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Short-Duration Rapid-Thermal-Annealing Processing of Tantalum Oxide Thin Films

Santhiagu Ezhilvalavan and Tseung-Yuen Tseng*

Department of Electronics Engineering and Institute of Electronics, National Chiao Tung University, Hsinchu-300, Taiwan, Republic of China

The effect of the rapid thermal annealing (RTA) processing time on the electrical properties of reactively sputtered tantalum oxide (Ta_2O_5) films that was deposited onto Pt/SiO_2/*n*-Si substrates, which resulted in the formation of a metal–insulator–metal (MIM) planar capacitor structure, was studied. The Ta_2O_5 MIM capacitors were subjected to different RTA processing times (30 s to 30 min) at temperatures in the range of 600°–800°C in an ambient oxygen-gas atmosphere. A very-short-duration RTA process at a temperature of 800°C in oxygen gas for 30 s crystallized the films, decreased the leakage current density (to 10^{-10} A/cm^2 at a stress field of 100 kV/cm), increased the dielectric constant (to 52), and resulted in the most-reliable time-dependent dielectric-breakdown characteristics. The decrease in leakage current density was attributed to the reduction of oxygen vacancies and the suppression of silicon diffusion from the SiO_2/*n*-Si substrate into the Ta_2O_5 grain and the grain boundary, because of the shorter-duration annealing. Increasing the annealing time to >30 s increased the leakage current density. The annealing duration of the RTA process was more crucial in regard to obtaining optimum dielectric properties and low leakage current densities. Time-dependent dielectric-breakdown characteristics indicated that Ta_2O_5 MIM film capacitors that were subjected to an RTA process at a temperature of 800°C for 30 s in oxygen gas can survive a stress field of 1.5 MV/cm for 10 years. The electrical and dielectric measurements in the MIM configuration showed that Ta_2O_5 is a good dielectric material and is suitable for use in future dynamic random-access memories.

I. Introduction

Tantalum oxide (Ta_2O_5) films have been demonstrated to be highly promising storage dielectrics in the next generation of dynamic random-access memories (DRAMs), because of their high dielectric constant and relatively good leakage current density.[1–3] Thermal treatments in various ambient environments have been performed after Ta_2O_5 film deposition, to reduce the leakage current and improve the quality of the film; these treatments include dry oxygen gas,[4–6] dry O_3,[7] UV-O_2,[8] UV-O_3,[8,9] UV-O_3 (UV-O_2),[8] N_2O plasma,[10] and oxygen-gas plasma.[11] During thermal treatment in the oxygen-containing ambient environment, oxygen diffuses into the Ta_2O_5 films, which leads to a decrease in the number of oxygen vacancies and a reduction of the number of weak spots in the Ta_2O_5 films.[12] When Ta_2O_5 film is deposited onto silicon, which forms a metal-oxide-semiconductor (MOS) capacitor structure, and is subjected to various post-deposition annealings, the process results in the formation of an interfacial silica (SiO_2) layer that has a low dielectric constant.[13] This event is one of the most serious problems for the application of Ta_2O_5 films to DRAM capacitors.

Rapid thermal annealing (RTA) is often used in the processing of semiconductor devices to minimize undesired diffusion and interfacial reactions.[14,15] Although there are some reports available in the literature about the RTA processing of Ta_2O_5 films, most of the research has been for Ta_2O_5 films that have a MOS capacitor structure and the duration of the RTA treatment was maintained at ≥60 s.[16–22] Also in the earlier reports, the importance of the RTA processing time on the leakage current density of Ta_2O_5 films was not studied in detail. Sun and Chen[16,17] demonstrated that Ta_2O_5 films that have been treated via RTA in N_2O are much less leaky than those that have been treated via conventional furnace annealing in oxygen gas. However, those researchers could only speculate about the reasons behind the improvement due to RTA treatment in N_2O. Lau *et al.*[18] reported that RTA treatment in N_2O is more efficient than RTA treatment in oxygen gas, in regard to providing free oxygen atoms. However, those researchers could not distinguish whether the decrease in silicon contamination or the decrease in oxygen vacancies was the principal factor that led to a lower leakage current. Pignolet *et al.*[19] observed a dielectric constant of 45 and leakage current density of 10^{-8} A/cm^2 for the RTA-processed reactively sputtered Ta_2O_5 films. However, those researchers did not study the effect of the RTA time duration on the leakage-current properties of Ta_2O_5 films. Ta_2O_5 storage capacitor films on rugged polysilicon that had been treated via RTA treatment in oxygen gas were reported by Kamiyama *et al.*[20] and Lo *et al.*[21] Their results showed that RTA treatment in oxygen gas reduces the density of the oxygen vacancies.

Very-short-duration (for ~30 s) RTA processing of Ta_2O_5 films in an ambient oxygen-gas atmosphere, having a MIM planar capacitor structure, are not reported in the literature. Decreasing the processing time will restrict the diffusion of silicon atoms, and using the MIM structure permits the elimination of the possible formation of the interfacial SiO_2 layer. In this investigation, we report the importance of the very-short-duration (30 s) RTA processing of Ta_2O_5 film that is formed on a platinum-coated SiO_2/*n*-Si substrate that has a MIM capacitor structure. Detailed investigation on the electrical and dielectric properties shows the high reliability and quality of the Ta_2O_5 film for possible incorporation into ultralarge-scale integration (ULSI) applications.

