

**The excel file contains raw data for the paper. The detailed description are below:**

Figure 1. (b) Electroforming process of the bilayer TiN/ZrO<sub>1.8</sub>/ZrO<sub>2</sub>/TiN device with a CC of 1 mA. (c) I-V characteristics of the device after forming. (d) The endurance characteristics of the bilayer device.

Figure 2. (a) Electroforming process of the bilayer TiN/ZrO<sub>1.8</sub>/ZrO<sub>2</sub>/TiN device with a CC of 1 mA. (b) I-V characteristics of the device for after forming. The endurance (c) and the retention (d) characteristics of the bilayer device.

Figure 3. Cumulative probability graphs of (a) VSET and VRESET and (b) HRS and LRS for both the positive bipolar (PB) mode and negative bipolar (NB) mode on the bilayer TiN/ZrO<sub>1.8</sub>/ZrO<sub>2</sub>/TiN device.

Figure 4. I-V characteristics and the SET process I-V curves in double-logarithmic plots of the (a) single layer TiN/ZrO<sub>2</sub>/TiN device and (b) bilayer TiN/ZrO<sub>1.8</sub>/ZrO<sub>2</sub>/TiN device formed with a CC of 1 mA.

Figure 5. I-V characteristics and the SET process I-V curves in double-logarithmic plots of the bilayer TiN/ZrO<sub>2-x</sub>/ZrO<sub>2</sub>/TiN devices with the composition of the top ZrO<sub>2-x</sub> layer being (a, d) ZrO<sub>1.2</sub>, (b, e) ZrO<sub>1.5</sub> and (c, f) ZrO<sub>1.8</sub> electroformed under a CC of 1 mA.

Figure 6. XPS spectra and their Gaussian fittings of the Zr 3d for (a) ZrO<sub>1.2</sub>, (b) ZrO<sub>1.5</sub>, (c) ZrO<sub>1.8</sub> and ZrO<sub>2.0</sub> films, respectively. Doublets are fitted with the spin-orbit splitting between the 3d<sub>5/2</sub> and 3d<sub>3/2</sub> peaks fixed to be 2.4 eV.

**Date of data collection: from 2015-2016**

**Information about geographic location of the data collection: University of Southampton**

**Date the file was created: 15/03/17**