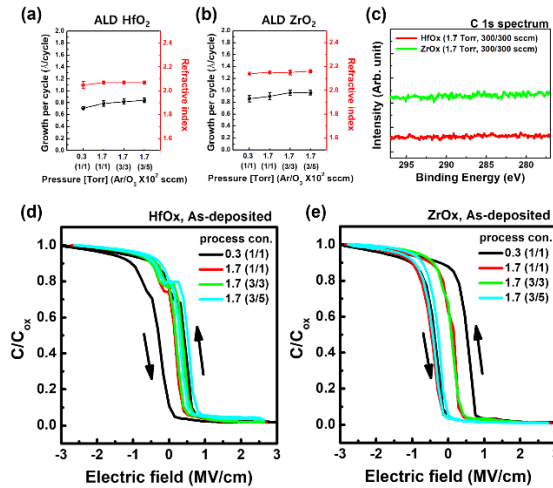
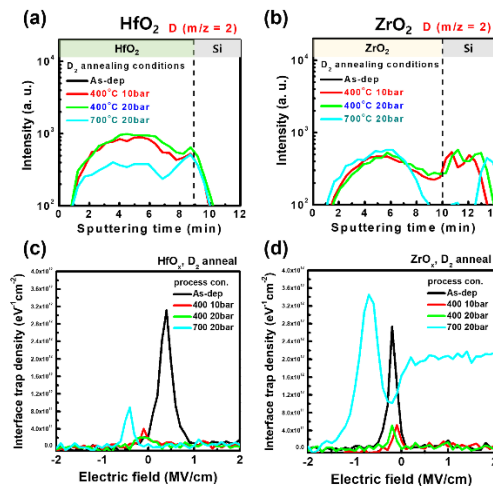


## Supplemental Document

The thicknesses and refractive index of high-k films were measured using spectroscopic ellipsometry (EliSE(UV)-FM8, Ellipso Technology). We used X-ray photoelectron spectroscopy (XPS, AXIS SUPRA+, KRATOS) to investigate the presence of Carbon. The top electrode, ITO 100 nm, of the MIS capacitor was deposited by RF sputtering. Subsequently, the MIS capacitor was fabricated using a lift-off process. C-V measurements of the MIS capacitors were conducted using a semiconductor parameter analyzer (4200-SCS, Keithley). the interface trap density was calculated using the conductance method.



**Figure 1.** GPC and RI values with modulated ALD process pressures for (a) HfO<sub>2</sub> and (b) ZrO<sub>2</sub>. (c) C content measured by XPS under modulated process conditions. C-V characteristics of MIS devices for high-k dielectrics with different process pressures: (d) HfO<sub>2</sub> and (e) ZrO<sub>2</sub>.



**Figure 2.** D-SIMS depth profiles of D<sub>2</sub> atoms across the insulator/Si interface before and after HPDA. D<sub>2</sub> profiles for (a) HfO<sub>2</sub>, and (b) ZrO<sub>2</sub> on Si interfaces. Calculated interface trap density using the conductance method for MIS devices with (c) HfO<sub>2</sub>, and (d) ZrO<sub>2</sub> films after HPDA.