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# Observation of multi-conductance state in solution processed Al/a-TiO<sub>2</sub>/ITO memory device

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#### ABSTRACT

In the present study, fabrication of memory devices using sol–gel derived amorphous titanium oxide (a- $TiO_2$ ) thin films and its characterization are presented. Titanium oxide thin films of thickness 50 nm were deposited on ITO substrates using a spin coating method. The films annealed at 200 °C for an hour in air atmosphere have shown good transparency in visible region. Bipolar resistive switching behaviors of Al/a- $TiO_2$ /ITO stacked structures were investigated using current–voltage characteristics. The observed current–voltage characteristics have described the bipolar resistive switching property of a- $TiO_2$  films. Multi-step conductance behavior has been observed in this Al/a- $TiO_2$ /ITO stacked structures and its mechanism has been analyzed.

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#### 1. Introduction

Resistive switching memory (ReRAM) devices based on binary transition metal oxides (TMO) such as TiO<sub>2</sub>, NiO<sub>x</sub>, and Cu<sub>x</sub>O have recently attracted extensive research interests in next generation nonvolatile memory applications. Particularly, they are attractive for low cost, low power consumption and fast operation; they are also easy for high density integration [1-3]. Comparing to  $Pr_{1-x}Ca_xMnO_3$ [4], La<sub>1-x</sub>Ca<sub>x</sub>MnO<sub>3</sub> [5] or Cr-doped SrZrO<sub>3</sub> [6] materials, the simple binary oxide TMO materials exhibit advantages not only for their simplicity in fabrication process, but also for their compatibility with CMOS processes. Among the large varieties of binary oxides, titanium dioxide (TiO<sub>2</sub>) is one of the most promising switching materials due to its simple composition and versatile resistive switching (RS) properties in both unipolar (URS) [7] and bipolar [1] resistive switching (BRS) devices. The solution processed TiO<sub>2</sub> has recently gained attraction considering its inexpensive synthesis route and large-area fabrication possibilities [8,9]. Many researchers have reported resistive switching characteristics of TiO<sub>2</sub> films prepared by solution process, and furthermore they have also suggested versatility of solution processed TiO<sub>2</sub> in the applications of memristors, ReRAMs and flexible ReRAMs [10-12]. In the memory devices, the multilevel switching is one of the interesting features that could be applied to future multibit memory devices [13].

In this work, we have reported multilevel switching property of Al/a-TiO<sub>2</sub>/ITO stacked structure using sol–gel prepared a-TiO<sub>2</sub>

\* Corresponding author. *E-mail address:* jkrhee@dgu.edu (J.-K. Rhee). films. The spin coated  $a-TiO_2$  thin films were characterized using XRD, SEM and UV–Vis analysis. The possible mechanism for the multilevel switching of the fabricated memory device has been discussed.

#### 2. Experimental details

Fig. 1 shows the schematic of ReRAM device fabricated with Al/ a-TiO<sub>2</sub>/ITO structure on a glass substrate. To fabricate this Al/TiO<sub>x</sub>/ ITO memory device, amorphous titanium oxide thin films of thickness 50 nm were deposited on a commercial ITO substrate by a spin coating method. Titanium (IV) isopropoxide (Ti[OCH(CH<sub>3</sub>)<sub>2</sub>]<sub>4</sub>, Junsei, Japan) was used as precursor and 2-butoxy ethanol(ethylene glycol mono butyl ether) used as a organic solvent in this study. The starting precursor solution of 4 ml titanium (IV) isopropoxide was mixed with 20 ml of 2-butoxy ethanol by constant stirring at 70 °C for an hour. Prior to the deposition of the TiO<sub>2</sub> film, the ITO substrate was cleaned in acetone and iso-propanol for 15 min each using an ultrasonic cleaner, and then after washing with deionized water dried using N2 gas. The solution cooled to room temperature was spin coated onto ITO substrate at a speed 3000 rpm for 30 s using a spin coater. The prepared films were then annealed at 200 °C for an hour in air atmosphere. For the fabrication of device, Al top electrodes (TEs) of thickness 100-nm with different areas were evaporated on the a-TiO<sub>2</sub> films using a shadow mask by thermal evaporation. X-ray diffraction (XRD) pattern of the film was obtained using XPERT-PRO X-ray diffractometer (XRD) with CuK $\alpha$  radiation (1.5406 Å). Optical transmittance spectrum of the a-TiO<sub>2</sub> film was measured using a UV-visible-near-IR





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Fig. 1. Schematic of constructed Al/a-TiO<sub>2</sub>/ITO memory device.

spectrophotometer. The electrical measurements on the fabricated Al/a-TiO<sub>2</sub>/ITO structures were performed using a Keithley 4200DC parameter analyzer. All the electrical measurements were done at room temperature in a clean room of class-1000.

#### 3. Results and discussion

#### 3.1. Optical and structural studies

Fig. 2 shows the room temperature optical transmittance spectrum of the a-TiO thin film deposited on a ITO substrate. The optical bandgap Eg can be determined using the absorption coefficient and considering the relation  $(\alpha h v)^2$  vs. hv for the direct allowed transition of TiO<sub>2</sub> [14]. Inset of Fig. 1 shows the  $(\alpha hv)^2$  vs. hv plot, the extrapolation of the linear portion of the plot to  $(\alpha h v)^2 = 0$ gives the optical bandgap value (Eg) of the material. The calculated band gap value (Eg = 3.65 eV) indicates that the prepared a-TiO<sub>2</sub> film is highly transparent in the visible region. XRD spectrum of the prepared TiO<sub>2</sub> films is shown in the Fig. 3; it depicts only ITO peaks indicating the amorphous nature of prepared TiO<sub>2</sub> film. The film prepared at room temperature was annealed at 200 °C for 30 min in air atmosphere, which is well below the crystallization temperature of the two major phases of TiO<sub>2</sub> such as anatase and rutile, hence the amorphous nature has been obtained. The inset of the Fig. 3 shows the morphology of the prepared a-TiO<sub>2</sub> film.

#### 3.2. Electrical measurements

Fig. 4(a) shows the typical *I–V* characteristic of an Al/a–TiO<sub>2</sub>/ITO memory cell measured in the voltage range ( $6 \sim -5$  V) under the dc voltage sweep at room temperature. During all the electrical



**Fig. 2.** Transmittance spectra of TiO<sub>x</sub> films coated on ITO substrate. Inset shows the calculated optical bandgap for direct allowed transition of the solution-processed  $TiO_x$  film.



Fig. 3. XRD diffractogram of the  $TiO_2$  films deposited on the ITO substrate (\* indicates ITO), inset: SEM micrograph of the film.

measurements, an external bias voltage was applied to the Al top electrode, and the ITO bottom electrode was electrically grounded. The observed I-V curve exhibits a clear hysteretic and asymmetric behavior indicating the bistable resistance switching between high-resistance state (HRS) and low resistance state (LRS) obtained by forward and reverse voltage scans, respectively. Switching of the device from HRS to LRS during the positive sweep of the applied electric field is known as the set process. The reset operation (LRS to HRS) is carried out by sweeping the electric field in the



Fig. 4. (a) single and (b) 10 consecutive dc voltage sweep of Al/a-  $\rm TiO_{x}$  /ITO memory device.

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