PERINTIS eJournal

PERINTIS eJournal, **2016**, Vol. 6, No. 2, pp. 91-100

OPTIMIZATIONS OF THERMOLUMINESCENCE RESPONSE OF DYPROSIUM DOPED LITHIUM MAGNESIUM BORATE DOSIMETER SUBJECTRED TO COBALT-60 GAMMA RAY

Ratna Suffhiyanni Omar*, Mohammad Alam Saeed*, Husin Wagiran, Hayder Khudhair Obayes

Department of Physics Sciences, Faculty of Science, Universiti Teknologi Malaysia, 81310 Skudai, Johor Bahru, Malaysia

*Corresponding Author: ratnasuffhiyanni@gmail.com; moalsd@gmail.com

ABSTRACT

There are a number of thermoluminescence (TL) materials have been investigated for its capability to be used as radiation dosimeter. These TL materials should be affordable cost and good reproducibility compared to available personal dosimeters nowadays. One of the TL materials that can be considered is using glass compound. In this study lithium as the modifier in magnesium borate glasses with the present of dysprosium as the activator was investigated to be performed as TL dosimeter. In this work, preliminary studied was carried out to determine the optimum time temperature profile (TTP) setting and annealing procedure as well as determination of optimum dysprosium concentration of the TLD. Lithium magnesium borate glass systems of (70-y) B_2O_3 - 20 Li_2O – 10 MgO - (y) Dy_2O_3 with 0.05 mol% ≤ y ≤ 0.7 mol% were prepared by melt-quenching technique. The amorphous structure of the TLD was determine by using powder X-ray diffraction (XRD) analysis and the TL measurements were carried out using Harshaw 4500 TLD reader. The TLD reader was setup at 5 °Cs^{-1} of heating rate for the readout process. The samples were irradiated to Co-60 gamma source at a dose of 50 Gy. 200 °C and 60 minutes of annealing temperature and annealing time respectively were applied for the annealing process of the samples. Optimum TL intensity was recorded with a heating rate of 5 °Cs^{-1}. The highest TL responses and sensitivity were found for the sample with 0.1 mol% of Dy concentration.

Keywords: *lithium magnesium borate, Co-60 gamma ray, thermoluminescence.*

1.0 INTRODUCTION

Luminescence phenomenon is the emission of light from solid called phosphors or it refers to the process of absorbing energy and converting it into visible light. The luminescence studies on borate compound have been carried since 1997 [1, 2]. Considerable studies were focused on borate glass to enhance its TL emission. The term TL or temperature-stimulated light emission after removal of excitation is refers to a case of phosphorescence observed under condition of steadily increasing temperature. These modifications were based on changing the modifier, or activator. Consequently, valuable features of magnesium borate materials such as near tissue-equivalency, high sensitivity and good performance subjected to X-ray, gamma, beta and neutron were the base for further improvement of this TL material [3]. Moreover, borate glass offers an important role in TL properties as it has good heat stability and lower melting temperature compare to other glasses [4]. In addition, due to the developments of borate glass in the field of radiation protection dosimetry, the application of borate glasses is highly focused nowadays as a new TL material. This is because, borate glasses are relatively chemically stable compound and showed no serious problems to be doped with impurities such as rare earth, copper, manganese ions and showed high sensitivity, linearity and good fading properties [5, 6]. The important phase to improve the glass properties is the addition of modifiers as it has ability of boron to combine with three or four oxygen atoms to produce a variety of atom groups [7, 8, 9, 10].

The technique (melt-quenching technique) used to prepare the glasses in this study considered easier compared the other glass preparation techniques because of some reasons such as the glass preparation and handling can be considered simple and the ability to produce wide variety of new oxide glasses is high [11]. Therefore, this newly prepared TL material is aim to choose the modifier and dopant concentration that will enhance the TL response.

