

**Supporting Information**

**One-Dimensional Piezoelectric BaTiO<sub>3</sub> Polycrystal of  
Topochemical Mesocrystal Conversion from Layered  
H<sub>2</sub>Ti<sub>4</sub>O<sub>9</sub>·H<sub>2</sub>O Single Crystal**

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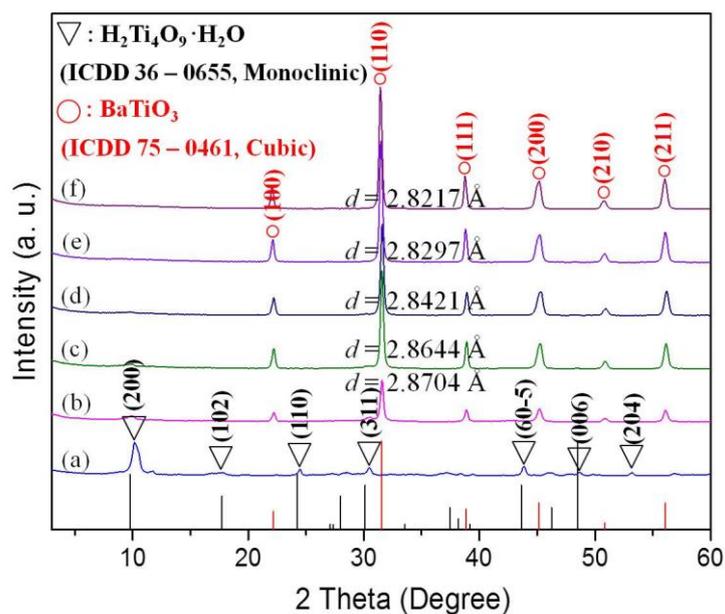
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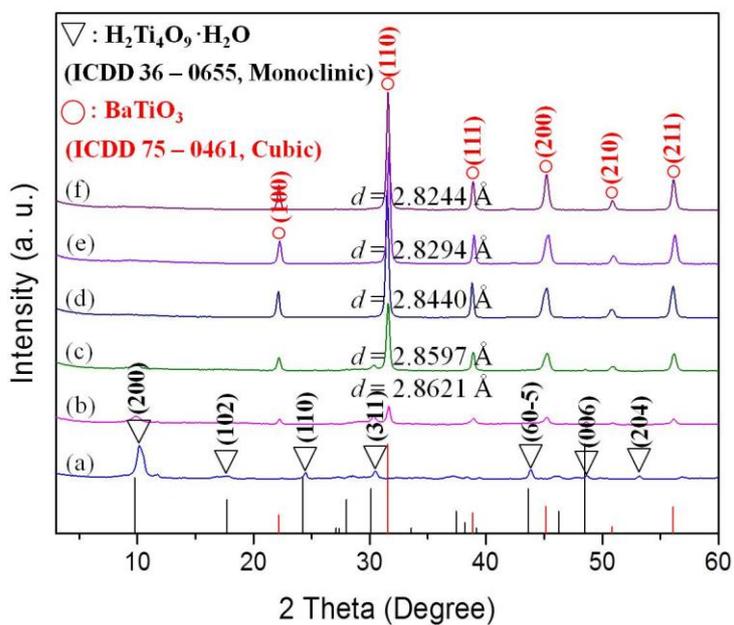
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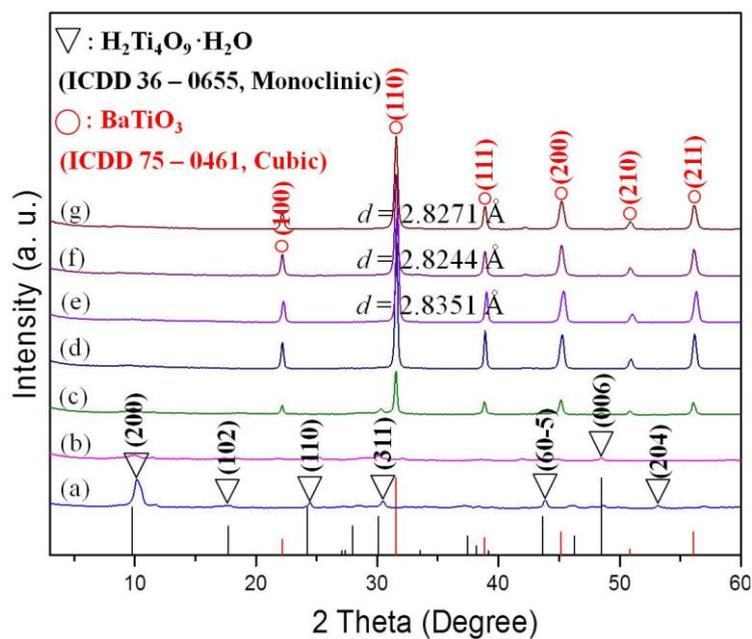
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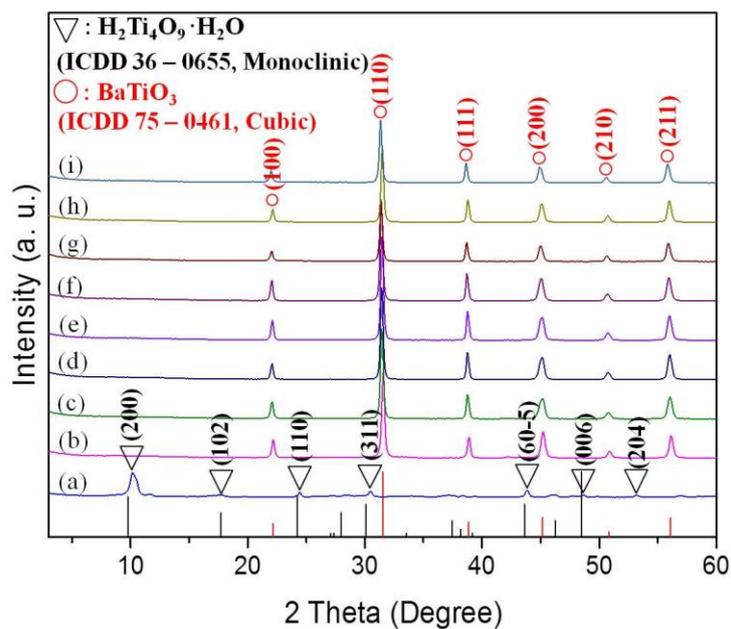
**Figure S1** XRD patterns of  $\text{H}_2\text{Ti}_4\text{O}_9 \cdot \text{H}_2\text{O}$  (HTO) single crystals (a) before and after hydrothermal treatments in  $0.2 \text{ mol} \cdot \text{L}^{-1}$   $\text{Ba}(\text{OH})_2$  water solution at  $150 \text{ }^\circ\text{C}$  for (b) 0.5, (c) 1, (d) 2, (e) 6, and (f) 12 h, respectively.



