Al_2O_3-Ta_2O_5 MULTILAYER THIN FILMS DEPOSITED BY PULSED DIRECT CURRENT MAGNETRON SPUTTERING FOR DIELECTRIC APPLICATIONS

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Abstract

This research aims at synthesizing multilayer oxide thin films made of Al_2O_3 and Ta_2O_5 for dielectric applications. Multilayer thin films made of two, four, or eight oxide layers are synthesized by physical vapor deposition, specifically the mid-frequency pulsed direct current magnetron sputtering. The thin films are made of stoichiometric Al_2O_3 and Ta_2O_5 layers having a specific morphology observed from cross-section images obtained by scanning electron microscopy (SEM). The Al_2O_3 layers have a columnar structure, whereas the Ta_2O_5 layers are uniformly dense. X-ray diffraction (XRD) characterizations show that these oxide layers have very limited crystallinity due to the experimental conditions used during the magnetron sputtering process, particularly the low temperature of the substrate.

The dielectric behavior of the multilayer oxide thin films is assessed by measuring their dielectric breakdown potential. The two-layer and four-layer systems have intermediate values compared to the dielectric breakdown potentials measured for a monolayer of Al_2O_3 and a monolayer of Ta_2O_5 produced under the same experimental conditions. In the case of the eight-layer system, the dielectric breakdown potential value is the highest one, even higher than that measured for a monolayer of Ta_2O_5.

Keywords: Reactive magnetron sputtering, aluminum oxide, tantalum oxide, multilayer thin films, dielectric breakdown

1. INTRODUCTION

The thin oxide coatings are widely studied in the microelectronics industry for their insulation properties characterized by high dielectric constants [1]. They are used for various applications such as CMOS transistors, flash memory, and surface passivation of solar cells [2]. The magnetron sputtered aluminum oxide (Al_2O_3) coatings are among the most common ones, mainly due to the low-cost production. On the other hand, the coating process is hard to control in order to achieve high reproducibility and the achieved dielectric strength is far from the ideal values measured for bulk Al_2O_3. Tantalum oxide (Ta_2O_5) coatings could be a valuable alternative since they have better dielectric properties and the reactive process is easier to control, but they are also more expensive [3,4].

This work aims to improve the dielectric properties of surface oxide layers and, at the same time, to maintain a reasonable production cost. A relevant solution is the combination of both oxides in a multilayer system that alternates Al_2O_3 layers and Ta_2O_5 layers. According to Martinez-Perdiguero *et al.*, the interface between two oxide layers is advantageous to improve the dielectric properties of surface oxide thin films [5]. They have observed a significant improvement in the dielectric breakdown potential of a multilayer thin film made of Al_2O_3 and SiO_2 compared to the average value of a single monolayer of each oxide. They explain that the interface between two oxide layers acts as a barrier able to slow down the electron flows inside the insulating material. The resulting dielectric breakdown potential is higher than the average value of both oxide layers [6].

Multilayer oxide thin films made of Al_2O_3 and Ta_2O_5 are barely studied in the literature. The present work explores a solution to synthesis this multilayer system at low temperature by pulsed direct current magnetron reactive sputtering. Multilayer thin films made of two, four, and eight layers are studied. The experimental conditions are described, and the synthesized thin films are characterized by scanning electron microscopy associated with energy-dispersive X-ray spectroscopy (SEM-EDS) and X-ray diffraction (XRD). Finally, the dielectric properties of the thin films are assessed by measuring their dielectric breakdown potential.

2. MATERIALS AND METHODS

The Al_2O_3 and Ta_2O_5 layers are deposited by pulsed direct current magnetron sputtering using the HVM Flexilab thin film deposition system (HVM Plasma spol s ro). The aluminum and tantalum targets are powered by a Pinnacle Plus + power supply (Advanced Energy) at 250 W. The Si (100) and steel substrates were ultrasonically cleaned for 3 min in acetone and 3 min in isopropyl alcohol. Then, they are placed inside the chamber of the deposition system. After pumping for two hours, a background pressure value of 10^{-3} Pa is reached inside the chamber. Then, argon and oxygen are continuously injected inside the chamber. The flow of Ar is 60 sccm. The flow of O_2 is 4 sccm in the case of the Al_2O_3 layers and 19.2 sccm in the case of the Ta_2O_5 layers. The corresponding pressures are 1.0 Pa and 1.2 Pa, respectively. The temperature of the substrates is maintained at 180 °C during deposition. The direct current is pulsed at a frequency of 350 kHz and a pulse reversal time of 0.6 µs.

