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INVESTIGATION OF THERMOLUMINESCENCE PROPERTIES OF CALCITE FROM KAMAN, KIRŞEHIR

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Abstract: This study investigates the thermoluminescence (TL) properties and dosimetric performance of calcite samples, with particular emphasis on dose response, heating rate (HR) effects, and reusability (RU). TL measurements were carried out using a Lexsyg Smart TL/OSL reader equipped with a $^{90}Sr/^{90}Y$ beta source. The dose-response relationship was analyzed by irradiating the sample with beta doses ranging from 0.1 to 500 Gy, revealed a progressive increase in TL intensity. The TL glow curve exhibited two broad peaks at 100-116 °C and 238 °C, with a shift in the first peak temperature at higher doses. A fitting procedure was applied to analyze the dose-response characteristics of the material, revealing distinct *b* values for different dose ranges. The effect of HR on the glow curve was also studied, showing an anomalous increase in TL intensity with rising HRs, contrary to expectations from the standard theoretical model. Additionally, the RU of the material was assessed by performing 10 consecutive TL measurements after a 50 Gy irradiation, demonstrating minimal degradation in TL intensity, with low deviations from the mean and first values. This remarkable RU is crucial for ensuring the material's effectiveness in long-term dosimetric applications.

Keywords: Thermoluminescence, Calcite, Anomalous hearing rate, Dosimetry, Reusability

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1. Introduction

Thermoluminescence (TL) is a well-established technique for radiation dosimetry, providing valuable information on the radiation dose absorbed by materials. It is widely used in various fields, including environmental monitoring, medical applications, archaeology and more (McKeever, 1985; Aitken, 1985; McKeever et al., 1995; Chen and McKeever, 1997). The technique relies on the ability of certain materials to trap charge carriers (electrons and holes) in their crystal lattice when exposed to ionizing radiation. Upon subsequent heating, these trapped charge carriers are released and recombine, emitting light (TL signal) which is proportional to the absorbed dose. Understanding the dose response and the factors affecting TL emission is crucial for optimizing its application in dosimetric measurements (Townsend and Kirch, 1989; Bailiff et al., 2004; Kim and Hong, 2014). Calcite (CaCO3) is a prevalent mineral that forms rocks and represents the most stable crystalline form of calcium carbonate, with aragonite being the alternative structure. It is commonly found in diverse geological settings, including sedimentary, igneous, and metamorphic rocks. While pure calcite is typically colorless or white, variations in its color often indicate the presence of different trace elements (Wintle, 1978; Chang et al., 1998). Natural calcite is widely utilized in the construction industry, serving as a key material in the production of cement and whitewashing powder, as well as in wall painting and industrial pollution management (Ponnusamy et al., 2012). Calcite has been extensively studied for its TL properties, owing to its favorable characteristics, such as high sensitivity and stability. In particular, it was reported that the TL properties of calcite make it a potential candidate for beta radiation dosimetry in industrial applications, with a reliable performance across a dose range of 0.01–104 Gy (Urbina et al., 1998). A study showed that the dose-response characteristics of calcite highlight its suitability as a dosimeter, particularly under varying thermal conditions, where thermal treatments affect its TL response (El-Faramawy et al., 2022). It was also shown that calcite's strong green emission band, with peaks at 255°C and 305°C, and its low fading rates position it as an important candidate for dosimetry applications (Toktamiş et al., 2014). Furthermore, it was found that calcite exhibits measurable and stable TL signals with a dose of 50 Gy, showing peaks at 150°C, 235°C, and 315°C, which make it effective for beta radiation dosimetry (Soliman and Metwally, 2006). Despite numerous studies, a comprehensive understanding of the impact of different factors, such as dose, HR, and RU, on calcite's TL response remains a topic of significant interest. Previous studies have highlighted the role of temperature and HR in influencing the TL signal, with the potential for anomalous HR effects that deviate from conventional TL behavior. These effects can alter the peak temperature and intensity of the glow curves, making it essential to consider the influence of heating conditions on the overall TL response. Therefore, further investigation into these factors is crucial for optimizing calcite's use in dosimetric applications. The calcite sample used in this study was obtained from the Kaman region in Kırşehir, Turkey. This study focuses on investigating the dose response, HR dependence, and RU of calcite as a thermoluminescent material. By performing a series of controlled experiments, the relationship between radiation dose and TL emission is explored, and the influence of different HRs on the glow curve characteristics is analyzed. Moreover, the RU of calcite is assessed through repeated measurements to evaluate the stability of its TL response over multiple cycles. The findings of this study aim to provide deeper insights into the performance and practical applicability of calcite for radiation dosimetry.

