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Types and Fabrication Methods of Borate-based Thermoluminescence Materials

Usha Shukla*

Abstract

Thermoluminescence (TL) is a kind of luminescence produced by certain crystalline materials that refers to the re-emission of energy received from electromagnetic radiation as light when the substance is heated. Thermoluminescence materials are used in a variety of fields, including environmental dosimetry, personal dosimetry, and medical research. Doping different rare earth impurities in different hosts causes the characteristics of materials suitable for diverse applications in many disciplines to change. Terbium-doped material has shown the highest TL sensitivity, with a wide dosimetry light peak at 240°C. The synthesized phosphors were tested for diametric features such as TL emission spectra, glow curves of thermoluminescence, fading studies, reproducibility, dose-response, and reusability. The method of preparing thermoluminescence diametric materials, as well as investigations and applications, are discussed.

Keywords: Thermoluminescence (TL), LMB (Lithium Magnesium Borate):TB^{3+}, SAB: CE, diametric features

INTRODUCTION

Thermoluminescence is a type of luminescence that occurs when certain crystalline materials are heated, and it refers to the re-emission of previously absorbed energy from electromagnetic radiation as light. After the stimulation is removed, thermoluminescence is temperature-stimulated light emission from a crystal. However, microscopically, this is far more complicated. Thermoluminescence is a ubiquitous and pervasive phenomenon defined as the emission of light at a specific temperature from samples subjected to electromagnetic radiation. Thermoluminescence can be seen in a variety of artificially created solid states, such as semiconductors and organic compounds. Thermoluminescence is commonly described in two stages. First, via energy absorption from the ultraviolet, the system is changed from an equilibrium to a meta-stable state. Second, the system can be returned to balance [1–3]. In 1602, alchemical texts discussed the thermoluminescence phenomenon. Sir Boyle spotted this for the first time in England in 1663. He heated the diamond in the dark and noticed that it was flowing. Thermoluminescence of minerals and other solid states was first studied scientifically in the first quarter of the twentieth century, and it has become a popular tool for studying energy storage in thermally stable states.

The process of thermoluminescence has been explored by many scholars in the seventeenth century, including Johann Sigismund Elsholtz, Robert Boyle, and Henry Oldenburg. In the eighteenth century, Dufay was the first researcher to examine thermoluminescence. He discovered that if a material is heated for a long time, it loses its thermoluminescence. Prominent scientists of thermoluminescence in the eighteenth century were De Saussure and Thomas Wedgwood. Heinrich discovered that when powdered materials were moderately heated, they could generate light. Theodor von Grotthus showed that

***Author for Correspondence**
Usha Shukla
E-mail: usha.shukla1@gmail.com

Assistant Professor, Department of Physics, Amity University, Lucknow, Uttar Pradesh, India

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thermoluminescence and eight essences are identical [4, 5]. Pearsall discovered a link between color and thermoluminescence. Thermoluminescence phenomena have also been exploited in prehistoric times [6].

When exposed to radiation, such as ultraviolet light or an electron beam, phosphor is a solid material that emits light or luminescence. Hundreds of thousands of phosphors have been produced, each with its distinct color of emission and duration of light emission after excitation has stopped. Electroluminescence occurs when certain phosphors glow as a result of electron excitation, and these phosphors are utilized to fabricate television screens and computer monitors. Phosphors activated by ultraviolet, visible, and infrared radiation are primarily used in fluorescent lamps, which are commonly used for illumination. UV phosphors absorb ultraviolet light and emit light in the visible spectrum. However, some materials may convert light to longer ultraviolet or infrared wavelengths. UV phosphors can be used for various purposes, including lighting, security marking, and product tagging for machine sorting. By converting the light from the current blue LEDs, blue LED phosphors can be used to create white LED lights. Phosphors can efficiently convert blue light to other colors because blue light has the highest energy of visible light [7].

Phosphor-emitted light was mixed with the remaining blue light to obtain the required white light. Because these materials absorb a large amount of blue light, they have bright colors in their bodies. IR emitters are commonly used in applications that require electronic phosphor detection. Security tagging and machine sorting of marked products are examples of these applications. Phosphorescence was observed in the glow-in-the-dark phosphors. Over time, the absorbed energy radiated. In contrast to fluorescence, energy is radiated instantly. An emergency signage that glows if the lights are turned off is one application. X-ray phosphors convert high-energy X-ray photons into visible light. These play a crucial role in the development of X-ray screens. Detecting X-rays using a standard photographic film is ineffective. The use of a phosphor layer to convert light has a significant impact on efficiency. Glow-in-the-dark phosphors work in a manner similar to storage phosphors. However, unless stimulated by infrared or ultraviolet beams, the material does not emit light. One such method is the detection of infrared light. These phosphors can detect a wider range of infrared light than the anti-Stokes phosphors. However, the phosphor must first store energy, and the activation of this energy depletes it. Over time, the strength of the resultant emissions decreases [8].

