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TEMPERATURE EFFECT ON THERMOLUMINESCENCE KINETIC PARAMETERS OF NANO-ALUMINA

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Abstract. This research explores the fundamental thermoluminescence characteristics of irradiated nano-α-alumina particles, investigating their response to varying heating rates. The study involves recording TL luminescence curves, revealing a distinct peak with a maximum at approximately 202°C. As dose levels increase, the peak consistently shifts towards lower temperatures, indicating adherence to non-first-order kinetics (b≠1). The crystallite size was calculated using XRD analysis and estimated as 40nm. To examine the impact of the heating rate on the TL glow curve and derive kinetic parameters for nano α-Al_2O_3, specimens were exposed to a 6 kGy dose. Subsequently, TL glow curves were documented over a temperature range from room temperature to 300°C, employing different heating rates (2, 4, 6, 8 and 12°C/s). The peak temperature of the glow peak shifts towards higher temperatures as the heating rate increases and the peak intensity continuously diminishes, aligning with TL theory. The observed decrease in TL glow peak intensity with escalating heating rates is attributed to thermal quenching, where quenching efficiency rises at higher temperatures. Normalizing maximum TL intensities to the lowest heating rate (2°C/s) reveals a substantial 22% decrease in peak intensity.

Keywords: *Activation energy, nano α-alumina, thermoluminescence, heating rate.*

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1. Introduction

The heating rate has a significant impact on the thermoluminescence (TL) properties of various materials. The effect of heating rate on the glow curve of MgB_4O_7:Tm,Dy has also been studied by González et al. (2022), who found that the kinetics parameters can be accurately determined using the sequential quadratic programming glow curve deconvolution (SQPGCD) method. This is consistent with the findings of Cruz-Zaragoza et al. (2011), who also observed changes in peak temperatures, peak intensities, and total area of glow peaks with increasing heating rates in other materials. Cruz-Zaragoza et al. (2011) found that as the heating rate increases, the peak intensity at the maximum decreases and shifts to a higher temperature. This was also observed by Ogundare (2005) in the case of fluorite, where the glow-peak temperatures increased with heating rate. Piters (1999) highlighted the influence of a temperature lag on the TL analysis, which can lead to a decrease in activation energy and frequency factor. Kitis (1993) studied the effects of heating rate on the TL glow-peaks of different

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phosphors, providing a comprehensive comparison of experimental results and theoretical calculations. These studies collectively demonstrate the significant impact of heating rate on TL properties.

The thermoluminescence glow curve of alumina (Al_2O_3) is influenced by the heating rate, with the response of single-crystal detectors decaying exponentially and ceramic detectors decaying linearly (Silveira, 2021). Thermoluminescence of α-Al_2O_3:C irradiated to low doses (0.04–7.20 mGy) similar to those measurable in the environment has been investigated. The glow curve consists of a dominant peak near 200°C and two additional glow peaks of weaker intensity at around 74 and 342°C, respectively for measurements made at 5°C/s. Analysis of the main peak using various methods shows that in this low dose range, the peak follows non-first order kinetics and also that the peak is unitary rather than being a composite of two overlapping peaks as is thought to be the case at comparatively higher dose. Its dose response was observed to be linear and the dependence of the luminescence intensity on heating rate showed that the thermoluminescence is subject to thermal quenching. However, further examination suggests that thermal quenching may be more of an effect with increase in luminescence excitation dose (Ogundare *et al*., 2013). However, Kumar (2006) cautions that the conservation of area under the glow curve is not always guaranteed and that the glow peak height can increase with heating rate.

Considering the preceding discussions, the objective of this study was to explore the thermoluminescence (TL) characteristics of alpha-alumina (α-Al_2O_3) when subjected to varying radiation doses and heating rates. The focus was on analyzing the dose-response relationship and conducting kinetic analysis of the primary glow peak.

2. Materials and methods

In this study, nano-sized α-Al_2O_3 particles with sizes of 40 nm commercially available from Skyspring Nanomaterials, Inc. were used as samples.

