Supporting Information

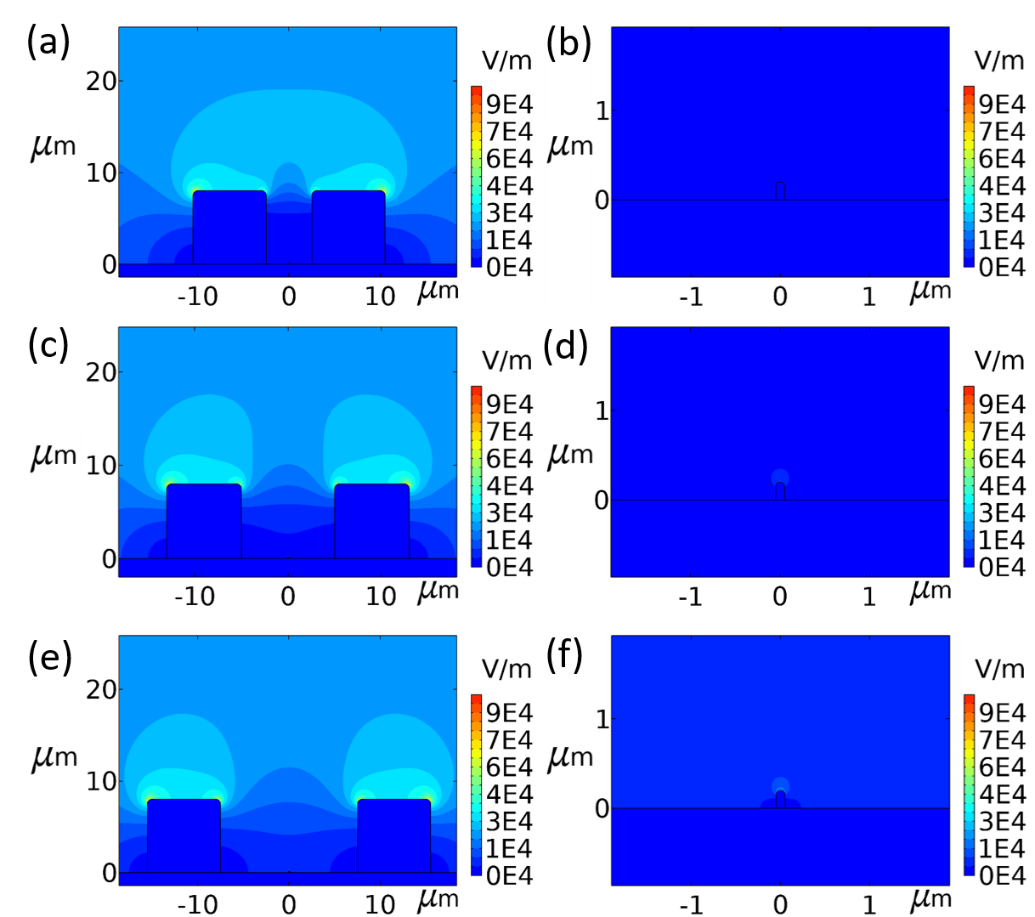
**High dielectric 3D scaffold to suppress Li-dendrites and increase the reversibility of Li-metal anodes**

