Nigerian Journal of Physics (NJP)
ISSN online: 3027-0936
ISSN print: 1595-0611
DOI: https://doi.org/10.62292/njp.v34i2.2025.409
Volume 34(2), June 2025

Influence of Annealing Temperature on the Thermoluminescence Properties and Kinetic Parameters of Synthetic Quartz

*[1]Fierkwap, S. B., [1]Makama, E. K., [2]Makama, R. E. and [1]Yohanna, E. M.

[1]Department of Physics, University of Jos, Jos, Nigeria
[2]Department of Civil Engineering, University of Jos, Jos, Nigeria

*Corresponding author's email: fierkwap@unijos.edu.ng

ABSTRACT
Thermoluminescence (TL) in synthetic quartz has long been a key tool in radiation dosimetry, geological applications, and archaeological dating due to its regulated flaw patterns and repeatable luminescence qualities. However, as demonstrated by earlier research, thermal annealing can impact TL sensitivity in a way that differs significantly from more substantial, long-term glow curve alterations. Nevertheless, there hasn't been much research done on how these changes might manifest in kinetic parameters such as frequency factors and activation energy. This study investigates the effects of annealing synthetic quartz specimens that had never been annealed at temperatures of roughly 500°C and 900°C on its TL characteristics. The TL glow curves were analyzed using five complementary kinetic techniques in order to reveal any changes in the kinetic model, order, recombination rate dynamics, and trap depth. The findings indicate a significant shift in the peak position of glow markings and a systematic drop in activation energy as the annealing temperature rises. The TL process surprisingly maintained first-order dynamics. The study addresses a critical gap by linking annealing-induced defect evolution with quantifiable kinetic changes, offering a pathway toward engineering synthetic quartz with tailored TL responses for specific technological applications.

Keywords:
Thermoluminescence,
Synthetic Quartz,
Annealing,
Activation Energy,
Trap Dynamics.

INTRODUCTION

Thermoluminescence (TL) is a well-established phenomenon in which certain materials emit light upon heating due to the release of trapped charge carriers from defect sites. This property has made TL materials, particularly quartz, invaluable in applications such as radiation dosimetry, archaeological dating, and geological studies (McKeever, 1985). Among the various forms of quartz, synthetic quartz is of particular interest due to its controlled defect structure, which allows for more reproducible and tunable luminescence properties compared to natural quartz (Yang and McKeever, 1990). Quartz's luminescence is tied inherently to its defect chemistry, including impurities, vacancies and interstitial atoms. These defects act as trapping and recombination centres that determine the conditions under which TL emission occurs (Agulló-López et al., 1988). Previous studies have demonstrated that thermal treatments, such as annealing, can significantly alter these defect configurations, thereby modifying the TL sensitivity, peak positions, and kinetic parameters (Galloway, 2002). In certain quartz samples, for example, annealing at moderate temperatures (such as 500°C) has been demonstrated to improve TL sensitivity, whereas higher temperatures (such as 900°C) may result in a decrease because of the annihilation or reconfiguration of flaws (Chithambo et al., 2011).

Recent advances in luminescence spectroscopy have further elucidated the role of annealing in synthetic quartz. Time-resolved optical studies by Chithambo and Niyonzima (2017) identified multiple emission bands in the radioluminescence spectra of annealed synthetic quartz, with intensities varying systematically with annealing temperature. These findings suggest that annealing not only affects the concentration of defects but also their relative dominance in the recombination process. However, the impact of annealing on the kinetic parameters of TL peaks, particularly the activation energy and frequency factor, remains less explored. Understanding these effects is crucial for optimizing synthetic quartz for dosimetric applications, where precise control over TL properties is essential.

The primary objective of this study is to investigate the influence of annealing temperature on the TL properties of synthetic quartz, with a focus on samples annealed at 900°C. Complementary measurements on unannealed and 500°C annealed samples provide a comparative framework to assess the thermal evolution of TL characteristics. Specifically, the study examines: The changes in glow curve structure, including peak positions and intensities; The kinetics of the main TL peak, evaluated using multiple analytical methods; The effect of annealing on activation energy and thermal quenching. By addressing these aspects, this work aims to contribute to a deeper understanding of defect engineering in synthetic quartz and its implications for TL-based technologies. The findings may also inform the development of advanced materials with tailored luminescence properties for specialized applications in radiation detection and beyond.

