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Study of dose response, luminescence kinetics, and reading modes combination advantage of TLD-100H radiation dosimeter

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► ***Short Report***

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ABSTRACT

Background: This study investigates various aspects of TLD-100H radiation dosimeter, including luminescence kinetics, dose response, and the effects of the combination of thermally stimulated (TL) and optically stimulated (OSL) luminescence modes. ***Materials and Methods:*** The TL kinetic parameters were determined using general order kinetics via computerized glow-curve deconvolution (GOK-CGCD). OSL decay curves and kinetics were also investigated through OSL signal decomposition. The dose response was studied for a practical dose range of 2-80 Gy. Interesting OSL and TL reading modes combination were also tested. ***Results:*** The identified dosimetric peak at ~560 K was deconvoluted into two main peaks (553K and 568K). The dose response study reveals a sublinear behaviour within the considered dose range. Two dominating types of fast and slowly decaying OSL traps were identified. Indicative values of de-trapping probabilities and photoexcitation cross sections of these OSL traps are thus provided. Results of TL and OSL reading modes combination demonstrate well that measuring TL signal before OSL resulted in almost complete erasure of the OSL signal. Additionally, an increase in TL integral intensity by ~+10% was observed when OSL measured firstly, likely due to the emptying of a limited number of lower temperature shallow traps by OSL. Indeed, these no longer applied a screen effect for the higher temperature traps, making their release more prominent in TL reading mode. Consequently, improving or optimizing the TL signal. ***Conclusions:*** These findings contribute to a better understanding of TLD-100H's behaviour and support its reliability as a radiation dosimeter in medical physics.

INTRODUCTION

Accurate measurements of ionizing radiation play a crucial role in various fields, such as medical diagnostics, radiation therapy, environmental monitoring, and space exploration. TLD-100 (LiF: Mg, Ti) and TLD-100H (LiF: Mg, Cu, P) thermoluminescent dosimeters have been used as a radiation dosimeter due to their reliable performances and wide dynamic range [(1)]. TLD-100 dosimeters are TLD100 are explicitly designed for medical physics and radiotherapy applications [(2-5)]. They show a linear response in the range of 10µGy to 10Gy with an annual fading not exceeding 5% [(1)]. TLD -100H presents the advantage of higher residual TL signal [(6)]. TLD-100H was originally developed by Nakajima *et al*. through the integration of Mg, Cu and P as dopants in the preparation process leading to a very sensitive LiF dosimeter [(7)]. LiF: Mg, Cu, P presents a very clear advantage in terms of sensitivity (30 times compared to LiF: Mg, Ti), thing which is particularly interesting given the low doses that it can measure in radiology application for example [(7)].

This study aims to investigate the luminescence kinetics of TLD-100H using general order kinetics (GOK) via computerized glow-curve deconvolution (CGCD) [(8-11)]. Additionally, the dose response of TLD-100H will be examined by analyzing the TL glow curves, while the optically stimulated luminescence (OSL) decay curves and their kinetics will also be investigated. Furthermore, the study will explore the very interesting effects of combining TL and OSL reading modes on TLD-100H, assessing the potential improvement or optimization of TL signal collection from deeper traps (residual signal). Combining Thermoluminescence (TL) and Optically Stimulated Luminescence (OSL) reading modes provides several advantages. In radiation dosimetry such reading modes combination leads to:

Improved Accuracy: With their different sensitivities, TL and OSL allow a more comprehensive assessment of radiation doses, leading to more accurate dose reconstructions.

Dose Rate Determination: TL and OSL can provide information not only about the total accumulated dose but also about the rate at which the dose was acquired by allowing understanding the dynamics of radiation fields in various environments.

Dose Reconstruction in Complex Environments: In environments with mixed radiation sources or varying radiation fields, combining TL and OSL readings can help disentangle the contributions from different sources and provide a more accurate reconstruction of the dose history.

Calibration and Validation: By comparing TL and OSL measurements accurate calibration is allowed that may enhance the reliability of TL&OSL dosimeters for use in various applications.

