Annealing effects on the optical and electrochemical properties of tantalum pentoxide films

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Received: September 27, 2020; Revised: January 25, 2021; Accepted: February 8, 2021

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Abstract: Tantalum pentoxide (Ta_2O_5) has attracted intensive attention due to their excellent physicochemical properties. Ta_2O_5 films were synthesized via electron beam evaporation (EBE) and subsequently annealed at different temperatures ranging from 300 to 900 °C. X-ray diffraction (XRD) results show that amorphous Ta_2O_5 thin films form from 300 to 700 °C and then a phase transition to polycrystalline β - Ta_2O_5 films occurs since 900 °C. The surface morphology of the Ta_2O_5 films is uniform and smooth. The resulted Ta_2O_5 films exhibit excellent transmittance properties for wavelengths ranging from 300 to 1100 nm. The bandgap of the Ta_2O_5 films is broadened from 4.32 to 4.46 eV by annealing. The 900 °C polycrystalline film electrode has improved electrochemical stability, compared to the other amorphous counterparts.

Keywords: Ta_2O_5 films; anneal; optical property; electrochemical stability

1 Introduction

Ta_2O_5 (tantalum pentoxide), which forms at either tetragonal (α - Ta_2O_5) or orthorhombic (β - Ta_2O_5) crystalline phase, is one of the important transition-metal oxides with high refractive index ($n = 2.02$ – 2.16), high dielectric constant ($\epsilon = 20$ – 46), wide bandgap (4.0–4.5 eV), and wide transmission spectrum [1]. Ta_2O_5 thin films have been widely used in new types of optoelectronic devices, electroluminescent devices [2,3], optical waveguides [4], optical coatings [5,6], and optical filters [7], owing its excellent thermal and chemical

stability, high mechanical strength, and good resistance to friction. For examples, a protective layer of Ta_2O_5 is often coated on thermal sensors utilizing its higher heat resistance and mechanical strength in satellites. Ta_2O_5 and its ternary alloys have been widely used as passivation layer in MOS-based devices [8]. Electrode materials in modern batteries often need a Ta_2O_5 cover layer to obtain long-term electrochemical energy storage and charge-and-discharge cycle toughness [9]. The crystalline structure obtained by annealing and the reduction of oxygen vacancies can increase the laser damage threshold. Thin-film saturable absorber materials typically need a coating of Ta_2O_5 on its top surface to maintain a high laser damage threshold and sustain higher output-power in ultrafast lasers [10,11]. Because

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of its compatibility with commercial integrated circuit technology, Ta₂O₅ is a promising non-silicon dielectric material for microelectronic devices and the ideal capacitive material for next generation dynamic random-access memory [12] and multi-chip modules [13,14]. With its high acidity, Ta₂O₅ can serve as a catalyst for important processes such as the oxidation of alkanes and the pollution reduction [15]. In addition, Ta₂O₅ is an important dielectric material for thin-film capacitors, and has been successfully used for anode oxidation of electrodes in lithium batteries [16].

The fabrication of Ta₂O₅ films using various methods has been investigated intensively and the potential application in industry has been developed during the past two decades. Hudner *et al.* [17] reported the growth of β -phase Ta₂O₅ films on Si(100) via electron beam evaporation in an atomic oxygen plasma at a substrate temperature of 650 °C. Mannequin *et al.* [18] discussed the difference in the structural and chemical properties of amorphous Ta₂O₅ thin films grown via EBE and RF sputtering. Using chemical vapor deposition (CVD), Devine *et al.* [19] developed a new plasma-enhanced process to obtain high-quality amorphous Ta₂O₅ thin films, which were used as gate insulators to make p-channel MOS transistors. Using atomic layer deposition (ALD) method, Chaneliere *et al.* [3] and Lukosius *et al.* [20] characterized the structural and electrical properties of Ta₂O₅ films and explored the possibility of applying Ta₂O₅ thin films in microelectronic devices. Pulsed laser deposition (PLD) was used by Krishnan *et al.* [21] to obtain crystalline SnO₂-doped Ta₂O₅ thin films with good optical properties at relatively low temperatures. Byeon *et al.* [22] and Reddy *et al.* [23] used magnetron sputtering method to synthesize Ta₂O₅ films and investigated the possibility of Ta₂O₅ films as dielectric materials in capacitors of integrated circuits. Xia *et al.* [16] chose anodizing method to fabricate a self-supporting porous Ta₂O₅ film for a battery electrode material with a large number of charge and discharge cycles. Ta₂O₅ films were also produced via the sol–gel method [24] and polycrystalline Ta₂O₅ film formed after heat treatment above 400 °C, which decreased optical absorption. The performance adjustment of Ta₂O₅ films was made at the same time. Xu *et al.* [10] improved the oxygen vacancy problem of evaporated Ta₂O₅ film via annealing, and increased the bandgap and the laser damage resistance threshold of the film. Zhang *et al.* [25] used spectroscopic ellipsometry (SE) to characterize

