(Supporting Information)

Ferroelectricity of 300°C Low Temperature Fabricated $Hf_xZr_{1-x}O_2$ Thin Films by Plasma-Enhanced Atomic Layer Deposition using Hf/Zr Cocktail Precursor

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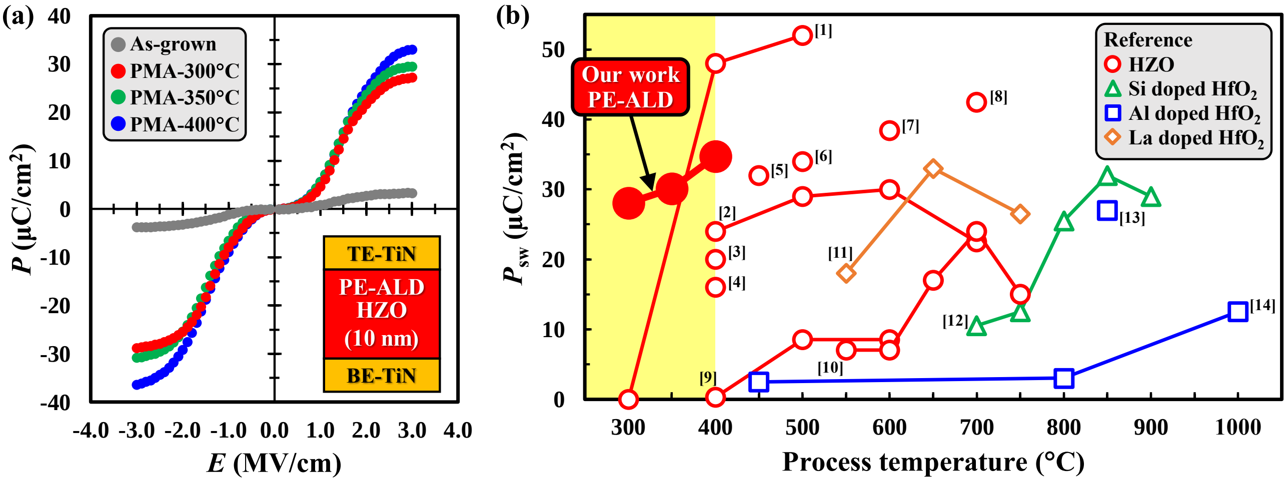

Fig. 1 (a) Pulse write/read results of as-grown and PMA-treated 10-nm-thick PE-ALD HZO films after wake-up cycling (10^3 cycles at 3.0 MV/cm). The PE-ALD HZO film after PMA at 300°C clearly exhibited ferroelectricity with P_{sw} of 28 μC/cm^2, while the as-grown HZO film showed smaller hysteresis with P_{sw} of 3.5 μC/cm^2. The P_{sw} increased as PMA temperature increased. (b) Relationship between the process temperature and the P_{sw} of the ferroelectric HfO_2-based thin films. In general, an annealing process at > 400°C is necessary to obtain HfO_2-based films with stable ferroelectricity. On the other hand, the superior ferroelectricity of HZO films was obtained using PE-ALD and a low temperature PMA even at 300°C.

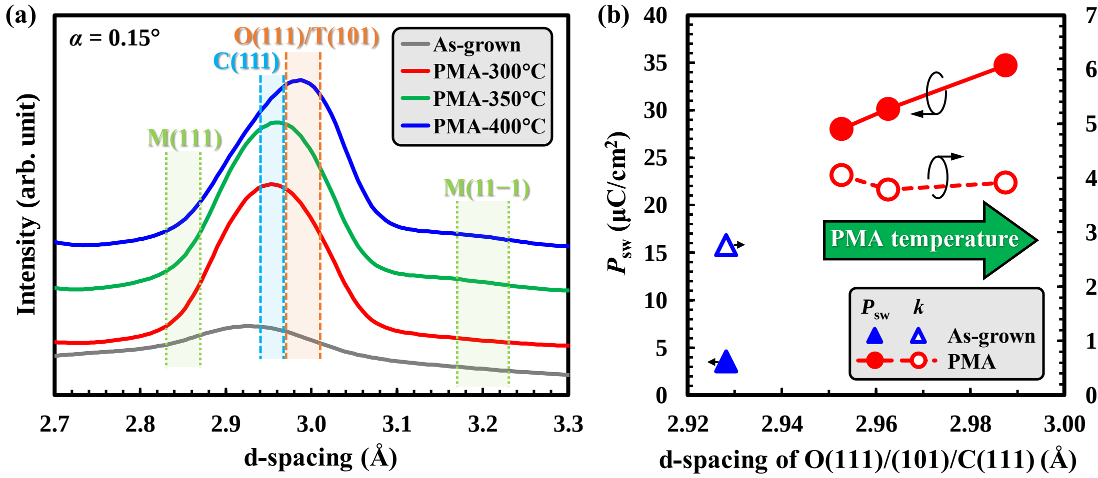

Fig. 2 (a) Synchrotron WAXS patterns of as-grown and PMA-treated 10-nm-thick PE-ALD HZO films. The WAXS pattern of the as-grown film showed a broad peak at ~2.94Å, indicating that the as-grown film had nanocrystalline structure with O/T/C phases. The peak position of O(111)/T(101)/C(111) was shifted from 2.95 to 2.99Å as PMA temperature increased. (b) P_{sw} and k value of as-grown and PMA-treated PE-ALD HZO films. The peak position of d-spacing for O(111)/T(101)/C(111), which extracted from Fig. 2 (a), increased as PMA temperature increased. Moreover, the P_{sw} of PE-ALD HZO films increased with PMA temperature, while those films showed almost the same k value of ~40. These results suggest that the phase transformation from C/T-phases to ferroelectric O-phase could occur during PMA process.

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