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Bipolar resistive switching memory using bilayer TaO_x/WO_x films

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ABSTRACT

Resistive switching properties of a memory device in an $IrO_x/TaO_x/WO_x/W$ structure have been investigated. High-resolution transmission electron microscopy image has shown the formation of a bilayer structure of TaO_x/WO_x which is further confirmed by energy dispersive X-ray spectroscopy and X-ray photo-electron spectroscopy analyses. The underlying switching mechanism is successfully explained by providing various electrical measurements such as device area dependency on set/reset voltage and low resistance state. A model based on oxygen ions migration is then proposed. Cumulative probability plots of essential memory parameters such as set/reset voltage and LRS/HRS show good distribution. The device has shown excellent read endurance of $>10^5$ times and data retention of $>10^4$ s with a resistance ratio of $>10^2$ at 85 °C.

1. Introduction

Being a powerful alternative to present conventional nonvolatile memory technologies, resistive memory (RRAM) devices have attracted considerable interest [1,2]. In such devices data bits can be stored as resistance making these devices easily scalable, faster and stackable. Other incentives of RRAM include its simple metal–insulator–metal (MIM) structure, compatibility with standard complementary metal–oxide–semiconductor (CMOS) process and lower power consumption. Recently, transition metal oxides especially binary oxides have been proposed for resistive switching memories. Among other various oxides such as NiO [3–5], TiO_x [6], HfO_x [7–10], Cr_2O_3 [11], Sm_2O_3 [12], CuO_x [13], ZrO_2 [14,15], AlO_x [16–18], WO_x [19], and so on, tantalum oxide (TaO_x) based devices are becoming appealing due to its high dielectric constant ($\varepsilon_r \sim 25$) and CMOS compatibility. However, the resistive switching characteristics using TaO_x material has been reported infrequently [20,21]. On the other hand, tungsten oxide (WO_x) is another promising material reported for RRAM applications due to its ease of fabrication and CMOS compatibility [19]. Although most of the reported literature on RRAM devices centered on single resistive switching layer, bilayer structures are advocated to improve the resistive switching performance [8,12,18].

In this study, we design a bilayer structure comprise of TaO_x and WO_x in an $IrO_x/TaO_x/WO_x/W$ RRAM stack keeping in mind the Gibbs free energies of formation of Ta_2O_5, WO_3 and WO_2 films which are −760, −529 and −506 kJ/mol at 300 K, respectively [22]. From Gibbs free energy values, Ta_2O_5 is energetically favorable and can take oxygen from both WO_3 and WO_2 making this layer oxygen deficient (WO_x). Consequently, WO_x layer will behave as conducting layer with abundance of oxygen vacancies (V_o) while TaO_x will be comparatively insulating. The resulting resistive memory device in an $IrO_x/TaO_x/WO_x/W$ structure has been confirmed by high-resolution transmission electron microscopy (HRTEM) and X-ray photo-electron spectroscopy (XPS) analyses. The RRAM device has shown good memory characteristics with small set/reset voltages of −2.1 V and 1.3 V, respectively. The statistical analysis of the essential memory parameters such as set/reset voltage, low resistance state (LRS)/high resistance state (HRS) has also been presented. The switching mechanism is successfully explained by conducting filament model which is in well agreement with various electrical characteristics. The device has shown excellent read endurance of $>10^5$ times and good data retention of $\geqslant 10^4$ s with a high resistance ratio of $>10^2$ at 85 °C.

2. Experimental

In this study, a resistive switching memory device in an $IrO_x/TaO_x/WO_x/W$ structure has been fabricated. A tungsten (W) metal layer of thickness 100 nm as a bottom electrode (BE) was deposited by rf magnetron sputter system on 8" SiO_2 (200 nm)/Si wafer. Then, a low temperature SiO_2 (LTO) layer with a thickness of 150 nm was deposited on W BE. Different via sizes ranging from 0.4 × 0.4 to 8 × 8 μm^2 were then defined by lithography and etching methods followed by another lithography step to pattern the

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device for lift-off. Next, a high-κ switching material layer of tantalum pentoxide (Ta_2O_5) with a thickness of approximately 10 nm was deposited by electron-gun evaporator using pure Ta_2O_5 granules. In order to deposit iridium oxide (IrO_x) as a top electrode (TE), IrO_x layer with a thickness of approximately 220 nm was deposited by reactive sputtering using Ir target (>99.999%). The ratio of Ar and O_2 mixture was 1:1 and working pressure was maintained at 20 mTorr. The rf power used during deposition was 50 W. Finally, lift-off process was performed to get the final device. The oxygen gas used during IrO_x deposition diffused through thin Ta_2O_5 film and oxidized tungsten surface of BE resulting a thin WO_x layer with a thickness of approximately 5 nm. This WO_x layer along with tantalum oxide layer forms bilayer. To know the film thickness and interface among deposited stack layers, HRTEM image was acquired using FEI Tecnai G2 F-20 field emission system with an operating voltage of 200 kV. The memory device for TEM observation was prepared using an FEI Helios-400s system with an operating voltage of 5 kV and Ga^+ ion source was used to thin down the device. The TEM image was acquired within a short time of <5 s. The elemental presence of deposited layers was investigated by energy dispersive X-ray spectroscopy (EDX). Material compositions were studied by XPS using Al Kα monochrome X-ray with energy of 1486.6 eV. The vacuum in analytical chamber was 1×10^{-9} Torr and a spot size of 650 μm in diameter was used to analyze the materials. In order to analyze W layer, the TaO_x layer with a thickness of approximately 9 nm was etched out using 3 keV Ar ion sputtering in 100 s. All the acquired spectra were calibrated by C1s peak at energy of 284.6 eV. Electrical measurements were performed using HP4156C precision semiconductor parameter analyzer and bias was applied on TE while BE was grounded.