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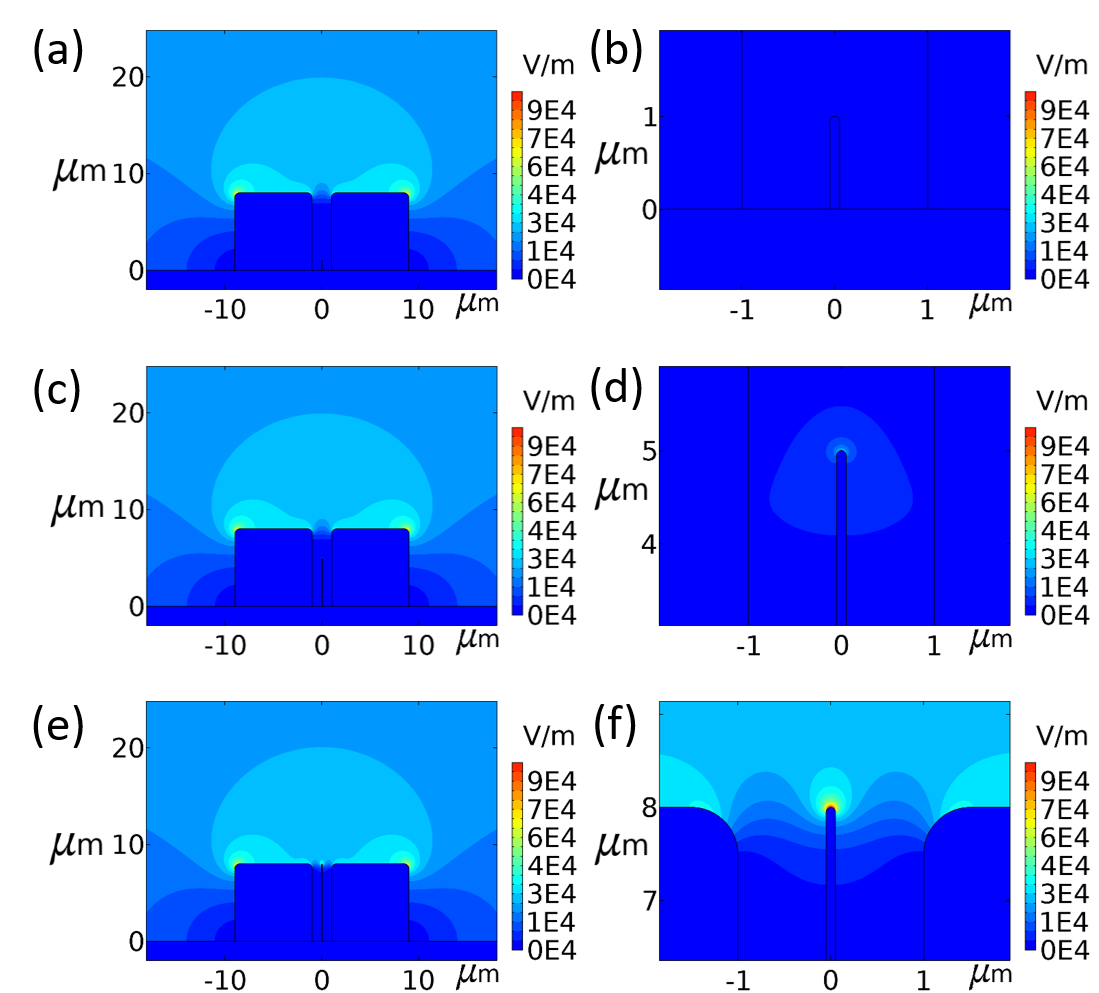
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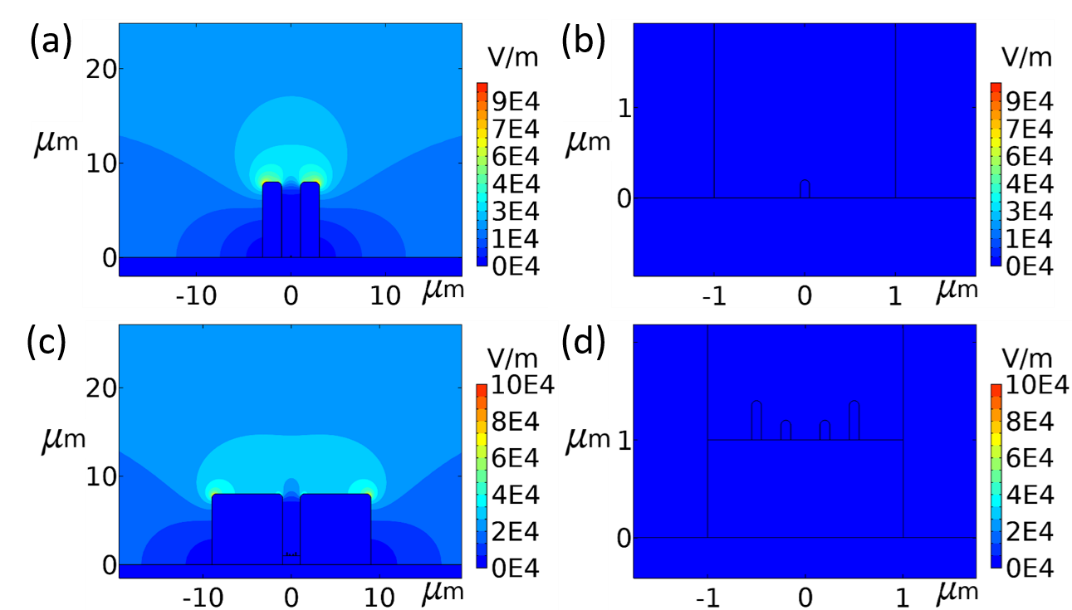
Faculty of Applied Sciences, Delft University of Technology.



