Journal of Alloys and Compounds 759 (2018) 44–51

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

Structural engineering of tantalum oxide based memristor and its electrical switching responses using rapid thermal annealing

Yawar Abbas, Andrey Sergeevich Sokolov, Yu-Rim Jeon, Sohyeon Kim, Boncheol Ku, Changhwan Choi*

Division of Materials Science and Engineering, Hanyang University, Seoul 04763, Republic of Korea

ARTICLE INFO

Article history:
Received 18 January 2018
Accepted 8 May 2018

Keywords:
Rapid thermal annealing
Multilevel RESET
Analogue switching
Pulse-paired facilitation
Post-tetanic potentiation
Hermann Ebbinghaus forgetting curve

ABSTRACT

We have demonstrated the co-existence of reliable analog and digital switching characteristics with tantalum oxide based memristor by appropriate rapid thermal annealing (RTA). The device without RTA exhibits a digital SET and multilevel RESET for positive and negative sweeps, respectively. On the other hands, the device shows only analog switching characteristics such that current level increases and decreases gradually for successive positive and negative voltage sweeps, respectively, before any electroforming process with the RTA in the nitrogen ambient at the crystalline temperature of tantalum oxide which is 700 °C for 60 s. Once electroforming process is done, the device exhibits a reliable digital switching with SET at positive sweep and RESET at negative sweep voltages. In the analog state of the device we successfully emulate the synaptic characteristic of the device like spike-rate dependent plasticity (SRDP), pulse-paired facilitation (PPF) and post-tetanic potentiation (PTP). Finally, the Hermann Ebbinghaus forgetting curve is obtained from these devices. The conversion of the device from the digital SET and multilevel RESET to analogue switching is attributed to structural transition of amorphous tantalum oxide to polycrystalline tantalum oxide, different defect density and interface variation in the device.

1. Introduction

One of the promising candidates for the next generation memory devices is resistive random access memory (RRAM), due to its high speed, low power consumption, high memory density and high switching speed [1–4]. RRAMs are two terminal nanoionics systems [5–7] that fall within broader definition of memristors in which switching layer is sandwiched between two appropriate electrodes to form a capacitor structure. Resistive switching phenomenon can be classified into two types, one is digital [8,9] and the other one is analog type switching [10,11]. For digital switching, the device switches from high resistance state (HRS) to low resistance state (LRS) when a suitable voltage with or without a compliance current is applied on the top electrode of the device. The voltage at which device changes its resistance by the abrupt current increase is called SET voltage (V_{SET}) while opposite polarity when its state changes from LRS to HRS is called RESET voltage (V_{RESET}).

In the case of analog switching, the device conductance or current level increases continuously with respect to consecutive sweeps for one polarity and its conductance or current level decreases continuously for the consecutive opposite polarity voltage sweeps. This switching behavior significantly depends upon the micro and nano-structure in the thin film, type of electrodes, deposition process and many other processes. The co-existence of bipolar and unipolar switching in digital switching is previously reported in the literatures [12,13]. However, the co-existence of analog switching and digital bipolar switching characteristics are not reported that much [13,14]. Some reports in literature discussed the thickness dependent analog and bipolar switching and current compliance dependence [11,15] in the same materials. Among the materials being investigated, tantalum oxide and its variant are popularly used for RRAM device due to their reported highest endurance characteristic until now and low power consumption [16]. The analog switching observation from tantalum oxide based RRAM devices is studied by doping and diffusion limiting layer processes [17,18].

The multilevel resistance states of the device are also obtained

---

* Corresponding author.
E-mail address: cchoi@hanyang.ac.kr (C. Choi).

https://doi.org/10.1016/j.jallcom.2018.05.106

using different materials [19–23]. These multilevel resistance states are attained by using different compliance currents or voltages modulation. The voltage modulated multilevel resistance states can be extracted by widening the voltage sweep spans during the successive resistance state transition from ON-state to OFF-state. Such resistive switching behavior indicates that the amount of oxygen migration at the metal/insulator interface can affect the degree of the rupture of conducting filament. Hence, by increasing the voltage sweep spans continuously, the more oxygen migration can be driven from interface to re-oxidize the residual conducting filaments gradually, and finally the lowest current level of the OFF-state can be achieved. By using devices with such memory characteristics, one can obtain the high density memory devices. In order to use the RRAM device as an artificial neural network chip, it is inevitable to engineer RRAM device such that its I-V characteristics appear to be tunable continuously [24–26].

In this work we engineered a tantalum oxide based RRAM device exhibiting analog resistive switching and digital switching with rapid thermal annealing (RTA) process under appropriate temperature and annealing time. The co-existence of analog and digital bipolar switching characteristics is found after the series of experiments by focusing on the RTA of the devices. In the analog switching state, the device successfully emulated the brain functions like pulse paired facilitation (PPF), post-tetanic potentiation (PTP), Hermann Ebbinghaus curves and others. The appearance of co-existence of switching types with RTA indicates that the nanostructures of the thin film and interfaces are affected leading to a suitable and favorable environment for the co-existed analog and digital switching characteristics.

