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Effect of Annealed Duration and Temperature on Crystallinity of Ta_2O_5 Thin Films

H. Darmasetiawan¹⁾, Irzaman¹⁾, M. N. Indro¹⁾, S. G. Sukaryo¹⁾, M Hikam²⁾, and N. P. Bo²⁾
¹⁾Department of Physics, FMIPA IPB,
Jl. Raya Pajajaran Bogor, Indonesia, e-mail : irzaman@student.fi.itb.ac.id
²⁾Department of Physics, FMIPA University Indonesia,
Kampus Depok, Indonesia, e-mail : hikam@makara.cso.ui.ac.id

Abstract
Tantalum oxide (Ta_2O_5) thin films were deposited on Si(100) and Si (100)/Pt (200) substrates using the chemical solution deposition (CSD) method. X-ray diffraction (XRD) measurement was employed to characterize the films. The growth condition of Ta_2O_5 was carried out by spin coating at 3000 rpm for 30 seconds, and then by annealing at 900°C for 4 hours and 15 hours. The crystallinity of thin films were investigated by XRD preferred orientation (001), (010), (100), (200) on Si (100) substrate and (001), (010), (100), (202) on Si (100)/Pt (200) substrate. Increasing the duration of annealing results in better quality of crystalline thin films and the effect of high thermal energy increases the grain size. The orthorhombic structure and crystallinity of Ta_2O_5 thin films on Si (100) substrates and Si (100)/Pt (200) substrates were analyzed by XRD preferred orientation (001), (010) and (100), giving lattice constants a = 3.708 Å, b = 3.716 Å, c = 4.114 Å.

Keywords : *Ta_2O_5, thin films, CSD method, annealed duration, orthorhombic.*

Abstrak
*Film tipis tantalum oksida (Ta_2O_5) pada substrat Si (100) dan Si (100)/Pt (200) ditumbuhkan dengan metode larutan kimia (*Chemical Solution Deposition* = CSD). Karakterisasi film dilakukan dengan difraksi sinar-x (XRD). Kondisi penumbuhan Ta_2O_5 dilakukan dengan* spin coating *pada kecepatan putaran 3000 rpm selama 30 detik dan kemudian dilakukan* annealing *(pemanggangan) pada suhu 900°C selama 4 jam dan 15 jam. Hasil kristal yang terbentuk diamati dengan XRD, bidang kristal (001), (010), (100), (200) terbentuk pada substrat Si (100) dan bidang kristal (001), (010), (100), (202) terbentuk pada substrat Si (100)/Pt (200). Makin lama waktu* annealing *ternyata hasilnya lebih baik dalam hal pembentukan kristal film tipis dan pengaruh energi termal yang tinggi menambah pembentukan ukuran butiran film tipis. Struktur kristal ortorombik dari film tipis Ta_2O_5 pada substrat Si (100) dan Si (100)/Pt (200) yang diamati dengan XRD pada bidang (001), (010) dan (100) diperoleh nilai konstanta kisi masing-masing a = 3,708 Å, b = 3,716 Å, c = 4,114 Å.*

Kata kunci : *Ta_2O_5, film tipis, metode CSD, lama annealing, ortorombik.*

1. Introduction

Tantalum oxide (Ta_2O_5) films are of considerable interest for a range of applications, including optical waveguide devices, high temperature resistors, oxygen sensors and relative humidity sensor¹⁾. Because of their high dielectric constant, in recent years they have received increasing attention as a possible alternative dielectric to replace thin SiO_2 layers as capacitor insulators in high density dynamic random access memories (DRAM) and in ultra large scale integrated (ULSI) devices¹⁾.

Tantalum oxide thin films can be formed by various methods, such as chemical solution deposition (CSD)^1,2,3)^, metal organic chemical vapor deposition (MOCVD)⁴⁾, plasma enhanced chemical vapor deposition (PECVD)^5,6)^, low pressure chemical vapor deposition (LPCVD)⁷⁾, and reactive magnetron sputtering⁸⁾. The properties of Ta_2O_5 thin films have been reported to be strongly dependent on the fabrication method, nature of substrate and electrode material, and post deposition-annealing treatment.

CSD Method is one of particular interest among other methods because of its good control of stoichiometry, easier to fabricate, low and high temperature synthesis. It is relatively new and requires a greater understanding to optimize the film quality. Crystallization mechanisms in CSD-derived thin films are different from phenomena associated with vapor phase epitaxy. It was reported that CSD derived thermodynamically is stable.

