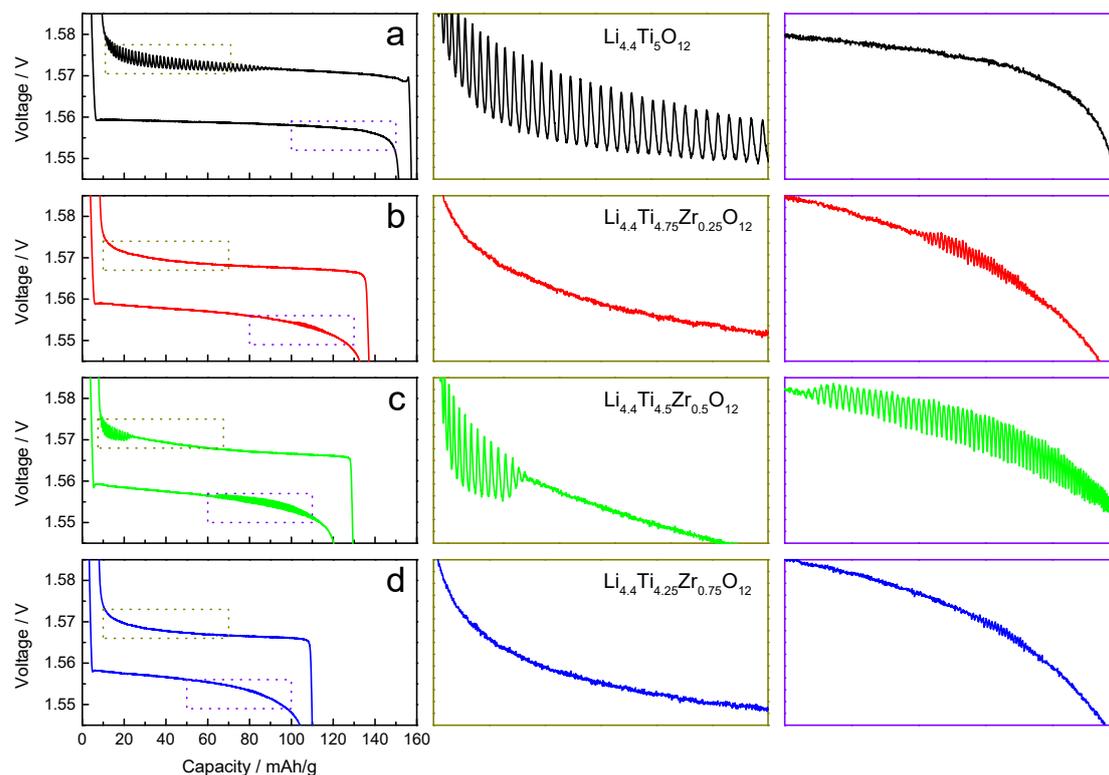


Figure S5. The discharge and charge curves of $\text{Li}_{4.4}\text{Ti}_5\text{O}_{12}$ (a), $\text{Li}_{4.4}\text{Ti}_{4.75}\text{Zr}_{0.25}\text{O}_{12}$ (b), $\text{Li}_{4.4}\text{Ti}_{4.5}\text{Zr}_{0.5}\text{O}_{12}$ (c) and $\text{Li}_{4.4}\text{Ti}_{4.25}\text{Zr}_{0.75}\text{O}_{12}$ (d) with a voltage range of 1.2–2.0 V vs. Li^+/Li and a current rate of 0.1 C, and the middle and right insets show the corresponding enlarged views in the end of charge and discharge plateaus, respectively.

