High-Power Recycling: Upcycling to the Next Generation of High-Power Anodes for Li-ion Battery Applications

Supporting Information

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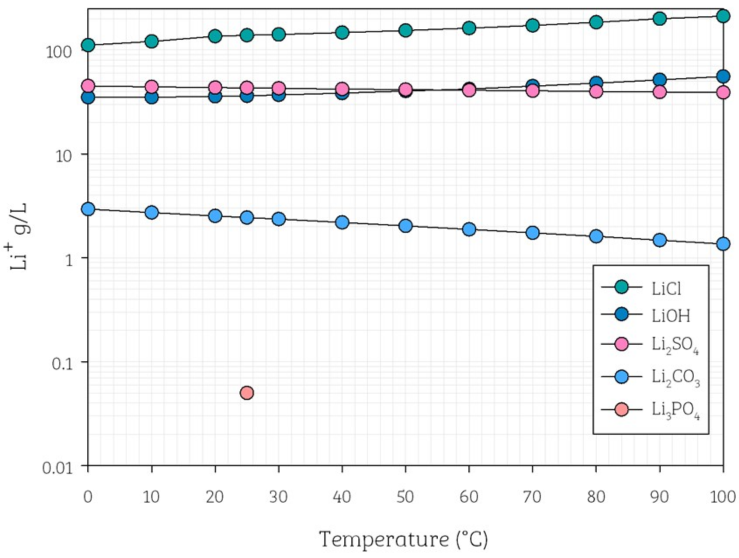
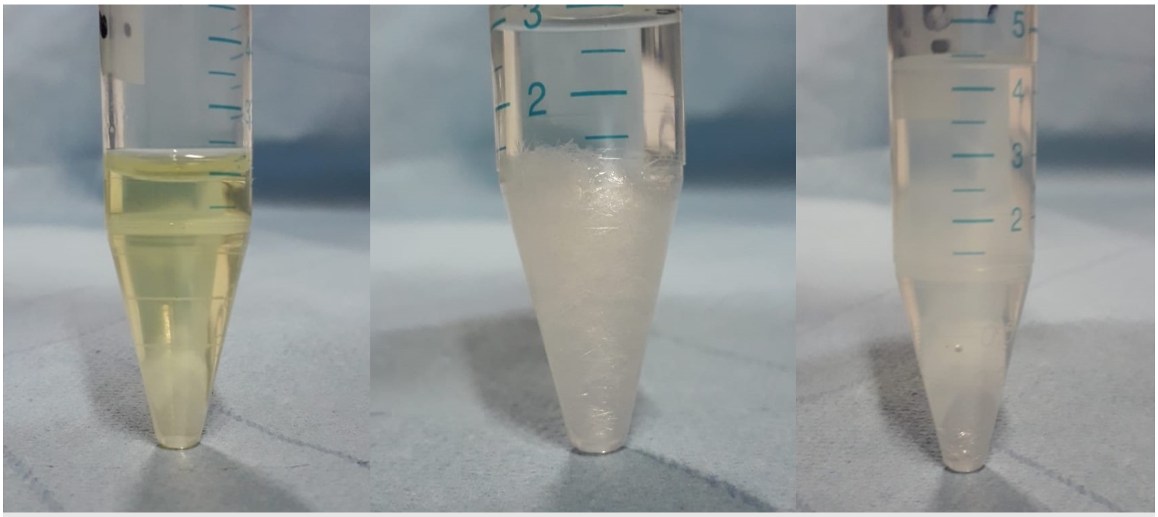
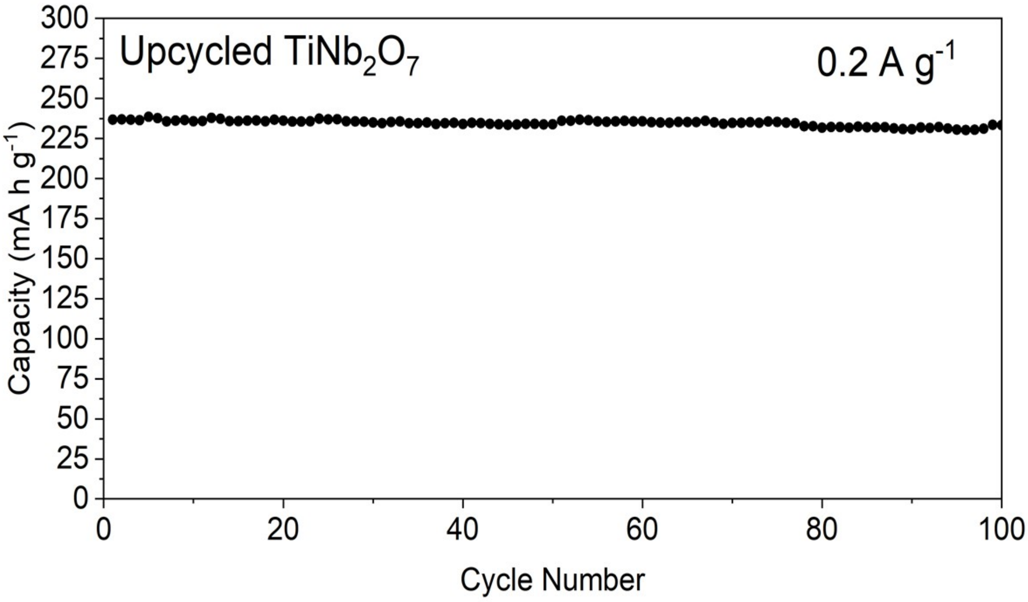