2. Materials and Methods

2.1. Mineral Calcite

The geological history and natural formation conditions of calcite play a crucial role in determining its luminescence characteristics, as variations in crystallization environments can lead to differences in trap depth distributions and luminescent efficiency. Therefore, detailed investigations of region-specific calcite samples are essential for understanding their potential use in dosimetric applications. The calcite sample analyzed in this study was sourced from Kaman, Kırşehir, Turkey (Orhan, 2020). Figure 1 presents a detailed visualization of the studied calcite sample. Specifically, Figure 1a displays the geological map of Turkey at a 1:100.000 scale, specifically the Kırşehir-J30 sheet, as provided by MTA (2001). Figure 1b shows a macroscopic photograph of the calcite sample with a scale of 1.5 cm, providing an overview of its physical appearance, while Figure 1c presents a microscopic image of the calcite captured using an optical microscope, with a scale of 500 µm, highlighting its finer structural details. X-ray diffraction (XRD) analysis was performed to determine the crystal structure and phase composition of the investigated calcite sample. Measurements were carried out using a MiniFlex 300/600 diffractometer under conditions of 40 kV and 15 mA, with a continuous scanning mode in the range of 3° to 90° and a step width of 0.020°. Qualitative analysis results indicated that the main phase in the sample is calcite ($CaCO_3$), which crystallizes in the R-3c (hexagonal) space group. The determined lattice parameters for calcite are a = 4.983 Å, c = 17.033 Å, with a unit cell volume of 366.34 Å^3 (Figure 2). The natural samples and XRD results, obtained through the project titled Investigation of Mineralogical, Geochemical, and Origin of Skarn Mineralization Related to Plutonic Rocks from the Kaman Region (Kırşehir, Central Anatolia) [NEUBAP Report No: 15/2F7, 2018] were provided for this study (Orhan and Orhan, 2018).

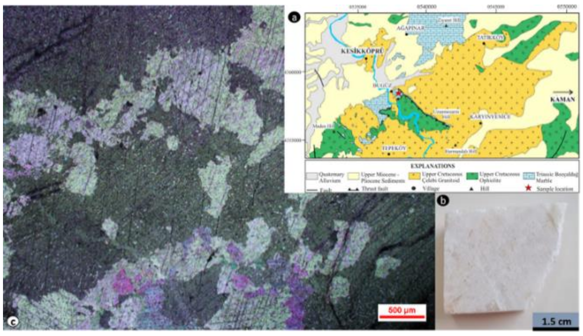

Figure 1. Visualization of the calcite sample: (a) Geological map of Turkey, Kırşehir-J30 sheet; (b) Macroscopic photograph of the calcite sample; (c) Microscopic image of the calcite under optical microscopy.

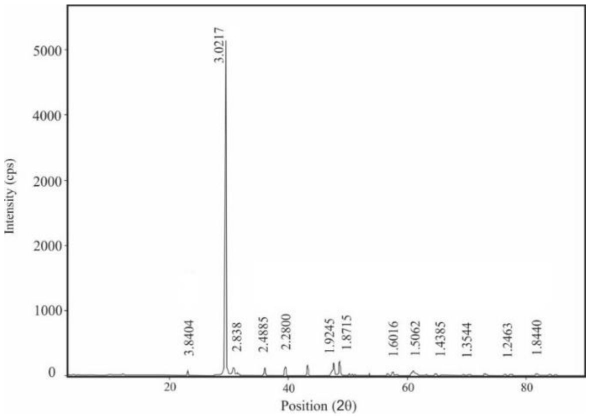

Figure 2. X-ray diffraction (XRD) analysis of the calcite sample.