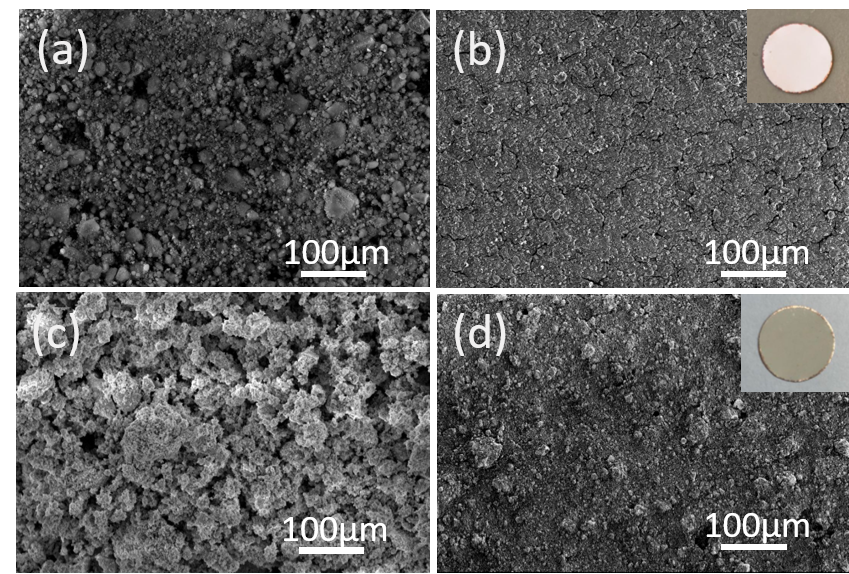
**Figure S1**  **Schematic diagram of the electric field distribution of BaTiO3 (BTO) coated Cu with different block gaps.** (a, b) with distance of 5 μm between blocks and zoomed-in figure, (c, d) with distance of 10 μm between blocks and zoomed-in figure, (e, f) with distance of 15 μm between blocks and zoomed-in figure.