2.0 METHOD

Lithium magnesium borate glass systems of (70-y) B_2O_3 - (20) Li_2O – 10 MgO - (y) Dy_2O_3 with 0.05 mol% $\leq$ y $\leq$ 0.7 mol% was prepared by melt-quenching technique. The raw materials used for glass preparation are borate oxide (B_2O_3), magnesium oxide (MgO), lithium oxide (Li_2O), and dysprosium (III) oxide (Dy_2O_3). The samples were prepared from chemically pure grades of materials supplied by Acros Organic and QReC (reagent grade) with 99.9 % of purity. The reagents were weighed and mixed to prepare 10 g for each sample. The samples then had undergone a milling process for 1 hour in order to obtain homogenous mixture. Next, the mixture reagent is then placed in porcelain crucible and melted in electronic furnace for 1 hour at 1100 °C. Consequently, the melted materials were stirred frequently to ensure the complete homogeneity of the mixtures. Then, the melted glass was poured on the preheated stainless steel plate and cool rapidly to the room temperature. The glass samples were annealed at 350 °C for 4 hours. At the end of annealing process, the furnace is switched off and the glasses were allowed to cool down gradually inside the furnace to avoid thermal stress at an average rate of 10 °Cmin^{-1} to room temperature. For safety purposes, crucible, tongs, facemask and thermal gloves were used during inserting and extraction of the crucible to the furnace.

Various nominal compositions of undoped and Dy doped lithium magnesium borate glasses system was prepared in this study as shown in Table 1. Most of the samples are in the glassy form and stable.

Table 1: Composition of prepared glass

GLASS NOTATION	COMPOSITION (mol %)			
	B_2O_3	MgO	Li_2O	Dy_2O_3
LMB	70.00	10	20	0.00
LMB: 0.05DY	69.95	10	20	0.05
LMB: 0.10DY	69.90	10	20	0.10
LMB: 0.30DY	69.70	10	20	0.30
LMB: 0.50DY	69.50	10	20	0.50
LMB: 0.70DY	69.30	10	20	0.70

The prepared samples were cut off into small pieces and each sample was weighted and the average mass of every sample should be in the range of 0.0200 - 0.0300 g. Next, the samples were undergone the annealing process by placing the samples into a metal block trays and heated in a TLD-FURNACE type LAB-01/400 model using THERMOSOFT computer program. The heating time was fixed at 60 minute and the temperature was varied to 100, 200, 300 and 400 ˚C. The TL response of each annealing temperature was determined and the best annealing temperature of producing highest TL response was recorded. The best annealing temperature obtained from earlier procedure was set and the heating time was altered to 10, 20, 30, 40, 50 and 60 minute. After the annealing process, each of the samples were placed in a capsule, stored in lightproof plastic bottle and in black toolbox to avoid the physical or environment factors such as to natural light, temperature and humidity. The samples were constantly kept inside the toolbox until the readout process was performed.

2.1 X-Ray Diffraction (XRD) Analysis

X-ray Diffraction (XRD) technique was performed to confirm the amorphous state of the samples. The micro size powder samples were scanned by mean of X-ray diffraction method using *Siemens Difffraktometer* D5000, equipped with diffraction software analysis with CuK_α radiation operating at 40 kV and 30 mA with Bragg–Brentano geometry at room temperature. The samples were grinded into powder form by using mortar and pestle. The diffraction patterns are measured in steps 0.05° with 2 s counting time per step, for 2-theta (2 θ) ranging from 10° to 90°.

2.2 Determination of Time Temperature Profile (TTP) Setting of the TL Reader

One of the TTP setup of a TLD reader is heating rate. According to first order kinetics of a TLD, the increase of the heating rate may influence the decreases of the TL peak height and increase the width or full width at half maximum (FWHM) of the glow curve. Whereas, for second order kinetic, as the heating rate was elevated, a shift of the TL peak to lower temperature and the increase in the TL intensity can be observed [12]. Therefore, TL glow curve of second-order kinetic, the TL peak is wider and more symmetric than a first-order peak. This can be understood from the consideration of the fact that in a second-order kinetic process, a significant concentration of released electrons is re-trapped before recombined that can delay the thermoluminescence emission and spreading out of the emission over a wider temperature range [13].

