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Enhanced Linear Response in Radiation Monitoring Using Mn-Doped Magnesium Borate Nanostructured TLD Phosphors

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Abstract: ***Accurate radiation monitoring is imperative for worker safety in ionizing radiation environments. This study presents a novel nanomaterial phosphor, magnesium borate doped with Mn impurity, synthesized via a chemical approach. Structural and morphological analyses were conducted using X-ray diffraction and field emission scanning electron microscopy (FESEM). Thermoluminescence analysis revealed the phosphor's superior linear response to absorbed doses of gamma rays ranging from 50 Gy to 3 kGy, surpassing traditional phosphors. The enhanced performance is attributed to its nanostructure, offering a broader linear dose response range compared to bulk materials. This research addresses limitations in existing thermoluminescent phosphors, showcasing the potential of nanotechnology in radiation monitoring materials. It builds upon a century of valuable research in this field, highlighting ongoing global efforts to advance radiation safety through innovative materials and techniques.***

Keywords: Ionizing radiation, dose response, gamma rays, magnesium borate

1. Introduction

Ionizing radiation has harmful effects on all living organisms, including humans, animals, and plants. In humans and animals, these effects can persist for decades and cause serious damage, including to the DNA mutations. Therefore, precise monitoring of absorbed radiation is essential for workers in research centers, medical facilities, and industries to prevent injuries. In recent times, the world has witnessed the threat of bioweapons like Covid-19, but ionizing radiation can be an even more powerful weapon capable of devastating large populations [1-3].

Significant advancements have been made in radiation monitoring over the past century, particularly with developments in material science and nanotechnology. Currently, active radiation monitoring devices rely on electronics and require a continuous power source to record accumulate dose data over years for radiation workers. Electronic devices are prone to failure, and the loss of accumulated radiation data is unacceptable under any circumstances. Consequently, passive techniques like thermoluminescence dosimeters (TLDs) have become crucial for recording accumulated doses for radiation workers [4-7].

TLDs are reusable and compact, about one-tenth the size of a cone, making them suitable as personal dosimeters. When ionizing radiation interacts with TLD material, electrons become excited and are trapped in predetermined locations between the valence and conduction bands of wide-bandgap materials, typically insulators. These trapped electrons retain information about the absorbed dose for extended periods. TLD dosimeters are periodically checked, typically every few months to a year. When heated, the trapped electrons absorb thermal energy, freeing them from the traps. Upon recombination with luminescence centers, they emit light, which can be monitored as a function of temperature. The intensity of emitted light at a specific temperature provides information about the accumulated absorbed dose.

While there are several commercially available TLD dosimeters, commonly used standard dosimeters include LiF: Mg,Cu,P (TLD-700H), Al2O3 (TLD- 500), CaSO4:Dy (TLD-900), and CaF2:Dy (TLD-200) [8,9]. LiF is preferred for personal dosimetry as it is similar to human tissue. Recent advancements in nanotechnology have led to the development of new TLD phosphors that offer significant advantages over the standard TLDs mentioned above [10].A recently developed material, magnesium borate doped with rare earth elements, has become crucial in radiation monitoring, particularly in luminescence. Notably, it has also exhibited favorable thermoluminescence properties. This material demonstrates remarkable stability even under extreme weather conditions, including high temperatures and moisture. It offers excellent reusability and minimal loss of trapped electrons during long-term storage [11, 12].

In recent decades, numerous methods have been developed for synthesizing nanomaterials, encompassing both physical techniques and chemical approaches. Among these, chemical co-precipitation stands out as a cost-effective, straightforward, and reliable method for producing polycrystalline nanomaterials. Consequently, chemical co-precipitation was employed in this study [13, 14].

The research work delves into the synthesis of Mn-doped magnesium borate using the chemical co-precipitation method. The resulting polycrystalline nanostructured material was thoroughly characterized through X-ray diffraction and field emission scanning electron microscopy (FESEM) to investigate its structural and morphological properties. Following these characterizations, a set of synthesized materials was prepared and utilized for thermoluminescence studies. The material exhibited outstanding thermoluminescence properties, coupled with its reusability, making it a promising candidate for future commercial dosimeters (TLDs).

