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TL-response and Trap Parameters Calculation of B_2O_3 +P_2O_5 + Li_2O + Al_2O_3+Na_2O+K_2O Doped with La_2O_3 Boro-Phosphate Glass.

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Abstract: The structure and thermoluminescence (TL) characteristics of the newly prepared La_2O_3 doped boro-phosphate glass were explored. La_2O_3 was added in 0.7 mol. % concentration to the glass structure. A reasonable response was obtained with an abroad peak contained at about 194 ºC. X-ray diffraction (XRD) was used to characterize the prepared sample. Deconvolution was made to obtain the other dosimetric characteristics; trap parameters, using the PeakFit program. Four peaks were obtained from the deconvolution at about 150,169, 183, and 195 ºC. The activation energy was calculated and found to range between 0.788 - 1.45 eV for the deconvoluted peaks. In addition, the frequency factor (s) was estimated and found to be 4.07×10^{15} -6.095×10^{7} s^{-1}. The obtained results showed reasonable TL- characteristics of the prepared samples.

Keywords: Boro-phosphate, Deconvolution, PeakFit program.

1 Introduction

Radiation dosimetry is routinely carried out for personnel and radiation workers' occupational safety because of the danger of radiation exposure[1]. As a result, different dosimeters are used worldwide to detect various types of radiation, including X-rays, gamma, and neutrons. The thermoluminescence (TL) technique has been used for several purposes, including dating archeological artifacts, identifying defects in solids, and measuring and detecting absorbed radiation[2]. It is still widely used as a dose-measurement system in hospitals and medical facilities worldwide to measure entrance doses for a handful of patients and in industrial applications, including nuclear reactors, petroleum, businesses, and radiation safety officers (RSOs)[3].

Glass is a reasonable class of TLD material because of its ease of fabrication, chemical stability, and tunable optical properties [4]. As the primary network formers, Boro-phosphate glasses have shown significant potential for TL applications due to their excellent trapping and recombination mechanisms[5]. The addition of modifiers such as Li_2O, AlF_3, Na_2O, and K_2O enhances the structural and optical properties of the glass network, improving its sensitivity and luminescence efficiency. Furthermore, doping with rare-earth oxides, particularly La_2O_3, introduces unique trap levels within the glass matrix, which are crucial for the TL response[6]. The introduction of La_2O_3 into the glass system has improved both the luminescence yield and the stability of TL signals, making it a promising dopant for TL materials[7].

The present research will focus on studying the role of adding lanthanum with 0.7% mol%. Concentration to glass constituents (B_2O_3-P_2O_5–Li_2O–Al_2O_3-Na_2O-K_2O), and calculating the trap parameters using the PeakFit program contributing to advancements in radiation detection technologies.

2 Experimental

A glass system with stoichiometric composition (54.3 %B2O3 – 25% P_2O_5 – 12% Li_2O – 2% Al_2O_3-3%Na_2O-3%K_2O-0.7% La_2O_3) mol. %, all of the raw

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material was melted in an electric oven at 1100 ºC. The molten mixture from each type was poured into a stainless steel plate and annealed for two hours at 400 degrees Celsius in a furnace.

2.1 Characterization

Before irradiation and thermoluminescence measurements, the boro-phosphate glass sample was characterized using X-ray diffraction (Shimadzu X-ray diffractometer, 600, forty kV, and 15 mA, having X-ray tube and copper -Kα target with 2θ=10°-90°) to ensure the morphism of the powdered glasses.

2.2 Irradiation

The sample was irradiated using 137Cs γ-ray source with a dose rate of 0.33 Gy/min at 25 ± 5 oC. It is available at the National Centre for Radiation Research and Technology (NCRRT) in Cairo, Egypt. Before irradiation, the prepared sample was annealed up to 400 ºC for 30 minutes.

2.3 TL-measurements

The properties of the prepared sample were measured by The TL-reader Thermo Scientific Harshaw Model 3500 was used to perform the TL measurements. The reader is operated by the WinREMS program, which runs on a computer that is connected to the reader and cooled by a nitrogen supply. TL glow curves were acquired at a chosen heating rate of 5 K s−1 and in the temperature range of 323 to 673 K.

2.4 Trap parameters calculation.

By tracking the mobility of electrons between trapping level and recombination sites, the thermoluminescence (TL) technique is a useful tool for researching how ionizing radiation interacts with defects created inside the material lattice. This mobility may be simple, like when an electron releases from a trap level and recombines into a recombination center, or it may be complex, involving numerous events like electron escape, trapping, releasing, tunneling, and recombining through trap levels and recombination centers before light emission [8]. For every phosphor, a fingerprint TL glow curve is created by charting the intensity of light emitted by irradiated TL materials across the heating temperature. The dosimetric kinetic characteristics of the trap centers inside the lattice, including the order of kinetic b, activation energies E (eV), and frequency factor S (s-1)[9]. Activation energy is the amount of heat energy needed to release trapped holes and electrons. In addition, the frequency factor is a constant characteristic of the electron trap known as the "pre-exponential frequency factor." It is proportional to the frequency of electron collisions with the lattice phonons.

