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Thermoluminescence (TL) dosimeter of dysprosium doped strontium borate glass for different glass modifiers (Na, Li, Ca) subjected from 1 to 9 Gy doses

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Abstract. This article reports TL response for different glass modifier and doping concentration. Alkali oxides (Na_2O and Li_2O) and alkali earth oxide (CaO) will be used as a glass modifier for strontium borate based glass. The samples were prepared by melt quenching technique. Dy_2O_3 concentrations ranging from 0.00 to 0.70 mol% and exposure doses of 1 to 9 Gy will be varied. All glass samples exhibit the prominent peak temperature positioned at 186 ºC to 232 ºC. From all the samples, one of the samples shows an excellent linearity dose response, higher TL and show good reproducibility after 5 cycles exposure which is sodium strontium borate doped with 0.1 mol% Dy_2O_3 (optimum concentration).

1 Introduction

Thermoluminescence dosimeter (TLD) has been widely applied in clinical, personal and environmental of ionizing radiation areas. Borate glass for better dosimeter materials were chosen due to is near tissue equivalent absorption coefficient, high thermal stability, low melting point and good solubility of rare earth ions [1]. The hygroscopic behavior of pure borate glass represents a serious drawback since humidity has often gives contrary effect on the performance of TL materials. Therefore, many investigations have been conducted to improve the performance of borate based dosimeters and explore the new TL materials. Strontium tetraborate is one of the potential candidates since it is not hygroscopic as normally exhibited by other borate compounds [2]. It is reported that the sensitivity of polycrystalline dysprosium doped strontium tetraborate is five times higher compared to LiF: Mg, Ti [3].

Up to now, there are no research study TLD properties on strontium borate glass system. So, in this study strontium borate glass with different modifiers alkali oxides (Na_2O and Li_2O) and alkali earth oxide (CaO) in glassy form and have investigated for the first time its thermoluminescence dosimeter (TLD) properties. Alkali oxide and alkali earth oxides are very good materials for modifying borate glasses because of their possible applications in radiation absorbent. [4,5]. The addition of dysprosium (Dy) as an activator was chosen in this research to enhance the high TL efficiency [6]. The aims of this study is to investigate the TL properties of dysprosium doped with different modifiers (Na_2O, Li_2O and CaO) of strontium borate glasses exposed to ^{60}Co Gamma irradiation. The best modifier of strontium borate glass with optimum concentration of Dy doping will gives the best TL properties such as glow curve, TL dose response, reproducibility, sensitivity to radiation and fading characteristics of the response. A small amount of dysprosium oxide (Dy_2O_3) was added to confirm the role as activator on TL dosimetry response.

2 Materials and Methods

2.1 Sample Preparation

In this study, the glass sample of Dy_2O_3 doped with different modifiers (Na_2O, Li_2O and CaO) of strontium borate at various Dy_2O_3 concentrations ranging from 0 to 0.7 mol% was synthesized. Table 1 shows the nominal compositions of the prepared samples. Melt quenching technique was used to prepare the glasses of compositions (70-x) B_2O_3- 20SrO -10 Na_2O - $x$$Dy_2O_3$, 10$Li_2O$ - 20 SrO - (70-x) B_2O_3 - (x)Dy_2O_3 and 10 CaO - 20 SrO - (70- x) B_2O_3- (x) Dy_2O_3, where, (x = 0.0, 0.1, 0.3, 0.5, 0.7) mol%. The commercially available raw materials in powder form were used. The high pure analar grade chemicals $SrCO_3$ (Aldrich 99.99%), B_2O_3 (Acros Organic 99%), Na_2CO_3 (Merck 99%), Li_2CO_3 (Acros Organic 99%),

$CaCO_3$ (QReC 99.9%) and Dy_2O_3 (Aldrich 99.99%). The required proportions of Na_2O, Li_2O, CaO, SrO, B_2O_3 and Dy_2O_3 powder was weighted using sensitive analytical balance. An appropriate mixture of 20 grams compound was placed in a bottle container. Then the mixture was mixed mechanically by milling machine in a homogenizer for about 1 hour to ensure the homogeneity of the mixture. The mixture was then poured into an alumina crucible and melted in high temperature furnace at 1100 °C for 20 minutes using the melt-quenching technique. The purpose of a high temperature anneal is required to clear the dosimetric traps of the residual signal, which may cause unwanted background during subsequent use of the dosimeters.