When ionizing radiation impacts an insulating crystal, it deposits some of its energy in the lattice at defect sites, color centers, and other spots. When the crystal is heated, part of the stored energy is released, and some of it may be emitted as visible light (before blackbody radiation begins at higher temperatures, i.e., > 250°C). This phenomenon is known as thermoluminescence. Within certain limits, the amount of light released was proportional to the radiation dose previously received by the TL material. When energy from an ionizing radiation source is absorbed by an insulating or semiconducting material, holes and free electrons are excited and nine of these electronic species are trapped in defects or meta-stable states. After a meta-stable trapped charge such as an electron absorbs thermal energy, the system is stimulated to return to its equilibrium state. The electronic charge of the 180-luminescence type phenomenon and their consequences for trapped holes recombine with the trapped hole during the relaxation phase, and the luminescence is generated if the recombination is radiative [9].

Borates have received much attention as hosts for light-emitting compounds when exposed to ultraviolet (UV) radiation. The explanation for their being at the focal point of examination is their assortment of construction types, straightforwardness to a wide range of frequencies, and high optical quality. Additionally, borates have phenomenal properties as hosts because of the innate traits of enormous band holes and covalent security energy. An assortment of borates with materials doped with uncommon earth elements and different particles has been considered and detailed [10–12]. The synthesis of borate compounds is difficult. Several borate compounds are available in both crystalline and glassy forms. Usually, crystalline forms are necessary to fabricate efficient luminous materials; however, glasses have been used in Photostimulated Luminescence (PSL), optical data storage, lasers,

and other applications. The use of boric acid as a boron source allows the manufacture of the constituents without melting them, which is necessary to avoid glass formation. Thus, longer reaction times are required. Non-stoichiometry can occur when boric acid evaporates. Recently, self-heating technologies, also known as combustion synthesis, have been used to produce oxide materials. The synthesis uses the heat generated by an exothermic chemical reaction. Some borate chemicals have also been used in this technique [13].

APPROACHES FOR PREPARING BORATE-BASED PHOSPHORS

Luminescent materials, such as glass and phosphors in microcrystalline and nanocrystalline forms, have been synthesized using a variety of processes.

Technique for Quenching Melts

Melt quenching, wet chemical processes, and drying techniques are examples of these methods. Melt quenching is the oldest method for fabricating glass, in which the molten material is rapidly cooled to inhibit crystal development. The starting components are fully combined in this manner and then melted at an appropriately high temperature in a furnace. The melt was then poured over a brass plate and pounded swiftly on another plate. Consequently, translucent glass was produced. The type of dopant added to the initial components determines the color of the glass. Another component that affects glass formation is the pace of melt quenching; the faster the rate of melt quenching, the better the glass formation.

Wet Chemical Method

The precursors were completely dissolved in the appropriate solvent during the wet chemical procedure, and the desired compound was dried using a gradual heating process. Coprecipitation is a wet chemical synthesis method. The elements were dissolved in a suitable solvent. The solution auto-precipitates, rather than generating a clear solution. In the liquid state, molecular movement and chemical reactions can happen very 11 quickly. In the right solvent, the acid-base reaction frequently produces a precipitate of the desired chemical. Lower reaction temperatures are attained in the self-heat generation method by acquiring reactants in fine form. The heat generated in the exothermic chemical reaction can be used for synthesis, which is a clever technique for lowering the operating temperature. The key benefit of this method is that it does not require external heating. Refractory muffles, insulators, and crucible materials were no longer required. The reactants were combined and squeezed to produce a bar. A flame was used to heat one end of the bar to a high temperature. The exothermic process spreads across the length of the bar once it begins. In such processes, temperatures as high as 7000°C have been used to develop TL materials.

Synthesis of Combustion

Combustion synthesis is a straightforward and cost-effective method to synthesize nanomaterials. It entails heating an aqueous solution of the appropriate metal salts to boiling until the combination ignites, resulting in a dry, usually crystalline, fine-particle oxide powder. This process produced extremely pure products. The combustion synthesis process is used to prepare lithium tetraborates doped with Cu+ and boron ions, such as $Li_2B_4O_7$:Cu:B. Solid-state reaction methods, such as tray drying at room temperature, direct heating, heating in a constant level water bath, heating in an oil bath, drying by blowing air at room temperature, drying by blowing hot air, spray drying, azeotropic distillation, and the freeze-drying process, are all used in the synthesis of phosphors.

