

DEVELOPMENT OF Dy³⁺-DOPED BORO-TELLURITE THERMOLUMINESCENCE GLASSES FOR ENHANCED RADIATION DOSIMETRY APPLICATIONS

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Abstract

This study presents the development of boro-tellurite glasses doped with varying concentrations of Dy₂O₃ (0.0, 0.1, 0.5, 1.0, and 1.5 mol%) using a conventional melt-quenching technique, targeting advanced thermoluminescence materials for radiation detection. Structural characterization via X-ray diffraction confirmed the amorphous nature of all samples, while comprehensive optical investigations-including UV-Vis-NIR absorption and photoluminescence spectroscopy-revealed distinct intra-4f electronic transitions of Dy³⁺ ions. The incorporation of Dy₂O₃ not only altered the optical absorption and emission characteristics (exhibiting prominent blue and yellow bands) but also significantly modified the glass network, influencing density, molar volume, and the formation of defect-related electron traps. Thermoluminescence measurements, performed under controlled irradiation with an Am-241 source at a dose of 1 mGy and a heating rate of 5°C/s, exhibited a well-defined glow peak whose intensity and activation energy were strongly dependent on the Dy concentration. An optimal performance was observed at 1.0 mol% Dy₂O₃, where the activation energy peaked at 1.17 eV, indicative of deep and stable traps favorable for dosimeter applications. These results demonstrate that precise control of Dy₂O₃ doping in boro-tellurite glasses can significantly enhance their performance, making them promising candidates for reliable radiation dosimetry in medical.

Keywords: Boro-tellurite; Dosimeter; Thermoluminescence

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Introduction

Radiation dosimetry plays a crucial role across multiple fields, including medical radiotherapy, industrial radiation processing, environmental monitoring, and nuclear safety regulation. Accurate and reliable radiation detectors are vital for measuring radiation exposure and ensuring compliance with safety standards. Among various radiation detection materials, glass-based thermoluminescence dosimeters (TLDs) have gained prominence due to their transparency, excellent durability, ease of fabrication, and low production cost.

Recently, boro-tellurite glasses have emerged as highly promising candidates for advanced radiation dosimetry applications. These glasses combine exceptional optical properties, high chemical stability, and ease of doping with rare-earth ions, which collectively enhance their performance as thermoluminescence dosimeters (Kaur *et al.*, 2021a). The incorporation of tellurium oxide (TeO₂) contributes to a high refractive index and low phonon energy, substantially improving luminescence efficiency (Said Mahraz *et al.*, 2014). Incorporating B₂O₃ as a glass former endows the material with excellent glass-forming ability, a low melting point, improved rare-earth solubility, high thermal stability, and superior optical transmission (Kashif and Ratep, 2023). As a fundamental component within the glass matrix, ZnO confers structural stability and durability upon the material (Mondal *et al.*, 2020). Another important role of BaO is its increased density and increased optical transparency (Kalenda *et al.*, 2020) and Bi₂O₃ has a high refractive index (Prasanth *et al.*, 2022). Finally, the glass composed of Li₂O as a network modifier was found to be bubble-free, highly stable, and moisture-resistant, suitable for systematic analysis (Anjaiah *et al.*, 2019). Dysprosium oxide (Dy₂O₃) doping has received significant attention due to its potential to enhance thermoluminescence properties in boro-tellurite glasses (Hathot *et al.*, 2024). Dy³⁺ ions exhibit efficient energy storage capabilities because of their distinct intra-4f electronic transitions, allowing effective photon emission after radiation exposure. Nevertheless, achieving optimal Dy₂O₃ doping levels is essential; insufficient doping may result in weak luminescence, while excessive doping can lead to concentration quenching, structural distortions, and reduced luminescence intensity. Therefore, understanding the balance between doping concentration and thermoluminescence response is critical for maximizing dosimetry performance.

This study systematically investigates the effect of varying Dy₂O₃ doping levels on the structural, optical, and thermoluminescence properties of boro-tellurite glasses. By identifying an optimal doping concentration, this research aims to develop glass compositions with superior thermoluminescence performance, significantly advancing their potential for use in high-performance radiation detection and dosimetry applications.