the refractive index of evaporated Ta₂O₅ films with different thicknesses and studied the optical properties of ultrathin film as a function of thickness. Masse *et al.* [26] studied Ta₂O₅ thin films at different annealing temperatures ranging from room temperature up to 900 °C and found mutual diffusion of oxygen, silicon, and Ta at high temperatures. He *et al.* [27] studied the effects of oxygen pressure on film roughness and optical transmission of PLD-grown Ta₂O₅ films.

Technically, the synthesis of Ta₂O₅ film is feasible using any of the above methods. Annealing of the deposited Ta₂O₅ films is necessary to improve film crystallinity as well as the refractive index, absorption coefficient, and the electrical properties of the films [26,28–32]. In this work, EBE, a simple film-growth method, had been employed to deposit amorphous Ta₂O₅ films on quartz and Si substrates to produce optical coatings with uniform, smooth surfaces and a high film density [33–35]. However, when evaporated Ta₂O₅ films undergo high temperature annealing, the effects on the film's structural, morphological, optical, and electrochemical properties are still not clear. Therefore, our purpose is to investigate the properties of Ta₂O₅ samples at different annealing temperatures, and the research work should bear the significance to improve the long-term stability of the photocatalyst/electrocatalyst electrodes.

2 Experimental

Ta₂O₅ films were synthesized with the following procedure. The source materials were small Ta₂O₅ pieces (purity 99.99%), which were put into a pure copper crucible in a commercial high-vacuum EBE system (ZZS700, Xing Nan Technology Co., China). The experimental device is shown in Fig. 1. Optically-polished Si(100) substrates with 4 k Ω ·cm² and the thickness of 650 μ m (for the structural and morphological characterization) and quartz with the thickness of 500 μ m (for transmittance studies) were cut into pieces (10 mm $\times$ 10 mm) and cleaned (ultrasonic cleaning in acetone, alcohol, and DI water, respectively, for 15 min). They were placed on a rotating workpiece which was heated with a contact heater in the upper vacuum chamber to maintain a constant substrate temperature of 300 °C. The substrate temperature was monitored by a thermocouple with an accuracy of ± 1 °C in real time. The evaporation source-to-substrate distance

Fig. 1 Physical object and schematic diagram of the experimental device.

was ~ 130 cm, and the evaporation source-to-electron-gun-filament distance was ~ 25 mm. Before the evaporation, the substrate surface was cleaned for 120 s using an ion source. The vacuum system consists of a low-vacuum mechanical pump and a high-vacuum molecular pump. The pump and vacuum monitor can (in real time) display the vacuum level in the system pipeline and the evaporation chamber. When the evaporation chamber was pumped down below 6×10^{-4} Pa, the materials started to be evaporated.

