Cumhuriyet Science Journal

Cumhuriyet Sci. J., 44(2) (2023) 389-395
DOI: https://doi.org/10.17776/csj.1226987

| csj.cumhuriyet.edu.tr | Founded: 2002 ISSN: 2587-2680 e-ISSN: 2587-246X Publisher: Sivas Cumhuriyet University

Investigation of Thermal Quenching Effect for Lithium Fluoride (LiF) Type Dosimeters

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Research Article

History
Received: 30/12/2022
Accepted: 10/06/2023

ABSTRACT

The thermal quenching effect was investigated for LiF:Mg,Ti (TLD-100), LiF:Mg,Cu,P (TLD-100H), ^{6}LiF:Mg,Ti(TLD-600) and ^{7}LiF:Mg,Ti (TLD-700) at two different doses (10, 1000mGy) using ^{90}Sr/^{90}Y beta source in this study. TLD-100, TLD-600 and TLD-700 showed different thermal quenching behavior according to dose values, while TLD-100H had the same characteristics for both doses. TLD-100, TLD-600, and TLD-700 showed thermal quenching based on the total area at 10mGy, while they did not show thermal quenching when ROI was used. All dosimeters were not affect thermal quenching at 1000mGy. In conclusion, it is recommended to use the lowest heating rate, to use the ROI, or to keep the same heating rate used in calibration and subsequent measurements at a low dose measurement (10mGy) for TLD-100, TLD-600, and TLD-700, while the desired heating rate can be used for high-dose measurement (1000mGy) for all LiF-type dosimeters.

Keywords: *Thermal quenching, LiF, Dosimetry, Thermoluminescence (TL), Heating rate.*

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Introduction

The thermal quenching phenomenon is defined as the decrease in luminescence efficiency with an increasing measurement temperature [1-3]. The luminescence intensity decreases with increasing heating rates in the presence of thermal quenching. In such a case, the heating rate to be used in the measurements becomes important because measurements taken at high heating rates may lead to erroneous dose assessments. Therefore, thermal quenching studies are of great importance not only for candidate dosimetric materials but also for the dosimeters in luminescence dosimetry research. The existence of a thermal quenching effect can be identified by monitoring the TL glow curves obtained at different heating rates under the same amount of radiation exposure [4]. Thermal quenching can also be understood with the change in the OSL signal obtained at different reading temperatures and luminescence lifetime measurements [5-9].

Lithium fluoride (LiF) type dosimeters have been widely used for radiation dosimetry for many years. They are chemically very strong materials due to properties having their resistance to corrosion and wear, and difficulty dissolving in water [2]. As well as its chemical advantages, the effective atomic number of LiF dosimeters is accepted to be tissue equivalent (effective atomic number of 8.31 compared to 7.35-7.65 for tissue), thus the dosimeters have a low energy response [10]. The LiF:Mg,Ti (TLD-100) and LiF:Mg,Cu,P (TLD-100H) dosimeters are used mainly in personnel dosimetry applications, while ^{6}LiF:Mg,Ti(TLD-600) and ^{7}LiF:Mg,Ti (TLD-700) dosimeters are used frequently in mixed fields neutron-gamma measurements due to having different neutron cross sections [2].

The thermal quenching effect for LiF-type dosimeters has been investigated by various researchers. The thermal quenching was investigated for TLD-100 irradiated with 3.5Gy by Taylor and Lilley [11]. The peaks 2 to 5 does not show thermal quenching according to this study. Pradhan and Bhatt [12] studied the influence of three different heating rates (4.5, 10, and 33°C/s) on TLD-100 irradiated with 0.9Gy and 9Gy. The results showed that all the peaks were not affected by thermal quenching. According to another study for TLD-100 performed by Caprile et al. [13], the variation in the area of dosimetric peak (peak 5) was investigated for four heating rates (7,10,15 and 25°C/s) for the dose of 0.2, 2 and 20cGy. No significant change was observed attributable to thermal quenching, although a slight decrease in intensity was observed. In a recent study by Singh and Kainth [14], TL intensity for TLD-100 irradiated with 186cGy was investigated for heating rates between 2 and 50°C/s. It was reported that the TL intensity decreased with increasing heating rate, indicating that TLD-100 undergoes thermal quenching in this study. However, when the literature was reviewed, TLD-100 does not show thermal quenching in most studies. For TLD-100H, the thermal quenching effect was investigated by Pradhan [15] for a heating rate between 1 and 20°C/s. Thermal quenching was not observed for TLD-100H according to this study. In another study on TLD-100H performed by Luo et al. [16], TL intensity was not change for heating rates between 1 and 30°C/s, thus the dosimeters did not show a thermal quenching effect.