The morphology and the chemical composition of the obtained thin films are studied from cross-sections of the samples with a scanning electron microscope (SEM, Tescan MIRA3) equipped with an energy dispersive X-ray spectrometer (EDS). The phases of the oxide layers are characterized by X-ray diffraction (XRD) with a Rigaku SmartLab Type F diffractometer using monochromatic $Cu_{K\alpha}$ radiation (λ = 0.15406 nm). The phases are identified from the diffraction files provided by the International Centre for Diffraction Data.

The dielectric breakdown potentials are measured from the multilayer oxide thin films deposited on the steel substrates. Each side of one sample is connected to one output of a DC power supply, progressively increasing the voltage between the two electrodes. Because the multilayer oxide thin film is an electrical insulator, there is no current flow through the material until dielectric breakdown occurs. The corresponding potential value is the dielectric breakdown potential of the multilayer oxide thin film. The measurement is repeated at ten different positions for each sample to obtain an average value.

3. RESULTS AND DISCUSSION

The cross-section SEM image of **Figure 1** shows the morphology of the oxide thin film made of two layers deposited on a silicon substrate. According to the EDS characterizations, the lower part of the thin film is stoichiometric aluminum oxide, and the upper part of the thin film is stoichiometric tantalum oxide.

The continuity at the interface is good, with no separation between the two oxide layers. The Al_2O_3 layer has a columnar structure. This type of growth is commonly observed for thin films deposited by magnetron sputtering [7].

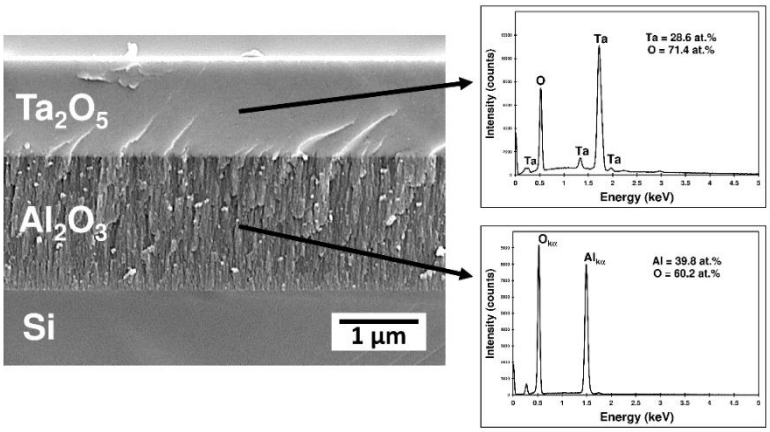

Figure 1 Cross-section SEM image and pointed EDS spectra of the bilayer oxide thin film deposited on a silicon substrate by pulsed direct current magnetron sputtering

This morphology indicates that the experimental conditions (target power, partial pressure, and substrate temperature) provide enough energy to the aluminum and oxygen atoms to direct the columnar growth of the Al_2O_3 layer. The morphology of the Ta_2O_5 layer looks different, being dense and uniform with no specific growth direction observable at this scale.

The four-layer and the eight-layer thin films are observed in the cross-sectional SEM images of **Figure 2**. The characteristic morphologies of the oxide layers are also observable, i.e., the columnar structure of the Al_2O_3 layers and the dense and uniform morphology of the Ta_2O_5 layers.

