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Effect of microwave irradiation power on resistive switching performance in solution-processed aluminum oxide resistive memory

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ABSTRACT

In this study, we fabricated ReRAM devices with the structure Ti/AlO_x/Pt by applying solution-processed AlO_x film as the switching layer. This enabled us to analyze the effect of the MWI power on the resistive switching performance of ReRAM. The AlO_x resistive switching layer deposited by the solution-process was subjected to PDA treatment with microwave power ranging from 600 W to 3000 W, and the resistive switching performance was compared with as-dep and CTA-processed ReRAMs. All AlO_x-based ReRAM devices exhibited bipolar resistive switching characteristics, and MWI-treated devices had larger memory windows than as-dep and CTA-treated devices. These solution-processed AlO_x ReRAMs were found to exhibit Ohmic conduction in the low-voltage range of both the LRS and HRS. The high-voltage range of HRS shows the Poole-Frenkel conduction mechanism. In addition, compared with the as-dep device, the PDA-treated devices exhibited stable endurance characteristics and uniform resistance distribution in 1000 cycles of the switching operation, and showed reliable retention characteristics for 10,000 s at both room temperature and high temperature. XPS measurements were performed to analyze the relationship between the resistive switching performance and chemical nature of the AlO_x switching layer by varying the microwave power and heat treatment method.

1. Introduction

In recent years, nonvolatile memory (NVM) has been widely applied in electronic devices such as cell phones, MP3 players, and laptops. NVM requires high-performance memory characteristics such as low-energy operation, fast write and read access, and high-density data storage [1,2]. However, typical flash memory has disadvantages such as high operating voltage, low endurance, and low write speed, and it is expected to run into physical limits in the near future owing to the demand for high-density storage [3]. Therefore, various studies on next-generation NVMs, such as phase-change random access memory (PRAM), magnetic random access memory (MRAM), and resistive random access memory (ReRAM) are being carried out actively [4–6]. In particular, ReRAM is emerging as a promising candidate for universal memory because of its low operating voltage, fast switching speed, high-density storage ability, and excellent scalability [7–10]. Various metal-oxide materials, such as ZnO_x, HfO_x, TaO_x, CuO_x, TiO_x, and AlO_x, have been applied to the active layer in ReRAM [11–16]. Among these metal-oxide materials, since AlO_x has advantages such as a low cost, stable resistive switching (RS) characteristic, wide bandgap, and good thermal and mechanical stability, it is attracting much attention as a promising material for nonvolatile ReRAM applications [17,18]. In general, sputtering, chemical vapor deposition (CVD), or atomic layer deposition (ALD) are widely used to form a metal-oxide layer of ReRAMs with excellent RS characteristics [19,20]. However, these methods require expensive vacuum equipment, have a long processing time, and require a high deposition temperature. In this sense, the chemical solution deposition method has been attracting the attention of researchers studying the ReRAM process because this process does not require expensive vacuum equipment; thus, initial investments and maintenance costs can be kept at a low level. In addition, the non-vacuum technology is simple, the process is fast and can be carried out at a low temperature, and it can be applied to large-area devices [21–24]. However, the lack of a high-purity vacuum environment during film deposition has to be compensated for by careful heat treatment to avoid undesired defects and contamination. The quality of the solution-derived film is generally improved in subsequent processing steps, where the material is annealed at higher temperatures. Commonly used heat treatment methods, such as conventional thermal annealing (CTA) using a furnace, has the disadvantage of a long heat treatment time and high temperature. Instead, microwave irradiation (MWI), which is a low-temperature heat treatment method with a short heat treatment time, excellent thermal uniformity, and good energy transfer efficiency, is attracting much attention [25,26]. Therefore,

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MWI technology is extensively used to improve electrical characteristic of devices in various fields where the solution process is more widely used such as oxide semiconductor thin-film transistors (TFTs), solar cells and light emitting diodes (LEDs) due to many advantages of microwave [27–29].

Therefore, in this study, we investigated the effect of microwave power on the resistive switching performance of ReRAM with a solution-processed AlO_x switching layer. Furthermore, we compared the electrical properties of as-dep and CTA treated ReRAM devices and confirmed the superiority of the microwave assisted post-deposition annealing (PDA) process.

2. Experimental method

As a starting material, (100) p-type silicon wafers with resistivity of 1–10 Ω cm were cleaned by using standard RCA cleaning and then a 300-nm-thick SiO_2 layer was grown by the thermal oxidation method. Ti and Pt layers were sequentially deposited as low electrodes using an e-beam evaporator at a thickness of 10 nm and 100 nm, respectively. The switching layer of ReRAM was prepared by spin coating 0.4 mol of a solution of AlO_x (Kojundo Chemical, Al-04) at 6000 rpm for 30 s. Subsequently, the samples were baked at 180 °C for 10 min to remove the solvent and impurities in the AlO_x, and MWI was performed at 2.45 GHz for 2 min in the range of 600 W–3000 W. On the other hand, for comparison, CTA was performed under an atmosphere of N_2 gas at 400 °C for 30 min. Table 1 shows the average temperatures of the silicon wafer measured from 600 W to 3000 W using an infrared thermometer (Gemini, P4.0 MTL LC2) to determine the actual temperature for the heat treatment conditions of the MWI. Finally, to form the top electrode, a Ti electrode with a diameter of 200 µm was deposited to a thickness of 100 nm using a shadow mask in an e-beam evaporator. The thickness of AlO_x film according to annealing conditions was measured using a TENCOR (Alpha-Step 500) surface profilometer. Also, the dielectric constant was measured using MIM capacitor configurations and Agilent precision 4284A LCR meter. The resistive switching (RS) characteristics and permittivity were evaluated by measuring the fabricated ReRAM devices in a dark box using an Agilent 4156B Precision Semiconductor Parameter Analyzer to avoid the effects of light and the external environment. In addition, an X-ray photoelectron spectroscopy (XPS) analysis was performed to confirm the chemical bonding state of the as-dep, CTA, and MWI treated solution AlO_x films. Fig. 1 shows the schematic structure of the solution-processed AlO_x ReRAM device fabricated in this study.