The thermal quenching effect has been investigated by several researchers for TLD-700 [15,17-18]. Pradhan [15] and [17] studied the influence of heating rates on various TLDs including TLD-700, for heating rates ranging from 1 to 50°C/s. The variation in the area value of the low dosimetric peak (200°C) and high temperature peak (270°C) of TLD-700 was investigated according to increasing heating rates. As a result, the area of the dosimetric peak does not change, while the area of the high-temperature peak is affected by variations in the heating rates. Therefore, thermal quenching was substantial for the high-temperature peak, while thermal quenching was not observed at the low dosimetric peak. On the other hand, it was seen that the thermal quenching studies were quite limited for TLD-600 when the literature was reviewed. A recent study performed by İflazoglu et al. [18] studied the thermal quenching for both TLD-600 and TLD-700 dosimeters. In this study, both dosimeters were irradiated with 15Gy and the variation in TL intensities was investigated considering the total area under the glow curve for heating rates ranging from 1 to 10°C/s. According to the results of this study, both dosimeters were affected by thermal quenching.

Generally, the thermal quenching studies for TLD-100 and TLD-100H were quite common in the literature, while it was quite limited for TLD-600 and TLD-700, especially TLD-600. Additionally, these studies were generally conducted on a single irradiation dose value and the heating rates used were either with relatively low heating rates (~10°C/s) or using several heating rates without a full sweep of the range used. The study aimed to investigate in detail the existence of thermal quenching effects over a wide heating rate range (1 to 50°C/s) for the TLD-100, TLD-100H, TLD-600, and TLD-700 dosimeters from the LiF family. Additionally, the behavior of the thermal quenching effect (10, 1000mGy) dosimeters will be investigated for all at two different doses (10, 1000mGy)..

Materials and Methods

Material

In this study, LiF:Mg,Ti (TLD-100), LiF:Mg,Cu,P (TLD-100H), ^{6}LiF:Mg,Ti (TLD-600) and ^{7}LiF:Mg,Ti (TLD-700) dosimeters purchased from Harshaw Chemical Company, U.S.A were used. Their dimensions were the same with of 3.2×3.2×0.9mm^3 in the chip form. The TLD-100 and TLD-100H consist of two elements of ^{6}Li (7.4%) and ^{7}Li (92.6%), respectively. On the other hand, TLD-600 and TLD-700 have ^{6}Li (95.6%) and ^{7}Li (4.4%) and ^{6}Li (0.1%) and ^{7}Li (99.9%), respectively. Dosimeters were irradiated using a ^{90}Sr/^{90}Y beta source with two doses at 10 and 1000mGy. TL measurements were performed with a Harshaw TLD-3500 reader controlled with the WinREMS program and consisted of a glass filter, a photomultiplier tube (PMT) and a heating element (planchet). A thermo theldo furnace was used for the annealing and preheat process of the dosimeters.

Calibration steps & TL measurements

Dosimeters were firstly subjected to the annealing process before starting the measurement. Annealing &preheat temperature and their duration times were given in Table 1 for each type of dosimeter. The preheat process was applied in the machine for TLD-100H, while it was applied in the oven for other dosimeters.

Table 1. Annealing and preheat temperature & time

Dosimeter Type	Annealing temperature & time	Preheat temperature & time
TLD-100H	240°C+10min	135°C+10s
TLD-100, TLD-600, TLD-700	400°C+1hour followed 100°C+2hour	100°C+10 min

The TL measurement protocol was presented in Table 2. The readout time in Table 2 was given according to 1°C/s. In other cases, the total readout time was determined according to the heating rate value.

