In-Situ Stress Measurement during Heat Treatment of Amorphous Tantalum Oxide

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Abstract

Tantalum capacitors are widely used in various electrical systems. Amorphous tantalum oxide (ATO) serves as the dielectric. The use of tantalum capacitors in high reliability applications generated intensive research on the electrical properties of the ATO, specifically on phenomena related to dielectric breakdown in high electric fields. Oxide defects such as interstitial oxygen ions and oxygen vacancies can significantly influence electrical and mechanical properties of ATO. Heat treatment below the crystallization temperature of ATO is known to improve its electrical properties. However, some of the details of the oxide transformation and structural change of ATO during heat treating below its crystallization temperature remain unclear. Temperature and atmosphere are the main parameters controlling the oxide transformation. Specifically, it is known that heating the as-grown, amorphous ATO to high temperatures around will result in the formation of crystallization oxide. Phase transformations and reactions in solids are typically accompanied by volume changes that can induce stresses when the material is subjected to a mechanical constraint. In Ta capacitors, this occurs because the ATO dielectric is constrained by the underlying Ta metal. In-situ stress measurements have been previously employed to explore the details of phase transformations and reactions in thin solid films. In our recent work, this approach has been used to study the amorphous-to-crystalline phase transition in ATO under high electrical fields. For example, compressive stress of 0.06 GPa was generated in the ATO along with morphological changes that lead to cracking in the dielectric. In the present study, this approach is used to examine stress evolution in the ATO during heat treatments. Temperature, atmosphere, and the temperature ramp rate were varied. In addition, scanning electron microscopy and X-ray diffraction were used to further characterize samples after a series of heat treatments.

Experimental

Preparation of the ATO/Ta film, ATO/Ta foil and high surface area ATO samples.— The 400 nm Ta films were deposited on quartz glass substrates (1 inch in diameter and 250 μm in thickness, Quartz Plus) by electron-beam evaporation (Ta pellets 99.95%, Kurt J. Lesker). The ATO films were fabricated by anodizing the Ta films on quartz substrate. For anodization, an adhesive Cu belt (5 mm wide, 50 mm long) was first attached and sealed as the electrical contact and then removed after anodization. The surface area of Ta film for anodization was 4.6 cm². Anodization was controlled using a DC power supply (Kepco BHK1000-0.2MG) with an output current of 0.3 mA. The target potential of the power supply was set to 85 V in order to obtain an ATO with a thickness of 135 nm (assuming an anodization rate of 1.6 nm/Volt).

High surface area samples were produced by pressing 7.5 g of commercial tantalum powder with a capacitance-voltage per gram rating of 12000 μF/V/gram to a press density of about 6 g/cm³. Subsequently, the samples were sintered at 1550 °C in vacuum. The pressed and sintered sample was then immersed in forming electrolyte to form the ATO with a target potential of 260 V. At 120 Hz and in a test electrolyte with a conductivity of about 30 mS/cm at T = 22 °C, a capacitance reading of about 430 μF was obtained for these samples. Using a value of 25 for the dielectric constant of the ATO, an approximate oxide thickness of 450 nm and the capacitance of about 430 μF, the internal ATO surface area of the sintered slug can be calculated to approximately 0.8 m². After ATO formation, the samples were carefully rinsed with 60 °C DI water and dried at 60 °C in a convection oven.

Stress evolution in the ATO films during hydration and different electrical fields were measured in situ by a multi-beam optical stress sensor (MOSS), as described in our previous work. The same technique has been used here to study internal stresses generated in ATO films during the annealing. Here, this approach is implemented in a high temperature vacuum system. In general, the internal stress generated in a thin film produces curvature changes (Δκ) in the thin film-substrate system. Here, the ATO/Ta layer is the thin film and quartz is the substrate. The sensor monitors the in-situ curvature change during the tests. According to Stoney’s equation, the real-time curvature change can be related to the average stress change across the thickness of film (⟨Δσ⟩):

$$\langle Δσ_f \rangle = \frac{\int_0^h Δσ_f(z)dz}{h^2} = \frac{M_β h}{6} Δκ$$

where M_β is the biaxial modulus of the substrate (87.95 GPa for quartz), z is the out of plane coordinate, h is the thickness of the thin film and substrate, respectively. By convention, negative curvature values indicate compressive stress in the film. From Eq. 1, it can be seen that the measured curvature gives the “stress thickness”.
product (i.e., \(\sigma_1 \cdot h_1\)), which in itself can be used to characterize the film stress as tensile or compressive. In addition, the stress-thickness product can be used to obtain the average biaxial stress directly if the film thickness is known.