The comprehensive understanding of luminescence kinetics, dose response, and reading mode combinations will contribute to advancing radiation dosimetry techniques and reinforcing the reliability of TLD-100H as a dosimetric material.

MATERIAL AND METHODS

Dosimeter description

The study was performed on a square-shaped TLD -100H (LiF:Mg,Cu,P) from Freiberg Instruments GmbH company, Germany. The dimensions of the studied dosimeter are 3.2 mm × 3.2 mm × 0.9 mm. One of the advantages of such LiF based thermoluminescent (TL) material is its tissue-equivalent property allowing an effective use for dose measurement in radiation therapy and nuclear medicine.

TL and OSL measurement

The TL measurements were carried out by the Risø TL/OSL-DA-20 reader from Risø National Laboratory, Denmark. The used reader is equipped with a Varian VF-50J (50 kV/1 mA) X-ray unit with an average dose rate of 2 Gy/s and a bialkali EMI 9235QB PM tube with a maximum detection efficiency of 200–400nm. TL measurements are performed in the temperature region of 0-450 °C at a linear heating rate of 5 °C/s.

TL and OSL kinetics study

In this work, general order kinetics model with computerized glow-curve deconvolution (GOK-CGCG) are used for TL glow curve deconvolution and mains kinetics parameters extraction of the studied dosimeter [12,13]. The deconvolution of TL glow curve is performed using GOK based on model formulation given by equation (1) [13, 14].

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

Where; k is the Boltzmann constant, b is the order of kinetic T is the temperature, T_m is the maximum temperature of peak, E is the energy, $\Delta = \frac{2kT}{E}$, $\Delta_m = \frac{2kT_m}{E}$ and $1+(b-1)\Delta_m$. The necessary experimental data fittings were performed using Microsoft Excel software package available at Ankara University (Turkey) [15].

For the OSL kinetics study, the OSL signal will be decomposed into many components (OSL_i) and an OSL background (Bk) according to equation (2). Generally, the decomposition approach is applied according to the deconvolution results of the TL experimental dosimetry peaks.

$$I_{OSL}(t) = \sum_i OSL_i + Bk \quad (2)$$

TL and OSL reading modes combination

In this part of this work, reading sequences are programmed to test the effects of reading priority by alternating between TL and OSL. Thus, the tested sequences are: 1) X-ray Irradiation-TL-OSL and 2) X-ray Irradiation (same dose) -OSL-TL. TL signal is collected in the temperature interval of 0-450°C at a heating rate of 5°C/s (250 data-points). The OSL signal reading is done at 80% maximum light power (80Mw/cm^2) using blue LED (470 nm) for a stimulation time of 50s (100 data-points). X-ray irradiation was performed at a maximal energy of 50keV and a dose of 2Gy.

Statistical analysis

Errors of the determined physical value and parameters are corresponding to standard deviations obtained through three-time measurement repetition and uncertainty propagation. In TL glow curve deconvolution, the Figure of Merit (FOM) metric is used for statistical quality control of the deconvolution results [14]. Finally, in OSL kinetics study, the standard errors of the fitting functions are scaled with square root of the reduced Chi-squared (Chi-sqr).

RESULTS

GOK-CGCG TL deconvolution

Figure 1 shows the TL general-order kinetics deconvolution results of the 2 Gy X-ray irradiated TL glow curve of TLD-100H. The applied GOK-CGCD deconvolution decomposed the experimental TL glow curve into five (5) peaks. The deconvolution experimental dosimertic identified at ~560 K (286.5 °C) gives rise of two main peaks (peaks 4 and 5). The values of the peak positions (T_{max}), the activation energies (E_a), the kinetics orders (b), and the frequency factors (s^{-1}) for different deconvolution peaks are indicated in table 1.

OSL kinetics analysis

Based on TL kinetics study, the TLD-100H OSL signal is decomposed into two components, OSL_1 and OSL_2, by considering an OSL background signal (Bk). The used model is given by equation (3) [16,17].

$$I_{OSL}(t) = OSL_1 + OSL_2 + Bk = Bk + I_0(OSL_1)\exp\left(-\frac{t}{\tau_1}\right) + I_0(OSL_2)\exp\left(-\frac{t}{\tau_2}\right) \quad (3)$$

Where; I_0 is the initial OSL intensity and $\tau_i = 1/p_i$ is the lifetime of the luminescence for each considered TL peak (pi is the detrapping probability).

