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Item Type	Presentation;Presentation
Authors	Jeje, S.O.;Jeje, S.O.;Akinwunmi, O.O.;Akinwunmi, O.O.;Shongwe, M.B.;Shongwe, M.B.
Publisher	IOP Publishing;IOP Publishing
Rights	Attribution-NonCommercial-NoDerivatives 4.0 International
Download date	2024-10-04 22:09:14
Item License	http://creativecommons.org/licenses/by-nc-nd/4.0/
Link to Item	https://hdl.handle.net/20.500.14519/490

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To cite this article: S O Jeje *et al* 2018 *IOP Conf. Ser.: Mater. Sci. Eng.* **430** 012049

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Synthesis and Characterization of Tantalum Oxide Deposited By Metal-Organic Chemical Vapour Deposition (MOCVD)

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Abstract: Tantalum oxide thin-films have rapidly evolved as a potentially important film material in a wide range of industrial applications such as optical coatings, dielectric films, corrosion and heat resistant coatings, dental implants and coronary stents, or prosthesis. The preparation of an improved liquid source metal-organic precursor suitable for the deposition of high-quality tantalum oxide thin films on soda-lime glass using Metal-organic Chemical Vapour Deposition (MOCVD) was reported. Peroxo-hydroxyl-amino tantalum complex precursor was prepared by a method of ligand-exchange reactions between hydroxo-peroxo tantalum complexes and traditional chelating reagents. Preparation of the thin-films was achieved by the pyrolysis of the precursor at four different temperatures (380 °C, 400 °C, 420 °C, 450 °C) with a flow rate of 1.5 dm³/min for 2 hours deposition period. The deposited films were characterized using Ultraviolet-Visible spectroscopy, Scanning Electron Microscopy (SEM) and Energy Dispersive X-ray spectroscopy (EDX). A direct optical band gap of 4.16 eV – 4.70 eV was obtained from the analysis of the absorption spectrum of the thin-films. SEM micrographs revealed that the deposited film had no regular structure and the layers could be described as amorphous. The EDX results confirmed the presence of Tantalum and Oxygen in the thin-films.

1. Introduction

Tantalum oxides are high dielectric constant material that has reasonably low leakage current and higher breakdown strength [1-3]. Some of its unique properties include high corrosion resistance, piezoelectricity and high refractive index [4]. Thin films of tantalum oxides have triggered considerable interest from the research community, due to their valuable physical properties as well as the promise of compatibility with microelectronics processing [3]. Tantalum oxide thin films have rapidly evolved as a potentially important film material in a wide range of industrial applications such as optical coatings, dielectric films, corrosion and heat resistant coatings, as high-reflective film mirrors for solar cells, charge coupled devices (CCDs), high power laser equipment, dental implants and coronary stents, or prosthesis [2-4].



Tantalum is a very passive metal such that it is almost completely immune to attack by hydrofluoric acid, fluoride ions in acidic solution, free sulphur trioxide and hot/strong alkalis (such as boiling aqueous solutions of metal hydroxides) at a temperature below 150°C [5-7]. This made tantalum suitable for applications in process equipment meant for harsh chemical environments. Good examples of such are surgical tools and implants because of tantalum inertness to bodily fluids [8]. Tantalum has a high affinity for the interstitial gas like oxygen, it forms a dense, non-porous layer of tantalum oxide on the surface that passivates tantalum and gives it its superior corrosion resistance in aqueous media [7].

Tantalum oxide layers employed so far have been deposited by Physical Vapour Deposition (PVD). PVD deposition technique has the limitation of low deposition rate and relative non-conformal coverage in devices and coated tools, which results in non-uniformity of the surface with submicron features and high aspect ratio contacts and via holes. The volatile source of tantalum was the pentahalides. The most common of these was TaCl₅ and deposition was through thermal decomposition or hydrogen reduction [9]. Tantalum oxide thin film deposition was accomplished using different techniques such as electron-beam evaporation, ion-assisted deposition, reactive magnetron sputtering and ion beam sputtering [10]. Metal-organic chemical vapour deposition (MOCVD) entails thin solid film formation on a substrate material by a chemical reaction of vapour-phase organometallic precursor that contains a direct metal-carbon bond. MOCVD is preferred to other deposition processes because its step coverage of films had been found to be superior even at a lower temperature. This study is aimed at producing tantalum oxide thin films from peroxy- α -hydroxyl-amino tantalum complex precursor using MOCVD technique and also to describe the optical properties of the tantalum oxide thin films prepared.