2.2. Experimental Details

A Lexsyg Smart TL/OSL reader equipped with a $^{90}Sr/^{90}Y$ beta source delivering a dose rate of 0.097 Gy/s was utilized for all TL measurements in an N_2 atmosphere (Bulcar et al., 2024). To prepare the sample, 35 mg of powder was compressed into a pellet (6 mm in diameter, 0.75 mm thick) by applying a pressure of 2 ton-force/cm^2 for 10 minutes. A single aliquot, weighing 32.5 mg, was used in TL experiments. Glow curves were recorded from room temperature (RT) to 500 °C at a constant HR of 2 °C/s, except for the HR analysis. For the filter selection test, the sample was subjected to a 10 Gy beta dose, and different filter combinations were evaluated using the TL reader. Based on these tests, the IRSL-TL-wideband blue filter was chosen for all subsequent measurements. To assess dose response, the sample underwent beta irradiation with incremental doses ranging from 0.1 to 500 Gy. Additionally, to study the effect of HR, the pellet was irradiated at beta doses of 10, 30, and 50 Gy, followed by TL measurements at HRs varying between 1 and 5 °C/s. RU tests were carried out using a 50 Gy dose and a HR of 2 °C/s, with 10 consecutive TL readouts. The net TL intensity was determined by subtracting the background signal from the first measurement for all TL experiments

3. Results and Discussion

3.1. Dose Response

One of the primary steps in evaluating a dosimeter's applicability for dose measurement is determining its dose response, which should ideally follow a linear or well-defined function. The relationship between TL intensity and irradiation dose, representing the total radiation energy absorbed by the sample, offers insights into the trapping and thermal release mechanisms of carriers in the crystal lattice (Kirsh, 1992). When ionizing radiation interacts with the material, charge carriers are trapped within the crystal structure. The number of trapped carriers (or more specifically, the intensity of the TL emitted) is generally proportional to the amount of radiation dose absorbed by the phosphor (Aitken, 1985). A linear relationship between dose and TL intensity is a highly desirable feature for TL materials used in dosimetric applications. In order to examine this response, the sample was exposed to beta radiation doses ranging from 0.1 Gy to 500 Gy, after which luminescence emission curves were recorded (Figure 3).

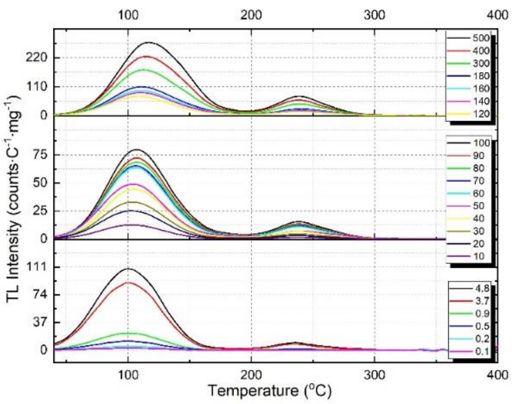

Figure 3. Temperature-dependent TL glow curves for beta-irradiated samples (0.1-500 Gy).

The results showed that emission intensity increased with dose up to 500 Gy, which was the maximum limit due to the saturation of the photomultiplier tube (PMT). The TL glow curves maintain their shape and exhibited two prominent and broad peaks, one occurring between 100–116 °C and the other at 238 °C. At lower doses, the peak with the highest intensity appeared at 100 °C, but as the dose increased, this peak shifted to 116 °C. This shift in the maximum TL peak temperature (T_m) with increasing dose can be attributed to several underlying physical mechanisms. Higher doses lead to a greater number of electrons being trapped in deeper energy levels within the material, requiring higher thermal energy for release. Additionally, as dose increases, retrapping effects become more significant, meaning that some released electrons are re-captured in deeper traps rather than recombining immediately, delaying the TL emission and shifting T_m to higher temperatures. Competition between recombination and thermal release also plays a role, as an increased density of trapped charges affects the kinetics of the TL process. Furthermore, high radiation doses can induce modifications in the local band structure or defect clustering, altering the activation energy required for charge release (Ducruet and Vass, 2009; Akça, 2020). For deeper analyze the dose response, a fitting procedure was applied using the equation 1:

$$I = aD^b \tag{1}$$

where I represents the integrated TL intensity, and D is the radiation dose. Upon plotting I_M (maximum value of intensity) as a function of D on a log-log scale, the equation produces a straight line with a slope b that could be equal to 1. (Oglakci et al., 2025). This analysis was performed separately for three dose intervals: 10–50 Gy, 50–100 Gy, and 10–100 Gy (Figure 4).