Figure 2 shows the OSL experimental data were after adjustment (fit) using fitting function given by equation 4 [17].

$$y = A_1 \exp\left(-\frac{x}{t1}\right) + A_2 \exp\left(-\frac{x}{t2}\right) + y_0 \quad (4)$$

The OSL kinetics study allows the determination of two interesting parameters: the electron de-trapping probabilities p_i ($p_i = 1/\tau_i$) and the photoionization cross-sections for the identified OSL_1 and OSL_2 components (traps). The de-trapping probabilities of traps are $4 s^{-1}$ for OSL1 and $6.06\times10^{-2} s^{-1}$ for OSL2. The determined photoionization cross-sections of fast (OSL1) and slowly (OSL2) decaying types of traps are 2.35×10^{-16} cm^2 and 3.57×10^{-18} cm^2.

Table 1. Kinetic parameters of main peaks of TLD-100H dosimeter.

	Peak #1	Peak #2	Peak #3	Peak #4	Peak #5
Tmax (K)	369±2	421±2	473±2	553±3	568±3
Ea (eV)	0.78±0.04	1.37±0.07	1.43±0.07	1.98±0.10	2.37±0.12
b	1.00000001	1.00000001	1.5	1.5	1.7
s^{-1}	1.33×10^{10}	1.15×10^{16}	6.31×10^{14}	3.67×10^{17}	4.64×10^{20}

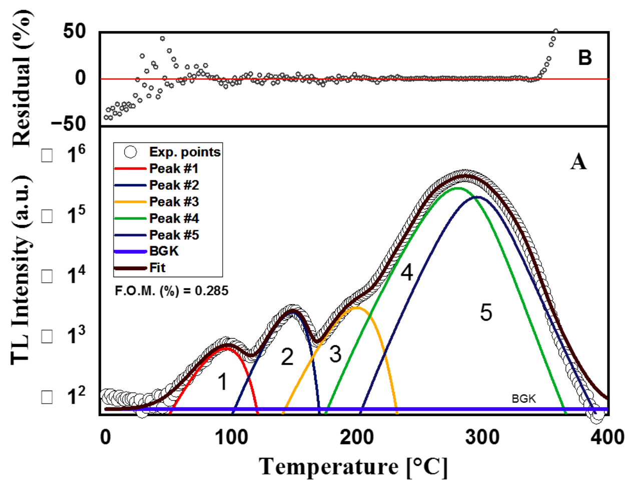

Figure 1. Computerized deconvolution (GOK-CGCD) results for TLD-100H. A) Semi-log graph showing the main dosimetric and fitting (deconvolution) peaks of TLD-100H when exposed to 2 Gy of 50 keV X-ray radiation. The experimental data are represented by open circles, while the fitting analysis is illustrated by the lines. B) Residuals data from the fit used to evaluate the deconvolution quality.

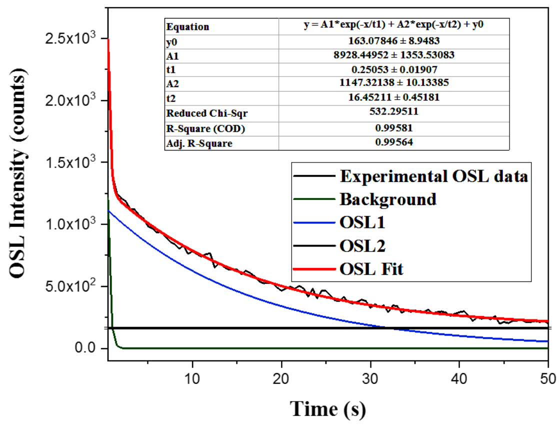

Figure 2. 2 Gy OSL curve decomposition into fast (OSL1) and slowly (OSL2) decaying types components. The data represent the fit of experimental signal by the proposed model. Model and fit function accuracy is evaluated using square root of the reduced Chi-squared (Chi-sqr) test.