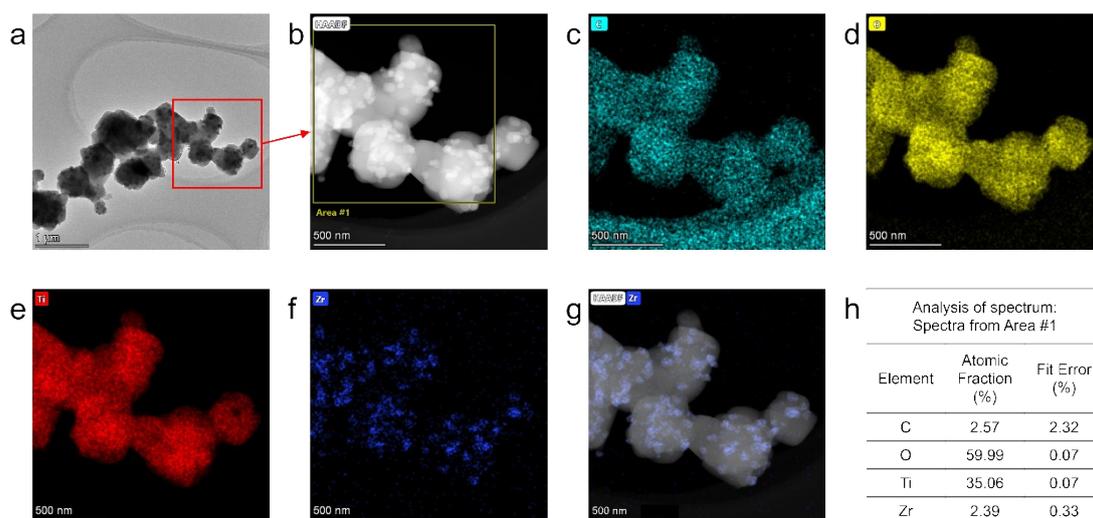


Figure S6. (a) The STEM image of $\text{Li}_{4.4}\text{Ti}_{4.5}\text{Zr}_{0.5}\text{O}_{12}$, the enlarged HAADF-STEM image (b) and the corresponding EDX maps of C (c), O (d), Ti (e), Zr (f), Zr overlapping the HAADF-STEM image (g) and element analysis of spectrum (h).

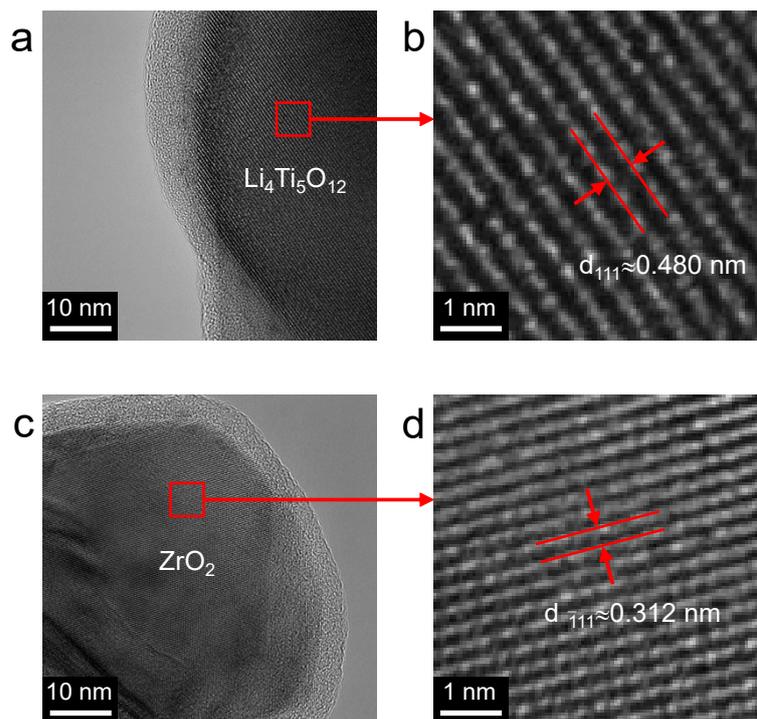


Figure S7. The high-resolution STEM images of $\text{Li}_{4.4}\text{Ti}_{4.5}\text{Zr}_{0.5}\text{O}_{12}$ for the $\text{Li}_4\text{Ti}_5\text{O}_{12}$ phase (a, b) and the ZrO_2 phase (c, d).

