Crystalline structure and optical properties of thin film LiTaO3

by Nani Djohan

Crystalline structure and optical properties of thin film $LiTaO_3$

IOP Conf. Series: Earth and Environmental Science **284** (2019) 012039 doi:10.1088/1755-1315/284/1/012039

Crystalline structure and optical properties of thin film $LiTaO_3$

N Djohan[1*], R Estrada[1], N Sevani[2], H Hardhienata[3] and Irzaman[3]

[1] Department of Electrical Engineering, Faculty of Engineering and Computer Science, Krida Wacana Christian University, Jl. Tj. Duren Raya No. 4, Jakarta 11470, Indonesia

[2] Department of Information Technology, Faculty of Engineering and Computer Science, Krida Wacana Christian University, Jl. Tj. Duren Raya No. 4, Jakarta 11470, Indonesia

[3] Department of Physics, Faculty of Mathematics and Natural Sciences, Bogor Agricultural University, Jl. Raya Dramaga, Bogor 16680, Indonesia

E-mail: nani.djohan@ukrida.ac.id

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 × 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 (λ_{Cu}) is 1.54060 Å. The spectrum was recorded at a diffraction angle of 2θ from a range of $10^o \leq \theta \leq 80^o$ 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 $LiTaO_3$ thin films with the Cramer and Cohen equations [26-28].

$$\begin{aligned} \sum \alpha \sin^2\theta &= C\sum\alpha^2 + B\sum\alpha\gamma + A\sum\alpha\delta \\ \sum \gamma \sin^2\theta &= C\sum\alpha\gamma + B\sum\gamma^2 + A\sum\gamma\delta \\ \sum \delta \sin^2\theta &= C\sum\alpha\delta + B\sum\gamma\delta + A\sum\delta^2 \end{aligned} \quad (1)$$

with: α, γ, δ= lattice parameters (Å)
θ = Bragg angle (degree)
A, B, C = lattice constants

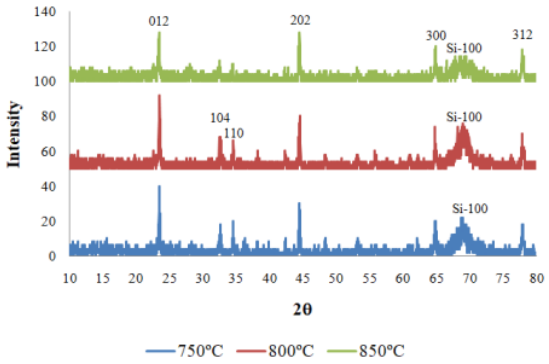

Figure 1. XRD patterns of $LiTaO_3$ thin films with different annealing temperatures.

Table 1 shows the lattice parameter values of $LiTaO_3$ 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 $LiTaO_3$ thin films.

Lattice parameter	$LiTaO_3$ annealing at 750°C (Å)	$LiTaO_3$ annealing at 800°C (Å)	$LiTaO_3$ annealing at 850°C (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 $LiTaO_3$ thin films can be calculated based on the FWHM of the diffraction peaks produced using the Scherrer equation [17]:

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

with: D = crystallite size (Å)
K = constant (0.96)
λ = wavelength (1.506 Å)
β = the graph width of FWHM (radian)
θ = 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 (°C)	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]:

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

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

with: α = 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 (6.626×10^{-34} J·s)
c = speed of light (2.998×10^{8} m·s^{-1})
λ = 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]:

$$n_e^{\,2}(\lambda,T) = A + \frac{B + b(T)}{\lambda^2 - [C + c(T)]^2} + \frac{E}{\lambda^2 - F^2} + \frac{G}{\lambda^2 - H^2} + D\,\lambda^2 \quad (5)$$

with: n_e = refractive index
A, B, C, D, E, F, G, H = constants (Sellmeier coefficients)
$b(T), c(T)$ = temperature dependent
λ = wavelength (μm)

