

Types and Fabrication Methods of Borate Based Thermoluminescence Materials

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Abstract

Thermoluminescence (TL) is a kind of luminescence produced by certain crystalline materials that refers to the re-emission of energy received from electromagnetic radiations 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: TB3+, SAB: CE, Diametric features

INTRODUCTION

Thermo-luminescence is a kind of luminescence that happens when certain crystalline materials are heated, and it refers to the re-emission of previously absorbed energy from electromagnetic radiations as light. After the stimulation is removed, thermoluminescence is a temperature-stimulated light emission from a crystal. Microscopically, though, it is far more complicated. Thermo-luminescence is a ubiquitous and pervasive phenomenon defined as the emission of light at a specific temperature from samples that have been subjected to electromagnetic radiations. Thermo-luminescence can be seen in a variety of artificially created solid states, such as semiconductors and organic compounds. Thermo-luminescence is commonly described in two stages: first, via energy absorption from the Ultraviolet, the system is changed from equilibrium to meta-stable state. Second, by releasing energy, the system can be brought back to balance [1-3]. In 1602 alchemical texts discussed the phenomenon of thermoluminescence. Sir Boyle spotted it for the first time in England in 1663. He heated a diamond in the dark and noticed that it was glowing. Thermo-luminescence of minerals and other solid states was first studied scientifically in the first quarter of the twentieth century, and it became a popular tool for studying energy storage in thermally stable states.

The process of thermoluminescence was 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 to examine thermoluminescence. He discovered that if the material is heated for a long time, it loses its thermoluminescence feature. The prominent scientists of thermoluminescence in the eighteenth century were De Saussure and Thomas Wedgwood. Heinrich discovered that when powdered materials are moderately heated, they can generate light. Theodar von Grotthus, a researcher, shown that

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thermoluminescence and its essence are identical [4-5]. Pearsall discovered a link between colour and thermoluminescence. The thermoluminescence phenomena was also exploited in prehistoric [6].

When exposed to radiation such as ultraviolet light or an electron beam, phosphor is a solid material that emits light, or luminesces. Hundreds of thousands of phosphors have been produced, each with its own distinct colour 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 utilised to make television screens and computer monitors. Phosphors activated by ultraviolet, visible, and infrared radiation are primarily used in fluorescent lamps, which are commonly used for general illumination. UV phosphors absorb ultraviolet light and often emit light in the visible spectrum. However, some materials may convert light to a longer wavelength of ultraviolet or even infrared. UV phosphors can be used for a variety of purposes, including lighting, security marking, and product tagging for machine sorting. By converting the light from current blue LEDs, blue LED phosphors are used to create white LED lights. Phosphors can efficiently convert some blue light to other colours because blue light has the highest energy of visible light [7].

The phosphor-emitted light is mixed with the remaining blue light to obtain the required white. Because these materials must absorb a lot of blue light, they will have bright colours in their bodies. IR emitters are most commonly used in applications that require electronic phosphor detection. Security tagging and machine sorting of marked products are examples of applications. Phosphorescence can be seen in glow-in-the-dark phosphors. Over time, the absorbed energy radiates away. In contrast to fluorescence, when the energy is radiated practically instantly. Emergency signage that glows if the lights go off is one application. X-ray phosphors convert x-ray photons with high energy into visible light. They played a crucial role in the development of x-ray screens. Detecting x-rays with 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 similarly to storage phosphors. However, unless stimulated by an infrared or ultraviolet beam, the material does not emit light. The detection of infrared light is one use. These phosphors can detect a wider range of infrared light than Anti-Stokes phosphors. However, the phosphor must first store energy, and activation of this energy depletes it. Over time, the strength of the resultant emission declines [8].

When ionising radiation impacts an insulating crystal, it deposits some of its energy in the lattice at defect sites, colour centres, 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 black-body radiation begins at higher temperatures, i.e. $> 250^{\circ}\text{C}$). This is known as thermoluminescence. Within certain limits, the amount of light released is proportional to the radiation dose previously received by the TL material. When energy from an ionising radiation source is absorbed by an insulating or semiconducting material, holes and free electrons are excited, and these electronic species are trapped in defects or metastable states. After a metastable trapped charge, like 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 trapped hole recombines with the trapped hole during the relaxation phase, and luminescence is generated if the recombination is radiative [9].