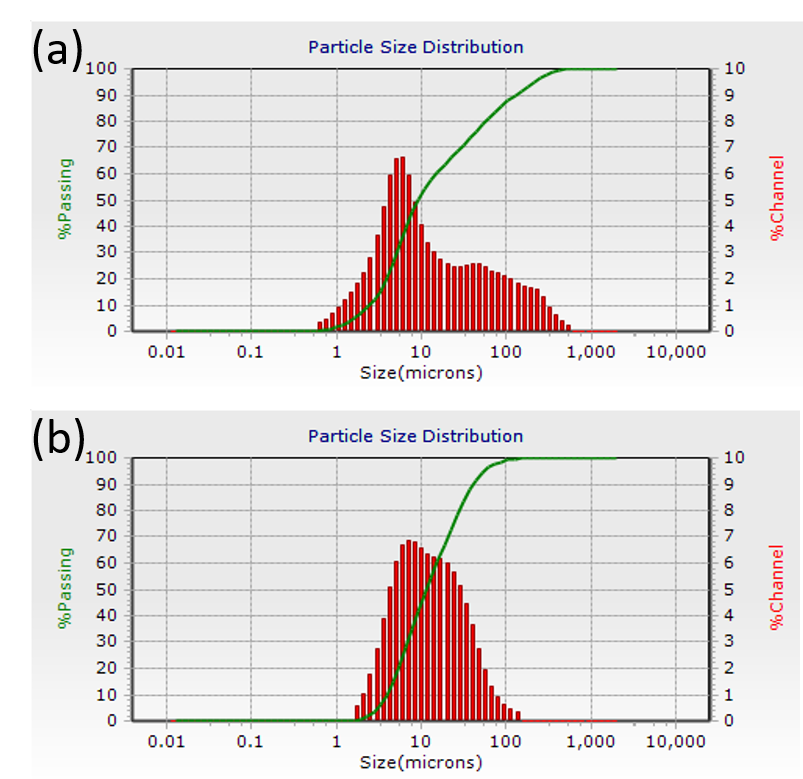
**Figure S2**  **Schematic diagram of the electric field distribution of BaTiO3 (BTO) coated Cu with different dendrite length.** (a, b) with a dendrite length of 1 μm and zoomed-in figure, (c, d) with a dendrite length of 5 μm and zoomed-in figure, (e, f) with a dendrite length of 8 μm and zoomed-in figure.



**Figure S3**  **Schematic diagram of the electric field distribution of BaTiO3 (BTO) coated Cu** (a, b) with a block width of 2 μm and zoomed-in figure, (c, d) with multiple dendrites and zoomed-in figure.



**Figure S4** **ball-milled starting materials and their coatings on copper.** (a, b) AO and AO coated on Cu,(c, d) BTO and BTO coated on Cu. Insets are the digital image of the electrode.



**Figure S5** Particle size distribution of (a) BTO and (b) AO as obtained using the Dynamic Light Scattering technique.

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**Figure S6** Nitrogen adsorption/desorption isotherms at 77 K of BTO and AO materials.

