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Thermoluminescence dosimetry properties and kinetic parameters of lithium potassium borate glass co-doped with titanium and magnesium oxides

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HIGHLIGHTS

- Lithium potassium borate glass doped with Ti and Mg was prepared.
- The material is close to soft tissues in terms of *Zeff*.
- The radiation sensitivity is about 12 times lower than that of TLD-100.
- The signal fades about 8% in 10 days and 17% in 3 months.

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ABSTRACT

Lithium potassium borate (LKB) glasses co-doped with TiO_2 and MgO were prepared using the melt quenching technique. The glasses were cut into transparent chips and exposed to gamma rays of ^{60}Co to study their thermoluminescence (TL) properties. The TL glow curve of the Ti-doped material featured a single prominent peak at 230 °C. Additional incorporation of MgO as a co-activator enhanced the TL intensity threefold. LKB:Ti,Mg is a low-*Z* material (Z_{eff}=8.89) with slow signal fading. Its radiation sensitivity is 12 times lower that the sensitivity of TLD-100. The dose response is linear at doses up to 10^3 Gy. The trap parameters, such as the kinetics order, activation energy, and frequency factor, which are related to the glow peak, were determined using TolAnal software.

1. Introduction

Lithium borate dosimeters show promising TL properties that passed the disturbance of phosphors and gave opulence applications in both medical and environmental fields (Magdalyna et al., 2004). Manganese was the first activator for lithium borate dosimeter proposed by Schulman et al. (1967). The thermoluminescence of the material was promising, but its radiation sensitivity was low. The low sensitivity was attributed to the incompatibility of the manganese emission (600 nm) with the spectral sensitivity of photomultiplier (PM) tube. An improvement in the sensitivity was achieved by replacing manganese with copper. The $Li_2B_4O_7$ powder doped with copper showed an emission band that was more compatible with the spectra of PM tubes (~400–460 nm) (Takenaga et al., 1980). A study conducted by Wall et al. (1982) proved that the sensitivity of the Cu-doped $Li_2B_4O_7$ to ^{90}Sr β-rays is nearly eight times higher than the sensitivity of $Li_2B_4O_7$ doped with Mn. The same study showed that, in 10 days after irradiation, the fading of the signals of $Li_2B_4O_7$:Cu and $Li_2B_4O_7$:Cu,Ag was about 85%. The dose response of the Mn-doped glass was essentially linear up to 1 Gy, whereas the dose responses of glasses doped with Cu, or Cu

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and Ag, were linear up to 10 Gy (Wall et al., 1982). Dosimetric advantages of this phosphor, either undoped or Cu-doped, were reported (Lakshmanan and Bhuwan Chandra, 1982, Takenaga et al., 1983, Srivastava and Supe 1989, Martini et al., 1995 and Alajerami et al., 2013). El-Faramawy et al. investigated the promising properties of $Li_2B_4O_7$:Cu, particularly the high linearity of its response and high sensitivity. They demonstrated the ability of $Li_2B_4O_7$:Cu to measure low doses down to 20 μGy. The sensitivity of home-made $Li_2B_4O_7$:Cu to ^{137}Cs radiation was twice as high as that of TLD-100, and the signal faded three months after the irradiation was 11%. The authors attributed the absence of supralinearity to copper in lithium borate (El-Faramawy et al., 2000). Recently, many researchers explored the mechanism of luminescence in borate glasses (Alajerami et al., 2013; Prokic, 2002; Liu et al., 2005; Elkholy, 2010; Puppalwar et al., 2012). In 2001, Furetta et al. used lithium borate with copper as a dopant and indium as a co-dopant. That material exhibited higher sensitivity and was capable to detect doses down to 10 μGy (Furetta et al., 2001). The dose responses of both $Li_2B_4O_7$:Cu,In and $Li_2B_4O_7$:Cu were linear in the range 10^{-4}–10^3 Gy (^{60}Co). Moreover, the fading was slower, about 10% in 3 months (Furetta et al., 2001).

Prokic (2002) determined characteristics of sintered $Li_2B_4O_7$ co-doped with several dopants, such as Cu, In; Cu, In, Ag; and Mg, Cu, P; exposed to ^{60}Co gamma radiation in the dose range 10^{-4}–3×10^3 Gy. The glow curve of $Li_2B_4O_7$ doped with Cu, Ag, and P featured a pronounced peak at 185–190 °C, and the radiation sensitivity of the material was five times higher than that of LiF: Mg, and Ti. The signal of $Li_2B_4O_7$:Cu, in faded 6% 3 months after irradiation; the fading of the signal of $Li_2B_4O_7$:Cu,In,Ag was 10% in the same period. The dose detection limit, defined as the dose producing a signal equal to three standard deviations of readings of unirradiated samples, was estimated to be 10 μ Gy (Prokic, 2002). Alkaline-earth borates, unlike the glasses with other modifiers, were less hygroscopic (Fukuda et al., 1984; Fukuda and Takeuchi 1985; Fukuda et al., 1986; McKeever et al., 1995). Venkateswara Rao et al. (2002) studied the thermoluminescence properties of a new borate glass system after an X-ray irradiation (35 kVp, 10 mA) at room temperature for 1 h. The phosphor was modified with lithium oxide and calcium fluoride (Li_2O–CaF_2–B_2O_3) and doped with MnO in various concentrations. They found that the TL intensity was decreasing with increasing MnO concentration. This quenching was attributed to the presence of Mn^{3+} ions (Venkateswara Rao et al., 2002). The authors assumed that there was a competition between the trapping of the electrons released during the excitation and the readout process. At the maximal concentration, the contiguous activator (traps) inhibited the radiative luminescence, and the opposite effect was observed in the TL intensity.

