Supplementary Information (SI) for Journal of Materials Chemistry C.

Supplementary Materials

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The estimation method on areal density, average width and height of Ta_2O_{5-x} nano-islands samples based on AFM results is described as follows:

1) Areal density: Draw a 200×200 nm^2 square in 2D AFM image and count the number of nano-islands inside the square using ImageJ software, and then convert the unit to cm^2 to obtain the areal density.

2) Average width: Measure the transverse size of all nano-islands in the above-mentioned 200×200 nm^2 square in 2D AFM image using ImageJ software, then calculate the average width of nano-islands and standard deviation. For 50-cycle sample, 1μm×1μm square is used.

3) Average height: Estimate the average height value based on the height curve of the line scan, for example, in Fig. 2(e) (red line indicated in Fig. 2(d)). Along the height curve, measure the relative height between every peak and the neighboring valley using ImageJ software and then calculate the average height value and standard deviation.

For 10-cycle Ta_2O_{5-x} nano-islands sample, the areal density and average width is estimated based on TEM result in Fig. S2. The average height still comes from the height curve of AFM in Fig. 2(b).

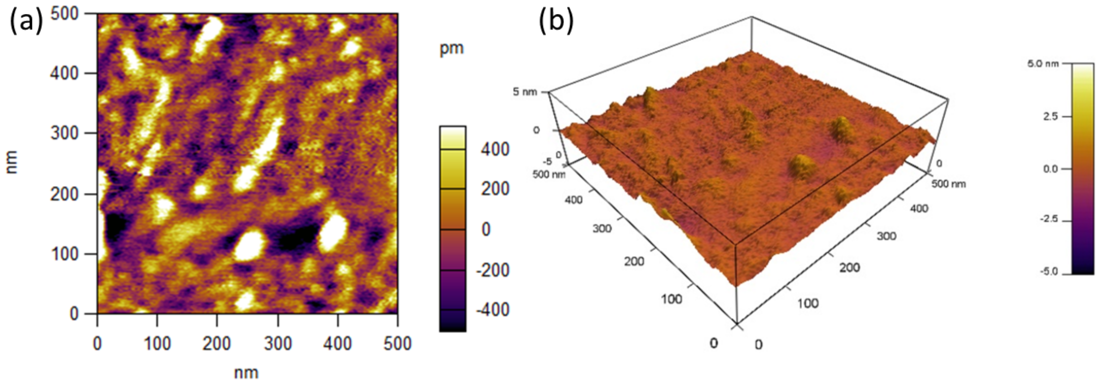

Fig. S1 (a) 2D and (b) 3D AFM images of ALD-derived Al_2O_3 thin films on Si.

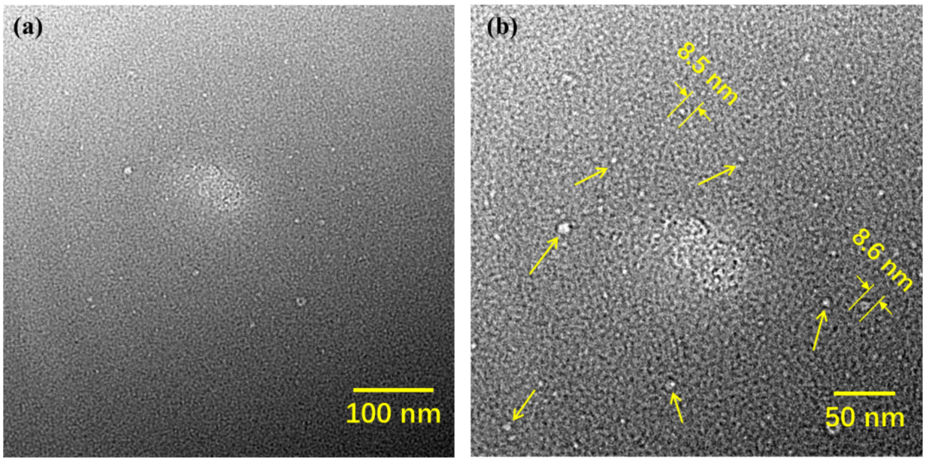

Fig. S2 (a)The surface TEM image and (b)partial enlarged detail of 10-cycle Ta_2O_5 nano-islands.

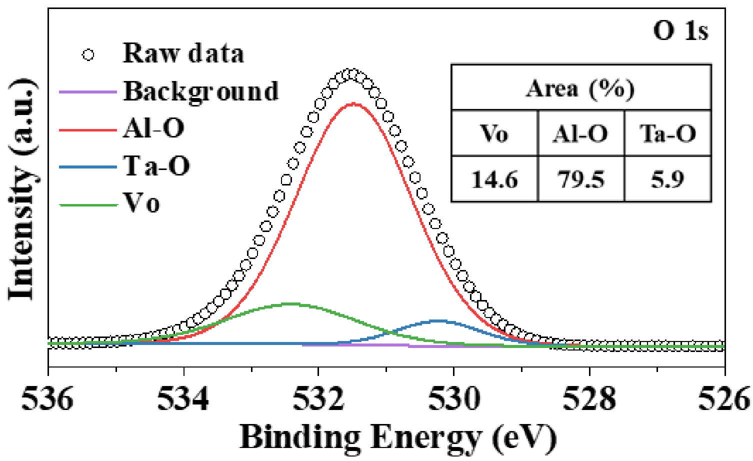

Fig. S3 XPS spectrum of O 1s of 10-cycle Ta_2O_{5-x} nano-islands on Al_2O_3 surface.

