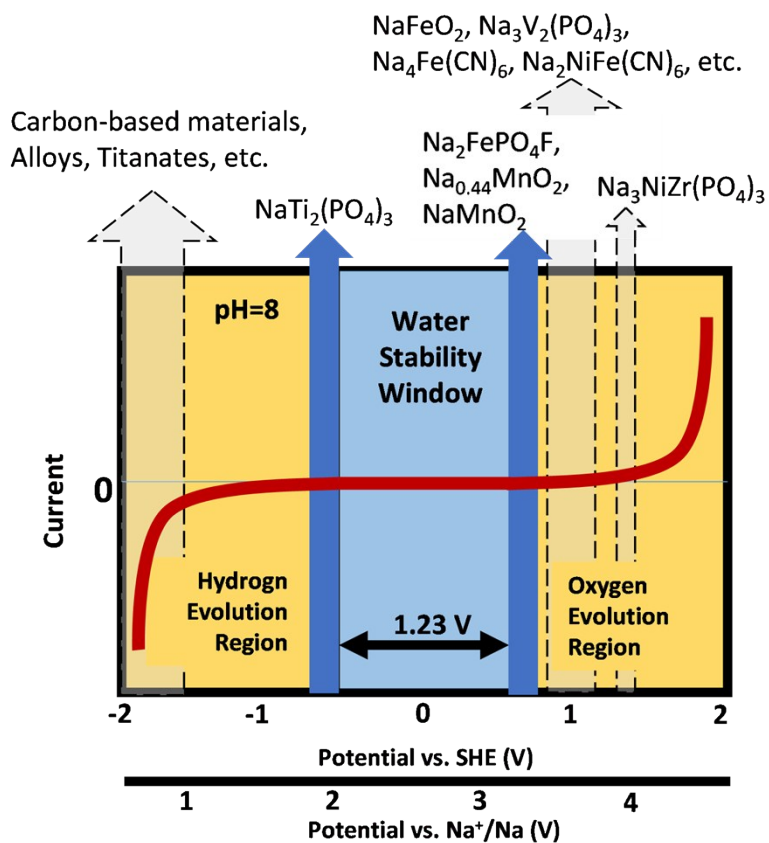
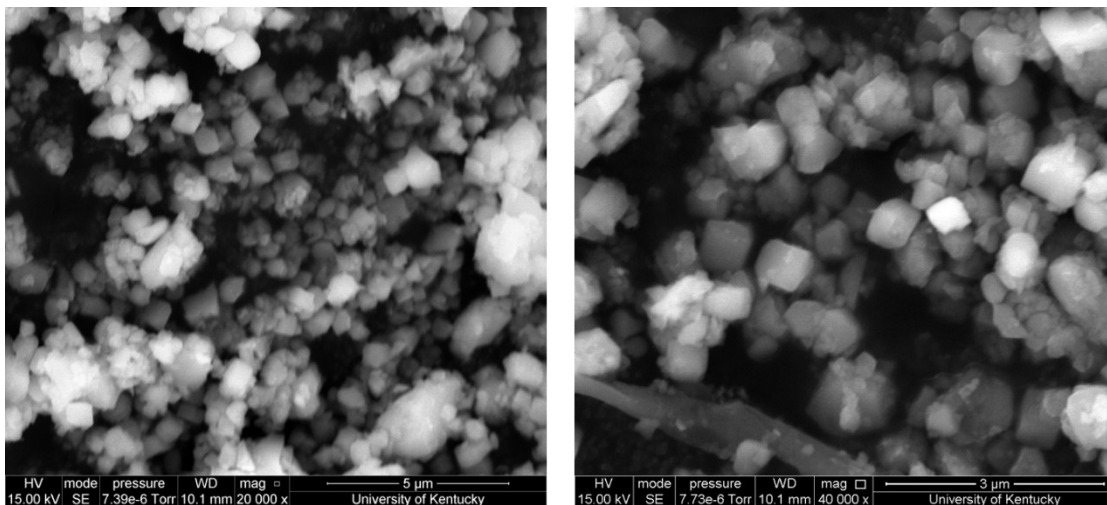