In this study, the TL glow curves of various heating rate from 1 to 20 ºCs^{-1} were investigated for dysprosium doped lithium magnesium borate subjected to Co-60 gamma irradiation.

2.3 Determination of Optimum Concentration of Dy in Lithium Magnesium Borate Glass

Determination of optimum glass compositions were implemented to (70-y) B_2O_3 - (20) Li_2O – 10 MgO -(y) Dy_2O_3 with 0.05 mol% $\leq y \leq$ 0.7 mol% glass series by observing the TL glow curve after the samples were irradiated to 50 Gy Co-60 gamma ray.

3.0 RESULTS AND DISCUSSION

3.1 X-ray Diffraction Analysis

The amorphous state of glass sample was confirmed by using X-ray diffraction (XRD) analysis. Figure 1 illustrates the XRD pattern observed undoped and dysprosium doped lithium magnesium borate (LMB: 0.1 mol% Dy). Generally, two major scattered peaks were observed at lower angle and shifted to higher angle as the dopant (Dy) is added to the glass system. It was also illustrated that that the XRD spectra exhibit a broad diffuse scattering at lower angles. From Figure 1, it can be observed that this glass sample revealed bands with no discrete or continuous sharp peaks which indicated its amorphous character.

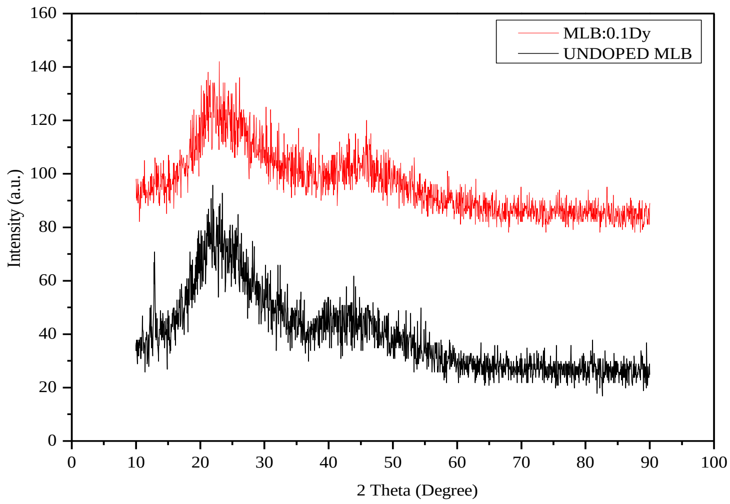


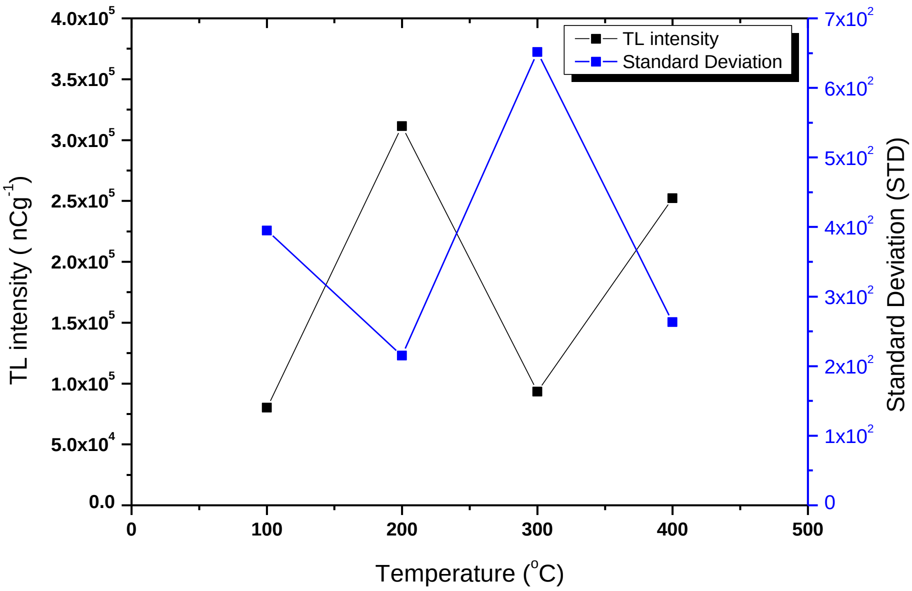
Figure 1: XRD pattern of undoped and dysprosium doped lithium magnesium borate (LMB : 0.1 mol% Dy)