FACTORS INFLUENCING THE DEMAND FOR THERMOLUMINESCENCE DOSIMETERS

A proper calibration must be used to develop the relationship between the signal and absorbed dosage to be evaluated in thermoluminescence dosimetry. With advancements in the production of solid thermoluminescent dosimeters and the instruments for reading them, thermoluminescent dosimeters have found growing applications. Currently, (Thermoluminescent Dosimeters) TLD-based systems are

widely utilized in routine personal dosimetry, environmental monitoring, and clinical radiation dosimetry. The high sensitivity of TL for detecting flaws as small as 107 μm within a specimen is advantageous for detecting low radiation levels encountered in personal and environmental monitoring. Thermoluminescent dosimeters are becoming more widely recognized for radiation dosimetry because of the existence of nearly tissue-comparable thermoluminescent materials. Its accuracy and sensitivity are sufficient for both personal and environmental monitoring. Commercially available compact solid detectors that can be processed manually or automatically are suitable for beta skin and extremity dosimetry. Materials with exceptional long-term stability under a variety of environmental circumstances are readily available, processing simplicity, and reusability; over a wide range of doses and dose rates, the response is linear [14].

Thermoluminescence-based radiation dosimetry has been studied for decades, primarily for personal, environmental, and therapeutic applications. Although numerous phosphors have been produced for dosimetric applications, much more research is needed to generate new low-cost materials with improved dosimetric capabilities. Because of the combined gamma and neutron ray responses, extremely cheap manufacturing costs, reduced synthesis temperatures, good thermal stabilities, reasonably high sensitivities, and ease of preparation, dosimetric studies of borate-based thermoluminescent materials are of interest. Zinc borate is a commonly used inorganic substance in a variety of applications, including flame retardancy, antibacterial activity, and as an additive to protect wood from insect/fungal attack [15].

TYPES OF THERMOLUMINESCENCE MATERIALS

Borate-based Glass

Owing to its advantages over other materials, bore glass was discovered to be a good candidate for scintillation and dosimetry experiments. Different amounts of Ce were used to prepare the cerium-doped strontium aluminum borate glass. Glow curves of thermally induced luminescence observations were obtained after the materials were subjected to 3.0×10^3 mGy X-rays. One notable peak at 90°C and two tiny peaks at 190°C and 270°C were observed for Ce concentrations of 0.01–0.1%. The 0.01 percent Ce concentration produced the maximum TL intensity. Furthermore, as the Ce concentration increases, the Thermally Stimulated Luminescence (TSL) intensity decreases. For X-ray doses ranging from 0.1 to 10^4 mGy, the samples displayed a linear TSL response. The TSL intensity significantly increased when the dose increased from 104 mGy. At approximately 466 K, both the 12 non-doped and rare earth-doped glass samples produced a single TL peak. For doped materials, there was a modest increase in the temperature at the climax, and an increase in TL intensity was observed. The maximum area covered by the glow curve was observed for Eu^{3+}-doped glass. Therefore, europium-doped cadmium borate glass has the potential to be used as a TLD material.

Phosphate-based Glass

The quenching melt method was used to make potassium aluminophosphate (KAP) glass. The TL response was the highest in KAP50 glass doped with 1.0 mol% MnO2. The TL emission curves of the non-doped KAP50, doped KAP30, and doped KAP50 glasses are displayed in Figure 1. The doped samples displayed two peaks in their TL emission curves, one at 150°C and the other at 364°C, whereas the non-doped KAP50 glass had only one peak at 348°C. The relative intensity of the high-temperature peak of the doped KAP50 glass was much higher than that of the non-doped glass. As a result, it was determined that Mn doping had a significant impact on the TL intensity.

Fluorophosphate-based Glass

Thomas et al. revealed the TL characteristics of K-Mg-Al-Zn fluorophosphate glasses. Two peaks were visible in the TL glow curve, one at 70°C and the other at 235°C. At the dose limit of 1–190 Gy, the TL response was hyperlinear. After 15 h, the primary peak decreased by up to 11%. According to the kinetic study, the activation energy of the higher temperature peak is 1.31 eV, whereas that of the lower temperature peak is 0.47 eV. Figure 2 shows the TL glow curves of K-Mg-Al-Zn fluorophosphate glass after different temperatures were applied to the synthesized sample. As the preheating temperature

increased, the TL peak migrated to higher temperatures, although the TL intensity steadily decreased. Figure 3 also shows the TL glow curves for various dosages in the ranges of (a) 1–10 Gy and (b) 40–190 Gy. It was discovered that as 13 the dose was increased, the intensity of both peaks increased, with the change being greater for the higher-temperature peak than for the lower-temperature peak.

