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THERMOLUMINESCENCE RESPONSE OF COPPER-DOPED POTASSIUM BORATE GLASS SUBJECTED TO 6 MEGAVOLT X-RAY IRRADIATION

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This study addresses the characteristics of Cu-doped and undoped potassium borate glass for use as ionizing radiation dosimeters by investigating and comparing the thermoluminescence responses, linearity, sensitivity and dose responses of the two types of glasses. A number of samples based on $xK_2CO_3 + (100 - x)H_3BO_3$*, where* $10 \le x \le 30$ *mol.%, have been prepared using a melt quenching technique. The amorphous phases were identified using X-ray diffraction (XRD). The undoped potassium borate samples* $20K_2CO_3 + 80H_3BO_3$ *(mol.%) and Cu-doped (0.5 mol.%) samples were placed in a solid phantom apparatus and irradiated with in X-ray tube under 6 MV accelerating voltage with doses ranging from 0.5 to 4.0 Gy. This beam was produced by the Primus MLC 3339 linear accelerator (LINAC) available at Hospital Sultan Ismail, Johor Bahru, Malaysia. The results clearly show the superiority of Cu-doped glass in terms of response and sensitivity to producing luminescence over undoped potassium borate glass. The sensitivity of Cu-doped glass is 6.75 times greater than that of undoped glass.*

Keywords: *amorphous materials, luminescence, radiation damage.*

Introduction. Thermoluminescence (TL) dosimetry has become the most widely used technology for evaluating personal and environmental radiation exposure. Currently, radiotherapy is the main modality for destroying cancer, and a high radiation dose may be given to the cancer tissue to maximize the probability of fully destroying the tumor [1]. Borate glasses have been studied due to their features as glass formers and because they are advantageous materials for radiation dosimetry applications [2]. The main advantages presented by these compounds are the effective atomic number, which is very close to that of human tissue ($Z_{eff} = 7.42$), and low cost and easy handling. These facts make borate materials adequate for use in developing medical and environmental dosimeters [3].

Many researchers have investigated the TL properties of lithium tetraborate ($Li_2B_4O_7$, LTB)-doped materials with different activators, such as Mn, Ag, and Mg [4–6]. Undoped and calcium-doped borate glass systems for TL dosimeters were studied by Rojas et al. [7]. The electron paramagnetic resonance spectroscopy of Mn^{2+} and Cu^{2+} centers in the glasses with $Li_2B_4O_7$ and $KLiB_4O_7$ compositions was performed [8]. Recently, we have studied effects of co-doped SnO_2 nanoparticles on the optical properties of Cu-doped lithium potassium borate glass [9]. The TL response of Ge and Al-doped SiO_2 optical fibres subject to 0.2–4.0 Gy dose external photon radiotherapy was studied by Hossain et al. [10]. Based on previous studies, this study aims to develop new potassium borate materials based on B_2O_3 glasses. The TL spectra of the potassium borate glass were investigated, and the glow curve of the Cu-doped potassium borate glasses was better than that of the undoped potassium borate glasses [11]. We focus on the investigation of the high dose (range of 0.5–4.0 Gy) photon irradiation responses of $K_2B_4O_7$:Cu, our interest being in its ability to measure doses in the periphery of typical treatment ranges.

Experimental Methods. Glass samples of undoped and Cu-doped potassium borate were prepared by a melt quenching technique, which is the most widely used technique in the glass industry. The potassium borate samples were made by mixing different compositions of potassium carbonate and boric acid after they had been weighed. The mass of each sample was measured using an electronic balance (PAG, Switzerland). This procedure allowed the TL yield to be normalized to unit mass of the sample. Table 1 shows the different compositions of the five undoped samples. The best-responding potassium borate sample (sample 3) was doped with 0.5 mol.% of pure copper (Cu). Before the melting process, the powders

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TABLE 1. Compositions of Five Undoped Samples of Boric Acid and Potassium Carbonate

Samples	H_3BO_3, mol.%	K_2CO_3, mol.%
1	90	10
2	85	15
3	80	20
4	75	25
5	70	30

were mixed and homogenized by milling the powders for 30 min. The undoped samples were then melted in an alumina crucible by an electric furnace at a temperature of 1000°C for 30 min until a clear homogeneous melt was obtained. The Cu-doped potassium borate samples were prepared using a high temperature furnace at a temperature of 1200°C for 30 min. The molten samples were then poured onto a steel plate in another furnace to be annealed at a temperature of 300°C for 3 h. This furnace was used at a different temperature for melting. The samples were left to be cooled inside the furnace until reaching room temperature to avoid thermal stress.