The activation energy (E) and frequency factor (S) can be evaluated in a variety of methods such as the Peak fit program (PFP) and the Peak Shape (PS) method.

2.4.1 PeakFit program.

The ability to apply computerized glow curve deconvolution (CGCD) in a variety of overlapping-peak glow curves without requiring heat treatment gives an advantage over experimental techniques. The PeakFit program is a simple and effective technique for extracting the crucial intrinsic trapping parameters, E(eV) and s (s-1), from the TL peaks of complex glow curves. The quality of the fitting will be estimated for trap parameters determination using the goodness fit (FOM) equation, which is determined [10, 11] as follows:

$$\mathrm{FOM} = \sum \frac{100\times \left|I_{exp} - I_{GCD}\right|}{\sum I_{GCD}} \qquad (1)$$

Where I_{exp} is the experimental point, I_{GCD} is the fitted points, and $\sum I_{GCD}$ Is the peak area of the fitted glow curve. The goodness of fitting depends on the value of FOM where a value of < 0.25 gives a good fitting. The deconvolution procedure has been conducted using the commercial software PeakFit v.4.12. Evaluation of (E) in terms of temperature of maximum peak intensity (Tm) and full width at half maximum (FWHM) [12-14] are provided by:

$$E = 2.417 k_B \frac{T_m^2}{\mathrm{FWHM}} - k_B T_m \qquad (2)$$

Where (KB) is Boltzmann's constant.

2.4.2 Peak shape method

By examining the glow curves' shapes, this technique could potentially be applied to determine the trap parameters' values [18]. The three temperature points Tm, T1 (a rising portion of the peak's temperature at half intensity), and T2 (descending part temperature of TL glow peak at half intensity) are used to extract shape parameters like ω (full half-width), δ (the half width on the side with lower temperatures), and half-width value at higher temperature (τ). These shape parameters can be used to determine symmetry's value factor (μg). Values of μg that correspond to general, first, and second-order kinetics, respectively, are 0.49, 0.42, and 0.52. The calculation of μg and E is as follows:

$$\mu_g = \frac{T_2 - T_m}{T_2 - T_1} \qquad (3)$$

$$E_\propto = C_\propto \left(\frac{k_B T_m^2}{\propto}\right) - b_\propto \left(2 k_B T_m\right) \qquad (4)$$

Where α represents the three parameters ω, δ and τ. Cα can be represented as follows:

$$C_\tau = 1.51 + 3\left(\mu_g - 0.42\right) \quad (5)$$

$$C_\delta = 0.976 + 7.3\left(\mu_g - 0.42\right);\ b_\delta = 0 \quad (6)$$

$$C_\omega = 2.52 + 10.2\left(\mu_g - 0.42\right);\ b_\omega = 1 \quad (7)$$

And Eα denoted the three sets of equations with various parameter values τ, δ, or ω. The average values of Eα gave the required activation energy. From activation energy we can calculate the frequency factor (s), decay time (τ_c) and probability of escaping (P) from the following equations [19] :

$$s = \frac{\beta E}{k_B T_m^2} \exp\left(\frac{E}{k_B T_m}\right) \quad (8)$$

Where β is the heating rate.

$$\tau_c = s^{-1} e^{E/KT} \quad (9)$$

Where τ_c is the decay time, and T is room temperature in kelvin (298ºK).

$$P = \frac{1}{\tau_c} \quad (9)$$

The recombination process's efficiency is determined by the escape probability [20].

3 Results and Discussion

3.1 X-ray diffraction analysis

X-ray diffraction analysis can establish a material's amorphous or crystalline nature. Figure 1 illustrates the glassy state of all powders. It became apparent that there were no diffraction peaks. Only wide bands were seen at about 2θ = 25º, approving the lack of crystalline peaks and confirming that the sample was glass.

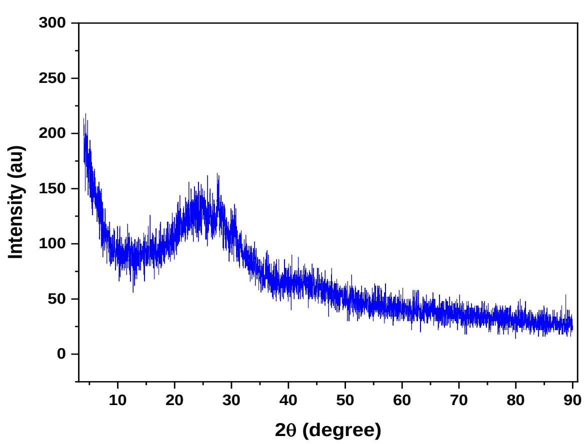

Fig. 1. XRD for (B2O3 – P2O5 – Li2O –Al2O3-Na2O-K2O doped with 0.7mol. % La2O3) sample.