Borates have gotten a lot of attention as a host for light-emitting compounds when exposed to UV radiation. The explanation for their being in a focal point of examination is their assortment of construction types, straightforwardness to a wide scope of frequencies, and high optical quality. Additionally, borates have phenomenal properties as host because of the innate traits of the enormous band hole and covalent security energy. An assortment of borate having materials doped with uncommon earth and different particles have 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 make efficient luminous materials, however glasses have been used in PSL, optical data storage, lasers, and other applications. The use of boric acid as a

boron source allows for the manufacture of the constituents without melting them, which is necessary to avoid glass formation. 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 create oxide materials. The synthesis uses the heat created by the exothermic chemical reaction. Some borate chemicals have also been added to the 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 methods. Melt quenching is the oldest method for making glass, in which the molten material is rapidly cooled to inhibit crystal development. Starting components are fully combined in this manner, and then melted at the appropriate high temperature in a furnace. After that, the melt is poured over a brass plate and swiftly pounded with another plate. As a result, translucent glass is produced. The type of dopant added to the beginning components determines the colour of the glass. Another component that impacts 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 are completely dissolved in the appropriate solvent in the wet chemical procedure, and the desired compound is then dried using a gradual heating process. Coprecipitation is a type of wet chemical synthesis. The elements are prepared to dissolve in a suitable solvent in this process. The solution auto-precipitates rather than generating a clear solution. In the liquid state, molecular movements and thus chemical reactions can happen very 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 the reactants in fine form. The heat created in the exothermic chemical reaction itself can be used for synthesis, which is a clever technique of lowering the operating temperature. The key benefit of this method is that it does not require external heat. Refractory muffles, insulators, and crucible materials are no longer required. The reactants are thoroughly combined and squeezed to produce a bar in this procedure. A flame is 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 achieved to develop TL materials.

Synthesis of Combustion

Combustion synthesis is a straightforward and cost-effective way to make 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 produces extremely pure products. The combustion synthesis process is used to make lithium tetraborates doped with Cu⁺ and boron ions, such as Li₂B₄O₇: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, or the freeze drying process, are all used in the synthesis of phosphor.

FACTORS INFLUENCING THE DEMAND FOR THERMOLUMINESCENCE DOSIMETERS

A proper calibration must be used to develop the relationship between the signal and the absorbed dosage to be evaluated in thermoluminescence dosimetry. With advancements in the production of solid thermoluminescent dosimeters and instruments for reading them, thermoluminescent dosimeters have found growing application. TLD-based systems are currently widely utilized in routine personal dosimetry, environmental monitoring, and clinical radiation dosimetry. The great sensitivity of TL for

detecting flaws as small as 107 within a specimen is advantageous for detecting low radiation levels encountered in personal and environmental monitoring. Because of the existence of almost tissue comparable thermoluminescent materials, thermoluminescent dosimeters are becoming more widely recognized for radiation dosimetry. Accuracy and Sensitivity are sufficient for both personal and environmental monitoring. Commercial availability as compact solid detectors that may be processed manually or automatically; suitability for beta skin and extremities dosimetry. Materials having exceptional long-term stability under a variety of environmental circumstances are readily available; Processing simplicity; 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 in the realms of personal, environmental, and therapeutic applications. Though numerous phosphors have been produced for dosimetric applications, much more research is needed to generate new low-cost materials with improved dosimetrist capabilities. Because of the combined gamma and neutron ray responses, extremely cheap manufacturing cost, reduced synthesis temperature, good thermal stability, reasonably high sensitivity, and ease of preparation, dosimetric studies of borate-based thermoluminescence materials are of interest. Zinc borate is a commonly used inorganic substance in a variety of applications, including flame retardant, antibacterial, and as an addition to preserve wood from insect/fungal attack [15].