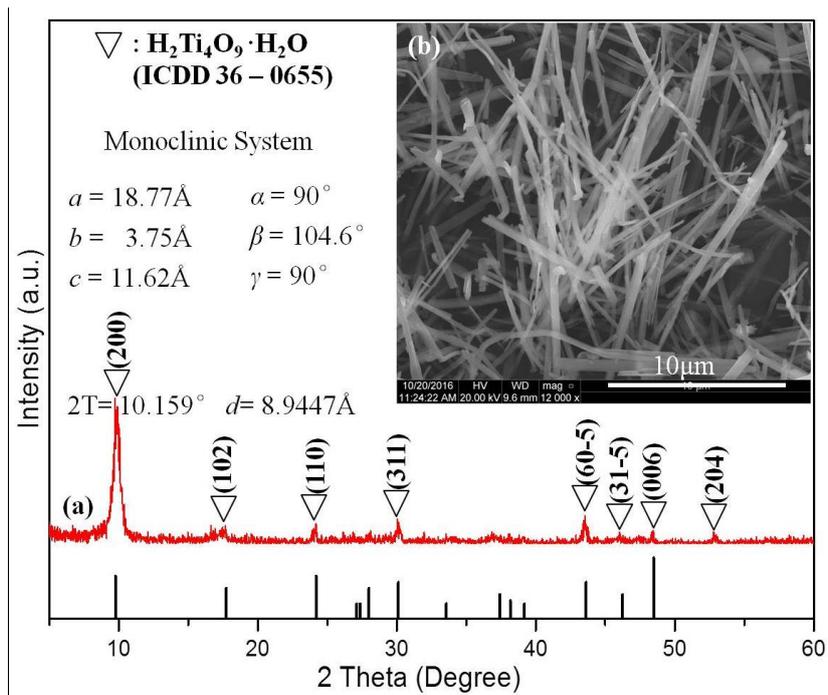
**Figure S2** XRD patterns of HTO single crystals (a) before and after hydrothermal treatments in  $0.1 \text{ mol} \cdot \text{L}^{-1}$   $\text{Ba}(\text{OH})_2$  solution at  $150 \text{ }^\circ\text{C}$  for (b) 0.5, (c) 1, (d) 2, (e) 6, and (f) 12 h, respectively.