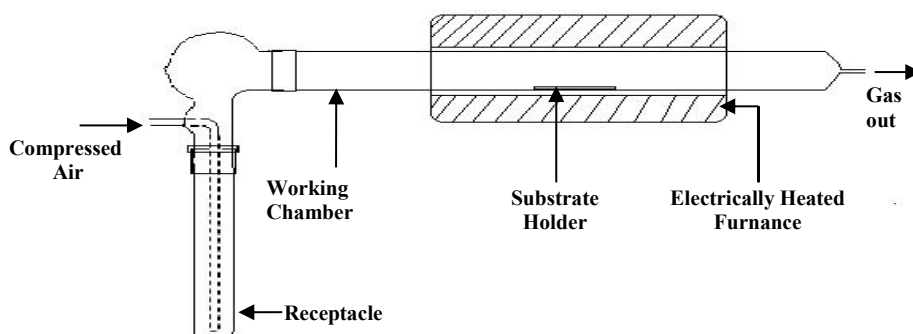
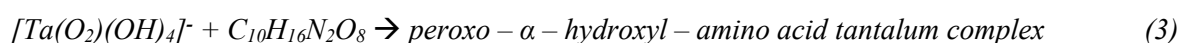
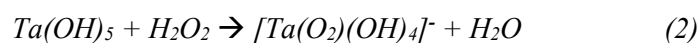
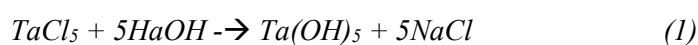


Figure 1. MOCVD thinfilms deposition setup

2. Experimental

The starting materials were tantalum pentachloride (TaCl₅), sodium hydroxide (NaOH), hydrogen peroxide (H₂O₂), ammonia (NH₃) solution (all supplied by Sigma Aldrich) and distilled water (H₂O). In a horizontal electrically heated furnace, tantalum pentoxide thin film was deposited on soda-lime glass by metal-organic chemical vapour deposition (MOCVD) technique using peroxy- α -hydroxyl-amino tantalum complex precursor which was prepared using the method of ‘Ligand-Exchange Reactions between Hydroxo-Peroxy Tantalum Complexes and Traditional Chelating Reagents’ related to the reaction as recorded for Titanium by Masato Kakihana et al. [11]. Equations (1) to (3) show the reaction for the precursor preparation.



The set up for the deposition and the flowchart for the precursor preparation are shown in Figure 1 and Figure 2 respectively. The clear liquid precursor was poured into an unheated receptacle and carrier gas (compressed air) was blown through the liquid precursor at 1.5 dm³/min flow. The compressed airborne precursor was transported into the working chamber maintained at a temperature of 380 °C by an electrically heated furnace for 2 hours; the process was repeated for three other temperatures (400 °C, 420 °C and 450 °C). This was carried out in a fume hood in order to curb problems associated with organic by-products. The thickness of the coating was determined using colour chart approach for thermally grown SiO₂ based on reflection theory of visible light. The thin film was characterized using UV visible spectroscopy, Scanning Electron Microscopy (SEM) and Energy Dispersive X-Ray (EDX) to determine the optical, morphological, and compositional analysis of the deposited films respectively.