II. Experimental Procedure

The deposition of Ta_2O_5 thin films in this study was performed on Pt/SiO_2/*n*-Si substrates, which forms a MIM planar capacitor structure via dc magnetron sputtering from a high-purity tantalum-metal target (diameter of 2.5 in.). More details on the deposition technique may be found in our earlier work.[23] The sputtering gas was a mixture of 80% argon and 20% oxy-

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gen, with a total pressure of 10 mtorr (~1.33 Pa). The purity of the gas was ascertained by using the same procedure as that reported by Oehrlein *et al.*[24] During deposition, the chamber was first filled with argon gas and then was used to presputter the target clean for at least 5 min. Then, the argon/oxygen gases were introduced into the chamber to attain a total pressure of 100 mtorr (~13.3 Pa). The substrates were not intentionally heated. The film thickness was estimated to be 100 nm by using both ellipsometry, with 632.8 nm wavelength He-Ne laser light source (Rudoph Research, Flanders, NJ), and an α stepper (Model 200, Tencor, USA); the thickness measurement was further verified by examining the cross section of the film via scanning electron microscopy (SEM). The platinum top electrode, with a thickness of 50 nm and a diameter of 350 μm, was patterned by using a shadow-mask process. The Ta_2O_5 capacitors were electrically characterized via computer control (Model HP4145B, Hewlett–Packard, Tokyo, Japan) for current–voltage (*I–V*) measurements. The dc leakage currents were evaluated using a step-voltage technique, and the measurement was performed with sufficient charging time to attain the steady-state dc leakage regime. The step-voltage technique, with a sufficiently long charging time, is generally accepted as revealing the most-accurate data.[25]

The capacitance–voltage (*C–V*) characteristics and the dielectric loss tangent were measured at a frequency of 100 kHz with a sweeping signal of 0.5 V (ac), using an impedance–gain phase analyzer (Model HP4194A, Hewlett–Packard). RTA was performed in an RTA furnace (Model HPC 700, Ulvac Sinku-Rico, Tokyo, Japan) at temperatures in the range of 500°–800°C for 30 s to 30 min in an ambient oxygen-gas atmosphere. The maximum heating rate (~100°C/s) was used. The structure and impurities that were distributed in the Ta_2O_5 films were analyzed by using X-ray diffractometry (XRD) (Model D5000, Siemens, Munich, FRG), SEM (Model S250 microscope, Hitachi, Tokyo, Japan, fitted for energy-dispersive X-ray analysis (EDX)), and secondary-ion mass spectrometry (SIMS) (Model IMS-4f, CAMECA, Courbevoie Cedex, France).

III. Results and Discussion

(1) Microstructural Features

Figure 1 shows the XRD patterns of as-deposited Ta_2O_5 films that have been subjected to RTA at different temperatures (600°–800°C) for 30 s in an ambient oxygen-gas atmosphere. The as-deposited films were amorphous, and the films were crystalline when subjected to RTA treatment at temperatures $\geq$600°C for 30 s. The crystallinity increased as the annealing temperature increased, in terms of an increase in the intensity of the diffracted peaks, whereas the thickness of the film was

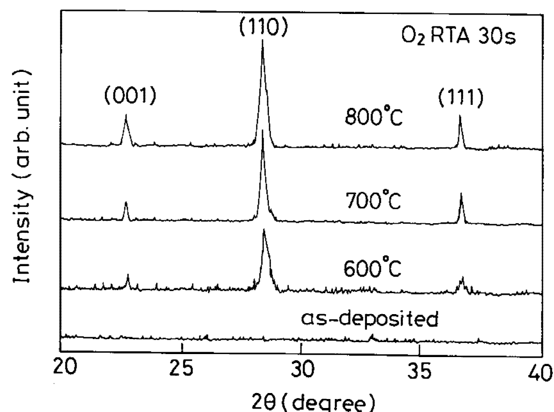

Fig. 1. XRD spectra of Ta_2O_5 films as-deposited and processed via RTA in oxygen gas at different temperatures for 30 s.

maintained at a constant value. The XRD spectra of the films in Fig. 1 correspond either to β-Ta_2O_5 (the orthorhombic crystal structure) or α-Ta_2O_5 (a high-temperature form of Ta_2O_5).[26] This uncertainty occurs because the spectra of the two phases are too similar to allow distinction between them on the basis of XRD alone.[26,27] In the RTA processing of amorphous films, during crystallization, the excess free energy of the amorphous state, relative to the crystalline phase, is released instantaneously, which results in explosive crystallization. Such a process often produces much-higher temperatures than the set values instantaneously in a localized area and provides a probable opportunity for the Ta_2O_5 to transform to the α-phase.[14,15,19] Because our RTA processing time is very short and also because the cooling and heating rates are very high (100°C/s, almost equivalent to normal quenching methods), we believe that there is a possibility of α-phase formation; this phase can appear as a minor phase and may be distributed in the grain boundaries. Such two-phase structures have been mentioned in research by Pignolet *et al.*[19] on RTA-processed Ta_2O_5 films.