2. Experimental

2.1 Synthesis

The synthesis of manganese (Mn) doped magnesium borate nanoparticles was accomplished through the chemical co-precipitation method. To prepare the precursor solution, two distinct solutions were formulated: one containing magnesium sulfate (1 mol/l, 99.9% purity, Sigma-Aldrich) and the other containing boric acid (4 mol/l, 99.50% purity, Sigma-Aldrich). Both of these solutions were dissolved in 200 ml of double-distilled deionized water. To introduce manganese doping, MnCl2 (2.0 mol%) acquired from Alpha Assar company was initially dissolved in the magnesium sulfate solution. The quantities of precursors used were determined according to the stoichiometric ratio indicated in the following reaction [15]:

$$MgSO_4 + 4H_3BO_3 \Delta MgB_4O_7 + \underset{\longrightarrow}{H_2SO_{4\uparrow}} + 5H_2O\uparrow$$

Subsequently, the boric acid solution was added dropwise into the magnesium sulfate solution. After the complete transfer of the boric acid solution, the reaction mixture was stirred continuously for a duration of 60 minutes. Subsequently, the solution was kept at 85 °C for 4 hours to ensure the completion of the reaction process during nanoparticle growth. Once the growth process was successfully finished, the sample solution was carefully removed from the heating source and allowed to cool naturally to room temperature. The resulting nanoparticles were thoroughly rinsed with double-distilled water after being centrifuged at 6000 rpm to remove any unreacted salts. Finally, the nanoparticles were dried in a nitrogen gas environment. All reagents were used as obtained without any further purification.

2.2 Characterizations

The morphologies and structural characteristics of the generated nanostructures were examined using a field emission scanning electron microscope (FESEM: MIRA3 TESCAN). The analysis of crystal structure was performed with a "Bruker D8" X-ray diffractometer, utilizing Cu K radiation with a wavelength of 1.5405 Å. For studying the radiation dosimeter properties of the grown nanostructured thin film, a thermoluminescence instrument made by Harshaw was employed. ^{137} Cs gamma radiation source was used as the irradiation source for the dosimetry study from inter-university accelerator center, New Delhi.