PERINTIS eJournal, **2016**, Vol. 6, No. 2, pp. 91-100

3.2 Annealing Procedure Before Irradiation Analysis

In this work the optimum annealing temperature and annealing time were determined to sample LMB: 0.1 Dy obtains the best annealing procedure of the sample. The sample was irradiated at a dose of 50 Gy subjected to Co-60 gamma ray. The TL responses and its standard deviations were plotted against annealing temperature and annealing time as shown in Figure 2(a) and Figure 2(b) respectively. It can clearly be seen that the highest TL intensity is achieved at temperature 200 ºC which gives lowest standard deviation (STD) compared to other annealing temperatures as shown in Figure 2(a). By using this optimum annealing temperature, the heating times were varied and the highest TL intensity was achieved at a heating time of 60 min. Therefore, the annealing procedure applied for this sample is 200 °C for 60 min.

a)



b)

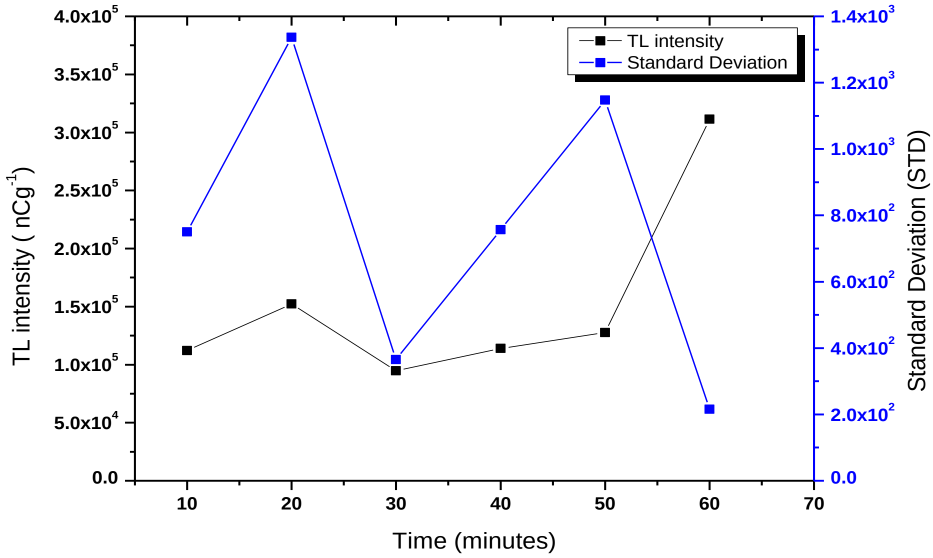


Figure 2: TL response and its standard deviation as a fuction of (a) annealing temperature and (b) annealing time of LMB : 0.1 mol% Dy