TYPES OF THERMOLUMINESCENCE MATERIALS

Borate-based Glass

Due to its benefits over other materials, bore glass was discovered to be a good candidate for scintillation and dosimetry experiments. Different amounts of Ce were used to make cerium doped strontium aluminum borate glass. Glow curve of thermally induced luminescence observations were done after the materials were subjected to 3.0×10^3 mGy Xrays. One notable peak at 90°C and two tiny peaks at 190°C and 270°C, respectively, were seen for Ce concentrations of 0.01–0.1%. The 0.01 percent Ce concentration produced the maximum TL intensity. Furthermore, as Ce concentration increased, TSL intensity dropped. For Xray doses ranging from 0.1 to 10^4 mGy, these samples displayed a linear TSL response. TSL intensity was significantly raised when the dose was increased from 104 mGy. At roughly 466 K, both 12 non-doped and rare earth doped glass samples produced a single TL peak. For doped materials, there is simply a modest increase in temperature at the climax. and a rise in TL intensity was observed. The maximum area covered by the glow curve was found in Eu³⁺ doped glass. As a result, 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 alumino phosphate (KAP) glass. The TL response was highest in KAP50 glass doped with 1.0 mol% MnO₂. The TL emission curves of nondoped KAP50, doped KAP30, and doped KAP50 glass are displayed in Figure 1. The doped samples displayed two peaks on their TL emission curves, one at 150°C and the other at 364°C, but the non-doped KAP50 glass had only one peak at 348°C. The relative intensity of the doped KAP50 glass's high temperature peak was way higher than that of the nondoped glass's peak. As a result, it was determined that Mn doping has a great impact on TL intensity.

Fluorophosphate Based Glass

Thomas and coworkers revealed the TL characteristics of K-Mg-Al-Zn fluorophosphate glass. Two peaks were visible on the TL glow curve, one at 70°C and the other at 235°C. In the dose limit of 1–190 Gy, the TL response was hyper linear. In 15 hours, the primary peak had faded by up to 11%. According to 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. With increasing preheat temperature, the TL peak migrated to higher temperatures, although TL intensity

steadily dropped. Figure 3 also shows the TL glow curves for various dosages in the ranges of (a) 1–10 Gy or (b) 40–190 Gy. It was discovered that as the dose was raised, the intensity of both peaks rose, with the change being greater for the higher temperature peak than for the lower temperature peak.