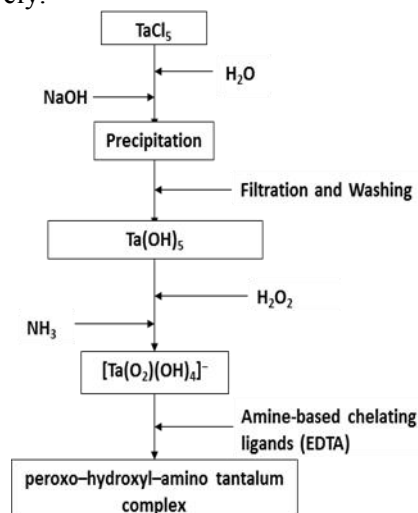


Figure 2. Flowchart for preparation of a water-soluble peroxo-hydroxyl-amino tantalum complex starting from TaCl₅

3. Results and Discussion

3.1 Film thickness

Colour chart approach for thermally grown SiO₂ for thickness measurement based on reflection theory of visible light was adopted in this work as it can be used for dielectric films other than SiO₂ or Si₃N₄ [12]. Using calibrated colour charts, thicknesses can be estimated between 10 to 20 nm accuracy and they are useful for oxide films thicker than about 80 nm [12]. The estimated value was calculated by using the related expression in equation 4.

$$n_a t_a = n_x t_x \quad (4)$$

where n_a = refractive index of SiO₂ = 1.46

t_a = SiO₂ film thickness from colour chart

n_x = estimated refractive index of the deposited film = 2.275

t_x = unknown film thickness of deposited film

540nm, 560nm, 570nm and 680nm represent SiO₂ film thickness from colour chart at deposition temperature at 380 °C, 400 °C, 420 °C and 450 °C respectively. Equation 4 was applied to calculate films thickness of the deposited Tantalum oxide film at temperature of 380 °C, 400 °C, 420 °C and 450 °C as 346.55nm, 359.38nm, 365.80nm, and 436.40nm respectively. The deposition rate as a function of the deposition temperature indicates that the growth rate for Tantalum oxide films increased with increasing deposition temperature which is in agreement with the work reported by Maeng *et al.* [13]; this can be attributed to the increase of kinetic reaction process in the reaction chamber as the temperature increases.

3.2 Optical characterization of thin films

The UV-visible spectra of the thin films at different temperatures on the soda-lime glass substrate are shown in Figure 3.