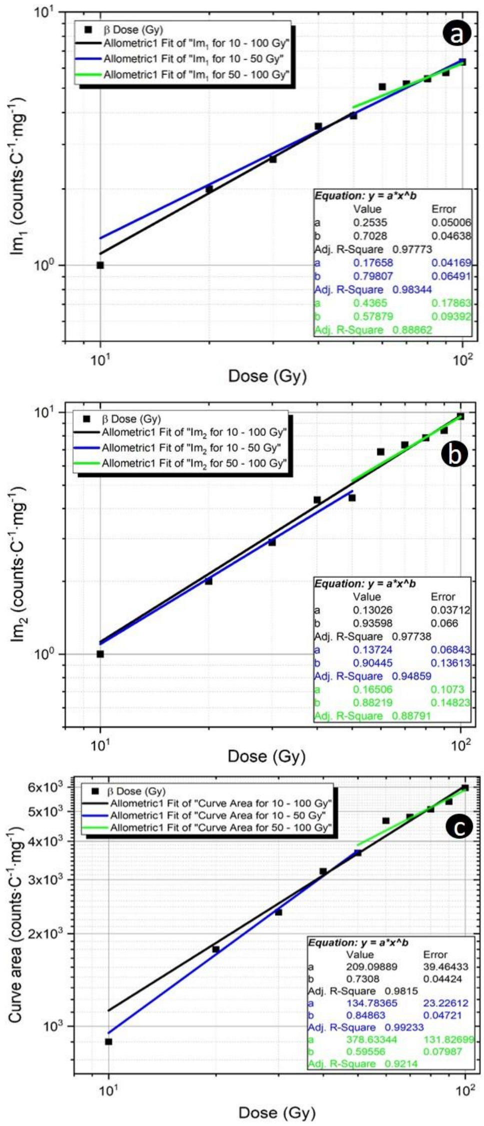

Figure 4. Dose-response analysis for 10–50 Gy, 50–100 Gy, and 10–100 Gy intervals: (a) first peak, (b) second peak, and (c) full glow curve.

The b parameter, which characterizes the nature of the dose-response relationship, was found to be 0.849 (R^2 = 0.9834) for the 10–50 Gy range, 0.596 (R^2 = 0.8886) for

50–100 Gy, and 0.731 (R^2 = 0.9777) for 10–100 Gy for the first peak (Figure 4a). As seen in Figure 4b, a similar fitting procedure was applied to the second peak at 238 °C, yielding b values of 0.904 (R^2 = 0.9486) for 10–50 Gy, 0.882 (R^2 = 0.8879) for 50–100 Gy, and 0.936 (R^2 = 0.9774) for 10–100 Gy. This peak demonstrated a stable dose response across the tested range. While analyzing individual peaks provides valuable information, evaluating the response of the entire glow curve offers a more comprehensive understanding of the material's radiation sensitivity. Therefore, the dose-response fitting for the full glow curve was performed and displayed in Figure 4c, resulting in b values of 0.849 (R^2 = 0.9923) for 10–50 Gy, 0.596 (R^2 = 0.9214) for 50–100 Gy, and 0.731 (R^2 = 0.9815) for 10–100 Gy. These findings suggest that the dose response for both individual peaks and the full glow curve follows a nearly linear relationship, with the b values varying across different dose intervals, and the I_m of the second peak (ultimately influencing the overall dosimetric properties of the material) showing the most consistent dose response (with b value 0,936) across the tested range.

3.2. Various Heating Rates (VHR)

Typically, TL glow curves shift toward higher temperatures as the HR increases during the TL readout process, as higher HRs cause traps within the bandgap to empty more rapidly. (Chen and Kirch, 1981). Figure 5 illustrates this trend in the TL low curves of calcite under different HRs (1-5 oC/s) with three different dose values. In order to achieve a deeper analysis, the maximum intensities of peak 1 and 2 (I_{m1} and I_{m2}), peak temperatures of peak 1 and 2 (T_{m1} and T_{m2}), full widths at half maximum of peak 1 and 2 ($FWHM_1$ and $FWHM_2$), and total curve area were plotted as functions of HR values in Figure 6a-c for 10, 30, and 50 Gy, respectively. According to TL theory, the FWHM should increase while the I_m decreases, though the total integrated intensity is expected to remain unchanged (Kitis et al., 1993).

