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TL/OSL studies of $Li_2B_4O_7$:Cu dosimetric phosphors

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HIGHLIGHTS

- $Li_2B_4O_7$:Cu as a personal dosimetry was firstly investigated by OSL techniques.
- Prepared phosphor has good OSL repeatability.
- Beta dose can be determined with less than 3–4% error by using OSL technique.
- This is the first study on dose measurement of $Li_2B_4O_7$:Cu using OSL technique.

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ABSTRACT

Dosimetric phosphors of Cu-doped lithium tetraborate ($Li_2B_4O_7$:Cu) were produced using a sintering technique in a laboratory environment and characterized using Scanning Electron Microscopy (SEM) and X-ray Diffractometry (XRD). The thermoluminescence (TL) and optically stimulated luminescence (OSL) properties of powdered ($Li_2B_4O_7$) phosphor doped with copper at different concentrations (0.020–0.025 wt %) were studied. The Cu-doped $Li_2B_4O_7$ phosphor material has two dominant TL glow peaks, and the maximum TL responses of the peaks are at 115 °C and 243 °C in the range of 0 °C–310 °C. The TL response of the Cu-doped lithium tetraborate is approximately 900 times more sensitive than undoped lithium tetraborate. The TL and OSL signal intensities increase as the beta radiation doses increase up to approximately 150.00 Gy and 76.50 Gy, respectively. The OSL dose–response curve is linear up to a dose range of 12.00 Gy for Cu-doped $Li_2B_4O_7$ dosimetric phosphors. The time-dependent fading behavior of the Cu-doped lithium tetraborate was found to be quite stable over long time durations. In addition, the repeatability of the OSL dose measurements were determined to be 2/3 lower compared to the TL measurements. The reproducibility of the OSL measurements was approximately 5%. Based on the TL and OSL results, the prepared phosphors can be used to measure beta doses ranging from 10 μGy to 150.00 Gy and 76.50 Gy, respectively, by using the TL and OSL techniques, with confidence limits of approximately 7% and 3–4%, respectively.

1. Introduction

The optically stimulated luminescence (OSL) technique has found widespread application in a variety of radiation dosimetry fields, including personal monitoring, environmental monitoring, retrospective dosimetry, and space dosimetry (Botter-Jensen et al., 2003). The use of OSL for personal dosimetry was first suggested several decades ago by Antonov-Romanovskii et al. (1956).

OSL only measures the component of the trapped electron population that is most sensitive to light (Botter-Jensen and Murray, 2001).

The use of OSL instead of thermoluminescence (TL) enables precise and real-time measurements to be performed (McKeever, 2001).

Various TL materials (e.g., lithium fluoride, calcium sulfate, and lithium borate) are currently in use as dosimeters in medical, personnel and environmental applications (McKeever, 1985; Yoshimura and Yukihara, 2006).

$Li_2B_4O_7$ is often used as a material for personal dosimetry due to its low effective atomic number ($Z_{eff} = 7.26$) and its similarity in density (2.44 g/cm^3) to that of biological tissues ($Z_{eff} = 7.4$) (Furetta et al., 2001). $Li_2B_4O_7$ is used in practice for personnel dose monitoring in various applications. Lithium tetraborate has been used in different forms (powder, single crystal, pellet and glass) in radiation therapy used in clinical practice involving X-ray and gamma rays

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with energies between 20 and 100 keV, and it is widely used as a radiation dosimeter.

The majority of previous studies have been focused on the photoluminescence (PL), thermoluminescence and dosimetric properties of various starting materials (copper, silver, magnesium or any combination of these dopants) doped $Li_2B_4O_7$ (El-Faramawy et al., 2000a, b; Huy et al., 2008; Prokic, 2001; Manam and Das, 2010; Laksmanan et al., 1981; Annalakshmi et al., 2011; Doull et al., 2013; Pekpak et al., 2011; Patra et al., 2013).

To our knowledge, OSL studies and the dosimetric properties of Cu-doped $Li_2B_4O_7$ dosimetric phosphors by beta irradiation have not been reported in the literature. In addition, there have been no published results concerning the optical stimulation readout of this phosphor material.

In the present work, we first report the OSL and dosimetric properties by beta radiation of undoped and Cu-doped $Li_2B_4O_7$ using the OSL technique.

The aim of this work was to study the TL and OSL properties of $Li_2B_4O_7$:Cu dosimetric phosphor material, which was prepared using a sintering technique; the samples were studied for their dosimetric properties in terms of the TL and OSL response to irradiation with different beta radiation doses, such as the linear response to beta exposure and fading behaviors. OSL under blue LED stimulation was observed for both doped and undoped $Li_2B_4O_7$ phosphors.

The repeatability and reproducibility of the characteristics of the $Li_2B_4O_7$ crystal after annealing were investigated using the TL and OSL techniques. The crystal structures of the samples were investigated using SEM and XRD methods. In addition, the OSL decay constants (t_1 and t_2) of the Cu-doped $Li_2B_4O_7$ samples were determined for different beta doses.

2. Experimental details

The Cu-doped $Li_2B_4O_7$ thermoluminescent material was prepared using a sintering technique in our laboratory according to a procedure given in the literature (Takenega et al., 1977, 1983). The prepared powdered phosphors were used for TL and OSL measurements. Each sample was measured three times. Each data point was the average of three measurements. The sample weight was 8 mg for all the TL and OSL measurements. Undoped and Cu-doped $Li_2B_4O_7$ crystals were annealed after each of the TL and OSL measurements. The heat treatment was performed in a RISO Model DA-20 TL/OSL reader.

XRD analysis, SEM analysis, and TL and OSL spectrometric analyses were performed during the course of the study.

All the measurements were performed in the dark to avoid the influence of the room light in the TL and OSL responses.