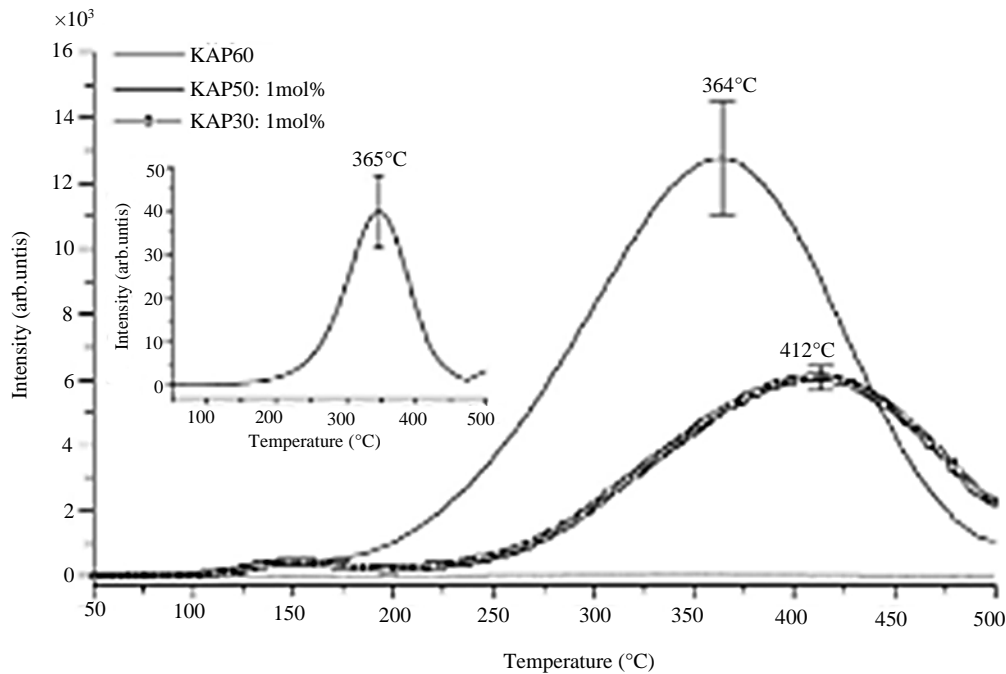


Figure 1. The non-doped KAP50 glass, as well as the doped KAP50 and KAP30 glass, have TL emission curves.

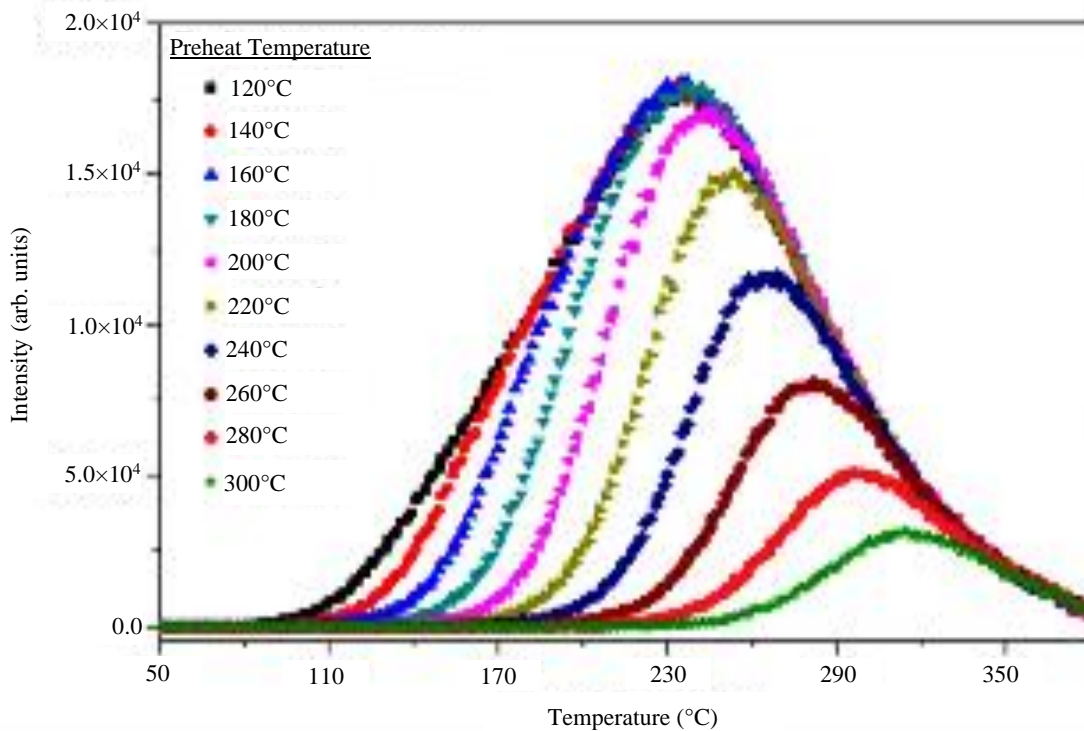


Figure 2. After preheating to different temperatures, the glow curves of KMgAlZn fluorophosphates glass were compared.

