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Oxygen annealing effect on resistive switching characteristics of multilayer CeO$_2$/Al/CeO$_2$ resistive random-access memory

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Abstract
Cerium oxide-based memristors have been extensively studied because of their compatibility with CMOS technology. Yet, inconsistency of resistive switching parameters is one of the main contests in development of nonvolatile memory for commercialization. Owing to filamentary nature of the resistive switching devices, variability of the resistive switching characteristics can be reduced by doping, where conductive filaments can easily grow due to reduction in the formation energy of oxygen vacancies. In this work, multilayer CeO$_2$/Al/CeO$_2$ films were prepared through radio-frequency (rf) sputtering at room temperature to study the effect of oxygen annealing on the resistive switching characteristics. Device with CeO$_2$/Al/CeO$_2$ multilayer structure after annealing exhibits reduction of defects and improved switching endurance, good data retention, and uniformity in operational parameters. The resistive switching characteristics have been simulated using space charge limited conduction and Schottky emission at high field region of the high resistance state, which is well fitted by linear curve fitting analysis. Improvement in the switching characteristics revealed that Al charge trapping layer has diffused into the CeO$_2$ matrix, which might have resulted in lower density of oxygen vacancies due to oxygen annealing. Experimental I–V analysis indicate that oxygen annealing is an effective approach to enhance the switching characteristics of RRAM devices.

1. Introduction

Oxide-based resistive random-access memory (RRAM) has attracted attention as a promising candidate for next generation nonvolatile memory due to its low switching voltage (<3), fast speed (<10 ns), excellent scalability (<10 nm), simple structure, and compatible with COMS technology [1]. As RRAM resistivity is electrically switched between high and low resistance state to display unipolar or bipolar switching depending on polarity of the applied voltage bias [2]. For better design of RRAM parameters, researchers concern involved an improved endurance, long data retention and good uniformity in RRAM switching parameters. And all are done by only bilayer or multilayer oxides of suitable materials [3]. By increasing the number of stable states with binary, ternary, bipolar or unipolar, we could bitterly enhance the design of multilevel storage of RRAM [4]. Up to now, researchers classify the RRAM into four types of mechanisms with respect to their resistive switching behaviors: (1) Anion-type RRAM: redox reaction and migration of oxygen ions, (2) Cation-type RRAM: redox reaction and migration of cations, (3) Carbon-based RRAM: the stretch of C–C bond lengths due to oxygen and hydrogen dual ions, (4) Oxide-based electrode: oxygen accumulation in oxide-based electrode [5].

Most commonly common resistive switching mechanisms proposed in RRAM devices can be categorized into oxygen ions-based oxide RRAM (OxRRAM) and conductive bridge RAM (CBRAM) [6]. In OxRRAM, the resistive switching mechanism is governed by formation and annihilation of conductive filaments formed by either oxygen vacancies or movement of ions inside the RRAM devices [7]. However, in CBRAM, a metal cation is necessary to develop resistive switching behavior by constructing and destructing the conductive bridges.
formed through electrochemical process [8–11]. Moreover, it is mandatory for IT industry to enhance electrical characteristics by exposing MOSFETs to high temperature annealing [12].

