Properties of Ta$_2$O$_5$ thin films prepared by ion-assisted deposition

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**Abstract**

Tantalum penta-oxide (Ta$_2$O$_5$) thin films were deposited onto highly polished and clean, fused silica glass substrates via ion beam-assisted deposition at room temperature using a high-vacuum coater equipped with an electron beam gun. The effects of ion beam parameters, oxygen flow rate, and deposition rate on the optical and structural properties as well as the stress of Ta$_2$O$_5$ films were studied. It has been revealed that Ta$_2$O$_5$ thin films deposited at 300 eV ion beam energy, 60 $\mu$A/cm$^2$ ion current density, 20 sccm oxygen flow rate and 0.6 nm/s deposition rate demonstrated excellent optical, structural and compressive stress.

1. Introduction

Ta$_2$O$_5$ thin films are characterized by excellent properties among which are high dielectric constant, high refractive index in the visible range and wide transmittance spectra region from 350 to 1000 nm. Due to their superior chemical stability, Ta$_2$O$_5$ thin films were additionally assessed from a high temperature resistance perspective. More recently, this material has received significant attention for its property as a capacitor [1] and having a high refractive index in interference coatings [2,3]. Ta$_2$O$_5$ thin film deposition was accomplished using different techniques such as electron-beam evaporation [4], ion-assisted deposition [5], reactive magnetron sputtering [6], and ion beam sputtering [7]. Various film characteristic properties were reported after employing these methods [8–10].

Ion-assisted deposition which involves ion bombardment of the surface is applied in connection with thin film deposition for decades. The amount and type of ion bombardment greatly influence film structure and composition. This, in turn, determines the optical, mechanical and electrical properties of the film. Bombardment can be achieved through several techniques, some of which are very simple. Despite the effortlessness of the techniques, the highest degree of critical parameter controls like ion flux, energy, species, and angle of incidence could only be attained with a broad beam Kaufman ion source [11–13]. This method has the advantage that the ion source gas discharge may be selectively separated and removed from the rest of the deposition system – something not applicable with other ion bombardment techniques utilized in film deposition. For this reason, the broad beam Kaufman ion source was selected for this research.

In the present study, the effects of ion beam parameters, oxygen flow rate and deposition rate on the optical and structural properties of Ta$_2$O$_5$ films were examined. An analysis of residual stress is also reported herein.

2. Experimental details

Ta$_2$O$_5$ thin films were deposited onto well cleaned, fused silica glass substrates at room temperature by ion beam-assisted deposition using a high vacuum coater equipped with an electron beam gun and a Kaufman ion source. Ta$_2$O$_5$ pellets were of high purity (99.99%) and served as the source material for evaporation. This study was conducted in a fully automatic turbo-pumped coating chamber outfitted with resistive sources, 6-pocket 270°, and 14 kW electron beam gun (E-gun) with the deposition pocket centered in the chamber. The deposition rate and film thickness were monitored and automatically controlled by a quartz crystal sensor linked to an electron beam power supply. Oxygen gas flow was managed by a mass flow controller. A Faraday probe placed into the tooling around the outer radius to characterize the Kaufman ion source. The basic equipment setup is shown in Fig. 1. The pressure during the evaporation process was roughly $5 \times 10^{-5}$ mbar, and was initiated at an ambient temperature of about 27°C. Nevertheless, the substrate temperature rose slightly as a result of the thermal energy from the E-gun and ion source.
During deposition, the ion current density and ion energy were varied from 10 to 80 μA/cm² and 100 to 400 eV, respectively.

