Supporting Information

Superior and Stable Ferroelectric Properties of Hafnium-Zirconium-Oxide Thin Films Deposited via Atomic Layer Deposition using Cyclopentadienyl-Based Precursors without Annealing

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We investigated the polarization characteristics of HZO thin films deposited using tetrakis(ethylmethylamido)Zr [TEMAZr] and tetrakis(ethylmethylamido)Hf [TEMAHf] in the same ALD chamber at 250 °C prior to and after rapid thermal annealing. As shown in Figure S1, the as-deposited HZO thin film did not exhibit ferroelectricity, and a hysteresis loop was observed after post-thermal annealing at 500 °C for 10 s.
B. Ferroelectric properties of HZO thin films deposited using a Cp-based cocktail precursor with a molar ratio of Hf[Cp(NMe$_2$)$_3$]:Zr[Cp(NMe$_2$)$_3$] = 50:50.

![Graph](image)

Fig. S2 Ferroelectric properties of as-deposited HZO thin films deposited using a Cp-based cocktail precursor with a molar ratio Hf[Cp(NMe$_2$)$_3$]:Zr[Cp(NMe$_2$)$_3$] = 50:50: (a) P-E loops of the HZO thin film for the pristine state and the subsequently measured cycles. (b) Variation of the remanent polarization and coercive field as a function of number of cycles measured at an applied electric field of 3.5 MV/cm.

To compare the cocktail precursor composed of a molar ratio of Hf[Cp(NMe$_2$)$_3$]:Zr[Cp(NMe$_2$)$_3$] = 35:65, we additionally investigated the ferroelectric properties of a HZO thin film deposited at 320 °C using a cocktail precursor with a molar ratio of 1:1 without post-annealing. Even in this case, ferroelectric loops were observed without annealing; however, the 2Pr value (~10 μC/cm$^2$) was relatively small.
C. Electrical properties of HZO thin films deposited using a Cp-based cocktail precursor with a molar ratio of Hf\([\text{Cp(NMe}_2\text{)}_3]\):Zr\([\text{Cp(NMe}_2\text{)}_3]\) = 35:65.

![Graphs showing current density-voltage (J-V) and capacitance-voltage (C-V) curves for HZO thin films.]

**Fig. S3** Electrical properties of HZO thin films deposited using a Cp-based cocktail precursor with a molar ratio of Hf\([\text{Cp(NMe}_2\text{)}_3]\):Zr\([\text{Cp(NMe}_2\text{)}_3]\) = 35:65: (a) current density-voltage (J-V) curve of the 10-nm-thick HZO thin film. (b) capacitance-voltage (C-V) curve of the 10-nm-thick HZO thin film measured at 100kHz.

We investigated the electrical characteristics of HZO thin films deposited using a Cp-based cocktail precursor with a molar ratio of Hf\([\text{Cp(NMe}_2\text{)}_3]\):Zr\([\text{Cp(NMe}_2\text{)}_3]\) = 35:65. As shown in Figure S3(a) and (b), the as-deposited HZO based MFM capacitors exhibited good leakage current properties, and a typical C-V butterfly-like curve, characteristics of ferroelectric capacitors, also observed.
D. Schematic diagram for applied pulse for endurance switching cycle test.

![Schematic diagram](image)

<table>
<thead>
<tr>
<th>Voltage</th>
<th>Rising time</th>
<th>Pulse width</th>
<th>Termination</th>
</tr>
</thead>
<tbody>
<tr>
<td>P-E</td>
<td>3.5V</td>
<td>1kHz frequency</td>
<td>Sine wave</td>
</tr>
<tr>
<td>Endurance</td>
<td>20ns</td>
<td>5μs</td>
<td></td>
</tr>
</tbody>
</table>

**Fig. S4** Schematic diagram for applied pulse used to measure P-E endurance switching cycle.

Referring to the previous paper, we conducted the P-E endurance switching cycle measurement with the following waveforms. In the measure section, a 1 kHz triangle wave field was used as in general P-E measurement. In stress section, rectangular wave field with a pulse width of 5 μs and rising/falling time of 2 ns were utilized to switching cycle.
E. Schematics of the transient measurement circuit diagram and HZO-based FeFET.

**Fig. S5** Schematics of (a) transient measurement circuit diagram, (b) the HZO-based FeFET.
References