In the current investigation, a Perspex sheet (tough transparent plastic material) containing 100 wells was covered by a thick black cover to avoid exposure to any light signals during transportation and storage or ultraviolet radiation from sunlight, which can raise the TL signal. Finally, the Perspex sheet, which contained the Cu-doped and undoped potassium borate glasses together, was placed on the beam axis at a depth of 10 cm in a solid phantom apparatus. The glass samples were exposed to X-ray irradiation emitted from a Linear Accelerator (LINAC) Primus MLC 3339 at accelerating voltage of 6 MV. The accelerator was provided by the Department of Radiotherapy and Oncology at Hospital Sultan Ismail, Johor Bahru. The beam field size was set to 10 × 10 cm^2 and placed at the standard source surface distance (SSD) of 100 cm. The dose delivered by the LINAC machine was 20 to 400 MU (Monitor Unit) using a constant dose rate of 200 MU/min. Each 50 MU dose is equivalent to 0.5 Gy, and complete exposure took 15 s. The dosimeter reading was performed 24 h after irradiation. The TL response was read using the TLD Reader 4500 at Ibnu Sina Institute, University of Technology Malaysia (UTM). In this experiment, the preheat temperature was 50°C for 10 s, and the readout temperature was 300°C for 33 s with a heating rate of 25°C/s. Finally, an annealing temperature of 300°C was applied for 10 s. Consequently, the characteristics of TLD were discussed and analyzed as follows.

Results and Discussion. The measurements were performed using a Siemens Diffractometer D5000 system to determine amorphous or crystalline structures of the samples. The XRD analysis showed that samples 3, which were composed of 80% boric acid + 20% potassium carbonate, were glasses because broad peaks appear on the spectra patterns, as shown in Fig. 1.

Figures 2a, b show the TL spectra for Cu-doped and undoped potassium borate glasses [11]. The functional dependence of intensity of the measured TL signal on the absorbed dose is known as the dose response, which is a very important characteristic of the material response. Figure 3 shows the TL responses obtained for the Cu-doped and undoped potassium borate glasses for X-ray irradiation with accelerating voltage 6 MV, presenting their dose response and linearity. The graph shows the data obtained for the high dose response in the dose range of 0.5–4.0 Gy. Each data point represents the average of three individual sample readings. It shows that, as the dose is increased, the TL response in (nC/g) increases, and vice versa. This shows that the TL response of each dosimeter depends on the dose. The idea is that more of the electron traps are filled at higher ionization (higher doses). This increases the electrons chance of finding a recombination center, where the electron can relax to its ground state when heated, resulting in thermoluminescence.

A linear relationship between TL emission and the absorbed dose is a very important characteristic for any TL dosimetric application. For many applications, a linear dose TL signal relationship between the TL signal and the dose is commonly favored. The dose responses of the Cu-doped potassium borate glass samples show a linear relationship between the delivered dose and the TL response, and the relationship shows a stronger TL response from irradiated Cu-doped than undoped potassium borate glasses. Actually, the TLD response to X-ray irradiation depends on the following relationship [12]:

$$\text{TLD response/Exposure} \propto f(E)/(\mu_{en}/\rho)_{air(E)} ,$$

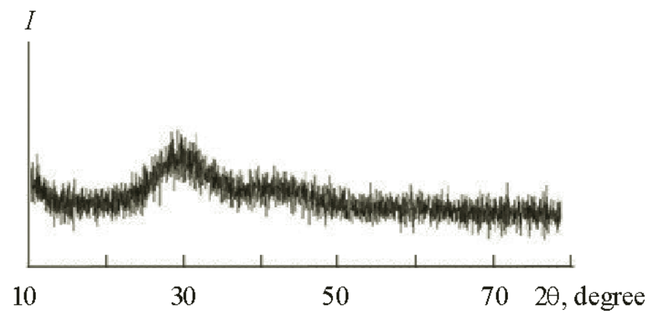


Fig. 1. XRD pattern of [20% K_2CO_3 + 80% H_3BO_3] glass sample 3.