A quartz crystal monitor (Inficon G10) was used to display the real-time film deposition rate. The target thickness value of the Ta_2O_5 film was pre-set to 50 nm twice in control panel of the EBE system (100 nm thick in total). The filament current of the electron gun was 310 mA. The film growth rate was set to be very slow (0.2–0.4 nm/s) so as to allow sufficient duration for film growth with uniform grain distribution. After the film growth was done and the chamber naturally cooled down, the samples were taken out of the chamber and subsequently annealed at 300, 500, 700, and 900 °C for 30 min in a tube furnace (with the temperature rising at 5 °C/min). After film deposition, the samples in the tube furnace are naturally cooling down in air to the room temperature with the cooling rate estimated to be about 2 °C/min.

The surface morphology of the samples was characterized using atomic force microscopy (AFM, Dimension ICON) and field-emission scanning electron microscopy (FE-SEM, Nova Nanosem 450). Phase identification was performed via XRD with the $\text{Cu K}\alpha$ radiation line ($\lambda = 1.5418$ Å, Shimadzu 7000SAS). The optical transmittance spectra of the Ta_2O_5 films with different annealing temperatures were measured via a UV–visible spectrophotometer (UV3300) for a wavelength range of 200–1100 nm. Using an electrochemical

workstation (CHI760E), the capacitive test was performed on the sample by cyclic voltammetry. The electrochemical test uses a three-electrode system for a scan rate range of 0.1–0.5 V/s, using a thin film sample as the working electrode, metal platinum as the counter electrode, and calomel electrode (SCE) as the reference electrode. The electrolyte uses 1 M NaCl electrolyte. The surface valence state is characterized by X-ray photoelectron spectroscopy (XPS, Kratos Analytical Ltd.), the binding energy was calibrated using C 1s (284.8 eV) spectrum of hydrocarbon that always remained in the XPS analysis chamber.

3 Results and discussion

Figure 2 shows the XRD diffraction patterns of the prepared Ta_2O_5 thin films on Si substrates at different annealing temperatures. When comparing these four curves (300–900 °C) with the Standard PDF Card #25-0922 [16,18,36], only 900 °C curve exhibits (001),

Fig. 2 XRD patterns of the $\text{Ta}_2\text{O}_5/\text{Si}(100)$ samples annealed at 300–900 °C.

(200), and (270) orientation peaks of orthorhombic-phase Ta_2O_5 ($\beta\text{-Ta}_2\text{O}_5$) film. The other three curves do not exhibit Ta_2O_5 peak except for a broad hump at $27^\circ\text{--}32^\circ$. The presence of this hump infers that the Ta_2O_5 films are amorphous in the temperature range of $300\text{--}700^\circ\text{C}$. A polycrystalline Ta_2O_5 film forms until 900°C . Considering that polycrystalline Ta_2O_5 films have been reported by annealing at 800°C [36], the crystallization of Ta_2O_5 films may start at less than 800°C . All the above results are consistent with previous reports [17,18,33,36,37].

Figure 3 illustrates AFM images of the planar morphology of $\text{Ta}_2\text{O}_5/\text{Si}(100)$ sample at $300\text{--}900^\circ\text{C}$ anneal. The R_a (the maximum roughness) values of these samples at $300, 500, 700$, and 900°C are $0.402, 0.865, 0.780$, and 2.263 nm , respectively; and the R_q (the root mean square roughness) values of these samples at $300\text{--}900^\circ\text{C}$ are $0.520, 1.151, 0.989$, and 3.132 nm , respectively. The overall color of the diagram for the $300\text{--}900^\circ\text{C}$ is dark brown, which indicates that the film surface is quite uniform and very dense [38]. Specifically, the light brown and white dots, representing the raised structures, are relatively small. The height errors are even within the scale of a single molecular layer, indicating the film surface has atomic flatness [39].