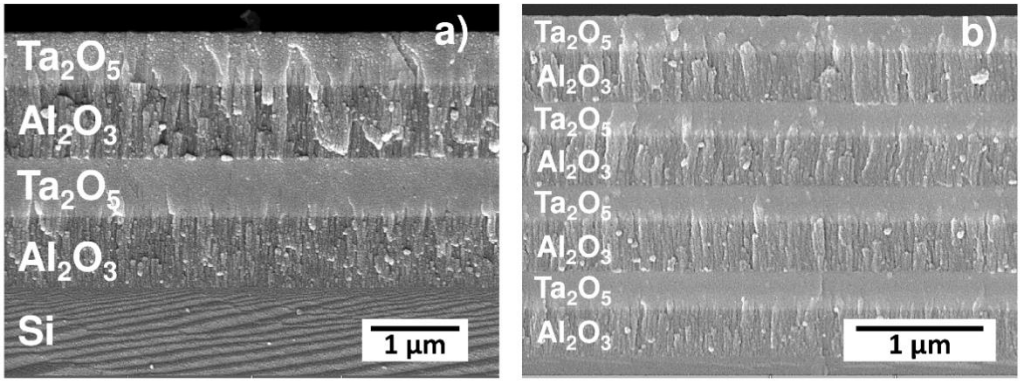

Figure 2 Cross-section SEM images of (a) the four-layers and (b) the eight-layers Al_2O_3-Ta_2O_5 thin films

The interfaces between the oxide layers are good for both samples, inducing continuity within the thin films without separation. All of these interfaces show that the tantalum oxide layers firstly grow by mimicking the columnar morphology of the aluminum oxide layer below. After a few nanometers, the Ta_2O_5 layer becomes more uniform with no specific growth direction as previously described for the bilayer oxide film (**Figure 1**).

The XRD patterns of the three multilayer oxide thin films are shown in **Figure 3**. Whatever the number of oxide layers, the XRD patterns show broad peaks corresponding to a nanocrystalline material. The low temperature used during the magnetron sputtering process induces a very low crystallization of the synthesized oxide layers [8]. However, a characteristic signal corresponding to Ta_2O_5 (pdf file # 00-025-0922) is observable for the three multilayer thin films corresponding to the upper oxide layer of each sample.

The dielectric breakdown potentials of the three multilayer oxide thin films are shown in **Figure 4** and compared to the values obtained for a monolayer of Al_2O_3 and a monolayer of Ta_2O_5 produced under the same experimental conditions.

As expected, the dielectric breakdown potential of the Al_2O_3 monolayer (100 V μm^{-1}) is lower than that of the Ta_2O_5 monolayer (171 V μm^{-1}). The two-layer thin films (152 V μm^{-1}) and the four-layer thin films (105 V μm^{-1})

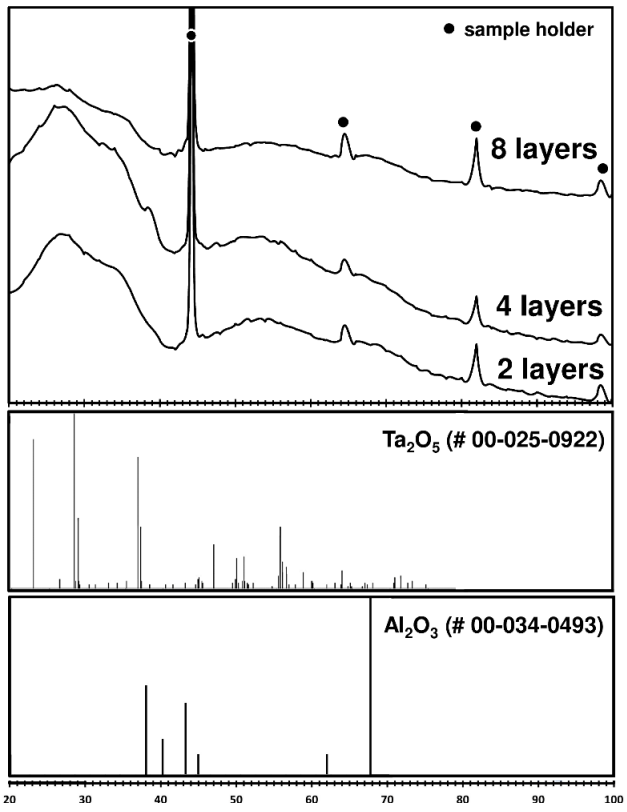

Figure 3 XRD patterns of the multilayer Al_2O_3-Ta_2O_5 thin films

have intermediate values compared to both oxide monolayers. In the case of the value of the eight-layer thin film, the dielectric breakdown potential value is the highest (184 V μm^{-1}), even higher than that measured for the Ta_2O_5 monolayer.