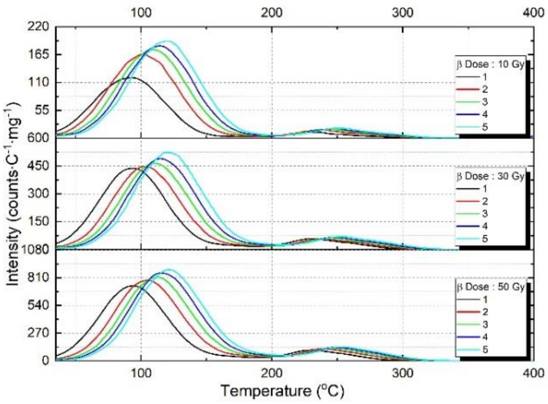

Figure 5. TL glow curves of calcite at different HRs (1-5 °C/s) and doses (10, 30, 50 Gy).

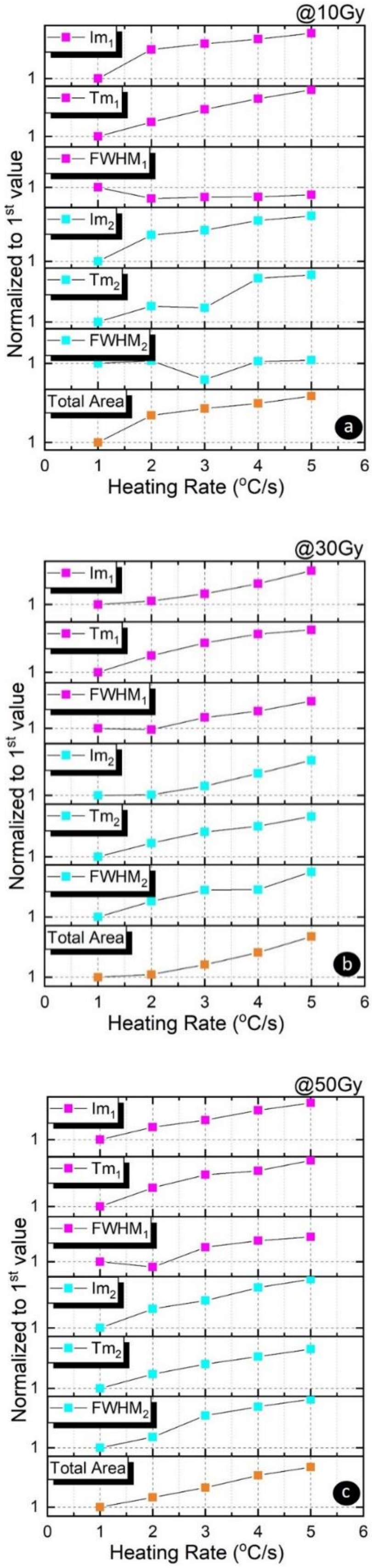

Figure 6. Variation of maximum intensities (I_{m1}, I_{m2}), peak temperatures (T_{m1}, T_{m2}), full width at half maximum ($FWHM_1$, $FWHM_2$), and total curve area as functions of HR for irradiated samples at (a) 10 Gy, (b) 30 Gy, and (c) 50 Gy.