The as-prepared 136 nm ATO films were firstly rinsed with DI water 9 times and then dried at RT. Then the samples were loaded in the furnace and annealed under a series of conditions as follows:

- **a.** Target temperature variations: 100 °C, 200 °C, 300 °C, 400 °C, 450 °C, and 500 °C with ramp rate of 5 °C per minute in air, respectively
- **b.** Ramp rate variations: 5 °C per minute and 1 °C per minute at 450 °C in air
- **c.** Atmosphere variation: air and vacuum (12 milli-Torr) with ramp rates of 5 °C per minute at 450 °C

**Other characterization methods.**—Species desorbing from the high surface area ATO samples during heating in vacuum were recorded using a mass spectrometer (Inficon, model Transpector). Two equally processed samples were heated simultaneously at a heat ramp rate of 20 °C per minute. The mass range between mass 1 and mass 100 was continuously scanned and recorded during subsequent scans. Due to the large combined surface area of the two samples (> 1.5 m²), a good signal-to-ratio was ensured in these measurements.

The leakage currents through the ATO dielectric were tested in forming electrolyte before and after the annealing. The electrical potential was controlled using a DC power supply (Kepco BHK1000-0.2MG). The leakage current passing through the ATO was recorded using a current meter (Agilent 34970A Data Acquisition Unit).

After annealing under the above set of conditions, the surface morphologies of ATOs were characterized by scanning electron microscopy (SEM, Zeiss LEO-1530 VP at 10 kV). The phase change of ATO/Ta film after being annealed was characterized by X-ray Diffraction (XRD Siemens D-5000).

### Results and Discussion

Fig. 1a–1f show the stress signals recorded during annealing for 5 ½ hours at 100, 200, 300, 400, 450 and 500 °C. In general, compressive stresses were generated during the temperature ramp-up and tensile stress-thicknesses during cool-down periods in all samples. The stresses during heating and cooling are mainly due to the thermal expansion mismatch between tantalum metal and quartz of 6.5E-06 and 0.6E-6, respectively. The sign and magnitude of the net stress due to heat-treatment in a sample can be calculated from the difference in the stress-thickness products after cool-down and at the start of the heat ramp. Net tensile stress-thicknesses were generated in samples heated to temperatures between 200 and 400 °C. This tensile stress-thickness is likely due to oxide densification accompanied by the desorption of the hydration species in the ATO during heat-treatment. The thermal desorption spectrum in Figure 2 shows the species desorbing from the as-formed ATO in this temperature range. It is clear from Figure 2a that the as-formed ATO releases significant amounts of water between 300 and 400 °C. The high desorption temperature indicates that the water is fairly strongly bound in the as-formed oxide. Figure 2b demonstrates that significantly lower water amounts are

![Stress-thickness evolution of ATOs heated in air to different temperatures](image)

**Figure 1.** Stress-thickness evolution of ATOs heated in air to different temperatures (a) 100 °C (b) 200 °C (c) 300 °C (d) 400 °C (e) 450 °C (f) 500 °C respectively.
desorbing from the ATO after annealing at 450 °C in air and recharging to the initial formation potential of 260 V in formation electrolyte for 2 h. Therefore, heat-treating the oxide in air at 450 °C permanently changes the oxide structure such that less water is admitted into the oxide upon subsequent hydration and charging in aqueous electrolyte.

Up to a temperature of 300 °C, the net tensile stress-thickness increases with increasing annealing temperature. From Figure 1 and Table I, the net tensile stress-thicknesses increase from 4 GPa±nm at 100 °C to 14 GPa±nm at 200 °C and 120 GPa±nm at 300 °C. It is known that little if any new surface oxide grows on evaporated Ta films held at temperatures below 350 °C in air. One mechanism that has been documented in this temperature range is oxygen diffusion from the metal-oxide interface into the substrate.32 This is potentially consistent with a maximum in the tensile stress-thickness as the temperature approaches 350 °C, i.e., where new oxide growth due to uptake from ambient air is not yet significant. According to Giber and Ochsner,33 oxygen diffusion rates into bulk Ta become appreciable at 300 °C, with values around 9×10^14 per cm^2 per hour. Oxygen diffusion rates may be even higher in the E-beam evaporated Ta substrates used in this study, because, as noted by Steidel and Gerstenberg,32 dense, sputtered films have an increased capability for oxygen uptake over bulk bcc tantalum. In proportion to the oxygen diffusion rate into the underlying tantalum substrate, oxygen vacancies are generated in the oxide at the metal-oxide interface. Hence, the thin, densely sputtered films will have elevated concentrations of oxygen vacancies with increasing heat-treatment temperatures. Due to the low oxygen diffusion coefficient in Ta_2O_5 at temperatures below 350 °C,34 these vacancies are only to a small extent refilled with oxygen diffusing in from the air interface and remain “frozen” at the interface after cooling. According to JCPDS card # 00-018-1304, the XRD spectra in Fig. 3 indicate that heat-treating at 300 °C in air promotes the growth of small crystalline entities in the ATO, likely at the ATO-tantalum interface. The thermal oxidation of tantalum produces substoichiometric TaO_y crystalline phases at the metal-oxide interface, formed due to and with the diffusing oxygen.

