Electronic Supplementary Information (ESI) available for:

Water-titanate intercalated nanotubes: Fabrication, polarization, and giant dielectric property

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Supporting Online Materials

- **S1.** XRD pattern of the starting material of anatase TiO_2 .
- **S2.** XRD patterns of Na-TNT and H-TNT.
- **S3.** TEM and HRTEM images of H-TNT.

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- S4. Impedance spectra of Na-TNT sample at selected temperature of 70, 80, and 90 °C.
- **S5.** TG data of Na-TNT and H-TNT.
- S6. Room-temperature impedance spectra of Na-TNT and H-TNT.
- **S7.** Temperature dependence of grain conductivity (σ_g) for Na-TNT and H-TNT.



Fig. S1 XRD pattern of the starting material of anatase TiO₂. Asterisks * denote the internal standard Ni. The bottom bars denote the standard diffraction data of anatase TiO₂ (JCPDS, No. 21-1272).



Fig. S2 XRD patterns of (a) Na-TNT and (b) H-TNT. Symbols "*" denote the internal standard Ni.



Fig. S3 (a) TEM and (b) HRTEM images of H-TNT.



Fig. S4 TG data of Na-TNT and H-TNT.



Fig. S5 Impedance spectra of Na-TNT sample at selected temperature of 70, 80, and 90 °C. Inset shows the enlarged portion of the high frequency spectra, respectively. It is clear that three spectra exhibited similarly non-zero-intercepted tubers at high frequencies, which are related to the grain ⁵ responses, respectively. At relatively low frequencies, the spectrum obtained at 70 °C only contains an arc, while the spectra obtained at 80 °C includes an arc, and also a short linear tail. When increasing the temperature to 90 °C, the tail becomes longer. The art and the tail denote the responses of grain boundary and electrode effect, respectively. Therefore, increasing the measurement temperature will enhance the electrode effect, which also imposes significant impact on the dielectric behaviors.



Fig. S6 Room-temperature impedance spectra of Na-TNT and H-TNT. Insets are the enlarged high frequency spectra (bottom) and the equivalent circuit model (top), respectively. According to the equivalent circuit model, the grain resistivity (R_g) is fitted to be $5.8 \times 10^4 \ \Omega$.cm for Na-TNT and ${}^{5} 4.3 \times 10^5 \ \Omega$.cm for H-TNT while their boundary resistivity (R_{gb}) is estimated to be $1.1 \times 10^8 \ \Omega$.cm for Na-TNT and $1.2 \times 10^8 \ \Omega$.cm for H-TNT. Obviously, for both Na-TNT and H-TNT, their R_{gb} is three or four larger than respective R_g , which underlies the heterogeneous systems.



Fig. S7 Temperature dependence of the grain conductivity (σ_g) for Na-TNT and H-TNT. For both Na-TNT and H-TNT, their grain conductivities, σ_g , have the similar features against temperature, i.e., it firstly increases and then decreases with increasing temperature, and finally keeps a near constant. However, there are 3 differences between Na-TNT and H-TNT: (1) the grain conductivity of former is one order larger than the latter in whole measured temperature region; (2) the maximum of conductivity is at 90 °C for the former while 100 °C for the latter; (3) conductivity begins to keep a constant at 135 °C for the former while this temperature for the latter is 170 °C. Excluding the effect from morphology and phase scale, such a giant conductivity discrepancy is attributed to the respective grain itself.