3.2 Glow curve of prepared glass

The experiment was carried out by exposing the prepared sample to 5 Gy γ-dose. The resulting TL intensity was normalized to its mass. From the illustrated figure, the sample shows high sensitivity. This glow curve showed an abroad peak centered at about 194 ºC. Therefore, this sample was selected for calculating trap parameters by PeakFit technique.

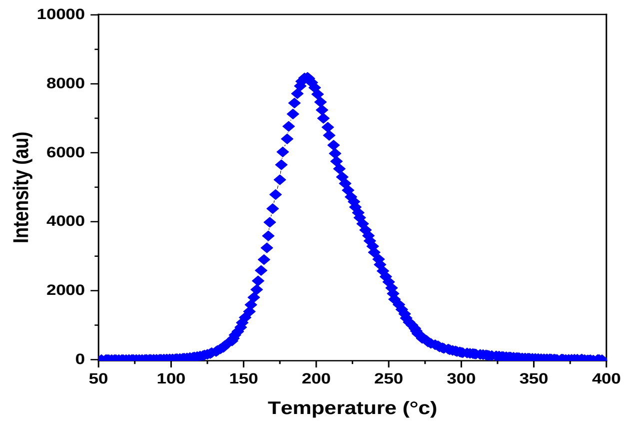

Fig. 2. Glow curve for (B2O3 – P2O5 – Li2O –Al2O3-Na2O-K2O doped with 0.7mol. % La2O3) sample.

3.3 TL-Kinetic Parameters calculations

The well-established TL method can be used to study defect characteristics. Research is ongoing to determine the trap parameters E, s, and b, in which, various methods have been employed to analyze the materials' glow curves.

3.3.1 Computed Glow Curve Deconvolution (CGCD)

TL glow curve data is subjected to CGCD to obtain clearly defined and separated peaks. One benefit of CGCD is that almost all parameters generate a theoretical curve from the experimental data. If a good deconvolution is achieved, as shown by its (FOM) value, values of the kinetic parameters can be modified. CGCD of glow curves of prepared sample previously irradiated at 10 Gy γ-doses were carried out by the PeakFit program (PFP) and represented in Figure 3. The deconvolution process resulted in four peaks from 1 to 4. Their position values and FWHM are explored in Table 1.

Using this program, the determination of E (eV) of each deconvoluted peak with different γ-doses as a function of Tm and FWHM was carried out using Eq. (2). The corresponding s (s-1) values were calculated using Eq. (8). Values of E (eV) and s (s-1) were listed in Table 1. The deconvolution of the TL signal revealed 4 different electron traps, with overlapping traps or even a quasi-continuous energy distribution from 0.788 up to 1.45 eV.

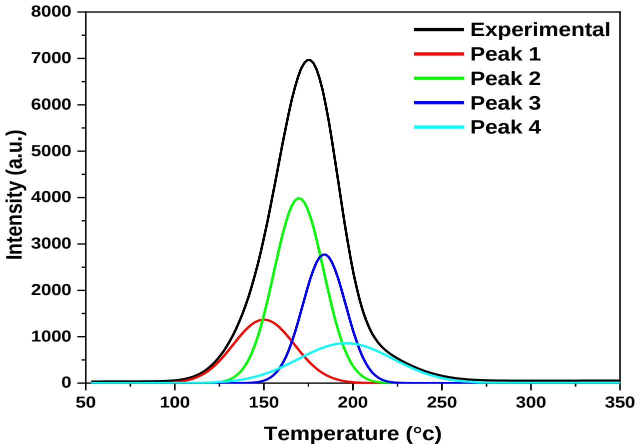

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Fig. 3. TL-Glow curve deconvoluted peaks (B2O3 – P2O5 – Li2O –Al2O3-Na2O-K2O doped with 0.7mol. % La2O3) sample irradiated at 10Gy.

Table 1. Deconvolution data for 80µm sample using PeakFit program.

Peak No.	Tm (ºk)	FWHM	E(eV)	S (s-1)
Peak 1	422.86	40.696	0.9114	$2.16*10^{10}$
Peak 2	442.63	33.11	1.1943	$1.4*10^{13}$
Peak 3	456.65	29.14	1.45	$4.07*10^{15}$
Peak 4	469.1	55.32	0.7881	$6.095*10^{7}$

4 Conclusions

This paper presents the thermoluminescence behavior of the doped boro-phosphate glasses with a La_2O_3 concentration of 0.7mol. %. The glass sample was prepared using a melt-quench method. The prepared glass sample was γ-irradiated with a dose of 5 Gy from γ-source to obtain their glow curves at a heating rate of 5 ºCs^{-1}. Results revealed that 0.7 mol. % of the La sample had very high sensitivity. The sample was characterized using XRD. Trap parameters were also calculated using the PeakFit program and revealed 4 different electron traps, with overlapping traps or even a quasi-continuous energy distribution from 0.788 up to 1.45 eV. The frequency factor (s) was estimated and found to be 4.07*1015 -6.095*107 s^{-1}.

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