

Supporting Information

Oxygen Scavenging of HfZrO₂-Based Capacitors for Improving Ferroelectric Properties

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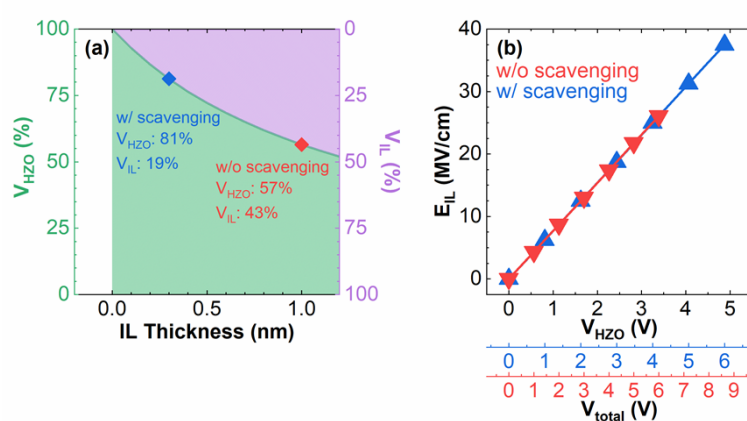


Figure S1. (a) Voltage drop distribution between HZO and IL according to the thickness of IL (d_{IL}). (b) The electric field across the IL (E_{IL}) as a function of the voltage drop across the HZO (V_{HZO}) and the total applied voltage (V_{total}).

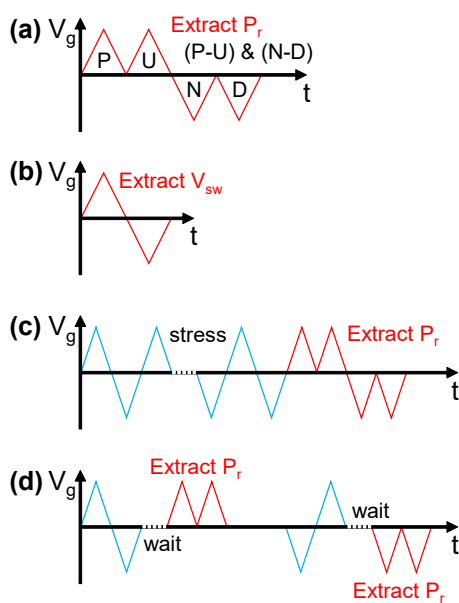


Figure S2. Schematic illustrations of pulse measurements. (a) Positive-up-negative-down (PUND) method using double triangular pulses for extraction P_r . (b) Bipolar triangular pulse for extraction of V_{sw} . (c) Endurance pulse train and PUND method. (d) Retention measurement.

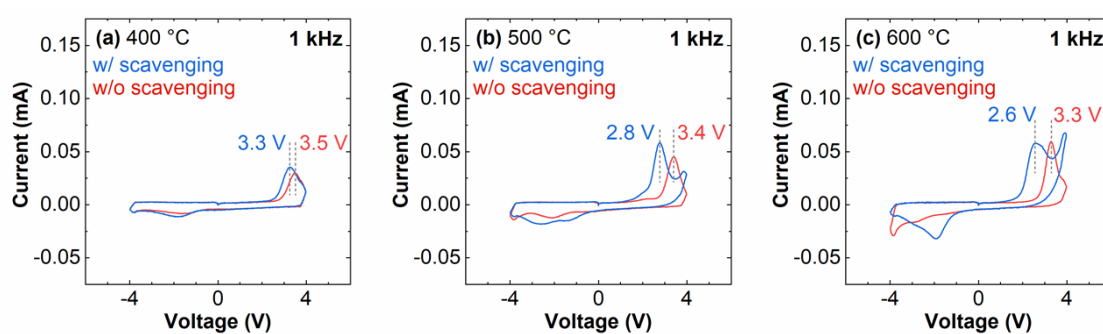


Figure S3. I - V curves of capacitors without and with scavenging according to PMA temperature of (a) 400, (b) 500, and (c) 600 °C measured by bipolar triangular pulse. In both capacitors, V_{sw} decreased as the PMA temperature increased. In particular, the decrease in V_{sw} was more pronounced in the capacitors with scavenging, implying that the oxygen scavenging is temperature-dependent. The capacitor with scavenging annealed at 600 °C exhibited the lowest V_{sw} , but the increase in leakage current was remarkable.