Experimental

Boro-tellurite thermoluminescence glasses with the composition (35-x)TeO₂ - 35B₂O₃ - 10ZnO - 10BaO - 5Li₂O - 5Bi₂O₃ - xDy₂O₃ (where x = 0.0, 0.1, 0.5, 1.0, and 1.5 mol%) were synthesized using the conventional melt-quenching technique. The raw materials were carefully weighed and mixed to achieve a homogeneous composition. The mixture was placed in a high-purity alumina crucible and heated in an electric furnace at 1100°C with a heating rate of 4.58°C/min to ensure complete melting and homogenization. The molten glass was poured onto a preheated graphite mold and rapidly quenched to room temperature to form bulk glass samples. The samples underwent annealing at 350°C for 3 h. to relieve internal stresses before being cooled gradually to room temperature. The prepared glass samples were then cut and polished with two sizes 1.0×1.5×0.3 cm³ and 0.3×0.3×0.1 cm³, to appropriate dimensions for characterization shown in Figure 1. The prepared boro-tellurite glasses doped with Dy₂O₃ are referred to as TDy glasses throughout the manuscript.

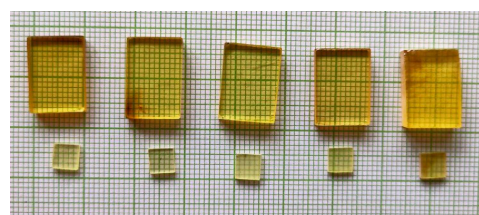


Figure 1. Photographs of the samples

The density (ρ) of the samples was determined with a densitometer (model AND HR-200), while the refractive index (n) was measured using an Abbe refractometer. Structural analysis was conducted through X-ray diffraction (XRD),

measuring in the range of 10 to 80 degrees on a Rigaku SmartLab X-ray Diffractometer to confirm the amorphous nature of the glass. Optical properties were evaluated to understand the effect of Dy_2O_3 doping on the glass. UV-Vis-NIR absorption spectra ranging from 500 to 2000 nm were recorded using a Shimadzu UV-3600 UV-VIS-NIR spectrophotometer to analyze the transitions. Excitation and emission spectra, as well as luminescence lifetimes, were measured with an Agilent Cary Eclipse fluorescence spectrometer to assess the luminescence behavior and efficiency of the samples. Thermoluminescence studies were performed to investigate the response of the glasses under radiation exposure using a thermoluminescence dosimeter reader (model RA'04400). These measurements provided valuable insights into how Dy_2O_3 doping affects the structural and optical properties of boro-tellurite thermoluminescence glass.

Results and Discussion

The density of the synthesized boro-tellurite glasses increased with Dy_2O_3 addition up to 0.5 mol%, from 4.46 to 4.52 g/cm^3 . This is attributed to the higher atomic mass of Dy compared to Te and the effective incorporation of Dy^{3+} ions into the glass matrix, which enhances atomic packing (Abouhaswa *et al.*, 2022). However, at concentrations above 0.5 mol%, the density decreased to 4.22 g/cm^3 (at 1.5 mol%). This reduction is likely due to the formation of Dy-O clusters or non-bridging oxygen species that disrupt the glass network and reduce structural compactness (Sadeq and Morshidy, 2019). In contrast, the molar volume slightly decreased from 31.8 to 31.5 cm^3/mol as Dy_2O_3 content increased, indicating a denser network at low concentrations. At higher doping levels, the trend suggests that the network becomes more open or distorted due to excessive dopant ions (Gurav *et al.*, 2024) occupying interstitial positions or causing structural strain. These results, shown in Figure 2, confirm that Dy_2O_3 significantly influences the packing density and network configuration of the glass.

X-ray diffraction analysis was performed to examine the structural nature of the synthesized boro-tellurite glasses. The obtained XRD patterns, depicted in Figure 3, exhibited broad diffraction humps rather than sharp crystalline peaks, confirming the amorphous nature of all TDy glasses (Sommat *et al.*, 2024). The absence of distinct Bragg reflections indicates that no long-range periodic order is present in the glass network. With increasing Dy_2O_3 concentration, no significant changes in the overall XRD pattern were observed,

suggesting that Dy incorporation does not induce crystallization within the glass matrix.