**Supplementary Information for**  
**Evolution of Solid/Aqueous Interface in Aqueous Sodium-Ion Batteries**

Xiaowen Zhan and Mona Shirpour

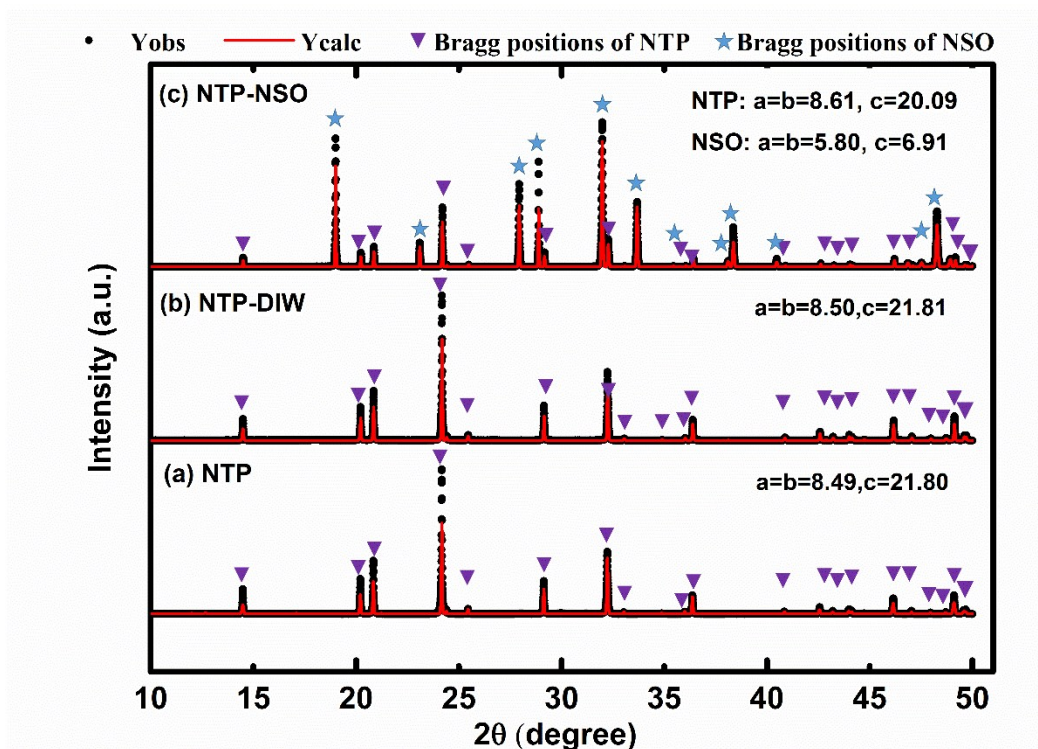


**Fig. S1.** Voltage stability window of water; the highlighted regions show the operating voltage of most of studied cathode and anode materials for sodium-ion cells.<sup>1-15</sup>



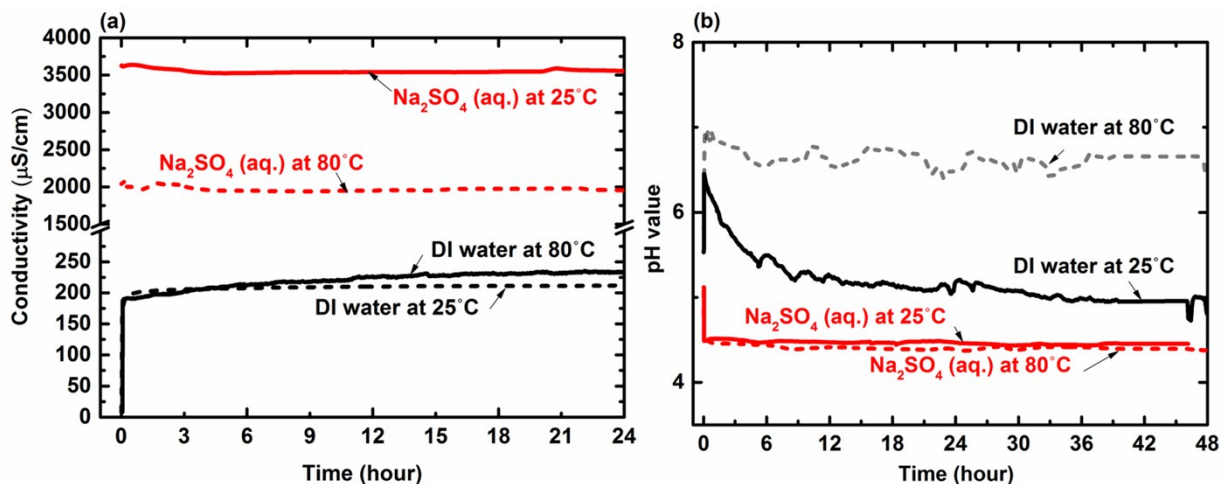
**Fig. S2.** SEM (Quanta 250) images of as-synthesized  $\text{NaTi}_2(\text{PO}_4)_3$