3.3 Time Temperature Profile (TTP) Analysis

Figure 3 indicates the TL glow curves of LMB: 0.1 mol% Dy glass at various heating rates after the sample was irradiated at a dose of 50 Gy subjected to cobalt-60 gamma ray. Differences in the shape of the TL glow curves were observed. It seems that broad peak become narrow as the heating rate increases. It can be seen that the highest TL response was observed when the heating rate was set at 1 °Cs^{-1}. However, it was also observed that the TL glow curve with heating rate of 1 °C s^{-1} the TL peak shifted to the lower temperature at around 150 °C. This peak temperature is not meets the requirement of TL peak for an ideal dosimetric characteristic as suggested by Garlick et al. [14]. Heating rate 7 °Cs^{-1} cannot be considered the best heating rate in this case because the glow curve is shifted to a higher temperature and it can be observed that the glow curve was not completed. Therefore, for LMB: Dy (0.1 mol%) the heating rate of 5 °Cs^{-1} was chosen as the best heating rate for the studied sample. This heating rate was selected due to it characteristic of the glow peak temperature which was located at around 180 – 220 °C compared to other heating rate. This heating rate can be considered to meet the requirement of the ideal dosimetric characteristic [14].

PERINTIS eJournal, **2016**, Vol. 6, No. 2, pp. 91-100

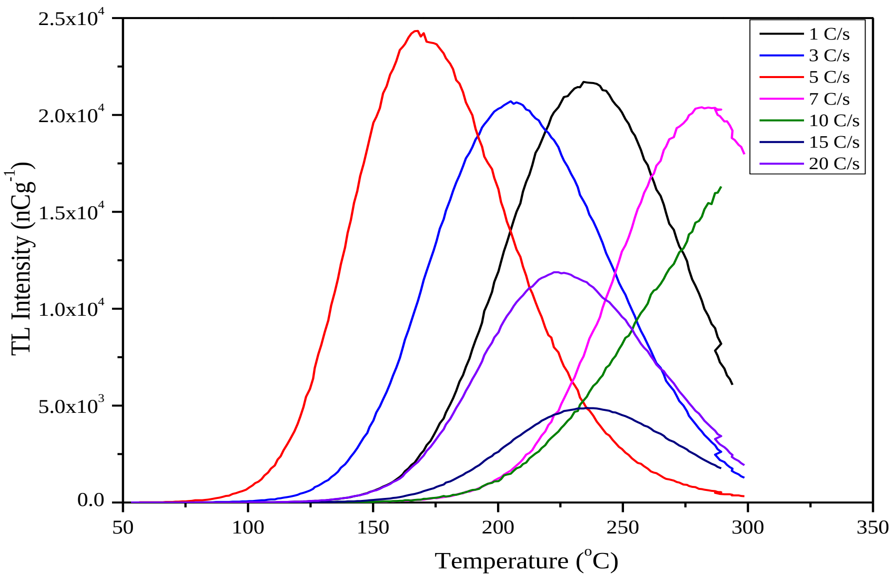


Figure 3: Graphs of TL glow curve for various heating rate

3.4 Optimum Dy concentration

Determination of optimum Dy concentration was carried out for (70-y) B_2O_3 - (20) Li_2O – 10 MgO-(y) Dy_2O_3 with 0.05 mol% $\leq$ y $\leq$ 0.7 mol% glass series by observing the TL glow curve response at different Dy concentration. The samples were irradiated to Co-60 gamma ray at dose of 50 Gy.

Figure 4 illustrates the glow curves of LMB: Dy glass samples at various Dy concentrations. From the figure, it can be seen clearly that the sample with 0.1 mol% Dy has highest TL response compared to the other samples. The TL glow curve peak was located at around 190 °C, which has meet the ideal TL glow curve and has a favourable property as a radiation dosimetry potential [15]. Therefore, LMB: Dy glass with a Dy concentration of 0.1 mol % was the best glass composition for TLD. Further investigated of its TL properties such as sensitivity, linearity, reproducibility, detectable limit, fading and etc. have to be carried to investigate the performance of this sample to be used as TLD.