Table 2. TL measurement protocol

Dosimeter Type	Measurement temperature (°C)	Readout time (s)
TLD-100H	240	11
TLD-100	350	300
TLD-600, TLD-700	400	350

Dosimeters were calibrated separately for both doses since there might be differences in the intensity values since two different dose values were studied. The element correction coefficient (ECC), which considers differences in the intensity between the dosimeters was obtained separately for two dose values (10 and 1000mGy) by Equation 1.

$$ECC_i = \frac{I_i}{I_{mean}} \quad (1)$$

where the I_i and I_{mean} were the luminescence intensity obtained from each dosimeter and the mean luminescence intensity obtained according to all dosimeters used, respectively. The final luminescence intensity (I_f) was determined by multiplying I_i and ECC_i values for both dose values. Calibration measurements were carried out at the lowest heating rate (1°C/s). Therefore, the measurement protocol for calibration was the same as given in Table 2.

Investigation of the thermal quenching effect

The thermal quenching effect was studied for varying heating rates (1, 2, 4, 6, 8, 10, 20, 40,and 50°C/s). For TLD-100H only, heating rates from 1 to 10 °C/s were used because higher heating rates require dosimeters to heat exceeding 240°C. The existence of thermal quenching was determined by both the Region Of Interest (ROI) and total area value for TLD-100, TLD-600, and TLD-700 at a dose value of 10mGy. The ROI was obtained over three peaks in the dosimetric peak region, while the total area value was determined under the glow curve.

Results and Discussion

Variation in the TL glow curves

Figure 1a,b,c,d show the variation of the glow curves for TLD-100, TLD-100H, TLD-600 and TLD-700 obtained at different heating rates for the dose value of 10mGy. The maximum peak temperatures (Tmax) for TLD-100, TLD-100H, TLD-600 and TLD-700 at 1°C/s were obtained at ~205°C, ~201°C, ~204°C and ~205°C, respectively. The glow curves of all dosimeters shifted to the right, that is, the peaks were obtained at higher temperatures with increasing heating rate (1 to 50°C/s). The peak shapes in the glow curves did not change according to the increasing heating rate. The total variation in Tmax was obtained at ~106°C, ~102°C and ~101°C for TLD-100, TLD-600 and TLD-700, respectively when the heating rate increased from 1 to 50°C/s. As a result, the temperature shifts can be considered the same for all three dosimeters. On the other hand, the variation in Tmax for TLD-100H was ~39°C for heating rates between 1 and 10°C/s. Another observation was that an increase in luminescence intensity was observed in TLD-600 and TLD-700 after 350°C due to the predominance of blackbody radiation. This situation is related to due to the lower intensity of the peaks in the glow curve obtained at a lower dose (10mGy) for TLD-600 and TLD-700. As the irradiation dose value increased to 1000mGy, the peaks in the dosimetric region become more intense. Here, the increase seen after 350°C was not observed in the glow curve becoming a lower intensity, although it was still present.