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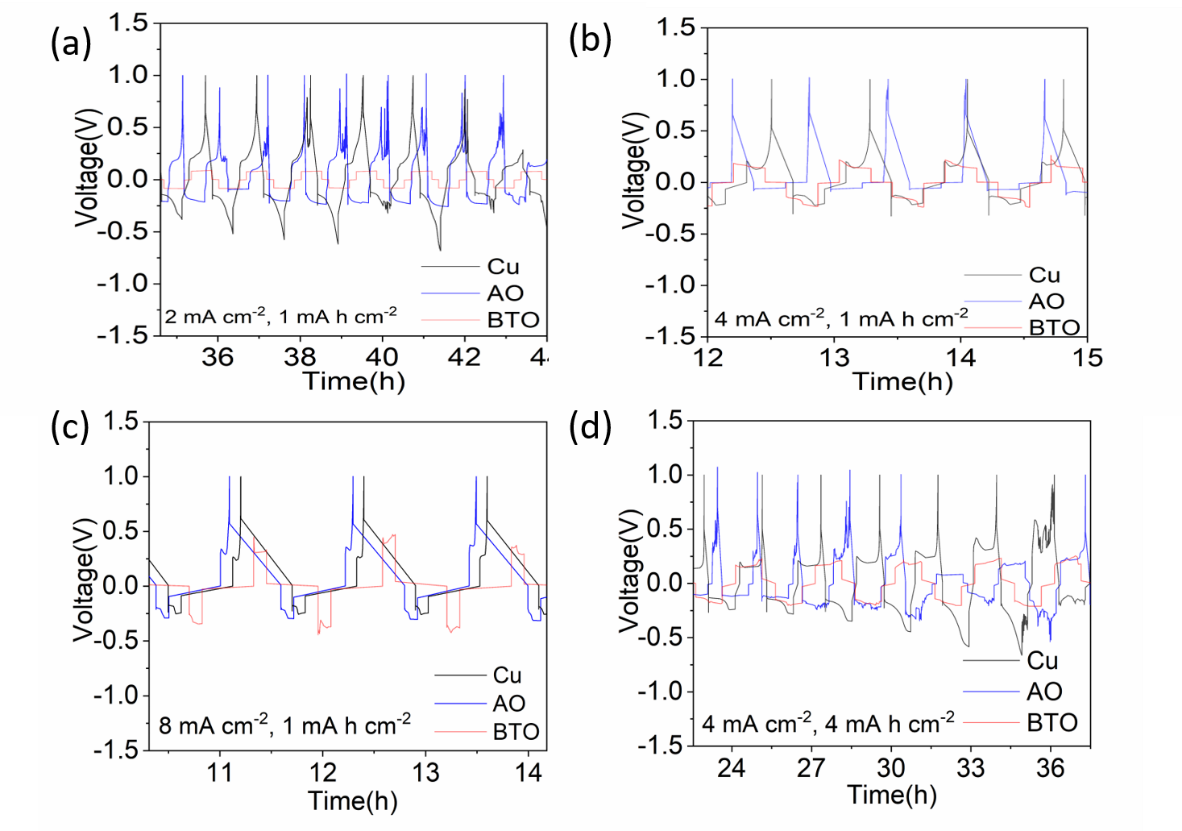
**Figure S7** Static 7Li NMR spectrum measured with a wide spectral window of a fresh BTO-LCO battery.



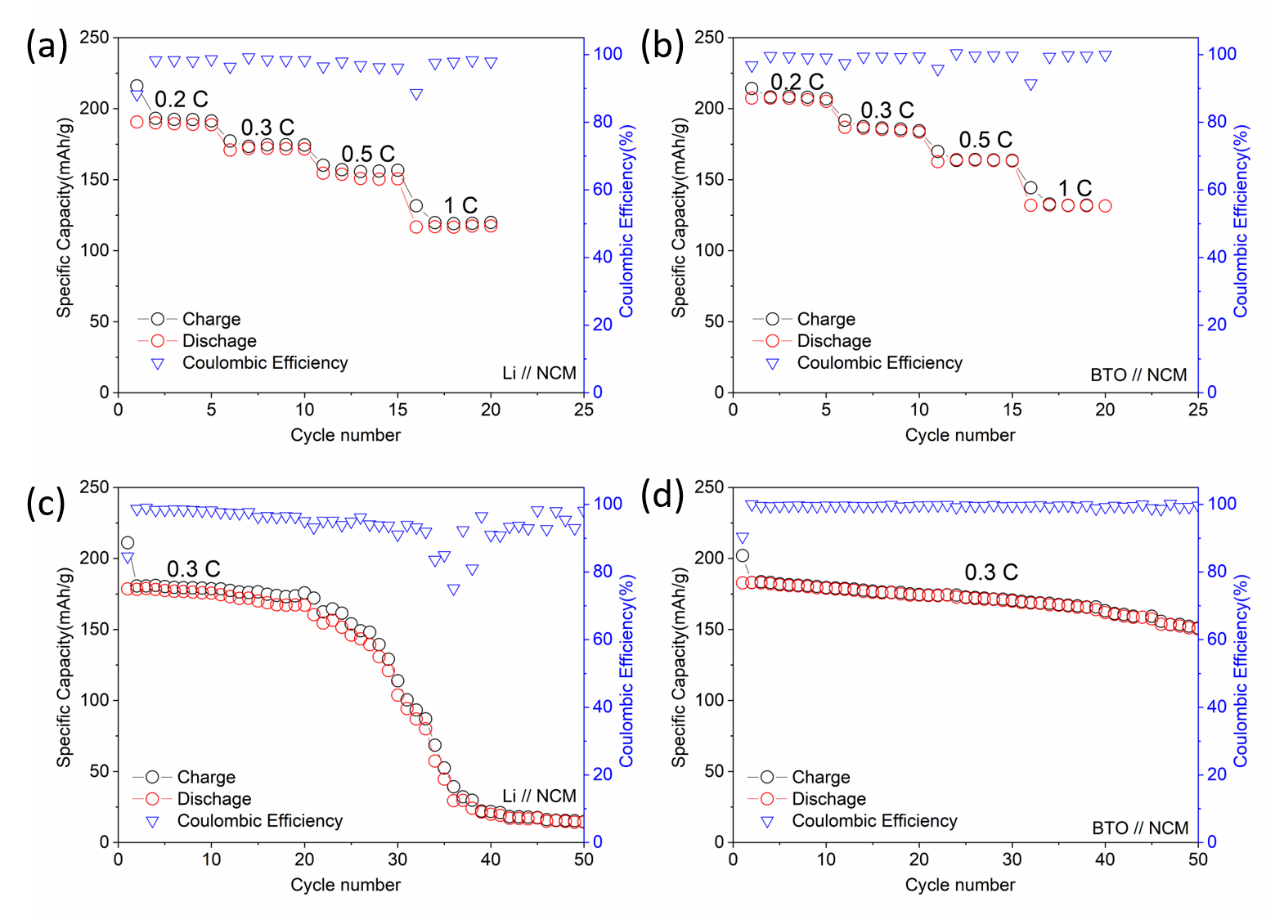
**Figure S8**   Cyclic voltammetry performance of different electrodes with a scan rate of 1mV s‑1 in the range of -0.5 V to 3 V.