2. Experimental

The tantalum oxide thin films with 12 nm thickness were deposited on the two substrates of structure $Pt/Ti/SiO_2$ by RF magnetron sputtering. The base pressure in the sputter chamber was 4×10^{-6} Torr and target was sputtered for 5 min in an argon atmosphere. The temperature of the substrate was 25 °C. The working pressure and RF power were maintained at 4 mTorr and 50 W, respectively. After the TaO_x thing films formation on the $Pt/Ti/SiO_2$ substrates, the UV-photolithography process was performed on the two samples to define $100\ \mu m \times 100\ \mu m$ patterns on the top of tantalum oxide thing film. The following was to deposit 70 nm-thick Ti as a top electrode on these patterned samples using DC magnetron sputtering for 20 min and 100 W DC power at room temperature. Finally, lift-off process was done using acetone for realizing the final $Ti/Ta_2O_5/Pt/Ti/SiO_2$ device structure. After device fabrication, one sample received the RTA annealing in the N_2 ambient at 700 °C for 60 s for modulating material properties while the other one was kept as pristine. In order to investigate the structural changes of the thin films after the RTA process in N_2 ambient, both as-deposited and RTA-processed thin films were examined using X-ray diffraction (XRD) by high resolution (HR) XRD of SmartLab having Cu source of 1.5406 A^o wave length. In addition, both devices were analyzed using high resolution transmission electron microscope (HRTEM) of JEOL 2100-F to know the thin film thicknesses, crystalline nature and elemental information. The TEM samples were prepared using focus ion beam (FIB) of FI system. The high energy gallium (Ga+) ions were used in order to etch out the lamellae from the device and mounted on the copper lift-out TEM grid to examine the structural nature of devices in the HRTEM chamber. The electrical data (including quasi-static and transient data) of the devices were analyzed with Keithley 4200 SCS semiconductor analyzer. During all electrical measurements bottom electrode Pt was grounded and transient and DC stimulations were applied on the Ti top electrode.

3. Results and discussions

Fig. 1 shows the XRD patterns of pristine and annealed-tantalum oxide thin films. Fig. 1 (a) is the XRD pattern of as-deposited tantalum oxide thin film with only one peak at $2\theta = 67.75°$ related to Pt (220) peak. Fig. 1 (b) exhibits the XRD pattern of tantalum oxide thin film annealed at 700 °C known as a most favorable crystalline temperature of tantalum oxide [27]. Besides the peak at $2\theta = 67.75°$ related with Pt (220), additional prominent peaks are observed at $2\theta = 46.6°$ and 40° degree, which are corresponded as Ta_2O_5 (002) and Ta_2O_5 (111), respectively [28,29]. Hence, RTA process on the tantalum oxide vividly leads to the transition of tantalum oxide thin film from amorphous to crystalline structure.

Fig. 2 shows the Half Angular Annular Dark Field (HAADF) Scanning TEM (STEM) images and z-contrast magnified TEM images of pristine and annealed devises. Fig. 2 (a) and (c) show the HAADF-STEM images of the pristine and annealed devices depicting the thicknesses of Ti, Ta_2O_5 and Pt as around ~70 nm, ~12 nm and ~70 nm, respectively. The HRTEM images of pristine and annealed devices from Fig. 2 (b) and (d), respectively, including the Fast Fourier Transform (FFT) of selected regions indicate that the poly-crystalline regions with different grain boundaries are observed in Ta_2O_5 region and at the Ti/Ta_2O_5 interfaces. The FFT of Ta_2O_5 regions is also with a close argument for the poly-crystallinity of thin film. In the light of TEM analysis one can argue that the annealing of device in N_2 ambient changed the amorphous nature of Ta_2O_5 thin film and Ti/Ta_2O_5 interface into ploy-crystalline nature. From the above XRD and HRTEM analysis, we can elucidate that the process of RTA in N_2 ambient does not only change the amorphous nature of as-deposited tantalum oxide thin film to poly-crystalline but also contribute to form the thicker and poly-crystalline TiO_x at the Ti/Ta_2O_5 interface. These poly-crystalline TiO_x at the interface and poly-crystalline Ta_2O_5 switching media along with their grain boundaries can offer a favorable environment for oxygen vacancies (V_{ox}) to accumulate at grain boundaries and transport much faster along these grain boundaries [30,31].

Fig. 3 shows the resistive switching characteristics of the pristine device. After the process of electro-forming at the sweep voltage of +3 V with compliance current of 5 mA, the device appeared to be SET in digital manner whereas for the RESET process we observed multiple resistance states when the negative voltages were swept incrementally. With the slight increase in magnitude of negative sweeps the multilevel resistance state can be generated in the pristine $Ti/Ta_2O_5/Pt$ device. These types of switching are also useful in the neuromorphic device application [25,32]. However, since our main focus is to explore the switching characteristics of the RTA processed device having analog and digital switching, we will discuss the electrical characteristics of the RTA processed devices further. The RTA processed device was investigated to extract the synaptic behaviors by applying suitable DC voltages and transient pulses. Following are the results and discussions which are extracted from the RTA annealed device which are divided into two parts, one is analog switching and another is digital switching.

Fig. 4 shows the typical analog type I-V characteristics of the RTA processed device. The DC sweeps were applied carefully to avoid any digital switching which is explained in the upcoming discussion. Fig. 4 (a) shows that the electric current levels increase gradually after every sweep when 8 positive consecutive sweeps of +2.5 V were applied. For the next 8 consecutive sweeps of −2.5 V the current level gradually decreases after each sweep cycles. The inset shows gradual decrease of the current with each consecutive negative sweep more clearly. The same trend of gradual increase and decrease in the conductance were observed when the