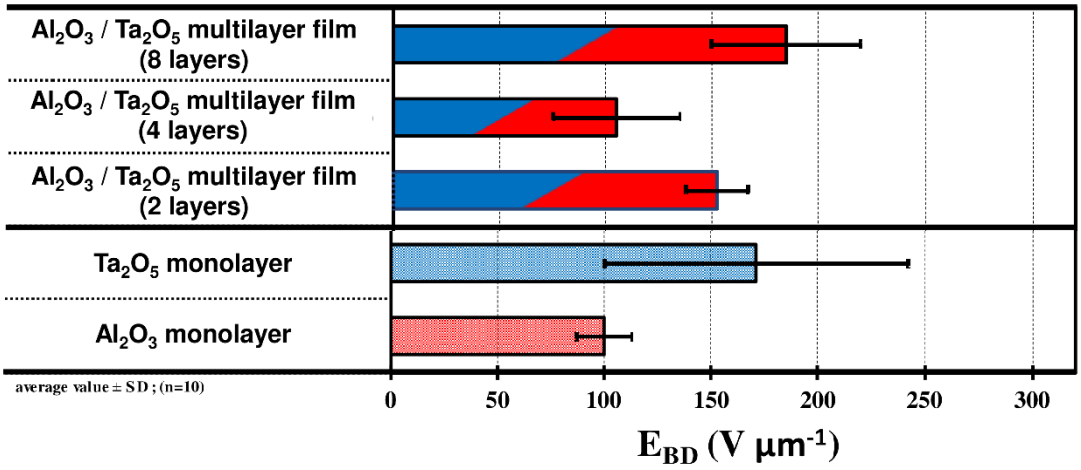

Figure 4 Dielectric breakdown potential (E_{BD}) of multilayer Al_2O_3-Ta_2O_5 thin films

The result of the four-layer oxide thin film is surprising in comparison with that of the other two multilayer thin films. The obtained dielectric breakdown potential value is barely higher than that of the Al_2O_3 monolayer despite the presence of Ta_2O_5 layers in the thin film in a multilayer system. This result may be due to some defects in the layers generated during deposition. This phenomenon is well known in reactive magnetron sputtering of oxide thin films [9]. The presence of oxygen in the atmosphere of the chamber contributes to the poisoning of the target, changing the surface of the metallic target to a dielectric oxide. The electric charges involved in the process are trapped in the dielectric at the surface of the target, and they accumulate until being released by arcing. This abrupt discharge results in an instability of the process, inducing uncontrollable damage to the deposited thin film [10]. However, the reason why this phenomenon occurs more intensively for the sample with four layers remains unanswered.

On the other hand, the two-layer and eight-layer thin films have higher dielectric breakdown potentials than the average value of the monolayers of Al_2O_3 and Ta_2O_5. This improvement is due to the impact of the interfaces in the multilayer thin films, which slow the propagation of the electric charges during an electrical breakdown. Gefle *et al.* have studied the mechanisms implied in this barrier effect. They describe the influence of several factors that prevent charge propagation at the interface between two solid media [11]. The main factors involved are local mechanical strain, space charge, permittivity gradient, and conductivity gradient. These factors are specific to the interfaces and increase the breakdown voltage and the breakdown time [12]. As a result, the dielectric strength of the multilayer oxide thin films is improved.

4. CONCLUSION

Thin films made of two, four, or eight alternate layers of Al_2O_3 and Ta_2O_5 were synthesized by pulsed direct current magnetron reactive sputtering. The process produced stoichiometric oxide layers of very low crystallinity with a characteristic morphology for each oxide layer. Thanks to the interfaces of the multilayer systems, the two-layer and eight-layer thin films show improved dielectric breakdown potentials compared to the average value measured for a monolayer of Al_2O_3 and a monolayer of Ta_2O_5. The value of the four-layer thin film is lower, certainly because of some defects in the layers generated during the magnetron reactive sputtering deposition.

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