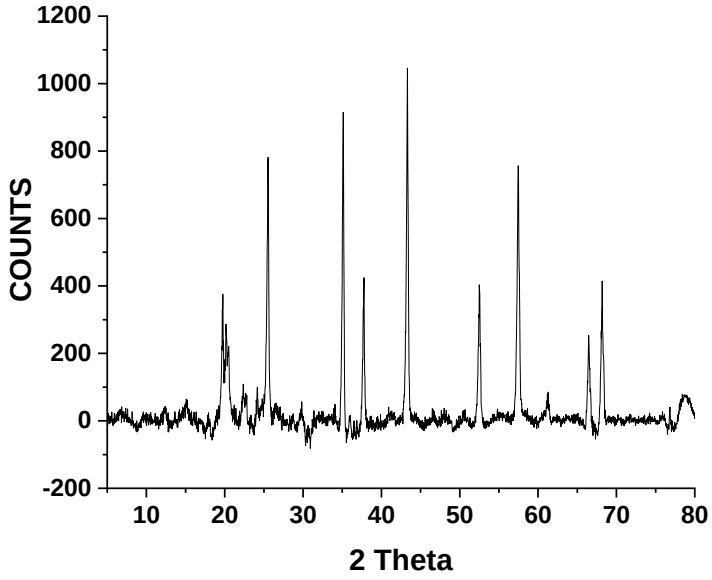

Figure 1. XRD pattern of nano-α-alumina with 40 nm particle size

The XRD pattern of the sample is shown in Figure 1. Samples were scanned in the range $5 \leq 2\Theta \leq 80°$ at 1.2°/min scan rate. Semi-quantitative estimates of mineral phase abundances were derived from the PXRD data using the intensity of specific reflections, density and mass absorption coefficients of the elements for CuKα radiation. The crystallite size was calculated using OriginPro according to the formula:

$$D = \frac{K\lambda}{\beta \cos\theta},$$

where D= Crystallite size; β=line broadening at FWHM in radians; θ (Bragg angle)=peak center; λ= X-ray wavelength; K (Dimensionless shape factor)=0.94 for spherical crystallites. The samples were irradiated at ambient temperature with a ^{60}Co gamma source with a dose rate of 1.76 Gy/s. The dose rate was determined using a Magnette Miniscope MS400 EPR spectrometer with individually packed BioMax alanine dosimetry films with barcode markings developed by Eastman Kodak Company. The Harshaw TLD 3500 Manual Reader was utilized to assess the TL sample characteristics using a linear heating rate of 2°C/s from 323K to 673K in an N_2 atmosphere with a Pilkington HA-3 heat-absorbing filter (Mammadov, 2024; Mammadov *et al.*, 2023). Three aliquots of 5 mg each were used for each measurement and the TL data points represented the average of the three aliquots. A thin layer of the sample powder was uniformly distributed on the planchet surface to ensure a uniform TL signal (Mammadov & Abishov, 2023).

3. Results and discussion

3.1. Dose response

The low-dose responses of nano α-Al_2O_3 with a particle size of 40 nm were evaluated within the dose range of 0.2 kGy to 8 kGy and TL glow curves were recorded (Figure 2). Figure 2 reveals a distinct and prominent peak in the TL glow curve, with its maximum occurring at approximately 202±2°C. Notably, this TL peak exhibits a tendency to shift towards lower temperatures with an increase in dose level, which is not within the bounds of experimental error.

