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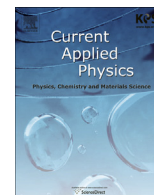

Effect of crystallization on the reliability of unipolar resistive-switching in HfO_2-based dielectrics

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The electroforming and unipolar resistive switching characteristics of as-deposited and annealed HfO_2 and $Hf_{0.6}Si_{0.4}O_2$ films are reported. The reliability of HfO_2 devices is shown to be significantly degraded by annealing at 600 °C, during which the film is observed to crystallize. In contrast, the characteristics of $Hf_{0.6}Si_{0.4}O_2$ devices subjected to the same annealing conditions are found to be unchanged, consistent with the fact that the films remain amorphous. These differences are attributed to the presence of grain boundaries and can have important implications for the use of HfO_2 in ReRAM applications.

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1. Introduction

Redox-based resistive switching represents a class of processes by which the resistance of a dielectric thin-film can be electrically switched between low and high resistance states. It is of particular interest for the fabrication of non-volatile resistive random-access-memory (ReRAM) where it offers to provide devices with better scalability, lower power consumption, faster switching, longer retention time, and simpler structure than conventional charge-storage technologies [1,2]. The physical processes underpinning resistive-switching are specific to particular material-systems but include filamentary processes in which the device resistance is controlled by a conductive filament spanning the dielectric film. In this case the low resistance state is independent of device area and switching is induced by breaking (reset) and reforming (set) part of the filament [3,4]. The switching mechanism is classified as bipolar or unipolar, depending on whether the reset and set operations are achieved with switching voltages of opposite polarity or the same polarity, respectively. Bipolar operation generally involves the motion of charged defects or ions, while unipolar operation is dominated by thermal processes induced by Joule heating [2,5].

Among the broad range of materials that exhibit resistive switching, transition metal oxides (TMOs), including NiO, TiO_2, ZrO_2, Cu_xO, TaO_x, WO_x and HfO_2, are of interest due to their simple chemical structure, ease of growth/deposition and excellent resistive-switching characteristics [6]. HfO_2 is of particular interest as it is already used in high-performance complementary metal-oxide semiconductor (CMOS) circuits as a high-k gate dielectric [7], exhibits both bipolar and unipolar switching [6], and has a large band-gap [7] (>6.0 eV) which is a prerequisite for a large resistance ratio between 'on' and 'off' states.) Its use as the active element in non-volatile memories would build on these technological advantages and reduce the number of materials required for integrated device fabrication. One potential problem with HfO_2, however, is the fact that it crystallizes at temperatures ≥400 °C. It is well known that grain boundaries act as preferential paths for atomic diffusion, current leakage and dielectric breakdown [8], and can therefore compromise device reliability and endurance. This could be a particular problem as ReRAM devices shrink to the point where individual memory cells contain few grain boundaries and the variation from cell to cell is maximized (e.g. neighbouring cells may have a few, one or no grain boundaries). When used as a gate dielectric HfO_2 is generally alloyed with SiO_2 to increase its

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crystallization temperature and ensure that it remains amorphous during thermal processing [7].

Given the nature of resistive switching it is reasonable to expect that grain boundaries may have a similar detrimental impact on the reliability of ReRAM devices and there have been several insightful studies of such effects [8–10]. To further understand the impact of grain boundaries on HfO_2-based ReRAM this study compares the forming and unipolar resistive switching characteristics of amorphous HfO_2 films with those of films crystallized by annealing at 600 °C for 1 h. The characteristics of as-deposited and annealed $Hf_{0.6}Si_{0.4}O_2$ films, which remain amorphous after annealing, are also measured for comparison.

2. Experimental

Metal–insulator–metal (MIM) test structures, consisting of Pt (200 nm)/$Hf_xSi_{1-x}O_2$ (60 nm)/Pt (200 nm) with $x = 1$ and 0.6, were fabricated on SiO_2/Si substrates by sputter deposition, as shown in Fig. 1. Direct-current (DC) magnetron sputtering was first used to deposit a thin (15 nm) Ti wetting layer and bottom Pt contact layer (200 nm thick). This was followed by the deposition of HfO_2 and $Hf_{0.6}Si_{0.4}O_2$ films using radio-frequency (RF) sources fitted with pure HfO_2 (99.99%) and SiO_2 (99.999%) targets. Half of each sample was maintained in its as-deposited state while the other half was annealed at 600 °C for 1 h in a quartz-tube furnace. Top contacts were subsequently fabricated on unannealed and annealed samples using a standard lift-off process, with Pt top contact layers deposited using DC sputtering. This ensured that the top contact layer underwent identical processing for all samples.

The structure of $Hf_xSi_{1-x}O_2$ films was determined by glancing-incidence x-ray diffraction (GI-XRD), while the thickness and composition of selected test samples (including $Hf_xSi_{1-x}O_2$films deposited directly onto Si substrates) was determined using Rutherford backscattering spectrometry (RBS) (data not shown) and scanning electron microscopy (SEM). Electrical characterization of the test structures was undertaken using an Agilent Technologies B1500A semiconductor parametric analyser in DC sweep mode. Positive bias was applied to the top electrode while the bottom electrode was grounded.