After the samples were totally melted, the glass samples were annealed for 4h at 400 °C below their calorimetric glass transition temperature to remove any external stresses and then cooled down to room temperature. Three batch series of samples with different modifiers and concentration of dysprosium dopant were prepared. The Dy_2O_3 as activator enhances the TL performance of the glass samples. After optimum concentration analysis of a single broad peak of TL glow curve is observed and it is located between 180 °C and 250 °C. It is in the same argument with previous research that an ideal glow curve should have a single sharp peak located between 180 °C and 250 °C [7]. The highest TL response with low standard error is observed and recorded for the Dy_2O_3 doped different modifiers of strontium borate glasses. After the analysis, it is found the highest TL response for different concentration Dy dopant were 0.1 mol% Dy doped sodium strontium borate glass, 0.1 mol% Dy lithium strontium borate glass and 0.3 mol% Dy doped calcium strontium borate glass. All of the highest TL response samples were labeled as **N1**, **L1** and **C2** respectively.

2.2 Gamma ray Irradiation

The glass samples were cut into small pieces with approximately same thickness and mass. They were weighted using an electronic analytical balance with an accuracy of 4 decimal places. Then each glass samples irradiated subjected to 50 Gy absorbed dose of ^{60}Co gamma ray at Universiti Kebangsaan Malaysia, UKM, Bangi. After 24 hours of irradiation, the samples were readout using TL reader Harshaw Model 4500 located at Nuclear Laboratory, Physics Department, Faculty of Science, Universiti Teknologi Malaysia. The linear time temperature profile (TTP) of each batch sample were different because different modifiers' presence in strontium borate glass. After few trials of TL readouts using various heating rates, N1 is 3 °Cs^{-1}, L1 is 5 °Cs^{-1}, and C2 is 7 °Cs^{-1} that gives the best resolution of the glow curve. The TL readings were then normalized to their mass.

3 Result and Discussion

3.1 X- ray Diffraction Analysis

X-ray diffraction analysis (XRD) was used to investigate the amorphous nature of the prepared glass samples. The powder samples of prepared glass were analyzed using Siemens Diffraction D5000, equipped with diffraction software analysis with Cu radiation (= 1.54045 Å) in range $5^o < 2\theta < 80^o$. This equipment is located in Material Science Laboratory, Faculty of Mechanical, Universiti Teknologi Malaysia (UTM). The XRD diffractograms were obtained by one second for each counting time powered at 40 kV and 30 mA at room temperature (300K) by step scanning 0.05^0. **Fig. 1** shows the XRD patterns of the samples. No sharp Bragg peaks are exhibited in any spectrum. The result proves that the samples were purely amorphous and did not exhibit any crystalline phase.

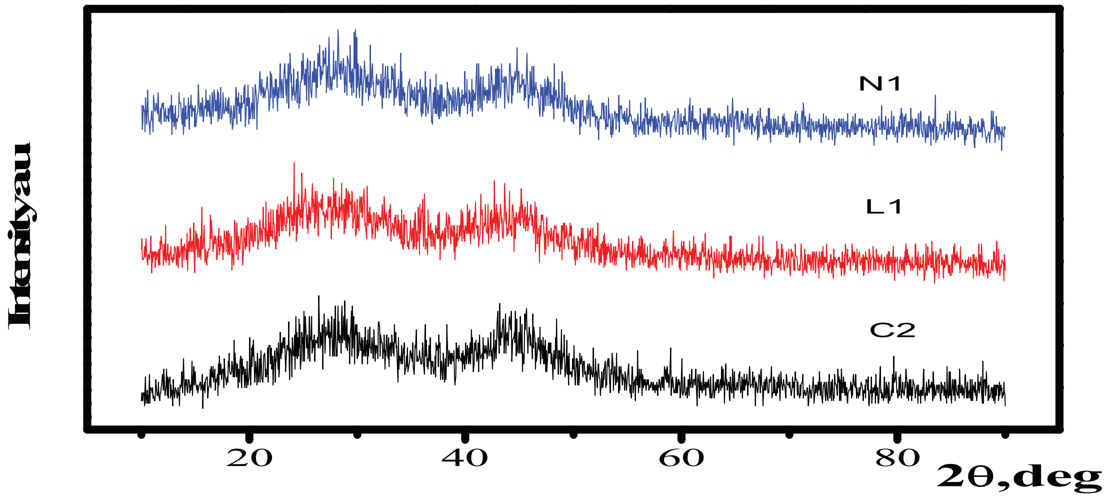

Fig 1:XRD patterns of optimum Dy-doped concentration on different modifiers of strontium borate glass.