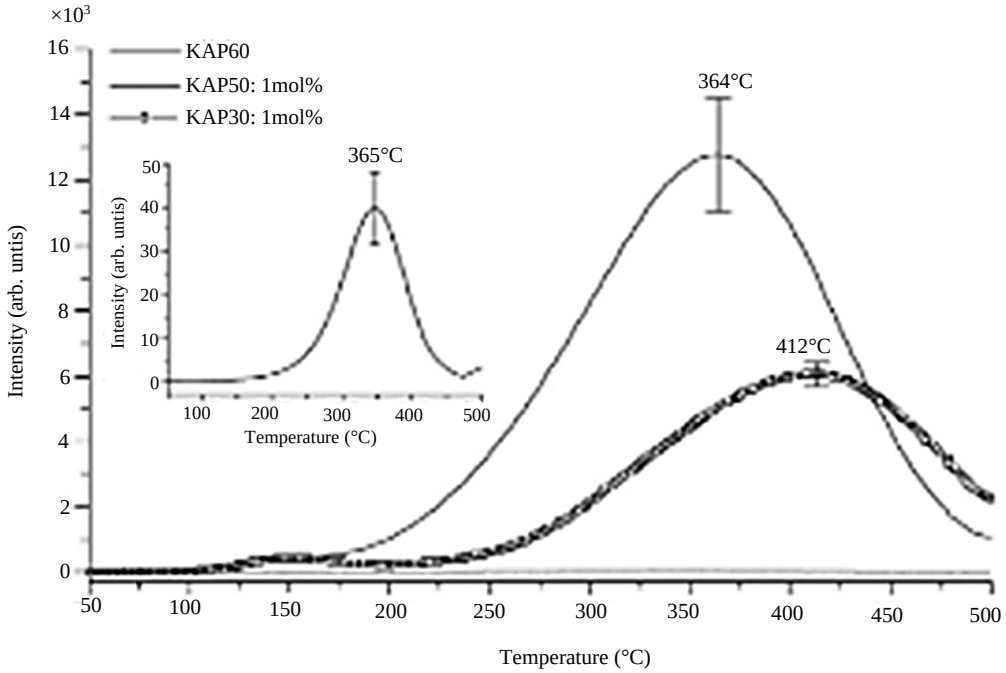

Figure 1. The non-doped KAP50 glass, as well as the doped KAP50 and KAP30 glass, have TL emission curves.

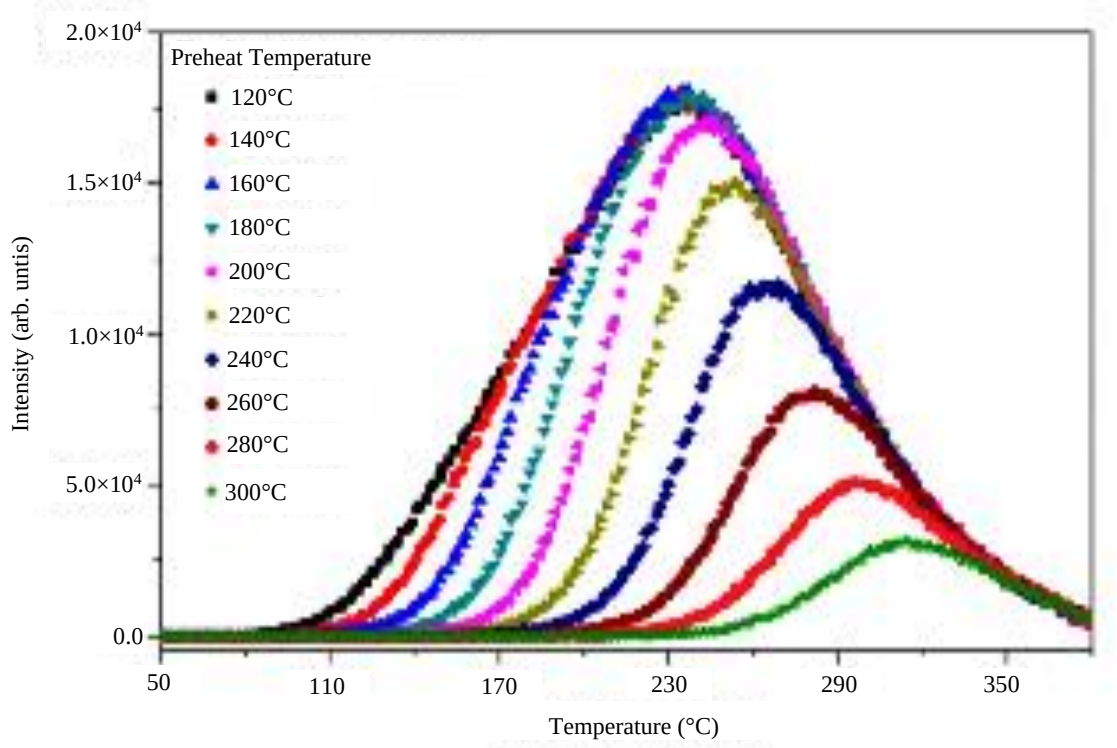

Figure 2. After preheating to different temperatures, the glow curves of K-Mg-Al-Zn fluorophosphate glass were compared.