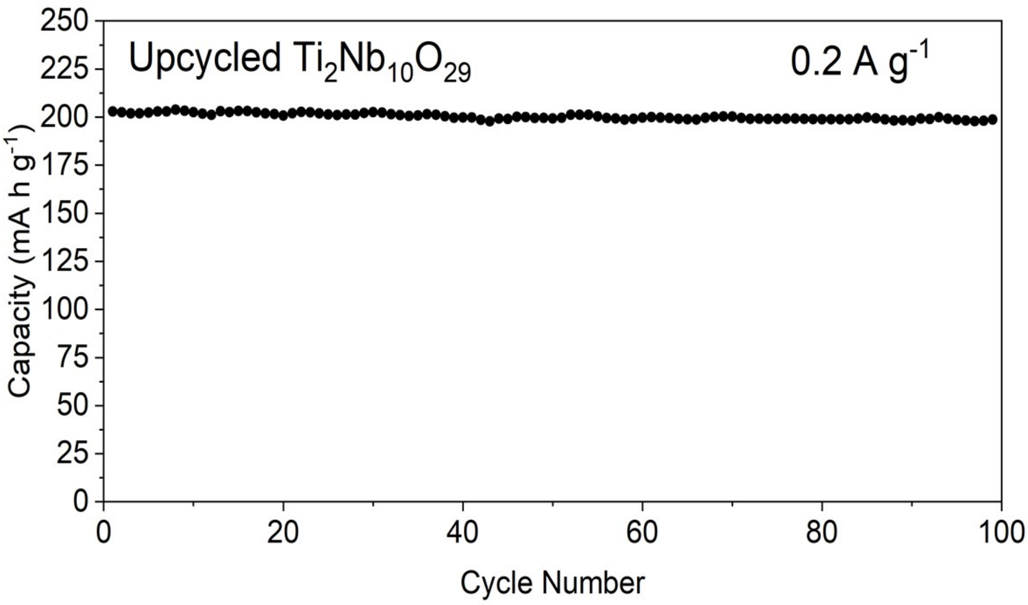
SI Figure 1: Solubility of lithium content vs temperature, for a variety of common lithium salts.



SI Figure 2: Photographs of the resulting solutions from acid exchange of the pristine LTO and the concentration step. The exchanges conducted with HCl (left), H_2SO_4 (middle) and H_3PO_4 (right).