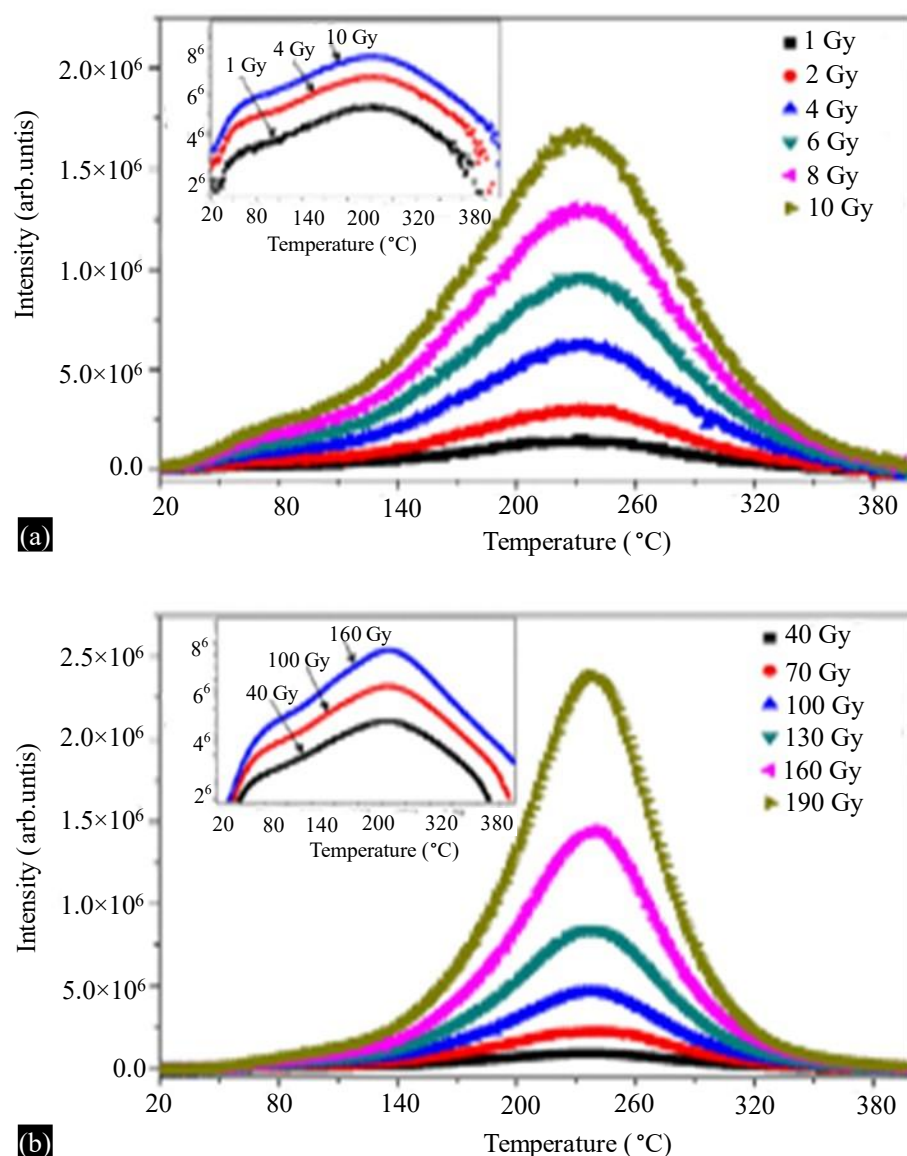


Figure 3. KMgAlZn fluorophosphate glass TL glow curves obtained at 1°C/s corresponding to 1–10 Gy (a) and 40–190 Gy (b).

