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Supplementary Information

Ultra-high Energy Storage Density and Scale-up of Antiferroelectric $TiO_2/ZrO_2/TiO_2$ Stacks for Supercapacitors

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Doping concentration of Ti in the ZrO_2 layer

Since the doping of Ti in the TZT*n* samples arises from the Ti diffusion from the TiO_2 interfacial layers or interlayers into ZrO_2, a non-uniform doping profile is expected. The doping percentage of Ti in the ZrO_2 layer is investigated by an XPS depth profile analysis. Fig. S1(a) shows the depth profile of the chemical composition in the TZT2 sample. The O/[Zr+Ti] ratio in the ZrO_2 layer is in the range of 1.84~1.99, which is near the stoichiometry of the oxides. The depth profile of the Ti/[Zr+Ti] percentage is shown in Fig. S1(b), which reveals that the doping percentage of Ti in the ZrO_2 layer approximately ranges from 7.9 to 18.6% and the average doping percentage is around 13.7%.

The chemical composition of the sample was analyzed by an X-ray Photoelectron Spectroscopy (XPS, Thermo Fisher Scientific Theta Probe) with an Al Kα X-ray source (1486.6 eV). Argon ions were used as the sputtering source for the depth profile analysis. The probing depth of the XPS is around 3~7 nm.

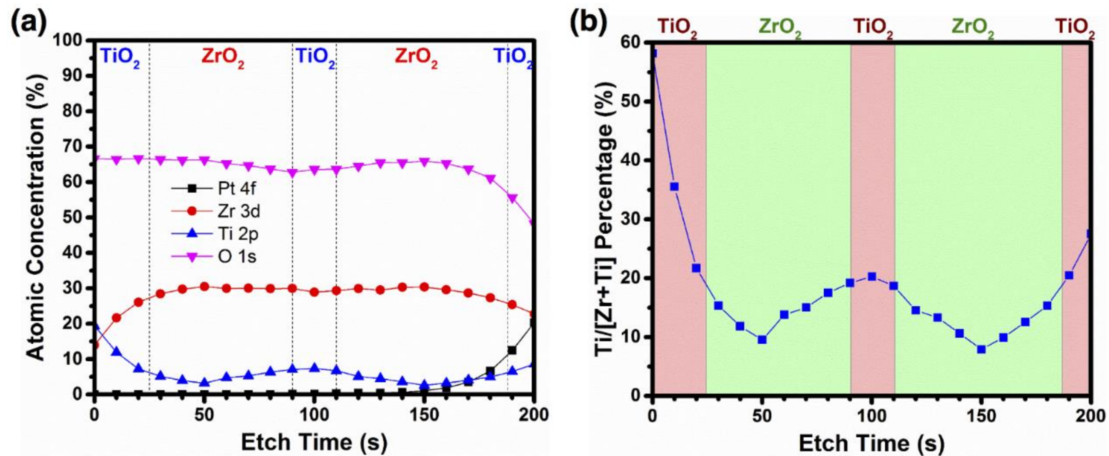

Fig. S1 (a) XPS depth profiles of the elements (Zr, Ti, O, and Pt) and (b) the depth profile of the Ti/[Zr+Ti] percentage in the TZT2 sample.

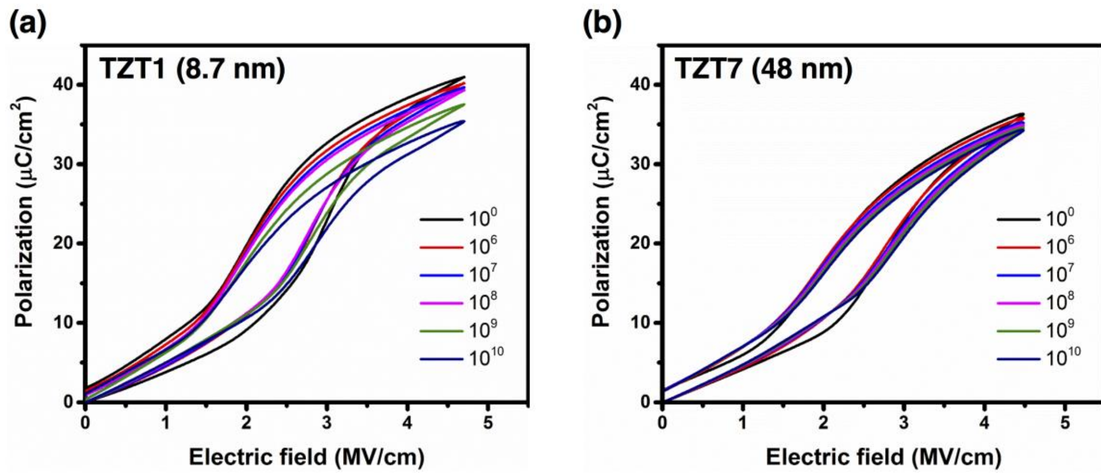

Fig. S2 Evolution of the *P*-*E* curves of the **(a)** TZT1 and **(b)** TZT7 capacitors with the fatigue cycling of unipolar rectangular pulses of 4.5 MV/cm at a frequency of 125 kHz.