Even through resistive switching characteristics have been realized in various rare-earth oxides, ceria (CeO$_2$) is considered as a promising switching material for RRAM applications due to its unique features such as high-k (dielectric constant ~26), thermally stable, bipolar switching, large memory window, and two valence cations states [13–15]. The CeO$_2$ is greatly used as the planer insulator which has cubic structure with lattice constant a$_0$ = 5.411 Å. The broad dispersion in switching parameters might cause overgrowth of conductive filaments. Formation of interfacial layer and readout hazards after long term operations hinder the use of RRAM for practical applications. Thus, looking for an effective approach to improve the uniformity of resistive switching parameters in oxide is crucial. So far, some methods have been adopted to minimize the dispersion in switching parameters such as, doping, bilayer structure, embedding nanoparticles, and oxygen annealing. Among them, oxygen annealing is one of most effective approach in reducing the strength of oxygen vacancies, which consequently enhances the switching parameters. Chen et al [16] demonstrated that water-soluble Cs$_3$PbBr$_3$ perovskite thin films were synthesized at low temperature through a facile chemical deposition technique to investigate transient resistive switching for nonvolatile memory. Hao et al [17] studied both resistive and magnetization switching in a Pt/NiFe$_{1.95}$Cr$_{0.5}$O$_4$/Pt device though manipulating the applied electric filed. Chang et al [18] investigated the role of Fe$_2$O$_3$ insertion layer to improve the reliability of Ag/Ta$_2$O$_5$/Pt resistive switching devices. Yao et al [19] examined the annealing atmospheric effect on resistive switching and magnetic properties of spinel Co$_3$O$_4$ films. Yawar et al [20] introduced rapid thermal annealing in atomic layer deposited ZrO$_2$ thin film, and found improvement in endurance, uniformity and ON/OFF ratio, respectively. Yang et al [21] reported that memory devices after rapid thermal annealing demonstrated remarkable improvement in switching characteristics as compared to as-fabricated devices. In our previous study, we reported that crystal structure of CeO$_2$ film is influenced by oxygen annealing treatment, and an amount of oxygen vacancies in the bilayer structures CeO$_2$/TiO$_2$ and ZnO/CeO$_2$ films can improve the switching parameters [22, 23]. Jin et al [24] found significant improvement in bipolar resistive switching characteristics after annealing at 300 °C in nitrogen atmosphere. According to our previous study and literature, effect of oxygen annealing environment on resistive switching characteristics of CeO$_2$-based RRAM device appears to be suitable for improving the switching parameters.

In this paper, multilayer CeO$_2$/Al/CeO$_2$ films were sandwiched between high oxygen affinity metallic top electrode (TaN) and high work function metal bottom electrode (Pt, 6.5 eV). The effect of oxygen annealing on resistive switching characteristics was systematically investigated. Characteristics of the annealed multilayer CeO$_2$/Al/CeO$_2$ thin film-based device show much better results as compared to the controlled device due to the decrease in thermal conductivity of Al ions. However, diffusion of Al into CeO$_2$ on annealing treatment leads to the somewhat improved performance like stability and better retention behavior. Both devices are explained in terms of altered status of Al atom diffusion in the CeO$_2$ matrix during oxygen annealing treatment, oxygen vacancies based filamentary model has been proposed to explain the switching mechanism.

2. Experimental procedure

A CeO$_2$ (~10 nm)/Al (~1 nm)/CeO$_2$ (~10 nm) multilayer structure was deposited on Pt/Ti/SiO$_2$/Si substrates by radio frequency (RF) sputtering at room temperature. The CeO$_2$ layers were fabricated by a reactive sputtering method (argon: oxygen = 6:18 sccm), while Al layer was deposited through a simple sputtering mode (argon = 20 sccm). More experimental details can be found in our previous publications [25, 26]. Subsequently, to study the effect of oxygen annealing on resistive switching characteristics, multilayer CeO$_2$/Al/CeO$_2$ films was thermally annealed at 400 °C for 30 min under oxygen (~1.2 × 10$^{-4}$ mbar) ambient in a horizontal furnace. As previously reported, the annealing in oxygen atmosphere is one of the effective approaches to create low density defects in the CeO$_2$ films. Then, using metal shadow mask with 150 μm × 150 μm size, TaN top electrodes were fabricated on top of CeO$_2$/Al/CeO$_2$/Pt by direct current magnetron sputtering at room temperature. The resistive switching behavior, forming, endurance characteristics, and data retention of the controlled (TaN/CeO$_2$/Al/CeO$_2$/Pt) and annealed (TaN/CeO$_2$/Al/CeO$_2$/Pt) devices were analyzed by using two probe measurement system with B1500A semiconducting analyzer.