TL glow curves and dose response of TLD100-H

The recorded TL glow curves associated for different X-ray doses are displayed in figure 3A. Corresponding integral TL intensities are plotted as a function of dose to illustrate the dose response of this dosimeter ihin the consdired dose interval of 0-80 Gy (figure 3B). Within the cosidred dose interval the dose reponse shows a slightly sublinear behaviour. When considreing low dose below 10 Gy, the dosimeter reppnse is quite linear.

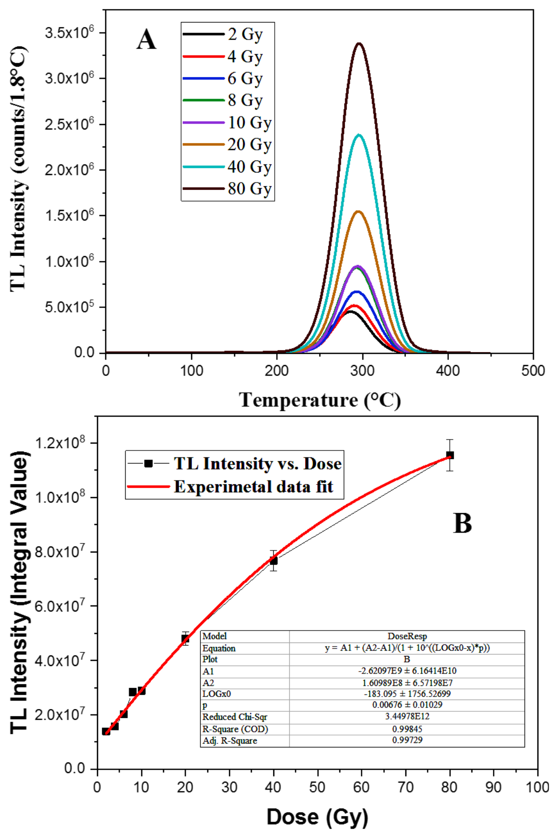

Figure 3. A) TL glow curves for different doses. **B)** Dose response of TLD-100H. The error bars on TL integral intensities represent the mean ± SD from three independent experiments.

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Importance of TL and OSL reading modes combination

The combination of the reading mode demonstrate well that these is an interest to read the OSL signal before the TL for one irradiation. Indeed, the TL integral collected is increased by almost 10% when the OSL signal is collected first. At the opposite, when TL signal is collected first the OSL signal decay completely to an insignificant and unusable level. This effect can be explained by the fact that when the OSL signal is collected first shallow traps are emptied. Thus, the electron shielding for de-trapping more deeply trapped electrons will be weaker allowing more ease in de-trapping such traps which are more populated than the shallow ones. This will ensure the observation of more intense TL peaks in the high-temperature range ensuring a greater TL signal. The optical stimulation (OSL) was found to be more effective in shallow traps empting when compared to low temperature stimulation (< 90°) by 71.70% when comparing the statistics of obtained TL signals before and after OSL reading (figure 4).

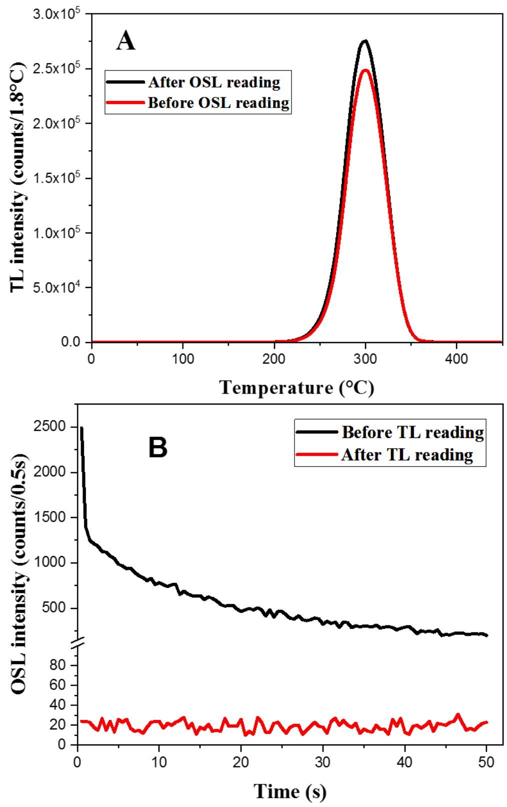

Figure 4. OSL and TL reading modes combination. **A)** Effect of OSL firstly readied on TL glow curve and intensity. **B)** Effect of TL firstly readied on OSL signal.