3. Results and discussion

GI-XRD spectra of as-deposited and annealed HfO_2 and $Hf_{0.6}Si_{0.4}O_2$ films prior to the deposition of the top contact layer are shown in Fig. 2. The spectra confirm the amorphous structure of the as-deposited films and show that while the $Hf_{0.6}Si_{0.4}O_2$ film remains amorphous after annealing at 600 °C for 1 h, the HfO_2 film crystallizes to form a polycrystalline layer of monoclinic-HfO_2. A systematic study of the thermal stability of the films showed that the HfO_2 film crystallizes at temperatures in the range 400–600 °C, while the $Hf_{0.6}Si_{0.4}O_2$ is stable up to temperatures exceeding 800 °C. This is consistent with earlier work that additionally

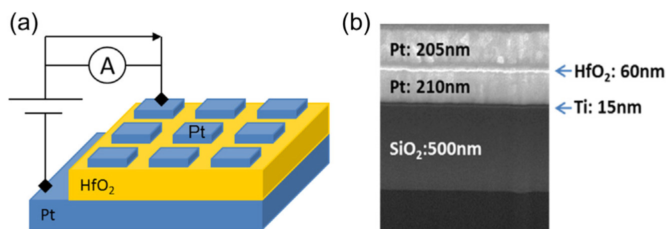

Fig. 1. a) Schematic diagram of sample configuration used for testing and b) an SEM image of a sample cross-section showing the sample structure.

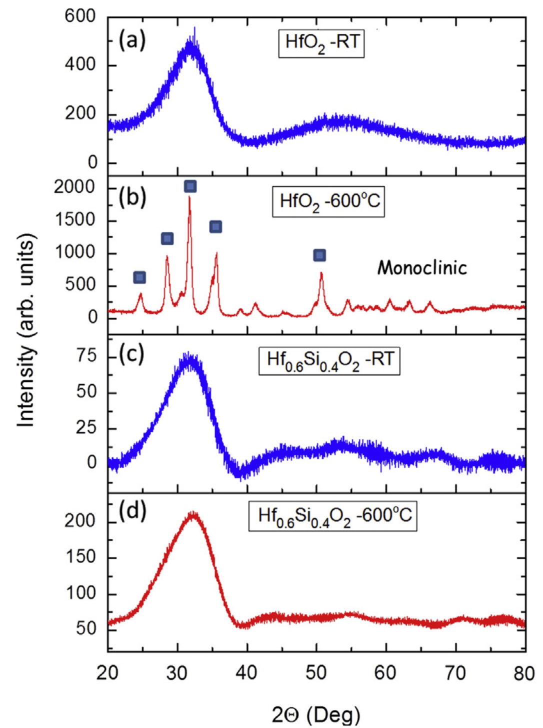

Fig. 2. GI-XRD spectra of dielectric films: a) as-deposited HfO_2, b) annealed HfO_2, c) as-deposited $Hf_{0.6}Si_{0.4}O_2$, and d) annealed $Hf_{0.6}Si_{0.4}O_2$. Measurements were performed with Cu Kα radiation ($\lambda = 0.1545$ nm) using an angle of incidence of 0.5^0 to the sample surface. The indicated peaks correspond to diffraction from (110), (−111), (111), (−211) and (220) planes of the monoclinic HfO_2 phase (from left to right).

showed that crystallization of $Hf_xSi_{1-x}O_2$ films involves phase separation of HfO_2 and SiO_2 to form polycrystalline HfO_2 in an amorphous SiO_2 matrix [7,11,12].

Forming was achieved by measuring the current–voltage characteristics of test structures during a linear voltage ramp, with typical results shown in Fig. 3a for an as-deposited HfO_2 film. Increasing the applied voltage produces a gradual increase in leakage current due to trap-assisted tunnelling [13], followed by a catastrophic increase in current at a critical 'forming' voltage (or field). Hard breakdown and irreversible failure of the device is avoided by limiting the maximum current, which in this case was limited to 50 μA. For the purpose of discussion, the forming voltage (V_f) is defined here as the voltage at which the current reaches the compliance limit (i.e. 50 μA). (NB: because the current rises rapidly during breakdown, the value of V_f is relatively insensitive to the choice of compliance current).

To compare the behaviour of different samples, cumulative probability distributions were derived from the measured V_f values for each of the samples investigated, assuming a Weibull cumulative probability distribution, which is given by: $P = 1 - \exp[-(V_f/V_o)^{\beta}]$, where P is the cumulative failure probability, and V_o and β are the Weibull scale and slope parameters, respectively. Note that the scale parameter provides an estimate of the median forming voltage, while the slope-parameter, β, is a measure of the dispersion in V_f, with higher β indicating smaller dispersion.

Fig. 3b shows measured distributions for as-deposited and annealed samples, together with least-squares fits to the Weibull cumulative probability distribution, clearly highlighting significant differences between the various samples. Of particular note is the large difference between as-deposited and annealed HfO_2 films. This includes a large shift in the median forming voltage, with V_o decreasing from 23.3 ± 0.2 V for the as-deposited film to 9.9 ± 0.5 V

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