3. Result and discussions

Results of direct current (DC) current-voltage (I-V) measurements are shown in figure 1. Before measuring the resistive switching characteristics, forming process is compulsory to activate all memory cells of controlled and annealed devices. Forming process is publicized in combination with typical switching sweep of the controlled and annealed devices. Noted that forming was required to activate both memory cells on negative polarity of the bias. After that both controlled and annealed devices were positively switched from high resistance state (HRS) to low
resistance state (LRS, SET process) and negatively switched from LRS to HRS, leading to a typical nonvolatile bipolar switching behavior. During electrical measurements, bias voltage was applied to the top electrode, while bottom electrode was grounded. Moreover, a compliance current of 10 mA was imposed to save the both devices from permanent breakdown during forming and SET processes. And RESET process of both devices are free from current compliance. As shown in figures 1(a), (b), forming voltages are much higher than SET-voltages of both devices. As can be seen the fresh state resistances (before forming) are almost similar to the HRS resistances after RESET process.

Figure 1(c) reveals the statistical distribution of forming voltages for both controlled and annealed devices, where 10 memory cells were selected for random measurements to evaluate the cell-to-cell uniformity of forming voltages of the both memory devices. We found that the average values of forming voltage are ∼8.5 V and ∼7.5 V for controlled and annealed devices, respectively. Figure 1(d) exhibits that HRS resistance of controlled memory devices is higher than that for annealed devices. The box plot for 10 memory cells of 20 cycles each for controlled and annealed devices are shown in figure 1(d). It can be seen that annealed devices have relatively better HRS resistance distribution as compared to controlled devices. This could be explained by the higher diffusivity of Al atoms into the CeO$_2$ films due to oxygen annealing as compared to controlled devices, which results in the large variations of filament formation and rupture in the controlled devices. Such variations in filament formation and rupture in turn result in larger HRS variations for controlled devices. This might happen because insertion of Al layer is expected to create imperfections such as interstitials and particularly oxygen vacancies [27] that cause a reduction of forming voltage. Furthermore, Al atoms loose electrons to become Al$^{3+}$ ions and diffuse [28] into CeO$_2$ layers on annealing which seems responsible for improving the switching characteristics. In addition, an improvement in the crystalline nature of the annealed devices could provide an easy path for the conduction of electrons since our earlier publications have shown that the as-fabricated CeO$_2$ based devices exhibit weak polycrystalline structure.

To further investigate the cycle-to-cycle switching performance for high density non-volatile memory applications, 100 consecutive DC switching cycles were performed on controlled and annealed devices, as shown in figures 2(a), (b). It is noticed that switching I-V curves are much scattered in the controlled device, while annealed device shows improved uniformity, in which overlapping could be observed. During programming
and erasing 100 consecutive DC switching cycles, spread of SET- and RESET-voltages are also investigated systematically, as shown in figures 2(c), (d). The magnitudes of SET- and RESET-voltages were collected from figures 2(a), (b) for further comparison. The histogram gives clue to elaborate the variability of voltages during the switching cycles. This information could be helpful to design the threshold voltage level to distinguish both SET and RESET processes. The standard deviation (\(\sigma\)) and mean value (\(\mu\)) for SET- and RESET-voltages are calculated. For both controlled and annealed devices, coefficients of variance (\(\sigma/\mu\)) for RESET and SET phenomena are 3.1\%, 5.2\%, and 3.5\%, 2.1\%, respectively. We can see that SET voltages of annealed devices are lower than those of controlled device, which can be explained by decreased formation energy of oxygen vacancies in the vicinity of Al atoms \([29]\). Here, we have shown that good uniformity can be achieved using

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**Figure 2.** Typical bipolar I-V curves depicting 100 consecutive switching cycles (positive SET and negative RESET processes) for (a) controlled and (b) annealed devices. Statistical distribution of SET- and RESET-voltages of (c) controlled and (d) annealed TAN/CeO\(_2\)/Al/CeO\(_2\)/Pt devices.