DISCUSSION

TL and OSL kinetics are essential aspects of TLD-100H dosimeters, as they determine how the dosimeter responds to radiation exposure and how the stored information can be read out. The TL glow curves of TLD-100H dosimeters is characteristic of the material and its impurities. Different peaks in the glow curve correspond to different energy levels of trapped electrons, allowing for discrimination between different types of radiation. The activation energy is a key parameter that influences the dosimeter's response. It determines the temperature at which the trapped electrons are released and the intensity of the emitted light. TL kinetics parameters extracted from GOK-CGCD TL glow curve deconvolution shows five mains peaks. The obtained value of TL kinetics parameters are depending on the specific manufacturing process, the purity of the material, and the irradiation conditions.

For the OSL kinetics study, our values of electron de-trapping probabilities and the photoionization cross-sections are close to those obtained by Jacob S. Nyemann *et al.* [18]. Thus, the probabilities and photoionization cross-sections of fast and slowly decaying types of traps are accurately determined. The photoionization cross-section varies with energy, so understanding these cross-sections is crucial for determining how different radiation types and energies interact with the dosimeter material. Accurate cross-section data helps in properly calibrating the dosimeter for the specific radiation type and energy encountered. In retrospective dosimetry, knowledge of electron de-trapping probabilities is vital. This information helps in understanding how quickly the trapped electrons are released upon exposure to stimulating light. Accurate de-trapping probabilities are necessary for reliable dose reconstruction.

Dose response results are in good agreement with works on similar dosimeter [19]. The dose response was found to be slightly sublinear in the considered dose range of 0-80 Gy but quiet linear for low doses. The behaviour of dose response at 0-10 Gy low dose range offers effective application of TLD-100H in such dose range that is generally used in medical application such as radiotherapy and radiology.

TL signal reading after OSL was found to be great interest because allowed keeping and to collecting the OSL signal and at the same time beneficiating from a more intense TL signal. Du *et al.* state that in the case of the studied luminescent material, the formation of deep-level traps contributed to the improvement of the stored energy, thus more energy could be released upon high temperature (> 200°C) external thermal stimulation [20]. Indeed, low temperature below 90°C or optical stimulation act as a fast-thermal-cleaning step to empty the relatively shallow traps [20]. In this work, optical stimulation was found to be more effective in shallow traps empting than low temperature thermal stimulation (T < 90°C)). Thus, the OSL&TL reading modes combination is of the most interest to improve TL signal intensity if reading is started firstly with OSL signal collection.

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CONCLUSION

An interesting feature of TL and OSL reading modes combination is the fact that corresponding light emission do not all originate from the same traps (shallow and deeper). Thus, it has been demonstrated that, even completely erased, the OSL signal if firstly collected has some benefit effect on the intensity of TL signal. Thus, the evaluation of dose based on the residual TL signal after OSL measurement gives more relevant results and additional security and documentation because two kinds of signal are recorded for the same irradiation. It should be noted that an insignificant OSL signal is left after heating the sample to 450 °C as it is the case in TL. The studied LiF: Cu, Mg, P dosimeter offers several advantages including an extended range of linearity at low doses (0-10 Gy), a human-tissue equivalence and a sensitivity to low energy photons allowing an effective use in clinical dosimetry.

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Conflicts of Interest: The authors declare that they have no conflicts of interest.

Authors' contributions: F.K. is the main investigator, directing experimental design and analysing results. A.M. performed the major of experiments and wrote the manuscript. C.Z.B. participated in experimental design and data collection. E.R. analysed obtained data and performed kinetics study. All authors read and approved the final manuscript**.**

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