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# Crystalline structure and optical properties of thin film LiTaO$_3$

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**Abstract**. The coatings of LiTaO$_3$ material on p-type Si (100) substrates were fabricated by using Chemical Solution Deposition (CSD) technique. In the fabrication process, the spin coater is used to flatten the LiTaO$_3$ solution on the p-type Si (100) substrates surface, which was set at 4000 rpm for 30 seconds. Furthermore, the coating on those substrates was annealed in the furnace (Nabertherm type B410) for 15 hours at 750ºC, 800ºC, and 850ºC temperature. The study of crystalline structure was obtained through the X-Ray Diffraction (Shimadzu XRD-7000) measuring with the interval of angular diffraction 2θ from 10º to 80º in the increment of 0.02º. Moreover, the study of optical properties was observed using the spectrometer (Ocean Optics USB4000-UV-VIS). Based on the results of the study, obtained a hexagonal angle diffraction peak for the crystal structure, when viewed from the refractive index, the optical properties are sensitive to visible and ultraviolet light. The results of the crystalline structure and optical properties that are sensitive to visible and ultraviolet light can be used to improve the performance of satellite systems in the term of the optical remote sensing.

## 1. Introduction

Lithium tantalate (LiTaO$_3$) is one of perovskite ferroelectric optical crystal materials [1-3]. The presence of external field derived from the sunlight spectrum may affect the position of atoms at the center of the perovskite structure, as it happens when overloaded on one side of the crystal (electro-optics) [4-8]. LiTaO$_3$ is very interesting to study because in practice it can be used as a sensor. However, over the last few years, LiTaO$_3$ has been studied intensively because it has unique properties [1, 4, 5]. LiTaO$_3$ is crystalline ferroelectric having an excellent of pyroelectric, ferroelectric, and coefficients that are non-linear optic [3, 9-11]. LiTaO$_3$ is crystalline ferroelectric which subjected to the process of high Curie temperature by 608ºC and it also has a high melting temperature at 1650ºC [2, 3, 12-14]. A good LiTaO$_3$ ferroelectric characteristic is used on electronic devices and optical devices [15, 16].

In order to choose the best intensity and high purity of $LiTaO_3$, using X-Ray Diffraction (XRD) method is suggested. X-Ray Diffraction methods can explain the lattice parameter, structure type, different atomic, the arrangement of crystals, presence of crystal imperfection, orientation, grain, and grain size. The XRD characterization aims to determine the crystal system [17, 18]. In this study, $LiTaO_3$ thin films were obtained from mixing lithium acetate and tantalum methoxide were coated on p-type Si (100) substrate using Chemical Solution Deposition (CSD) method, followed by spin-coating technique and annealed using furnace at variety temperature of 750ºC, 800ºC, and 850ºC, then characterized using X-Ray Diffraction (Shimadzu XRD-7000). Spectrometer (Ocean Optics USB4000-UV-VIS) was used to investigate the energy gap and the refractive index of $LiTaO_3$ thin films [2, 17, 18].

Related to the electromagnetic spectrum and the optical remote sensing on the satellite system, the purpose of this study is to fabricate and investigate the performance of $LiTaO_3$ thin film which can be applied for: (1) climate and environmental monitoring (oceanography field), which have required sensor with sensitivity in the visible light range; (2) thermal monitoring (volcanology field), which have required sensor with sensitivity in the ultraviolet light range; and (3) weather and climate monitoring (meteorology field), which have required sensor with sensitivity in the ultraviolet-visible light range [19-21].

## 2. Research method

In the first process, the used substrate, p-type Si (100), was cut to $1 \times 1$ cm$^2$ size using a glass cutter. Furthermore, the substrates were cleaned sequentially for 15 minutes with acetone, methanol, and deionized water in the tub of the ultrasonicator device [22-25]. Then, 0.16496 gram of lithium acetate [$Li(CH_3COO)$, 99.99%] and 0.84030 gram of tantalum methoxide [$Ta(OCH_3)_5$, 99%] were weighed using Kern analytical scale. The materials were then mixed and dissolved in 2.5 ml of 2-methoxyethanol ($CH_3OCH_2CH_2OH$), thereafter stirred using Vortex 3000. Next step, the solution mixture was homogenized using the ultrasonicator device for 30 minutes to obtain a homogenous $LiTaO_3$ solution.

