Supplementary Materials

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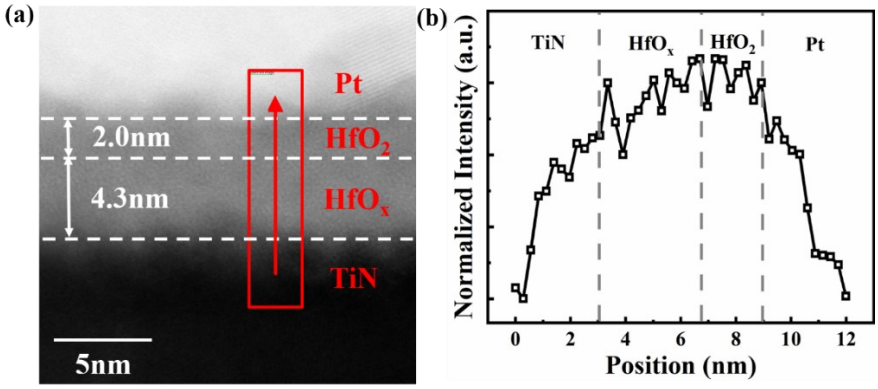

Fig. S1 (a) Cross-section HAADF-STEM image, (b) EELS O intensity in $Pt/HfO_2/HfO_x/TiN$ bilayer memistive devices with HfO_x layer having 20 s H_2 plasma treament time. The test position is the red rectangular area in Fig. S1(a) and the test direction is from bottom to the top.

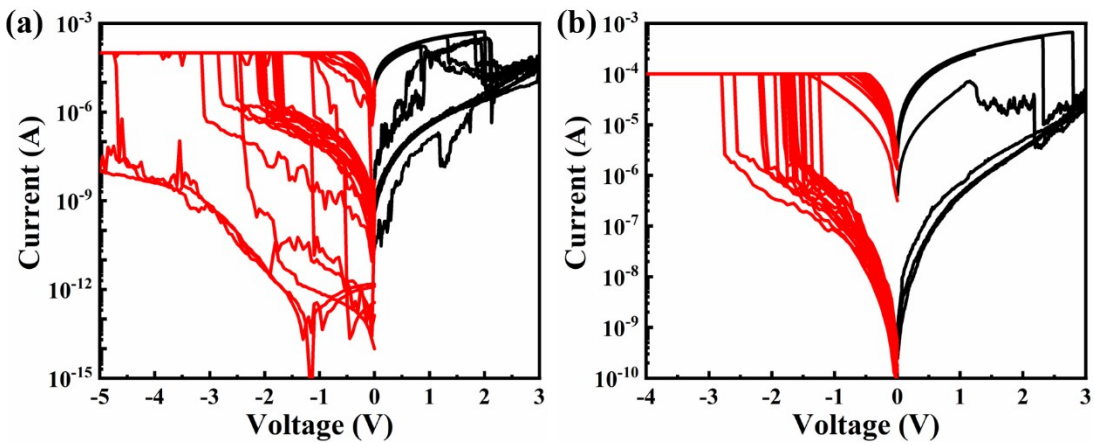

Fig. S2 I-V curves of (a) TiN/HfO_2/Pt and (b) TiN/HfO_x/Pt memristors for 30 times tests by DC double ramp sweep.

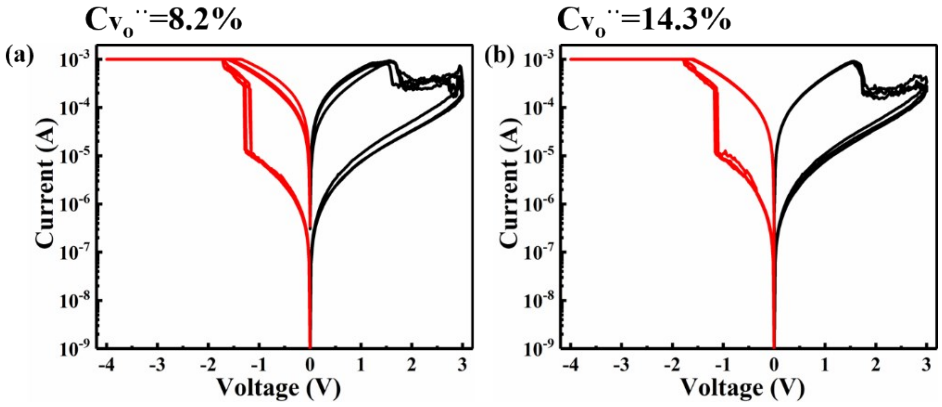

Fig. S3 Comparison of 30 times DC voltages sweep test IV curves between HfO_x layer having (a) 8.2% and (b) 14.3% oxygen vacancy contents in Pt/HfO_2/HfO_x/TiN memristors.