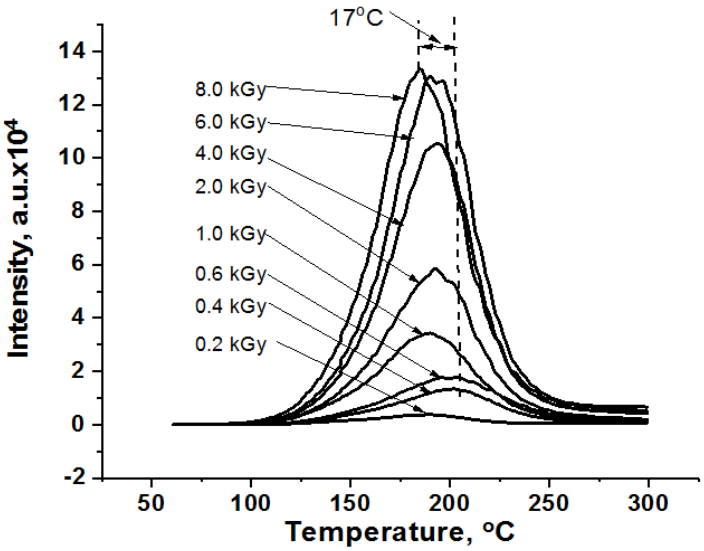

Figure 2. TL glow curves of nano-α-Al_2O_3 obtained with heating rate of 2°C/s

The peak maximum temperature ($Tmax$) at the maximum intensity ($Imax$) are fundamental characteristics of a thermoluminescence (TL) peak. It is both theoretically predicted and experimentally verified that ($Tmax$) shifts to higher temperatures as the readout heating rate increases. Additionally, it is theoretically predicted that ($Tmax$) shifts to lower temperatures as the radiation dose increases (Kitis *et al*., 2020). According to TL theory, peak temperatures are anticipated to change solely with the heating rate for first-order kinetics (order of kinetics b=1). Consequently, under a constant heating rate, the peak maximum should remain relatively stable, unaffected by other experimental parameters and within the limits of experimental errors (Yazici & Topaksu, 2003). Therefore, if the TL peak temperature decreases with rising dose levels, indicating non-first-order kinetics (b≠1), otherwise, it suggests first-order kinetics (b=1). In this case temperature shift is approximately 17°C. The relationship between TL response and dose for nano α-Al_2O_3 with a particle size of 40 nm is depicted in Figure 3.

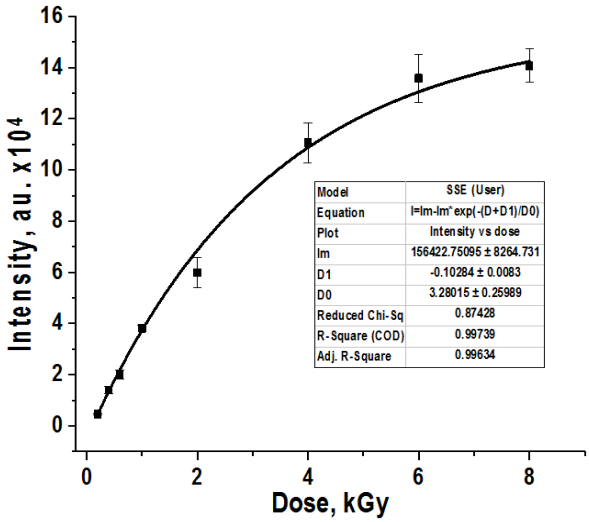

Figure 3. Dose dependence of TL intensity of nano-α-Al_2O_3 obtained at heating rate of 2°C/s

The one trap one recombination (OTOR) model is a basis for analyzing the effect of radiation dose on $Tmax$, which predicts the behavior of the TL curve as well as the dose-response pattern. However, the empirical general order kinetic equation (Kitis *et al*., 2020) helps to understand the nature of this effect. In this model, the dependence of $Tmax$ on radiation dose, which is mainly evident for second-order kinetics, gradually weakens with decreasing kinetic order and finally disappears for first-order kinetics. Despite the extensive focus on dose-response studies in the TL literature, the shift of $Tmax$ as a function of dose has not been clearly demonstrated experimentally.

3.2. Impact of Heating Rate

The influence of heating rate on TL glow curves is a fundamental experimental variable in TL measurements (Pagonis *et al*., 2006). The heating rate applied to dosimetric materials affects the variation in their TL sensitivity and consequently, the trends observed in the dose curve (Ogundare *et al*., 2005).

In TL dosimetry applications, alterations in heating rate impact the TL glow peak (or curve) area and TL glow peak height (Kitis *et al*., 2020). To assess the effect of heating rate on the TL glow curve and calculate kinetic parameters for nano α-Al_2O_3 with a

particle size of 40 nm, samples were irradiated with a 6 kGy dose and TL glow curves were recorded from room temperature to 300°C using various heating rates (2, 4, 6, 8 and 12°C/s).

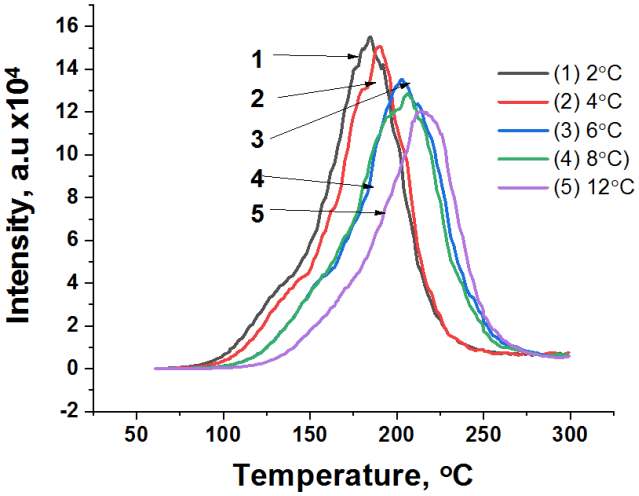

Figure 4. TL glow curve of nano-α-Al_2O_3 at different heating rates

Figure 4 displays TL glow curves of the irradiated samples, indicating that the peak temperature of the glow peak shifts towards higher temperatures as the heating rate increases and the peak intensity continuously diminishes, aligning with TL theory (Rasheedy & Zahran, 2006).