The substrate was placed on the center of spin coater plate with sellotape to cover 1/3 of the substrate surface [22]. The dripped $LiTaO_3$ solution on the p-type Si (100) substrate was fixed to the stage of spin coater [11, 22]. $LiTaO_3$ coating process on the surface of each substrate and rotating process using a spin coater at 4000 rpm speed for 30 seconds were repeated three (3) times (disposition and rotation) with one (1) minutes break in between [1, 6, 22-25]. $LiTaO_3$ thin film on p-type Si (100) substrate was then annealed using Nabertherm type B410 for 15 hours at 750ºC, 800ºC, and 850ºC temperature. The filming and crystallization of $LiTaO_3$ thin films were obviously affected by the heat treatment process [18].

After the growth and annealing process was completed, the formed thin films were characterized using X-Ray Diffraction (XRD), measuring it with the interval of angular diffraction 2θ from 10º to 80º in the increment of 0.02º and it was characterized using spectrometer (Ocean Optics USB4000-UV-VIS) to measure the reflectance.

## 3. Result and discussion

### 3.1. XRD pattern

X-Ray Diffraction (XRD) was used to observe the crystalline structure of $LiTaO_3$ thin films crystallized on the surface of the substrate p-type Si (100) with different annealing temperatures (750ºC, 800ºC, 850ºC). In this study, the thin films were characterized using X-Ray Diffractometer (Shimadzu XRD-7000) with Cu $K\alpha$ radiation whose wavelength ($\lambda_{Cu}$) is 1.54060 Å. The spectrum was recorded at a diffraction angle of 2θ from a range of $10º \leq \theta \leq 80º$ with the interval of 0.02º/minute. Measurement results (figure 1) show the peaks diffraction angle with the hexagonal crystal structure,

while the peak diffraction intensity showed the Miller index values. The Miller index is used to determine the lattice parameters of LiTaO₃ thin films with the Cramer and Cohen equations [26-28].

$$\sum \alpha \sin^2\theta = C\sum\alpha^2 + B\sum\alpha\gamma + A\sum\alpha\delta$$
$$\sum \gamma \sin^2\theta = C\sum\alpha\gamma + B\sum\gamma^2 + A\sum\gamma\delta \qquad (1)$$
$$\sum \delta \sin^2\theta = C\sum\alpha\delta + B\sum\gamma\delta + A\sum\delta^2$$

with:  $\alpha$, $\gamma$, $\delta$ = lattice parameters (Å)
  $\theta$ = Bragg angle (degree)
  $A$, $B$, $C$ = lattice constants
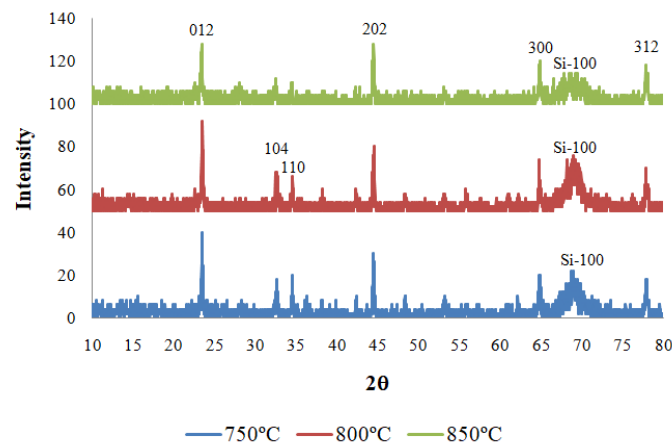


**Figure 1.** XRD patterns of LiTaO₃ thin films with different annealing temperatures.

Table 1 shows the lattice parameter values of LiTaO₃ thin films. Based on the table, the lattice parameter values show some differences from the JCPDS data. Small differences caused by the peaks diffraction value that appears at the 2θ diffraction angle are not the same as the diffraction angle from the JCPDS data. The obtained diffraction peaks experience a shift at an angle of 2θ. This diffraction angle shift was caused by the magnitude of the annealing temperature.

**Table 1.** Lattice parameter values of LiTaO₃ thin films.

| Lattice parameter | LiTaO₃ annealing at 750ºC (Å) | LiTaO₃ annealing at 800ºC (Å) | LiTaO₃ annealing at 850ºC (Å) | JCPDS (No. 29-0836) [29] (Å) |
|---|---|---|---|---|
| $c$ | 17.376750 | 17.453590 | 21.258570 | 13.750 |
| $a$ | 5.699490 | 5.722771 | 5.716996 | 5.153 |

The crystallite size of LiTaO₃ thin films can be calculated based on the FWHM of the diffraction peaks produced using the Scherrer equation [17]:

$$D = K \frac{\lambda}{\beta \cos\theta} \qquad (2)$$

with:  $D$ = crystallite size (Å)
  $K$ = constant (0.96)
  $\lambda$ = wavelength (1.506 Å)
  $\beta$ = the graph width of FWHM (radian)
  $\theta$ = maximum angle (radian)

Based on the Scherrer equation, the calculation of the crystallite size of $LiTaO_3$ thin films was listing in table 2. The average calculation value of the crystallite size of $LiTaO_3$ thin films on the surface of the substrate p-type Si (100) shows the decrement along with the increment of annealing temperature. It means that the increment of temperature can reduce the crystallite size.