Figure 4 shows planar and cross-sectional SEM images and EDS mapping of Ta_2O_5 thin films at different annealing temperatures. Table 1 shows the atomic weight percentage of oxygen and tantalum in EDS mapping energy spectra. In Fig. 4(a), it exhibits that the particle size of Ta_2O_5 films decreases greatly with increasing of the temperatures (from 300 to

700°C). Significant changes occurred at 900°C , where the film surface morphology becomes texturized structure in spite of the film surface keeping flat. This phenomenon is consistent with XRD results of the Ta_2O_5 film transition from amorphous phase to crystalline phase at high temperature. Figure 4(b) measures the sample thickness values of $157.5, 105.7, 113.5$, and 116.0 nm for $300\text{--}900^\circ\text{C}$ films. According to Ref. [25], the film thickness below 40 nm could affect the optical properties of Ta_2O_5 thin film dramatically. When the film thickness exceeds 40 nm , the refractive index of the film will be close to the value of bulk Ta_2O_5 . Thus, the optical properties of the obtained films are thickness-independent. In Fig. 4(c) and Table 1, it shows the elemental distributions of Ta and O on the film surface of four films, confirming high elemental distribution uniformity of all film surfaces.

Since the optical properties of Ta_2O_5 films can reflect the variation in the crystalline quality and energy bandgap of the films, the transmittance spectra of four films on the quartz substrate are plotted in the wavelength range of $200\text{--}1100\text{ nm}$ in Fig. 5. All the Ta_2O_5 films exhibit relatively good optical transmittance over the wavelength range of $310\text{--}1100\text{ nm}$. For 300°C curve, the transmittance rate of the film is around 0.6 to 0.77 (60% to 77%) in the wavelength range of $310\text{--}660\text{ nm}$. For 500 and 700°C curves, the transmittance rates of the two samples increase to $0.75\text{--}0.82$ ($75\%\text{--}82\%$) in the wavelength range of $310\text{--}660\text{ nm}$. This phenomenon occurs probably because the optical properties of the samples were

Fig. 3 AFM images of the planar morphology of the $\text{Ta}_2\text{O}_5/\text{Si}(100)$ samples at $300\text{--}900^\circ\text{C}$ anneal.

Fig. 4 (a) Planar and (b) cross-sectional SEM images; (c) EDS mapping energy spectra of Ta₂O₅ thin films annealed at different temperatures.

Table 1 Atomic weight percentage of O and Ta in EDS mapping energy spectra

	300 °C	500 °C	700 °C	900 °C
Ta (%)	92.318	90.914	92.885	91.870
O (%)	7.682	9.086	7.115	8.130

Fig. 5 Transmittance spectra of the Ta₂O₅/quartz samples within the incidence light wavelength of 200–1100 nm, and sample color contrast illustration. The inset: $(\alpha h\nu)^{1/2}$ versus incidence photon energy of Ta₂O₅ films.

relatively not so good with low density and there are many defects in the Ta₂O₅ film at low temperature anneal [10,36]. Another reason may be that 300 °C film is thicker (157.5 nm) than the other three films (around 110 nm). Unlike Ref. [26], the transmittance spectra of 500 and 700 °C are very similar probably because the hexagonal structured Ta₂O₅ (δ -Ta₂O₅) forms at 700 °C in Ref. [26] while the film crystallization of 500 and 700 °C does not occur yet. In addition, the optical transmittance continues to substantially increase in the wavelength ranges of 300–550 nm and 700–1100 nm, from 700 to 900 °C since crystalline Ta₂O₅ phase starts to form at 900 °C and thus there are fewer defects in 900 °C film. Finally, contrary to the results in Ref. [36], where the transmittance of the prepared films decreased when the annealing temperatures increased from 600 to 1000 °C, the transmittance in Fig. 5 generally increases with temperature. The film morphology of Ta₂O₅ may explain the difference: the surface roughness values of those films at 600, 800, and 1000 °C in Ref. [36] were 9.364, 31.367, and 43.699 nm, respectively, which are much rougher than those of 300–900 °C. It is clear that the rougher the film surface is, the stronger the light scattering in the film, or the smaller the film transmittance.

