



Resistive switching characteristics of NiO/Ni nanostructure

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ARTICLE INFO

Article history:

Available online 25 July 2012

Keywords:

Resistive switching
ReRAM
Nanowire

ABSTRACT

We have studied variation of switching voltage of resistive random access memory (ReRAM). Basically, the switching voltage at every switching has a different value because there are many conduction paths, which are randomly chosen at every switching with different conductivity in an oxide insulator between top and bottom electrodes. Limitation of the number of conductive paths is expected to lead the suppression of the variation of switching voltage. In this study, Ni nanorods buried into the nanoholes of an anodic aluminum oxide (AAO) film have been fabricated to restrict the formation of many filaments and the switching behaviors of the In/NiO/Ni nanorods has been investigated.

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1. Introduction

Resistive switching random access memory (ReRAM) is one of storage device with interesting reversible switching phenomena between two different resistance states (low-resistance state:OFF and high-resistance state:ON). These phenomena are observed in many kinds of transition metal oxide thin film, such as Al₂O₃ [1], NiO [2–5], and TiO₂ [6]. The ReRAM has great advantages, for example non-volatile, high-speed response [7], and low-power driving [8]. From these reason, in the case of ReRAM consisted of NiO materials, many kinds of devices have been extensively studied, such as a dependence of electrode sizes and types [2,3], IrO₂ doped NiO film [4], and NiO nanowire [5].

These curious phenomena have been understood in terms of a formation and rupture of numerous nanofilaments as conduction passes formed at a soft breakdown of an insulator [9]. When switching, several filaments with different conductivity and different length are randomly chosen from many choices. This must have been leading the poor reproducibility of switching voltages. For the practical application, it is required that the devices have the same switching voltages at every switching. One of solutions for suppression of the variation switching voltage is a restriction of filament formation. If the restriction is possible, extreme improvements are expected in the reproducibility. However, the variation of the SET and RESET voltages seem not to depend on electrode size in the region of 5×10^{-2} – $10^3 \mu\text{m}^2$ [2], because the electrode size was much larger than the filament size. It have been reported that the filament diameter is several nanometer [10].

Namely, fabrication of oxide thin film with several tens nanometer in diameter is thought to be suitable to restrict the different formation of the filament path. A nanorod which was prepared by electroplating to an anodic aluminum oxide (AAO) is an optimal material as the bottom electrode for the restriction [11], where AAO consisted of a barrier layer and a porous layer was obtained by anodization of Al film [12].

There are some reports about ReRAM device using a nanowire as building blocks already [5]. However, most of the previous studies employed thermal oxidation in forming oxide in the metallic nanowires. Such a high temperature process would cause metal diffusion problems into adjacent semiconductor materials.

In this study, Ni nanorod has been prepared by an electroplating method and then the surface of Ni nanorod is oxidized by oxygen plasma treatment for growth of oxide thin film. The current–voltage (*I*–*V*) characteristics of In/NiO/Ni nanorod capacitor have been investigated. The device using usual NiO/Ni film and the oxide also has been fabricated and studied for comparison of the restriction effects.

2. Experimental

Fig. 1(i)–(vi) show a schematic procedure of preparation of Ni nanorod ReRAM device. The AAO templates were prepared on Si substrates to fabricate Ni nanorod arrays. 10 nm-Ti, 40 nm-Au and 500 nm-Al films were successively sputtered on H-terminated *n*-Si (100) substrates which cut into $30 \times 30 \text{ mm}^2$. Usually, an amorphous alumina layer called the barrier layer exists at the bottom of AAO pores on the Si substrate, which hinders homogeneous electrodeposition in each pore. The barrier layer can be removed by

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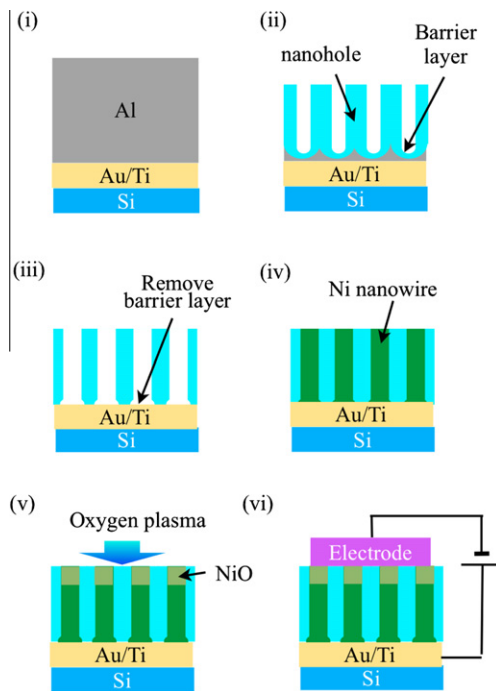


Fig. 1. Schematic procedure of preparation of NiO/Ni nanorod ReRAM device from (i)–(vi).

wet chemical thinning process of aluminum oxide after completion of anodization of Al [13]. Moreover, the Au functions as the protective layer for oxidation of the Si substrate during anodization. The Ti layer was introduced to improve adhesion strength between the Au film and Si substrate.