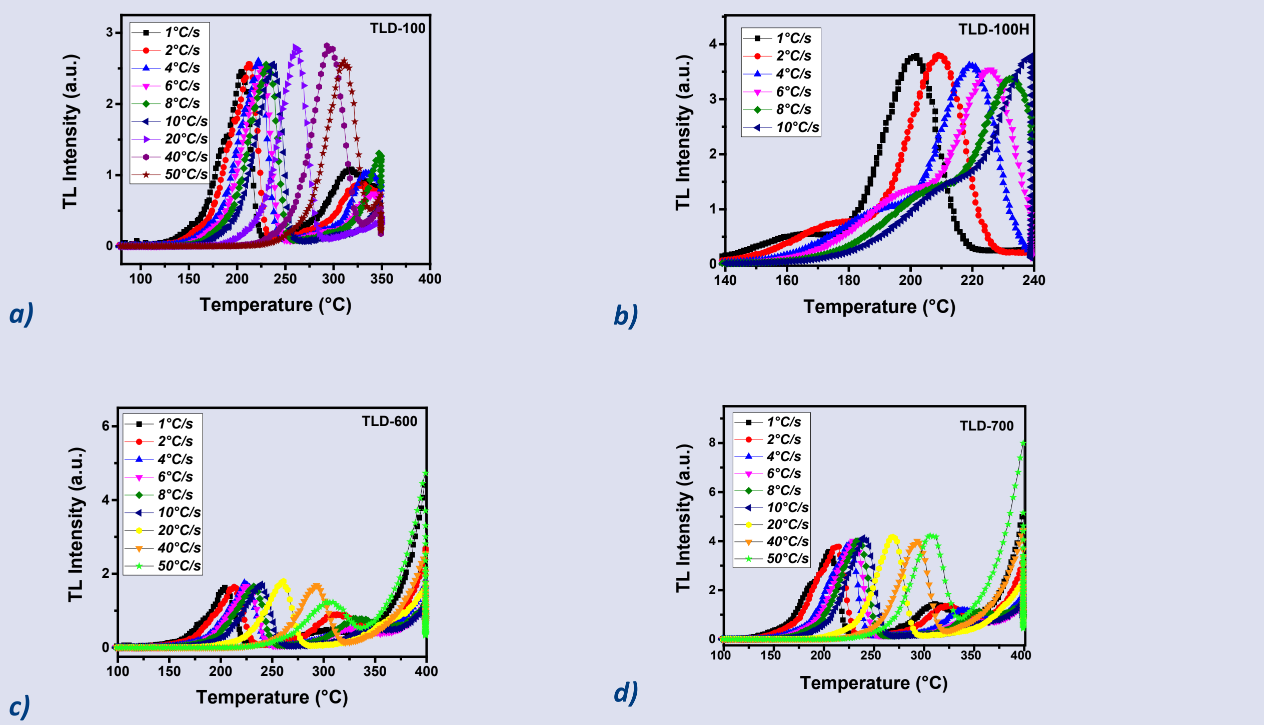

Figure 1. Variation of the glow curves for all dosimeters according to the increasing heating rate at 10mGy: a) TLD-100, b) TLD-100H, c) TLD-600, d) TLD-700

Figure 2a,b,c,d indicate the variation of the glow curves for TLD-100, 100H, TLD-600, and TLD-700 obtained at different heating rates for the dose value of 1000mGy. The Tmax of all dosimeters was around ~205°C. Therefore, the positions of the peaks did not change as the dose increased. Similar to Figure 1, the peaks shifted to higher temperatures as the heating rate increased for all dosimeters. The total variation in Tmax was observed ~110°C, ~108°C, ~102°C and ~37°C for TLD-100, TLD-600, TLD-700, and TLD-100H, respectively. Therefore, the temperature shifts in Tmax were unchanged compared to the dose value of 10mGy. Furthermore, the increase seen in high temperature for TLD-600 and TLD-700 in Figure 1c,d was not observed at high dose (1000mGy) in Figure 2c,d due to high luminescence intensity.