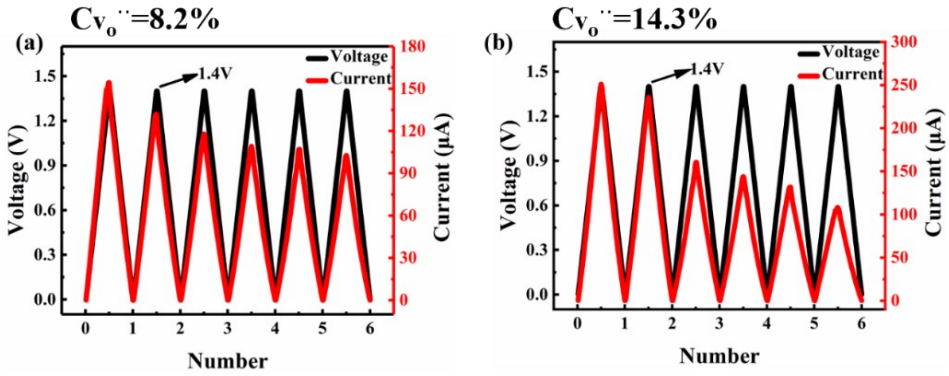

Fig. S4 Comparison of voltage and current versus +1.4 V switching cycle number between HfO_x layer having (a) 8.2% and (b) 14.3% oxygen vacancy contents in Pt/HfO_2/HfO_x/TiN memristors.

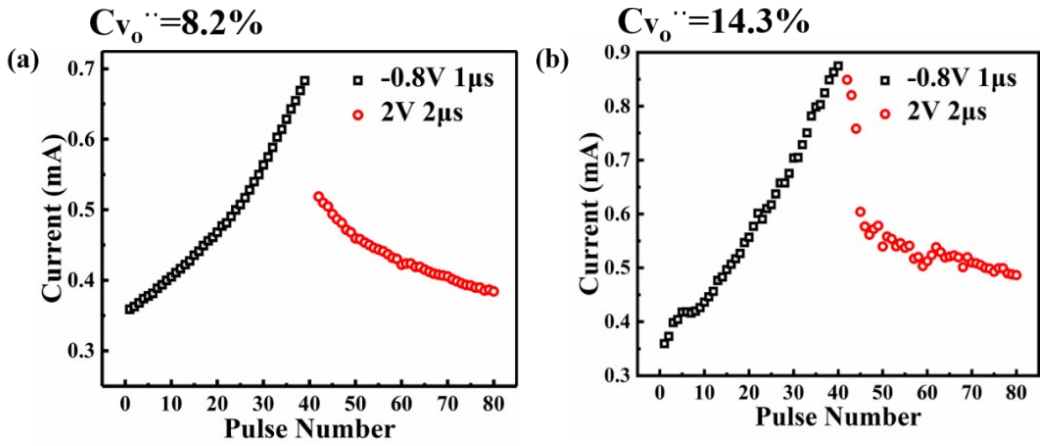

Fig. S5 Comparison of long-term potentiation / long-term depression simulation between HfO_x layer having (a) 8.2% and (b) 14.3% oxygen vacancy contents in $Pt/HfO_2/HfO_x/TiN$ memristors.

