Isothermal decay analysis of quartz

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The analysis of isothermal decay in thermoluminescence (TL) provides critical insights into the trapping and recombination mechanisms governing luminescence in materials. In first-order kinetics, charge carriers are released from traps without significant retrapping, resulting in a simple exponential decay. However, real-world systems often exhibit deviations from idealized behavior due to competing pathways, retrapping efects, or higher-order kinetics. The linear relationship between ln(I) and time in frst-order kinetics facilitates straightforward determination of activation energy (E), making it a cornerstone of TL analysis. The inclusion of general-order kinetics expands the applicability of these studies by accommodating complex TL behaviors, particularly in fields such as dosimetry and geochronology, where precise trap characterization is vital for accurate dose reconstruction and age determination. By analyzing decay curves at varying temperatures, the resulting slope differences ln(I) versus time plots ofer insights into the underlying kinetic processes.

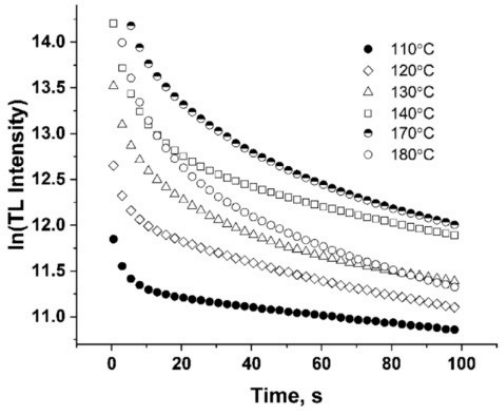


Fig.1 . Plot of the natural logarithm of TL intensity versus time at different Tstop temperature. Tstop temperatures are indicated in the legend

Figure 1 illustrates the time-dependent TL intensity decay at constant temperature for quartz samples preheated at 500 °C on a semi-log scale. Subsequently, samples were heated to a predefined Tstop temperature, followed by TL signal recording under isothermal conditions (100 s stabilization). For TL peaks governed by frst-order kinetics, isothermal decay follows an exponential trend, as described in. A linear dependency of ln (TL intensity) versus time plot confrms frst-order behavior, with the slope yielding−E/kT and the intercept corresponding to ln(s/β). Nonlinearity in such plots, however, indicates deviations from frst-order kinetics. The plots generated from the data in Fig. 1 demonstrated nonlinearity, indicating that the TL data does not conform to frst-order kinetics. The general-order kinetics equation suggests that plotting $(I_t / I_0)^{(1-b)/b}$ as a function of time should result in a straight-line relationship when an appropriate kinetic order parameter (b) is selected according to [2]:

$$(\frac{I(t)}{I_0})^{\frac{(1-b)}{b}} = 1 + (b-1)\frac{t}{\tau}b \neq 1$$

where I(t) is the intensity of the luminescence signal as a function of time; τ is the lifetime for isothermal decay; and b is the order of kinetics.

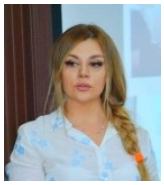
However, when the general-order kinetics approach was applied to the isothermal decay data in Fig. 1 using kinetic order parameters b=1.9,2.0,2.1, and 2.2, none of the tested values produced a statistically valid linear ft. This outcome highlights a fundamental challenge in analyzing isothermal decay data, as subtle graphical variations for different b values complicate the identification of an optimal kinetic order. The persistent lack of linearity across all tested b values strongly suggests that the TL decay does not conform to second-order kinetics. This conclusion was further reinforced by repeating the analysis on additional datasets in Fig. 1, all of which similarly failed to align with second order kinetic behavior.

To explore alternative explanations for the observed decay dynamics, the superposition of two or three exponential functions-each linked to distinct trapping centers-was evaluated [1]:

$$I_t = I_{01}exp(-P_1t) + I_{02}exp(-P_2t) + \dots$$

where Ion is the phosphorescence intensity due to electrons in the traps of energy En, Pn=s exp (- En/kT) is the probability of an electron escaping from a trap, k is the Boltzmann constant, and s is the escape frequency factor. A detailed decomposition of the decay curves into such components was attempted to resolve individual activation energies (E). However, the results revealed that the decay profiles cannot be accurately modeled as a sum of two or three first-order exponential terms, ruling out a simple superposition of first-order processes.

Subsequent efforts to deconvolute the curves using second order kinetic components also proved unsuccessful, further underscoring the complexity of the underlying trapping-



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recombination mechanisms. Intriguingly, the isothermal decay data were best described by combining two single exponential decay terms with a constant offset, a phenomenon previously reported in studies of natural quartz. For instance, successfully modeled similar isothermal decay profiles using three exponential components and a constant term.

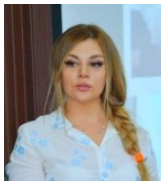
Collectively, the nonlinearity observed in Fig. 1 and analogous datasets in the literature implies that the TL signal arises from contributions of multiple trapping centers. The relative influence of these centers appears temperature dependent, leading to deviations from idealized exponential decay behavior. This complexity challenges conventional kinetic models and underscores the need for advanced analytical frameworks to account for heterogeneous trap distributions and dynamic recombination pathways.

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