2.1. Preparation of the $Li_2B_4O_7$:Cu phosphors

The optimal activator concentration for preparing $Li_2B_4O_7$:Cu was found to be 0.020–0.025 wt % Cu (El-Faramawy et al., 2000a, b). Cu (0.020 wt %) was added as an activator to the $Li_2B_4O_7$ powder (99.998%, Alfa Aesar). The Cu and $Li_2B_4O_7$ powder materials were mixed with alcohol. Then, the mixture was stirred and dried for 1 h at 100 °C using a Thermolyne oven and sintered by heating in a platinum crucible to 920 ± 1 °C for 2 hours under a nitrogen atmosphere. The mixture was then rapidly cooled to room temperature. The resultant glassy mass was heated to 650 °C and maintained at this temperature for 1 h to ensure complete crystallization. The prepared powdered phosphor sample was soft and white in color, with a very large grain size. The phosphor was ground, and the resultant powder was used for the TL and OSL dosimetric studies. Polycrystalline $Li_2B_4O_7$:Cu was then powdered using an agate mortar and pestle and sieved to obtain grain sizes of 74–100 μm.

To completely erase the TL signal and to restore the original TL sensitivity of the phosphors, fresh powdered samples were annealed in an oven at 300 °C for 30 min.

2.2. Irradiations

Irradiations of all the samples were performed at room temperature using a calibrated $^{90}Sr/^{90}Y$ beta source at the Sarayköy Nuclear Research Center of the Turkish Atomic Energy Authority in Ankara. A $^{90}Sr/^{90}Y$ beta source, which can be used together with a DA-20 Model RISO TL/OSL Reader, was used for beta irradiations. The activity of the source was 40 mCi, and the dose rate was 0.153 Gy/sec. All irradiated and unirradiated powdered samples were stored at room temperature in the dark until the OSL and TL measurements were performed. The room temperature storage conditions were 30% relative humidity at a temperature of 22 ± 3 °C.

2.3. TL measurements

All TL measurements were performed at room temperature using an automated RISO Model TL/OSL-DA-20 TL/OSL Luminescence Reader (Risø National Laboratory, Røskilde, Denmark), which allows up to 48 samples to be both individually heated to any temperature between room temperature and 700 °C and individually irradiated by a radioactive beta source ($^{90}Sr/^{90}Y$). The heating system is able to heat the samples up to 700 °C at linear heating rates from 0.1 to 10 °C/s. The heating strip is cooled by a nitrogen flow, which also protects the heating system from oxidation at high temperatures. Thermal stimulation is achieved using the heating element, which heats the sample and lifts the sample into the measurement position. Nitrogen was flushed in the heating chamber to reduce spurious TL arising from the presence of oxygen.

The glow curve is the most important property for the production of the TL dosimeters. The TL glow curves, which are plotted by software connected to TL measurement systems, illustrate the thermoluminescence intensity versus temperature.

All the undoped and Cu-doped $Li_2B_4O_7$ powdered phosphors were subjected to an annealing treatment at 300 °C for 30 min before the TL measurements were performed to erase the residual TL signal and to restore the original TL sensitivity of the powdered samples (Furetta et al., 2001). The TL glow curves were collected up to a temperature of 310 °C in a nitrogen gas atmosphere and were recorded by setting the linear heating rate of the TL reader at 4 °C/s. (TL 310 °C, m = 8 mg, no preheating). All the undoped and Cu-doped $Li_2B_4O_7$ powdered samples were stored at room temperature before and after the TL measurements, and the samples were not preheated. The measurements were performed 24 h after the irradiation process to eliminate the effects of the low temperature peak.

2.4. OSL measurements

The OSL measurements were performed using the same RISO TL/OSL (TL/OSL-DA-20) luminescence reader. All luminescence emissions were detected with a bi-alkali EMI 9235QA photomultiplier tube (PMT), which has an extended UV response with a maximum detection efficiency between 300 and 400 nm. To prevent scattered stimulation light from reaching the PMT, the reader is equipped with a 7.5 mm Hoya U-340 detection filter, which has a peak transmission at approximately 340 nm. Internal blue light emitting diodes (LEDs) (470 nm, 40 mW/cm^2) were used for stimulation, and the OSL signal was detected through Hoya U-340 filters. A detachable beta irradiator located above the sample carousel accommodates a 1.48 GBq (40 m Ci) $^{90}Sr/^{90}Y$ beta source, which emits beta particles with a maximum energy of 2.27 MeV. A $^{90}Sr/^{90}Y$ beta source provided a dose rate of 0.153 Gy/s. All the OSL measurements were performed in the continuous wave (CW-OSL) mode, and the power level was software controlled and set at 90% of the maximum stimulation power for the blue LEDs. The OSL measurements were performed at room temperature. The weight of the disc is 8 mg. The samples were irradiated with beta doses in the range of 0 Gy–76.50 Gy to obtain the dose response of the phosphor. Then, the prepared powdered crystals were stimulated with OSL blue LEDs under the following conditions: duration of 350 s, no preheat, and 90% of maximum stimulation power.

2.5. XRD studies

The characterization of the prepared pure and Cu-doped $Li_2B_4O_7$ powdered phosphors was performed using X-ray diffraction. The XRD measurements were performed at room temperature for the pure and Cu-doped $Li_2B_4O_7$ powdered phosphors. The phosphor grains were only sieved before the measurements were performed. In all our XRD experiments, powdered phosphor grains in the size range of 74–210 μm were used. The XRD patterns were obtained over a wide range of Bragg angle 2θ values ($10° \leq 2\theta \leq 90°$) using a Bruker AXS D8 Advance X-ray powder diffractometer (operated at 40 kV) with a Cu-Kα radiation source with a wavelength of 1.54056 Å. Scanning was performed in the 2θ mode with a step size 0.05° and 1.5 s per step. The determination of the phase structure and the unit cell parameters of the produced samples, along with the numerical calculations, were performed using the PDF 2009. 4 cA analysis program.

In this study, the crystal sizes of the prepared samples were calculated using the Debye-Scherrer equation.

$$D = (0.9\lambda)/(\beta \cos\theta) \qquad (1)$$

where D is the average grain size of the crystallites, λ is the incident wavelength (1.54056 Å), θ is the Bragg angle, and β is the diffracted full width at half maximum (FWHM). The hkl and 2θ values of the undoped and doped $Li_2B_4O_7$ were determined by analyzing the measured XRD patterns.

2.6. SEM studies

The structural and morphological characteristics (particle size and shape of undoped and Cu-doped $Li_2B_4O_7$ powdered samples) were studied using a Scanning Electronic Microscope (SEM) operated at 5–10 kV. In this study, samples in powder form (74–125 μm) were placed directly into a SEM for imaging. The measurements were performed using a JEOL-JSM7000F scanning electron microscope. SEM images of the undoped and Cu-doped $Li_2B_4O_7$ samples were recorded at both 100× and 2500× magnifications.