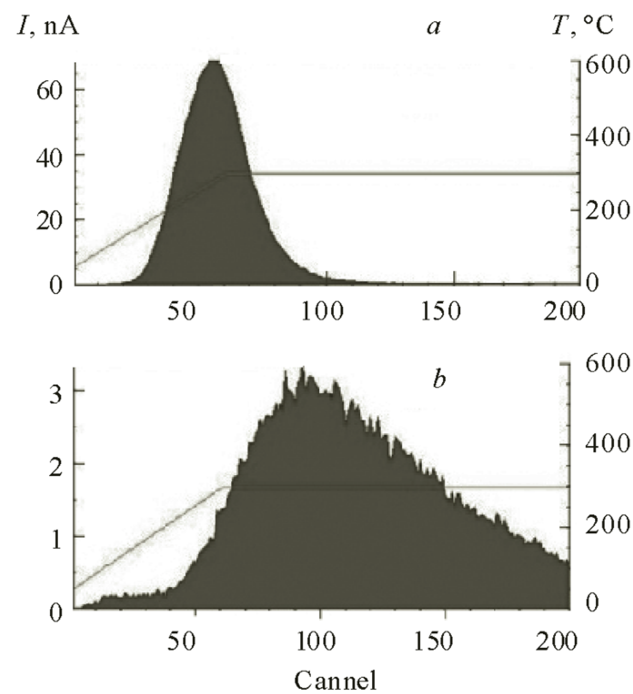


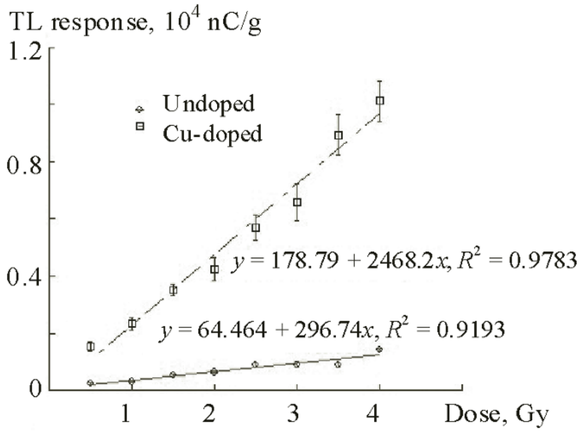
Fig. 2. The TL spectra for the Cu-doped (a) and undoped (b) potassium borate glasses under 6 MV accelerating voltage of X-ray irradiation.

where $f(E)$ is the average fraction of energy deposited in the TL material for any given energy, and $(\mu_{en}/\rho)_{air(E)}$ is the mass energy absorption coefficient for air as a function of the incident energy. It is much more probable that this difference is simply due to the dopant material. Usually, a chemical element is added to a TL material to enhance its luminescence efficiency.

The regression coefficient (R^2) is another indicator of good linearity. Figure 3 shows the R^2 values, which is 0.9783 for Cu-doped and 0.9193 for undoped glass. It is clear that the higher value is demonstrated by the Cu-doped glass, followed by the undoped potassium borate glass. The R^2 value is a measure of the data correlation. The closer the number is to 1, the more closely correlated the data are. It is concluded that the TL response depends strongly on the types of materials and doses.

The sensitivity of a TL material is another important characteristic that can yield information about material responses. It can be described in terms of TL yield per unit dose per unit mass of the sample.