Optical transmittance was set in the 350–2500 nm range and measured with a Perkin-Elmer Lambda 19 spectrometer. The films’ refractive index and extinction coefficient were calculated with an envelope technique using the transmission spectra [14]. The residual stress, σf, in the oxide thin films, was evaluated by simulating the change in curvatures before and after deposition, and was determined using Stoney’s Equation (1) [15].

\[
\sigma_f = \frac{E_s \cdot t_s^2}{6(1 - \nu_s)} \cdot \frac{t_f}{R}
\]

where \(E_s\) and \(\nu_s\) are the Young modulus and Poisson ratio, respectively, \(t_s\) and \(t_f\) are the thickness of the substrate and film, respectively, and \(R\) is the spherical radius of the curvature in the disk-shaped glass substrate. The expression is valid for film that is substantially thinner than the substrate.

X-ray diffraction measurement (Cu Kα, 40 kV, 20 mA) was used to obtain the crystal structure. The diffraction angle 2θ was scanned in a range of 20–80° with a step of 0.05°, while the Ta₂O₅ film grain size was estimated in accordance to the Scherrer function.

3. Results and discussion

3.1. Optical analysis

The refractive index (n) values at λ = 500 nm for Ta₂O₅ coatings bombarded with 200, 300 and 400 eV oxygen ions are plotted in Fig. 2 as a function of oxygen ion current density.

The plot demonstrates that the energy from each type of bombarding ions follows a distinguished trend. For instance, at 200 eV the refractive index seems to be at a standstill. However, at 300 eV it increases tremendously from 2.18 to 2.27 and vice versa for 400 eV, whereby the refractive index diminishes at a higher ion current density.

The refractive index versus ion current density trend is considered, especially for 60 μA/cm². When the coating is bombarded with 200 eV, the refractive index remains at 2.18. The value seems to increase for greater bombarding energy, something obvious at 300 eV. On the other hand, the index drops if the energy is further increased to 400 eV.

The rising refractive index (n) values relative to the increasing oxygen ions current density indicates that ion bombardment during deposition modifies the films’ columnar microstructure growth, resulting in film densification. The result indicates that the coatings bombarded with 300 eV oxygen ions had larger values of n than those bombarded with 400 eV oxygen ions. The diminishing refractive index at 400 eV is potentially due to the degrading effect that the highest ion bombardment energy has on the coating’s refractive index. The result in Fig. 2 indicate that only minor densification occurs for coatings bombarded with 200 eV oxygen ions during deposition.

A comparable dependence of the refractive index on bombarding ion energy has been reported for ion-assisted CeO₂ films [16]. The current density value (for fixed ion energy) at which the maximum refractive index occurs is termed the “critical value.” The results in Fig. 2 illustrate that the film index values for ion current density decrease after exceeding a critical value, i.e. 60 μA/cm². An explanation for this may be the result of degrading film stoichiometry, the appearance of closed isolated voids, or oxygen incorporation into the films. The largest decrease is noted for 400 eV ion bombardment, but the decline is less significant for 200 and 300 eV ions in the case of Ta₂O₅. This energy dependent refractive index drop is consistent with the dependence of the average ion penetration depth and preferential sputtering yield on ion energy. Similar results with decreasing refractive index values for current densities beyond the critical values have been reported for ion-assisted ZrO₂ [17] and CeO₂ films [16].

Fig. 3 contains the plotted graphs of extinction coefficient, k, versus ion current density for ion bombardment at 200, 300 and
400 eV. The values of $k$ were calculated at $\lambda = 500$ nm for an average thickness of 250 nm. It is noted that the lowest value of $k$ at $2.0 \times 10^{-4}$ is too small to be deemed reliable due to the minimum sensitivity of the measurement technique. Generally, when increasing the ion current density, all experimental energies exhibit rising $k$ values that reflect relatively good optical characteristics. The values seem to consistently increase beyond the “critical value” of 60 $\mu$A/cm$^2$.

Fig. 4 illustrates the film’s residual stress plotted against oxygen ion current density. Only selected bombardment energies at 300 and 400 eV are considered. It is apparent that the stress type is compressive and gradually increases with increasing levels of ion current density. A slight reduction is obvious though at an ion current density of 60 $\mu$A/cm$^2$ before it begins to increase again. In another study, Hoffman and Gaerttner [18] bombarded evaporated Cr films during deposition with 11.5 keV argon ions. A similar trend was observed, whereby tensile stress changed to compressive, and it kept consistently increasing with higher ion bombardment. The researchers attributed the results to modifications in film microstructure.

