## Reactive Sputtering Deposition of TiO<sub>x</sub> for Memristive Devices

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 ${
m TiO_x}$ -based memristive devices have attracted considerable attention as one potential choice for the next generation non-volatile random access memory [1]. Such memristive devices possess a simple metal/insulator/metal (MIM) structure: a  ${
m TiO_x}$  functional layer is sandwiched by two metal electrodes. The properties of the  ${
m TiO_x}$  switching layer and the oxide/electrode interfaces are critical to the device performances, which in turn highlights the importance of the thin film preparation process. Here we systematically studied reactive sputtering deposition of  ${
m TiO_x}$  thin films using a mixture of Ar and  ${
m O_2}$  gases under different ratios of  ${
m O_2}$  flow. With this simple technique, desired oxygen vacancy profiles inside the switching materials can be engineered and designed switching properties can be implemented in single or bi-layer memristive devices.

In this study, the Pt bottom electrode (BE) was shared for all devices on the same wafer and the Pt top electrodes (TEs) were isolated metal disks of 50 µm diameter. Both the BEs and TEs were deposited in an electron beam evaporator, and a metal shadow mask was employed during the deposition of the TEs. Both single switching layer and bilayer devices were made in this work, and their geometries (from bottom to top) were 5 nm Ti/10 nm Pt //20 nm TiO<sub>x</sub>//20 nm Pt and 2 nm Cr/20 nm Pt //20 nm TiO<sub>x</sub>//20 nm TiO<sub>x</sub>//20 nm TiO<sub>x</sub>/ layers were deposited through the reactive sputtering process while the top TiO<sub>2</sub> layer in the bilayer devices was deposited by RF-sputtering from a ceramic TiO<sub>2</sub> target, all at room temperature. During the measurements, the bottom electrodes were grounded while the top electrodes were biased.

Deposition rate dependence on the  $O_2$  flow ratio was shown in Fig. 1. The target changed from a metallic mode to an oxide mode when the  $O_2$  flow ratio was increased beyond a certain value (40%), together with a sudden drop on the deposition rate. Single layer devices based on these thin films exhibited a wide spectrum of electrical behavior such as Ohmic, rectifying and resistive switching (Fig. 2). Thin films deposited at the metallic target mode can be applied to engineer the electrical performance of bilayer memristive devices. With a low  $O_2$  flow ratio (7% for example), the bilayer device exhibited typical bipolar resistive switching behavior (Fig. 3(a)). When the  $O_2$  flow ratio was increased to higher than 15% for the bottom  $TiO_x$  layer (25% for example), the bilayer device was unipolar (Fig. 3(b)).

The above evolutions in both single layer and bilayer devices are attributed to the different chemical compositions of sputtered films under different flow ratios. Physical characterizations including X-ray photoelectron spectroscopy analyses have been conducted as solid supports.

[1]. Yang, J. J et al. Nature Nanotech. 3, 429 (2008).

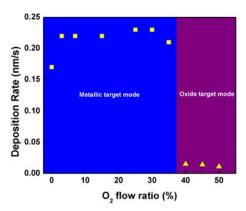


Figure 1. Deposition rate dependence on the  $O_2$  flow ratio. The deposition changed from a metallic target mode into an oxide target mode when the oxygen flow ratio is 40% or higher. The blue and purple colors in the chart represent the plasma color at the metallic and oxide target modes, respectively.

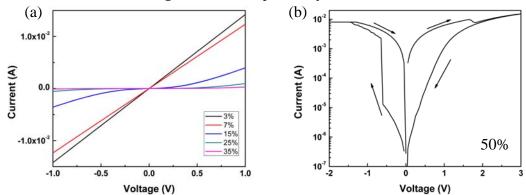


Figure 2. Electrical behavior for single layer devices made from TiOx prepared with different oxygen flow ratios. (a) I-V curves from single layer devices based on films prepared with  $O_2$  flow ratios varied from 3% to 35%. A transition from linear to nonlinear behavior is observed. (b) Typical I-V curve from a single layer device based on the film deposited with 50%  $O_2$  flow ratio. Bipolar resistive switching behavior is observed in this device. The black arrows indicate the switching directions.

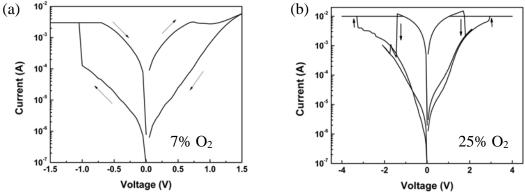


Figure 3. Electrical behavior for double layer memristive devices with  $TiO_x$  as the base layers. (a) Typical bipolar resistive switching I-V curve from the bi-layer device with the base layer prepared under an  $O_2$  flow ratio of 7%. (b) Typical unipolar resistive switching I-V curve from the bi-layer device using the base layer prepared with 25%  $O_2$  flow ratio. The black arrows indicate the switching directions.