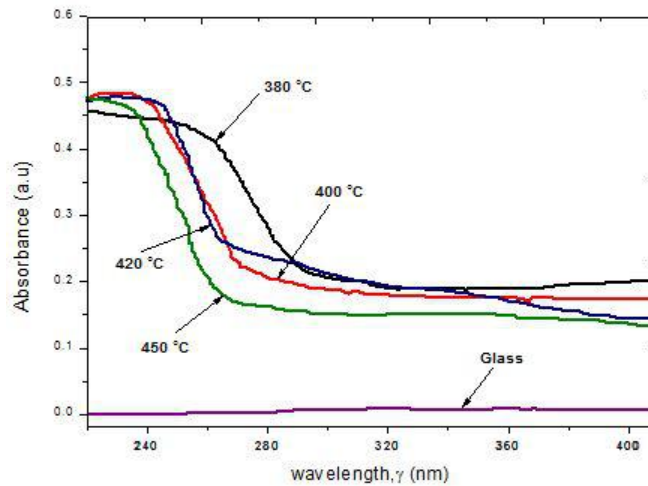


Figure 3. Absorbance versus wavelength for the thinfilms

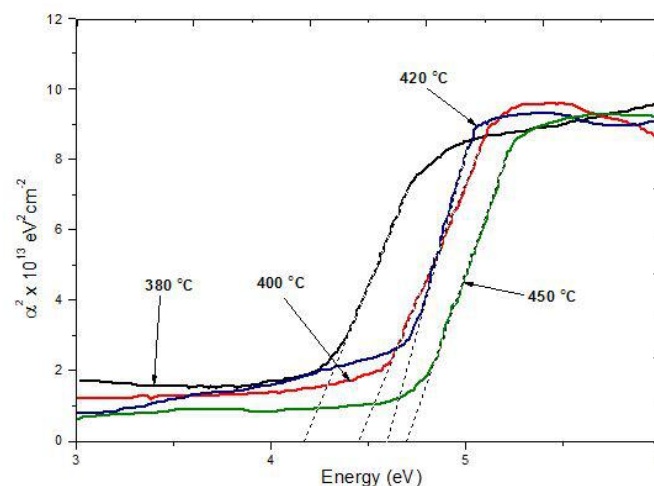


Figure 4. α^2 versus energy of the thinfilms

Each spectrum is a plot of absorbance against incident photon wavelength at normal incidence and room temperature. The UV-Visible spectra showed that the optical absorbance of the films increases with increasing energy of the photon, indicating that the film is highly absorbing in the UV region. The Transmittance, Energy (eV) and the absorption coefficient was determined by equation (4), (5) and (6) respectively, where T = transmittance; A = absorbance; h = Planck's constant; c = speed of light; λ = wavelength (nm); and t = film thickness. The absorption coefficient squared (α^2) was plotted against photon energy (eV) in figure 4 to determine the energy band gap of the deposited film. The resulting Tauc plot (figure 4) has a distinct linear regime that indicates the onset of absorption. Thus, extrapolating the linear region to the abscissa yields the energy of the optical band gap of the material. The extrapolations of the linear part of the Tauc plot to the x-axis were 4.16 eV, 4.45 eV, 4.59 eV and 4.7 eV for temperature 380 °C, 400 °C, 420 °C and 450 °C respectively.

$$T = 10^{-A} \quad (4)$$

$$E = hc/\lambda \quad (5)$$

$$A = [\ln(1/T)]/t \quad (6)$$

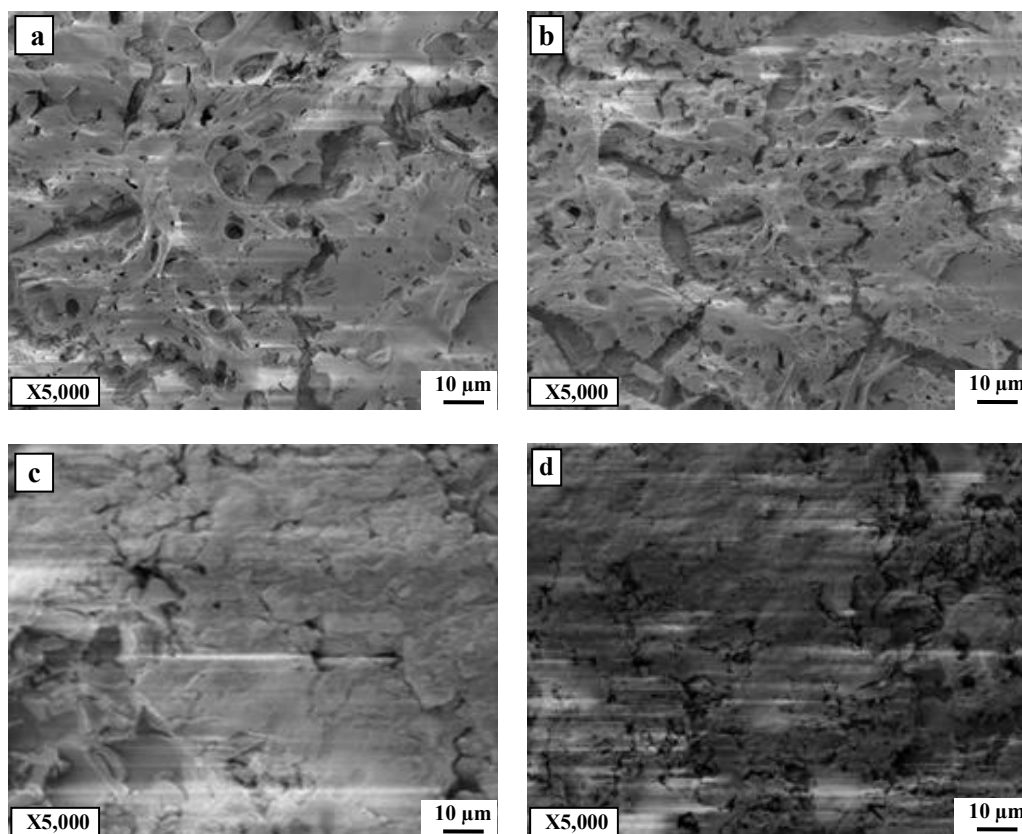


Figure 5. SEM Micrograph of Thin Film at (a) 380 °C (b) 400 °C (c) 420 °C and (d) 450 °C