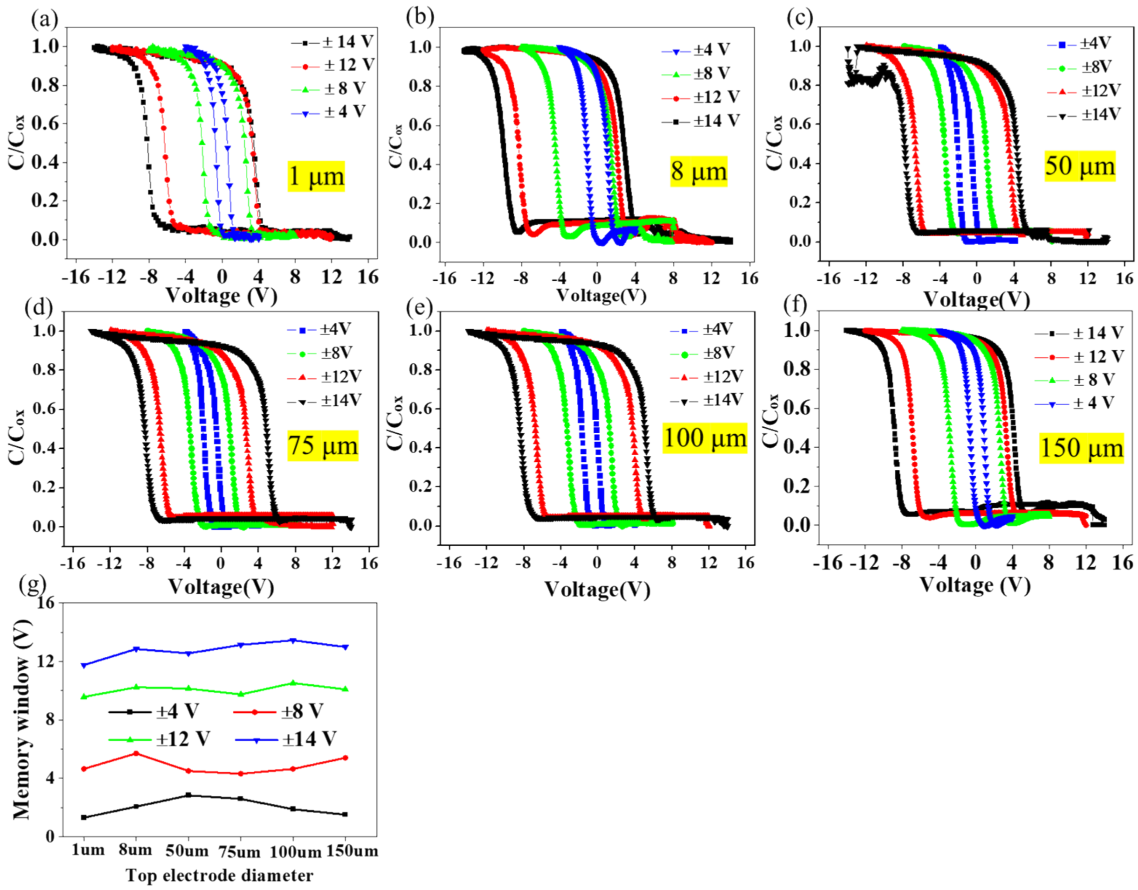

Fig. S4 C-V curves of Ta_2O_{5-x} nano-islands CTM devices with different top electrode diameters: (a) 1 μm, (b) 8 μm, (c) 50 μm, (d) 75 μm, (e) 100 μm and (f) 150 μm. (g) Dependence curve of memory window on sweeping voltage at different top electrode diameters.