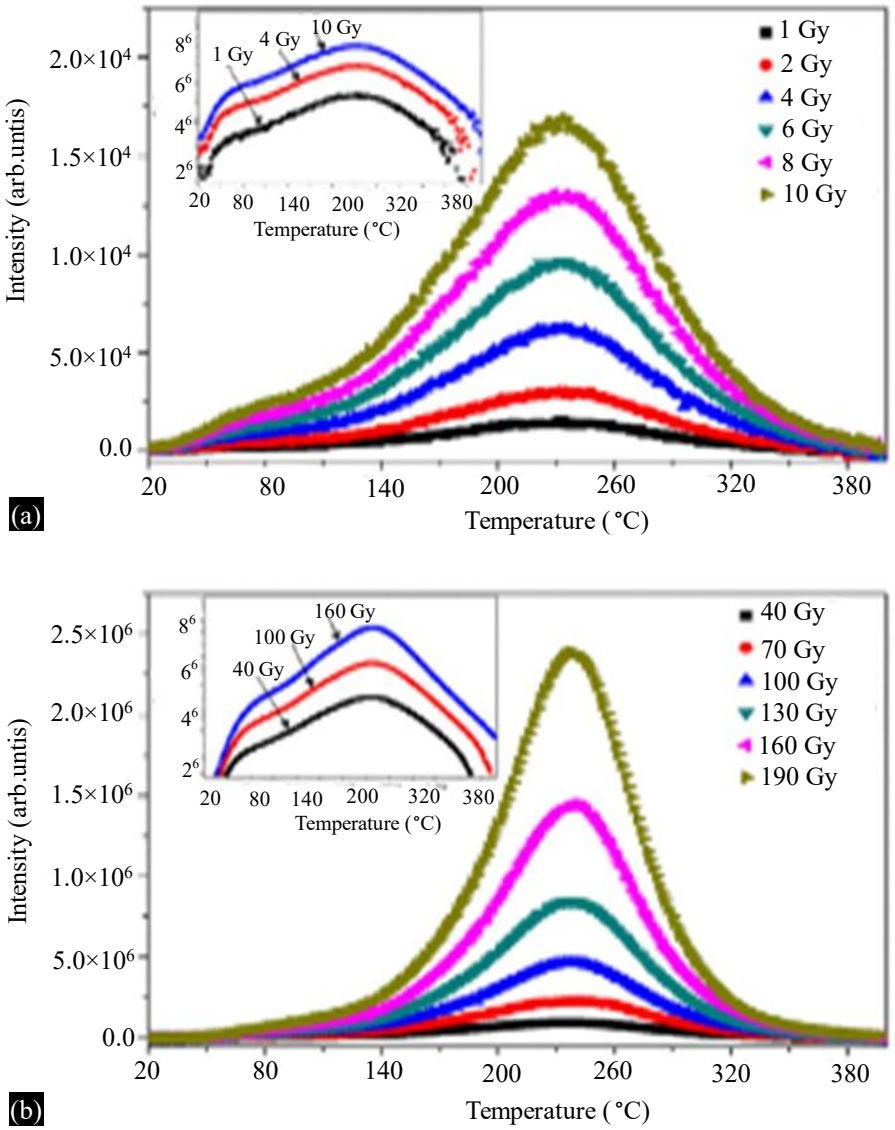

Figure 3. K-Mg-Al-Zn fluorophosphate glass TL glow curves obtained at 1°C/s corresponding to 1–10 Gy (a) and 40–190 Gy (b).

TLD Phosphors with High Sensitivity

Only a few very sensitive TLD phosphors have been produced using solid-state diffusion methods, such as $K_2Ca_2(SO_4)_3$:Eu, $Li_2B_4O_7$:Cu, $K_3Na(SO_4)_2$:Eu, MgB_4O_7:Dy, $KMgF_3$:Eu, and $Sr_2B_5O_9Cl$:Eu. The glow curve of $K_2Ca_2(SO_4)_3$:TL revealed a large 14 peak at 420 K, which was seven times more sensitive than that of $CaSO_4$: Dy, as well as a tiny peak at 434 K. Because of its simple light curve shape and linear TL response, $K_2Ca_2(SO_4)_3$:Eu, Ce phosphor is a highly sensitive phosphor for radiation dosimetry. The $K_3Na(SO_4)_2$: Eu phosphor was shown to be three times more sensitive than ordinary $CaSO_4$:Dy phosphors, with TL light maxima at 423 K and 475 K.