3. Results and discussion

3.1. XRD results

The formation of doped and undoped $Li_2B_4O_7$ crystals was confirmed by studying the X-ray diffraction (XRD) patterns shown in Fig. 1 (a) and (b). The XRD pattern of the undoped crystals matches with PDF 2009. 4 cA data (Card No. 076–0768). The undoped and Cu-doped $Li_2B_4O_7$ powdered samples have tetragonal crystal structures with lattice parameters of $a = b = 9.477$ Å, $c = 10.286$ Å and $\alpha = \beta = \gamma = 90°$, having the space group I41cd (110). Pekpak et al. (2011) also obtained similar results for $Li_2B_4O_7$ samples. The results indicate that the lattice size of the crystal changed with the addition of the Cu dopant.

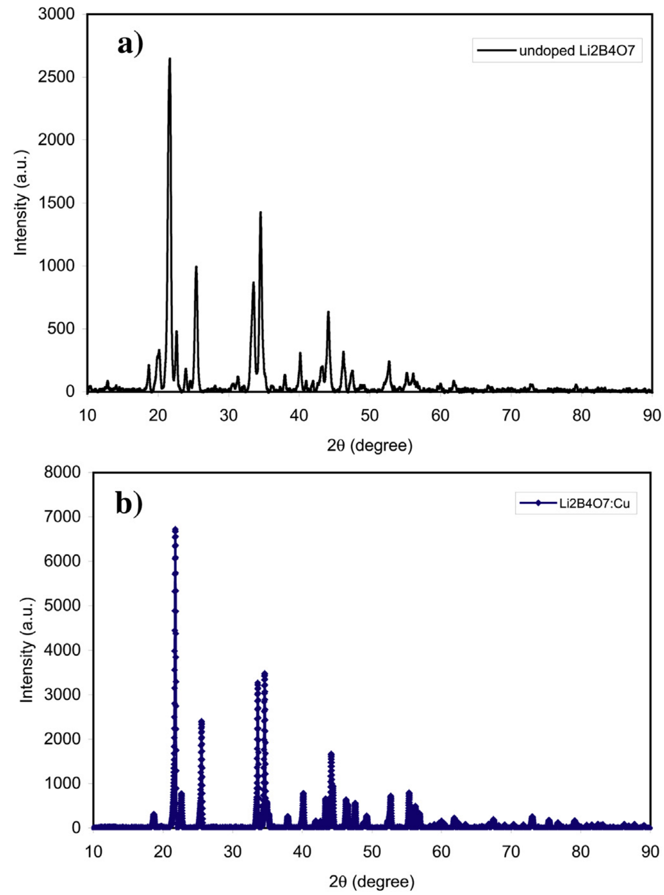

Fig. 1. XRD pattern of $Li_2B_4O_7$ samples at room temperature. a) Undoped $Li_2B_4O_7$ b) Cu doped $Li_2B_4O_7$.

The average grain sizes of the undoped and doped $Li_2B_4O_7$ were calculated to be approximately 29.13 nm and 68.59 nm, respectively. The crystallite size of Cu-doped lithium tetraborate is larger than that of undoped lithium tetraborate. The values of the grain sizes, reflections, and 2-theta values of the prepared phosphors based on the XRD patterns are listed in Table 1. The crystal size directly provides information about the crystalline quality, and the diffraction peak obtained by the XRD peak width is inversely proportional to the half-height. The diffraction peaks are very narrow, indicating that the crystals in this case exhibit good crystalline quality.

Table 1
The crystal unit cell parameters, reflection, 2-theta values and crystallite sizes of pure and Cu doped $Li_2B_4O_7$.

Crystal type	2-Theta (degrees)	Reflection (hkl)	a (nm)	b (nm)	c (nm)	Crystalite sizes (nm)
Pure $Li_2B_4O_7$	21.670	112	947.7	947.7	1028.0	29.13
	25.464	202				
	34.565	312				
Cu doped $Li_2B_4O_7$	21.760	112	947.9	947.9	1028.0	68.59
	25.461	202				
	34.564	312				

3.2. SEM results

The crystal sizes of the undoped and Cu-doped $Li_2B_4O_7$ powdered samples were determined using SEM, as shown in Fig. 2. The luminescence efficiencies are related to the phosphor material with a crystallite size in the range of 74–125 µm. According to the SEM images, the Cu-doped $Li_2B_4O_7$ powdered samples apparently form clusters.

3.3. TL results

One of the important aspects to be investigated for any TL dosimetry material is its glow curve. TL glow curves were not observed in the unirradiated $Li_2B_4O_7$ powdered samples. The TL glow curves of the undoped and Cu-doped $Li_2B_4O_7$ powdered samples irradiated with beta radiation doses of 1.50 Gy are shown in Fig. 3 (a) and (b), respectively. The TL glow curve of the pure $Li_2B_4O_7$ phosphor in powdered form irradiated with a beta dose of 1.50 Gy has one TL glow peak at approximately 103 °C, while the Cu-doped $Li_2B_4O_7$ phosphor exposed to the same dose has two dominant TL glow peaks, with the maximum TL response of the peaks at 115 °C and 243 °C in the range of 0 °C–310 °C. The lower temperature peak of the Cu-doped $Li_2B_4O_7$ occurs at approximately 103 °C, which is too low for use in practical TLDs. The peak activated with Cu occurs at 243 °C, which is sufficiently high for routine TL dosimetry applications. The dosimetric peak of the Cu-doped $Li_2B_4O_7$ was also observed to be approximately 205 °C by Takenega et al. (1983).

The TL response of the Cu-doped lithium tetraborate is approximately 900 times more sensitive than that of the undoped lithium tetraborate.

3.4. OSL results

The OSL signal is set to the background level after 350 s. The stimulation is very efficient, with no long tail observed. The $Li_2B_4O_7$ powdered phosphor OSL signal was read out two or three times, with the last read out, being considered the background of the system (reader and crystal), subtracted from the first one to obtain the background-free signal.