TLD Phosphors with High Sensitivity

Only a few very sensitive TLD phosphors have been produced utilizing solid-state diffusion methods, such as $\text{K}_2\text{Ca}_2(\text{SO}_4)_3:\text{Eu}$, $\text{Li}_2\text{B}_4\text{O}_7:\text{Cu}$, $\text{K}_3\text{Na}(\text{SO}_4)_2:\text{Eu}$, $\text{MgB}_4\text{O}_7:\text{Dy}$, $\text{KMgF}_3:\text{Eu}$, $\text{Sr}_2\text{B}_5\text{O}_9\text{Cl}:\text{Eu}$, and others. $\text{K}_2\text{Ca}_2(\text{SO}_4)_3:\text{TL Eu}$'s glow curve revealed a large 14 peak at 420 K, which was seven times more sensitive than $\text{CaSO}_4:\text{Dy}$, as well as a tiny peak at 434 K. As a consequence of its simple light curve shape and linear TL response, the $\text{K}_2\text{Ca}_2(\text{SO}_4)_3:\text{Eu}$, Ce phosphor is a highly sensitive phosphor for radiation dosimetry. The $\text{K}_3\text{Na}(\text{SO}_4)_2:\text{Eu}$ phosphor was shown to be three times more sensitive than ordinary $\text{CaSO}_4:\text{Dy}$ 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 microcrystalline $\text{CaSO}_4:\text{Dy}$ 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 $\text{CaSO}_4:\text{Dy}$ nanoparticles at higher fluencies. The TL light curve of nanocrystalline $\text{LiF}:\text{Mg}$, Cu, P revealed a strong peak at 410 K, as well as 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 the primary emission peak at 368 nm and a faint emission peak in the 410–430 nm range. The peak of the PL emission at 368 nm was typical of Eu²⁺ ions. The Sr₂B₅O₉Cl: Eu TL glow curve revealed a strong peak at high temperatures at 495 K. The strength of this peak was roughly 70% that of the CaSO₄: Dy dosimetry peak. In the blue portion of the spectrum, TL emission of 495 K was recorded at 409 nm. The 495 K exhibited very little fading. As a result, all of the phosphors discussed have been found to be more sensitive than standard phosphors. At high temperatures, strong TL glow peaks were found, which makes them useful for ionization radiation dosimetry. Furthermore, nanocrystalline phosphors performed better than microcrystalline phosphors at high doses [16]

CHARACTERIZATION AND XRD DATA ANALYSIS

Since 1967, researchers have been studying the luminescence of borate compounds. Borate chemicals are mostly employed in radiation dosimeters and nonlinear optics that use thermoluminescence to stimulate luminescence. Ionizing radiation monitoring with TL dosimeters has been widely used in clinical, personal, and environmental settings. A number of researches have looked at the TL properties of tissue equivalent lithium and magnesium tetra borates doped with Mn, Cu, or rare earth metals. These borates have several unwanted properties in terms of radiation dosimetry, such as hygroscopicity, light induced fading, and so on. Borates could also be used as a birefringent crystal in the manufacture of optical communication components. The boron atom can combine with three or four oxygen atoms to produce a compound. X-ray diffraction patterns in powder recorded in a STOE diffractometer employing Cu-K α of wavelength 1.5406 angstrom at room temperature are used to describe the materials' crystalline growth. An automated TSL reader developed by Nucleonic India Ltd., India, model TSL Analyzer intended for powder samples is 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.