Upon increasing the annealing temperature to 400 °C, the net tensile stress-thickness reduces from 120 GPa±nm to 76 GPa±nm (Fig. 1d). This is likely due to the additional oxide formation with oxygen taken up by the ATO from the ambient air during heat-treatment. Hence, the tensile stress-thickness signal due to the “thinning out” of the oxide at the metal-oxide interface is partially compensated by compressive stress due to oxide growth at the oxide-air interface at 400 °C. Upon increasing the annealing temperature further from 400 °C to 450 °C and

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**Table I.** The leakage currents of ATO before and after annealing at different temperatures and the corresponding net stress after the respective annealing.

| Temperature (°C) | 100  | 200  | 300  | 400  | 450  | 500  |
|-----------------|------|------|------|------|------|------|
| Net stress thickness (GPa±nm) | 4    | 14   | 120  | 76   | 103  | 295  |
| Original current (μA) | 25   | 25   | 25   | 25   | 25   |
| Current after annealing (μA) | 13.2 | 11.5 | 4.9  | 8.4  | 11.2 | 17.5 |

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**Figure 3.** XRD of ATOs after heat-treatment at different temperatures in air.
500 °C, increasing net compressive stresses are generated in ATOs. This is shown in Fig. 1e and 1f. The increasing net-compressive stress goes along with increasing thermal oxide growth in this temperature region which requires oxygen uptake from the ambient atmosphere.

In the temperature range between 300 °C and 500 °C, the concentration and the diffusivity of neutral oxygen vacancies increase rapidly. After diffusing through the oxide layer to the oxide-air interface, oxygen vacancies promote the uptake of oxygen from the ambient atmosphere. Hence, the slopes and slope changes of the stress-thickness over time curves recorded during the constant temperature time window are directly related to the prevailing diffusion processes at the metal-oxide interface and in the oxide:

i) **Positive slope.** A positive slope indicates that diffusion from the oxide into the substrate base metal is the dominant process. Concurrently, oxygen vacancies are generated in the oxide at the metal-oxide interface.

ii) **Zero slope.** A flat- or zero slope either indicates a low overall concentration of vacancies or a balance between tensile stress-thickness from vacancy generation and compressive stress from oxide formation at the oxide-air interface. Importantly, oxygen diffusion into the base metal is by necessity accompanied by substoichiometric oxide formation in the substrate. This in turn creates compressive stress with increasing oxygen density.

iii) **Negative slope.** A negative slope is due to new oxide formation at the oxide-air interface being the dominant process.

The net-stress in the samples before and after heat-treatment is similarly informative. Here, a net tensile stress-thickness indicates that oxygen diffusion from the oxide into the base metal dominates with vacancies remaining “frozen” at the metal-oxide interface after cooldown. A net compressive stress indicates new oxide growth at the oxide-air interface dominates.

Indeed, inspection of Figure 1a reveals a largely flat slope in the constant temperature time window, likely due to very low to non-existent generation of oxygen vacancies at 100 °C. In Figure 1b, a small, but rapid slope increase is followed by an almost flat, but still slightly increasing slope at the target heat-treat temperature of 200 °C. This likely indicates that oxygen diffusion into the base metal and concurrent vacancy generation in the oxide is rapid initially and slows down as the oxygen density in the metal adjacent to the metal-oxide interface increases. Essentially the same is observed in Figure 1c for a heat-treat temperature of 300 °C. A rapidly increasing slope corresponding to tensile stress-thickness is observed first. This is followed by a gradual slope increase, indicating the slowing of oxygen diffusion into the metal and the buildup of substoichiometric oxide producing compressive stress. Increasing the temperature further to 400 °C (see Figure 1d) changes the stress balance in favor of compressive stress, due to new oxide formation. However, closer inspection of Figure 1d reveals that new oxide is evidently only formed after a delay time. Likely, this delay corresponds to the time that vacancies need to diffuse through the oxide. Eventually, at higher temperatures, new oxide growth prevails, due to high concentrations of neutral vacancies at the oxide-air interface and the increased diffusivity of oxygen in the oxide at temperatures of 400 °C and above. The time required for vacancy diffusion through the 135 nm oxide decreases rapidly with increasing temperature so that at 450 °C and 500 °C, new oxide growth is already setting in even before the target temperatures are reached (Figure 1e and 1f, respectively). The sharp stress reversal feature in Figure 1e, right before the constant temperature time window is reached (i.e., while the oven temperature is still increasing), is due to oxygen diffusion into the base metal and the subsequent onset of strong oxide growth at the oxide-air interface: The slowing oxygen diffusion into the base metal produces the weakening tensile stress-thickness signal. Diffusion of large vacancy concentrations through the oxide subsequently promote the strong oxide growth which in turn produces the compressive stress signal. Slowing down the heat-ramp rate from 5 °C per minute to 1 °C per minute will therefore simply lengthen the time required for the stress reversal. This is demonstrated in Figure 4c: With the slow rate, approximately 10000 sec are required to produce the stress reversal. Only about 1000 sec are needed when a fast ramp rate is used (Figure 4a) The stress reversal is also apparent upon heating to 500 °C (Figure 1f). At 500 °C, vacancy diffusion is so rapid that tensile stress-thickness due to vacancy generation at the metal-oxide boundary and compressive stress due to oxide growth at the oxide-air interface are balanced for a short time before the oxide growth due to oxygen uptake becomes dominant.