According to an interesting paper by Pekpak et al. (2009), the glow curve consists of several peaks, with the main peak at 185–235 °C. The range of the linearity of the dose response extended as far as to 1000 Gy, and the signal lost about 5% of its intensity in one month (the dosimeters were exposed to the ^{90}Sr-^{90}Y beta radiation at a dose rate of 0.5 Gy/min at room temperature for 5 minutes). Elkholy (2010) showed that adding magnesium oxide to the lithium borate glass produced a dosimeter capable to work in the dose range of 0.1–20 kGy (^{60}Co). However, there was no significant research to investigate the effect of TiO_2 on the TL properties (dose response) of the borate glass. Despite the general agreement between the studies of the role of titanium ions in the glassy host, the mechanism of the TL enhancement is far from being understood. It is assumed that titanium occurs in the glass mixture as Ti^{3+} and Ti^{4+}. In a material with a higher relative Ti^{4+} concentration, some of the electrons released by the thermal treatment would be effectively captured by the Ti^{4+} ions, whose energy levels are close to the Fermi level, by non-radiative recombination. The remaining free electrons would be trapped by other holes on the network. It is well known that hosts with high concentrations of Ti^{4+} have weaker TL responses (Balaji Rao et al., 2004; Balaji, et al., 2005; Nageswara Rao et al., 2005). The aim of the present work was to study a new material, namely, a borate glass modified with lithium and potassium carbonate and co-doped with titanium and magnesium oxides (LKB:Ti,Mg). Of the primary interest were potential applications of this glass in the dosimetry of high-energy photons and electrons.

2. Experimental

2.1. Glass preparation

Boric acid was used as a glass former; lithium and potassium carbonate were added as glass modifiers. This mixture was initially activated with a small amount of titanium oxide (0.3–1.0 mol%). Then, magnesium oxide was added as a co-activator in a concentration between 0.05 and 0.50 mol% to provide the optimal TL response. All the used reagents were of high purity (main component concentration at least 99.99%) and obtained from Sigma-Aldrich (Germany). The composite was well mixed for 40 min and then kept in an alumina crucible in an electrical furnace with the temperature around 1200 °C for 45 to 60 min (depending on the dopant concentration). The melt was poured into a steel mold (400 °C) and kept on it for 3 h to reduce the mechanical stress. The glass transition temperature was determined by thermal analysis of a preliminary sample; it was slightly lower than the first annealing temperature. Table 1 lists the compositions of the prepared samples and their thermal parameters.

2.2. Dosimeter preparation and irradiation

All the prepared glass samples had a rectangular form ($2.5 \times 2.5 \times 1$ mm^3) and were transparent. The transparency of the glass varied with the dopant concentration. Transparency of a TLD dosimeter facilitates the collection of recombination light from its inner parts (Bos, 2007). The optical properties of these mixtures have been previously described (Alajerami et al., 2012). Measurements of glow curves of the samples were performed with a Harshaw TLD reader Model 4500 at a secondary standard dosimetry laboratory (SSDL) at the Malaysian Nuclear Agency. The reader had an infrared filter to absorb the thermal noise. Background signals were subtracted from all the TL glow curves. The samples were irradiated with a ^{60}Co source to various doses in a “solid water” phantom (dose rate ~5.232 mGy min^{-1}) at room temperature. The samples were at d_{max}, SSD was 100 cm, and the filed size was 10×10 cm^2. Initially, the glow curves were recorded at a fixed heating rate (20 °C s^{-1}) in the temperature range 50–400 °C. A continuous flow of gaseous nitrogen was used during the readouts to reduce chemiluminescence in the heated samples. Before the TL measurements, the irradiated samples were kept in a

Table 1
The compositions and thermal properties of the glass samples.

Glass	Composition (mol%)					Thermal characteristics, °C		
	Li_2CO_3	H_3BO_3	K_2CO_3	TiO_2	MgO	T_g	T_c	T_m
Sample 0 (S0)	20.00	70.00	10.00	–	–	490	633	900
Sample 1 (S1)	20.00	69.70	10.00	0.30	–	497	628	910
Sample 2 (S2)	20.00	69.50	10.00	0.50	–	505	625	913
Sample 3 (S3)	20.00	69.30	10.00	0.70	–	512	620	920
Sample 4 (S4)	20.00	69.00	10.00	1.00	–	510	645	927
Sample 5 (S5)	20.00	69.45	10.00	0.50	0.05	515	635	936
Sample 6 (S6)	20.00	69.40	10.00	0.50	0.10	520	632	928
Sample 7 (S7)	20.00	69.25	10.00	0.50	0.25	530	627	913
Sample 8 (S8)	20.00	69.00	10.00	0.50	0.50	540	620	915