Improved Resistive Switching Behavior in Solution-processed AlO$_x$ based RRAM

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Abstract—**In this study, a comprehensive comparison in light of the switching behavior has been made to the AlO$_x$ dielectrics in resistive random access memory (RRAM) devices, which were fabricated with solution-processed (SP) and atomic-layer-deposition (ALD) based techniques under the same temperature (250 ℃). The improved resistive switching properties such as smaller V$_{set}$/V$_{reset}$ (1.2~1.8 V/ -0.9~-1.3 V), a narrower R$_{HRS}$/R$_{LRS}$ distribution (2~80 kΩ/0.8~1 kΩ) and a higher resistance ratio (8~230) under 5 mA compliance current (CC) have been achieved with SP method. Therefore, the solution-based fabrication method has the potential application for the flexible memory, due to the fabrication merits of the low-temperature, low cost, large area implementation and being environmental friendly.**

Keywords- RRAM, Resistive Switching, Aluminum Oxide, Solution-processed, ALD

I. INTRODUCTION

Resistive random access memory (RRAM) as one of the revolutionary nonvolatile memory (NVM) technologies has attracted extensive attention due to its excellent memory performances such as simple sandwich structure, fast switching speed, strong reliability and high-density scaling feasibility [1-4]. Al$_2$O$_3$ as the typical dielectric material has been explored due to the superior switching performance in recent years and is promising to extend its applications in the synaptic memory using the controllable conductance change [2-4]. Despite extensive investigation has been made to the atomic-layer-deposition (ALD) fabricated AlO$_x$ thin films

oxidized aluminium. Solution-processed (SP) method of oxide dielectric layers has received recently a great deal of interest due to its intrinsic advantages of low process temperature, simplicity of the process and cost-effective mass production. Although this method has been widely used in the fabrication of thin-film transistors (TFTs) [5], few works systematically studied the performances of AlO$_x$-based RRAM fabricated by SP methods.

In this work, the resistive switching (RS) characteristics of SP aluminum oxide dielectric with TiN/AlO$_x$/Pt structure were investigated. It has been found that the environmental-friendly low-temperature solution-based fabrication method has the potential for flexible memory applications.

II. EXPERIMENTAL

The prepared AlO$_x$ precursor solution was spin-coated onto the Pt (200 nm)/Ti/SiO$_2$/Si substrate at a spin rate of 4500 rpm for 40 s and annealed at 250 °C for 60 min under ambient conditions. The AlO$_x$ precursor solution was prepared by dissolving ~18.76 g of aluminum nitrate nonahydrate (Al (NO$_3$)$_3$·9H$_2$O) in 20 mL of deionized water without any additives. Then the solution was stirred vigorously for 30 min under air ambient conditions and filtered through 0.45 μm polyethersulfone (PES) syringe filters before spin-coating. As a control sample, the AlO$_x$ layer was deposited at 250 °C on the ALD system, model MNT/F-200. Finally, a 100 nm TiN thin film and a 70 nm Al thin film with a diameter of 0.1 mm were deposited for both samples using an e-beam evaporation, respectively.
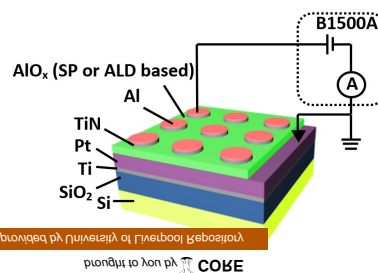

Figure 1. A schematic of structure and I-V measurement setup of Al/TiN/solution-processed or atomic-layer-deposition based AlO$_x$/Pt devices.

Fig. 1 shows the Al/TiN/AlO$_x$/Pt structure for current-voltage (I-V) measurements. The resistive switching behavior was characterized using a two-probe measurement system with Agilent B1500A high-

precision semiconductor analyzer (Fig. 1) in the dark and at room temperature with a Faraday Cage surrounding the wafer prober.
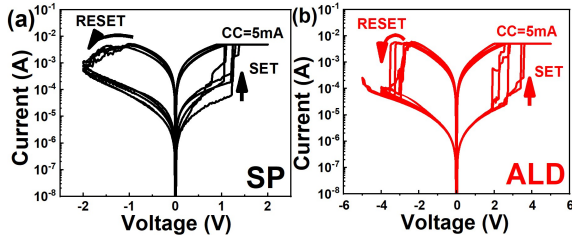
III. RESUSULTS AND DISCUSSION

Figure 2. Typical bipolar I-V characteristics of several TiN/AlO$_x$/Pt devices fabricated by (a) SP and (b) ALD methods for 5 times tests.