SEM analysis readily shows that the as-deposited Ta_2O_5 film is amorphous, whereas the annealed Ta_2O_5 films are polycrystalline. Figure 2 shows SEM micrographs of the amorphous Ta_2O_5 thin film (Fig. 2(a)) and crystalline Ta_2O_5 thin films that have been annealed at a temperature of 800°C for 30 s and 30 min (Figs. 2(b) and (c), respectively). The average grain size was estimated to be 0.3–1 μm. The grains for the film that was subjected to RTA processing for 30 s are angular and have thin grain boundaries, whereas the films that were annealed for a longer duration (~30 min) exhibit larger grain-size distributions. These results demonstrate that RTA that is conducted for a longer duration leads to sufficient grain growth; in contrast, RTA treatment for 30 s causes restricted grain growth and grain-boundary formation. This result is obvious because an RTA processing time of 30 s is very short and is too rapid for possible nucleation and growth kinetics to occur.

(2) Electrical Characteristics

Figure 3 shows the *I–V* characteristics of Ta_2O_5 thin-film MIM capacitors that have been subjected to RTA processing for 30 min in an ambient oxygen-gas atmosphere at temperatures of 600°, 700°, and 800°C. The leakage current of amorphous Ta_2O_5 is lower than that of polycrystalline Ta_2O_5 films that have been annealed at higher temperatures for a long time (30 min). The as-deposited amorphous Ta_2O_5 films show good electrical properties, in terms of leakage current density (10^{-10} A/cm^2 at a stress field of 100 kV/cm) and dielectric constant (31). The leakage current that flows through the Ta_2O_5 film increases from 10^{-10} A/cm^2 to 10^{-7} A/cm^2 following the annealing. Detailed analysis of the leakage-current mechanisms of amorphous and polycrystalline Ta_2O_5 films at different electric-field regimes were reported in our recent paper.[23] The increase in the leakage current density in the crystallized film is attributed to silicon that has penetrated into the Ta_2O_5 grain and the grain boundary from the underlying SiO_2/*n*-Si substrate material.

Figure 4 shows the variation in the leakage current density with the applied electric field for the Ta_2O_5 films that have been subjected to RTA processing at different temperatures for 30 s in an ambient oxygen-gas atmosphere. The *I–V* curve shifts to a higher leakage current density as the annealing temperature increases, up to 700°C. Furthermore, increasing the annealing temperature to 800°C results in *I–V* characteristics with a very low leakage current. This phenomenon can be related to the degree of crystallization, because the as-deposited amorphous films begin to crystallize at temperatures $\geq$600°C and they undergo complete crystallization at a temperature of 800°C. This observation is supported by the XRD results, which indicate complete crystallization of the films that have been annealed at a temperature of 800°C (by way of a higher intensity of the diffracted peaks), in comparison to the films that have been annealed at temperatures of 600° or 700°C. In addition, RTA that is performed at higher tempera-

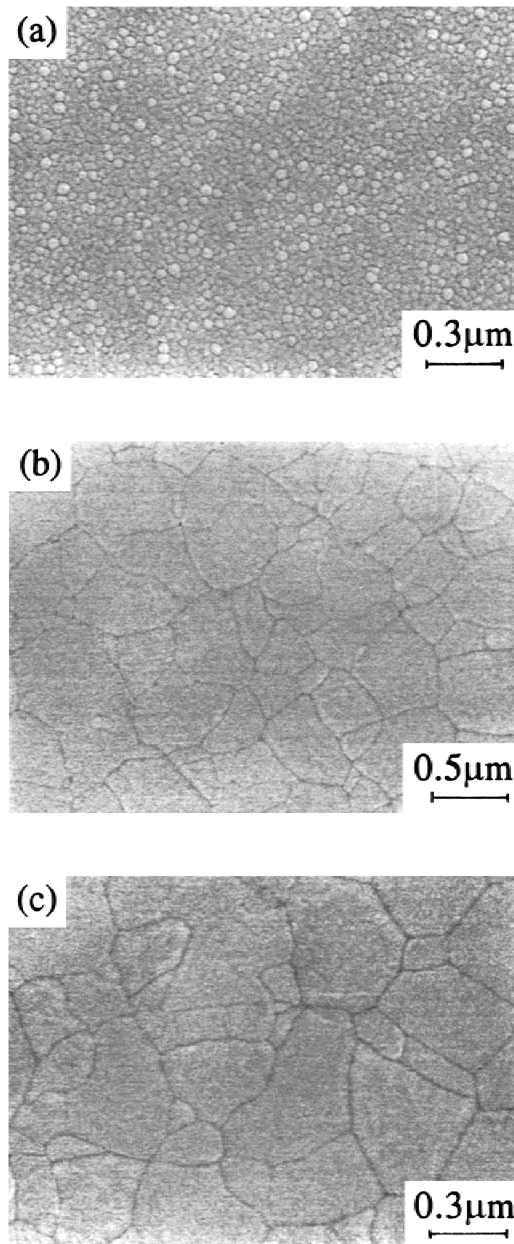

Fig. 2. SEM micrographs of Ta_2O_5 films ((a) as-deposited amorphous film, (b) film after RTA treatment for 30 s at 800°C in oxygen gas, and (c) film after RTA treatment for 30 min at 800°C in oxygen gas).

