Supporting Information:

Sp^2-clustering induced improvement of resistive switching uniformity in Cu/amorphous carbon/Pt electrochemical metallization memory

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XPS analysis of the amorphous carbon films

The C 1s core level of the as prepared a-C films are decomposed into two components at 284.3 eV and 285.5 eV, which were identified with the sp^2 and sp^3 hybrids forms of carbon. The sp^2/sp^3 hybridization ratio can be estimated from the area of the two components [1]. Our analysis suggests that the a-C has a concentration of sp^2 hybrids and sp^3 hybrids of 73% and 27% respectively.

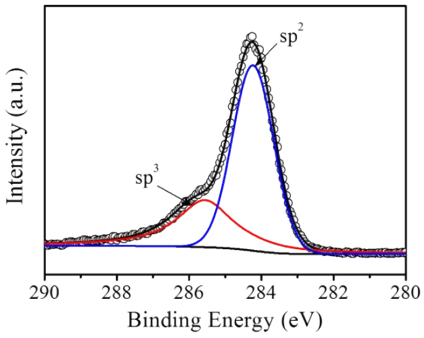

Fig. S1. C 1s XPS spectra of the sputter-cleaned a-C thin film.

Estimation of the resistivity of Cu filament from the LRS.

Size effects come into play as the lateral dimension of the Cu filament becomes comparable to or smaller than the mean free path of the electrons. In the calculation of the resistivity of Cu CF, the electron scattering at the surface and grain boundaries of the CF should be taken into account [2, 3]. According to the model proposed by Fuchs-Shondheimer and Mayadas-Shatzkes [2], the resistivity ρ of Cu filament with a

circular cross section is given by

$$\rho = \rho_0[1 + \frac{3}{4}(1-p)\frac{\lambda_0}{D} + \frac{\rho_0}{1 - \frac{3}{2}\alpha + 3\alpha^2 - 3\alpha^3 \ln(1+\frac{1}{\alpha})}] \quad (1)$$

Where ρ_0 is the resistivity of the bulk Cu, p is the fraction of electrons scattered at the surface, D is the diameter of the Cu CF, λ_0 is the mean free path of the bulk Cu, $\alpha = (\lambda_0 / w)[r/(1-r)]$, w is the mean grain size, and (1-r) is the fraction of electrons scattered by the potential barrier at a grain boundary.

The diameter D of the Cu CF is given by:

$$D = 2\sqrt{\frac{L\rho}{\pi R_{LRS}}} \quad (2)$$

Where L is the a-C film thickness, R_{LRS} is the low resistance state of the device. The resistivity ρ of the Cu filament was calculated using equation (1) and (2). The values of the parameter are given as using p = 0.5, λ_0 = 40 nm, w = 25 nm, and r = 0.34, which were taken from the literature [4].

The repeating set/reset *I-V* curves of the Cu/a-C/Pt device

The repeating set/reset *I-V* curves (one *I-V* curve for each 50 cycles) of the Cu/a-C/Pt device under the compliance current of 10 mA are shown in Fig S2, indicating no cycling degradation in the continuous 1000 switching cycles.

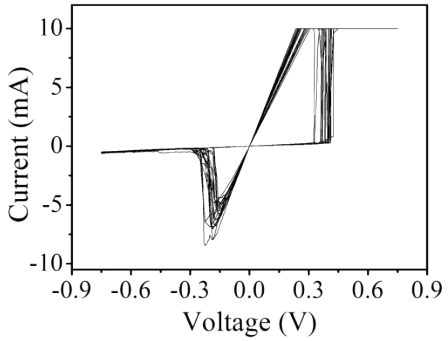

Fig. S2. 1000 (every 50 cycles) repeating set/reset *I-V* curves of the device.

References:

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[2]. Tsuruoka T, Terabe K, Hasegawa T, Aono M. Forming and switching mechanisms of a cation-migration-based oxide resistive memory. Nanotechnology, 2010, 21(42):425205.

[3]. Steinhögl W, Schindler G, Steinlesberger G, Engelhardt M. Size-dependent resistivity of metallic wires in the mesoscopic range. Physical Review B, 2002, 66(7): 075414.

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