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Dedicated to Academician Professor Dr. Emil Burzo on His 80th Anniversary

THERMOLUMINESCENCE PROPERTIES OF THE $0.5P_2O_5$ - xBaO - (0.5-x)K_2O GLASS SYSTEM. A POSSIBLE DOSIMETRIC MATERIAL

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ABSTRACT. Thermoluminescence (TL) properties of freshly β irradiated phosphate glasses doped with BaO and K_2O oxides at various concentrations were investigated. Barium-doped glasses ($0.5P_2O_5$ - 0.5BaO) show two TL peaks centered at 180 ^{0}C and 380 ^{0}C due to the defects generated by modifier Ba^{2+} ions inserted into the glass network. In the case of potassium-doped glasses ($0.5P_2O_5$ - $0.5K_2O$) an intense TL peak at 280 ^{0}C with an weak shoulder at 150 ^{0}C appear. The TL emission of the other phosphate glasses, $0.5P_2O_5$ - xBaO - (0.5-x)K_2O with $0.1 \leq x \leq 0.4$, containing both type of the network modifier ions (K^+, Ba^{2+}) consist from the overlap of the above – mentioned luminescence spectra depending on the local energetic level diagrams of the luminescence centers. A linear dependence ($R^2 > 0.99$) of the integral TL signals with the absorbed doses were evidenced for all the investigated glasses which can be considered as good materials for dosimetry in the 0 – 50 Gy range.

Keywords: *P_2O_5 - BaO – K_2O glasses, TL dosimetry*

INTRODUCTION

Radiation dosimetry with solid luminescent materials is a well-established technique of monitoring ionizing radiation. Successful applications of thermoluminescence (TL) dosimetry are a result of search for materials that can be used as detectors of ionizing radiation and analysis of their properties [1].

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Inorganic crystals such as LiF: Mg, Ti and LiF: Mg, Cu, P have been traditionally used for gamma and X-ray TL dosimetry applications. Various TL materials are in use today as dosimeters; however, as far as optically stimulated luminescence (OSL) is concerned, Al_2O_3:C is virtually the only synthetic material currently used in medical, environmental and personal dosimetry [2]. OSL has certain advantages over TL, such as higher precision, flexibility and ease of use, as well as the possibility of performing real-time measurements and future dose reassessments, if desired.

Extensive research is underwayto introduce new dosimetric systems into traditional in-phantom measurements, mailed dosimetric services for radiotherapy beam calibrations and two-dimensional TL dosimetry. Promising solutions are also being developed for online in-vivo dosimetry using OSL systems [3].

TL and OSL glass dosimeters are of particular interest because of their optical transparency, which results in an overall improvement of the efficiency of phosphor. Other characteristics that make them very suitable are their relatively simple preparation, easy shaping and long-term stability. Due to these advantages many studies have been dedicated to investigation and improvement of luminescent properties of glass systems [4-9].

Thus the effect of the Fe_2O_3 concentration on the TL properties of PbO–Sb_2O_3–As_2O_3 glasses in the light of different oxidation states of iron ions has been reported in the paper [10]. Also the Judd–Ofelt theory has been successfully used to characterize the absorption and luminescence spectra of Tb^{3+} ions in BaO–M_2O_3(M = Ga, Al, In)–P_2O_5 glasses [11, 12].

The thermoluminescence effect of the MgO at different concentrations on the B_2O_3-Li_2O glass system and TL properties of Cu-doped lithium potassium borate glasses have been reported in the papers [13] and [14], respectively.

The recent investigation of the Sm^{3+} - doped cadmium borate glass shown that this can be considered as a possible TL – dosimeter [15]. The Dy^{3+} - doped of alkali – silicate glasses [16] and of lithium magnesium borate glasses [17] suggested that these materials can be also used in radiation dosimetry measurements.

This paper presents the results of an investigation of TL properties of the $0.5P_2O_5$-xBaO-(0.5–x)K_2O glass systems ($0.1 \leq x \leq 0.4$) undertaken to evaluate their usability as luminescence dosimetric materials.

EXPERIMENTAL

Starting materials to obtain the $0.5P_2O_5$-xBaO–(0.5–x)K_2O glass systems with $0.1 \leq x \leq 0.4$ were reagent grade $(NH_4)_2HPO_4$, $BaCO_3$ and K_2CO_3. The samples were prepared by mixing powders of the components in suitable proportions and melting the mixture in sintered corundum crucibles at 1200 ^oC for 1h. The mixtures were put into a furnace already stabilized at this temperature. The obtained glass samples were quenched by pouring the molten glass onto a stainless steel plate. The obtained transparent, homogenous and color-free glass pellets with a thickness of 1 mm were used in the experiments.