2. Experimental Procedure

A Tantalum oxide (Ta_2O_5) thin film was deposited on Si (100) and Si (100)/Pt (200) substrates respectively by chemical solution deposition (CSD) method. In this experiment, tantalum ethoxide ($Ta(OC_2H_5)_5$) was initially dissolved in 2-methoxyethanol ($H_3COCH_2CH_2OH$) in a reactive tube. For getting a clear solution mixing by the ultrasonic model Branson 2210 was used for 1 hour. Finally acetic acid was added to prevent rapid hydrolysis and metal oxide. After 2 hours of aging at room temperature the solution was applied on 25 mm x 25 mm of Si (100) and Si (100)/Pt (200) substrates respectively and then was prepared by spin coating at 3000 rpm for 30 seconds. Finally the films were annealed in furnace at 900°C for 4 hours and 15 hours in an air atmosphere.

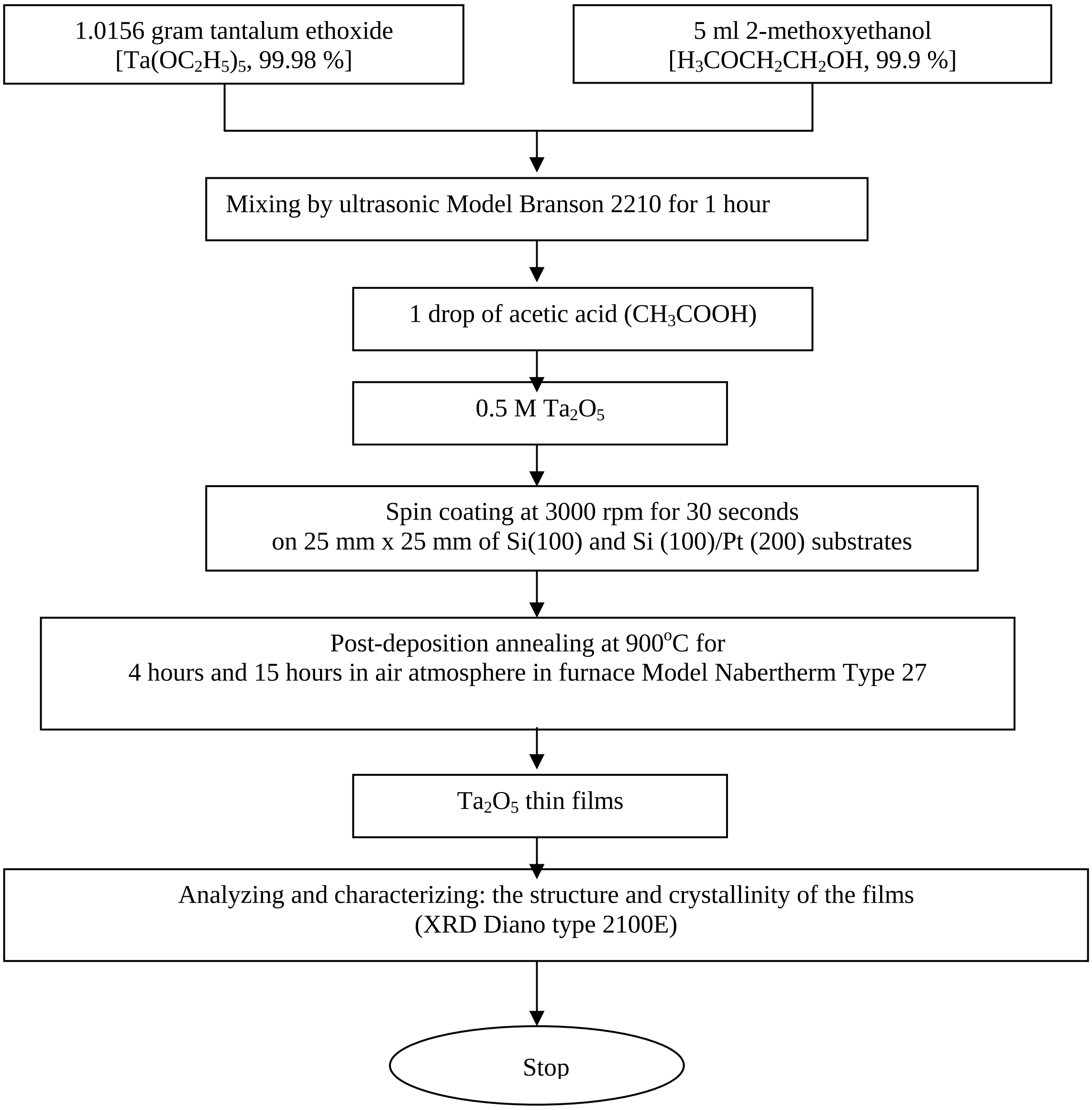

Figure 1. Flow diagram of the research

Table 1. Treatment of the growth of Ta_2O_5 thin films using CSD method

Substrates	Treatment	Annealing duration (hour)	Angular velocity (rpm)	Rotation time (second)	Annealing temperature (°C)
Si (100)	I	4	3000	30	900
	II	15			
Si (100)/ Pt (200)	III	4			
	IV	15			

The structure of the films was analyzed by x-ray diffraction (XRD). The XRD pattern was recorded on a Diano type 2100E diffractometer using CuKα radiation at 30 kV and 30 mA (900 watt). Figure 1 shows the flow diagram of research[9,10] and Table 1 shows four treatment of the growth of Ta_2O_5 thin films.