3.2 Differential Thermal Analysis (DTA)

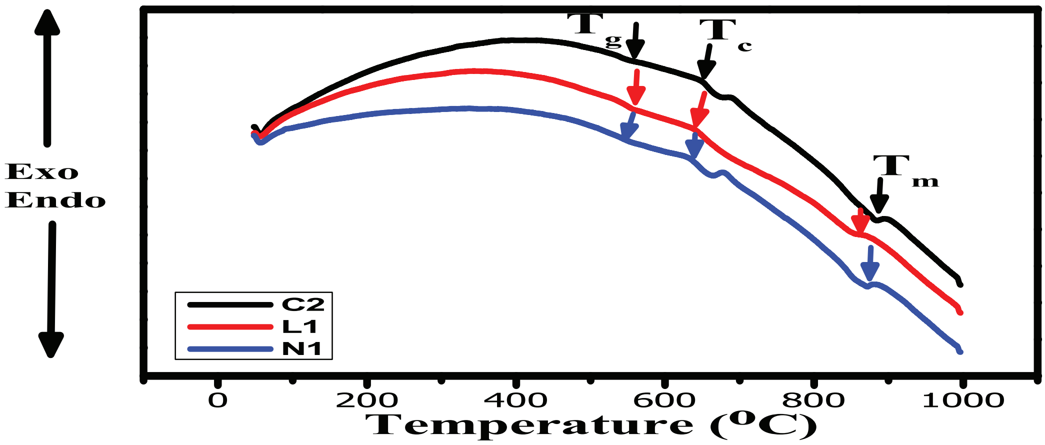

Fig 2: DTA traces of Dy-doped on different modifiers of strontium borate glasses.

Glass forming ability and stability of the prepared glass samples were measured using differential thermal analysis (DTA) curves Pyris STA 8000 analyzer. The endothermic peak corresponding to the glass temperature, T_g, melting temperature, T_m, exothermic peak of the crystallization temperature, T_c and glass transition temperature, T_{rg}, of the glass samples were traced using DTA analysis curves (**Fig. 2**).

Table 1: Thermal stability glass forming ability and Hruby number of the optimum dy-doped different modifier of strontium borate glasses

Sample No.	$T_g \pm 0.01(^oC)$	$T_c \pm 0.01\ (^oC)$	$T_m \pm 0.01(^oC)$	$\Delta T \pm 0.01(^oC)$	$T_{rg} \pm 0.01(^oC)$	$H_g \pm 0.01(^oC)$
N1	542.69	653.96	864.80	111.27	0.63	0.53
L1	549.99	637.88	854.52	87.89	0.64	0.41
C2	551.18	671.30	881.11	120.12	0.63	0.58

Glass forming ability is estimated using Kauzmann relation:

$$T_{rg} = T_g / T_m \tag{1}$$

In this study, the values of T_{rg} appear in the range of 0.63 to 0.64 and Hurby relation, H_g varies from 0.53 to 0.58. The calculated glass forming ability and thermal stability are summarized in **Table 1**. Glass forming ability is said to be good if it is in the range $1/2 \leq T_{rg} \leq 2/3$. Kauzmann criterion is well-obeyed for the present glass composition. Glass stability is said to be very poor for $H_g \leq 1$ and superior for $H_g \geq 0.5$.

3.3 TL dose response

The TL response of sample (L1,N1 and C2) were exposed from 1 Gy to 9 Gy doses range of gamma irradiation. Every data point represent an average of five individual glass readings. The TL response is slightly energy dependent, and the value of TL response tends to spread wider at higher energies from 1 Gy up to 9 Gy doses. **Fig. 3** show that sample C2 gives a linear and the highest TL dose response at high energies compared to sample N1 and L1. As the dose range increases from 1 Gy to 9 Gy, the TL intensity of glass sample N1 also increases linearly.