However, as illustrated in Figure 6, increasing HR leads to a decrease in $FWHM_1$ and $FWHM_2$ for the 10 Gy dose (Figure 6a), accompanied by an increase in I_{m1}, I_{m2}, and the total curve area, whereas for the 30 Gy (Figure 6b) and 50 Gy doses (Figure 6c), all these parameters exhibit an increasing trend, which contradicts the expected outcome predicted by the OTOR model. This behavior, referred to as the anomalous HR effect, has been explained by Pagonis et al. (Pagonis et al., 2013) using Mandowski's semi-localized transition (SLT) model (Mandowski, 2005). Their study introduced an additional non-radiative transition pathway from the directly excited state to the recombination center, describing what is known as the anti-quenching effect. When traps release electrons upon heating, these electrons may transition thermally into the conduction band, where they can either be recaptured or recombine with holes in the recombination centers. In the first scenario, this recombination generates a TL signal, whereas in the second, the electron recombines directly with a hole without emitting radiation. The relative likelihood of radiative versus non-radiative transitions depends on the HR; as the HR decreases, the proportion of non-radiative transitions increases, and vice versa. This effect is attributed to the two-step process of charge carrier excitation from traps. At higher HRs, the probability of thermal excitation into the conduction band increases, reducing the time electrons spend in the excited state. The anomalous effect arises due to competition between radiative and non-radiative processes. Since charge conservation must be maintained, an increase in total TL intensity corresponds to a proportional decrease in the non-radiative component. Consequently, the rise in integrated TL intensity reflects an increase in the probability of radiative transitions relative to non-radiative ones (Mandowski, 2011; Pagonis et al., 2013; Portakal Uçar, 2021; Oglakci et al., 2023).

3.3. Reusability (RU)

The RU of the material was evaluated by performing successive thermoluminescence (TL) measurements on calcite samples, where each measurement was conducted after irradiating the sample with a 50 Gy dose at a HR of 2 °C/s. This process was repeated 10 times to assess the stability of the TL response over multiple cycles and shown in Figure 7. For the 1st peak, the deviation from the mean value was calculated to be 1.01%, while the deviation from the first reading was 2.75% (Figure 8a). Similarly, for the 2nd peak, the deviation from the mean value was 1.06%, and the deviation from the first reading was 1.08% (Figure 8b). However, a more comprehensive analysis of the entire glow curve rather than individual peaks gives a better representation of the material's RU. When considering the entire glow curve, the deviation from the mean value was calculated to be 1.40%, while the deviation from the first value was 3.97% (Figure 8c) (Oglakci et al., 2023).

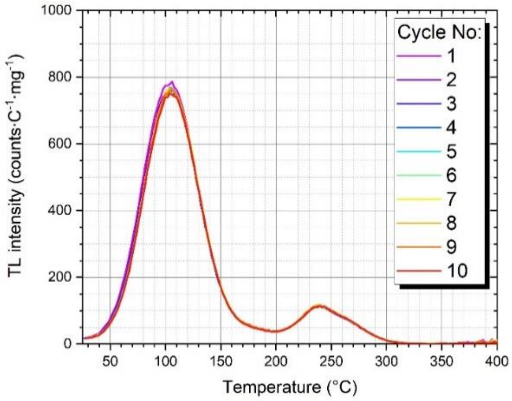

Figure 7. Stability of the TL response in calcite after 10 successive irradiations with a 50 Gy dose at a 2 °C/s HR.

