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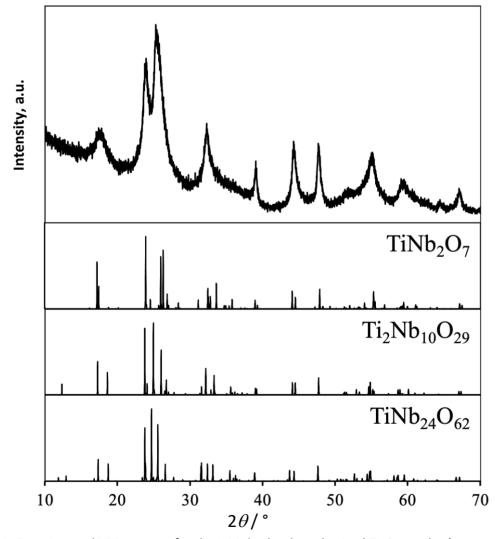
Supplementary material to

## Impact of carbon coating processing using sucrose for thick binder-free titanium niobium oxide lithium-ion battery anode

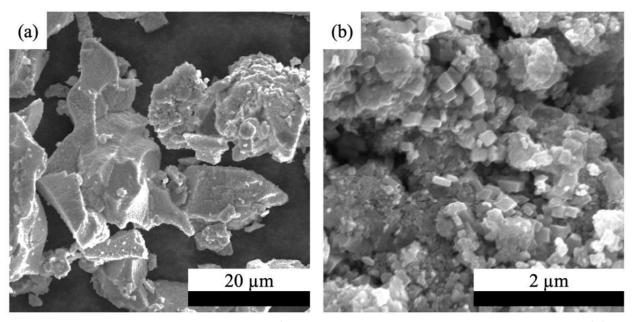
Chen Cai, Patrick McCormack, Ziyang Nie, Gary M. Koenig Jr. <sup>™</sup>

Department of Chemical Engineering, University of Virginia, 102 Engineers Way, Charlottesville, 22904-4741, VA, USA

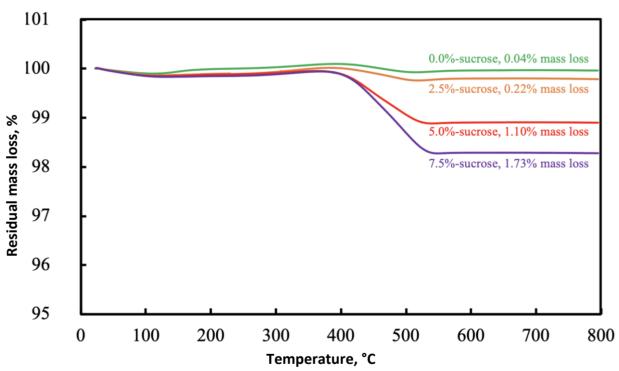
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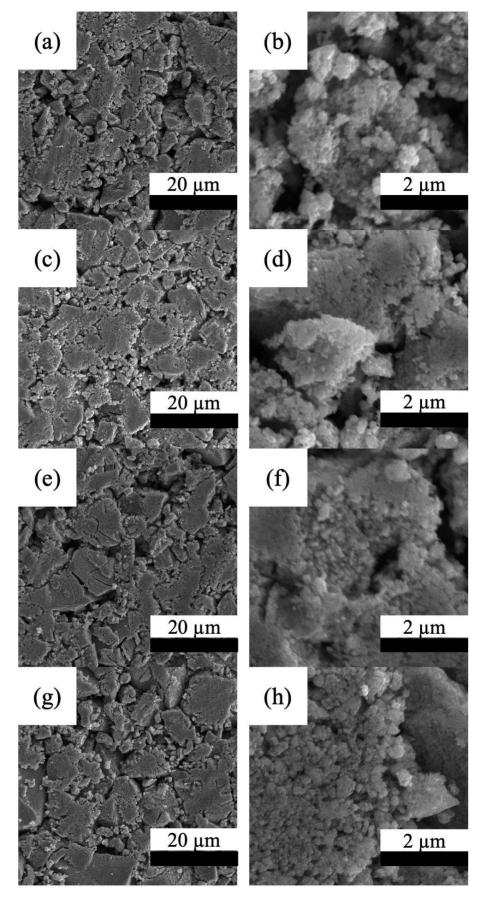
**Figure S-1.** Experimental XRD pattern for the initial sol-gel synthesized TNO powder (top pattern) and relevant indicated reference patterns [1–3]



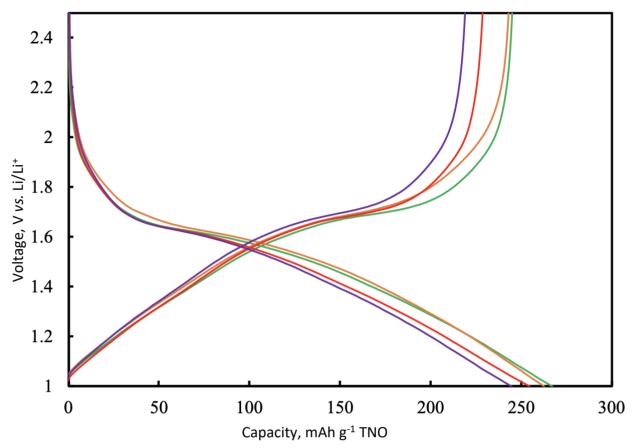
**Figure S-2**. Scanning electron micrographs of initial sol-gel synthesized TNO powder at relatively (a) lower and (b) higher magnifications