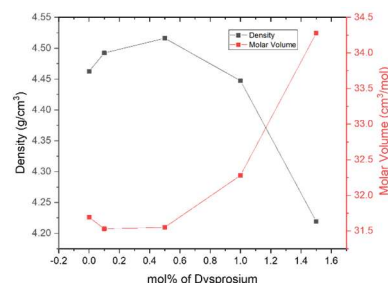


Figure 2. Trend of density and molar volume of TDy glasses

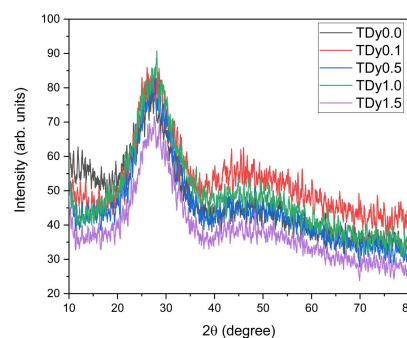


Figure 3. X-ray diffraction pattern of TDy glasses at different concentrations of dysprosium

The UV-Vis-NIR absorption spectra of the synthesized boro-tellurite glasses were recorded to investigate the optical transitions of Dy^{3+} ions in the glass matrix. According to the spectra shown in Figure 4, the measurement was carried out in the range of 500-2000 nm, which covers both the visible and near-infrared (NIR) regions. The spectra revealed characteristic absorption bands corresponding to the electronic transitions of Dy^{3+} , confirming the successful incorporation of the dopant into the glass network. The observed absorption bands were identified at specific wavelengths, corresponding to the transitions from the ground state $^6\text{H}_{15/2}$ to the excited states: $^6\text{F}_{3/2}$ (759 nm), $^6\text{F}_{5/2}$ (804 nm), $^6\text{F}_{7/2}$ (903 nm), $^6\text{H}_{7/2} + ^6\text{F}_{9/2}$ (1099 nm), $^6\text{H}_{9/2} + ^6\text{F}_{11/2}$ (1282 nm), and $^6\text{H}_{11/2}$ (1684 nm) (Carnall *et al.*, 1968). These peaks indicate the intra-4f transitions of Dy^{3+} ions, which are characteristic of rare-earth-doped glasses. With increasing Dy_2O_3 content, the intensity of these absorption bands showed a gradual increase, indicating an enhancement in the concentration of optically active Dy^{3+} ions.

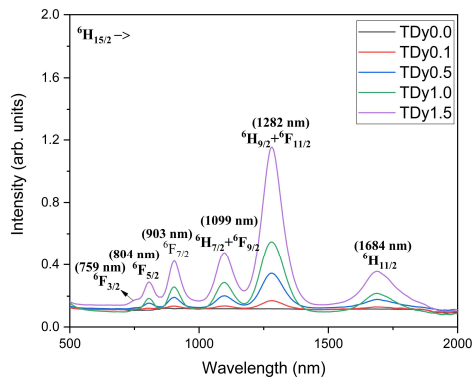


Figure 4. Absorption spectra of TDy glasses at different concentrations of dysprosium

The excitation spectra of the TDy glasses were obtained by monitoring the emission intensity at a fixed emission wavelength characteristic of Dy³⁺. The recorded spectra, as shown in Figure 5, reveal several well-defined peaks that correspond to the intra-4f transitions of Dy³⁺ ions. In particular, transitions from the ground state $^6H_{15/2}$ to various excited states, such as $^6P_{3/2}$ (325 nm), $^6P_{7/2}$ (351 nm), $^4P_{3/2}$ (366 nm), $^4K_{17/2}$ (388 nm), $^4G_{11/2}$ (427 nm), $^4I_{15/2}$ (453 nm), and $^4F_{9/2}$ (473 nm), are observable (Carnall *et al.*, 1968). The intensity of these peaks shows a noticeable variation with increasing Dy₂O₃ concentration, reflecting changes in the optical absorption behavior. This variation suggests modifications in the ligand field and symmetry around Dy³⁺ ions due to the dopant's effect on the glass network, as supported by previous studies on Dy-doped glasses (Shoaib *et al.*, 2020). This sensitivity to dopant concentration suggests that careful optimization of Dy₂O₃ is essential to maximize the luminescence efficiency.

