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Resistive switching characteristics in manganese oxide and tantalum oxide devices

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ABSTRACT

The monodisperse manganese oxide nanoparticles with an average diameter of 30 nm were chemically synthesized. The nanoparticles assembled as a close-packed monolayer on Pt bottom electrode by dip-coating and annealing process. The Ta/Ta_2O_5/MnO/Pt device was fabricated. The bipolar resistive switching behaviors could be caused by the formation and rupture of conductive filaments in the switching layers. The stable self-compliance property was demonstrated which can be attributed to the high resistance Ta/Ta_2O_5 interface, the Schottky barrier of Ta/Ta_2O_5, and the discontinuity of conduction band at Ta_2O_5 and MnO interface. The retention characteristics of Ta/Ta_2O_5/MnO/Pt device were investigated. The conduction mechanisms of Ohmic conduction, space charge limited conduction, Schottky conduction and Poole–Frenkel emission had been investigated for resistance switching mechanism.

1. Introduction

Resistive switching random access memory (RRAM) has been widely investigated as one of the most promising candidates for next-generation nonvolatile memory applications due to its simple structure, high scalability, high speed and low power consumption, and high packing density [1–8]. A RRAM cell is usually composed of a resistance switching layer sandwiched by two electrodes. In general, both organic and inorganic materials are available for the resistance switching layer. Organic materials have been demonstrated to have significant advantages in flexibility, simple fabrication, and low cost, while inorganic materials can be prepared by a variety of fabrication methods and exhibit excellent switching stability [1]. As one group of the inorganic materials, the binary transition metal oxides, such as NiO [2,9,10], HfO_2 [8,11,12], Ta_2O_5 [3,4,13,14], TiO_2 [5,15,16], AlOx [6], ZrO_2 [7,17], Fe_2O_3 [18], and MnO [19,20], have attracted significant attention for RRAM applications owing to their simple fabrication process [21]. Long et al. reported the characterization and modeling of the set and reset statistical distributions of RRAM devices [10–12]. The reset switching statistics in NiO-based RRAM devices were ascribed to thermal dissolution of conductive filament [10]. In our previous research, the stable and reliable bipolar resistive switching behaviors of manganese oxide nanoparticles have been reported [20]. The resistance switching can be easily affected by the nanoscale redistribution of defects in the nanoparticle assembly. Therefore, using nanoparticles as resistance switching elements can give an appropriate control in the RRAM devices [20]. Ta_2O_5 became a potential material for RRAM application due to its simple composition, reliable properties and chemical stability [3,4,13,14]. Recent researches on RRAM devices with oxide heterostructures suggest that superior performances over single-layer based devices could be obtained. The cycling stability can be improved. Non-linear switching, complementary resistive switching and self-compliance can be achieved. In addition, the oxide heterostructure can modulate each layer and interface on the switching properties [22–26]. Self-compliance can both lower the power consumption and improve the scalability for an array structure [27,28]. However, in the previous researches, most studies focused on single-layer based devices rather than heterostructure based ones.

In this study, Ta_2O_5/MnO heterostructures with Ta top electrode and Pt bottom electrode had been fabricated. The bipolar resistive switching characteristics of the device were investigated. The repeatable self-compliance property was observed in Ta/Ta_2O_5/MnO/Pt device.

2. Experimental

The manganese oxide nanoparticles, stabilized by a surfactant of oleic acid, were chemically synthesized by thermal decomposition of manganese acetate at high temperature [29]. Oleic acid acted as a surfactant to prevent the aggregation of the nanoparticles. Because the hydrophobic tail of oleic acid was faced with the solvent, the nanoparticles were well dispersed in the nonpolar hexane solvent. The size of the nanoparticles was characterized using transmission electron microscopy (TEM, JEM2100F).

The manganese oxide nanoparticles with monolayer were assembled on the Pt (100 nm)/Ti (10 nm)/SiO_2/Si bottom electrode by dip-

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coating and annealing procedures. Ta_2O_5 film was prepared by radio frequency sputtering with Ta_2O_5 target (99.9%) at room temperature. The top electrode of Ta was deposited at room temperature by magnetron sputtering by a round-shaped shadow mask with the diameter of 400 μm.

The nanoparticle layer on the Pt electrode surface was analyzed with scanning electron microscopy (SEM, Hitachi S-4800). Current–Voltage (I–V) characteristics of the fabricated devices were measured by an Agilent 4156B semiconductor parameter analyzer by sweeping the voltage at Ta electrode as bottom Pt electrode grounded.

3. Results and discussion

Fig. 1 showed the bright-field image TEM micrograph of the manganese oxide nanoparticles. Nanoparticles with an average diameter of 30 nm were chemically synthesized. Fig. 2(a) showed the plan-view SEM micrograph of manganese oxide nanoparticles on Pt bottom electrode. The thickness of the nanoparticle assembly was ~30 nm. The nanoparticles were assembled as a monolayer on Pt bottom electrode by dip-coating process. According to the previous research, the adsorption of MnO nanoparticles was thought to be driven by van der Waals interaction between nanoparticle and bottom electrode and the interaction energy was calculated to be ~80.9 kT, which was much higher than thermal fluctuation at room temperature [20]. Therefore, the MnO nanoparticles could adsorb on the surface of Pt electrode and closely assembled as a monolayer. Fig. 2(b) exhibited the cross-sectional view SEM micrograph of Ta_2O_5/MnO on Pt electrode. The thickness of the oxide layers was ~60 nm.