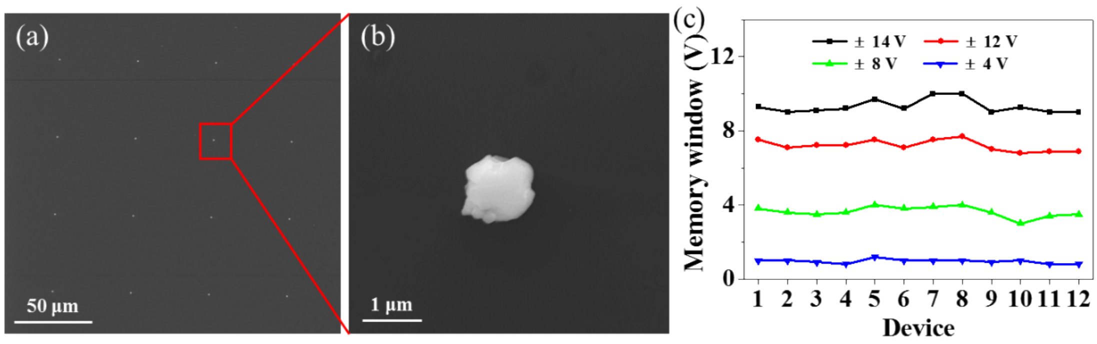

Fig. S5 SEM images of (a) 16 CTM devices with 1 μm top electrodes, (b) an enlarged view of a single CTM one with 1 μm top electrode. (c) Device-to-device variability for memory window on sweeping voltage of 12 CTM devices with 1 μm top electrode diameter.

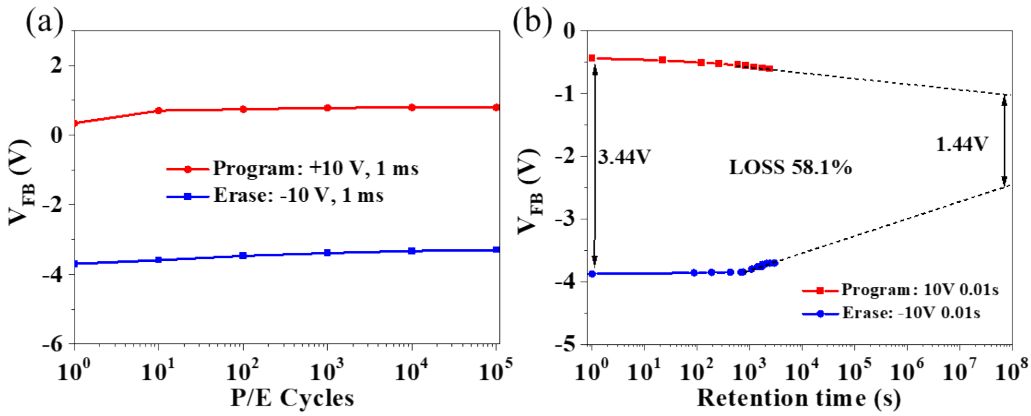

Fig. S6 (a) Endurance and (b) retention of 20-cycle Ta_2O_{5-x} nano-islands CTM device at 0.01 ms and 0.01 s, respectively.

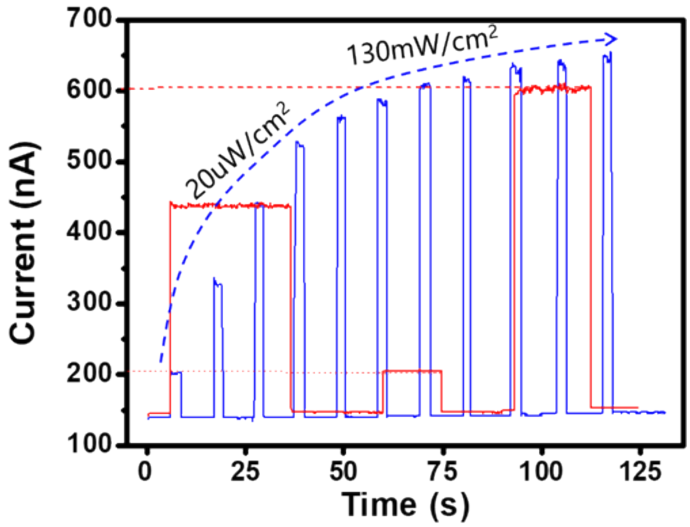

Fig. S7 Photocurrent response of Ta_2O_{5-x} nano-islands CTM under 450 nm light at 0.1 V bias voltage.

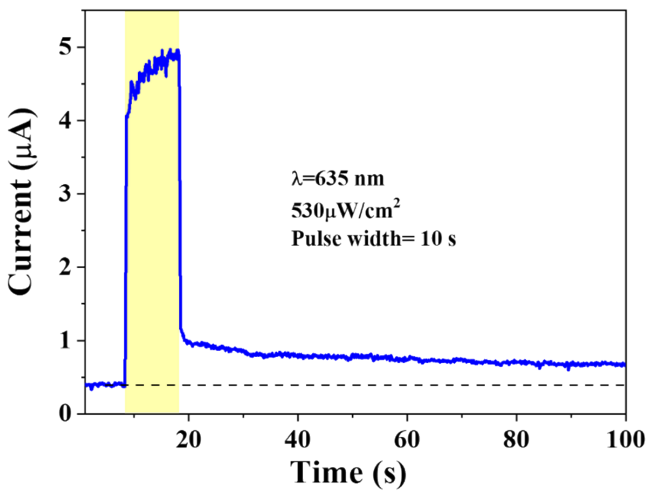

Fig. S8 Photocurrent response of Ta_2O_{5-x} nano-islands CTM under 635 nm light at 2 V bias voltage.