MATERIALS AND METHODS

Sample Preparation and Annealing Protocol

The experimental investigation was carried out using commercially sourced synthetic quartz (Sawyer Research Products, Ohio, USA). The quartz crystals were mechanically ground to grain sizes ranging from 90 to 500 µm to ensure uniformity in irradiation and thermal exposure during analysis.

Annealing treatments were conducted in a high-temperature furnace. One batch of samples was heated to 500 °C and another to 900 °C, both for 10 minutes duration, followed by natural cooling in air. These annealing temperatures were selected based on previous findings that suggested significant structural and luminescent modifications at elevated temperatures (Pagonis et al., 2006). A set of unannealed samples was retained as a baseline for comparison.

Irradiation and TL Measurement Setup

Thermoluminescence measurements were performed using a Riso TL/OSL DA-20 Luminescence Reader. This setup includes a photomultiplier tube (EMI 9635QA) for signal detection and a Hoya U-340 filter (transmission range: 250–390 nm) to isolate the relevant TL emission bands. All samples were exposed to beta irradiation using a 90Sr/90Y source at a constant dose rate of 0.10 Gy/s, up to a total dose of 40 Gy, to induce trapped charge carriers within the quartz lattice. The heating rate was fixed at 1°C s^{-1}, consistent with prior studies (Pagonis et al., 2006).

Kinetic Analysis Methods

To determine the influence of annealing on TL response, glow curves were recorded at a heating rate of 1 °C/s. Subsequent kinetic analysis focused on the main glow peak observed in the low-temperature region (<100 °C), using a variety of established methods: initial rise, peak shape, whole glow peak integration, variable heating rate, and glow curve deconvolution. These methods were applied consistently across the three sample types to ensure reliable comparison. The experiment was repeated thrice and the results were reported as mean ± standard deviation. Each technique provided estimates for key kinetic parameters, including activation energy (E), frequency factor (s), and kinetic order (b), which collectively describe the charge carrier recombination processes underlying the observed luminescence.

i. The Initial Rise Method holds that the low-temperature edge of a TL peak follows the relationship $I(T) \propto \exp(-E/kT)$. Intensities from the rising side (50–80°C for the 70°C peak; 60–95°C for the 86°C peak) were extracted and a plot $ln\,(I)$ vs. $1/kT$ was linearly fitted to determine E from the slope.

ii. The Whole Glow Peak Method entails that the order of kinetics is iteratively solved for b = 1 (first order) and b = 2 (second order) to identify the best-fit kinetics. This was done by modifying Pagonis et al. (2006) equation:

$$ln\left(\frac{I}{n^b}\right) = \left(\frac{S'}{\beta}\right) - \left(\frac{E}{kT}\right) \quad (1)$$

Where n is the area under the peak.

iii. For the Peak Shape Method, the following parameters were used for the calculation of the activation energy in Eq. 2: $\tau = T_m - T_1$ (low-temperature half-width), $\delta = T_2 - T_m$ (high-temperature half-width), $\omega = T_2 - T_1$ (full width at half maximum).

$$E_\alpha = C_\alpha\left(\frac{kT_m^2}{\alpha}\right) - b_\alpha(2kT_m) \quad (2)$$

Where $\alpha \in \{\tau, \delta, \omega\}$ and C_α, b_α are constants (McKeever, 1985).

iv. Variable Heating Rate Method: Glow curves at heating rates (β) of 0.2, 0.5, 1, 2, and 5°C s^{-1} were recorded and a plot of $ln(T_m^2/\beta)$ vs. $1/kT_m$ for each peak was obtained from which E and S were obtained from the slope and the intercept, respectively.

v. Curve Fitting with General-Order Kinetics was done using Kitis et al. (1998) model, given as Eq. 3

$$I(T) = I_m b^{b/(b-1)} exp\left(\frac{E}{kT} - \frac{T-T_m}{T_m}\right)\left[(b-1)(1-\Delta)\frac{T^2}{T_m} exp\left(\frac{E}{kT} - \frac{T-T_m}{T_m}\right) + Z_m\right]^{-b/(b-1)} \quad (3)$$

where $\Delta = 2kT/E$. This process was achieved using MATLAB's nonlinear least-squares solver and the goodness-of-fit was evaluated via the figure of merit (FOM < 5%).