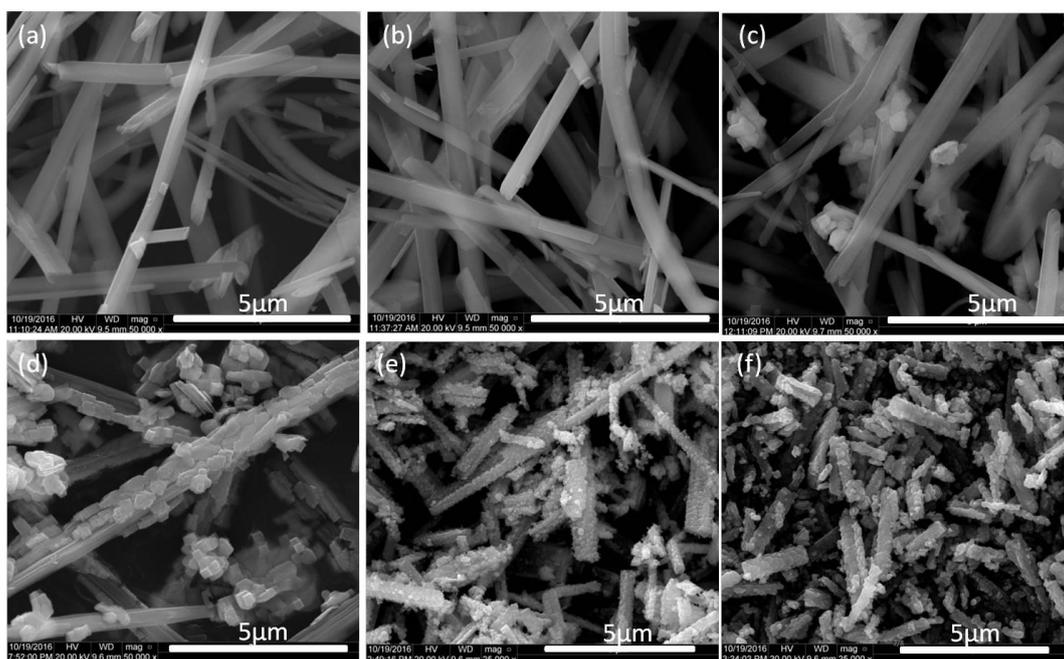
**Figure S3** XRD patterns of HTO single crystals (a) before and after hydrothermal treatments in  $0.1 \text{ mol} \cdot \text{L}^{-1} \text{ Ba(OH)}_2$  water solution at (b) 50, (c) 80, (d) 100, (e) 120, (f) 150, and (g) 200 °C for 12 h, respectively.