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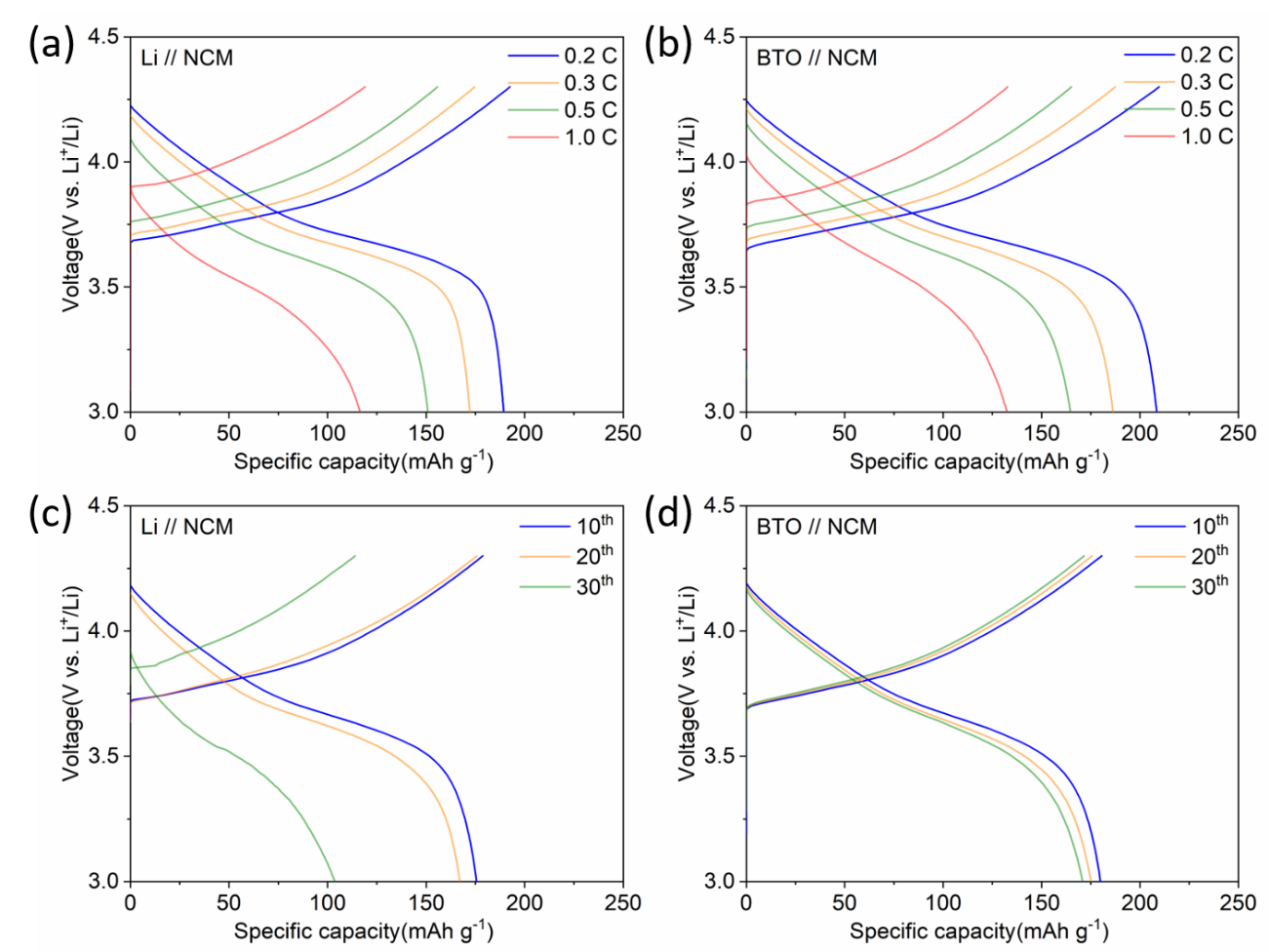
**Figure S9**   XRD patterns of BTO raw material and BTO electrodes before and after 1st and 100th cycles.