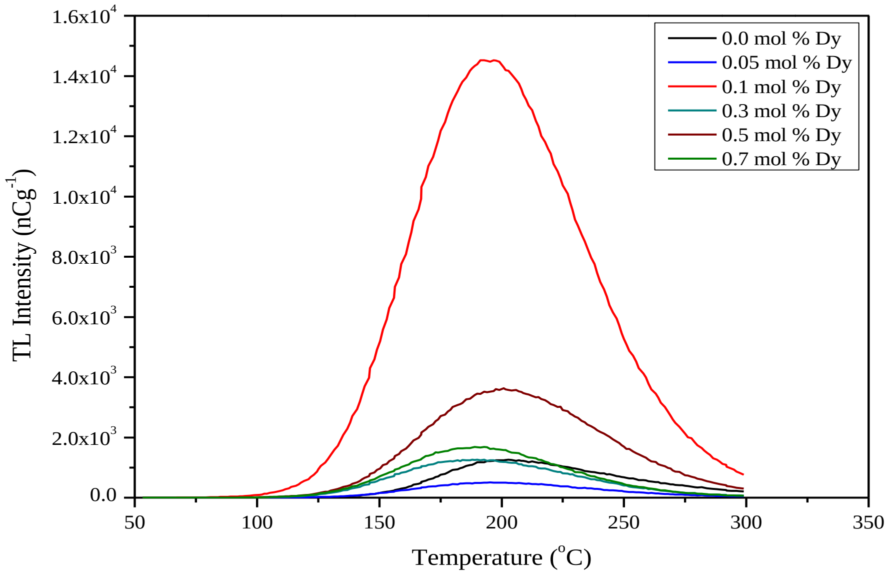


Figure 4: Graph of TL glow curve of optimum concentration of Dy in lithium magnesium borate glass system

3.5 Sensitivity

The intrinsic TL sensitivity of a TLD is expressed as the TL yield or intensity per unit mass per unit dose (nCg-^{1}Gy^{-1}). The sensitivities of Dy doped lithium magnesium borate at various Dy concentrations subjected to Co-60 gamma irradiation at a dose of 50 Gy are calculated and the results are plotted as shown in Figure 5. It was found that the highest TL sensitivity was recorded for a sample with 0.1 mol% Dy concentrations. The sensitivity of this sample was about 8.7 times higher than that of undoped sample. Therefore, the existence of dysprosium doped lithium magnesium borate glass contributed to the enhancement of the TL response.