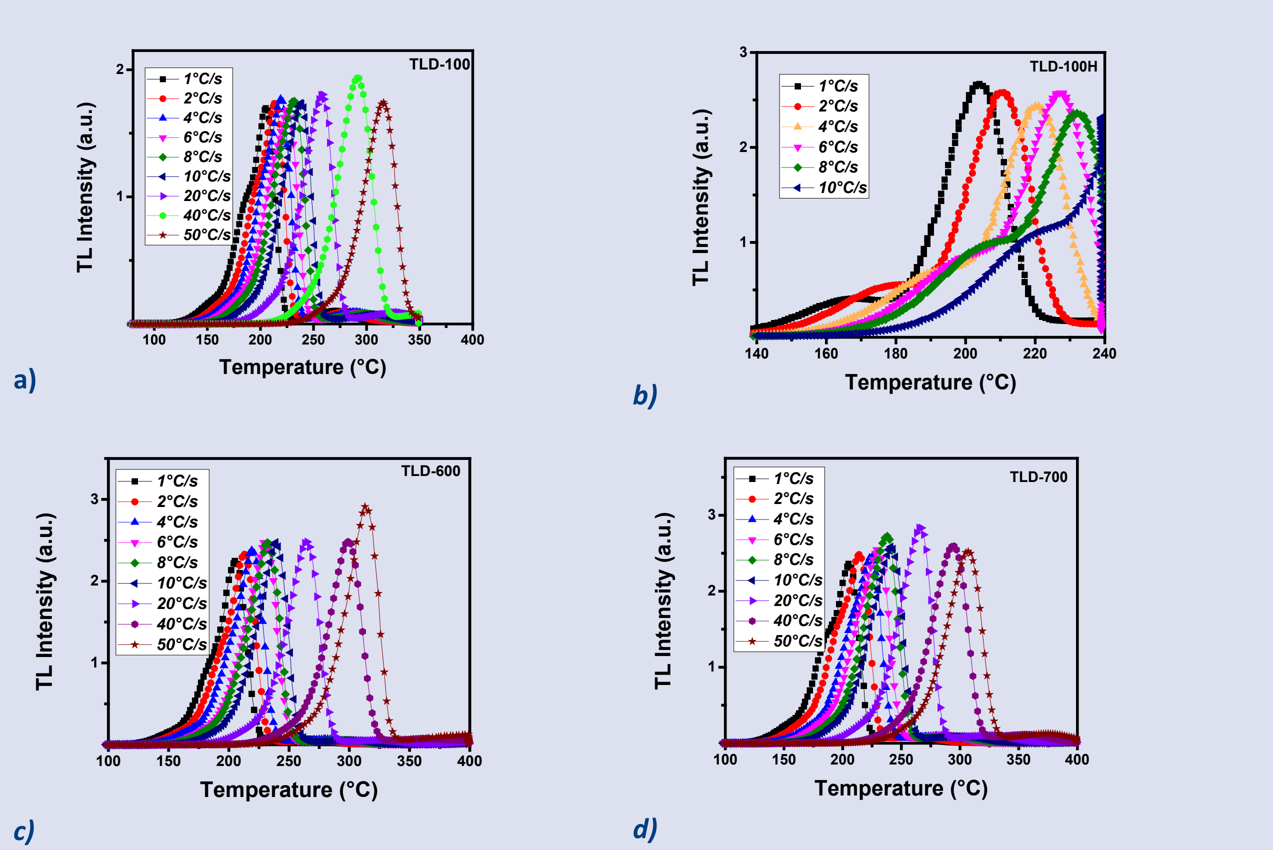

Figure 2. Variation of the glow curves for all dosimeters according to the increasing heating rate at 1000mGy: a) TLD-100, b) TLD-100H, c) TLD-600, d) TLD-700

Standard deviations in ECC values were also monitored for both dose values, taking into account the total area under the glow curve. Accordingly, the deviation in ECC values at 10mGy ranged between 10% and 15% for the TLD-100, TLD-600, and TLD-700, while it was <5% for TLD-100H. On the other hand, the deviation in ECC values obtained for 1000mGy was ~5% for each type of dosimeter. Therefore, the standard deviations in ECC values decreased significantly as the dose increased. These results showed the importance of selecting ROIs for TLD-100, TLD-600, and TLD-700 at 10mGy.

Thermal quenching results

Figure 3 indicates the variation in the area values obtained at different heating rates for all dosimeters at the 10mGy. In the figure, normalization was done according to the heating rate of 1°C/s. According to Figure 3a, TLD-100 shows thermal quenching considering the total area condition. The intensity obtained at 10°C/s showed a 30% reduction compared to 1°C/s. After 10°C/s, the area value behaved stable. Notable thermal quenching was observed in TLD-100 in the case of the total area condition. On the other hand, thermal quenching was not observed in TLD-100 when the ROI was used. As for TLD-100H, the total area value was around 1.00 for all heating rates used, thus thermal quenching was not observed (Figure 3b). According to Figure 3c,d, both TLD-600 and TLD-700 showed different behavior according to the total area and ROI similarly seen in TLD-100. Thermal quenching was observed for both dosimeters considering the total area, while thermal quenching was not seen when ROI was used. The intensity value obtained for TLD-600 at 10°C/s decreased by 35% according to 1°C/s, while no significant decrease was observed when considering the ROI. TLD-700 showed also similar behavior by showing a 35% reduction in the total area, again no thermal quenching was observed when ROI was used. On the other hand, a significant increase in intensity was observed for both TLD-600 and TLD-700 for heating rates of 40 and 50°C/s in the case of total area. This increase was thought to be due to the more pronounced increase in blackbody radiation with increasing heating rates in TLD-600 and TLD-700.