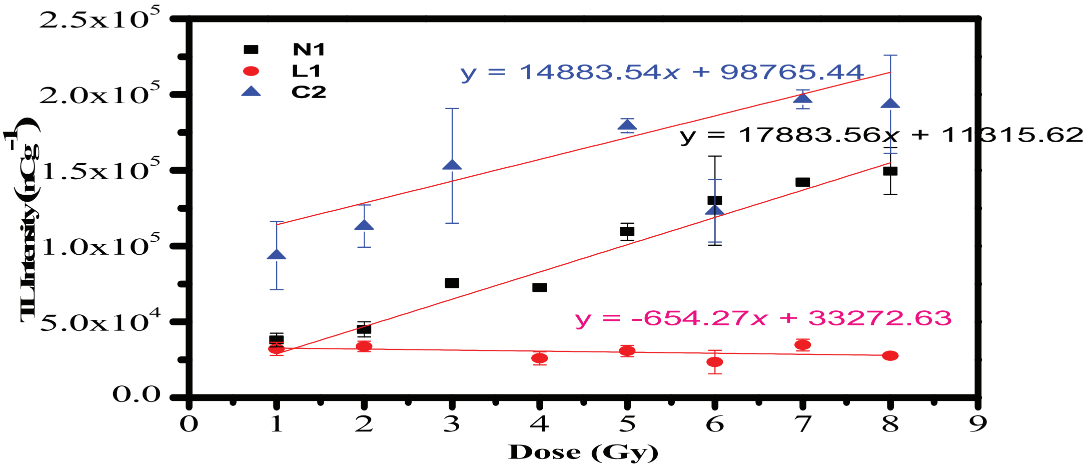

Fig. 3. TL dose response for low dose range from 1 Gy to 9 Gy doses range.

3.4 Sensitivity

Table 2: Sensitivity and the relative sensitivity of low dose energies of N1, L1, and C2 glass samples from 1 to 9 Gy.

TLD Material	Range of Dose	Sensitivity (nC g^{-1} Gy^{-1})	Regression Coefficient, R^2	Relative Sensitivity with Respect to TLD-100
N1		0.018 x 10^6	0.92	0.016
L1	1 to 9 Gy of ^{60}Co gamma ray	0.0065 x 10^6	0.19	0.00058
C2		0.015 x 10^6	0.85	0.013
TLD-100		1.12 x 10^6	0.93	1

TL sensitivity is defined as TL intensity signal over certain temperature region per unit absorbed dose per unit mass (nC g^{-1} Gy^{-1}). The TL sensitivity signal can directly determine from the slope of the graph of TL intensity response as a function of dose. **Table 2** summarized sensitivity and the relative sensitivity of low dose energies of N1, L1, and C2 glass samples from 1 to 9 Gy doses. From our observation, sample N1 has the highest relative sensitivity. It shows that the sensitivity sample. N1 is slightly near to TLD-100. TL materials with high sensitivity allow for their utilization in the assessment of low doses [8,9].

3.5 Reproducibility

Reproducibility is one of the essential characteristics of TL properties to determine the reusability of the glass samples. Thus, TL intensity signal of a particular dose should be approximately the same after several measurements. A TL material is expected to not suffer any physiochemical change due to repeated irradiation, readout and annealing process [10]. **Fig 5** parade different samples (N1, L1, C2) at a dose 5 Gy of gamma irradiation after five times sequential cycles. There is no difference between the results of the sequential measurement of low dose at 5Gy. The result confirms that these glass materials are reusable in radiation dosimeter.

