Supporting Information

Towards environmentally stable solutionprocessed oxide thin-film transistors: rare-metalfree oxide-based semiconductor/insulator heterostructure and chemically stable multistacking

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Figure S1. XPS spectra (Zn 2*p*, Sn 3*d*, and O 1*s*) from the Sn-poor ZTO films immersed in pH-engineered acidic Al_2O_3 precursor solutions. O 1*s* peak is deconvoluted into three peaks corresponding to lattice oxygen (M–O) at 530.2 eV, oxygen vacancies (V_O) at 531.2 eV, and oxygen associated with hydroxyl groups (M–OH) at 532.3 eV, respectively.





Figure S2. The O 1*s* XPS spectra from sol-gel processed Al₂O₃ insulators annealed from the pH-engineered Al₂O₃ precursor solutions via sequential soft- (120 °C for 10 min) and hard-bake (300 °C for 30 min) processing. The O 1*s* peak is deconvoluted into two peaks corresponding to lattice oxygen (Al–O) at 531.0 eV and oxygen associated with hydroxyl groups (Al–OH) at 532.3 eV.



Figure S3. The O 1*s* XPS spectra from sol-gel processed nitrate-based Al_2O_3 insulators annealed at various hard-bake temperatures (150–300 °C). O 1*s* peaks are deconvoluted into two peaks corresponding to lattice oxygen (Al–O) at 531.0 eV and oxygen associated with hydroxyl groups (Al–OH) at 532.3 eV.



Figure S4. The XPS spectra (Zn 2*p*, Sn 1*s*, and O 1*s* peaks) for Zn, Sn, and O from the Sn-modulated ZTO films immersed in acidic nitrate-based Al_2O_3 precursor solutions. The O 1*s* peaks are deconvoluted into three peaks corresponding to the lattice oxygen (M–O) at 530.2 eV, oxygen vacancies (V₀) at 531.2 eV, and the oxygen associated with hydroxyl groups (M–OH) at 532.3 eV, respectively.



Figure S5. Leakage current-voltage curves of thickness-controlled Al_2O_3 films (7, 14, and 21 nm) fabricated on p⁺⁺-Si substrate via sol-gel process.



Figure S6. The stress-induced V_{Th} shift (ΔV_{Th}) in Sn-eqi ZTO single-layer (Sn-eqi ZTO) and Sn-eqi ZTO/thin Al₂O₃ (7 nm) heterostructure TFTs under dry-air ambient (RH = 33%) in response to positive bias stress (PBS), negative bias (NBS), negative bias temperature stress (NBTS), and negative bias illumination stress (NBIS).