All TL signals were recorded at a controlled heating rate of 5 $^oC/s$ in nitrogen atmosphere with a RISØ TL/OSL DA-20 machine. Luminescence emissions have been detected using a bialkali EMI 9235QA photomultiplier tube using a HoyaU-340 filter (transmission between 290 and 390nm).

Irradiations were carried out at room temperature in a homogenous field of a ^{60}Co gamma source with a dose rate of 5.2Gy/h or individually and automatically in the luminescence reader using a ^{90}Sr–^{90}Y beta source with a dose rate of 0.05 Gy/s and which had been preliminarily calibrated against the gamma source.

RESULTS AND DISCUSSION

TL glow curves recorded at a controlled heating rate (5 $^oC/s$) immediately after β - irradiation to 25Gy are shown in Fig.1. No TL signals appear in the simple phosphate glass (P_2O_5). In the case of freshly irradiated $0.5P_2O_5$ - 0.5BaO glass, a narrow peak centered at 180 ^oC and also a broad intense peak in vicinity of 380 ^0C can be observed. An intense TL peak centered around 280 ^0C with an weak shoulder at 150 ^0C appear in $0.5P_2O_5$ - $0.5K_2O$ glass.

The differences between TL spectra of the two types of phosphate glasses may be attributed to the differences which appear in the energetic level diagrams of luminescent centers (defects) generated by the modifier ions (Ba^{2+}, K^+) inserted into the phosphate glass network.

TL glow curves of the mixed $0.5P_2O_5$ – xBaO - (0.5–x)K_2O glasses with $0.1 \leq x \leq 0.4$ consist from the overlapping of the above-mentioned peaks. In the case of glasses containing both modifiers ions (Ba^{2+}, K^+) the structures of energetic levels characteristics to luminescent centers are changed and thus also the positions of luminescent peaks. The shapes of luminescent spectra are conditioned by the realized local structure of inserted cations (Ba^{2+}, K^+) and proper energetic level diagrams of luminescent centers in phosphate network. The local structural changes of these glasses with different Ba^{2+}, K^+ ions content are also evidenced by FT-IR and FT-Raman measurements. These results are in progress and will be published.

TL glow curves after freshly irradiated with ^{90}Sr – ^{90}Y, integral TL output as function of the absorbed dose and reproductibility tests after 10 measurement cycles when the sample was gamma irradiated of 10 Gy dose and then immediately heated to 500 ^oC for barium – phosphate, potassium – phosphate and mixed (Ba, K) phosphate glasses are shown in Figs. 2, 3.