RESULTS AND DISCUSSION

This section presents the thermoluminescence (TL) response of synthetic quartz under different annealing conditions: unannealed, annealed at 500 °C, and annealed at 900 °C. The glow curve characteristics, kinetic order, and activation energies were systematically analyzed using a range of techniques.

Glow curve characteristics

Figure 1 shows the TL glow curve of synthetic quartz annealed at 900 °C and measured at a heating rate of 1 °C/s after irradiation to 40 Gy. A prominent single peak is observed at approximately 86 °C. The inset compares this curve with those from unannealed and 500 °C-annealed samples, which exhibit dominant peaks at approximately 70 °C.

The observed shift in the main TL peak from 70°C (unannealed and 500°C samples) to 86°C (900°C sample) suggests fundamental changes in the trap structure. This shift could be attributed to several factors including the creation of new trapping centres due to defect reorganization at high temperatures, the annihilation of existing traps and formation of more stable configurations, or changes in the relative dominance of different recombination pathways.

The reduction in intensity of the respective secondary peaks (110°C, 180°C, 310°C) after 900°C annealing indicates that higher temperatures may preferentially eliminate certain defect types while stabilizing others. This result corroborates the observation reported by Yang and McKeever (1990) on thermal quenching effects in quartz. All three samples exhibit three peaks, but the position and intensity of the main peak shift with annealing temperature. These variations indicate that annealing affects trap depth and recombination mechanisms. The focus of further analysis is on the main low-temperature peak in each sample.

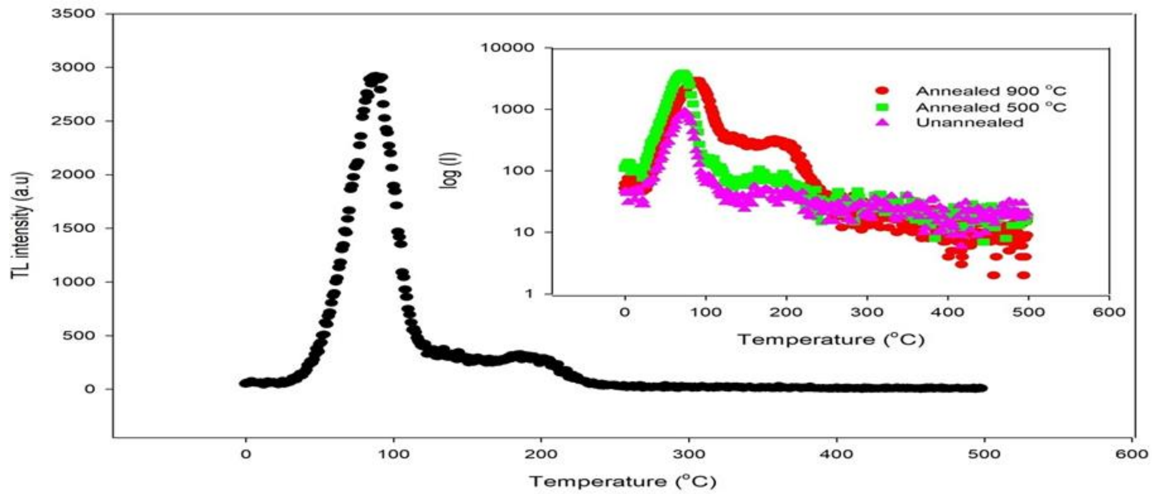

Figure 1: Glow curve of synthetic quartz annealed at 900°C recorded at 1 Cs^{-1} after irradiation to 40 Gy. Inset shows a semi logarithmic plot of the same glow curve for: Annealed 900°C, Annealed 500°C, and unannealed

Kinetic Analysis

Activation Energy

Five different analytical techniques were used to determine the activation energy (E) associated with the main thermoluminescence (TL) peak of synthetic quartz under three different sample conditions: unannealed, annealed at 500°C, and annealed at 900°C. These techniques included Initial Rise (IR), Whole Glow Peak (HGP), Peak Shape (PS), Variable Heating Rate (VHR), and Curve Fitting (CF). The results are summarized in Table 1. All methods indicate a consistent decrease in activation energy with increasing annealing temperature as presented in Figure 2. While the values from each technique vary slightly due to methodological assumptions, the downward trend across the board is robust and statistically supported by the reported uncertainties.