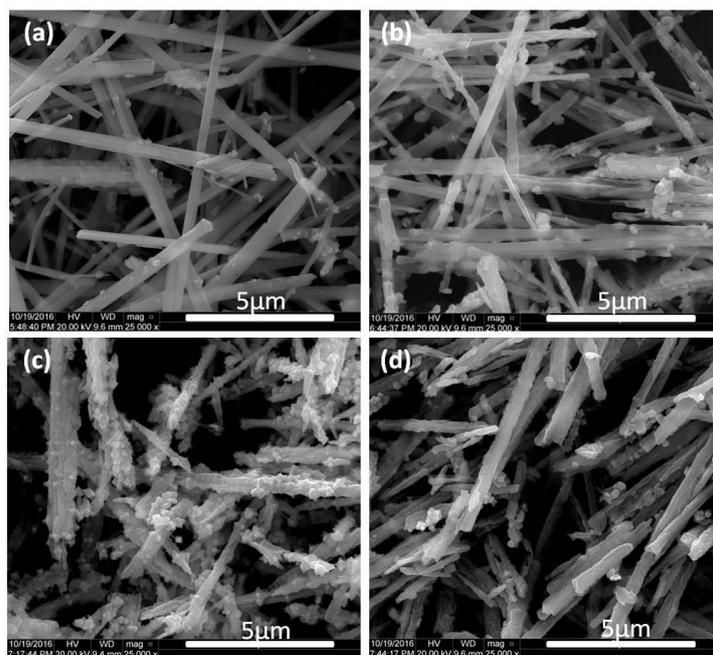
**Figure S4** XRD patterns of HTO single crystals (a) before and after hydrothermal treatments in (b) 0.1, (c) 0.2, (d) 0.5, (e) 1.0, (f) 1.5, and (g) 2.0  $\text{mol} \cdot \text{L}^{-1} \text{ Ba(OH)}_2$  water solution at 150 °C for 12 h, respectively.



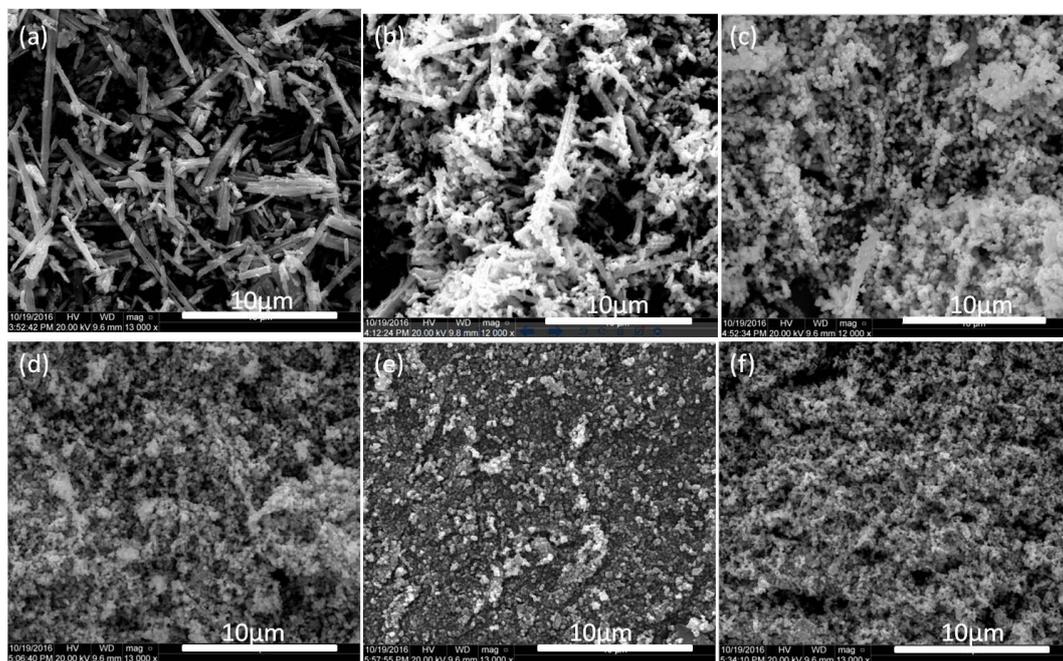
**Figure S5** (a) XRD pattern and (b) SEM image of protonated HTO single crystals.