**Table 2.** The crystallite size of $LiTaO_3$ thin films.

| Annealing temperature (ºC) | Diffraction angle 2θ (degree) | FWHM | Crystallite size (Å) |
|---|---|---|---|
| 750 | 23.5198 | 0.11 | 770.8524654 |
| | 32.6350 | 0.09 | 961.0708700 |
| | 34.5885 | 0.09 | 966.0269819 |
| | 44.4985 | 0.15 | 597.9111105 |
| | 64.8000 | 0.09 | 1092.264842 |
| | 77.9800 | 0.09 | 1186.404787 |
| *Average Crystallite Size (ACS)* | | | 929.0885095 |
| 800 | 23.5480 | 0.14 | 605.7008116 |
| | 32.6900 | 0.14 | 617.9180784 |
| | 34.5800 | 0.12 | 724.5035234 |
| | 44.4795 | 0.19 | 472.0031116 |
| | 64.8166 | 0.13 | 756.2528047 |
| | 77.8850 | 0.15 | 711.3662991 |
| *Average Crystallite Size (ACS)* | | | 647.9574381 |
| 850 | 23.4316 | 0.16 | 529.8764096 |
| | 34.5000 | 0.12 | 724.3464565 |
| | 44.4700 | 0.18 | 498.2086376 |
| | 64.8500 | 0.18 | 546.2835689 |
| | 77.8650 | 0.19 | 561.5258775 |
| *Average Crystallite Size (ACS)* | | | 572.0481900 |

*3.2. Energy gap and refractive index*

The energy gap of $LiTaO_3$ thin films was presented in figure 2, which determined through extrapolation by withdrawing straight line on the coordinate axis using the following equation [30]:

$$\text{y-axis: } 2\,\alpha\,d = \left[ ln\left( \frac{R_{max} - R_{min}}{R - R_{min}} \right) \right]^2 \tag{3}$$

$$\text{x-axis: } E_g = \frac{h\,c}{\left(1.602 \times 10^{-19}\ J\right)(\lambda)} \tag{4}$$

with:  $\alpha$ = absorbance coefficient
  $d$ = film thickness (μm)
  $R$ = reflectance values (from measurement using an Ocean Optics USB4000-UV-VIS)
  $R_{max}$ = maximum reflectance value
  $R_{min}$ = minimum reflectance value
  $E_g$ = energy gap (eV)
  $h$ = Planck's constant ($6.626 \times 10^{-34}$ J·s)
  $c$ = speed of light ($2.998 \times 10^8$ m·s$^{-1}$)
  $\lambda$ = wavelength (m)