Two glow peaks were seen in synthesized nanoparticles, one at 166°C and the other at 210°C. Both peaks were identical to those seen in the microcrystalline $CaSO_4$: Dy nanoparticles, with the exception that the peak at 210°C was more prominent in the microcrystalline sample at low fluencies, whereas the peak at 166°C dominated in the $CaSO_4$: Dy nanoparticles at higher fluencies. The TL light curve of nanocrystalline LiF: Mg, Cu, and P revealed a strong peak at 410 K and four smaller overlapping peaks at 570, 609, 638, and 663 K.

The thermoluminescence of the Sr2B5O9Cl: Eu phosphor was studied using a solid-state diffusion approach, with a primary emission peak at 368 nm and a faint emission peak in the 410–430 nm range. The PL emission peak at 368 nm is typical of Eu^{2+} ions. The Sr2B5O9 Cl: Eu TL glow curve exhibits a strong peak at high temperatures at 495 K. The strength of this peak was approximately 70% of that of the $CaSO_4$: Dy dosimetry peak. In the blue portion of the spectrum, TL emission at 495 K was recorded at 409 nm. The 495 K sample exhibits very little fading. As a result, all the phosphors discussed have been found to be more sensitive than standard phosphors. At high temperatures, strong TL glow peaks were observed, making them useful for ionization radiation dosimetry. Furthermore, nanocrystalline phosphors perform better than microcrystalline phosphors at high doses [16].

CHARACTERIZATION AND XRD DATA ANALYSIS

Since 1967, researchers have studied the luminescence of borate compounds. Borate chemicals are mostly employed in radiation dosimeters and nonlinear optics, which use thermoluminescence to stimulate luminescence. Ionizing radiation monitoring using TL dosimeters has been widely used in clinical, personal, and environmental settings. A number of studies have investigated the TL properties of tissue-equivalent lithium and magnesium tetraborates doped with Mn, Cu, or rare earth metals. These borates have several undesirable properties in terms of radiation dosimetry, such as hygroscopicity and light-induced fading. Borates can also be used as birefringent crystals in the manufacture of optical 15 communication components. A boron atom can combine with three or four oxygen atoms to produce a compound. Powder X-ray diffraction patterns recorded in a STOE diffractometer employing Cu–Ka of wavelength 1.5406Å at room temperature were used to describe the crystalline growth of the materials. An automated TSL reader developed by Nucleonic India Ltd., India, model TSL Analyzer intended for powder samples, was used to record the g-ray-driven TL glow curves of the polycrystalline powder. Electrical heating in a Kanthal tray produces TL light curves of the samples at a linear heating rate of 10 1C/s.

The area under the dosimetric peak in the 150–340°C range was used to compute TL intensity. Two separate gamma sources were employed for mild and high doses of gamma irradiation. Isothermal heating was used to acquire the TL emission spectra of the gamma-irradiated materials. The TL emission spectra of the gamma-irradiated samples were acquired by isothermal heating of the samples in a TL reader heating tray at each glow peak temperature. The light emitted from the sample was collected using a fiber optical cable and transferred to the spectrofluorometer emission monochromator to record the spectrum, with the xenon lamp turned off. Borate radicals with two or four boron atoms were produced in lithium magnesium borate samples using various quantities of boric acid. The starting chemical, which supplies lithium, magnesium, and boron atoms in the final result, is preserved in a 2:2 ratio for the synthesis of these materials. Figure 4 shows the X-ray diffraction (XRD) patterns of the various materials. As a result, even if the starting recipe contains varying amounts of boric acid, the borate radicals generated in the (lithium magnesium borate) LMB phosphor may be the same. The thermoluminescence glow curves of gamma-irradiated LMB phosphors with and without rare-earth element doping are shown in Figure 5. These are the results of a 21.4 Gy dosage of 60Co gamma rays at ambient temperature. Two or more peaks were observed in every glow curve. The TL peaks in the undoped sample were observed at 115, 235, and 290°C. Tb^{3+} doping, Gd^{3+} doping, Dy^{3+} doping, Mn^{2+} doping, Pr^{3+} doping, and Ce^{3+} doping in LMB phosphors have increased the strength of the prominent dosimetric peak visible around 220°C. Table 1 lists the relative TL sensitivity of the dosimetric peaks detected in the LMB for various dopants.