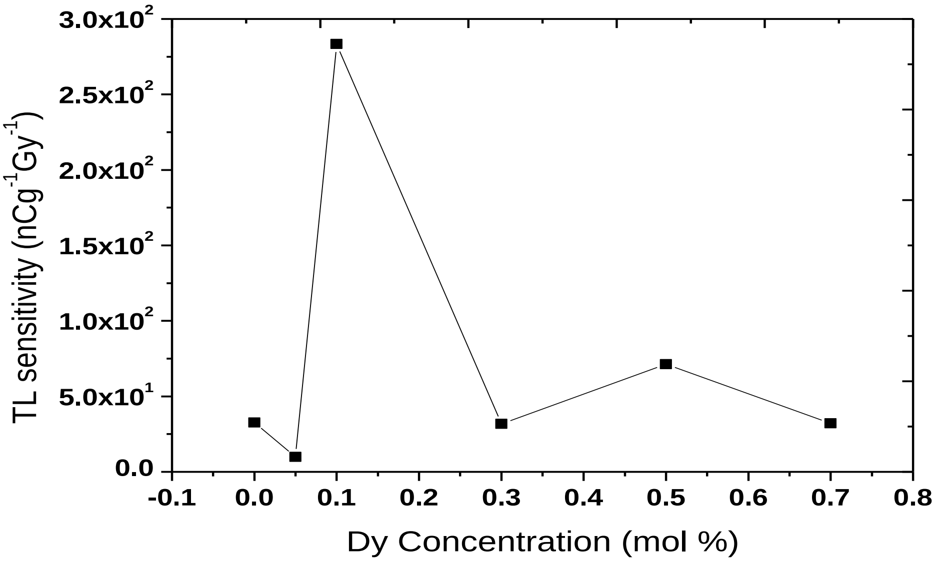


Figure 5: TL sensitivity versus concentration of Dy in LMB glass

4.0 CONCLUSION

The present work was carried out towards a systematic study of the TL properties of lithium magnesium borate glass doped with Dy to be performed as TLD. Preliminary work was done to determine the optimum TTP settings parameters of the TLD. The optimum concentration of Dy in lithium borate was also determined in this work. The best TL response and sensitivity were recorded for the glass sample that doped with 0.1 mol% Dy.

ACKNOWLEDGEMENT

The authors would like to express their appreciation to the Malaysia Ministry of Education (MOE) and Universiti Teknologi Malaysia for the support of the sponsors with Project No Q.J130000.2526.12H87.

REFERENCES

[1] Kazankaya, V. A., Kuzmin, V. V., Minaevaand, E.E., and Sokolov, A. D. (1974). Proceeding of Fourth International Conference on Luminescence Dosimetry. Krakow, Poland: *Polish Academy of Sciences*, 581.

[2] Furetta, C. and Weng, P.S. (1998). Operational thermoluminescence dosimetry. London: World Scientific.

[3] Prokic, M. (1980). Development of highly sensitive CaSa4: Dy/Tm and MgB4O7: Dy/Tm sintered thermoluminescent dosimeters. *Nucl. Instrum. Meth*, 175, 83-86.

[4] Zarand, P., and Polgar I. (1984). The relative standard deviation of TLD systems. *Nucl. Instrum Meth*, 222, 567-573.

[5] Garrett, D. E. (1998). Borates: Handbook of deposits, processing, properties, and use. Academic Press.

[6] Anishia, S. R., Jose, M. T., Annalakshmi, O., & Ramasamy, V. (2011). Thermoluminescence Properties of Rare Earth doped Lithium Magnesium Borate Phosphors. *Journal of Luminescence*, 131(12), 2492-2498.

[7] Rasid, A. A. (2014). Dosimetric Properties of Dyprosium-doped lithium sodium borate and lithium borate glasses subjected to photon irradiation. *Doctor Philisophy*, Universiti Teknologi Malaysia, Skudai.

[8] Wright, A. C., Simmons, C. J., and El-Bayoumi, O. H. (1993). Experimental techniques of glass science. *The American Ceramic Society*, 205.

[9] Dubovik, M.F., Tolmachev, A.V., Grin, L.A., & Dolzhenkov, E.F. (2000) Quant. Electron. *Optoelectron*, 3(3), 420.

[10] Dietzel, A. (1968). Glass Structure and Glass Properties. *Glastech. Ber*. 22, 41.

[11] Sahar, R. M. (2000). Fizik Bahan Amorfus. *Penerbit Universiti Teknologi Malaysia Press*, 34-36.

[12] Furetta, C., Kitis, G., Weng, P. S, and Chu T. C. (1999). Thermoluminescence characteristics of MgB4O7 : Dy,Na. Nuclear Instruments and Methods in *Physics Research. Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 420(3), 441–445.

[13] Chen, R., and McKeever, S.W.S. (1997). Theory of Thermoluminescence and Related Phenomena. Singapore. World Scientific.

[14] Garlick, G.F.J., and Gibson, A.F. (1948). The electron traps mechanism of luminescence in sulphide and silicate phosphors. *Proceeding of the physical society*, 60, 574-589.

[15] Wu, L., Chen, X.L., Tu, Q.Y., He, M., Zhang, Y., and Xu, Y.P. (2002). Phase Relations In The System Li2O–MgO–B2O3. *Journal of Alloys and Compounds*, 333(1-2), 154-158.