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Optically stimulated luminescence of borate glasses containing magnesia, quicklime, lithium and potassium carbonates

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ABSTRACT

The OSL characteristics of three different borate glass matrices containing magnesia (LMB), quicklime (LCB) or potassium carbonate (LKB) were examined. Five different formulations for each composition were produced using a melt-quenching method and analyzed in terms of both dose-response curves and OSL shape decay. The samples were irradiated using a $^{90}Sr/^{90}Y$ beta source with doses up to 30 Gy. Dose-response curves were plotted using the initial OSL intensity as the chosen parameter. The OSL analysis showed that LKB glasses are the most sensitive to beta irradiation. For the most sensitive LKB composition, the irradiation process was also done using a ^{60}Co gamma source in a dose range from 200 to 800 Gy. In all cases, no saturation was observed. A fitting process using a three-term exponential function was performed for the most sensitive formulations of each composition, which suggested a similar behavior in the OSL decay.

1. Introduction

The use of glass in the dosimetry of ionizing radiation has been amply investigated, particularly when high radiation doses are involved. Radiation detection applications are pointed out as one of the emerging research areas in glass science (Mauro et al., 2014). In particular, over the past decade, borate glasses have been studied as thermoluminescent dosimeters (TL) (Rojas et al., 2006; Ayta et al., 2010, 2011).

Borate glass matrices based on lithium carbonate, potassium carbonate and boric acid (LKB), doped or co-doped with different materials, have shown good thermoluminescence characteristics in terms of response linearity and sensitivity; moreover, their effective atomic number (Z_{eff}) is similar to that of biological soft tissue (Alajerami et al., 2013a, 2013b; Hashim et al., 2014). Optically stimulated luminescence (OSL), also known as photostimulated luminescence, was also observed in some borate glasses (Qiu et al., 1997), and opened new possibilities for radiation dosimetry (Nanto et al., 2015; Marini et al., 2015). However, borate glasses present high hygroscopicity, which can significantly limit their practical applicability as dosimeters.

The only two OSL dosimeters that are commercially available are both crystalline (Al_2O_3:C and BeO). To the best of our knowledge, no vitreous matrices are currently in use as OSL dosimeters. Glass matrices can be appealing compared to crystalline materials due to characteristics as easy preparation, inexpensive production, possibility to cast large and uniform pieces, and high transparency. These matrices are focus of our research, and we hereby present a comparison between the OSL characteristics of various formulations of three borate glasses: Li_2CO_3–K_2CO_3–B_2O_3 [LKB], Li_2CO_3–MgO–B_2O_3 [LMB], and Li_2CO_3–CaO–B_2O_3 [LCB].

In this work, we investigated the effect of adding MgO in high molar ratios to borate glasses. Magnesia (MgO) has been proposed as a dopant increasing the luminescent signal of glass matrices (Alajerami et al., 2013b, 2013c). In our case, we added magnesia also because it is known to reduce glass hygroscopicity, as confirmed by Marini et al. (2015).

Quicklime was added because it is used in glass production for increasing the final product durability and chemical wear resistance. Since Ca and Mg have similar valence characteristics, we also tried this oxide to our formulations in view of investigate its impact on the OSL signal, which is not reported in the literature.

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Table 1
Produced glasses and purity level of the reagents.

Reagents (purity level)	Li2CO3–K2CO3–B2O3 [LKB] mol%	Li2CO3–MgO–B2O3 [LMB] mol%	Li2CO3–CaO–B2O3 [LCB] mol%
B_2O_3	20Li2CO3. 10K2CO3. 70B2O3	20Li2CO3. 10MgO. 70B2O3	20Li2CO3. 10CaO. 70B2O3
(> 98%)	**(L10KB)**	**(L10MB)**	**(L10CB)**
Li_2CO_3	20Li2CO3. 15K2CO3. 65B2O3	20Li2CO3. 15MgO. 65B2O3	20Li2CO3. 15CaO. 65B2O3
(> 99%)	**(L15KB)**	**(L15MB)**	**(L15CB)**
K_2CO_3	20Li2CO3. 20K2CO3. 60B2O3	20Li2CO3. 20MgO. 60B2O3	20Li2CO3. 20CaO. 60B2O3
(99%)	**(L20KB)**	**(L20MB)**	**(L20CB)**
MgO	20Li2CO3. 25K2CO3. 55B2O3	20Li2CO3. 25MgO. 55B2O3	20Li2CO3. 25CaO. 55B2O3
(97%)	**(L25KB)**	**(L25MB)**	**(L25CB)**
CaO	20Li2CO3. 30K2CO3. 50B2O3	20Li2CO3. 30MgO. 50B2O3	20Li2CO3. 30CaO. 50B2O3
(98%)	**(L30KB)**	**(L30MB)**	**(L30CB)**

2. Materials and methods

Table 1 shows our borate glass formulations and the purity level of our reagents. The glasses were produced using a fixed amount of lithium carbonate (Li_2CO_3; 20 mol%) and various amounts of the glass former boron oxide (B_2O_3) as well as various percentages of three additives (K_2CO_3, MgO, and CaO).

All glass formulations were produced using a melt-quenching method. The mixtures were first kept at 1100 °C for 30 min in a platinum crucible, and then cooled down by placing them between two bronze sheets at 0 °C. After vitrification, the compositions were thermally treated at 350 °C for 24 h to reduce residual stresses.