Moreover, the transmittance drops rapidly to zero

from 310 to 273 nm, which is due to strong light-absorption in this region. The minimum absorption is below 273 nm. If the energy that corresponds to the 273 nm absorption edge is defined as the bandgap of the film, then, according to the formula:

$$E = hc/\lambda \quad (1)$$

The bandgap of the samples is ~ 4.54 eV, which is close to the 4.2 eV of bulk Ta₂O₅ [21]. In addition, the Ta₂O₅ film coverage reduces the substrate transmittance slightly and the film color of 300–900 °C turns to light blue from dark blue in Fig. 5. As the temperature increases, the optical transmittance of the Ta₂O₅ film generally increases. The transmittance of 900 °C film remains above 90% from 374 to 1100 nm (slightly below 90% at 500–600 nm), and reaches a maximum of 93 % at 400 nm. Thus, annealing facilitates removal of the voids and holes of thin films and the consequent improvement of the film optical quality as well as the film color.

Further, Fig. 5 shows the bandgap (E_g) variation of the Ta₂O₅ films with annealing temperatures. The film E_g value can be derived using Tauc [40] formula:

$$\left(\frac{\alpha h\nu}{K}\right)^{1/2} = h\nu - E_g \quad (2)$$

where α is the absorption coefficient, K is a constant, and $h\nu$ is the photon energy. α can be obtained using [41]:

$$\alpha = -\frac{\ln T}{d} \quad (3)$$

Here, d is the film thickness and T is the transmittance. The insets in Fig. 5 are plotted based on Eq. (2). The E_g value of 300 °C for Ta₂O₅ films on quartz substrate is 4.32 eV, while those of 500–900 °C increase slowly from 4.32 to 4.46 eV. The E_g values of 300–900 °C for Ta₂O₅ films on Si substrate are from 4.46 to 4.51 eV. The slight increase in the band gap of Ta₂O₅ film on Si substrate is related to the weak diffusion of Si into Ta₂O₅ film forming SiO₂ and/or tantalum silicate [10,21].

Annealing processing also affects the electrochemical properties of Ta₂O₅ films. In a typical cyclic voltammetry test, the redox peak current (i_p) and the scan rate (ν) satisfy the following relationship [42]:

$$i_p = a\nu^b \quad (4)$$

where a and b are adjustable constants. When $b = 0.5$,

the charge/discharge is controlled by diffusion-controlled electrode process; when $b = 1.0$, the charge/discharge is adsorption-controlled electrode process; if $0.5 < b < 1$, the electrode reaction of NaCl electrolyte is an irreversible process driven by both adsorption and diffusion [42–44]. Take the logarithm of Eq. (4) at both sides of equal sign, we get the following equation:

$$\lg i_p = b \lg v + \lg a \quad (5)$$

Figure 6 plots the cyclic voltammograms (CVs) of 300 and 900 °C samples dipped into 1 M NaCl neutral electrolyte collected at potential scan rates from 0.1 to 0.5 V/s. In Fig. 6(a), redox peaks appearing at (–0.5)–0.7 V represent the oxidation/reduction of Ta ion species on the anode surface. For the scan rate of 0.5 V/s anodic peak is broad, about 500 mV and cathodic peak is relatively narrow, about 375 mV. The maximum peak currents of anodic and cathodic peaks are 3.0 and –3.7 μA, respectively. When the scan rate decreases from 0.4 till 0.1 V/s, the cathodic peak potentials shift from –0.5 to 0.2 V in a positive direction (the negative shift of the peak potentials signifies a greater ohmic resistance in the thicker oxide coating) and the peak-to-peak potential separation decreases. While both the anodic/cathodic current peaks decrease in intensity, the cathodic peaks decrease faster. In Fig. 6(b), previous redox peaks are not present. When the scan rate increases from 0.1 to 0.5 V/s, the peak current gradually increases, and their shapes remain unchanged, reflecting good electrochemical stability of 900 °C electrode. The maximum peak currents are –0.8 and 0.8 μA, respectively. This phenomenon occurs probably because the film defects or vacancies are enormously eliminated with 900 °C

annealing, and the capacity of the film surface to capture carriers decreases, leading to performance degradation of the film conductivity and peak current.