In conclusion, TLD-100, TLD-600, and TLD-700 have thermal quenching when considering the total area, while thermal quenching is not observed when ROI is used in the 10mGy. The reason for not seeing thermal quenching at 10mGy in the case of ROI could be related to dosimetric peak regions being more stable to changing heating rates than higher temperatures in the glow curve. In other words, higher peaks in TLD-100, TLD-600 and TLD-700 are more affected by thermal quenching. On the other hand, thermal quenching was not observed in TLD-100H.

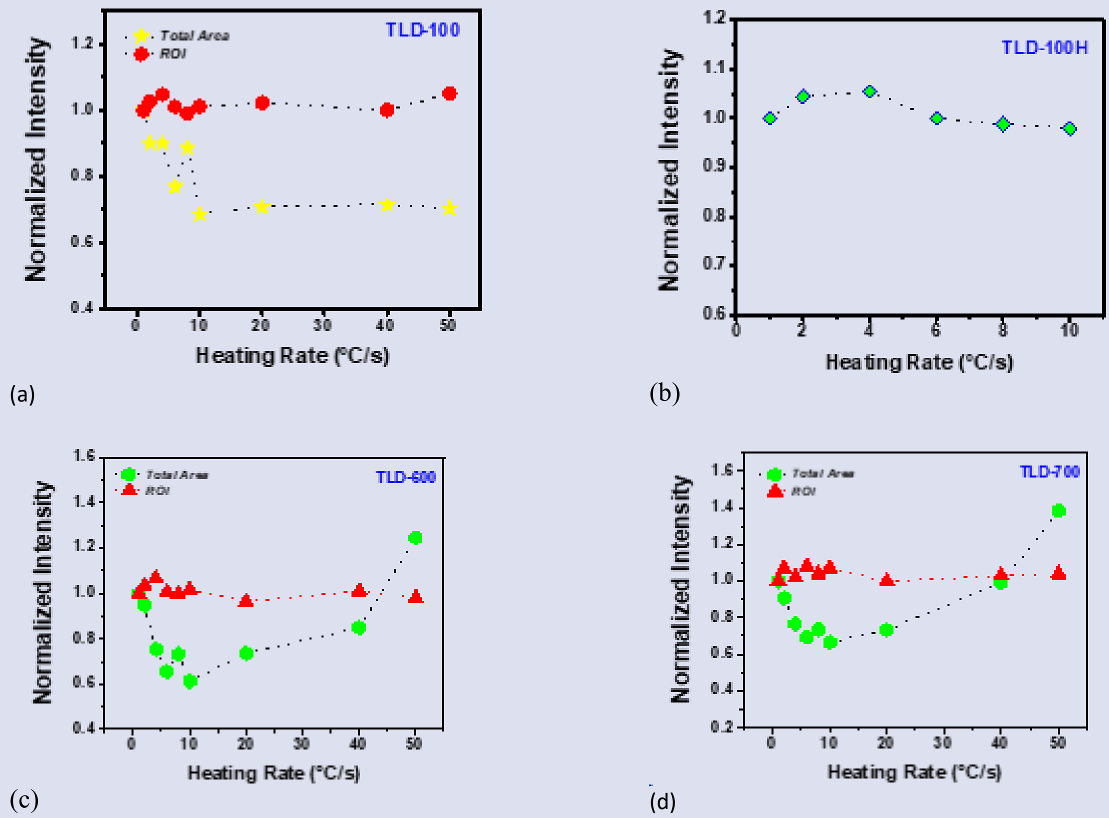

Figure 3. The variation of the area values according to the different heating rates for the 10mGy: a) TLD-100, b) TLD-100H, c) TLD-600, d) TLD-700.