3. Results and discussion

Fig. 1a shows typical cross-sectional TEM image of fabricated resistive switching memory device. The memory device size is approximately $2 \times 2\ \mu m^2$. Fig. 1b shows the HRTEM image of the selected area in Fig. 1a where stack layers and their interfaces are clearly visible. The thicknesses of the TaO_x and WO_x layers are approximately 9 and 5 nm, respectively. The high-κ TaO_x film is amorphous while the WO_x is polycrystalline due to the presence of nano-grains. The presence of oxygen (O) peak at energy of approximately 0.52 keV in EDX spectra shown in Fig. 1c indicates the formation of tantalum oxide and tungsten oxide layers. The composition of both the layers is confirmed by the XPS analysis. The XPS spectra of TaO_x and WO_x layers are shown in Fig. 2a and b respectively. The peaks of Ta_2O_5 4f doublet with peak binding energies of 26.70 eV ($Ta_2O_54f_{7/2}$) and 28.60 eV ($Ta_2O_54f_{5/2}$) with peak separation of 1.9 eV are observed and are in well agreement with reported values [23]. Additionally, peaks corresponding to metallic tantalum (Ta) with peak binding energies of 21.77 eV ($Ta4f_{7/2}$) and 23.74 eV ($Ta4f_{5/2}$) [24] are also observed (Fig. 2a). The peaks of Ta_2O_5 along with metallic Ta ensure the sub-stoichiometric tantalum oxide (TaO_x) film. The peaks corresponding to WO_3 with peak binding energies of 36.21 eV ($WO_34f_{7/2}$) and 38.23 eV ($WO_34f_{5/2}$) in addition to peaks corresponding to metallic W 4f doublet with peak binding energies of 31.63 eV ($W4f_{7/2}$) and 33.88 eV ($W4f_{5/2}$) are observed in the XPS spectra of WO_x layer (Fig. 2b) which confirm the formation of tungsten oxide (WO_x). These values of binding energies of W4f for W^0 in metallic tungsten and for W^{+6} in WO_3 are in well agreement with the published literature [25]. Due to the bilayer structure of TaO_x/WO_x films, good

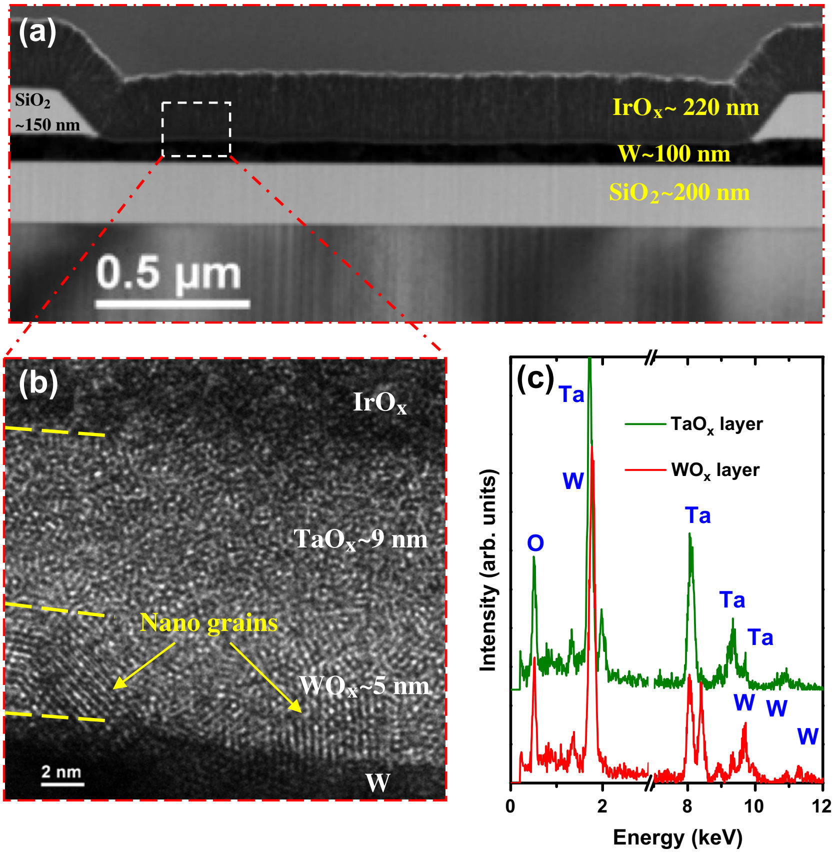

Fig. 1. (a) Cross-sectional TEM image of resistive memory stack in $IrO_x/TaO_x/WO_x/W$ structure. The effective device area is $2 \times 2\ \mu m^2$. (b) HRTEM image of memory stack showing bilayer structure. The thicknesses of TaO_x and WO_x layers is approximately 9 and 5 nm, respectively. (c) EDX spectra of TaO_x and WO_x layers.