3. Results and discussion

Table 2 shows the thickness and permittivity of solution-processed AlO_x films due to heat treatment. It is found that when the solution-deposited AlO_x film is annealed, the thickness is reduced and the permittivity is increased. These changes are attributed to the densification and quality improvement of films as the solvent is removed by heat treatment.

Fig. 2 shows the bipolar resistive switching (BRS) characteristics (a) and on/off ratio of solution-processed AlO_x ReRAM devices (b) as a function of the MWI power. The as-dep and CTA results are also shown for comparison. The BRS characteristics were measured by applying

Table 1
Average temperatures of silicon wafer measured using an infrared thermometer.

	600 W/2′	1200 W/2′	1800 W/2′	2400 W/2′	3000 W/2′
Average temperature [°C]	347.09	413.15	477.47	515.92	564.87

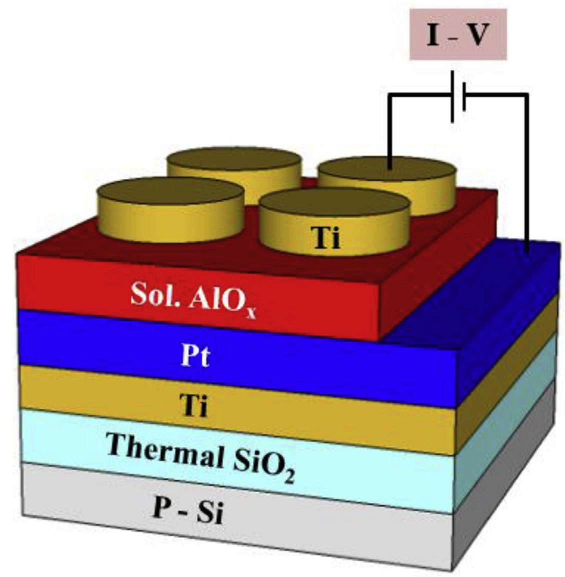

Fig. 1. Schematic of the solution-processed AlO_x ReRAM device with a Ti/AlO_x/Pt structure.

Table 2
Thickness and permittivity of AlO_x film according to annealing conditions.

	as-dep	400 °C	600 W	1200 W	1800 W	2400 W	3000 W
Thickness (nm)	159	132	130	120	118	110	106
Permittivity	6.75	6.88	6.94	7.17	7.32	7.42	7.48

positive or negative voltages to the top electrode and by grounding the bottom electrode. When the compliance current (CC) is 10 µA and positive voltage is applied to the upper electrode, the resistance suddenly changes from a high state to a low state at the critical voltage (forming voltage). The forming voltage is the smallest in the as-dep device and increased after heat treatment. Further, the higher the MWI power the larger the forming voltage. This is due to the effect of removing impurities in the solution-deposited AlO_x thin film and densifying the film by post-deposition heat treatment. Fig. 2 (b) shows the average on/off current ratio of solution-processed AlO_x ReRAMs extracted from the BRS characteristics and is defined as the difference between the HRS and LRS at 0.2 V. The average on/off ratio was the lowest in the as-dep device and was the highest in the device subjected to 3000 W MWI. In addition, the CTA device and the 600-W MWI device showed a similar on/off ratio. Nevertheless, it can be seen that the solution AlO_x layer is effectively heat-treated by MWI, and thus the MWI has advantages as a PDA process.

Fig. 3 shows the power consumption during the set and reset processes according to heat treatment conditions calculated from Fig. 2. The set operation power was defined as $P_{set} = V_{set} \times I_{cc}$, and the reset operation power was defined as P_{reset} = maximum (I × V) during the Reset process. As a result, the set operation power was less than 10 mW and the P_{set} decreased with increasing the microwave power. Also, although the reset operation showed the power consumption of larger than 10 mW in the CTA processed device, but the MWI processed devices showed a power consumption smaller less than 10 mW.

Fig. 4 shows the I-V characteristics on a log-log scale to investigate the current conduction mechanism in solution-processed AlO_x ReRAMs. In the LRS and HRS in the low-voltage range, all ReRAM devices exhibited a linear slope close to 1. These results confirmed that the LRS and the low-voltage range HRS follow the Ohmic conduction mechanism. On the other hand, the HRS in the high-voltage range has a nonlinear slope, suggesting that its conduction mechanism differs from