The I–V properties of Ta/Ta_2O_5/MnO/Pt device were measured and shown in Fig. 3. The device exhibited bipolar resistive switching characteristics. Positive voltage set the device to LRS and negative voltage reset it to HRS. For Ta/Ta_2O_5/MnO/Pt device, the voltage was applied from 0 V to + 3 V without any external compliance current to decrease the resistance from high resistance state (HRS) to low resistance state (LRS) and no hard breakdown was seen. After more than 300 repeated switching cycles, repeatable self-compliance was observed and it indicated the good reproducible and reliable behaviors of Ta/Ta_2O_5/MnO/Pt device.

To further understand the reliability of the device, the switching cycling was examined. A stable switching property with the switching cycle up to 300 cycles was observed. Fig. 4 showed the resistance of HRS and LRS *versus* the number of switching cycles at the reading voltage of + 1.0 V. The average resistance ratio of HRS to LRS was ~21. The average current density at HRS and LRS were 29.7 pA/μm^2 and 610 pA/μm^2, respectively.

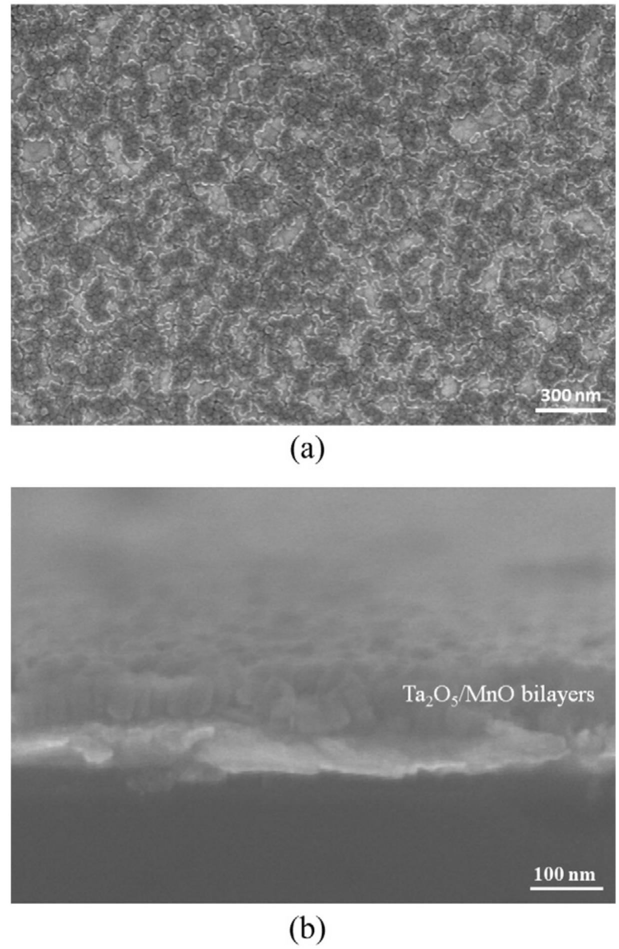

Fig. 2. (a) The plan-view SEM micrograph of MnO nanoparticle assembly on Pt electrode. (b) Cross-sectional view SEM micrograph of Ta_2O_5/MnO on Pt electrode.

According to the previous study, the resistive switching characteristics of the manganese oxide nanoparticle device were attributed to the formation and rupture of the conductive filaments in the nanoparticle assembly [20]. When a positive voltage was applied, the oxygen ions migrated to the interface between top electrode and nanoparticle

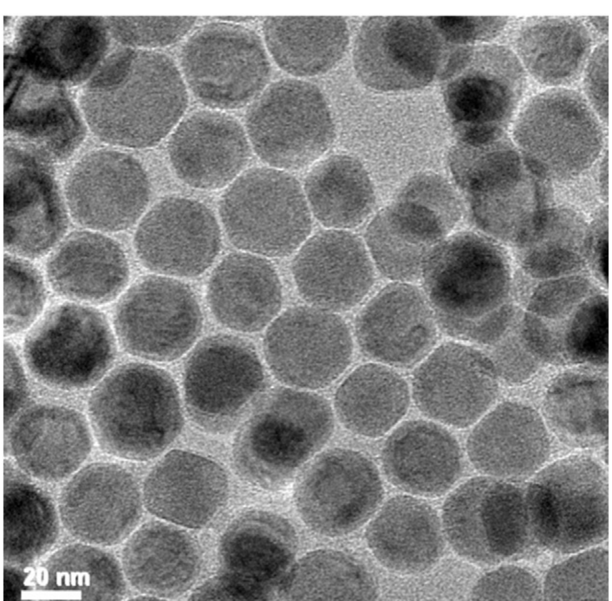

Fig. 1. Transmission electron micrograph of manganese oxide nanoparticles.

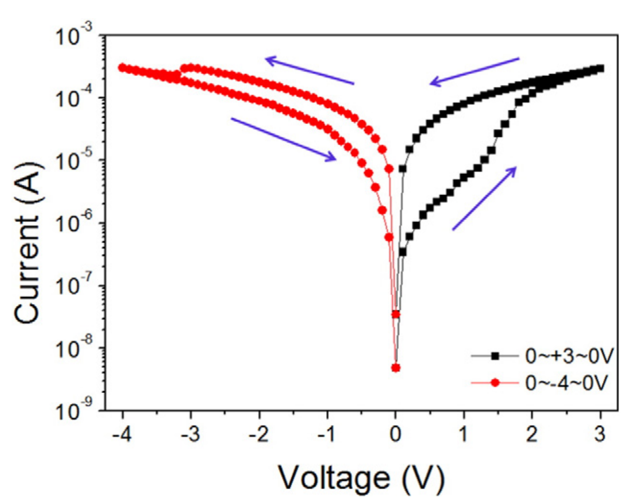

Fig. 3. I–V curves of Ta/Ta_2O_5/MnO/Pt device.