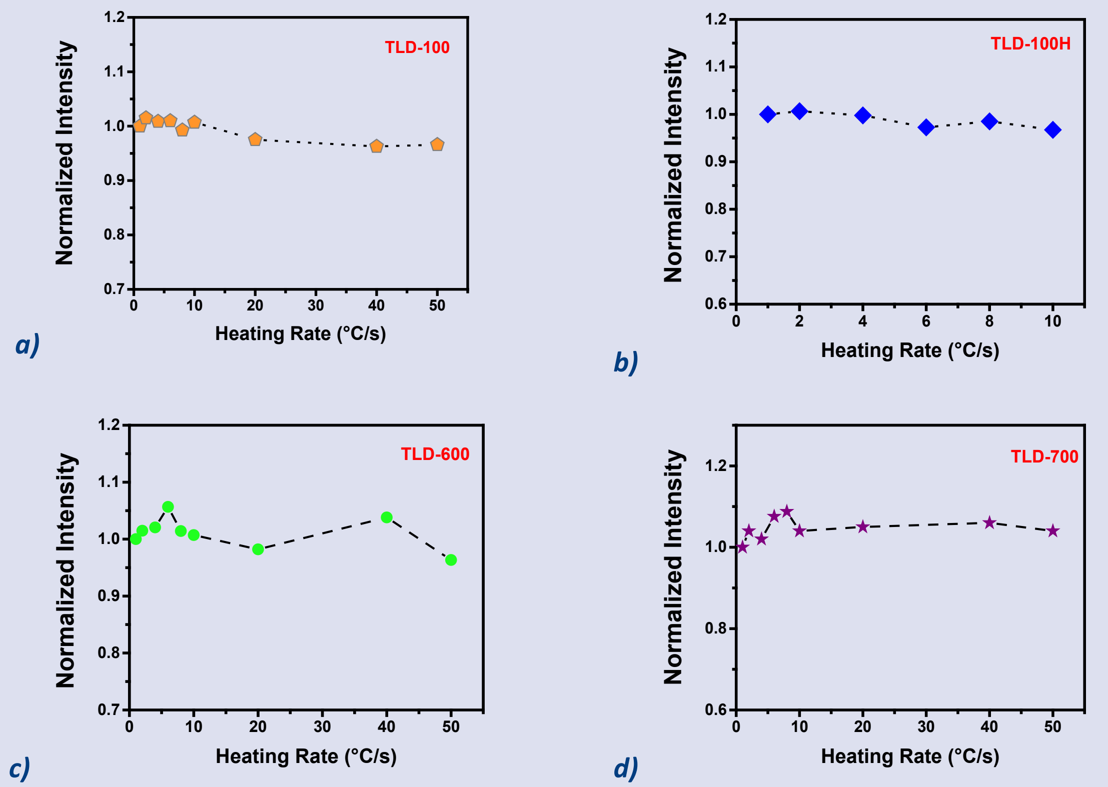

Figure 4. The variation of the area values obtained according to the different heating rates for the 1000mGy: a) TLD-100, b) TLD-100H, c) TLD-600, d) TLD-700.

Figure 4 shows the variation in the area values obtained at different heating rates of all dosimeters for 1000mGy. The normalization was performed according to 1°C/s. The area values varied from 0.95 to 1.00 for both TLD-100 and TLD-100H (Figure 4a,b). Although partial increases were observed in the initial heating rates for TLD-600 and TLD-700, the area values in general varied between 1.00 and 1.10 (Figure 4c,d). As a result, all dosimeters did not show thermal quenching effect at 1000mGy as opposed to 10mGy over the total area