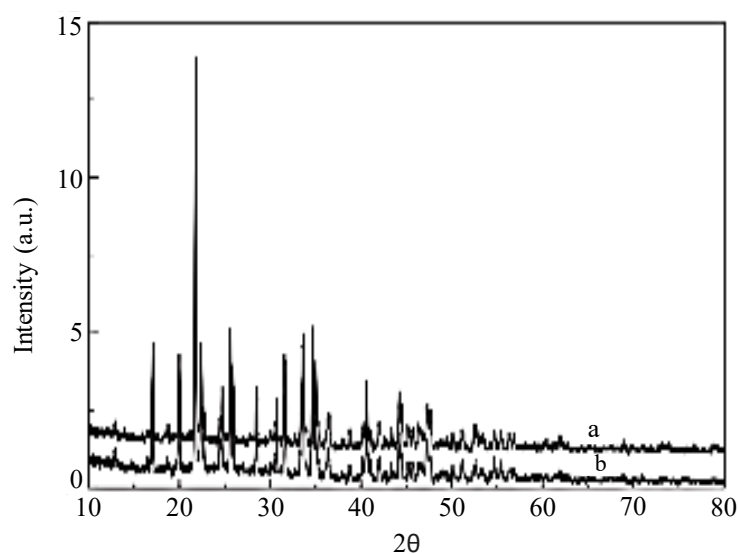
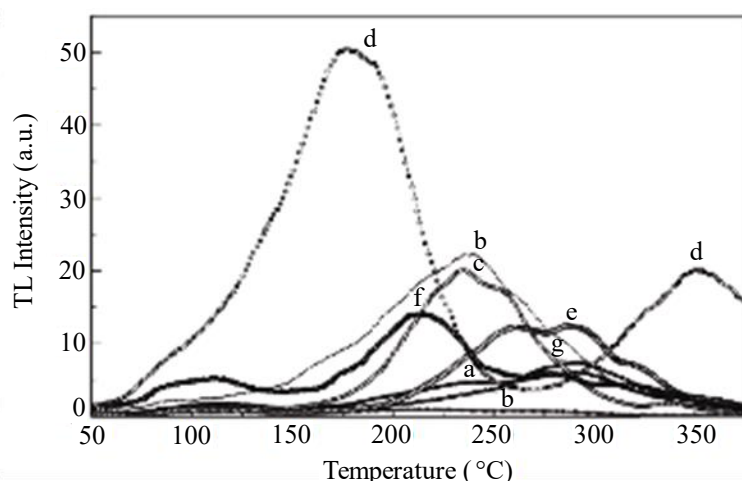
The area under dosimetric peak in the 150–340°C range was used to compute TL intensity. Two separate gamma sources are employed for mild and high doses of gamma irradiation. Isothermal heating is used to acquire the TL emission spectra of gamma irradiation materials. The TL emission spectra of the gamma irradiation samples are acquired by isothermal heating the samples in a TL reader heating tray at each glow peak temperature. The light emitted from the sample is collected using a fiber optical cable and transferred to the spectrofluorometer's emission monochromator to record the spectrum, with the xenon lamp turned off. Borate radicals with two or four boron atoms are produced in lithium magnesium borate samples using various quantities of boric acid. The starting chemical, which supplies the 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 XRD patterns of various materials. As a result, even if the starting recipe contains varying amounts of boric acid, the borate radical generated in (lithium magnesium borate) LMB phosphor may be the same. Thermoluminescence glow curves of gamma irradiation 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 given at ambient temperature. There are two or more peaks in every glow curve. The TL peaks in the undoped sample are at 115, 235, and 290°C. Tb³⁺ doping, Gd³⁺ doping, Dy³⁺ doping, Mn²⁺ doping, Pr³⁺ doping, or Ce³⁺ doping in LMB phosphor has increased the strength of the prominent dosimetric peak visible around 220°C. Table 1 shows the relative TL sensitivity of the dosimetric peaks detected in LMB for various dopants.

TL DOSAGE VS. INTENSITY

One of the most significant characteristics of a TL dosimeter material is that it is flexible. The TL intensity and the absorbed dose has a linear relationship. The TL intensity of many TL materials grows nonlinearly with time absorbed dosage in a specific dose range. The connection relationship Tb³⁺ doped LMB TL intensity and gamma dose. Figure 6 shows the phosphor. To keep track of the dose–response relationship phosphor has been bombarded with various gamma levels at the glow curve provides the 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: Tb³⁺.

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: Tb³⁺ 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 are employed for irradiation of various dosage rates and gamma energies. From the low dose in mGy to the saturation dose at 103 Gy, the 240°C peak exhibits a nearly linear response. When compared to other tissue analogous TL phosphors such as LiF and Li/ Mg borates, LMB:Tb³⁺ material has greater values in terms of dose linearity and saturation dosage. Figure 7 shows

the glow curves of LMB: Tb³⁺ powder exposed to various gamma dosages. These glow curves are offered to illustrate the variations in shape and peak temperature as a function of absorbed dosage.

TL intensities are not in the same scale for comparison. This material has 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 upwards as exposure increased, reaching 250°C at 26 Gy. The glow peak changes southward as the dose is increased above 26 Gy, reaching 210°C at 6 kGy. The glow curves show that the peak shift is not accompanied by a significant change in the glow curve shape, with the exception of a minor rise in the low temperature shoulder peak. The complete width at half maximum (ω), low temperature half-width (δ_1), high temperature half-width (δ_2), and the symmetry factor (μ) of the glow peak, which is determined by $\mu = \delta_2 / \omega$, have all been used to examine the variance in peak form. Table 2 shows the glow peak properties of the phosphor as exposure increases.