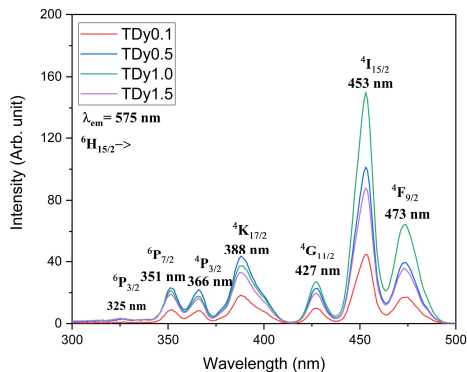


Figure 5. Excitation spectra of TDy glasses at different concentrations of dysprosium

Emission spectra were recorded under excitation at a wavelength corresponding to one of the prominent excitation peaks, which is 453 nm. The spectra, as shown in Figure 6, exhibit characteristic emission bands arising from transitions of the Dy³⁺ ions from the excited states back to the ground state $^6H_{15/2}$. Typically, two main emission bands are observed: one in the blue region (483 nm), corresponding to the $^4F_{9/2} \rightarrow ^6H_{15/2}$ transition, and another in the yellow region (575 nm), corresponding to the $^4F_{9/2} \rightarrow ^6H_{13/2}$ transition (Griebenow *et al.*, 2021). In addition to these dominant peaks, two smaller peaks are also noticeable. One of these appears in the red region, which can be attributed to a transition such as $^4F_{9/2} \rightarrow ^6H_{11/2}$ (663 nm), and the other likely arises from the $^4F_{9/2} \rightarrow ^6H_{9/2}$ (753 nm) transition. At higher Dy₂O₃ concentrations, the emission intensity shows a noticeable decline due to concentration quenching. This phenomenon occurs when the dopant ions are in closer proximity, facilitating nonradiative energy transfer processes (such as cross-relaxation) between Dy³⁺ ions. These processes lead to energy migration toward quenching centers where the energy is lost as heat rather than light (Bajaj *et al.*, 2022). As a result, there is an optimal dopant concentration, beyond which further increases in Dy³⁺ ions lead to enhanced quenching and a reduction in emission efficiency. In addition to analyzing the conventional yellow-to-blue (Y/B) emission intensity ratio, we introduce the emission density to matrix distortion (ED/MD) ratio as a complementary metric. This ED/MD ratio not only reflects the balance between the characteristic emission bands but also serves as an indicator of the local asymmetry in the glass network surrounding the Dy³⁺ ions (Zaman *et al.*, 2019). For instance, in the TDy1.0 sample, the ED/MD ratio is observed to be approximately 1.95. This relatively high ratio suggests that the Dy₂O₃ doping in TDy1.0 induces a beneficial level of asymmetry within the glass matrix.

The Commission Internationale de l'Éclairage (CIE) diagram was employed to evaluate the chromaticity of the light emitted by the Dy³⁺-doped glasses, thereby linking the observed spectral features to their perceived color. The color coordinates were calculated using the transformation equation proposed in the literature (Krishnaiah *et al.*, 2013). This equation provides a robust correlation between the measured emission spectra and the perceived color by converting the spectral data into precise (x, y) coordinates. For example, one of the optimized samples that is shown in Figure 7 exhibited coordinates of

approximately ($x = 0.33$, $y = 0.35$), confirming that the overall emission is dominated by a balanced contribution from the blue and yellow components, which results in a near-white appearance. This approach not only validates the spectral measurements but also enables a more accurate interpretation of the color quality inherent to the TDy glasses.