3.3 Morphological and compositional characterization

Figure 5a – d shows the SEM micrograph of the thin films at 380 °C, 400 °C, 420 °C and 450 °C respectively. It is observed that the deposited layers are less of cracks and pores and the grains have no regular structure. The layers can be described as amorphous and the porosity decreases with increment in temperature up to 450 °C. Films that are amorphous do not require lattice matching and epitaxy, therefore, uniform and defect-free multi-layers of amorphous films can be grown even on sharp interfaces down to small individual layer thickness [14]. The EDX spectrum (not shown here) confirmed the presence of Tantalum and Oxygen as the elements present. Other elements (Na, Mg, Si, Ca) in the spectrum are present in the sodalime glass slide used as the thin film substrate. The stoichiometry of the tantalum oxide film could not be ascertained using EDX.

4. Conclusion

A novel single liquid source precursor peroxo–hydroxyl–amino tantalum complex has been synthesized from tantalum chloride and solvents by modifying the method of ‘Ligand-Exchange Reactions between Hydroxo–Peroxo Titanium Complexes and Traditional Chelating Reagents’. The tantalum oxide thin films obtained has little or no impurity compound at different deposition temperatures of 380 °C, 400 °C, 420 °C and 450 °C respectively. The thickness of deposited films was found to vary slightly in the range of 346.55 nm to 436.40 nm, therefore the thickness increased with increase in deposition temperature. The direct energy band gap of the deposited films increased from 4.16 eV to 4.70 eV with respect to increasing the deposition temperature from 380 °C to 450 °C.

Acknowledgments

This research was supported by MOCVD laboratory, Physics department, Obafemi Awolowo University, Ile-Ife, Nigeria and Institute of Nano Engineering Research (INER), Tshwane University of Technology, South Africa.

References

- [1] Piret F, Singh M, Takoudis C G and Su B L 2008 *Che. Phy. Lett.* **453** 87-91.
- [2] Jong-Ho Y and Shi-Woo R 1997 *Thin Solid Films* **292** 324-29.
- [3] Vladoiu R, Ciupina V, Mandes A, Dinca V, Prodan M and Musa G 2010 *J. of App. Phys* **108** 093301.
- [4] Audier M, Chenevier B, Roussel H, Vincent L, Peña A and Lintanf Salaün A 2011 *J. of Solid State Chemistry* **184** 2033-40.
- [5] Cardonne S M, Kumar P, Michaluk C A, and Schwartz H D 1995 *Int. J. of Ref Met and Hard Materials* **13** 187-94.
- [6] Patnaik P 2003 *McGraw-Hill Companies Handbook-of-Inorganic-Chemicals*.
- [7] Namur R S, Reyes K M and Marino C E B, 2015 *Mat. Research* **18** 91-97.
- [8] Rahmati B, Sarhan A D, Basirun W J, and Abas W A B W 2016 *J. of Alloys and Compounds* **676** 369-76.
- [9] Levesque A, and Bouteville A 2004 *Chemical Vapor Deposition* **10** 23-28.
- [10] Shakoury R, and Willey R R 2016 *Appl Opt* **55** 5353-7.
- [11] Kakihana M, Kobayashi M, Tomita K, and Petrykin V 2010 *Bull. of the Chem.l Society of Japan* **83** 1285-308.
- [12] Schroder D K 2006 *Semiconductor Material and Device Characterization, Third Edition* (Wiley) 595.
- [13] Maeng W J, Park Sang-Joon, and Kim H 2006 *J. of Vacuum Sci. & Tech. B: Microelectronics and Nanometer Structures* **24** 2276.
- [14] Gu Z G and Zhang J, 2017 *Coordination Chemistry Review: Epitaxial growth and applications of oriented metal–organic framework thin films* 2017/10/26/.