The typical DC I-V curves of the TiN/AlO$_x$/Pt devices fabricated by SP and ALD methods for 5 times tests are plotted in Fig. 2. Compliance current (CC) is set at 5 mA. The majority values for set voltage (V$_{set}$) for SP samples are around 1.2 V while the ones for ALD samples are up to 4 V. In reset process, nearly all reset voltages (V$_{reset}$) of SP samples are -1 V approximately. The ones for ALD samples are ranging from -2 V to -3.8 V. Furthermore, more gradual reset behavior can be observed for SP samples, of importance for application in the bionic circuit simulated by synapses and neural networks [4].

To study the effect of fabrication techniques on the RS behavior of TiN/AlO$_x$/Pt devices, we also carried out a statistic analysis of resistive switching parameters, such as V$_{set}$/V$_{reset}$, high/low resistance state (HRS/LRS) resistance (R$_{HRS}$/R$_{LRS}$, measured at 0.1 V) and resistance ratio as shown in Fig. 3. The analysis is based on the 40 times tests of data for 8 device units on each sample. SP samples exhibit a smaller average V$_{set}$ (1.49 V) and a smaller average V$_{reset}$ (-1.14 V) as shown in Fig. 3(a). The set and reset voltage values of ALD samples are 2.4 V and -1.5 V, respectively. Furthermore, SP samples have a sharper reset voltage distribution. Fig. 3(b) further illustrates the cumulative probability of R$_{HRS}$/R$_{LRS}$ for the SP and ALD fabricated samples. A very small range of LRS resistance level distribution (one order) can be seen for SP samples while R$_{LRS}$ variations are nearly up to two orders of magnitude for ALD samples. Moreover, the difference is nearly one order on scale in the high resistance region of HRS (90-100%). The R$_{HRS}$ values of some ALD samples are larger than 100 kΩ. The uniformity of RS parameters plays a significant role in high-density memory application. The randomness of conductive filaments (CFs) formation locations or the complex CF formation mechanism during the set process may result in the large V$_{set}$ dispersion. Furthermore, it can be seen that SP method also reduces the dispersion of V$_{set}$/V$_{reset}$, R$_{HRS}$/R$_{LRS}$. Finally, variations of the resistance ratio for TiN/AlO$_x$/Pt devices are presented in Fig. 3(c).

It can be observed that the magnitude of resistance ratio tends to be enhanced for SP samples (8~230) in comparison to that of ALD samples (4~130).
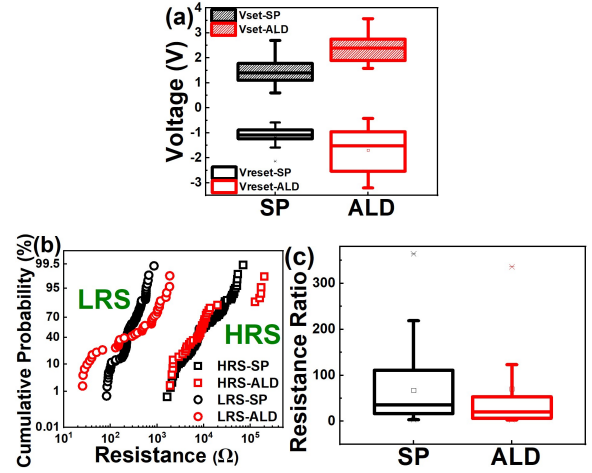

Figure 3. (a) The variations of set/reset voltages; (b) distribution of HRS/LRS resistance measured at 0.1 V;(c) variations of the resistance ratio for the TiN/AlO$_x$/Pt devices fabricated by SP and ALD methods.

IV. CONCLUSION

In conclusion, a comprehensive comparison in light of the switching behavior has been conducted on the AlO$_x$ dielectrics in RRAM devices, which were fabricated by SP and ALD under the same temperature (250 °C). The SP samples exhibited a sharper distribution of set/reset voltage (1.2~1.8 V/-0.9~-1.3 V), a narrower distribution of HRS/LRS resistance (2~80 kΩ/0.8~1 kΩ) and a larger resistance ratio (8~230) in comparison to ALD samples. The results point to the environmental-friendly low-temperature solution-based fabrication method as the contender for potential applications in the flexible memory.

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