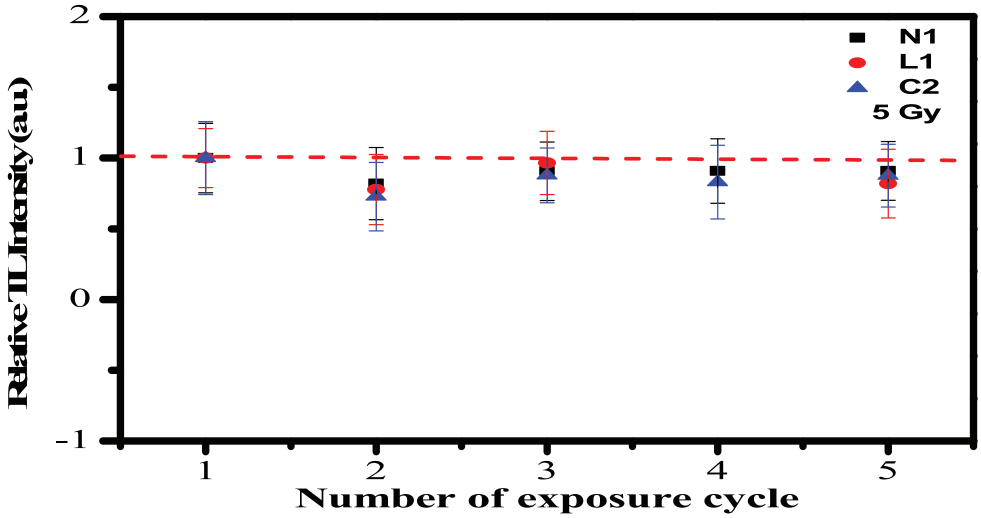

Fig 5. Reproducibility of different glass samples irradiated at 5 Gy doses of gamma irradiation.

3.6 Thermal Fading

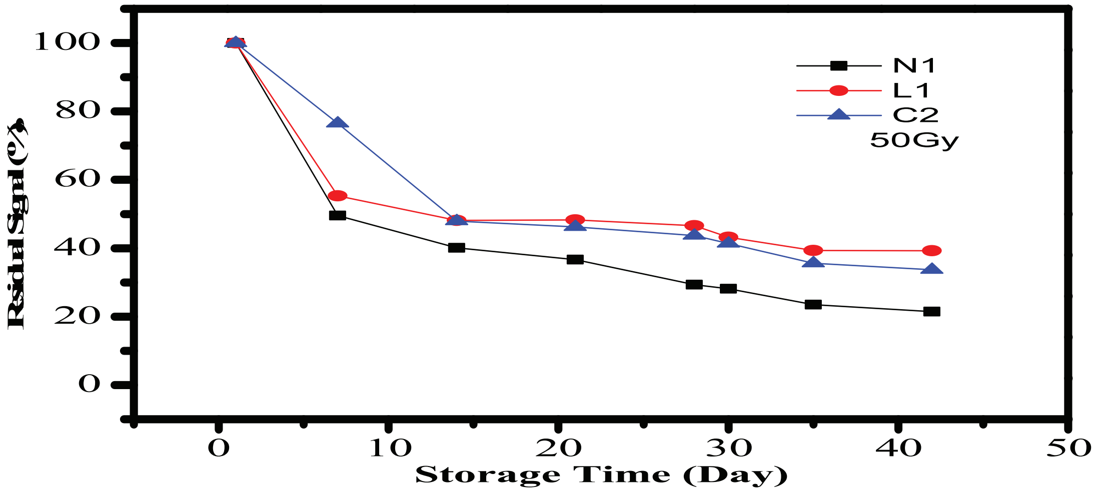

Fig 6. Thermal fading pattern after 42 day of exposure at 5 Gy for L1, N1 and C2.

Thermal fading is a reduction of the TL intensity signal due to the effect of ambient temperature after a period [11]. The fading behavior of samples (N1, L1, C2) up to 42 day is illustrated in **Fig. 6**. After irradiation, the samples were stored in a dark room at room temperature to minimize the possible escape of trapped electrons due to the simulation of temperature and time. It can be seen the TL loss signals after 42 day. It shows that the TL signal rapidly drops in the first 7 days and decrease slowly until day 42 are observed in every sample. Therefore, the samples need to be readout immediate after irradiation.

4. Conclusion

The basic TL dosimeter properties of strontium borate glass system doped with various concentration of Dysprosium (Dy) has been investigated subjected to 1 Gy to 9 Gy doses range of ^{60}Co gamma irradiation. The optimum concentration of Dy has been identified for $NaSr(BO_3)_3$, $LiSr((BO_3)_3$ and $CaSr((BO_3)_3$ are 0.1 mol% (N1), 0.1 mol% (L1) and 0.3 mol% (C2). The XRD analysis confirms the amorphous state of the glass samples and DTA analysis shows for each glass samples in a stable state. The variation of TL dose response, sensitivity, reproducibility and thermal fading has been identified for all batch series of glass samples. Comparison on the three modifiers shows that sodium strontium borate glass doped with 0.1 mol% of dysprosium (N1) oxide gives the best TL dosimeter response. It proves that the addition of different modifier influences the TL dosimeter response.

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