The final sample structure was investigated by X-ray diffraction (XRD) scanning grains below 75 μm with a Rigaku diffractometer RINT-Ultima Plus 2000/PC unit from 10° to 80° (2θ), with a scan rate of 1°/min, at ambient temperature. A thermal characterization was done by differential thermal analysis (DTA) with a DTA – 50 Shimadzu unit, placing grains below 53 μm in an alumine crucible and a nitrogen atmosphere and using a heating rate of 20 °C/min from 30 °C to 800 °C.

For each formulations, twelve pellets were prepared according to the procedures described by Marini et al. (2015) and their optically stimulated luminescence (OSL) was examined. A minimum of three cycles of irradiation/reading/bleaching were done on each batch using a fixed dose. Only pellets presenting a standard deviation within 10% of the mean were selected.

The dosimetric analysis was done using a Risø TL/OSL reader. A $^{90}Sr/^{90}Y$ beta source built into the reader, producing a dose rate of 0.1 Gy/s, and a ^{60}Co gamma source producing a dose rate of 0.933 kGy/h, were used for the sample irradiations. A blue LED with a peak emission at 470 nm (FWHM=20 nm) was used as stimulation light and a Hoya U-340 filter (transmission between 290 and 390 nm) was used for the emitted luminescence. The OSL measurements were performed with continuous wave stimulation (CW-OSL) applied right after the irradiations. The initial OSL intensity (the integral over the first 0.16 s of the OSL emission curve) was the parameter chosen for the dose-response. The LMB and LCB batches were irradiated with doses ranging from 5 to 30 Gy, while LKB was irradiated with doses ranging from 1 to 7 Gy, in all cases using the $^{90}Sr/^{90}Y$ source. The L15KB samples were also irradiated to doses ranging from 200 to 800 Gy with the ^{60}Co source; in this case, a collimator with a 1 cm opening was used in front of the photomultiplier to avoid saturation damage.

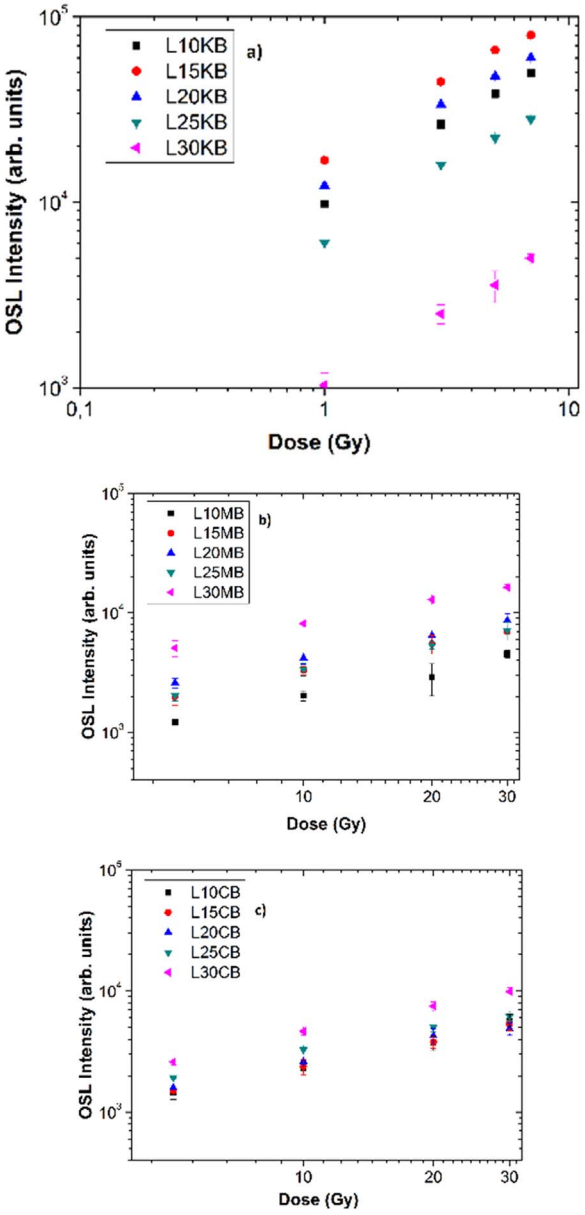

Fig. 1. Pellet dose-response curves. Samples of a) LKB, b) LMB, and c) LCB.

3. Results and discussion

The XRD analysis showed the presence of broad bands instead of

Table 2
Glass transition temperature (Tg), crystallization temperature (Tc), and a measure of glass stability (Ts) for each produced composition.

Sample	Tg (°C)	Tx (°C)	Ts (°C)	Sample	Tg (°C)	Tx (°C)	Ts (°C)	Sample	Tg (°C)	Tx (°C)	Ts (°C)
L10KB	456.9	481.6	24.7	**L10MB**	516.9	546.2	29.3	**L10CB**	516.7	542.0	25.3
L15KB	432.7	459.1	26.4	**L15MB**	507.2	539.1	31.9	**L15CB**	519.2	546.3	27.1
L20KB	418.4	439.3	20.9	**L20MB**	522.3	550.1	27.8	**L20CB**	526.3	551.2	24.9
L25KB	407.0	422.4	15.4	**L25MB**	516.4	545.3	28.9	**L25CB**	513.5	538.1	24.6
L30KB	394.7	409.5	14.8	**L30MB**	504.0	536.5	32.5	**L30CB**	509.7	535.1	25.4

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