Table 1: Activation Energy (*E*, in eV) from Various Methods for the Three Sample Conditions

Sample Condition	Activation Energy, *E* (eV)				
	Initial Rise	**Whole Glow Peak**	**Peak Shape**	**Variable Heating Rate**	**Curve Fitting**
Unannealed	0.91 ± 0.01	0.90 ± 0.02	0.92 ± 0.03	0.91 ± 0.03	1.03 ± 0.03
500 °C-annealed	0.83 ± 0.02	0.91 ± 0.01	0.89 ± 0.92	0.82 ± 0.03	0.96 ± 0.02
900 °C-annealed	0.77 ± 0.03	0.67 ± 0.02	0.67 ± 0.68	0.52 ± 0.02	0.80 ± 0.04

The observed decrease in activation energy (E) with rising annealing temperature reflects substantial modifications in the trap structure of synthetic quartz. For the unannealed samples, E values clustered around 0.90 to 1.03 eV, indicating relatively deep and stable traps. Annealing at 500 °C caused minor changes in E, suggesting partial restructuring of the trap system. However, annealing at 900 °C resulted in a significant drop in activation energy (to as low as 0.52 eV), implying a shift toward shallower and potentially more thermally unstable traps.

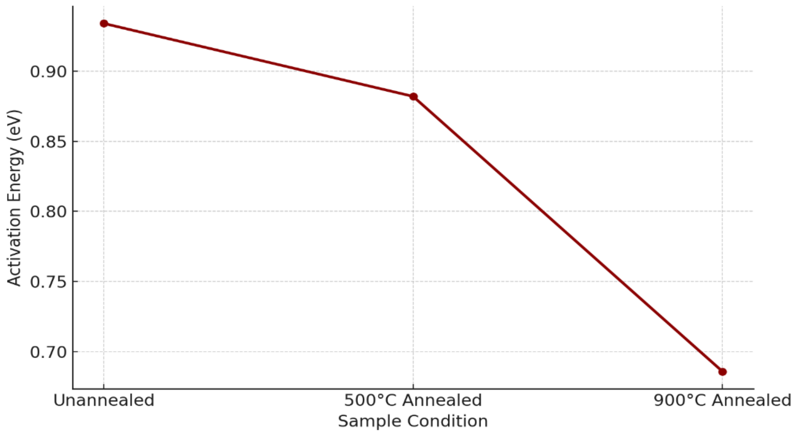

Figure 2: Variation of Activation Energy, $E(eV)$ with Annealing Temperature

These changes are attributed to high-temperature-induced defect reorganization, including the annealing out of deeper traps and the formation of new, energetically favorable recombination centres.
Such behavior is comparable with previous findings in annealed synthetic quartz and related silicate materials, where considerable changes in thermoluminescence (TL) properties have been attributed to trap rearrangement, as reported by Chithambo et al. (2011) and Galloway (2002).

Frequency Factors
The frequency factor (s), often linked with the attempt-to-escape frequency, serves as a critical parameter in characterizing the kinetics of charge carrier release from traps. For the unannealed sample, curve-fitting evaluation yields a frequency factor on the order of ~10^{13} s^{-1}, indicative of a relatively high escape probability. In comparison, the frequency factors for samples annealed at 500 °C and 900 °C are estimated to be approximately ~10^{12}.8 s^{-1} and ~10^{12}.2 s^{-1}, respectively, suggesting a slight reduction in escape dynamics likely due to thermally induced modifications in trap depth or structure.
The trend of decreasing s with increasing annealing temperature complements the observed reduction in activation energy. It implies that the likelihood of carrier release per unit time also diminishes after high-temperature treatments, which could result from reduced trap populations or structural reconfigurations within the lattice that alter phonon coupling mechanisms.