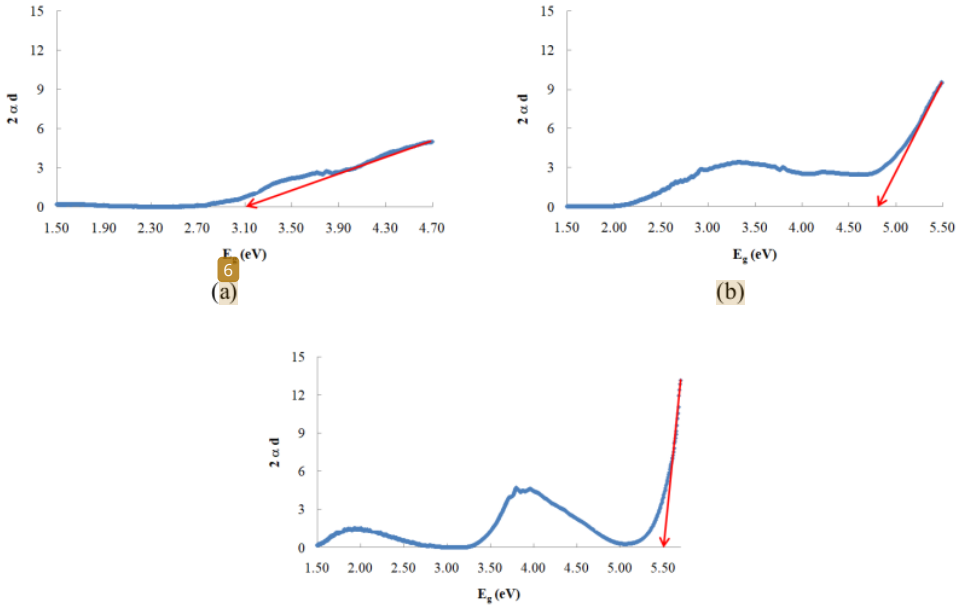

Figure 2. Energy gap of $LiTaO_3$ thin films: (a) 750ºC (b) 800ºC (c) 850ºC.

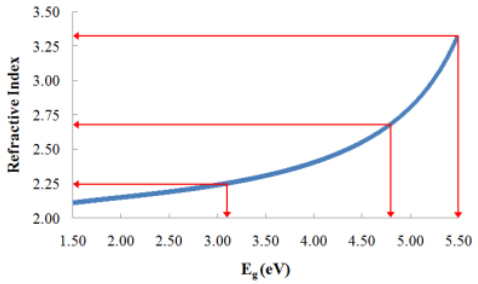

Figure 3. Refractive index of $LiTaO_3$ thin films with annealing treatment at 750ºC, 800ºC, and 850ºC.

Table 3. The energy gap and the refractive index of $LiTaO_3$ thin films.

Annealing temperature (°C)	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.

Acknowledgment

This study was supported by the Ministry of Research, Technology and Higher Education of the Republic of Indonesia through the Research Grant No. 005/KM/PNT/2018.

References

[1] Djohan N, Estrada R, Sari D, Dahrul M, Kurniawan A, Iskandar J, Hardhienata H and Irzaman 2017 The effect of annealing temperature variation on the optical properties test of $LiTaO_3$ thin films based on Tauc Plot method for satellite technology *IOP Conf. Ser.: Earth Environ. Sci.* **54** 012093

[2] Irzaman, Pebriyanto Y, Apipah E R, Noor I, and Alkadri A 2015 Characterization of Optical and Structural of Lanthanum Doped $LiTaO_3$ Thin Films *Integrated Ferroelectrics* **167** 137

[3] Ismangil A, Jenie R P, Irmansyah, Irzaman 2015 Development of lithium tantallite ($LiTaO_3$) for automatic switch on LAPAN-IPB Satellite infra-red sensor *Procedia Environ. Sci.* **24** 329

[4] Ismangil A, Irmansyah, Irzaman 2016 The diffusion coefficient of lithium tantalite ($LiTaO_3$) with temperature variations on LAPAN-IPB satellite infra-red sensor *Procedia Environ. Sci.* **33** 668

[5] Irzaman, Maddu A, Syafutra H, dan Ismangil A 2010 Uji Konduktivitas Listrik dan Dielektrik Film Tipis Lithium Tantalate ($LiTaO_3$) yang Didadah Niobium Pentaoksida (Nb_2O_5) Menggunakan Metode Chemical Solution Deposition *Prosiding Seminar Nasional Fisika* 175