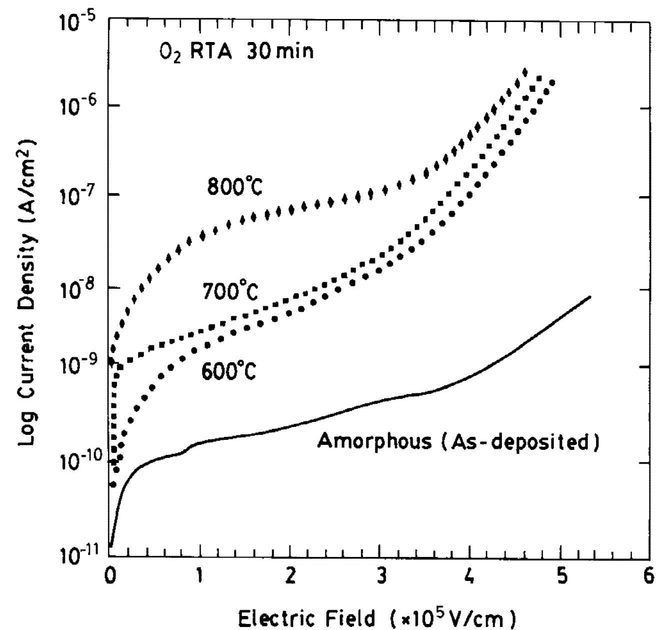

Fig. 3. Leakage current density versus applied electric field for amorphous and polycrystalline Ta_2O_5 films processed via RTA for 30 min in an ambient oxygen-gas atmosphere at different temperatures.

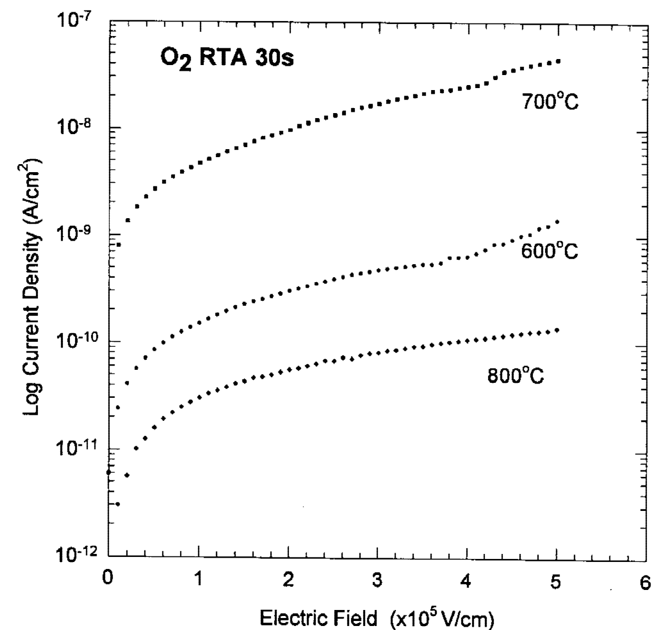

Fig. 4. Leakage current density versus applied electric field for Ta_2O_5 films subjected to RTA processing at different temperatures for 30 s in an ambient oxygen-gas atmosphere.

ture in an ambient oxygen-gas atmosphere provides greater grain growth and also corresponds to greater diffusion rates and greater amounts of oxidation, thereby decreasing the oxygen vacancies and, hence, reducing the leakage current density.

The effect of the RTA processing time duration—i.e., the holding time of the films at higher annealing temperature—on the leakage current property of Ta_2O_5 films was also studied. Figure 5 shows the leakage current density as a function of the RTA processing time of Ta_2O_5 films that have been processed at a temperature of 800°C in an ambient oxygen-gas atmosphere for 30–120 s. A shorter processing time (~30 s) clearly leads to better electrical characteristics, by way of reducing the leakage current density from 10^{-7} A/cm^2 to 10^{-10} A/cm^2 at a stress field of 100 kV/cm. The present results demonstrate that a much-shorter duration of RTA processing (~30 s) is sufficient for complete crystallization of the films and also for reducing the oxygen vacancies; the shorter RTA processing duration also minimizes the interface reactions through the grain boundaries.

