## **Supplemental**

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**Figure 1.** (a) Schematics of the memristive Ru/HfO<sub>x</sub>/Pt devices deposited by e-beam evaopartor (Pt), ALD at 250 °C (HfO<sub>x</sub>), and DC sputtering (Ru). Especially, for the HfO<sub>x</sub> deposition, we used the carbonated  $H_2O_2$  as an oxygen precursor for achieving the carbon composited films. (b) Current density–voltage (J–V) characteristics of the demonstrated devices without any annealing process. After the high voltage-based formation of HfO<sub>x</sub> film, the devices exhibit the memristive behavior, *i.e.*, ReRAM operation. Unlike the carbon-less HfO<sub>2</sub> operation, the high leakage current was observed in the films. (c) J–V characteristics of the demonstrated devices with RTA at 450 °C for 1 m under N<sub>2</sub>. After annealing, the forming voltage-free could be demonstrated, which is different from the forming behavior in ReRAM devices with respect to the magnitude of switching voltage and current level.



**Figure 2.** (a) Non-polarization switching behavior of the demonstrated devices, investigated by applying same/opposite polarities of operating voltage. (b) Cycle-to-cycle variation of the unipolar operation in the devices without applying forming voltage. The set and reset voltage ( $V_{set}$  and  $V_{reset}$ ) is ~1.6 and ~0.6 V during the cycles. (c) Bipolar operation of the devices, highlighting the great merits of non-polarization switching behavior.