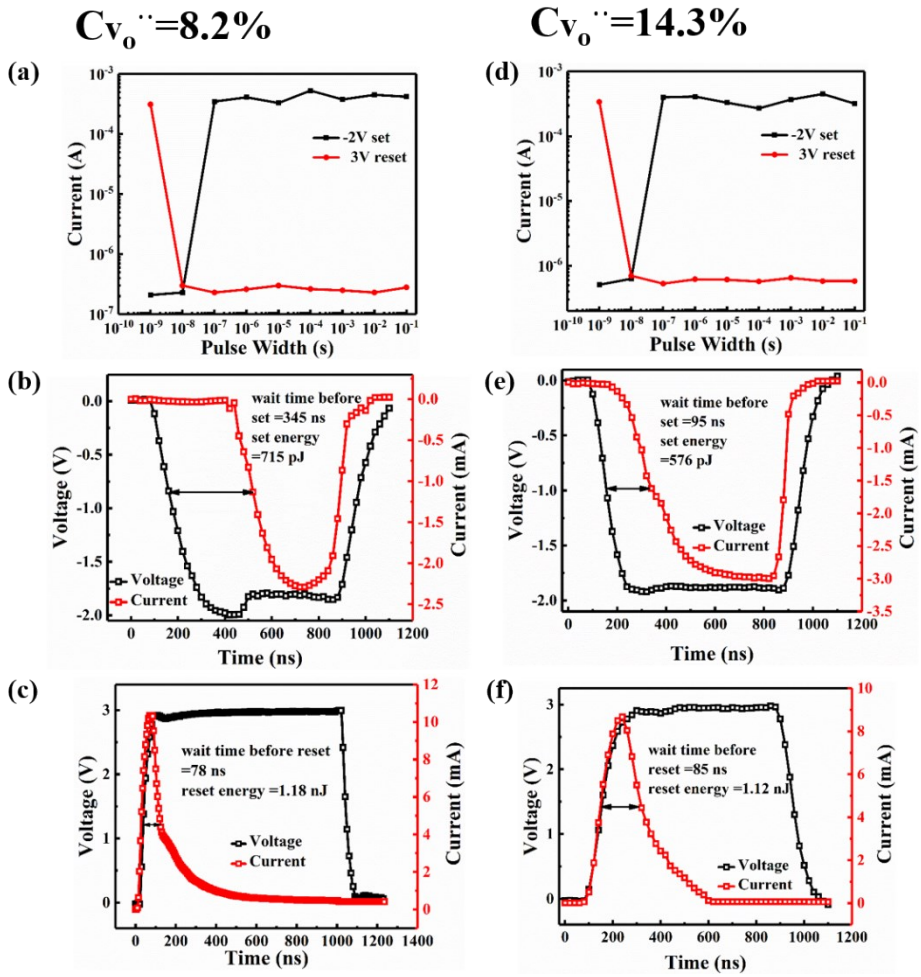

Fig. S6 Comparison of switching speed test and transient current response between HfO_x layer having (a-c) 8.2% and (d-f) 14.3% oxygen vacancy contents in $Pt/HfO_2/HfO_x/TiN$ memristor. Switching speed test of the (a) $C_{Vo^{\cdot\cdot}}$ = 8.2% and (d) $C_{Vo^{\cdot\cdot}}$

= 14.3% synaptic devices. The voltages for set/reset operation in memristors are about -2 V and +3 V, respectively. The two devices can be switched in less than 100 ns for set and less than 10 ns for reset. Transient current response on applied voltage pulse for a set operation between (b) $C_{Vo^{\cdot\cdot}}$ = 8.2% and (e) $C_{Vo^{\cdot\cdot}}$ = 14.3% synaptic devices. All set pulse amplitude is -2 V. Transient current response on applied voltage pulses for a reset operation between (c) $C_{Vo^{\cdot\cdot}}$ = 8.2% and (f) $C_{Vo^{\cdot\cdot}}$ = 14.3% synaptic devices. Reset pulse amplitude is set to be 3 V. All the pulse width, rising time, and falling time are set to be 1000 ns, 20 ns, and 20 ns, respectively.

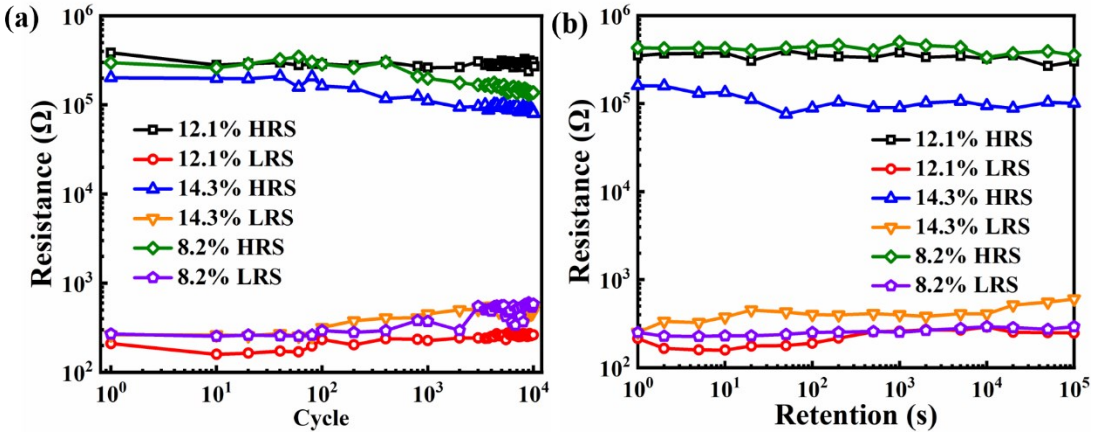

Fig. S7 (a) Endurance and (b) retention characteristics for different oxygen vacancy concentration samples ($C_{Vo^{\cdot\cdot}}$ = 8.2%, 12.1%, and 14.3%) after 10^4 cycles and 10^5 seconds, respectively.