The CW-OSL decay curves of $Li_2B_4O_7$:Cu dosimetric crystal in powdered form is shown in Fig. 4 for a beta dose of 3.06 Gy. The best correlation is obtained for second-order exponential decay functions [$(I = I_{0a}.\ \exp(-Dt_1) + I_{0b}.\ \exp(-Dt_2))$]. The OSL decay curve is composed of the sum of two exponential functions. In this function, I and D represent the signal intensity and the beta radiation dose (in Gy), respectively. The parameters t_1 and t_2 are decay constants. The parameters t_1 and t_2 were calculated as 31.87 and 145.79 (see Table 2.), respectively, with a correlation coefficient of $R^2 = 0.99973$ and $Chi^2 = 8.145 \times 10^{-8}$ from the fitting procedure. The amplitudes I_{0a} and I_{0b} of the fitting exponential components were calculated to be 0.04725 a.u. and 0.03486 a.u., respectively. Of the total OSL signal, 57.54% is composed of component t_1 and 42.46% is composed of component t_2 for a beta dose of 3.06 Gy.

The analysis of the OSL signal indicates that it is composed of two components: a fast component, assigned to electrons that recombine directly with holes, and a slow component due to the presence of shallow traps in the structure, in which electrons are retrapped for several seconds before being recombined with holes.

Table 2 also indicates that the decay constants (t_1 and t_2) become faster at the low beta doses and become slower with increasing beta dose.

The OSL luminescence intensity is proportional to the trapped charge, and the trapped charge is related to the radiation dose (energy deposited). The normalized OSL decay curves of the Cu-doped $Li_2B_4O_7$ phosphor for various absorbed doses using a $^{90}Sr/^{90}Y$ beta source are shown in Fig. 5a and b. The OSL intensity increases with the increase of different beta doses.

After irradiation with the $^{90}Sr/^{90}Y$ beta source, poor OSL signal was observed from the pure compound.

3.5. Dose response

To study the TL response, the $Li_2B_4O_7$ phosphor was investigated over the beta dose range from 0.20 Gy to 150.00 Gy. The TL responses of the Cu-doped lithium tetraborate with beta doses of up to 150.00 Gy are shown in Fig. 6. The relative TL intensity increased with increasing beta doses. In Fig. 6, a fit to the data using a 3rd degree polynomial ($y = 1.77073 + 1.25927x - 0.07609x^2 - 0.03387x^3$ and correlation coefficient of $R^2 = 0.9994$ from the fitting procedure) is shown. The slope of the line of 0.996 (between 0.20 Gy and 10.00 Gy) indicates a linear dependence of the TL response, which is highly desirable for dosimetry applications because it ensures an accurate estimation of the dose. The TL output of the Cu-doped $Li_2B_4O_7$ is linear for beta radiation exposures up to approximately 10.00 Gy, and it becomes sublinear above this dose. Among the many TL phosphors exhibiting a supralinear response for doses over 10.00 Gy (Furetta et al., 2001), the prepared Cu-doped $Li_2B_4O_7$ phosphor exhibits an exceptional behavior. For beta radiation doses above 150.00 Gy, the TL output saturates,

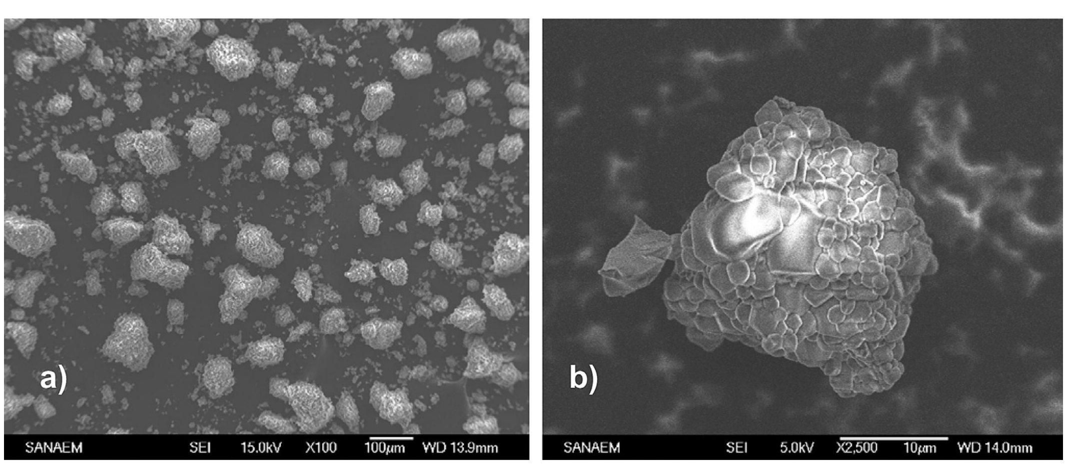

Fig. 2. a, b) SEM image of undoped $Li_2B_4O_7$; c, d) SEM image of Cu doped $Li_2B_4O_7$ samples at room temperature.

