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# Oxygen-doped zirconium nitride based transparent resistive random access memory devices fabricated by radio frequency sputtering method



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# ABSTRACT

In this work, we present a feasibility of bipolar resistive switching (RS) characteristics for Oxygen-doped zirconium nitride (O-doped ZrN<sub>x</sub>) films, produced by sputtering method, which shows a high optical transmittance of approximately 78% in the visible region as well as near ultra-violet region. In addition, in a RS test, the device has a large current ratio of  $5 \times 10^3$  in positive bias region and  $5 \times 10^5$  in negative bias region. Then, to evaluate an ability of data storage for the proposed memory devices, we measured a retention time for  $10^4$  s at room temperature (RT) and 85 °C to ~ $10^3$  at RT. This result means that the transparent memory by controlling the working pressure during sputtering process to deposit the ZrN<sub>x</sub> films could be a milestone for future see-through electronic devices.

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## 1. Introduction

In recent, it is necessary to develop new memory concept with two terminals, not conventional three-terminal devices, i.e., transistor-based memory like including floating gate (FG) type memory and charge trap flash (CTF) memories, since the rapidly elevated demand for high-density devices by scaling them down is expected to be a major issue because of technical and physical limitations [1–3].

In order to solve the mentioned issues, several memory concepts such as phase change random access memory (PRAM), ferroelectric RAM (FRAM), magnetic-resistive memory (MRAM), and resistive RRAM (RRAM) have been studied. Among them, a conducting-filament (CF) based RRAM has attracted much attention as one of the most promising candidates for new memory concept because of its simple structures, low voltage operation, high density, transparency, and fast speed [4–6]. In general, to achieve fully transparent RRAM devices, wide-bandgap resistive switching materials as well as transparent top/bottom electrodes should be employed. Recently, although related some studies have been reported based on metal oxides and metal nitride, their resistive switching (RS) properties (i.e., set/reset voltages and current) fluctuate seriously in a limited numbers of operating cycles, and the conduction mechanism should also still be clearly identified. Therefore, at the moment it is still a pending challenge to realize a stable memory operation in active layers. Especially, to realize fully transparent RRAM (T-RRAM) cells applying for the transparent electronic applications, it is required to develop the transparent active layer as well as transparent electrode. In some previous literature related, indium tin oxide (ITO) has been employed as both bottom and top electrodes to form a high transmittance for whole structures of RRAMs. But in this case, although it is possible to improve its transparency, the device should be worked in unipolar switching mode only, not operating in bipolar switching mode. When considering a reliability (i.e., disturb phenomena) in array structures and power consumption, it is much more favorable to adopt RRAM structures having bipolar switching behavior; generally bipolar RRAM can operate in lower voltage/current level and obtain higher read-margin.

In our earlier work [7,8], we reported a feasibility of stable bipolar RS characteristics in a  $ZrN_x$  films as RS materials and successfully demonstrated. In this work, so as to realize a fully T-RRAM cells, we optimized the transmittance of  $ZrN_x$  films that is increased

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by increasing a working pressure in sputtering process and used the ultra-thin metal electrode of 5 nm as top/bottom electrodes. To examine RS properties of T-RRAM devices, we measured current–voltage (I-V) curve characteristics in a direct-current (DC) mode and the retention time. We also discuss a possible mechanism for bipolar RS properties by applying the Poole-Frenkel and space charge limited conduction models.

## 2. Experimental details

To prepare the samples, 5 nm-thick ultra-thin Ti-bottom electrodes were deposited on quartz substrates using a radio frequency (RF) sputtering system. Then the 50 nm-thick oxygen-doped  $ZrN_x$  (O-doped  $ZrN_x$ ) films were deposited using a reactive RF sputtering system with ZrN target in Ar/N<sub>2</sub> (15 sccm/5 sccm) ambient at room temperature, by controlling working pressure in a range of 5 mTorr-20 mTorr. Finally, to achieve a fully transparent MIM structure, we employed an ultra-thin 5 nm-thick Pt as top electrode. The devices have a 50  $\mu$ m-diameter. For electrical measurement, the top electrodes of the samples were connected to the Agilent 4156B parameter analyzer while the bottom electrodes were grounded. Fig. 1(a) shows the schematic drawings of the proposed Pt/O-doped ZrN<sub>x</sub>/Ti memory cell and in Fig. 1(b) we can

**(a)** 



Fig. 1. (a) Schematic drawings and (b) fabrication procedure in detail of Pt/O-doped  $\rm ZrN_x/Ti/quartz$  T-RRAM cells.

see the sequence of a manufacturing process in detail.

#### 3. Results and discussion

First, in order to optimize a transparent property of  $ZrN_x$  films for T-RRAM applications, we investigated a change of atomic concentration of O, N, and Zr with different stoichiometry of  $ZrN_x$  films that was controlled by changing a working pressure (WP) in RF sputtering process. Note that native oxygen concentration in a chamber of RF sputter can be increased by increasing the WP. In this work, we changed the WP in a range of 5 mTorr–20 mTorr when depositing  $ZrN_x$  films. As shown in Fig. 2(a), we investigated an atomic concentration of  $ZrN_x$  films deposited in the WP of 5, 10, and 20 mTorr by analyzing an auger electron spectroscopy (AES). In this result, Zr and N concentrations were reduced from 50.86% (5 mTorr) to 34.43% (20 mTorr) by increasing the WP, while the increased O concentration from 6.87% (5 mTorr) to 25.3% (20 mTorr) was detected in a higher WP due to a higher native O concentration than that of films deposited in a lower WP.

And then we fabricated T-RRAM by employing the 50 nm-thick O-doped  $ZrN_x$  film deposited in 20 mTorr and estimated its transparency as shown in Fig. 2(b). In this figure, in  $ZrN_x/Ti$  films, we



**Fig. 2.** (a) Atomic concentration of nitrogen, oxygen, and zirconium in the ZrN<sub>x</sub> films according to the working pressure in a range of 5 mTorr–20 mTorr, measured by an auger electron spectroscopy (AES). (b) Transparency of Pt/O-doped ZrN<sub>x</sub>/Ti memory structures fabricated by 20 mTorr before and after annealing process.

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