**Figure 3.** DC endurance characteristics of (a) controlled and (b) annealed TAN/CeO\(_2\)/Al/CeO\(_2\)/Pt devices. (c) The retention performance of annealed TAN/CeO\(_2\)/Al/CeO\(_2\)/Pt devices.
oxygen annealing treatment, which suggests a feasible way of optimizing the operational parameters of RRAM devices.

Figures 3(a), (b) reveal the endurance characteristics of controlled and annealed devices, resistance values of HRS and LRS are extracted at reading voltage of 0.2 V. Endurance characteristics were carried out for 100 consecutive DC switching cycles on both controlled and annealed devices. The controlled devices demonstrate severe fluctuations in HRS, as shown in figure 3(a), which may be due to unstable defects formed by inserting the Al layer in between the two CeO₂ layers. In contrast, a clear memory window with weak fluctuations in HRS is observed in annealed devices, as obvious from figure 3(b). Such a good cycle-to-cycle uniformity of the annealed device gives a clear evidence of the device stability, which may be beneficial from the improvement point of view due to reduction of formation energy of oxygen vacancies near the Al atoms. In short, configuration of conductive filaments appeared to be regular due to oxygen annealing as compared to controlled device. It is widely recognized that localized conductive filaments formed by oxygen vacancies or metallic ions play important role in the resistive switching characteristics. In the controlled device, randomly formed/distributed metallic filaments would lead to large fluctuations in switching parameters, such as distribution of SET/RESET voltage, HRS and LRS etc [30]. However, in the annealed device, oxygen annealing stimulates diffusion of Al³⁺ ions in the CeO₂ layer forming seeding (or nucleation) regions for monotonous formation and rupture of conductive filaments within switching layers, leading the conductive filaments to easily form and rupture near the locations of Al³⁺ ions, which could reduce the randomness of the conductive filaments formation/rupture processes and improve the switching parameters. To further verify the influence of Al³⁺ ions diffusion in annealed devices, time dependent retention characteristics were examined. The resistance values of both HRS and LRS were monitored up to 10⁴ s at room temperature. The readout of LRS and HRS resistances was performed after SET and RESET processes, respectively. Then, the resistance values were recorded after every 10 s with the read voltage of ±0.2 V. As shown in figure 3(c), no failure in the resistance values of HRS and LRS was observed even after 10⁴ s at room temperature. The improved endurance characteristics and stable retention properties are indicating the high feasibility of present annealed device as the resistive switching element in RRAM applications.
To investigate the electrical transport mechanisms of the resistive switching behaviors, the I-V curves for the typical switching cycles of the controlled and annealed devices are replotted in double logarithmic scale, as shown in figure 4. In the LRS of SET- and RESET- processes, log(I)-log(V) indicates a linear fit with slope values of ∼1.03, ∼1.06 for controlled devices, and ∼1.03 and 1.02 for annealed devices, respectively, indicating the Ohmic behavior which suggested well-arranged formation of conductive filaments due to electron conduction in LRS [31], as shown in figures 4(a)–(d). Nevertheless, the I-V curves in HRS could be divided into two parts as presented by the different slopes of the linear fitting, as shown in figures 4(a)–(d). In the low voltage regions of SET- and RESET-processes of controlled and annealed devices, the injected carrier density is lower than thermally generated carrier density and so conduction follows Ohm’s law (I ∝ V) [32] in the early stages of space-charge-limited conduction. With increasing the voltage bias, the injected carriers become dominant. When all of the traps (defect centers) are filled by injected carriers under the higher voltage, a sudden jump occurs in current and then I-V behavior follows the Child’s law [33], as shown in figures 4(a), (d). Therefore, for HRS in high field regions of SET- and RESET-processes, I-V linear characteristics have slopes of ∼2 (S ∝ V^2), which indicates that at high field regions current might be dominated with trap-controlled space-charge-limited conduction (SCLC) [34]. To further elucidate the conduction mechanism in high filed region, I-V characteristics can be related to either Schottky emission or Poole-Frenkel emission [35]. All plots in high field regions (controlled and annealed) illustrate linear relationships which follow Schottky equation as compared to Poole-Frenkel equation (results are not shown here). These analyses reveal that transport mechanism in the high field regions of HRS during SET- and RESET-processes operative dominantly is caused by Schottky emission, which might lead to a symmetrical switching.