3. Results and Discussion

3.1 XRD

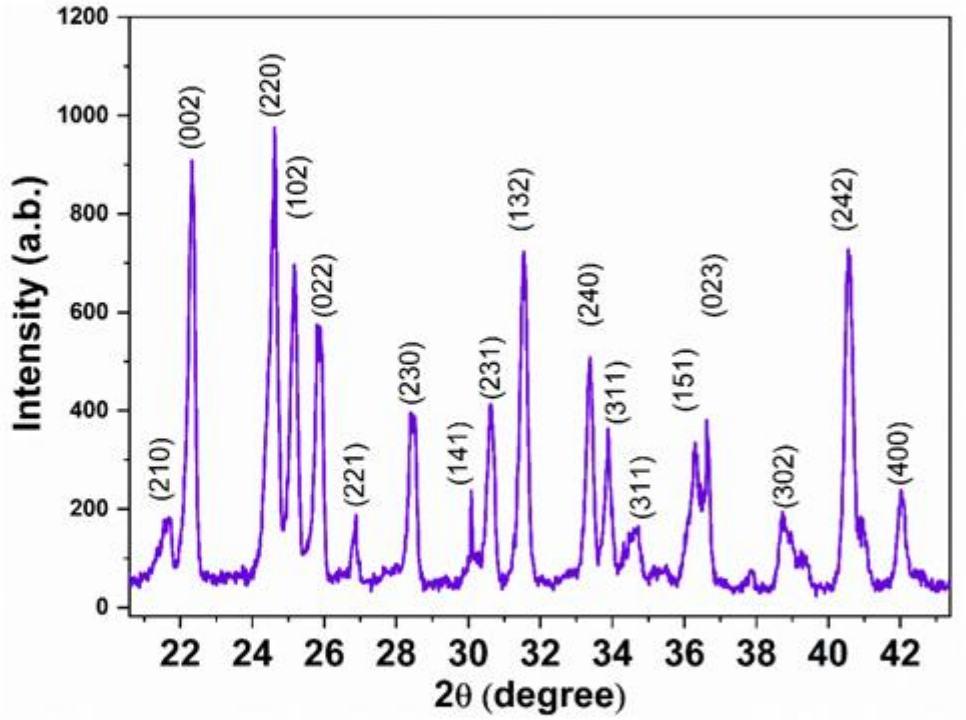

Figure 1: X-ray diffractogram of prepared magnesium borate nanoparticles

Figure 1 displays the X-ray diffractogram of the Mn-doped magnesium borate nanoparticles synthesized in this study. The successful synthesis of the material has been confirmed by comparing the XRD pattern of the prepared sample with the data available in the JCPDF (Joint Committee on Powder Diffraction Standards) file number 31-0787, which is consistent with previously reported literature. All the diffraction peaks were matched and indexed, and they closely corresponded to the JCPDF data [16,17]. The nanoparticles exhibit an orthorhombic crystal structure with the space group Pbca. Notably, there were no additional peaks observed that could be attributed to the presence of Mn dopant in the diffractogram and it confirms the perfect doping conditions.

3.2 FESEM

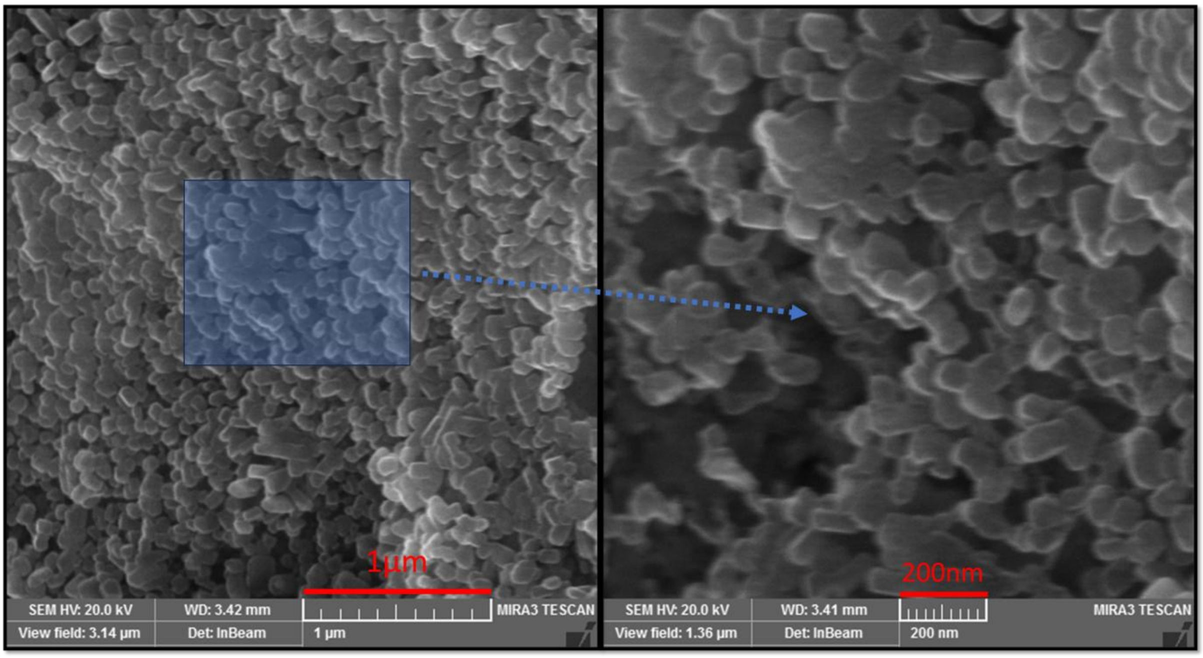

Figure 2: Represent the FESEM images of as synthesized magnesium borate nanoparticles