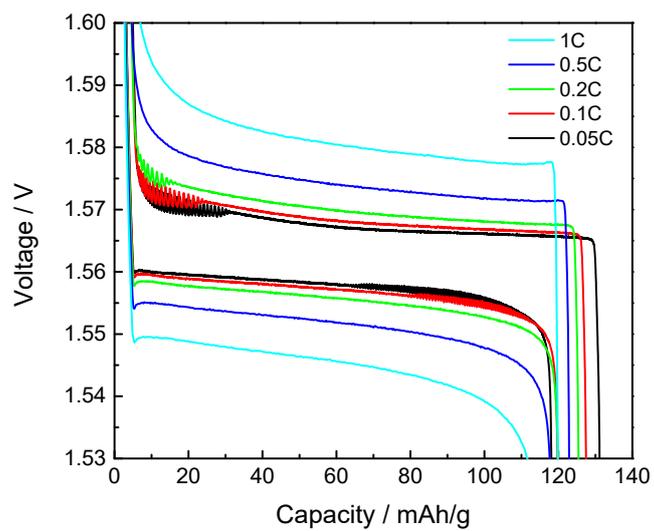


Figure S8. The discharge and charge curves of $\text{Li}_{4.4}\text{Ti}_{4.5}\text{Zr}_{0.5}\text{O}_{12}$ at different rates of 1C, 0.5C, 0.2C, 0.1C and 0.05C.

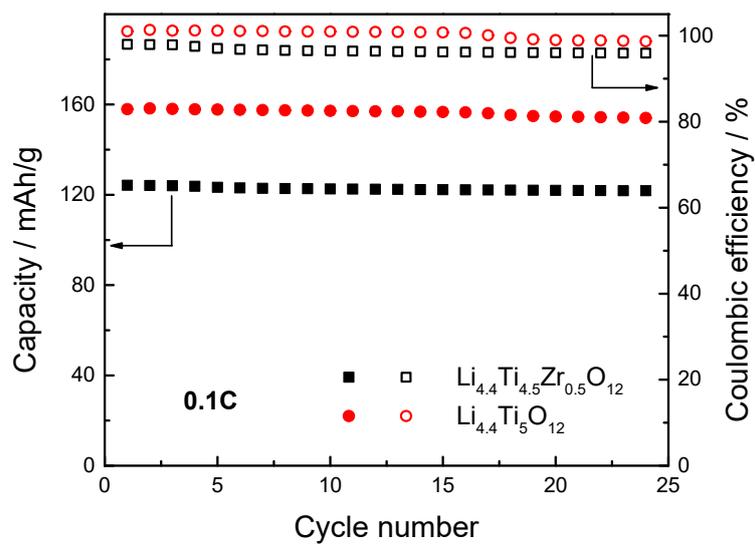


Figure S9. The Cycle performance of $\text{Li}_{4.4}\text{Ti}_5\text{O}_{12}$ and $\text{Li}_{4.4}\text{Ti}_{4.5}\text{Zr}_{0.5}\text{O}_{12}$ with a voltage range of 1.2–2.0 V vs. Li^+/Li and a current rate of 0.1 C.

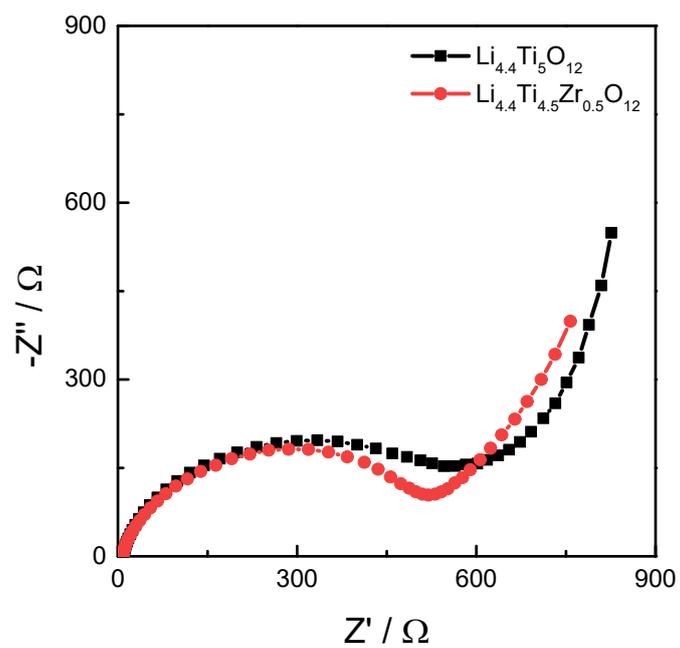


Figure S10. Nyquist plots of EIS spectra for $\text{Li}_{4.4}\text{Ti}_5\text{O}_{12}$ and $\text{Li}_{4.4}\text{Ti}_{4.5}\text{Zr}_{0.5}\text{O}_{12}$.