A large number of grain interiors and grain boundaries were scanned via EDX for possible differential distributions of silicon between these regions (Fig. 6). The solid line in Fig. 6 represents the film that was subjected to RTA treatment at a temperature of 800°C in oxygen gas for 30 min, and the dotted line denotes the film that was subjected to RTA treatment for 30 s at a temperature of 800°C in oxygen gas. Typical EDX tracings indicate that silicon was detected both in the grain boundary and in the grain interior; the silicon distribution at the grain boundary was larger than that within the grain. The EDX distribution profile also shows that the silicon concentration in the Ta_2O_5 films that were subjected to RTA processing for 30 s is very low, in comparison to that of films that were subjected to RTA processing for 30 min.

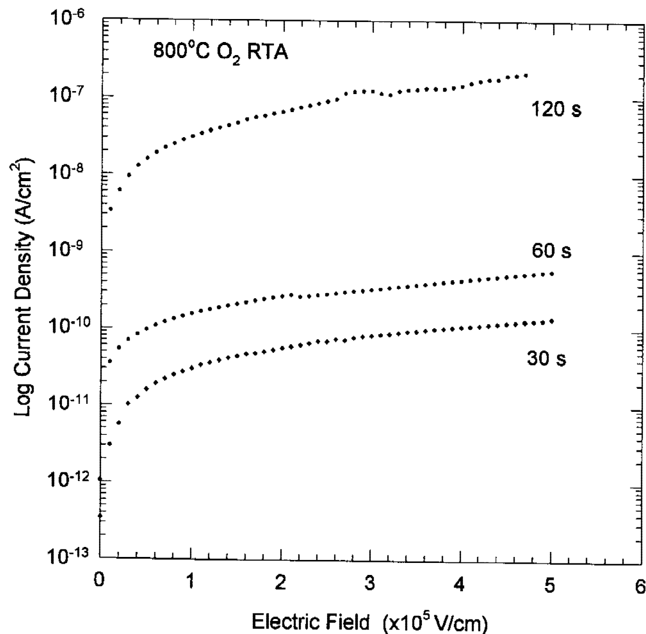

Fig. 5. Leakage current density versus applied electric field for Ta_2O_5 film after RTA processing at 800°C in oxygen gas for different time durations.

SIMS was performed on a set of Ta_2O_5/Pt/SiO_2/*n*-Si samples to confirm the presence of silicon contamination in the Ta_2O_5 films that were subjected to RTA processing at high temperatures. Figure 7 shows the SIMS depth profile of the Ta_2O_5 film on a Pt/SiO_2/*n*-Si substrate. The dashed curve in the figure represents as-deposited Ta_2O_5 film, the solid curve represents the Ta_2O_5 film that was annealed at a temperature of 800°C for 30 min in oxygen gas, and the dotted curve represents the film that was subjected to RTA processing at a temperature of 800°C for 30 s in oxygen gas. A trace amount of silicon was detected in the RTA-processed film. The film that was processed via RTA for 30 min in oxygen gas shows a higher silicon concentration and a lower oxygen concentration. Also, the silicon concentration at the interface is increased after annealing and it further extends into the Ta_2O_5 film. In contrast, the film that was subjected to RTA processing for 30 s shows a lower concentration of silicon at the interface and in the film, in comparison to the Ta_2O_5 film that was processed via RTA for 30 min. The reason for this difference is the underlying SiO_2/*n*-Si substrate, from which silicon probably diffuses into the Ta_2O_5 film through the very thin layer of platinum (1500 Å (150 nm)) during the high-temperature annealing. This result suggests that Ta_2O_5 crystallizes rapidly and silicon diffuses along the grain boundary and further penetrates into the grain. This result also suggests that the diffusion process of silicon is dependent on the temperature and the time duration of RTA processing. The details of the oxidation of these diffusing silicon atoms—which results in an increased concentration of oxygen vacancies, thereby increasing the leakage current of the polycrystalline Ta_2O_5 films—were reported in our earlier paper.[23] Because the presence of silicon is the only definite difference between the amorphous and polycrystalline Ta_2O_5 films, we have given more attention to silicon, to clarify the leakage current that flows in polycrystalline Ta_2O_5 films.

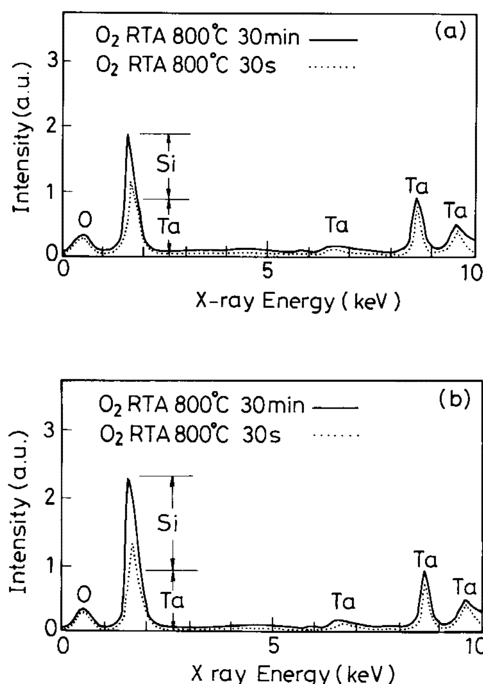

Fig. 6. EDX spectra for a Ta_2O_5 thin film subjected to RTA processing at 800°C for 30 s ((a) grain interior and (b) grain-boundary region).

