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

Figure 1. XRD patterns of the as deposited ZrO2/ZrO2-x/ZrO2 tri-layer structure (red), oxygen-deficient ZrOx layer (green), stoichiometric ZrO2 layer (blue) and pure metallic Zr layer (pink).

Figure 2. XPS spectra of (a) Zr 3d and (b) O 1s for the ZrO2/ZrO2-x/ZrO2 tri-layer structure over an etch time of 700 s. (c) XPS depth profile of the ZrO2/ZrO2-x/ZrO2 tri-layer structure.

Figure 3. (a) ADF-STEM image of the cross section of the sample and (b) STEM-EDX elemental line profiles of the TiN/ZrO2/ZrO2-x/ZrO2 structure.

Figure 4. (a) Schematic of the tri-layer TiN/ZrO2/ZrO2-x/ZrO2/TiN memory. (b) *I-V* characteristics of the electroforming process for the TiN/ZrO2/ZrO2-x/ZrO2/TiN device. (c) *I-V* characteristics of the interfacial switching after forming. (d) Current as the function of device size for both HRS/LRS in the interfacial switching mode.

Figure 5. (a) *I-V* characteristics of the transformation process (orange) from the interfacial switching (red) to the filamentary switching (blue). (b) Cumulative probability graph of HRS and LRS for both interfacial (VRESET = 6 V) and filamentary switching modes. The SET process *I-V* curves of the (c) interfacial and (d) filamentary switching modes in double-logarithmic plot.

Figure 6. (a) Schematic of the single layer TiN/ZrO2/TiN device. (b) *I-V* characteristics of the electroforming process for the TiN/ZrO2/TiN device. (c) *I-V* characteristics of the TiN/ZrO2/TiN device after forming. (d) *I-V* curves of the SET process in double logarithmic plot with linear fitting.

Figure 7. Schematics of the switching mechanism on the tri-layer TiN/ZrO2/ZrO2-x/ZrO2/TiN device for the interfacial switching mode (a-e), the transformation from interfacial mode to filamentary mode (f) and the filamentary mode (g-h).

Figure 8. (a) *I-V* characteristics of the interfacial switching for the tri-layer TiN/ZrO2/ZrO2-x/ZrO2/TiN device with varying RESET voltages. (b) Endurance test of 100 cycles for the interfacial switching with different RESET voltages.

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

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

**Date the file was created: 17/05/17**