Figure 2 illustrates FESEM images used for the examination of the morphology of the developed nanostructures. The nanoparticles that have been cultivated exhibit a remarkable level of uniformity, as clearly depicted in the figure. These nanoparticles possess an average size ranging from approximately 20 to 30 nanometers, with some minor interconnected structures being observable. It is worth noting that the size of these nanoparticles is a critical factor influencing the linear thermoluminescence (TL) response of the nanoparticles even when exposed to very high levels of radiation [18].

3.3 Thermoluminescence

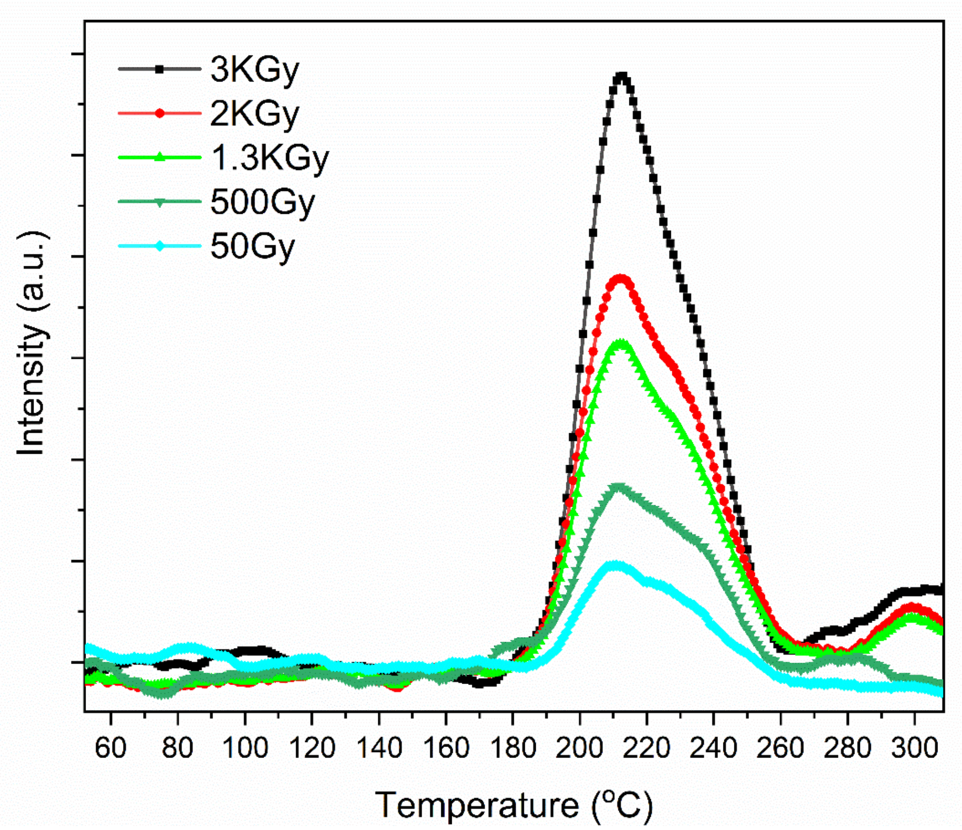

Figure 3: TL grow curve of Mn doped magnesium borate nanoparticles irradiated with Υ-rays

The structure of the TL glow curve and the reactivity of a thermoluminescent dosimeter (TLD) phosphor material are influenced by both the impurity levels within the forbidden region of the material's band structure and the ionic state of these impurities, which are typically rare earth elements, within the host matrix. In this specific case, nanomaterial magnesium borate was intentionally doped with 2.0 mol% of Mn as an impurity. Subsequently, it was exposed to varying radiation doses ranging from 50 Gy to 3 kGy, generated by a ^{137}Cs γ-source, and its thermoluminescence (TL) was recorded. It's worth noting that the material's sensitivity to gamma (γ) rays demonstrated an increase as the impurity concentration reached up to 2 mol% of Mn, but beyond this concentration, it declined.