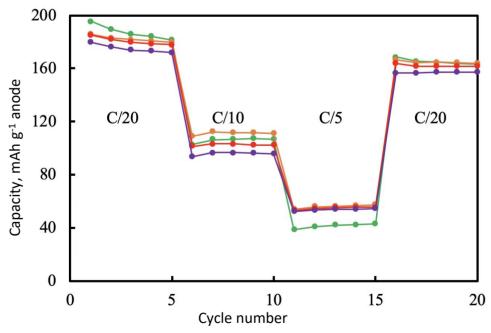
**Figure S-3**. Thermogravimetric analysis of the BF pellets without sucrose (green), and with 2.5 % (orange), 5.0 % (red), and 7.5 % (purple) sucrose used when processing the pellet before the thermal treatment step. The mass losses indicated on the plot were calculated from the mass differences at 800 °C and at room temperature



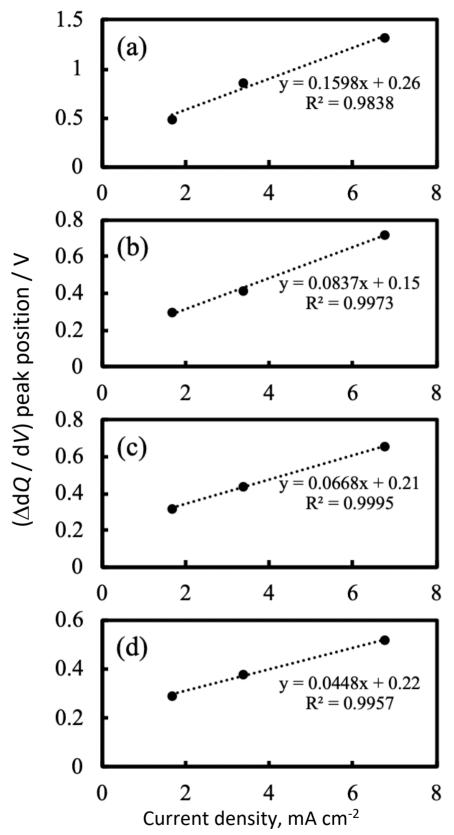
**Figure S-4.** Scanning electron micrographs of the surfaces of TNO pellets processed without sucrose at relatively (a) lower and (b) higher magnifications, 2.5 % sucrose at relatively (c) lower and (d) higher magnifications, 5.0 % sucrose at relatively (e) lower and (f) higher magnifications, 7.5 % sucrose at relatively (g) lower and (h) higher magnifications



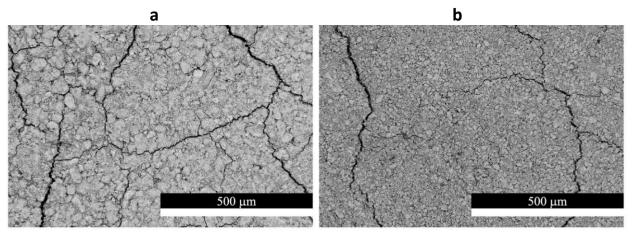
**Figure S-5**. First discharge and charge cycles without sucrose (green), and with 2.5 % (orange), 5.0 % (red) and 7.5 % (purple) sucrose TNO materials in composite cathodes paired with Li metal anodes. The charge and discharge rate were both C/20 (C/20 ranged between 4.1-6.9 mA cm<sup>-2</sup> depending on actual active material loading in each cell). The voltage window was 1.0-2.5 V (vs. Li/Li<sup>+</sup>)



**Figure S-6**. Discharge capacity at different rates (e.g. rate capabilities) for cells containing TNO BF anodes without sucrose (green) and 2.5 % (orange), 5.0 % (red) and 7.5 % (purple) sucrose cycled between 1.0-3.2 V paired with LCO BF cathodes. LCO mass loading was ~1.9 times greater than any of the TNO anodes to ensure an anode limited system. C/20 corresponded to a current density of ~1.7 mA cm<sup>-2</sup>. All charge and discharge cycles were at the same rate



**Figure S-7**.  $\Delta dQ/dV$  peak position as a function of current densities of cells with BF LCO cathodes and BF TNO anodes (a) without sucrose, and with (b) 2.5 %, (c) 5.0 % and (d) 7.5 % sucrose. Each plot contains the result of a linear least squares regression and the corresponding  $R^2$  values.  $\Delta dQ/dV$  peak positions were calculated from the dQ/dV from analysis of the cells cycled in Figure S6 between charge and discharge cycle at increasing rates



**Figure S-8**. SEM images of the surfaces of the thinner BF TNO used for cycle life testing after 100 charge/discharge cycles at a rate of C/2 (a) without sucrose and (b) with 5.0 % sucrose.

## References

- [1] R. S. Roth, A. D. Wadsley, Mixed oxides of titanium and niobium: the crystal structure of TiNb<sub>24</sub>O<sub>62</sub> (TiO<sub>2</sub>·12Nb<sub>2</sub>O<sub>5</sub>), *Acta Crystallographica* **18** (1965) 724-730. https://doi.org/10.1107/S0365110X65001664
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- [3] M. Gasperin, Affinement de la structure de TiNb<sub>2</sub>O<sub>7</sub> et répartition des cations, *Journal of Solid State Chemistry* **53** (1984) 144-147. https://doi.org/10.1016/0022-4596(84)90238-X