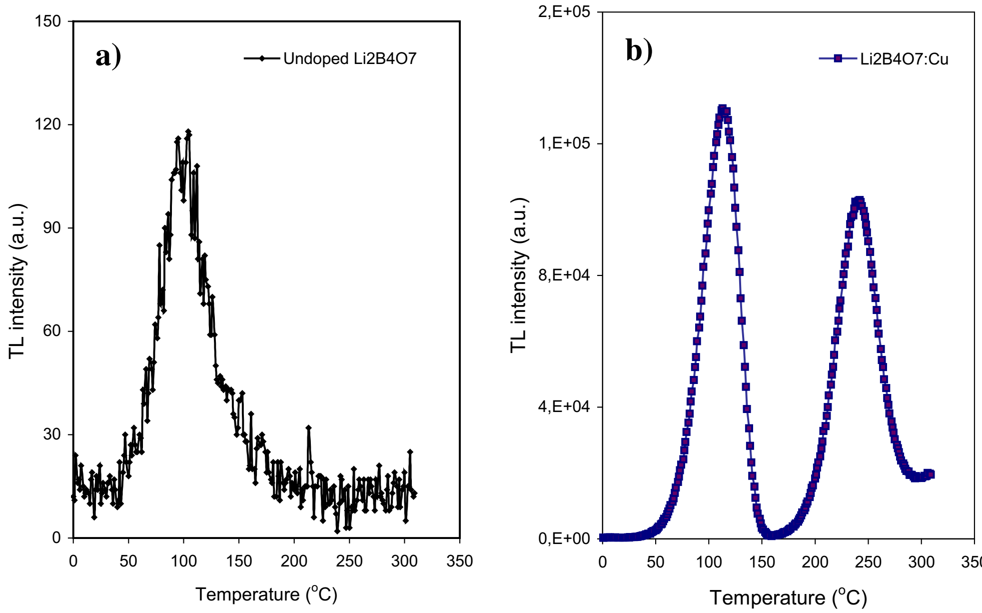

Fig. 3. TL glow curve of $Li_2B_4O_7$ powder irradiated with 1.50 Gy at room temperature with beta source ($m = 8$ mg, no preheat, TL 310 °C, heating rate: 4 °C/s). a) TL glow curve of undoped $Li_2B_4O_7$ b) TL glow curve of Cu doped $Li_2B_4O_7$.

indicating a saturation of traps related to the TL response in the crystals.

To construct the OSL dose response curves of the Cu-doped $Li_2B_4O_7$ in powdered form, dosimetric crystals having a mass of 8 mg were irradiated using different beta doses up to 76.50 Gy. The OSL dose response curves of Cu-doped $Li_2B_4O_7$ samples are presented in Fig. 7 (a) and (b).

The OSL dose response curve of the powder-form $Li_2B_4O_7$:Cu dosimetric crystal irradiated at 0 Gy –11.48 Gy with a beta source and stimulated by $\lambda = 470$ nm blue LEDs (blue light) (350 s, 90% of maximum stimulation power) is presented in Fig. 7(a). A straightforward linear relationship between the OSL integrated intensity and the dose was observed up to 11.48 Gy. The OSL dose–response curve of Cu-doped $Li_2B_4O_7$ was best described by a linear function of dose, $y = a + bD$. In this function, y and D represent the OSL signal intensity and the applied beta radiation dose in Gy, respectively, and the parameters a and b, which are calculated from fitting procedures for Cu-doped $Li_2B_4O_7$, were found to be −75051 and 481863 (correlation coefficient $R^2 = 0.9994$), respectively. In other words, this material exhibits excellent linearity in this dose range.

To present the OSL dose response curves of Cu-doped $Li_2B_4O_7$, the powder-form dosimetric crystals were irradiated over a dose range of 1.07 Gy–76.50 Gy at different beta doses and stimulated by $\lambda = 470$ nm blue LEDs (blue light). The OSL dose–response curves of the Cu-doped $Li_2B_4O_7$ are presented in Fig. 7 (b) using a logarithmic scale. The OSL intensity increased with increasing beta irradiation doses between 1.07 Gy and 60.00 Gy; Fig. 7 (b) shows a fit of a 3rd degree polynomial ($y = 10.898x^3 - 4539.7x^2 + 512149x - 94329$, $R^2 = 0.9997$) to the data. For beta radiation doses above 60.00 Gy, the OSL output saturates, indicating a saturation of the traps.

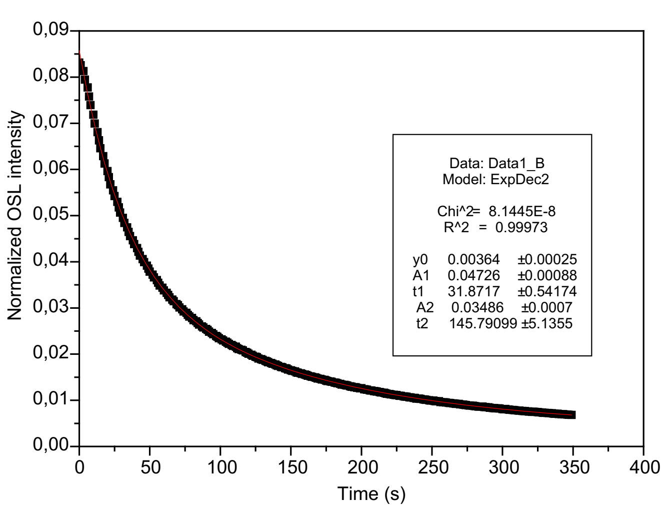

Fig. 4. CW-OSL decay curves of $Li_2B_4O_7$:Cu in powder form dosimetric crystal for a beta dose of 3.06 Gy.

3.6. TL repeatability studies

The repeatability measurements of the prepared phosphor are an important topic of dosimetric studies. In the TL repeatability studies, the $Li_2B_4O_7$:Cu powder phosphors were first annealed at 300 °C for 30 min. Then, the annealed $Li_2B_4O_7$ materials were irradiated with beta doses of 7.50 Gy for the measurements. The TL measurements were performed for 20 cycles at $t = 310$ °C and at a heating rate of 4 °C/s. The dosimetric peak intensities, which were

Table 2
OSL decay constants change of the Cu doped $Li_2B_4O_7$ with the different beta doses.

Beta dose (Gy)	t_1 (s^{-1})	t_2 (s^{-1})
0.153	21.09	105.95
0.306	24.64	116.04
0.765	27.33	126.58
1.530	29.12	131.08
3.060	31.87	145.79
4.590	36.07	161.75
15.300	37.84	173.22
30.600	38.12	175.28
45.900	38.12	175.28
76.500	37.89	173.55