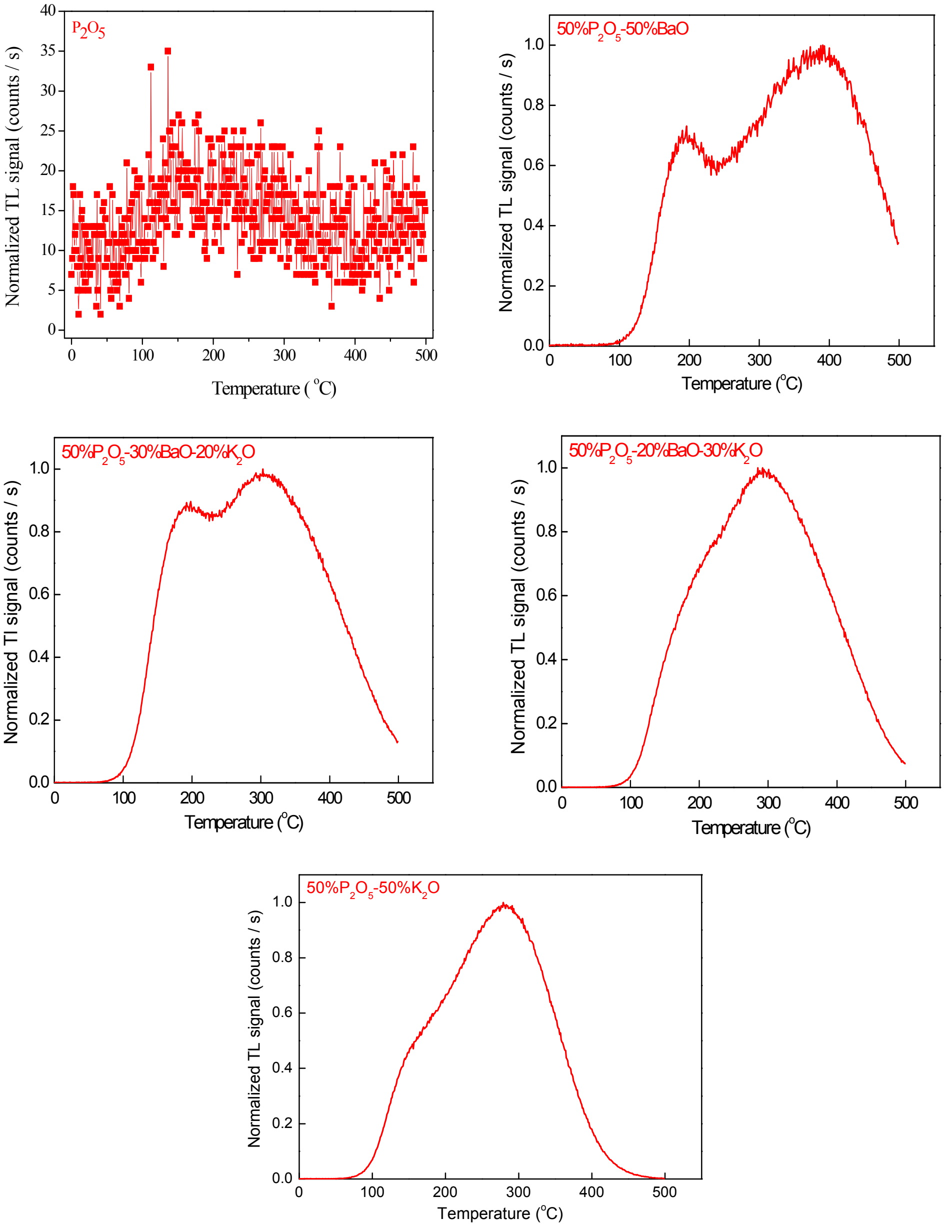

Fig. 1. TL glow curves of the P_2O_5 – BaO – K_2O glass system.

All measurements were performed with a single aliquot of each glass specimen because it had been observed that the process of recording the TL signal (ramp heating to 500 °C) reduces all signals to a negligible level (3% of the response recorded after an irradiation to 10 Gy).

Phosphate glass doped with barium oxide is at least one order of magnitude more sensitive to radiation than the glass doped with potassium oxide. An analysis of twelve aliquots showed that intensity ofthehigh-temperature peak produced by a 1-g portion of $0.5P_2O_5$–0.5BaO is about 50 times lower than the intensity produced by the traditional LiF:Mg, Ti(TLD100) pellet irradiated to the same dose and measured with the same setup.

However, very good linear dependences ($R^2 > 0.99$) of the integral TL signals can be observed for both dosimetric peaks of $0.5P_2O_5$ - 0.5BaO up to 50Gy (Fig. 2). The linear response at doses above 10Gy is a common characteristic of many TL materials what are very attractive for high-dose measurements [18].

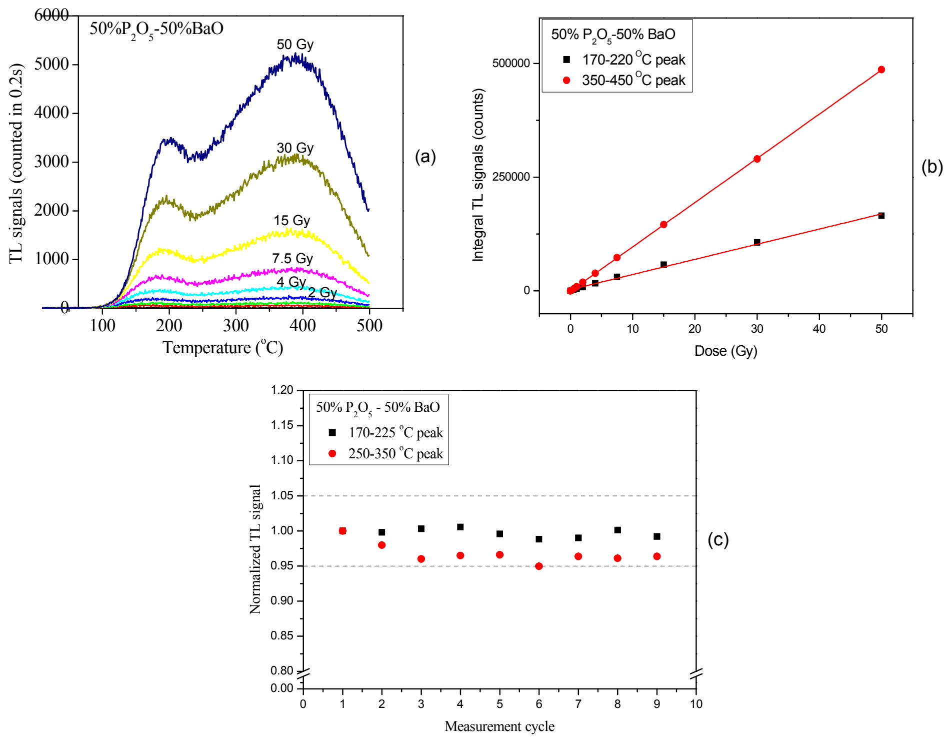

Fig. 2. Responses of β - irradiated $0.5P_2O_5$ – 0.5BaO glass: (a) – TL glow curves after freshly irradiation with ^{90}Sr – ^{90}Y; (b) – Integral TL output as a function of the adsorbed dose; (c) – Reproductibily tests after 10 measurement cycles when sample was gamma irradiated of 10Gy dose and then immediately heated to 500°C.