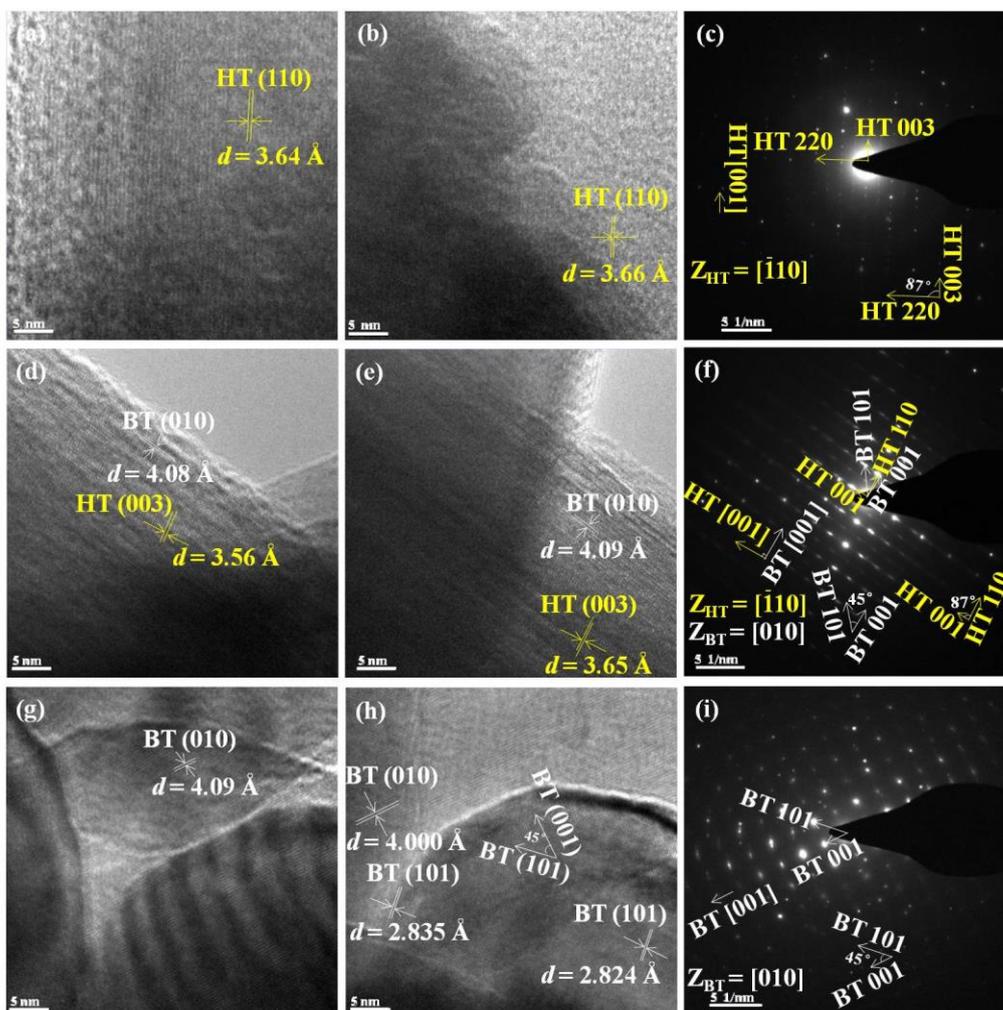
**Figure S6** SEM images of samples obtained by hydrothermal treatments of HTO single crystals in  $0.1 \text{ mol} \cdot \text{L}^{-1} \text{ Ba(OH)}_2$  water solution at (a) 50, (b) 80, (c) 100, (d) 120, (e) 150, and (f) 200 °C for 12 h, respectively.



**Figure S7** SEM images of samples obtained by hydrothermal treatments of HTO single crystals in  $0.1 \text{ mol} \cdot \text{L}^{-1} \text{ Ba(OH)}_2$  water solution at  $150 \text{ }^\circ\text{C}$  for (a) 0.5, (b) 1, (c) 2, and (d) 6 h, respectively.



**Figure S8** FE-SEM images of samples obtained by hydrothermal treatments of HTO single crystals in (a) 0.1, (b) 0.2, (c) 0.5, (d) 1.0, (e) 1.5, and (f)  $2.0 \text{ mol} \cdot \text{L}^{-1} \text{ Ba(OH)}_2$  water solution at  $150 \text{ }^\circ\text{C}$  for 12 h, respectively.



**Figure S9** (a, b, d, e, g, h) HRTEM images, (c, f, i) SAED patterns of hydrothermal treatments of HTO single crystals in  $0.1 \text{ mol} \cdot \text{L}^{-1} \text{ Ba(OH)}_2$  water solution at (a-c) 80, (d-f) 100, and (g-i) 150  $^\circ\text{C}$  for 12 h, respectively.