In the current study, the sensitivities for both the Cu-doped and undoped potassium borate glasses were calculated as the averages of the eight (TL yield, $g^{-1} \times Gy^{-1}$) readings. The sensitivity as a function of dose is shown in Fig. 4. The average sensitivity of the Cu-doped glass is 2324.73. In contrast, the sensitivity of the undoped potassium borate glass is 344.27. The



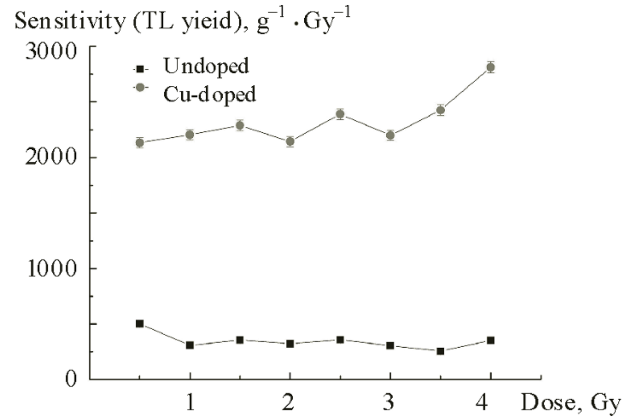


Fig. 3. TL response versus dose of Cu-doped and undoped potassium borate glasses under 6 MV accelerating voltage of X-ray irradiation.

Fig. 4. TL sensitivity versus dose of Cu-doped and undoped potassium borate glasses under 6 MV accelerating voltage of X-ray irradiation.

Cu-doped TL dosimeter shows a TL sensitivity that is 6.75 times greater than that using the undoped potassium borate glass for 6 MV photon irradiation.

Conclusions. This research has investigated the thermoluminescence response, linearity, sensitivity and dose response properties of Cu-doped and undoped potassium borate glasses when exposed to X-ray irradiation with accelerating voltage 6 MV in a dose range of 0.5 to 4.0 Gy. The linearity was also confirmed by the regression coefficient (R^2). The R^2 values were 0.9783 and 0.9193 for Cu-doped and undoped potassium borate, respectively. The sensitivity of the Cu-doped potassium borate glass is 6.75 times higher than that of the undoped potassium borate glass. The Cu-doped potassium borate glass is promising for use as a material for radiation dosimetry.

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REFERENCES

1. M. Oberhoffer and A. Schermann, *Applied Thermoluminescence Dosimetry*, Bristol, Adam Hilger Ltd (1981).
2. M. Santiago, C. Grasseli, E. Caselli, M. Lester, A. Lavat, and F. Spano, *Phys. Status Solidi, A*, **185**, No. 2, 285–289 (2001).
3. G. V. Rao, P. Y. Reddy, and N. Veeraiah, *Mater. Lett.,* **57**, 403–408 (2002).
4. M. Ishii, Y. Kuwano, S. Asaba, T. Asai, N. Senguttuvan, T. Hayashi, and M. Koboyashi, *Radiat. Measur.*, **38**, 571–574 (2004).
5. G. Corradi, V. Nagirnyi, A. Kotlov, A. Watterich, M. Kirm, K. Polgár, A. Hofstaetter, and M. Meyer, *J. Phys. Condens. Matter,* **20**, 025216-24 (2008).
6. G. Kitis, C. Furetta, M. Prokic, and V. Prokic, *J. Phys. D: Appl. Phys.*, **33**, 1252–1262 (2000).
7. S. S. Rojas, K. Yukimitu, A. S. S. de Camargo, L. A. O. Nunes, and A. C. Hernandes, *J. Non-Cryst. Solids*, **352**, 3608–3612 (2006).
8. B. V. Padlyak, W. Wojtowicz, V. T. Adamiv, Y. V. Burak, and I. M. Teslyuk, *Acta Phys. Pol. A*, **117**, 112–117 (2010).
9. H. Aboud, H. Wagiran, I. Hossain, R. Hussin, and S. Saber, *Mater. Lett.,* **85**, 21–24 (2012).
10. I. Hossain, H. Wagiran, and A. N. H. Yaakob. *J. Appl. Spectrosc.,* **80**, No. 4, 635–638 (2013).
11. I. Hossain, N. K. Shekaili, and H. Wagiran, *Adv. Mater. Sci. Eng.*, **278934**, 1–4 (2013).
12. N. H. Yaakob, H. Wagiran, I. Hossain, A. T. Ramli, D. A. Bradley, S. Hashim, and H. Ali, *Nucl. Instrum. Meth. Phys. Res. A*, **637**, 185–189 (2011).