Impurity constituents that have limited solubility (e.g., silicon) help to form coherent grain-boundary layers and will help to increase the density of trapping states, which influences the field dependence of the potential barrier very strongly at high electric fields and results in increased current density. This mechanism is very similar to that which occurs at the grain boundaries in classical varistor materials.[28] The Si^{4+} anion that substitutes into the Ta^{5+} site prefers a four-coordinated tetrahedral site and is expected to form an acceptor state. Lau *et al.*[29] reported the possible, energetically most-shallow defect state (0.3 eV) in Ta_2O_5 on silicon capacitors, where its density increases as the post-deposition annealing temperature increases. This defect state is related to the silicon contamination from the silicon substrate. This observation is consistent with the present results, because our SIMS and EDX results clearly indicate the presence of silicon, both in the grain interior and in the grain-boundary regions. The significance of the present observation is as follows. The silicon concentration is increased by the RTA processing of the sputtered films in oxygen gas for long durations (≥30 min). The diffused silicon atoms are then oxidized by the oxygen that is contained in Ta_2O_5 itself and is also supplied from the gas phase, which results in Ta_2O_5 films with an increased concentration of oxygen vacancies, as shown in the following defect equation:

$$2SiO_2(s) \xrightarrow{Ta_2O_5} 2Si'_{Ta} + V_O^{\cdot\cdot} + 4O_O \quad (1)$$

Therefore, the presence of silicon is partially charge compensated by the oxygen vacancies, whereas the remaining oxygen vacancies continue to participate in enhancing the leakage current of the films that were annealed for long durations. In the case of very-short-duration (30 s) RTA processing of Ta_2O_5 films in oxygen gas, the amount of silicon diffusion is restricted to a smaller level and annealing in oxygen gas decreases the number of oxygen vacancies, thereby reducing the leakage current density.

(3) Dielectric Constant

The dielectric constants of the Ta_2O_5 films were calculated from the accumulation capacitance at a frequency of 100 kHz, the known area of the platinum electrodes of the MIM capacitor, and the ellipsometrically determined oxide thickness. Figure 8 shows the dielectric constants and dielectric loss of Ta_2O_5 films as a function of RTA temperature for a duration of 30 s in an ambient oxygen-gas atmosphere. The effective dielectric constant of Ta_2O_5 film that has been annealed at a temperature of 800°C is larger than that of films that have been annealed at temperatures of 600° or 700°C. Similarly, the loss factor shows

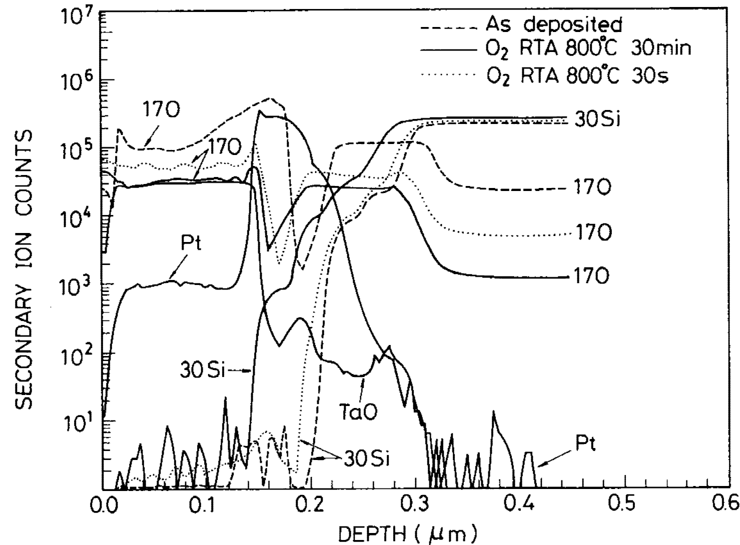

Fig. 7. SIMS depth profiles of an amorphous Ta_2O_5 thin film and polycrystalline Ta_2O_5 thin films.

an increase, from 0.01 to 0.017. This observation suggests that the fully crystalline film has a larger dielectric constant than the partially crystallized film. Furthermore, increasing the RTA processing time for a given RTA temperature results in an increase of the dielectric constant and the dielectric loss factor (from 0.012 to 0.035), as shown in Fig. 9. However, increasing the RTA processing time to >30 s increases the leakage current density (see Figs. 5 and 7). Hence, the best RTA processing time, ~30 s, was optimum for MIM Ta_2O_5 capacitor films to obtain a higher dielectric constant (52), yet preserving a relatively low leakage current density of 10^{-10} A/cm^2 at a stress field of 100 kV/cm.