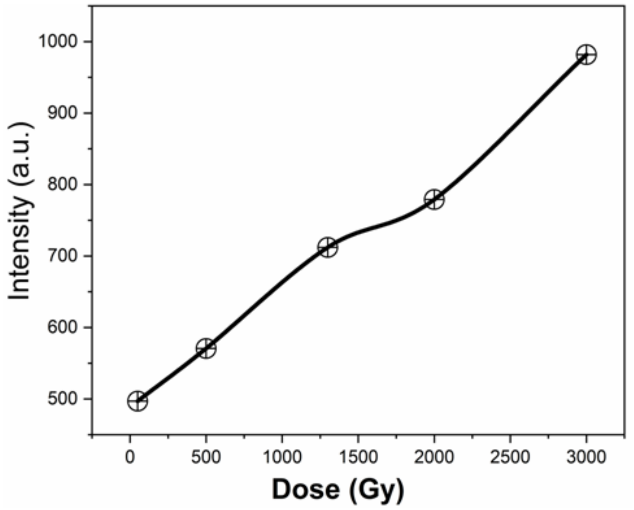

Figure 4: TL response curve drown from the TL glow curve represented in figure 3

This report exclusively focuses on the optimized impurity concentration for TL studies.The Mn doped magnesium borate nanoparticles functions as a highly sensitive and thermally stable thermoluminescent dosimeter (TLD) phosphor. An ideal optimized concentration of dopant, specifically 2 mol%, has been has been used to get the highest thermoluminescence (TL) sensitivity.The TL glow curves are represented in figure 3. It exhibited a straightforward pattern, primarily featuring a prominent peak at approximately 205 °C, accompanied by one smaller shoulder around 240 °C. Thermoluminescence (TL) measurements were conducted after subjecting pallets of synthesized nanoparticles to gamma (γ)-ray irradiation in the dose range of 50 Gy to 3 kGy.

The thermoluminescence (TL) peaks observed in this study are notably sharp, and their peak height consistently and proportionally increases as the radiation dose escalates. Figure 4 presents a response curve derived from the peak intensity of the TL glow curve across various radiation doses. Notably, this graph exhibits a high degree of linearity over a broad range of irradiation doses. It's worth highlighting that the nanoparticle-based phosphors investigated in this report display a significantly higher dose tolerance compared to standard micro-particle phosphors, which tend to saturate at much lower radiation doses.Importantly, these temperatures are significantly higher than room temperature and therefore it can be stored for several months without significant loss of TL information’s [20, 21].

4. Conclusions

The experimental findings indicate a successful synthesis of 30 nm highly uniform magnesium borate nanoparticles verified by X-ray and FESEM techniques. The Mn doped magnesium borate functions as a highly sensitive and thermally stable thermoluminescent dosimeter (TLD) phosphor. An ideal optimized concentration of dopant, specifically 2 mol%, has been has been used to get the highest thermoluminescence (TL) sensitivity. The TL glow curve exhibited a straightforward pattern, primarily featuring a prominent peak at approximately 205 °C, accompanied by one smaller shoulder around 240 °C. The nanoparticles exhibit a linear response across an extremely wide range of irradiation doses. Importantly, these temperatures are significantly higher than room temperature and therefore it can be stored for several months without significant loss of TL information’s.The study confirms that Mn-doped magnesium borate has significant potential as an excellent thermoluminescent dosimeter (TL dosimeter). It can find practical applications in commercial settings for monitoring radiation levels, ranging from small to very high doses of ionizing radiation. This includes extreme conditions such as those encountered in scenarios like nuclear bomb explosions or proximity to nuclear reactors.

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