The comparison of the stress-thickness time diagrams obtained upon heating the sample to 450 °C in air and in vacuum in Figure 4 is consistent with this picture: No net new oxide growth
can be formed in vacuum. Hence, the net stress after cool-down is tensile in vacuum, even after heating to 450 °C, due to the generation of oxygen vacancies and compressive in air due to the generation of new oxide. The detail of the stress signal in the constant temperature time window is also consistent: new oxide growth causes the compressive stress at 450 °C in air. A small amount of substoichiometric oxide growth in the substrate metal causes the slightly compressive stress observed at 450 °C in vacuum.

The leakage currents of ATOs after heat treating at the different temperatures are listed in Table I. The leakage currents through the ATO decrease after heat-treating at 100 °C, 200 °C and 300 °C in air from 13.2 μA to 11.5 μA to 4.9 μA respectively. Evidently, dehydration of the ATO and generating neutral oxygen vacancies at the metal oxide interface reduces the leakage current. According to Smyth et al.,19,20 the reason for the decrease in leakage current is a heat-treatment-induced shift in the conductivity profile through the dielectric: As discussed above, heat-treatment produces neutral oxygen vacancies at the metal-oxide interface. Due to the two electronic charges associated with each vacancy, the effect of the neutral vacancies is analogous to creating n-type semi-conductivity. Hence, a conductivity-profile is established across the dielectric, with high conductivity at the metal-oxide interface and low conductivity at the oxide-air interface. According to Smyth et al.,20 the overall change in conductivity is on the order of 1020–1025. Applying a bias voltage with our thermal desorption experiment (Figure 2) suggests that in addition to this well-known explanation by Smyth et al.,20 the dehydration of the dielectric may also be significant. In analogy to the observed increase in the electric conductivity of the aluminum oxide dielectric upon hydration, the electrical conductivity of hydrated tantalum pentoxide is likely higher than that of non-hydrated or de-hydrated oxide. Therefore, dehydration will also decrease the leakage current. Figure 2b demonstrates that once the oxide is dehydrated, subsequent re-hydration does not occur due to the permanent structural change in the oxide. The observed increase in leakage current from 4.9 μA upon increasing the heat-treatment temperature from 300 °C to 500 °C is likely due to an increasing number of oxide distortions and cracks. In turn, these distortions are likely produced by the strong compressive stress due to the new oxide formation at the oxide-air interface. The increasing formation of oxide distortions upon increasing the heat-treatment temperature is demonstrated in the SEM images in Figure 5. No such distortions are observed upon heating to high temperatures around 450 °C and above in vacuum. In addition, it is known that the originally amorphous oxide has an increasing propensity to crystalize at higher temperatures with a concurrent increase in the electronic conductivity.

Conclusions

In-situ stress measurements were used for the first time to evaluate changes in amorphous tantalum oxide films during heat treatments. The majority of the stress effects observed on the heat-treated ATO/Ta systems are explained on the basis of oxygen vacancy generation and oxygen uptake mechanisms at the metal-oxide and the oxide-air interfaces, respectively. For heat-treatments up to 300 °C, tensile stress-thickness is generated in the ATO. The tensile stress-thickness is due to dehydration of the oxide at the air interface and oxygen vacancy creation in the oxide at the metal-oxide interface. Direct evidence for the dehydration of the anodically grown tantalum pentoxide dielectric during heat-treatment is, to our knowledge, demonstrated for the first time in this paper. Upon heating to 400 °C and above, the ATO begins to take up oxygen from the ambient air, producing an increasingly net-compressive stress-thickness. The compressive stress-thickness is accompanied by an increase in leakage current and oxide surface distortions.

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Figure 5. SEM images of ATOs after heat-treating in air at different temperatures (a) 300 °C (b) 400 °C (c) 450 °C (d) 500 °C respectively.
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