[6] Estrada R, Djohan N, Pasole D, Dahrul M, Kurniawan A, Iskandar J, Hardhienata H and Irzaman 2017 The optical band gap of $LiTaO_3$ and Nb_2O_5-doped $LiTaO_3$ thin films based on Tauc Plot method to be applied on satellite *IOP Conf. Ser.: Earth Environ. Sci.* **54** 012092

[7] Inaguma Y, Matsui Y, Shan Y -J, Itoh M, Nakamura T 1995 Lithium ion conductivity in the perovskite-type $LiTaO_3$–$SrTiO_3$ solid solution *Solid State Ionics* **79** 91

[8] Su Y, Lang J, Du C and Wang X 2016 *Perovskite Materials - Synthesis, Characterisation, Properties, and Applications* ed L Pan and G Zhu (Rijeka: InTech) **Chapter 16** 489

[9] Tarafder A, Annapurna K, Chaliha R S, Tiwari V S, Gupta P K, Karmakar B 2010 Structure, dielectric and optical properties of Nd^{3+}-doped $LiTaO_3$ transparent ferroelectric glass–ceramic nanocomposites *J. Alloy. Compd.* **489** 281

[10] Aoyagi S, Osawa H, Sugimoto K, Iwata M, Takeda S, Moriyoshi C, and Kuroiwa Y 2015 Crystal structure analysis of $LiTaO_3$ under electric field *Jpn. J. Appl. Phys.* **54** 10NB03
[11] Sari F I W, Djohan N, Kurniawan A, Rohaeti E, Irzaman 2015 Analisis Energy Gap dan Indeks Bias $LiTaO_3$ Didadah Ga_2O_3 Berdasarkan Metode Reflektansi *Prosiding Seminar Nasional Fisika dan Aplikasinya (Jatinangor)* **1** FM-12
[12] Irzaman, Irmansyah, Syafutra H, Arif A, Alatas H, Astuti Y, Nurullaeli, Siskandar R, Aminullah, Sumiarna G P A, Jamal Z A Z 2014 Effect of annealing times for $LiTaSiO_5$ thin films on structure, nano scale grain size and band gap *Am. J. Mater. Res.* **1** 7
[13] Fang S, Wang B, Zhang T, Ling F, Wang R 2006 Growth and photorefractive properties of Zn, Fe double-doped $LiTaO_3$ crystal *Opt. Mater.* **28** 207
[14] Irzaman, Darvina Y, Fuad A, Arifin P, Budiman M, and Barmawi M 2003 Physical and pyroelectric properties of tantalum-oxide-doped lead zirconium titanate $[Pb_{0.9950}(Zr_{0.525}Ti_{0.465}Ta_{0.010})O_3]$ thin films and their application for IR sensors *Phys. Stat. Sol. (A)* **199** 416
[15] Misbakhusshudur M, Ismangil A, Aminullah, Irmansyah, Irzaman 2016 Phasor diagrams of thin film of $LiTaO_3$ as applied infrared sensors on satellite of LAPAN-IPB *Procedia Environ. Sci.* **33** 615
[16] Wendong Z, Qiulin T, Jun L, Chenyang X, Jijun X, Xiujian C 2010 Two-channel IR gas sensor with two detectors based on $LiTaO_3$ single-crystal wafer *Opt. Laser Technol.* **42** 1223
[17] Fakhri M A, Al-Douri Y, Hashim U, Salim E T 2016 XRD Analysis and Morphological Studies of Spin Coated $LiNbO_3$ Nano Photonic Crystal Prepared for Optical Waveguide Application *Adv. Mater. Res.* **1133** 457
[18] Gou J, Wang J, Huang Z H and Jiang Y D 2013 Preparation of $LiTaO_3$ Nano-Crystalline Films by Sol-Gel Process *Key Eng. Mater.* **531-532** 446
[19] McClain C R, Meister G, Monosmith B 2014 Satellite Ocean Color Sensor Design Concepts and Performance Requirements *Exp. Methods Phys. Sci.* **47** 73
[20] Watson I M, Prakash A 2015 *Monitoring Volcanoes in the North Pacific: Observations from Space* ed K G Dean and J Dehn (Berlin: Springer-Verlag Berlin Heidelberg) **Chapter 2** 27
[21] Thies B and Bendix J 2011 Satellite based remote sensing of weather and climate: Recent achievements and future perspectives *Meteorol. Appl.* **18** 262
[22] Irzaman, Siskandar R, Nabilah N, Aminullah, Yuliarto B, Hamam K A, and Alatas H 2018 Application of lithium tantalate ($LiTaO_3$) films as light sensor to monitor the light status in the Arduino Uno based energy-saving automatic light prototype and passive infrared sensor *Ferroelectrics* **524** 44
[23] Irzaman, Sitompul H, Masitoh, Misbakhusshudur M, and Mursyidah 2016 Optical and structural properties of lanthanum doped lithium niobate thin films *Ferroelectrics* **502** 9
[24] Irzaman, Siskandar R, Aminullah, Irmansyah, and Alatas H 2016 Characterization of $Ba_{0.55}Sr_{0.45}TiO_3$ films as light and temperature sensors and its implementation on automatic drying system model *Integr. Ferroelectr.* **168** 130
[25] Irzaman, Putra I R, Aminullah, Syafutra H, Alatas H 2016 Development of ferroelectric solar cells of barium strontium titanate ($Ba_xSr_{1-x}TiO_3$) for subtituting conventional battery in LAPAN-IPB Satellite (LISAT) *Procedia Environ. Sci.* **33** 607
[26] Cullity B D, Stock S R 2014 *Elements of X-Ray Diffraction* (Harlow: Pearson Education Ltd)
[27] Suryanarayana C and Norton M G 1998 *X-Ray Diffraction: A Practical Approach* (New York: Springer Science+Business Media)
[28] Spinolo G and Maglia F 1999 Cohen's method revisited *Powder Diffr.* **14** 208
[29] Korotcov A, Chen R -S, Hsu H -P, Huang Y -S, Tsai D -S, and Tiong K -K 2007 Well-Aligned IrO_2 Nanocrystals *J. Nanomater.* **2007** 84845
[30] Mulyadi, Rika W, Sulidah, Irzaman, Hardhienata H 2017 Barium Strontium Titanate Thin Film Growth with rotational speed variation as a satellite temperature sensor prototype *IOP Conf. Ser.: Earth Environ. Sci.* **54** 012094