The Al films on the Si substrates were anodized using single step anodization method in 0.3 M oxalic acid under constant voltage conditions of 40 V. Then, the AAO nanopore array with the diameter of about 30 nm and the density of 80 G pores/inch² was obtained [14]. During anodization, the electrical contact to the Al film was formed from the back surface of the Si substrate, which was tightly contacted with a Cu anode plate. The anodization was stopped when the AAO pores reached Au surface. The temperature during anodization process was controlled at 5 °C. Subsequently, the AAO films were slightly etched in 5 wt.% phosphoric acid solution to remove the amorphous alumina barrier layer that existed at the bottom of the nanohole. At this process, the pores were widened from 30 to 70 nm. Although the diameter of the pores was increased, we were able to obtain the AAO templates without the barrier layer on the Au/Ti/Si substrates.

Ni nanorod arrays were formed in the AAO templates by pulsed electroplating technique (voltage = 3 V, total time = 5 min, pulse width = 10 ms, interval = 990 ms) at 53 °C [11], where the electrolytes were a mixture of nickel sulfate, nickel chloride, and boric acid, and the bottom Au layer acted as working electrode during Ni deposition. The Ni electroplating was continued until complete filling of AAO pore with Ni. On the other hand, Ni film with thickness of 50 nm was prepared by DC sputter for comparison of morphological dependence of ReRAM. Both the Ni films and the nanorod samples were treated with 200 W oxygen RF plasma for 10 min at room temperature. The thickness of NiO layer is about 10 nm. Subsequently, the top indium electrodes of diameter of about 300 μm are attached on NiO nanorods. The surface morphology of the both sample was evaluated by scanning electron microscopy (SEM). The *I*–*V* characteristics were examined by a conventional two-probe method at room temperature, where the current compliance of the Ni nanorod and Ni film samples limited

to 100 and 10 mA to guard the device from complete dielectric breakdown, respectively. Compliance current limit depends on the electrical conductivity of capacitors. In this study, the contact area is the almost the same in both devices but effective area of the device using usual Ni film is much larger than that of the device with nanorod. Large effective area requires relatively high current limit. A monopolar operation was adopted instead of an ordinary bipolar operation.

3. Results and discussion

Fig. 2 shows the SEM image of the cross-section of the Ni nanorod in AAO templates. Ni metal is found in the nanoholes with 70 nm in diameter, 500 nm in length. Fig. 3 shows SEM image of the cross-section of Ni film sample. The thickness of Ni film is about 90 nm. The oxide layer formed by oxygen plasma was too thin to observe by SEM.

Fig. 4 shows *I*–*V* characteristics of the nanorod sample. The sample clearly exhibits switching phenomena and *I*–*V* hysteresis loops with large “window” in the set (a) and reset (b) processes. During set operation, the current compliance limit is required to prevent permanent breakdown of oxide insulating layers. A maximum and a minimum of the set region are 1.42 V and 0.84 V, respectively. On the other hand, the reset behavior occurs between 0.80 and 0.34 V.

Fig. 5 shows *I*–*V* characteristics of Ni film sample. The sample shows the switching phenomena with the large variability of switching voltage (set region = from 4.3 to 13.2 V, reset region = from 2.3 to 4 V). Switching voltages of the nanorod sample distribute at the narrow voltage region owing to the restriction of the filament formation by nanostructure. The variability of switching voltages of the nanorod sample clearly is suppressed, comparing with that of the Ni film sample.

Fig. 6 shows cycle number dependence of switching voltages. As shown in Fig. 6(a), the sample using NiO/Ni nanorod shows one hundred stable switching behaviors. Meanwhile, after twenty-two times cycles, the Ni film sample broke, as illustrated in

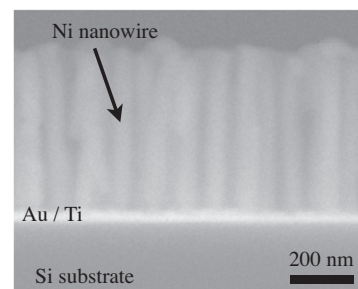


Fig. 2. SEM image of cross-section of the nanorod sample.

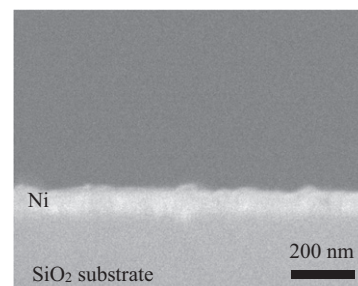


Fig. 3. SEM image of cross-section of Ni film sample.

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