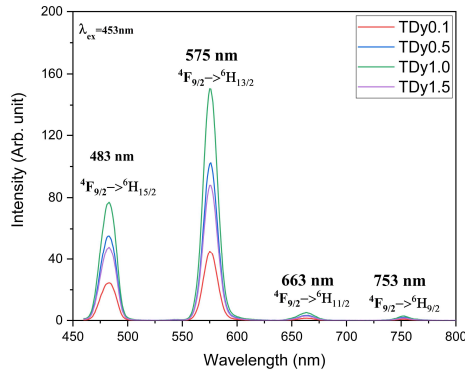


Figure 6. Emission spectra of TDy glasses at different concentrations of dysprosium

is attributed to enhanced nonradiative energy transfer processes, such as cross-relaxation among closely spaced Dy^{3+} ions, which become more prominent at higher concentrations (Carvalho Pinto *et al.*, 2020). Consequently, the increased probability of energy migration to quenching centers results in a diminished radiative decay component.

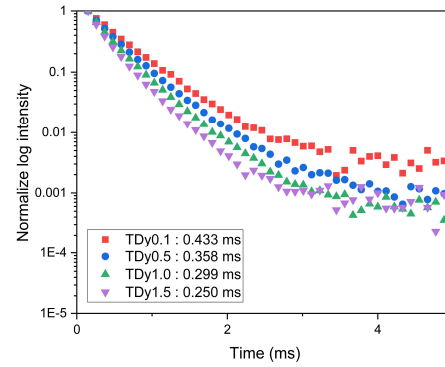


Figure 8. Decay time of TDy glasses at different concentrations of dysprosium

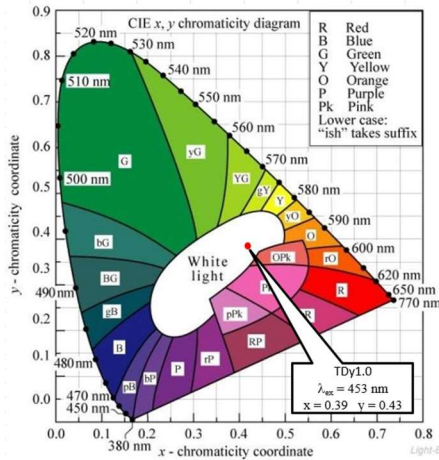


Figure 7. CIE diagram of TDy1.0 glass

Time-resolved luminescence measurements were conducted to evaluate the decay behavior of the TDy glasses at different Dy_2O_3 concentrations. The decay curves, as shown in Figure 8, were measured under 453 nm excitation and monitoring with emission 575 nm, exhibit a near-single exponential behavior at lower dopant levels, indicating a relatively uniform distribution of Dy^{3+} ions within the glass matrix. However, as the concentration of Dy increases, the observed decay times gradually decrease. This reduction in lifetime

Thermoluminescence (TL) is the emission of stored energy in the form of light from insulating or materials upon heating, following exposure to ionizing radiation. This phenomenon occurs due to the release of electrons from defect-related traps within the material. When exposed to radiation, electrons in the material absorb energy and are elevated from the valence band to the conduction band, subsequently becoming trapped at defect centers. Upon heating, these trapped electrons are thermally stimulated and recombine with luminescent centers, emitting photons. The activation energy (trap depth, E) can be estimated using the peak shape method, represented by the following equation (Kitis *et al.*, 2000; Vallejo *et al.*, 2020):

$$E_{\tau,\delta,\omega} = C_{\tau,\delta,\omega} \frac{kT_M^2}{\tau_{\delta,\omega}} - b_{\tau,\delta,\omega}(2kT_M) \quad (1)$$

$$C_{\tau} = 1.510 + 3.0(\mu_g - 0.42); \quad b_{\tau} = 1.58 + 4.2(\mu_g - 0.42) \quad (2)$$

$$C_{\delta} = 0.976 + 7.3(\mu_g - 0.42); \quad b_{\delta} = 0 \quad (3)$$

$$C_{\omega} = 2.52 + 10.2(\mu_g - 0.42); \quad b_{\omega} = 1 \quad (4)$$

Where $E_{\tau,\delta,\omega}$ is the activation energy (trap depth)

k is the Boltzmann constant (8.617×10^{-5} eV/K)

T_M is the glow peak temperature (in Kelvin)
 τ the half-width at the lower temperature side of the glow peak
 δ the half-width at the higher temperature side of the glow peak
 ω is the total half-width of the glow peak
 $b_{\tau,\delta,\omega}$ is the shape factor determined from the glow curve
 μ_g is the geometric factor