Quite frequently, the dielectric constant that is reported for Ta_2O_5 film is ~25–35. However, dielectric constants of >35 have also been reported in the literature.[30,31] For example, Wu *et al.*[30] reported that their Ta_2O_5 films had a dielectric constant in the range of 20–45. Pignolet *et al.*[19] reported that a sample that was subjected to RTA processing in oxygen gas exhibited a dielectric constant of 45, and Lau *et al.*[18] observed that films that were subjected to RTA processing in oxygen gas exhibited a dielectric constant of 40. Treichel *et al.*[31] reported that the dielectric constant is a function of the film thickness and the annealing parameters; they also reported that, for relatively thick Ta_2O_5 films (>60 nm), a dielectric constant as high as 40 was possible.[31] It should be noted that the Ta_2O_5 films that have been used for the present study have a thickness of ~100 nm. Therefore, it may be realized that the dielectric constant of 52 that has been obtained for the present Ta_2O_5 films is higher than that which is conventionally observed.

(4) Time-Dependent Dielectric Breakdown

Time-dependent dielectric breakdown (TDDB) is also called resistance degradation of dielectrics; this process shows a slow increase of leakage current under a dc field stress. TDDB is a characteristic of the intrinsic materials, the procedures, and the quality of the processing and electrode materials.[32,33] Lifetime extrapolation, by using constant voltage-stress time-dependent dielectric-breakdown studies, predicts a lifetime of 10 years at

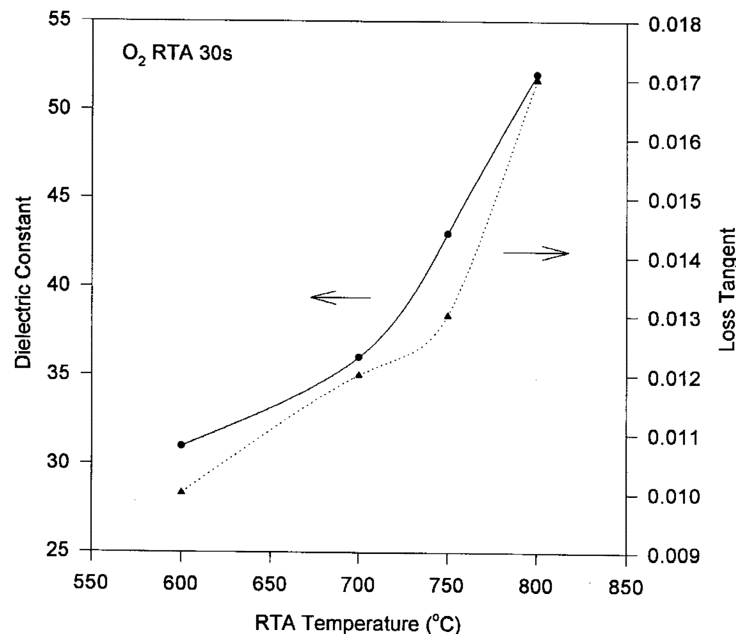

Fig. 8. Plot of effective dielectric constant of Ta_2O_5 film versus RTA temperature.

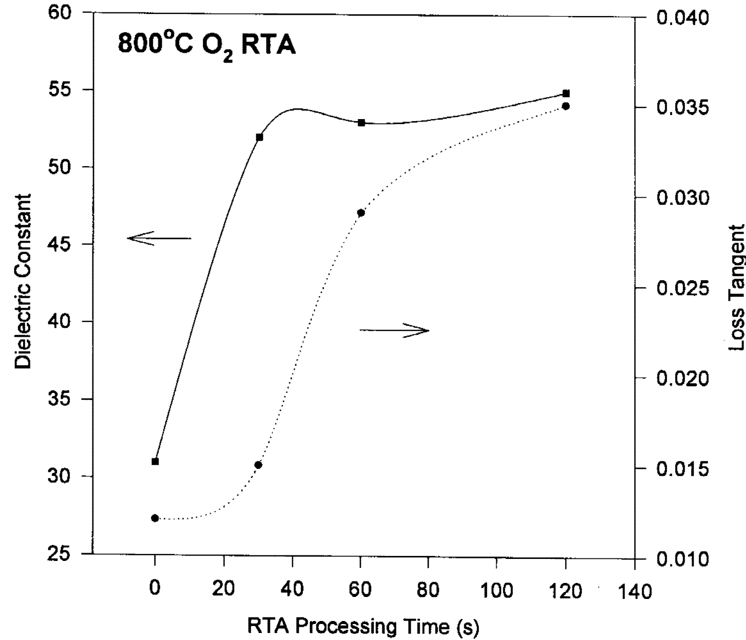

Fig. 9. Plot of effective dielectric constant of Ta_2O_5 film versus RTA processing time.