Peak width ω has varied by roughly 20°C between different exposures, with the lowest value in the 10²–10³ Gy exposure range and greater values at lower and higher doses. The increase in ω is attributed to a substantial increase in δ_1 up to a maximum of 24°C, whilst δ_2 exhibits a progressive reduction with increasing dose. At dosages below 10 Gy and beyond 103 Gy, the expansion of the low temperature shoulder peak causes the greater values of ω and δ_2 .

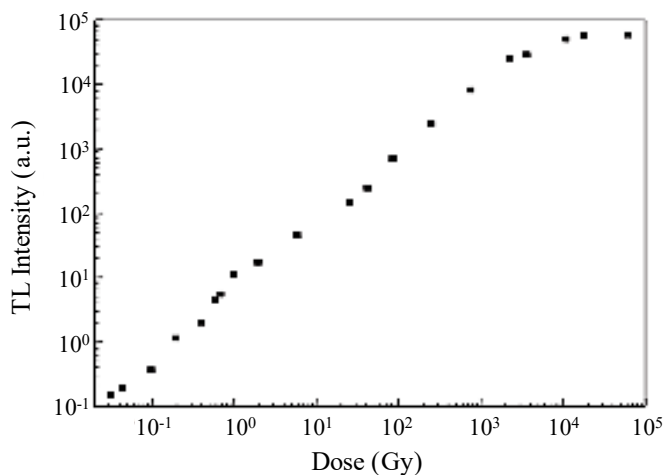


Figure 6. The dosimetry light peaks at 240°C with integral TL.

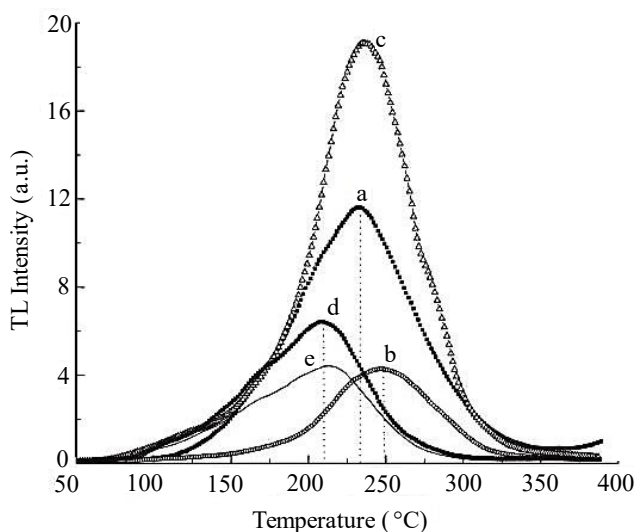


Figure 7. LMB: Tb³⁺ TL glow curve at various doses.

Table 2. With increasing gamma dose, TL glow peak characteristics of LMB: Tb³⁺ phosphor.

Absorbed dose (Gy)	T _{max} (°C)	T ₁ (°C)	T ₂ (°C)	δ ₁	δ ₂	FWHM (ω)	μ = δ ₂ / 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 report was to study about Thermoluminescence of borate-based phosphors. In this review report, mainly the properties and characteristics of borate-based phosphors has been discussed. The simple solid state diffusion approach was used to make lithium magnesium borate phosphor doped with various rare earth dopants. The best among the RE dopants, LMB: Tb³⁺, displays a very stable dosimetrist peak at 240°C, according to TL glow curves. The phosphors' XRD diffraction pattern revealed a crystalline structure. For the identification of crystal structure and exact chemical formula, a detailed investigation based on X-ray diffraction is now underway. A variety of TLD materials, including glass, microcrystalline, and nanocrystalline phosphors, have been examined. In comparison to existing materials, studies on TL dosimetry materials were found to be 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 a variety of applications, the TL mechanism is crucial and must be considered. Scientists hope to create technologies that are both dependable and efficient. Research is still on to develop new gadgets and improve existing ones in order to fulfil future demand and bring technology to the masses for everyday usage.

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