TL DOSAGE VS. INTENSITY

One of the most significant characteristics of TL dosimeter materials is their flexibility. The TL intensity and absorbed dose exhibited a linear relationship. The TL intensity of many TL materials grows nonlinearly with the time-absorbed dosage in a specific dose range. Connection relationship Tb_3 doped LMB TL intensity and gamma dose. Figure 6 shows the phosphor. To keep track of the dose-response relationship, the phosphor was bombarded with various gamma levels at the glow curve to provide room temperature and TL intensity.

Table 1. Temperature of additional glow peaks and relative TL sensitivity of dosimetric peaks for different LMB phosphors normalized to LMB:Tb^{3+}.

TL phosphor material	Dosimetric peak temperature (°C)	Relative TL sensitivity	Temperature of other glow peaks (°C)
LMB:Undoped	230	0.3	115, 218
LMB:Tb	240	1.0	-
LMB:Gd	235	0.8	117,338
LMB:Dy	350	0.8	178
LMB:Pr	260	0.5	290
LMB:Mn	212	0.6	120, 280
LMB:Ce	290	0.3	124

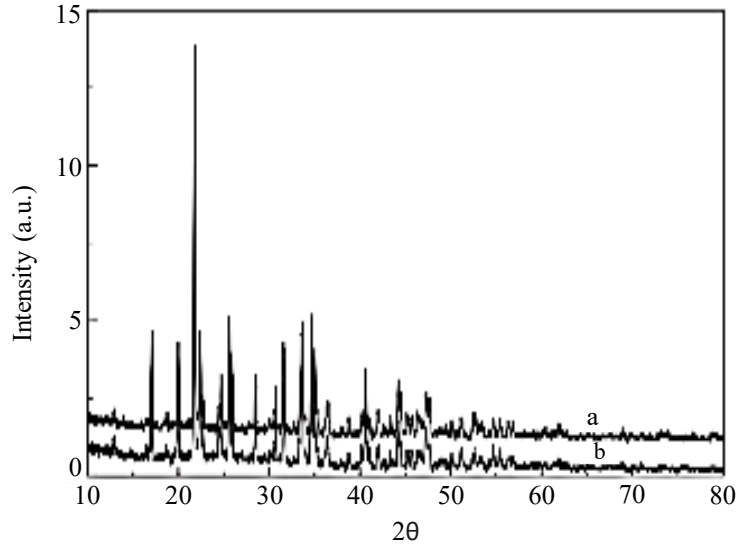

Figure 4. LMB: Tb^{3+} phosphor X-ray diffraction patterns generated with various mole concentrations of boric acid.

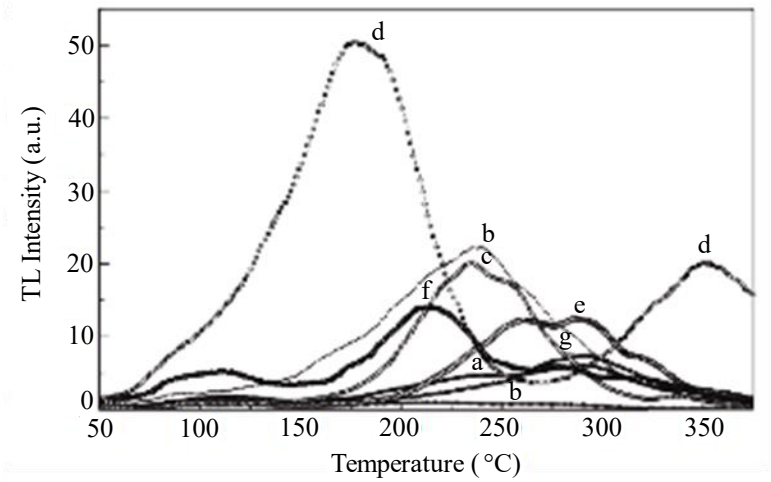

Figure 5. LMB TL lighting curves with various dopants.

Two sources of irradiation were employed at various dosage rates and gamma energies. From the low dose in mGy to the saturation dose at 103 Gy, the 240°C peak exhibited a nearly linear response. Compared to other tissue-analogous TL phosphors such as LiF and Li/Mg borates, LMB:Tb^{3+} has greater values in terms of dose linearity and saturation dosage. Figure 7 shows the glow curves of LMB:

Tb^{3+} powder exposed to various gamma dosages. These glow curves illustrate the variations in the shape and peak temperature as a function of the absorbed dosage.

The TL intensities were not on the same scale for comparison. This material exhibited no severe glow-curve aberrations or peak shifts. The glow peak, which was near 230°C at low doses (235°C at 0.5 Gy), shifted upward as exposure increased, reaching 250°C at 26 Gy. The glow peak changes southward as the dose increases above 26 Gy, reaching 210°C at 6 kGy. The glow curves show that the peak shift was not accompanied by a significant change in the glow curve shape, with the exception of a minor increase in the low-temperature shoulder peak. The complete width at half maximum (ω), low-temperature half-width (δ1), high-temperature half-width (δ2), and symmetry factor (μ) of the glow peak, which is determined by $\mu = \delta 2/ \omega$, have all been used to examine the variance in the peak form. Table 2 lists the glow peak properties of the phosphor with increasing exposure.