an operating voltage of 1.5 V. This result indicates that very-short-duration (30 s) RTA-processed film has a longer lifetime than that of film that has been annealed for a long duration (30 min). Figure 10 shows the lifetime extrapolation from the dependence of cumulative failure on TDDB stress time for Ta_2O_5 films that have been subjected to RTA processing for 30 min in oxygen gas. The plotted points follow straight lines, and random failure modes are not observed, which indicates that the sputtered MIM Ta_2O_5 capacitors are of high quality and good uniformity. Repeated measurements for more films demonstrate that the films that were annealed for a long time (30 min) at a temperature of 800°C have the shortest breakdown time. Also, the breakdown time for amorphous or partially crystallized films (such as that which was subjected to RTA processing at a temperature of 600°C for 30 min in an oxygen gas) is longer than that of films that were subjected to RTA processing for 30 min in oxygen gas for temperatures of 700° or 800°C. Higher temperature and longer RTA processing times lead to more grain growth (Fig. 2) and higher silicon diffusion into the grain and the grain-boundary region (Fig. 3), which results in increased leakage current density; these factors are considered to be responsible for the shorter TDDB lifetimes. TDDB lifetime characteristics for the Ta_2O_5 films that were subjected to RTA processing for a very short time (30–120 s) in oxygen gas are shown in Fig. 11. The Ta_2O_5 film capacitor that was subjected to RTA processing for 30 s in oxygen gas seems to have a longer lifetime than films that were subjected to RTA processing for 30 min in oxygen gas. The extrapolated long-term lifetime indicates that Ta_2O_5 MIM capacitors that are subjected to RTA processing for 30 s at a temperature of 800°C in oxygen gas can survive 10 years at a stress field of 1.5 MV/cm.

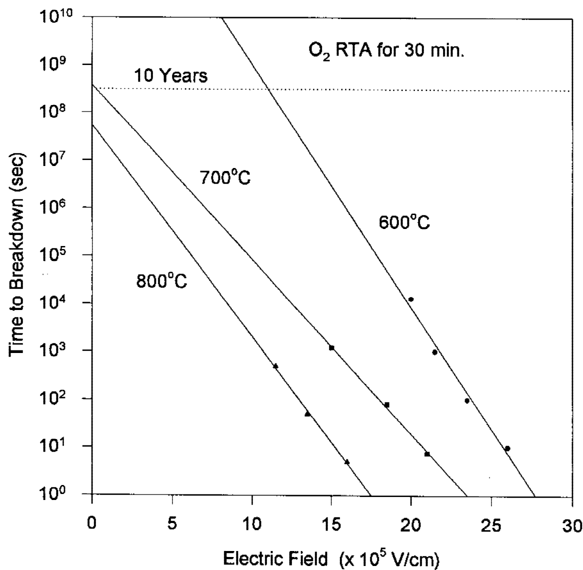

Fig. 10. TDDB lifetime, as a function of electric field, for Ta_2O_5 films processed via RTA in oxygen gas for 30 min at different temperatures.

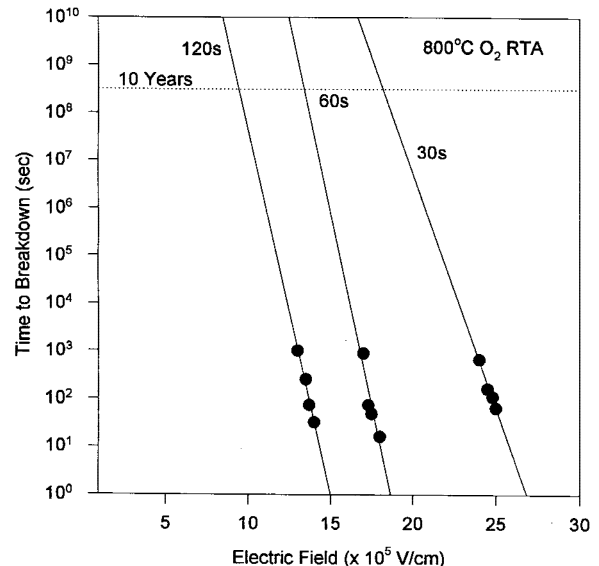

Fig. 11. TDDB lifetime, as a function of electric field, for Ta_2O_5 films processed via RTA in oxygen gas at 800°C for 30–120 s.

IV. Conclusions

We have investigated the electrical characteristics of tantalum oxide (Ta_2O_5) thin films that have been deposited on Pt/SiO_2/*n*-Si substrates (forming a metal–insulator–metal (MIM) planar capacitor structure) and their improvement via very-short-duration high-temperature rapid thermal annealing (RTA) processing, from the standpoint of application to ultra-large-scale integrated (ULSI) circuits. Present results demonstrate that a longer processing time (~30 min) results in increased leakage current density, whereas a shorter processing time (30 s) leads to lower leakage characteristics. Very-short-duration RTA processing is sufficient for the complete crystallization of the films; yet, it minimizes the interface reactions through the grain boundaries. The decrease in leakage current density is attributed to the reduction of oxygen vacancies and the suppression of silicon diffusion from the SiO_2/*n*-Si substrate into the Ta_2O_5 grain and the grain boundary, because of the shorter annealing time. Time-dependent dielectric breakdown characteristics indicate that Ta_2O_5 MIM film capacitors that are subjected to RTA processing for 30 s in oxygen gas can survive 10 years at a stress field of 1.5 MV/cm. It can be concluded in the present experiment that RTA processing at a temperature of 800°C for 30 s in an ambient oxygen-gas atmosphere is the most effective method for simultaneously maintaining a large dielectric constant and a lower leakage current density.

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