Thermoluminescence measurements of TDy glass were irradiated using an Am-241 source at a dose of 1 mGy and measured at a heating rate of 5°C/s. Figure 9 demonstrates the TL glow curves, clearly illustrating the single prominent glow peak for each glass composition. The glow curves indicate a clear correlation between Dy₂O₃ concentration and the intensity and position of the glow peak. The activation energies calculated using equations (1), (2), (3), and (4) and summarized in Table 1, ranged from 0.88 eV (TDy0.1) to a maximum of 1.17 eV (TDy1.0), decreasing slightly at higher dopant concentrations (TDy1.5). The clear TL glow peaks suggest well-defined electron traps within the glass matrix, significantly influenced by the Dy doping concentration. Increasing Dy content initially improves electron trapping efficiency, with the optimal TL performance observed at 1.0 mol% Dy₂O₃. A slight decrease in activation energy at the highest concentration (TDy1.5) indicates the presence of dopant clustering, which introduces non-radiative recombination pathways. The TDy1.0 glass demonstrated optimal thermoluminescence properties, characterized by a higher activation energy, implying more stable and deeper traps suitable for radiation dosimetry applications. This makes TDy1.0 a promising material for further investigation as a reliable radiation dosimeter. Moreover, the trend of decreasing glow peak temperature (T_m) and intensity at higher Dy₂O₃ concentrations can be explained by changes in trap depth and ion clustering behavior. At low Dy³⁺ doping levels (TDy0.1 and TDy0.5), the dopant ions are uniformly dispersed, with maximum electron crosssection value, maximum recombination probability on luminescence centers, and transportation probability of electrons from trap centers to luminescence centers and resulting in prominent TL peaks (Kaur *et al.*, 2021b). However, as the concentration increases (particularly in TDy1.5), Dy³⁺ ions tend to cluster, which causes the formation of shallower trap sites and facilitates non-radiative recombination. These clustered regions reduce trap depth, leading to lower T_m values.

Additionally, cross-relaxation and energy migration among closely spaced Dy³⁺ ions further quench the thermoluminescence signal, diminishing the peak intensity. These effects collectively account for the observed decrease in both glow peak temperature and intensity at high dopant levels (Kaur *et al.*, 2021b).

Table 1. Data on various trap depth parameters of TDy glasses

Sample	T_m (°C)	μ_g	E_T	E_δ	E_ω	E_{av}
TDy0.1	247.85	0.49	0.84	0.91	0.88	0.88
TDy0.5	240.85	0.50	0.94	0.99	0.97	0.96
TDy1.0	238.85	0.51	1.16	1.18	1.18	1.17
TDy1.5	224.85	0.50	0.97	1.01	0.99	0.99

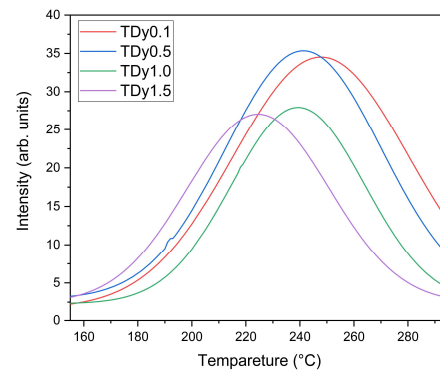


Figure 9. Glow curve of TDy glasses at different concentrations of dysprosium

Conclusions

The investigation of Dy³⁺-doped boro-tellurite glasses revealed significant insights into the interplay between glass structure and luminescent properties. The optimal doping concentration was identified as 1.0 mol% Dy₂O₃, demonstrating the highest thermoluminescence efficiency with a prominent glow peak and activation energy of 1.17 eV. Structural analysis confirmed the amorphous nature of the glasses, unaffected by the doping levels used. The luminescent properties exhibited distinct transitions suitable for efficient radiation detection. These findings suggest that Dy³⁺-doped boro-tellurite glasses have considerable potential for application as radiation dosimeters, with further exploration warranted to optimize their performance for practical dosimetry applications.

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