The linear relationship between $\lg i_p$ and $\lg v$ and the slope b values were obtained from the insets of Fig. 6. The b values of ~0.625 for 300 °C film and ~0.877 for 900 °C film indicate that Ta₂O₅ film electrode behaves are driven by both adsorption and diffusion. Further, the b value of Ta₂O₅ film electrode increases with annealing temperatures. The possible reason is that annealing reduces the oxygen vacancies, improves the flatness and uniformity of the film, and weakens the ability of the electrode surface to adsorb ions.

Figure 7 shows the XPS results of 300 and 900 °C electrodes before and after the CV tests and their area ratio graph after Ta⁵⁺/Ta⁴⁺ peak position fitting. In Fig. 7(a), the characteristic peak positions of Ta⁵⁺ of Ta 4f_{7/2} and 4f_{5/2} of the samples are 26.2 and 28.0 eV [45,46]. The presence of surface states or oxygen vacancies may cause partial Ta⁵⁺ to lower chemical valence state in the formula of Ta₂O_{5-δ} [47]. After fitting the Ta 4f spectral line of (a), a small amount of Ta⁴⁺ peaks at 25.7 and 27.9 eV [46] are obtained. In Fig. 7(b), the two pairs of peaks in the sample after the CV test are located at 25.4, 25.9, 27.7, and 27.9 eV, respectively. Among them, the 25.9 and 27.9 eV peaks represent Ta⁵⁺ in the Ta 4f spectrum. Further, the Ta⁴⁺ content of the sample has increased from 5.27% to 7.73% for 4f_{7/2} and from 3.95% to 5.80% for 4f_{5/2}, which infers that a certain amount of Ta⁴⁺ in the 300 °C sample was produced after the electrochemical test. Thus, the anodic/cathodic peaks in CV test (Fig. 6) may be attributed to the redox transitions of Ta⁵⁺/Ta⁴⁺ [45,48]. How the specific reactions take place still needs further

Fig. 6 Cyclic voltammograms of Ta₂O₅/Si anode electrode at (a) 300 °C and (b) 900 °C anneal and their fitting curves in 1 M NaCl electrolyte at different potential scan rates. The insets: log(peak current) vs. log(scan rate) of Ta₂O₅/Si electrode at different annealing temperatures.

Fig. 7 XPS spectra for 4f energy levels of Ta on Ta₂O₅/Si anode electrodes at 300 and 900 °C anneal (a, c) before and (b, d) after the CV tests, with fitting peaks of Ta⁵⁺/Ta⁴⁺ and their contents on film surfaces, respectively.

investigation. With larger reduction peak in Fig. 6(a) than oxidation peak, it might be concluded that the reduction process proceeds faster than the oxidation process and thus, more Ta⁴⁺ was generated. In Figs. 7(c) and 7(d), the peak position and content of Ta 4f do not change much. Due to the symmetrical CV curve of Fig. 6(b), the Ta⁵⁺/Ta⁴⁺ redox transition seems to be reversible, which suggests better chemical stability and cyclical performance of the electrode material after 900 °C heat treatment.

4 Conclusions

Ta₂O₅ films were synthesized on Si(100) and quartz substrates via EBE method, followed by annealing at different temperatures in air. Amorphous dense films formed at temperatures between 300 and 700 °C with enlarged band gap values from 4.32 to 4.46 eV. As the annealing temperature increased, the film transmittance was improved due to the higher film density and less defects in the film. Film crystallization of β-Ta₂O₅ occurred at 900 °C, which produced the highest transmittance rate among these Ta₂O₅ films. The smooth and flat film-surface, high film-density, less defects in the film, and crystallization may play

vital roles in maintaining the excellent transmittance properties of Ta₂O₅ films. The crystallization of amorphous Ta₂O₅ film by annealing may lead to the reversible electrochemical reaction inside electrolyte.

Acknowledgements

This work was supported by the Joint Research Funds of Department of Science & Technology of Shaanxi Province and Northwestern Polytechnical University (Grant No. 2020GXLH-Z-029).

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