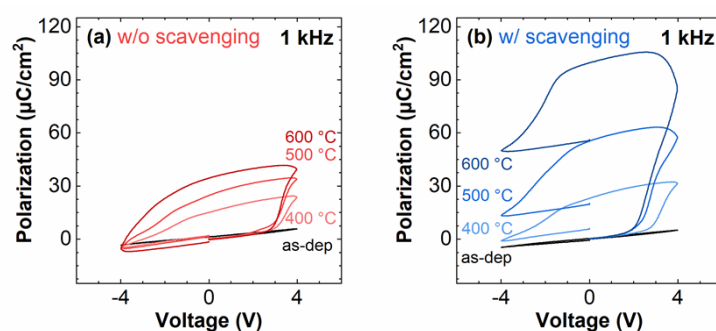


Figure S4. P - V curves of capacitors (a) without and (b) with scavenging according to PMA temperature of 400, 500, and 600 °C measured by bipolar triangular pulse. The as-deposited capacitors showed the P - V curve of a typical dielectric material without polarization hysteresis. In both capacitors, P_r increased as the PMA temperature increased. As shown in Figure S3, the leakage current increased as the PMA temperature increased in the capacitors with scavenging, resulting in an open loop.

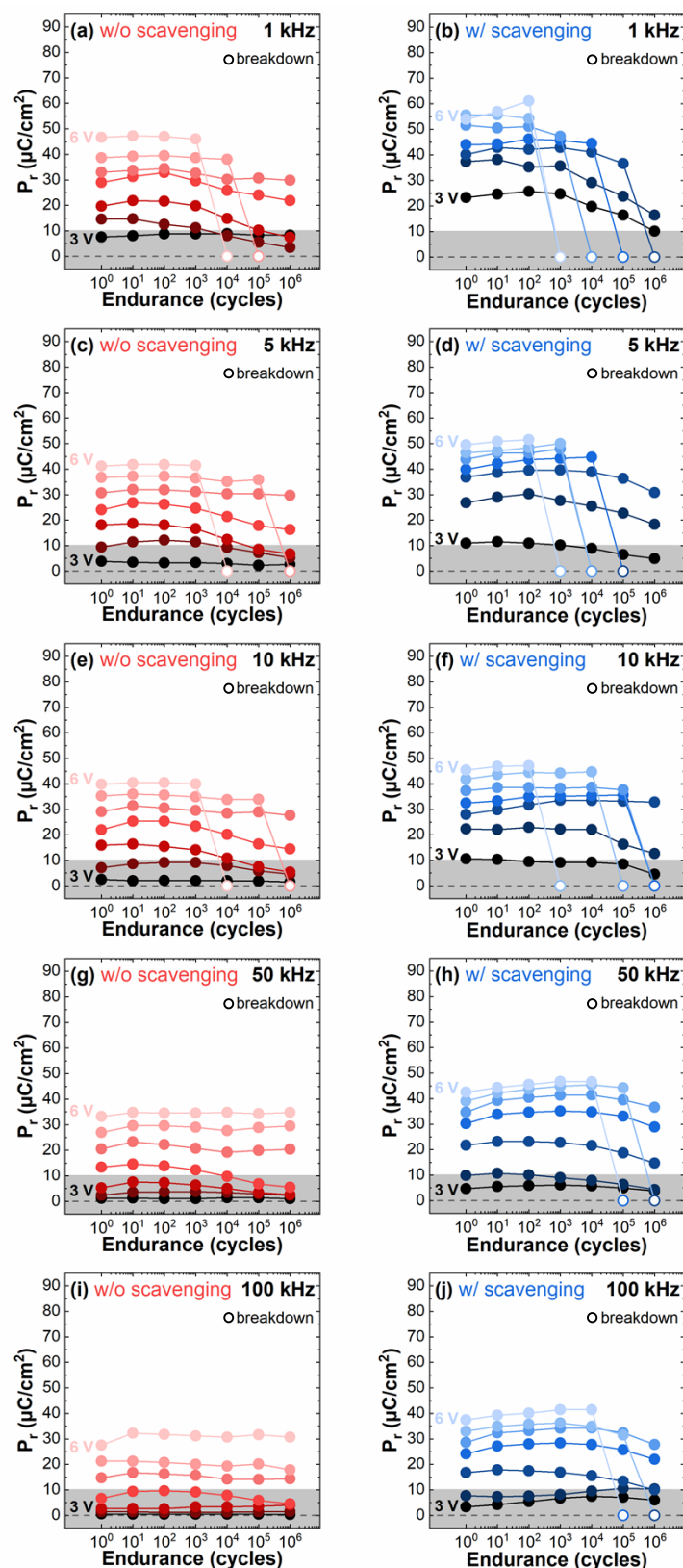


Figure S5. Endurance properties of capacitors (a) without and (b) with scavenging according to pulse amplitudes and frequencies. The breakdown is indicated by empty circles.