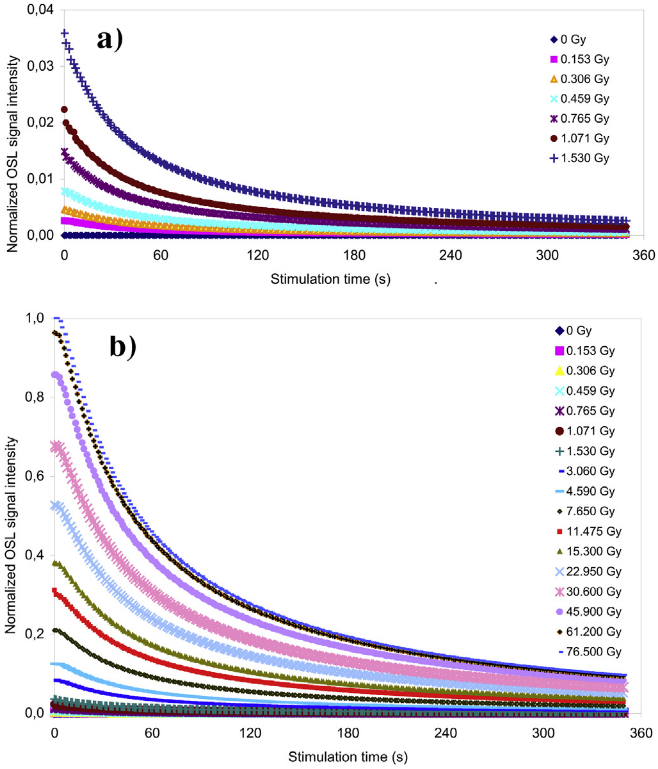

Fig. 5. OSL decay curves of $Li_2B_4O_7$:Cu in powder form dosimetric crystal irradiated at different beta dose under blue light stimulation. The weight of the disc is 8 mg (OSL blue LEDs 350 s, no preheat, 90% of the maximum stimulation power). a) Irradiated between 0 Gy and 1.53 Gy with beta source b) irradiated between 0 Gy and 76.50 Gy with beta source. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

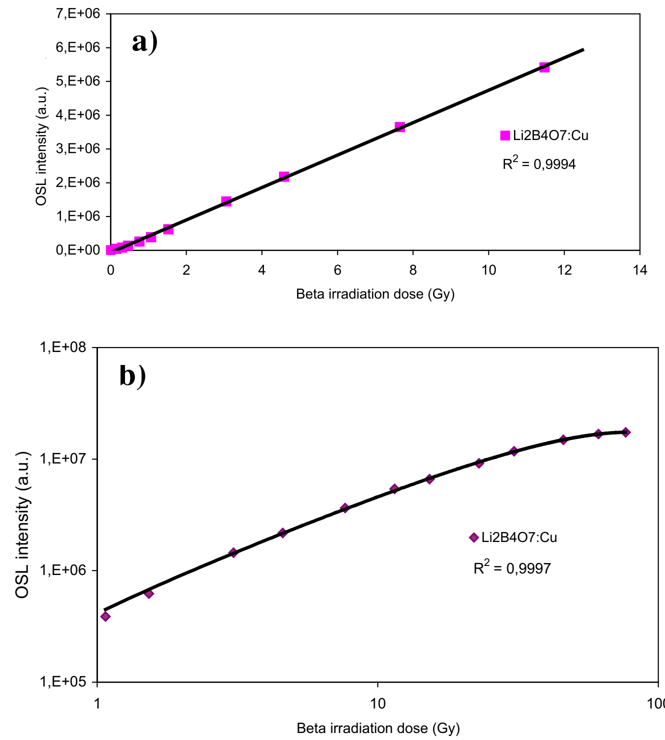

Fig. 7. OSL dose response curves of Cu doped $Li_2B_4O_7$ in powder form dosimetric crystal irradiated at different beta doses taken at room temperature (m = 8 mg, OSL blue LEDs 350 s, no preheat, 90% of the maximum stimulation power). a) irradiated between 0 Gy and 11.48 Gy with beta source b) irradiated between 1.07 Gy and 76.50 Gy with beta source. Symbols (experimental) and solid lines are the best-fitting lines. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

obtained at 243 °C, were used for the repeatability measurements. Fig. 8(a) shows the glow curve variation with the number of repeating cycles. Neither the shape of the TL glow curve nor any shift in the glow peak temperature was observed at the end of the 20 cycles in the studied dose range. The TL repeatability obtained of the Cu-doped $Li_2B_4O_7$ crystal was 7.0% (2σ) for 20 sequential measurements. The slight variation in the maximum TL intensity

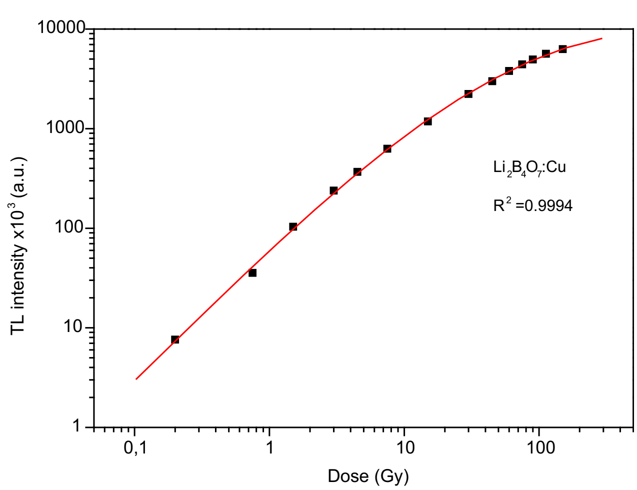

Fig. 6. Beta dose response curve of Cu doped $Li_2B_4O_7$ (0.20 Gy –150.00 Gy) (no preheat, TL 310 °C, heating rate: 4 °C/s). Symbols (experimental) and solid lines are the best-fitting lines.

during the 20 cycles indicates that both phosphors are reusable in the TL studies.

3.7. OSL repeatability studies

If the sensitivity of the phosphors does not change after several cycles of exposure and readouts, then it is considered to be as a good dosimetric phosphor. The repeatability of the OSL measurements of the undoped and Cu-doped $Li_2B_4O_7$ crystals, which were irradiated at 7.50 Gy with the beta source, was obtained by taking 20 measurements for each of samples (7.50 Gy beta doses, m = 8 mg, 350 s, no preheat, 90% of maximum stimulation power, blue LEDs). The variation of the normalized OSL signal intensity vs. cycle number for undoped $Li_2B_4O_7$ crystal is shown in Fig. 8(b). The repeatability obtained for the undoped $Li_2B_4O_7$ crystal was 7.8% (2σ). The repeatability of the OSL measurements for the undoped $Li_2B_4O_7$ crystals was determined to be 1.12 times higher compared to the TL measurements. The variation of the normalized OSL signal intensity vs. cycle number for Cu-doped $Li_2B_4O_7$ crystal is shown in Fig. 8(c). The repeatability obtained of the Cu-doped $Li_2B_4O_7$ crystal was 3–4% (2σ). The repeatability of the OSL measurements was determined to be 2/3 lower than that of the TL measurements.

