Supplemental

In-situ Half-Cycle Study of High Purity H_2O_2-based HfO_2 Atomic Layer Deposition on TiN Substrate

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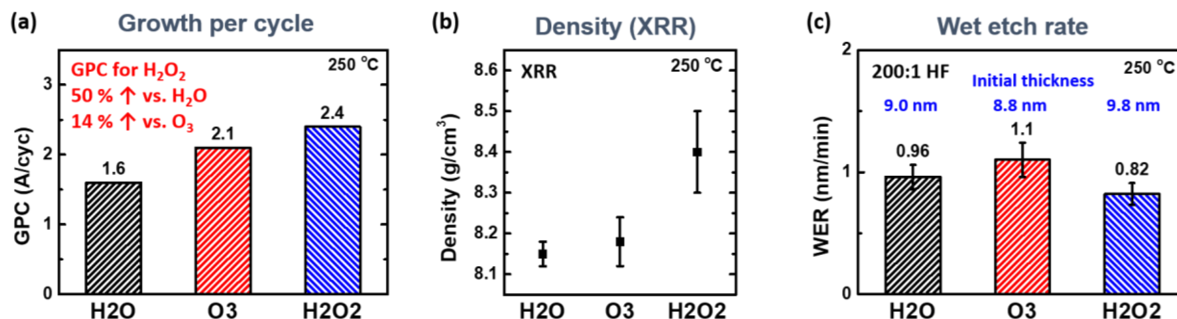

Figure 1. HZO film properties after deposition using H_2O. O_3, and H_2O_2. (a) Growth rate of HZO using TDMA-Hf/Zr supercycle at 250 °C. (b) Density of HZO deposited at 250 °C evaluated by XRR. (c) Wet etch rate of HZO in 200:1 HF.

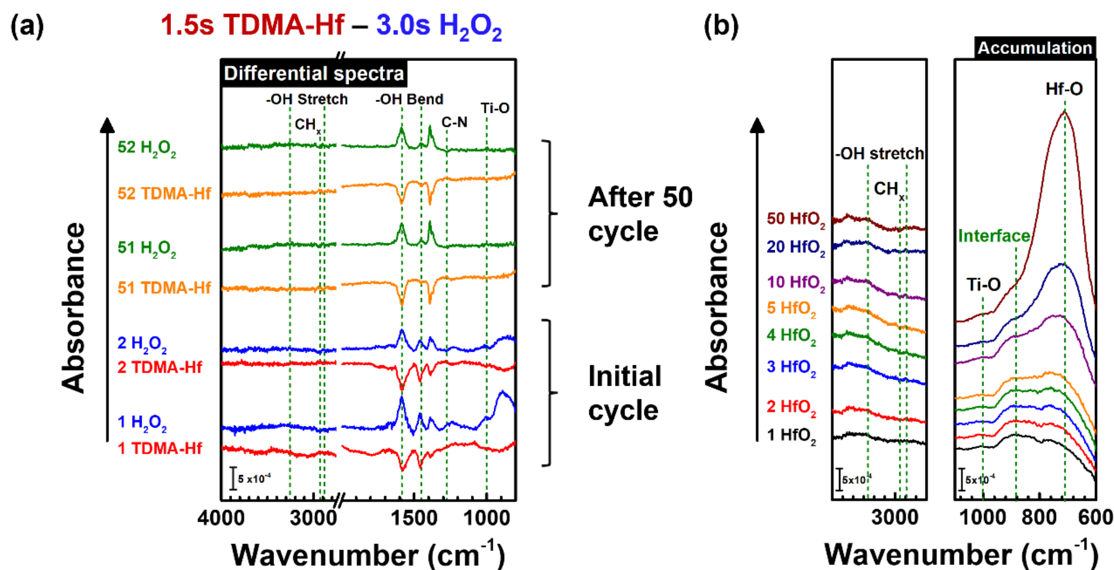

Figure 2. (a) Differential Half-cycle study of ALD cycle using TDMA-Hf and H_2O_2 with *in-situ* RAIRS system. H_2O_2 spectra show strong intensity in OH bending modes, increasing with H_2O_2 exposure and decreasing after TDMA-Hf cycle OH bending modes. Moreover, the initial cycle shows the precursor reacting with oxidant and the reaction between the substrate surface and the TDMA-Hf and H_2O_2. After 50 cycles, the surface reaction disappears, and the ALD reaction becomes dominant. Furthermore, using differential spectra, the growth of a peak around 880 cm^{-1} is observed in initial cycles but disappears quickly after a few cycles, which might be showing the interface formation. (b) Accumulation and differential spectra of ALD full cycles up to 50 cycles. By increasing the cycles, the increase of Hf-O bond is observable, which can be used to compare the bonding density with other oxidants.