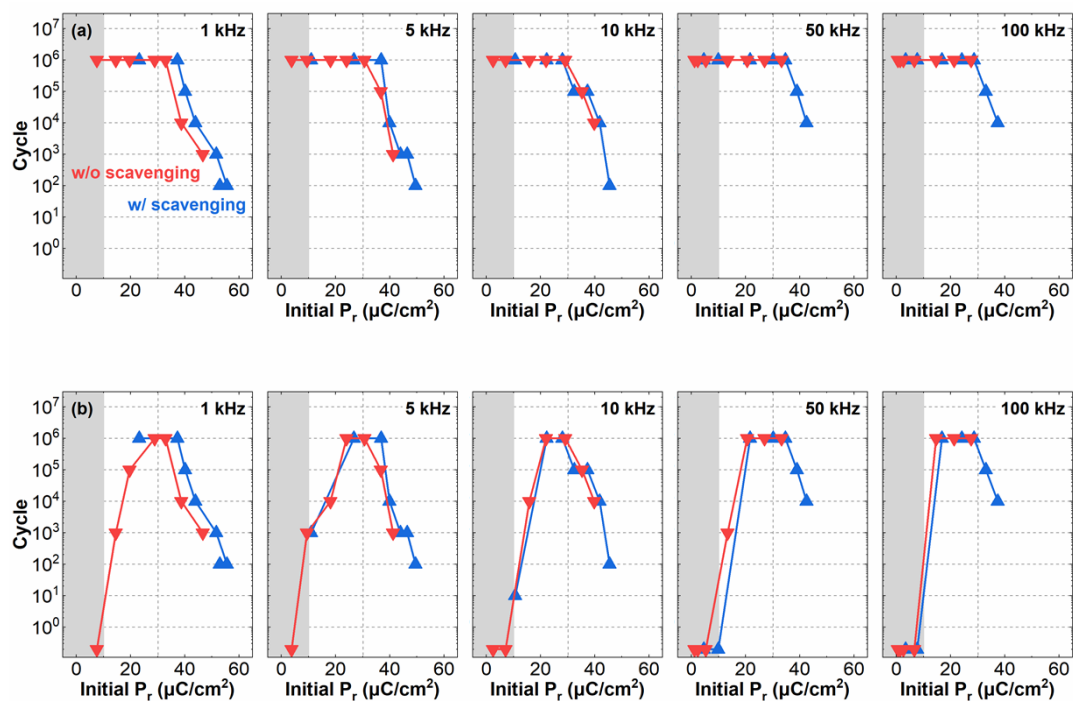


Figure S6. Endurance properties according to initial P_r and pulse frequency until (a) breakdown occurred and (b) P_r reached $10 \mu\text{C}/\text{cm}^2$. Pulses with amplitudes of 3 to 6 V were applied.

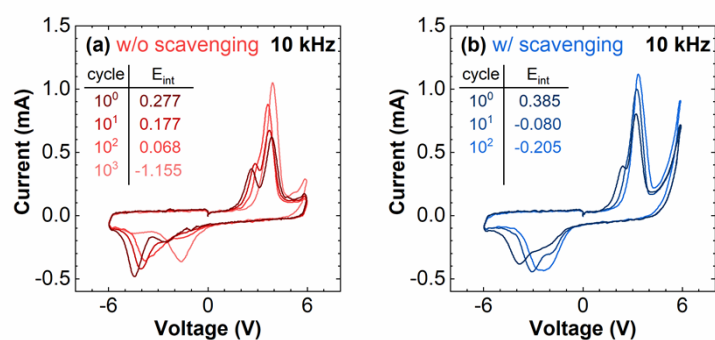


Figure S7. P - V curves of capacitors (a) without and (b) with scavenging during endurance test with the pulse of 6 V/10 kHz. The table insets are calculated E_{int} according to the endurance cycles.

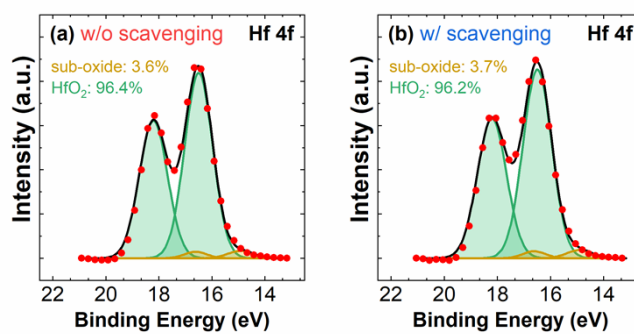


Figure S8. XPS Hf 4f spectra deconvoluted with HfO₂ and sub-oxide at the top interface of capacitors (a) without and (b) with scavenging.

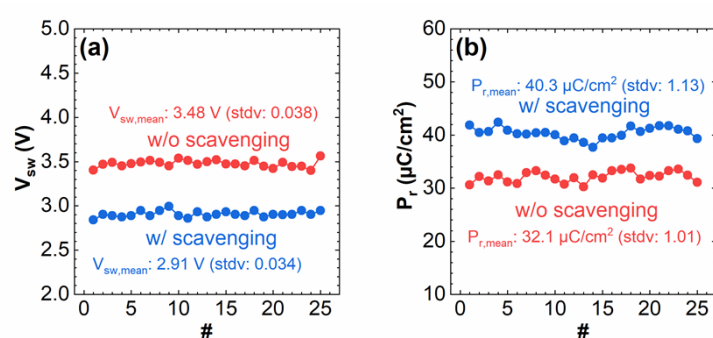


Figure S9. Plots of (a) V_{sw} and (b) P_r of 25 capacitors without and with scavenging and their mean and standard deviation.