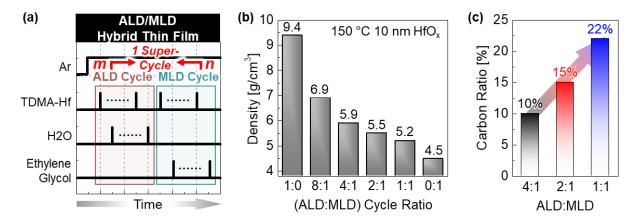
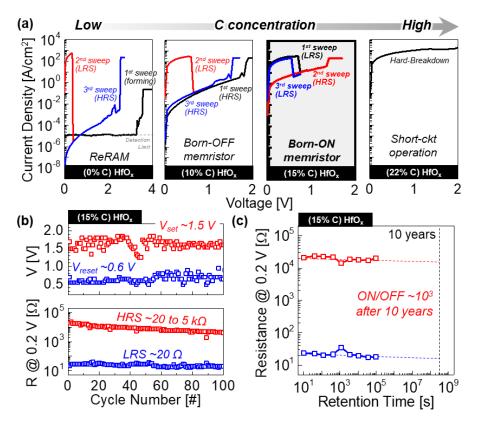
## Supplemental

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**Fig. 1.** (a) Conceptual figure illustrating the super-cycle approach during ALD and MLD cycles. Controllable properties include (b) film densities and (c) carbon composition by adjusting super-cycle protocols. Film densities can range from  $4.5 \text{ g/cm}^3$  to  $9.4 \text{ g/cm}^3$ , and carbon ratio can be cotrolled up to 22% and above.



**Fig. 2.** (a) J–V characteristics of 10 nm-thick HfO<sub>x</sub> memristive devices characterized by different carbon concentrations in HfO<sub>x</sub>. The approach for doping carbon involves incorporating multiple precursors and/or reactants during the ALD process. (b) Endurance of both set/reset and low/high resistance states (LRS/HRS) for up to 100 cycles. Both V<sub>set</sub> and V<sub>reset</sub> exhibit excellent reproducible results of ~1.5 and ~0.6 V, respectively, regardless of the cycle numbers. While the LRS remained constant at 20  $\Omega$ , the HRS decreases from 20 k $\Omega$  to 5 k $\Omega$ . Nevertheless, the ON/OFF ratio is still above 10<sup>2</sup>, which is comparable to other memory devices. (c) Time-dependent retention characteristic of 15% carbon doped HfO<sub>x</sub> memristors. The ON/OFF ratio is expected to be ~10<sup>3</sup> after 10 years.