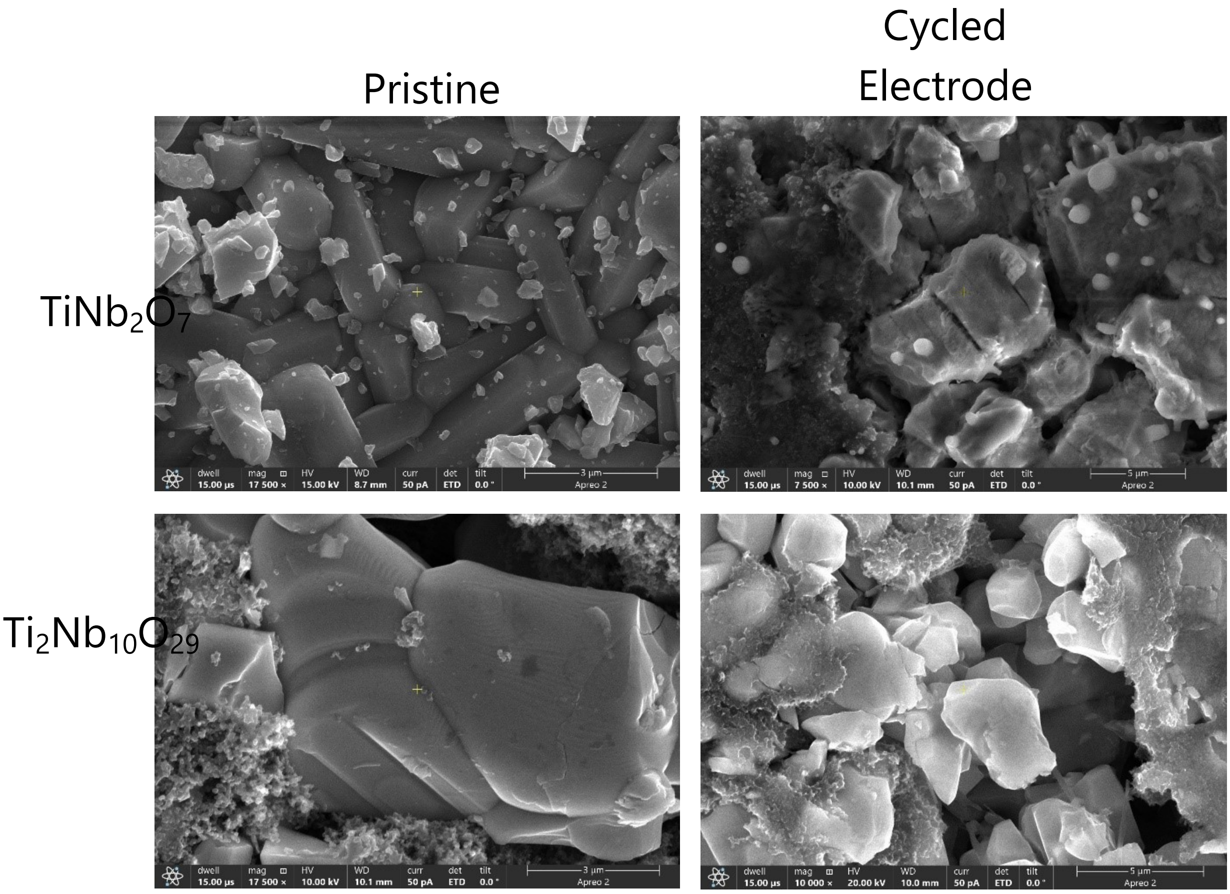


SI Figure 3: Long term cycling data for upcycled TNO. 100 cycles were done at a current density of 0.2 A g^{-1}.



SI Figure 4: Long term cycling data for upcycled $Ti_2Nb_{10}O_{29}$. 100 cycles were done at a current density of 0.2 A g^{-1}.

SEM images of the pristine powders/electrodes and the cycled electrodes, accessed from disassembled cells were taken (**SI Figure 5**). The images were collected on a ThermoFisher Scientific Apreo 2 SEM. Cracking of the $TiNb_2O_7$ is visible on the cycled electrode image, which can be attributed to the production of soft shorts resulting in capacity uplift on the return to 0.1 A g^{-1} after rate cycling.