The various sets of planes in a lattice have various values of interplanar spacing. The planes of large spacing have low indices. The interplanar spacing (d_{hkl}) is a function of both the planes indices (*hkl*) and lattice constants (*a, b, c*). For calculating lattice constants *a, b* and *c* Equation (1) and (2) below were used: [11,12]

$$\lambda = 2d\sin\theta, \tag{1}$$

$$\frac{1}{d^2} = \frac{h^2}{a^2} + \frac{k^2}{b^2} + \frac{l^2}{c^2}. \tag{2}$$

where : d = interplanar spacing; a, b, c = lattice constants; h, k, l = plane indices; λ = wave length (Cu element = 1.542 Å); θ = angle of diffraction.

3. Results and Discussion

A polycrystalline film of orthorhombic phase was obtained at an annealing temperature of 900°C for 4 hours and 15 hours and showed the different crystal planes.

The crystallinity and the orientation of the annealed Ta_2O_5 films were determined by x-ray diffraction. The Ta_2O_5 film that was annealed at 900°C for 15 hours indicated better crystallinity and grain size as shown in Figures 2 and 3. In fact, the effect of high temperature and varying the duration of annealing caused one peak increase in intensity. Increase of annealing duration increased the diffraction peak corresponding to (001) plane and decreased the full width of its half maximum (FWHM). This indicates enhanced crystallinity and increased grain size. Besides, the diffraction from (200), (201), (020) and (1 11 2) planes became relatively weaker.

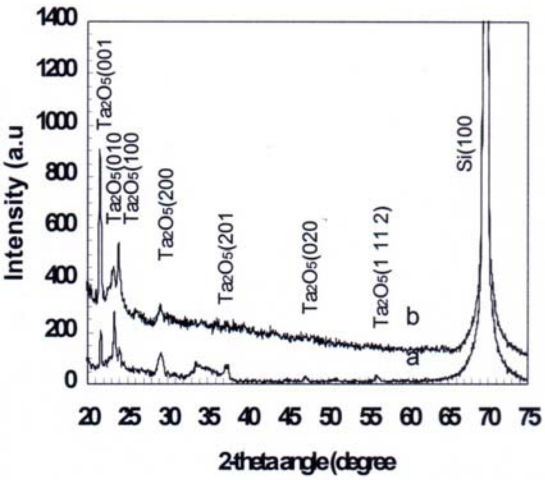

Figure 2. The structure and crystallinity by XRD of Ta_2O_5 thin films on Si (100) substrate at 3000 rpm for 30 seconds, annealing temperature 900°C, (a). Annealing duration = 4 hours, (b). Annealing duration = 15 hours.

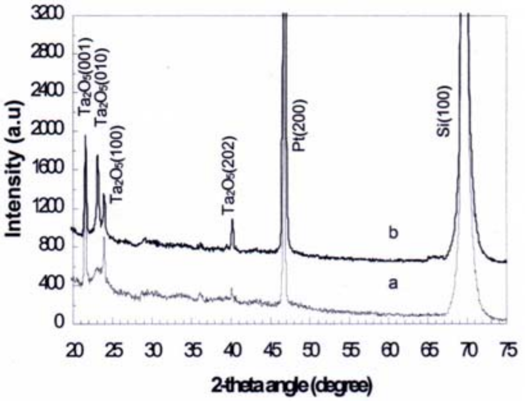

Figure 3. The structure and crystallinity by XRD of Ta_2O_5 thin films on Si (100)/Pt (200) substrate at 3000 rpm for 30 seconds, annealing temperature 900°C, (a). Annealing duration = 4 hours, (b). Annealing duration = 15 hours.

The diffraction peaks of (001), (010) and (100) crystal planes on Si (100) substrate and on Si (100)/Pt (200) substrate were used for calculating lattice constants *a*, *b* and *c*. By using Equation (1) and (2) the lattice constants of *a*, *b* and *c* were 3.708 Å, 3.716 Å, 4.114 Å respectively (in literature shown a = 6.225 Å, b = 39.8521 Å, c = 3.8609 Å [3] and a= 6.20 Å, b = 3.66 Å, c = 3.89 Å [4]).

The XRD pattern of one film showed an additional peak at 2θ = 40° that was related to the (202) plane as shown in Figure 3.

4. Conclusion

A polycrystalline film of orthorhombic phase was obtained at an annealing temperature of 900°C using 4 hours and 15 hours annealing. Annealing for 15 hours indicated better crystallinity and grain size than annealing for 4 hours. The lattice constants of the Ta_2O_5 film on Si (100) substrate and on Si (100)/Pt (200) substrates were a = 3.708 Å, b = 3.716 Å, c = 4.114 Å.

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