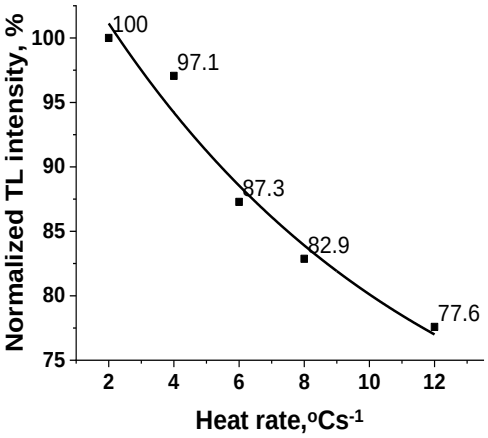

Figure 5. Normalized TL glow curve intensity at different heating rates

Numerous studies have reported a decrease in TL glow peak height (or area) intensity with an increase in heating rate. This phenomenon is attributed to thermal quenching, whose efficiency escalates with rising temperatures (Ogundare *et al.*, 2005; Spooner & Franklin, 2002; Dogan *et al.*, 2017). Normalizing the maximum TL intensities of the glow peaks to the lowest heating rate (2°C/s) reveals a roughly 22% decrease in peak intensity (Figure 5).

The maximum value of the main dosimetry peak temperature is close to that determined for alumina doped with carbon, with a predominant TL peak centered at 471

K (Rodriguez *et al*., 2011) and α-Al_2O_3, where T_m=465 K (Mammadov *et al*., 2024; Mammadov *et al*., 2023; Mishra *et al*., 2007), but significantly higher than that determined for alumina crystal (T_m=450 K) (Kitis *et al*., 1993), bauxite (T_m=412 K) (El-Taher *et al*., 2013) and Al_2O_3 doped with Sr, Li and Ge (T_m=448 K) (Saharin *et al*., 2017). Natural diaspores exhibit TL glow curves with a low-temperature maximum peaked at 453 K and a wide broad curve above 490 K (Garcia-Guinea *et al*., 2005). Diapores samples show a discrete distribution of electron traps at a lower temperature (~463 K) and a continuous structure of traps at a higher temperature (above 500 K), which is due to dehydroxylation and oxidation of the chromophore (Garcia-Guinea *et al*., 2001). Al_2O_3 nanoparticles doped with Cr (particle size of 25 nm) show a prominent peak at approximately 474 K and a linear response from 100 Gy to 20 kGy (Saharin *et al*., 2011).

4. Conclusions

This study explored the fundamental TL and XRD properties of irradiated nano-α-alumina particles, particularly their response to varying heating rates. The recorded TL glow curves revealed that as the radiation dose increased, the peak consistently shifted towards lower temperatures. This behavior indicates that the TL peak follows non-first-order kinetics, providing new insights into the kinetic processes of nano-α-alumina.

Heating rates emerged as a critical experimental variable, significantly influencing the dosimetric performance of nano α-Al_2O_3. The TL glow peak temperature shifted to higher values as the heating rate increased, while peak intensity diminished. This behavior aligns with established TL theory, offering valuable information for optimizing the use of nano α-Al_2O_3 in dosimetric applications under different heating conditions.

The decrease in TL glow peak intensity with higher heating rates was attributed to thermal quenching, where quenching efficiency rises with temperature. This finding highlights the importance of considering thermal quenching effects in the development and refinement of TL materials.

Notably, the study revealed an exponential increase in TL intensity with increasing radiation dose for the exposed nanoparticles. This suggests that alumina nanoparticle powder could serve as a promising substrate material for ionizing radiation dosimetry, opening up new possibilities for its application in radiation monitoring and dosimetric technologies. Further development could focus on enhancing sensitivity and exploring its use across a broader range of radiation environments.

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