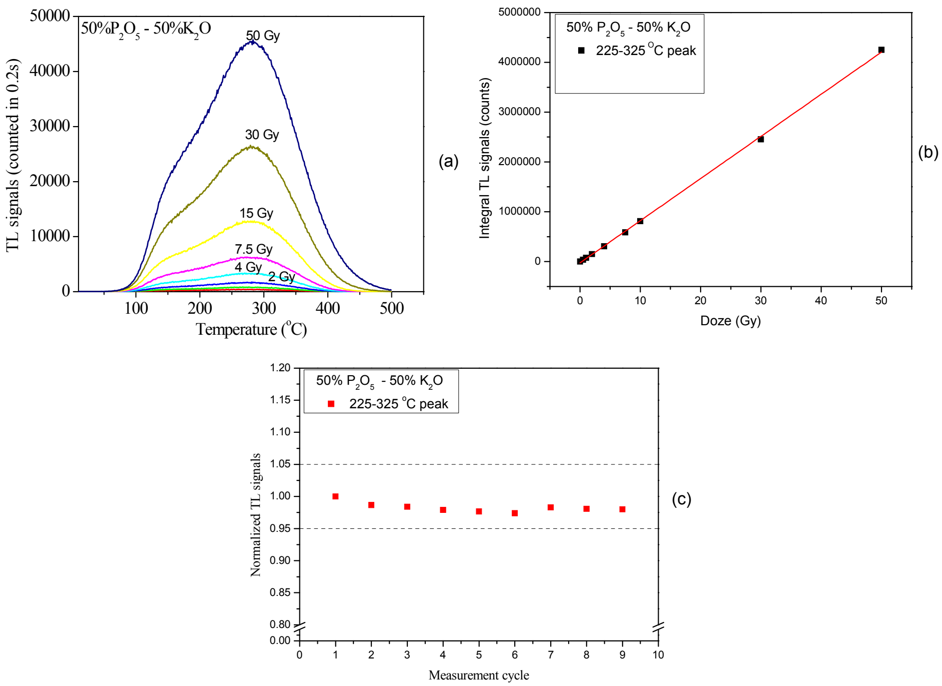

Fig. 3. Responses of β - irradiated $0.5P_2O_5 – 0.5K_2O$ glass: (a) – TL glow curves after freshly irradiation with ^{90}Sr – ^{90}Y; (b) – Integral TL output as a function of the adsorbed dose; (c) – Reproductibily tests after 10 measurement cycles when sample was gamma irradiated of 10Gy dose and then immediately heated to 500°C.

An analogous dependence of the integral TL signals on the absorbed dose was also found in the case of $0.5P_2O_5–0.5K_2O$ (Fig.3) and $0.5P_2O_5$ - 0.3BaO - $0.2K_2O$ glasses. This might be an indication that the mechanism of the underlying luminescence process can be described by the first-order kinetics [19].

Reproducibility tests were also performed with 12 aliquots, each one repeatedly gamma irradiated of 10 Gy and then immediately heated to 500 °C over the course of 3 months for each type of the investigated glasses. Figs (2c-3c) show the average behavior of 12 aliquots over 10 measurement cycles. None of the investigated aliquots showed a pronounced sensitization or desensitization trend.

To estimate the threshold dose and the minimal detectable dose, we recorded the TL signals from a set of 10 unirradiated glass samples. The threshold dose D_0 was calculated as $D_0 = (B + 2\sigma_B)F$, where F is the calibration factor expressed in Gy/TL, B is the mean TL signal obtained for the background and σB is the standard deviation of the background signal. For the high-temperature peak, a value of 0.2Gy was obtained. The low limit of detection for this peak was estimated to be 0.4Gy at 95% confidence level according to Eq.(8) in the paper [20].

CONCLUSIONS

Thermoluminescence properties of the investigated glsses depend on the relative ratio content of the incorporated network modifiers oxides (BaO, K_2O).

Two TL peaks centered at 200°C and 400°C were observed for $0.5P_2O_5$ – 0.5BaO glass, and only an intense TL peak at 280°C with an weak shoulder at 150°C appear in the case of $0.5P_2O_5$ – $0.5K_2O$glass.

TL emissions of the other phosphate glasses containing both type of detects generated by modifier Ba^{2+}, K^+ ions consist from the overlap of the above – mentioned luminescence spectra.

All the investigated glasses can be considered as good materials for dosimetry until 50 Gy due to the linear dependence ($R^2 > 0.99$) of the integral TL signals with the absorbed doses.

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