Considering the existence of Al^{3+} ions and their diffusion in CeO_{2} films and migration of oxygen ions and oxygen vacancies, resistive switching behavior of the annealed and controlled devices can be demonstrated by the schematic energy band diagrams, as shown in figures 5 and 6, respectively. In this regard, negative electroforming can be associated with combined effect of field induced diffusion of the positively charged Al^{3+} ions and oxygen thermal annealing.
ions and negatively charged oxygen ions [36], which contribute in creation of optimum conducting channels. Figures 5(a), (b) and 6(a), (b) display the change in energy band diagrams during SET and RESET processes according to bias polarities and caused by strong/weak oxygen vacancy gradients. Whereas, figures 5(c), (d) and 6(c), (d) illustrate switching mechanisms for SET- and RESET-processes (bipolar switching). During SET process, when positive voltage is applied to TaN top electrode, oxygen ions could be released from the CeO$_2$/Pt bottom interface and are driven to Al:CeO$_2$ layer and then to the anode TaN. The endured oxygen vacancies firstly create conductive tips near the Pt cathode region. Then, from these tips conductive filaments begin to grow with increasing electric field and finally extend to TaN top electrode, which would result in larger area close to Pt bottom electrode and small bottleneck like area toward TaN top electrode, leading the devices to switch from HRS to LRS. Conversely, when negative voltage is applied to TaN top electrode, conductive filaments are ruptured near the thin region of active electrode, which corresponds to TaN/CeO$_2$ interface, leading the devices to switch from LRS to HRS (RESET process).

Smaller electroforming, SET/RESET-voltages and improved endurance characteristics of the annealed device are due to the fact that optimum conductive filaments are formed, leading to great enhancement/improvement in the switching parameters [37]. The optimum annealing temperature (400 °C) causes sufficient diffusion of Al$^{3+}$ ions within the switching layer (CeO$_2$) due to relatively lower ionic radii of Al as compared to that of Ce [38]. Furthermore, thermal conductivity of aluminum decreases by annealing, so the diffusion of Al$^{3+}$ ions within CeO$_2$ layer is enhanced, which appears to be responsible for strong improvement of RS behavior of the annealed device. Another reason could be associated to binding energy since binding energy of Ce-vacancy (0.65 eV) is smaller than that of Al-vacancy (0.80 eV) [38, 39]. This means that a strong interaction exists between Ce atoms and oxygen vacancies, and the binding energy is much larger than thermal energy, $k_BT$ ($k_B$ is Boltzmann constant and $T$ is absolute temperature). Therefore, possibility of Ce-vacancy decomposition caused by thermal fluctuations can be ignored and thus Ce and vacancies can form an inseparable complex in which vacancies move only around Ce atoms [38]. On the other hand, in the controlled device, the formation and
rupture of conductive filaments are highly random caused by inserted Al layer and unstable defects, which lead to wider dispersion in switching parameters and instability in endurance performance. Diffusion of Al$^{3+}$ ions through oxygen annealing assists in easy generation of conductive filaments responsible for improvement in the all key parameters of RRAM devices [40].

4. Conclusions

In summary, we have investigated the effect of oxygen annealing on the resistive switching characteristics of the TaN/CeO$_2$/Al/CeO$_2$/Pt devices. Both annealed and controlled devices exhibit the bipolar switching behavior. Due to lower density of oxygen vacancies through oxygen annealing, optimum and suitable ON mechanisms are Ohmic expected to contribute in the switching after annealing process. The governing electrical conduction mechanisms are Ohmic (LRS and low field regions of HRS), trap-controlled space charge limited conduction dominated by Schottky conduction (in high field regions of HRS).

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