In this study, the refractive index values of LiTaO$_3$ thin films were pointed out in figure 3 and listed in table 3. In figure 3, the coordinate axes were plotted to form the curve of the energy gap with the refractive index through the temperature dependent dispersion relation by using the Sellmeier equation [31, 32]:
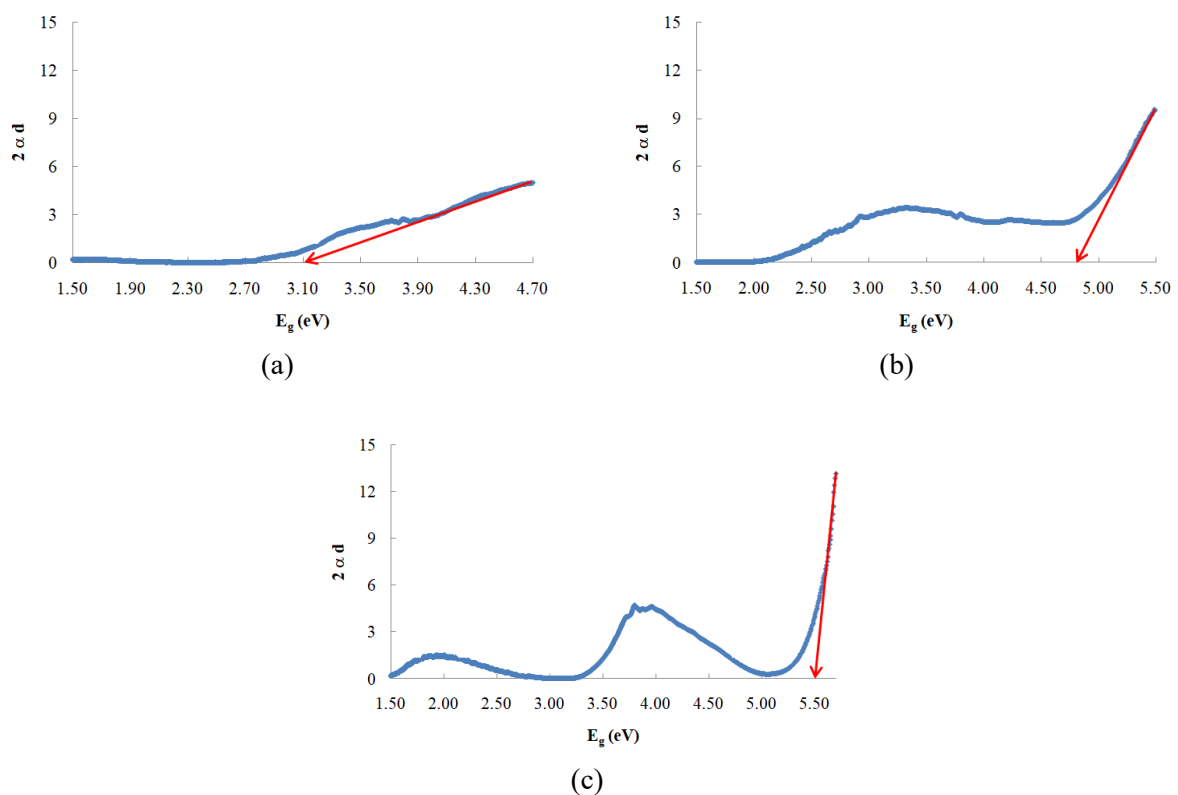
$$n_e^2(\lambda,T) = A + \frac{B + b(T)}{\lambda^2 - [C + c(T)]^2} + \frac{E}{\lambda^2 - F^2} + \frac{G}{\lambda^2 - H^2} + D\,\lambda^2 \tag{5}$$
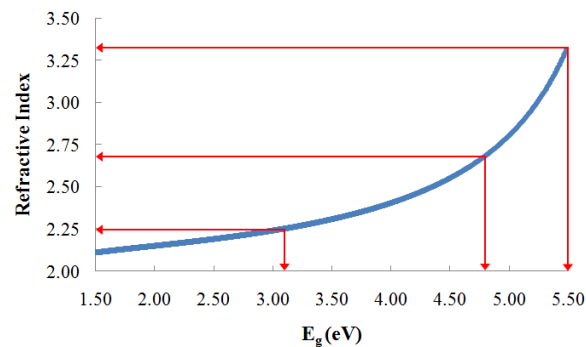
with: $n_e$ = refractive index

    $A, B, C, D, E, F, G, H$ = constants (Sellmeier coefficients)

    $b(T), c(T)$ = temperature dependent

    $\lambda$ = wavelength (μm)



(a)



(b)



(c)

**Figure 2.** Energy gap of LiTaO$_3$ thin films: (a) 750ºC (b) 800ºC (c) 850ºC.



**Figure 3.** Refractive index of LiTaO$_3$ thin films with annealing treatment at 750ºC, 800ºC, and 850ºC.

**Table 3.** The energy gap and the refractive index of LiTaO$_3$ thin films.

| Annealing temperature (ºC) | Energy gap (eV) | Refractive index |
|---|---|---|
| *750* | 3.10 | 2.24 |
| *800* | 4.80 | 2.68 |
| *850* | 5.50 | 3.32 |

## 4. Conclusion

The thin layer from LiTaO$_3$ solution was deposited on p-type Si (100) substrates using Chemical Solution Deposition (CSD) technique and it was flattened at 4000 rpm for 30 seconds using spin coater device. Then, crystallization of LiTaO$_3$ thin layer was formed because of annealing temperature treatment. Further, the crystalline structure was characterized by X-Ray Diffraction (XRD) and the optical properties were identified based on measurement using spectrometer (Ocean Optics USB4000-UV-VIS). The study of crystalline structure exhibits that thin films were polycrystalline with the direction of orientation along (104) and (110). Moreover, optical properties indicated the sensitiveness to the visible and ultraviolet light. For its application as the visible and ultraviolet light sensor, these thin films can improve the performance of satellite systems that contribute to the optical remote sensing.

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