Synchrotron-based high-resolution X-ray diffraction (HRXRD) patterns of NTP powder in water and in 1M Na<sub>2</sub>SO<sub>4</sub> solutions were collected on beamline 11-BM at the Advanced Photon Source at Argonne National Laboratory. The samples were loaded into Kapton capillaries and rotated during the experiment. The measurement conditions were 295.0 K, a wavelength of 0.414161 Å, and a 2θ step-size of 0.001° from -6.5° to 28.0°. HRXRD patterns were analyzed using a full pattern fitting technique using the General Structure Analysis System (GSAS) with the EXPGUI interface (Fig. 3).<sup>16, 17</sup>



**Fig. S3.** Synchrotron-based high-resolution X-ray diffraction (HRXRD) patterns of (a) as-made NTP, (b) NTP powder after stirring in DI water, and (c) NTP powder after stirring in Na<sub>2</sub>SO<sub>4</sub> solution. The 2θ is converted to corresponding angle for λ = 1.54 Å (Cu-Kα) from the real wavelength λ = 0.414161 Å used for synchrotron XRD experiments.

The conductivity of suspensions containing ~2 wt% of as-prepared NTP powder in deionized water and in 1M Na<sub>2</sub>SO<sub>4</sub> aqueous solution was monitored using a Vernier conductivity probe (Order code: CON-BTA) at room temperature and at 80° C. The pH evolution was similarly recorded using a Vernier pH sensor (Order code: PH-BTA; accuracy: ±0.2 pH units).



**Fig. S4.** (a) Conductivity and (b) pH values at room temperature and at 80° C for the aqueous suspensions as a function of time.

A Varian Vista Pro Inductively Coupled Plasma/Optical Emission Spectrometry (ICP-OES) was used to determine the release of cations from NTP powders into solutions.

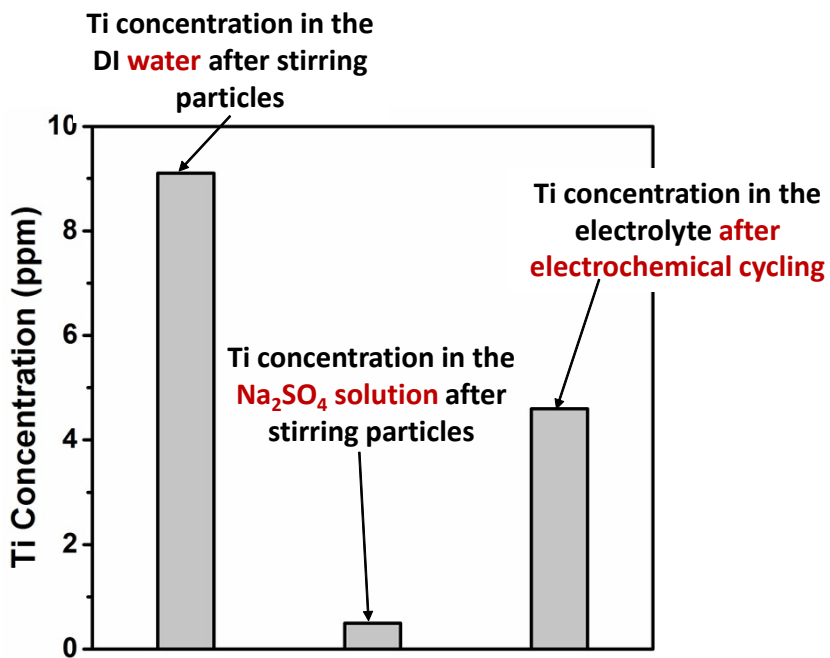


Fig. S5. Release of Ti cations from NTP powders into solutions.

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