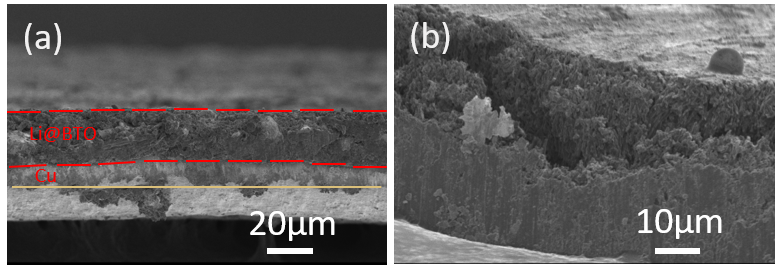
**Figure S10 Detailed voltage profiles of the anode-less Cu, AO and BTO scaffolds vs a Li-metal anode.** Evolution of the voltage during Li plating/stripping for the bare Cu and the AO and BTO scaffolds at different current densities (a) 2 mA cm-2, (b) 4 mA cm-2, (c) 8 mA cm-2 with a fixed areal capacity of 1 mA h cm-2, and (d) an increased areal capacity of 4 mA h cm-2 at a current density of 4 mA cm-2.



**Figure S11**   **Cycling performances and coulombic efficiency of Li-metal and BTO scaffold when paired with NCM cathode**.(a,b) Rate test from 0.2 C to 1 C. (c,d) Cycling test at 0.3 C.



**Figure S12**   **Voltage profiles of Li-metal and BTO scaffold when paired with NCM cathode**.(a,b) Voltage profiles when cycled with different current densities. (c,d) Voltage profiles of different cycles when cycled at 0.3 C.



**Figure S13** Cross-section of BTO (a) and Cu (b) electrode after depositing 1 mA h cm‑2 Li-metal at a current density of 2 mA cm-2.