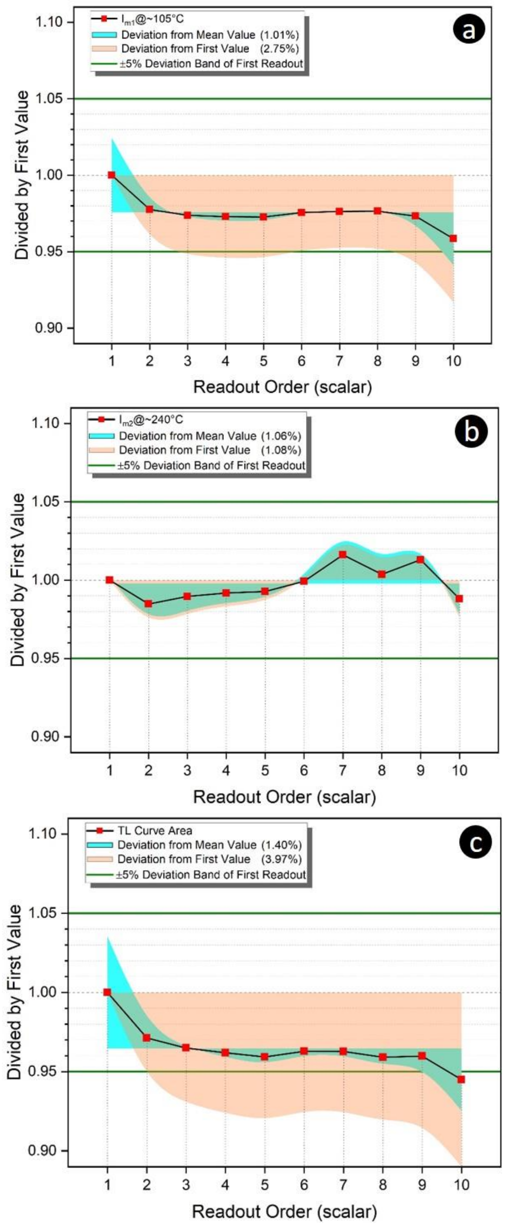

Figure 8. (a) Deviation of the first peak from the mean and first reading; (b) Deviation of the second peak from the mean and first reading; (c) Deviation of the entire glow curve from the mean and first reading.

These low deviations indicate that the material demonstrates outstanding RU, as it maintains a stable TL response even after multiple irradiation and readout cycles, showing minimal degradation in its thermoluminescent properties. Such remarkable RU is essential for practical dosimetric applications, where materials need to maintain performance over time without significant loss of sensitivity or efficiency.

4. Conclusion

The calcite material demonstrated stable thermoluminescent properties, with a well-defined dose-response relationship and an anomalous HR effect that deviated from conventional predictions. The results indicate that as the radiation dose increases, both the density of trapped charges and recombination dynamics contribute to shifts in the maximum TL peak temperature. The study also highlighted the importance of evaluating the entire glow curve for more reliable dosimetric characterization. RU tests revealed that the material retains its TL intensity over repeated measurements, confirming its robustness and suitability for practical dosimetric applications. The low deviations observed in successive measurements emphasize the material's excellent RU, ensuring reliable performance without significant sensitivity loss over time. This study contributes valuable insights into the performance and stability of calcite as a dosimeter for radiation measurements, showcasing its potential for extended use in radiation monitoring and dosimetry.

Author Contributions

The percentages of the author’s contributions are presented below. The author reviewed and approved the final version of the manuscript.

	S. B.
C	100
D	100
S	100
DCP	100
DAI	100
L	100
W	100
CR	100
SR	100
PM	100
FA	100

C=Concept, D= design, S= supervision, DCP= data collection and/or processing, DAI= data analysis and/or interpretation, L= literature search, W= writing, CR= critical review, SR= submission and revision, PM= project management, FA= funding acquisition.

Conflict of Interest

The author declared that there is no conflict of interest.

Ethical Consideration

Ethics committee approval was not required for this study because of there was no study on animals or humans.

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References

Aitken MJ. 1985. Thermoluminescence Dating. Academic Press, London, UK, pp: 26.

Akça S. 2020. Determination of thermoluminescence properties of ZnB_2O_4:Tm^{3+}, Li^+ for dosimetric purposes. Appl Radiat Isot, 157: 109041.

Bailiff IK, Stepanenko VF, Gosku HY, Bøtter-Jensen L, Brodski L, Chumak V, Correcher V, Delgado A, Golikov V, Jungner H, Khamidova LG, Kolizshenkov TV, Likhtarev I, Meckbach R, Petrov SA, Sholom S. 2004. Comparison of retrospective luminescence dosimetry with computational modeling in two highly contaminated settlements downwind of the Chernobyl NPP. Health Phys, 86: 25–41.

Bulcar K, Oglakci M, Altowyan AS, Bilgin R, Yucel A, Sezer S, Hakami J, Depci T, Topaksu M, Can N. 2024. Exploring beta irradiation responses in Sr_2^+-doped hydroxyapatite: A thermoluminescence study. Ceram Int, 50(17B): 31300–31312.

Chang LLY, Howie RA, Zussman J. 1998. Rock Forming Minerals, Non-silicates—Sulphates, Carbonates, Phosphates, Halides, Vol. 5. Geological Society of London, London, UK, 2nd ed, pp:25.

Chen R, Kirsh Y. 1981. The Analysis of Thermally Stimulated Processes. Pergamon Press, Oxford, UK, pp: 53.

Chen R, McKeever SWS. 1997. Theory of Thermoluminescence and Related Phenomena. World Scientific, Singapore, pp: 42.

Ducruet JM, Vass I. 2009. Thermoluminescence: experimental. Photosynth Res, 101: 195–204.