3.8. OSL and TL reproducibility studies

The batch uniformity of the prepared dosimetric phosphors was also investigated. To determine the reproducibility of the preparation procedure, different batches (4 batches) of the Cu-doped $Li_2B_4O_7$ powdered phosphor was used for the OSL sensitivity measurements. The reproducibility of the OSL response of

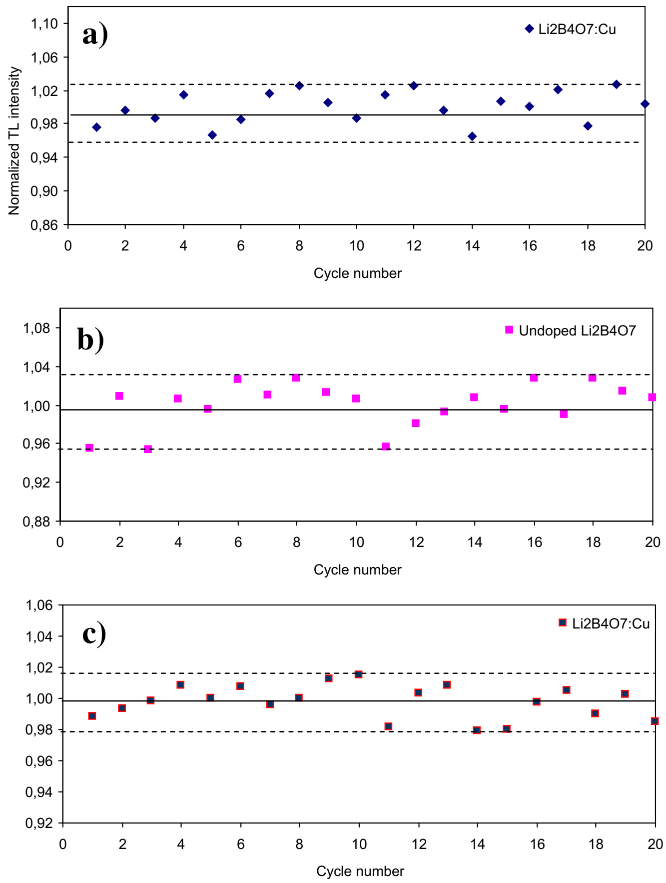

Fig. 8. a) Variation of normalised TL intensity for Cu doped $Li_2B_4O_7$ crystal irradiated at 7.50 Gy with number of reuse cycles (TL 310 °C, no preheat, heating rate: 4 °C/s). b) Variation of normalised OSL signal intensity for undoped $Li_2B_4O_7$ crystal irradiated at 7.50 Gy with number of reuse cycles (OSL blue LEDs 350 s, no preheat, 90% of the maximum stimulation power). c) Variation of normalised OSL signal intensity for Cu doped $Li_2B_4O_7$ crystal irradiated at 7.50 Gy with number of reuse cycles (OSL blue LEDs 350 s, no preheat, 90% of the maximum stimulation power). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Cu-doped $Li_2B_4O_7$ phosphors was obtained from three measurements for each sample. First, the annealed $Li_2B_4O_7$ materials were irradiated using beta doses of 7.50 Gy for the measurement of the OSL sensitivity. The OSL signal intensities were used for the reproducibility measurements ($m = 8$ mg, 350 s, no preheat, 90% of maximum stimulation power, blue LEDs). We did not observe any change in the OSL signals shapes and positions. Table 3 shows the OSL sensitivity changes for the different batch samples. The reproducibility characterized by the standard deviation of the results was good for the four samples. The results from the reproducibility experiments indicate that the changes of the OSL sensitivities of the Cu-doped $Li_2B_4O_7$ crystals were in the range of approximately 5.0% (2σ) for the four batches. Our experimental measurements indicated that the changes of OSL sensitivity were within the error bars of the measurements.

Table 3
OSL sensitivity of the Cu doped $Li_2B_4O_7$ phosphor synthesized in different batches and irradiated with 7.50 Gy ($m = 8$ mg, 350 s, no preheat, 90% of the maximum stimulation power, blue LEDs).

Batch no	Relative OSL sensitivity
1	0.98 ± 0.01
2	0.97 ± 0.01
3	1.02 ± 0.01
4	0.97 ± 0.01

3.9. *Fading studies*

An ideal TL and OSL material should have deep thermally stable traps for the long-term storage of dosimetric information without significant fading. To determine the room temperature stability of the glow peaks for the Cu-doped $Li_2B_4O_7$ phosphors, the samples were investigated after different periods.

To increase the sensitivity of the Cu-doped $Li_2B_4O_7$ crystal in powdered form, the samples were heated at 300 °C in an oven for 30 min. TL measurements were performed on the samples immediately after the heat treatment. The Cu-doped $Li_2B_4O_7$ crystals were ready for use again after the heat treatment. The variations in the TL intensity could be neglected for the cases of measurements immediately after the annealing procedure and before the irradiation process.

The samples were stored under dark conditions at room temperature until the completion of the measurements. The results of the studies on fading were normalized to 100% with the first measurements taken 64 h after irradiation for the low temperature peak. Fading of the TL intensity of the dosimetric peak was investigated for 40 days.

A decrease of the TL intensity was observed relative to the maximum values measured at the end of the storage time for undoped $Li_2B_4O_7$.

Fig. 9(a) and (b) show the fading of the TL glow curve and the variations of the % TL intensity of Cu-doped $Li_2B_4O_7$ samples, which were stored at room temperature, irradiated with a beta radiation dose of 1.50 Gy as function of temperature and storage time, respectively.

The first peak (low-temperature peak) of the TL glow curve of the $Li_2B_4O_7$:Cu samples irradiated with 1.50 Gy fades completely when stored at room temperature in the dark for 24 h, whereas the second peak (dosimetric peak) slowly decreased over the storage time. In other words, fading of the second temperature TL peak is negligible over the time of storage, and the TL peaks did not shift with storage time ($m = 8$ mg, no preheat, TL 310 °C).