The peak width ω varies by approximately 20°C between different exposures, with the lowest value in the 10^2–10^3 Gy exposure range and greater values at lower and higher doses. The increase in o is attributed to a substantial increase in δ1 up to a maximum of 24°C, whereas δ2 exhibits a progressive reduction with increasing dose. At dosages below 10 Gy and above 103 Gy, the expansion of the low-temperature shoulder peak caused greater values of ω and δ2.

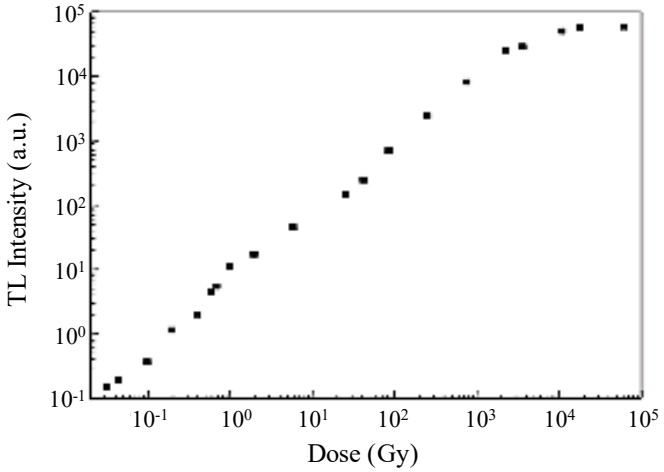

Figure 6. The dosimetry light peaks at 240°C with integral TL.

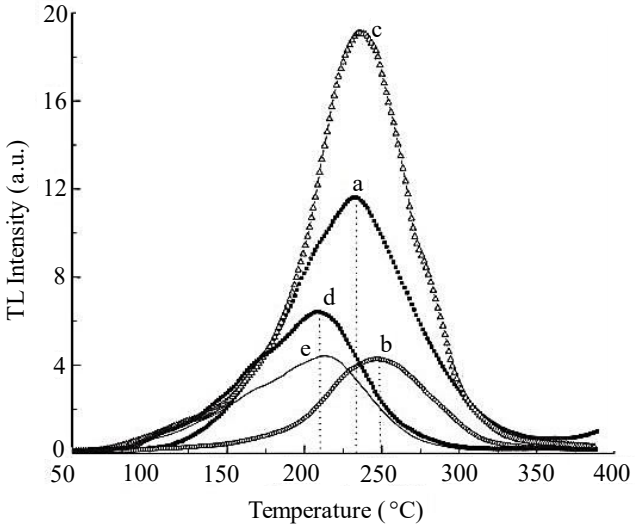

Figure 7. LMB: Tb^{3+} TL glow curve at various doses.

Table 2. With increasing gamma dose, TL glow peak characteristics of LMB: Tb^{3+} phosphor.

Absorbed dose (Gy)	Tmax (°C)	T1 (°C)	T2 (°C)	δ1	δ2	FWHM (ω)	μ = δ2/ ω
0.5	233	184	275	49	42	91	0.47
26	247	210	289	37	42	79	0.52
770	235	210	273	34	38	72	0.53
11, 175	208	155	243	53	35	88	0.40
63, 325	213	155	245	58	32	90	0.35

CONCLUSION

The main objective of this study is to investigate the thermoluminescence of borate-based phosphors. In this review, the properties and characteristics of borate-based phosphors are discussed. A simple solid-state diffusion approach was used to prepare lithium magnesium borate phosphors doped with various rare-earth elements. Among the RE dopants, LMB: Tb^{3+} displays a very stable dosimetrist peak at 240°C according to the TL glow curves. The XRD patterns of the phosphors revealed a crystalline structure. A detailed investigation based on XRD is currently underway to identify the crystal structure and the exact chemical formula. Various TLD materials, including glass, microcrystalline, and nanocrystalline phosphors, have been examined. In comparison with existing materials, studies on TL dosimetry materials have primarily focused on the creation of more efficient phosphors. Glow-curve simplicity, a wide range of TL response linearity, and little fading should all be priorities in the quest for novel TL materials. Furthermore, while developing innovative high-performance materials for various applications, the TL mechanism is crucial and must be considered. Scientists hope to develop technologies that are both dependable and efficient. Research is still on developing new gadgets and improving existing ones to fulfill future demand and bring technology to the masses for everyday usage.

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