[31] Bruner A, Eger D, Oron M B, Blau P, Katz M, Ruschin S 2003 Temperature-dependent Sellmeier equation for the refractive index of stoichiometric lithium tantalate *Opt. Lett.* **28** 194

[32] Weng W -L, Liu Y -W, Zhang X -Q 2008 Temperature-Dependent Sellmeier Equation for 1.0 mol% Mg-Doped Stoichiometric Lithium Tantalate *Chin. Lett.* **25** 4303

Crystalline structure and optical properties of thin film LiTaO3

ORIGINALITY REPORT

12%	9%	9%	4%
SIMILARITY INDEX	INTERNET SOURCES	PUBLICATIONS	STUDENT PAPERS

PRIMARY SOURCES

1	Submitted to Institut Pertanian Bogor Student Paper	2%
2	prame.be Internet Source	2%
3	www.researchgate.net Internet Source	1%
4	www.tandfonline.com Internet Source	1%
5	dokumen.pub Internet Source	1%
6	Encyclopedia of Sustainability Science and Technology, 2012. Publication	1%
7	Irzaman, Yunus Pebriyanto, Epa Rosidah Apipah, Iman Noor, Ahmad Alkadri. " Characterization of Optical and Structural of Lanthanum Doped LiTaO Thin Films ", Integrated Ferroelectrics, 2015 Publication	1%

8	repository.ipb.ac.id Internet Source	1 %
9	Weng Wen-Le. "Temperature-Dependent Sellmeier Equation for 1.0 mol% Mg-Doped Stoichiometric Lithium Tantalate", Chinese Physics Letters, 12/2008 Publication	1 %
10	digilib.unimed.ac.id Internet Source	1 %
11	www.mdpi.com Internet Source	1 %

Exclude quotes	Off	Exclude matches	< 1%
Exclude bibliography	On		