Most thermal quenching studies for TLD-100 in the literature generally do not show thermal quenching for in the order of Gy [11-13]. In the study performed by Caprile et al. [13] in the order of mGy, the area of the peak did not change according to the heating rate, that there was no thermal quenching. Apart from this, there are rare studies reporting existence of thermal quenching in TLD-100 [14]. In our study, TLD-100 irradiated with 1000mGy did not show thermal quenching. Similarly, TLD-100 was not affected by thermal quenching at 10mGy when ROI was used. In this regard, the results obtained for TLD-100 were consistent with the literature of [11-13]. However, thermal quenching was observed only at the 10mGy when only the total area value was considered. The reason for thermal quenching observed in the total area condition at low dose is thought to be because the peaks after the dosimetric peak region are more affected by the increasing heating rates. TLD-100H was not shown thermal quenching for both 10 and 1000mGy. These results were compatible with studies of [15,16].

TLD-700 showed thermal quenching according to the studies of [15,17-18]. In our study, thermal quenching was observed only in the 10mGy over total area conditions for TLD-700. However, thermal quenching was not observed in both TLD-600 and TLD-700 at 1000mGy and using ROI. When the results obtained for TLD-600 and TLD-700 in our study are compared directly with the study of İflazoğlu et al. [18], thermal quenching was reported in İflazoğlu et al. [18], but no thermal quenching in our study in the order of Gy. As a conclusion, the findings obtained in the above mentioned studies [15,17-18] in the order of Gy do not seem compatible with those obtained in our study. The Tmax values for the main dosimetric peak for both TLD-600 and TLD-700 were reported at ~245°C for the heating rate of 1°C/s in these studies of [15,17-18]. However, Tmax values for the main dosimetric peak obtained in our study were obtained at ~205°C for both TLD-600 and TLD-700 for 1°C/s, which was almost same with that of TLD-100. The Tmax values were reported ~205°C for TLD-600 and TLD-700 by Oster et al. [19]. In another study, Tmax value was again ~205°C for TLD-600 by [20-21]. Therefore, the Tmax values obtained in our study for TLD-600 and TLD-700 were in agreement with the studies of [19-22]. The reason why thermal quenching is seen in the studies of [15,17-18] for TLD-600 and TLD-700, but not in our study may arise from previous use of the dosimeter depending on the thermal treatment, the readout process, irradiation and/or possible differences in the impurities during the production conditions. Therefore, these differences between the dosimeters could cause not only the change in the peak maximum temperatures but also change the behavior of the peaks under different heating rates by leading to the thermal quenching effect.

In a nutshell, TLD-100H did not show any change when considering the thermal quenching effect at low and high doses, while TLD-100, TLD-600, and TLD-700 showed differences according to the total area. This dose-related change may be related to the fact that thermal quenching is more effective at a low dose at high temperature peaks, and this effect decreases as the dose increases. In order to better understand this situation, additional experiments are required on subjects such as analyzing each peak in the glow curve doing deconvolution methods and investigating the effects of deep traps in dosimeters on the heating rates. According to the outcome of the study, it is recommended to use the lowest heating rate, to use the ROI, or to keep the same heating rate used in calibration and subsequent measurements for low dose measurement (10mGy). On the other hand, any heating rate can be preferred for high dose measurement (1000mGy) for all LiF-type dosimeters.

Conclusion

LiF-type dosimeters have been widely used for radiation dosimetry for many years. TLD-100 and TLD-100H are used mainly in personnel dosimetry, while TLD-600 and TLD-700 are preferred for the mixed fields neutron-gamma measurements. The decrease in intensity with increasing heating rate, namely thermal quenching, is of great importance for accurate dose evaluation. In this study, thermal quenching at two different dose values (10 and 1000mGy) was elaborately investigated for LiF-type dosimeters. Considering the total area values at a low dose (10mGy), thermal quenching was observed in all dosimeters except TLD-100H, whereas thermal quenching was not seen in any dosimeter at a high dose (1000mGy). According to the outcome of the study, it was recommended to use the lowest heating rate, to use the ROI, or to keep the same heating rate used in calibration and subsequent measurements for low-dose measurement (10mGy). On the other hand, all types of LiF dosimeters can be read with the desired heating rate at high-dose measurement (1000mGy), regardless of the total area and ROI.

Conflicts of interest

There are no conflicts of interest in this work.

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