The influence of film thickness on Ta$_2$O$_5$ thin film stress deposited at 300 eV and 60 $\mu$A/cm$^2$ is visible in Fig. 5. The film stress is approximately constant; this behavior is potentially attributed to the development of void fractions in the films deposited at critical ion beam parameter values. Clearly, film thickness beyond 200 nm has less influence on film stress, which is in accord with other reported results [19].

The same deposited Ta$_2$O$_5$ thin film was further characterized. The dispersion data of the films at $\lambda = 500$ nm deposited at various oxygen flow rates are shown in Fig. 6. Here, the extinction coefficient reaches a minimum upon approaching 20 sccm oxygen flow rate, and starts to increase again beyond this value. However, although the refractive index initially decreases with oxygen flow rate, it later becomes constant beyond 20 sccm. This is attributed to the incorporation of oxygen into the grain boundaries as opposed to the modification of the coating’s columnar microstructure.

The variation in refractive and extinction coefficient at $\lambda = 500$ nm for Ta$_2$O$_5$ films deposited at the critical ion beam parameter values (i.e. 300 eV, 60 $\mu$A/cm$^2$) as a function of deposition rate is depicted in Fig. 7. It is clearly shown in the figure that the deposition rate has a marginal effect on both refractive index and extinction coefficient. This behavior could be explained on the basis that the coatings bombarded during deposition at the 300 eV and 60 $\mu$A/cm$^2$ exhibit good optical characteristics.
Table 1: Grain size of Ta2O5 films determined from XRD spectrum using Scherrer function.

<table>
<thead>
<tr>
<th>Ion energy (eV)</th>
<th>Grain size from (100) peak (nm)</th>
<th>Grain size from (101) peak (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>Amorphous</td>
<td>Amorphous</td>
</tr>
<tr>
<td>100</td>
<td>32</td>
<td>40</td>
</tr>
<tr>
<td>200</td>
<td>130</td>
<td>140</td>
</tr>
<tr>
<td>300</td>
<td>141</td>
<td>150</td>
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Elevating the ion energy facilitates the enhancement of coating surface mobility and atom diffusion into the nucleation sites, thus increasing the coating’s crystallization. It is apparent that the samples’ grain size enlarges at higher ion energies. Finally, the improved grain size in the thin films is attributed to the oxygen ion bombardment during deposition, which strongly oxidizes substrates to form stoichiometric oxides by removing certain defects and oxygen vacancies.

4. Conclusion

Ta2O5 thin films were prepared on fused silica glass substrate using ion-assisted electron beam evaporation. The effects of ion-beam parameters, oxygen flow rate and deposition rate on the optical and structural properties as well as film residual stress of Ta2O5 films were investigated. The optical properties of Ta2O5 thin films were found to be strongly dependent on the ion beam energy, current density (critical parameters) and deposition rate during the deposition process. The refractive index value increased with increasing oxygen ion energy from 200 to 300 eV, after which the value decreased from 400 eV. The ion beam parameters had an effect on the film’s compressive stress. The films were stressed upon incrementing the level of ion bombardment. As per the XRD results, the thin films deposited with no ions were amorphous, whereas those deposited under ion bombardment were crystalline δ-Ta2O5. The crystallinity and grain size enlarged at higher ion energies. It can be concluded that the high refractive index, low extinction coefficient and low compressive stress of crystalline Ta2O5 film deposited at an ambient substrate temperature were feasible when using critical ion beam parameters. The identified parameters were 300 eV ion beam energy, 60 μA/cm² ion current density, 20 sccm oxygen flow rate and 0.6 nm/s deposition rate.

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