The % TL intensity decreased due to the time-dependent changes in the TL intensity of the first TL peak after 16 h. The first TL peak intensity of the $Li_2B_4O_7$:Cu sample decreased rapidly after 2.6 h. In contrast, the intensity of the dosimetric TL peak (second peak) only decreased by approximately 8–10% after 40 days. As expected from the very low temperature of the peak, the peak itself is affected by rapid fading at the end of the study period.

3.10. *Minimum detectable dose*

The lowest level of detection, known as the minimum detectable dose, was calculated from the following relation (McKeever et al., 1995).

$$D_o = 2.26 \cdot \sigma \cdot S \quad (2)$$

where σ is the standard deviation of the background reading value of unirradiated samples in units of nC, and S represents the conversion factor (=2.310) in units of mGy/nC. The experimentally determined minimum detectable dose (M.D.D) is defined as three times the standard deviation of the zero dose readings of the dosimeters, and it has a value of approximately 10 μGy for the $Li_2B_4O_7$ phosphor. At the end of the readout, there are no residual TL and OSL signals.

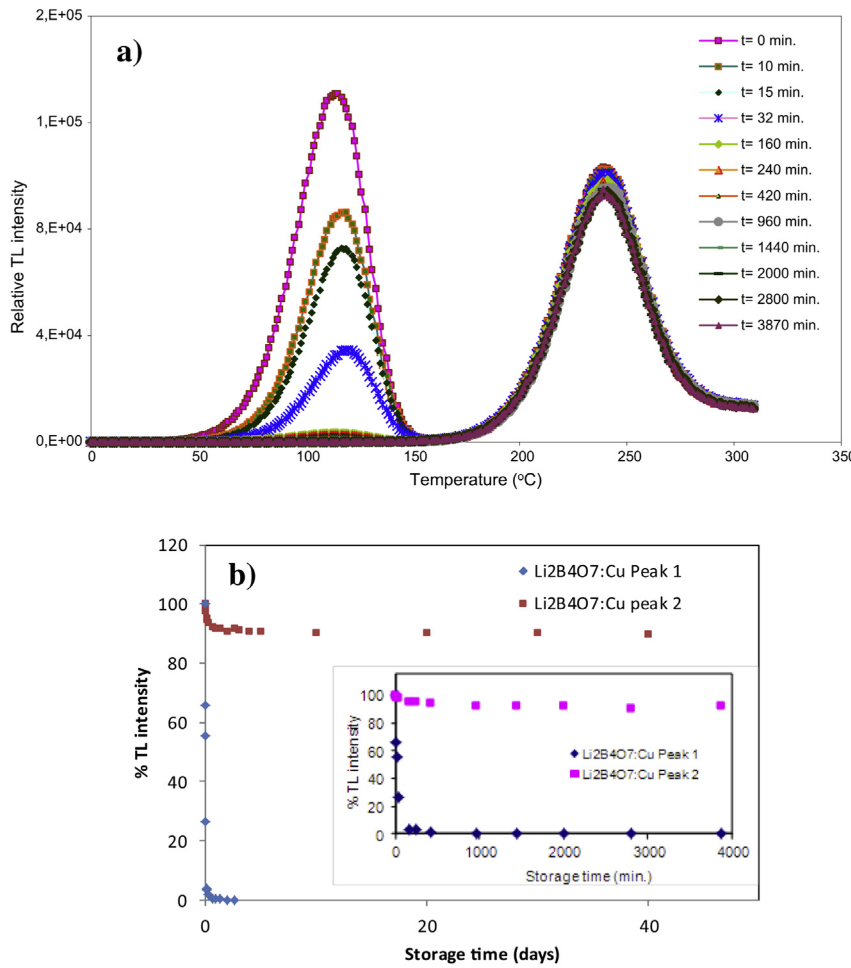

Fig. 9. a) Fading of TL glow curve of Cu doped $Li_2B_4O_7$ irradiated with 1.50 Gy beta radiation which is storage time at room temperature (m = 8 mg, no preheat, TL 310 °C, heating rate: 4 °C/s). b) Variation of %TL intensity with storage time for Cu doped $Li_2B_4O_7$ irradiated with 1.50 Gy beta radiation which is storage time at room temperature (m = 8 mg, no preheat, TL 310 °C, heating rate: 4 °C/s). The insert shows the short storage time.

4. Conclusions

This work investigated the TL and OSL dosimetric properties of $Li_2B_4O_7$:Cu phosphors synthesized using a sintering technique. From detailed Cu doping studies, the TL response was found to be approximately 900 times more sensitive than that of undoped lithium tetraborate.

The TL and OSL intensities of the prepared crystalline samples increased with increasing beta radiation doses. The OSL dose–response curve is linear up to a dose range of 12 Gy for the Cu-doped $Li_2B_4O_7$ dosimetric phosphors, with a correlation coefficient of 0.9994.

The OSL and TL peak intensities exhibited the same reduction from the time-dependent changes and were not dependent on the irradiated beta doses. In contrast, the main dosimetric peak occurred at a higher temperature (t = 243 °C) for the Cu-doped $Li_2B_4O_7$ and was found to exhibit little fading with storage time; its fading properties are satisfactory for use in dosimetry applications.

Using TL measurements repeated 20 times on the copper-doped lithium tetraborate phosphors irradiated at a beta irradiation dose of 7.50 Gy, the measurement error was found to be 7%; this error was measured to be 3–4% by for the OSL technique. The repeatability of the OSL dose measurements was determined to be 2/3 lower than the TL measurements. The reproducibility of the OSL measurements was found to be approximately 5%. Beta doses as low as 10 μGy can be measured using the prepared phosphors, and 76.50 Gy can be measured with a precision on the order of 3–4% error by using the OSL technique. The TL and OSL signal intensities of lithium tetraborate have a dosimetric peak that could be neglected under room temperature storage conditions.

The results indicate that the crystal parameters were not changed with the addition of Cu dopants. The size of the crystal changed with the addition of Cu dopants.

The studied $Li_2B_4O_7$ dosimetric phosphors can provide high sensitivity, linear response, good repeatability and reproducibility, batch uniformity, low fading and low read error in dosimetry applications, and it may be used as an OSL personal dosimetry monitor to measure beta radiation doses.

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