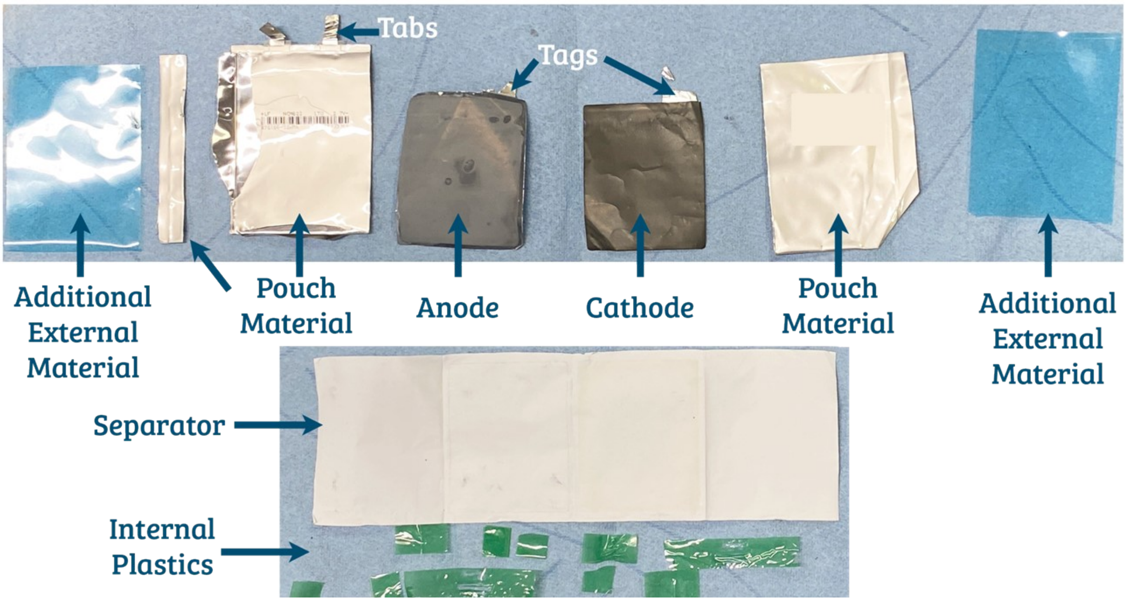
SI Figure 5:SEM images taken of the upcycled Ti-Nb-O materials. From top left clockwise: pristine $TiNb_2O_7$ powder, cycled electrode of $TiNb_2O_7$, cycled electrode of $Ti_2Nb_{10}O_{29}$, and a pristine electrode of $Ti_2Nb_{10}O_{29}$.

Application (Commercial Cell)

To confirm the process and validity of the upcycling process, a commercial 32 mA h pouch cell (with dimensions of 108.9 mm x 66.5 mm) of LTO – NMC622 cell was purchased from LiFun (China). The active material: carbon black: binder (AM:CB:B) ratio of the LTO was reported by the supplier to be 93:3:4. The cell was cycled extensively before being manually disassembled into its constituent cell components to access the LTO anode, with the cell at 0% state of charge.

The single stack pouch cell was manually dismantled, through cutting of the pouch material around the blue plastic region (pouch cell visible in **SI Figure 6**). The single sheets of electrode material (on an aluminium current collector) were removed, along with the separator and pouch material. The cathode (NMC622), anode (LTO) and the plastics, were placed individually into a beaker and completely submerged with IPA for 20 minutes, to remove any electrolyte residue. The materials were then placed into a bespoke heated vacuum temperature vessel, where all components were dried at ca. 75°C at 100 mbar overnight. The electrodes and plastics after this process are shown in **SI Figure 7**. The anode coating was delaminated from its current collector through a water-ultrasonic delamination process, before the addition of HCl acid to leach the lithium. The collected powder was dried and heated to 650°C to remove the carbon additives to produce anatase (TiO_2). Due to the small size of the pouch cell, and the low anode loading, limited material was obtained to allow for upcycling to Ti-Nb-O phases and electrochemical testing.

SI Figure 6: Photograph of the commercial LTO-NMC622 commercial cell supplied from LiFun, after extensive cycling and before manual dismantling.



SI Figure 7: Photograph of the electrodes and the pouch material/separator after the dismantling process.