El-Faramawy N, Alazab HA, Gad A, Sabry M. 2022. Study of the thermoluminescence kinetic parameters of a β-irradiated natural calcite. Radiat Phys Chem, 190: 109793.

Kim KB, Hong DG. 2014. Kinetic parameters, bleaching and radiation response of thermoluminescence glow peaks separated by deconvolution on Korean calcite. Radiat Phys Chem, 103: 16–21.

Kirsh Y. 1992. Kinetic analysis of thermoluminescence. Phys Status Solidi A, 129(1): 15–48.

Kitis G, Spiropulu M, Papadopoulos J, Charalambous S. 1993. Heating rate effects on the TL glow-peaks of three thermoluminescent phosphors. Nucl Instrum Meth Phys Res A, 73: 367–372.

Mandowski A, Bos AJJ. 2011. Explanation of anomalous heating rate dependence of thermoluminescence in YPO_4:Ce^{3+}, Sm^{3+} based on the semi-localized transition (SLT) model. Radiat Meas, 46: 1376–1379.

Mandowski A. 2005. Semi-localized transitions model for thermoluminescence. J Phys D Appl Phys, 38: 17–21.

McKeever SWS, Moscovitch M, Townsend PD. 1995. Thermoluminescence Dosimetry Materials: Properties and Uses. Nuclear Technology Publishing, Ashford, UK, pp: 63.

McKeever SWS. 1985. Thermoluminescence of Solids.

Cambridge University Press, Cambridge, UK, pp: 41.

Oglakci M, Akça-Özalp S, Portakal-Uçar ZG, Correcher V, Benavente JF, Sonsuz M, Can N, Halefoglu YZ, Topaksu M. 2025. Thermoluminescence study of Nd^{3+} doped lanthanum tri-borate phosphor. J Alloys Compd, 1013: 178570.

Oglakci M, Portakal-Uçar ZG, Akça-Özalp S, Correcher V, Benavente JF, Sonsuz M, Can N, Halefoglu YZ, Topaksu M. 2023. Thermoluminescence behavior of Ce^{3+}-doped lanthanum tri-borate phosphor for dosimetry applications. Ceram Int, 49(22B): 36092–36102.

Orhan A, Orhan A. 2018. Investigation of mineralogical, geochemical and origin of skarn mineralization related plutonic rocks from the Kaman region (Kırşehir, Central Anatolia). NEUBAP Project Report No: 15/2F7, pp: 85.

Orhan A. 2020. Mineralogy, fluid inclusions and carbon isotopes of the Kaman iron deposits, Central Anatolia, Turkey: Implication for ore genesis and hydrothermal evolution. Ore Geol Rev, 127: 103808.

Pagonis V, Blohm L, Brengle M, Mayonado G, Woglam P. 2013. Anomalous heating rate effect in thermoluminescence intensity using a simplified semi-localized transition (SLT) model. Radiat Meas, 51–52: 40–47.

Ponnusamy V, Ramasamy V, Jose MT, Anandalakshmi K. 2012. Effect of annealing on natural calcitic crystals—A thermostimulated luminescence (TSL) study. J Lumin, 132(4): 1063–1075.

Portakal Uçar ZG. 2021. Thermoluminescence characteristics and kinetic analysis of beta irradiated $Ca_4LaO(BO_3)_3$ phosphor. Cumhuriyet Sci J, 42(3): 702–714.

Soliman C, Metwally SM. 2006. Thermoluminescence of the green emission band of calcite. Radiat Eff Defects Solids, 161(10): 607–613.

Toktamiş H, Toktamiş D, Yazıcı AN. 2014. Thermoluminescence studies of calcite extracted from natural sand used in making roasted chickpea. J Lumin, 153: 375–381.

Townsend PD, Kirsh Y. 1989. Spectral measurement during thermoluminescence—An essential requirement. Contemp Phys, 30: 337–354.

Urbina M, Millán A, Beneitez P, Calderón T. 1998. Dose rate effect in calcite. J Lumin, 79(1): 21–28.

Wintle AG. 1978. A thermoluminescence dating study of some Quaternary calcite: potential and problems. Can J Earth Sci, 15(12): 1884–1893.