Order of Kinetics
The order of kinetics (b), obtained via curve fitting, are close to unity. The values for the three samples: unannealed, 500 °C-annealed, and 900 °C-annealed, are 1.12, 1.06, and 1.14 respectively. This consistency reinforces the classification of the TL process in synthetic quartz as predominantly first-order kinetics, even after thermal modification. First-order behavior suggests that trap occupancy does not significantly influence recombination probability, meaning each electron is equally likely to recombine regardless of trap concentration, a desirable trait for reproducibility in TL dosimetry.

Implications and Material Optimization
The unannealed and 500 °C-annealed samples retain higher activation energies and more stable traps, making them better candidates for long-term radiation recording. The 900 °C-annealing on the other hand, modifies the trap environment substantially, which could be leveraged for applications requiring quicker signal release or higher sensitivity at lower energies. The transition between 500 °C and 900 °C appears critical, marking a threshold where significant structural reconfigurations begin to dominate, likely involving irreversible changes to the defect population. However, a finer exploration between 600–850 °C and detailed spectroscopic analysis of defect types could yield deeper insights into trap evolution and allow for engineered luminescent properties.

CONCLUSION
The kinetic analysis demonstrates that annealing temperature profoundly influences the activation energy,

frequency factor, and trap dynamics of synthetic quartz. While the order of kinetics remains first-order across all conditions, suggesting stable recombination mechanics, the quantitative shifts in E and s highlight the potential of thermal treatments to tune TL performance for specific applications. Understanding and controlling these parameters is essential for optimizing synthetic quartz in dosimetric and photonic technologies. While this study provides valuable insights, certain limitations are worthy of note. For instance, the analysis focused only on the main peak of the TL with only two annealing temperatures studied between room temperature and 900°C. Moreover, the exact nature of defect modifications was not characterized spectroscopically. Dose response and fading characteristics for dosimetric applications may be a worthy future study.

REFERENCES

Agulló-lópez, F., Catlow, C. R. A., & Townsend, P. D., 1988: Point defects in materials. *Academic Press*.

Chithambo, M. L., 2004: Time-resolved luminescence from annealed synthetic quartz under 525nm pulsed green light stimulation. *Radiation Measurements*, 38(5-6), 553-555. https://doi.org/10.1016/j.radmeas.2004.02.014

Chithambo, M. L., 2014: A method for kinetic analysis and study of thermal quenching in thermoluminescence based on use of the area under an isothermal decay-curve. *Journal of Luminescence*, 151, 235-243. https://doi.org/10.1016/j.jlumin.2014.02.037

Chithambo, M. L., & Niyonzima, P., 2017: Radioluminescence of annealed synthetic quartz. *Radiation Measurements*, 106, 35-39. https://doi.org/10.1016/j.radmeas.2017.02.005

Chithambo, M. L., Sane, P., & Tuomisto, F., 2011: Positron and luminescence lifetimes in annealed synthetic quartz. *Radiation Measurements*, 46(3), 310-318. https://doi.org/10.1016/j.radmeas.2010.12.003

Chithambo, M. L., Seneza, C., & Kalita, J. M., 2017: Phototransferred thermoluminescence of Al_2O_3:C: Experimental results and empirical models. *Radiation Measurements*, 105, 7-16. https://doi.org/10.1016/j.radmeas.2017.08.009

Galloway, R. B., 2002: Luminescence lifetimes in quartz: Dependence on annealing temperature prior to beta irradiation. *Radiation Measurements*, 35(1), 67-77. https://doi.org/10.1016/s1350-4487(01)-00258-x

Kitis, G., Gomez-Ros, J. M., & Tuyn, J. W. N., 1998: Thermoluminescence glow-curve deconvolution functions for first, second and general orders of kinetics. *Journal of Physics D: Applied Physics*,31(19), 2636-2641. https://doi.org/10.1088/0022-3727/31/19/037

Mckeever, S. W. S., 1985: Thermoluminescence of solids. *Cambridge University Press*. https://doi.org/10.1017/CBO9780511564994

Pagonis, V., Kitis, G., & Furetta, C., 2006: Numerical and practical exercises in thermoluminescence. *Springer Science & Business Media*. https://doi.org/10.1007/0-387-30090-2

Yang, X. H., & McKeever, S. W. S., 1990: Point defects and the pre-dose effect in quartz. *Radiation Protection Dosimetry*, 33(1-4), 27-30. https://doi.org/10.1093/oxfordjournals.rpd.a080687