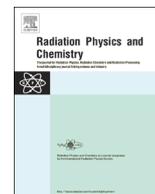




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An ultra-high dose of electron radiation response of Germanium Flat Fiber and TLD-100

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HIGHLIGHTS

- The supralinearity of GFF and TLD-100 was highly dependent on the UHD.
- The maximum supralinearity $f(D)_{max}$ of TLD-100, occurs around 10 kGy.
- TL kinetic model can be used to explain the glow peak behavior of doses $< f(D)_{max}$.
- The supralinearity response decreases for doses above the critical dose limit.

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ABSTRACT

The thermoluminescence (TL) response of Germanium Flat Fiber (GFF) and TLD-100 irradiated with 2.5 MeV electrons for the doses up to 1 MGy were studied and compared. The aim was to evaluate the TL supralinearity response at an ultra-high dose (UHD) range and to investigate the change in kinetic parameters of the glow peaks, as the doses increases up to 1 MGy. It is found that the critical dose limit (CDL) of GFF is 5 times higher as compared to TLD-100. CDL is determined by the dose at the maximum supralinearity, $f(D)_{max}$. It is also found that annealing the TLD-100 and GFF with temperature more than 400 °C is required to reset it back to its original condition, following radiation doses up to 1 MGy. It is also noticed the strange behavior of Peak 4 (TLD-100), which tends to be invisible at the lower dose (< 10 kGy) and starts to be appeared at the critical dose limit of 10 kGy. This result might be an important clue to understand the behavior of TLD-100 at extremely high dose range. For both samples, it is observed that the TL intensity is not saturated within the UHD range studied.

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1. Introduction

Thermoluminescence (TL) dosimetry at high dose radiation has attracted much attention as increasing needs in food safety, radiation protection on an extreme dose field and nuclear reactor dose monitoring. Therefore, it is crucial to reveal the performance of a dosimeter under circumstances of high dose deposition, and investigate the TL mechanisms, including the TL glow curve response and glow peak behavior, especially in the supralinearity dose response.

There are some findings from the Institute of Nuclear Physics (IFJ), Poland related to MTS-N which is equivalent to TLD-100 (IJF); (1) The TL response of TLD-100 (IJF) following high doses of ^{60}Co gamma rays revealed a new peak at the temperature of 420 °C for doses exceed 700 kGy by Khoury et al. (2011); (2) Bilski et al. (2010) found that two glow peaks dominated at 5 kGy with peak temperatures of 340 °C and 370 °C and saturated at 50 kGy; (3) A small peak was observed near 500 °C following irradiation of 20 kGy reported by Obryk et al. (2010) using 24 GeV/c protons. On the other hand, Montañó-García and Gamboa-deBuen (2006) investigated the response of TLD-100 (Bicron) to high doses of ^{60}Co gamma rays and observed a significant increase in TL intensity of the peaks 5 and 7 within the range of 2.5 kGy to 7 kGy and then TL decreases with further increases in dose. Glow curves were deconvoluted into single first-order peaks using GlowFit[®], a program developed by IFJ (Puchalska and Bilski, 2006).

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However, none of these works have adequately addressed the limitation of TL response at extremely high doses of electron radiation. Despite much excellent work on TL dosimetry such as dose linearity response and TL glow curve, scholars examining the TL behavior in the clinical dose range, have not yet fully explored the importance of supralinearity and critical dose limit, in an ultra-high dose radiation field. Yet, without such an understanding, we are left with an inadequate analysis that creates the condition for misinterpretation of the TL glow curve and main dosimetric peak evaluation and improper of kinetic model implementation.

This study will remedy this gap in the literature by examining the TL supralinearity response and determine the critical dose limit of GFF and TLD-100, in order to reveal more connections between the supralinearity behavior and the kinetic parameters of the specific high dose radiation field. The aim of this paper is to evaluate the TL glow curve response by using WinREMS[®] software and determine its kinetic parameters by using WinGCF[®] software deconvoluted glow peaks in the UHD region of interest.

2. Experimental procedure

The TLD-100 chips (3.0 mm × 3.0 mm × 0.89 mm) were obtained from the Harshaw-Bicron Company. The GFF samples used were fabricated with 6 wt% of Germanium dopant, by the Fiber Optic Research Center, Multimedia University, Malaysia. In this study, the samples were annealed using a Nabertherm Program Controlled S27 Furnace (Nabertherm[®], Germany) at 400 °C for 1 h. At the end of the heat treatments, the samples were quickly cooled in air to the room temperature (RT) by placing them on an aluminum block. The cooling rate is estimated at 16 °C per minute.

The samples were irradiated with 2.5 MeV electrons at doses ranging from 1 kGy to 1 MGy at RT (ALURTRON[®], Nuclear Malaysia Agency, Bangi, Malaysia). The TL yield as a function of temperature, referred to as the TL glow curve, were obtained by WinREMS[®], which is the operating system for the Harshaw[®] TLD reader, model 3500 (Thermo Fisher Scientific Inc, U.S.A). During readout the following parameters were used: preheat temperature of 60 °C for 5.0 s; acquisition temperature of 400 °C for $116\frac{2}{3}$ s and a linear heating rate of 3 °C s⁻¹. All readings were taken under N₂ gas flow, to suppress oxidation and creating an isothermal contact between the sample and planchet. The temperature distribution inside the sample is assumed to be homogeneous. All TLDs were subjected to the homogeneity test with the method as described by Alawiah et al. (2015).

Samples were placed at a distance of 100 cm from the source, and irradiated in air with a beam width of 60 cm and the dose ranging from 1 kGy to 1 MGy. The irradiation process was carried out at an ambient temperature using accelerating voltage of 2.5 MeV and the beam current of 1.0 mA. Before the irradiation, the beam output was verified according to the dosimetry protocols of TRS 398 (IAEA, 2000). The measurement was carried out using an electrometer (Wellhofer, Scanditronix) with the cavity volume of 0.65 cm³ (FC65-G) Farmer-type ion chamber. During the sample irradiation measurement, the delivered dose was separately measured by an ionization chamber to check the accuracy of the delivered dose.

Ten samples are allocated for one group of dose range and all samples were read for each TL measurement. A standard clean density glass filter was installed in the reader between the sample and photomultiplier tube, to eliminate unwanted infrared light emitted from the heater component. The samples were readout 24 h post-irradiation to minimize the low temperature peaks contribution in the glow curve. The glow curves were analyzed by a curve fitting computer program that is known as WinGCF[®]

software, which then produce the deconvoluted glow peaks for the kinetic parameters evaluation. WinGCF[®] is the commercial software for the TL spectra data analysis. The computerized glow curve deconvolution (CGCD) is an established and a powerful tool for analyzing the TL glow curves and WinGCF[®] is mainly based on the first order kinetics equation (Randall and Wilkins, 1945).

3. Results and discussion

The experimental glow curves by WinREMS[®] and the deconvoluted glow peaks by WinGCF[®] are presented. The uncertainty of TL response has been determined as ± 1 standard error of the mean and the coefficient variation did not exceed ± 5%, as required for radiotherapy clinical applications (ICRU 24, 1976) and the homogeneity of the samples are maintained. The TLDs also show good reproducibility with a standard deviation less than 3.0%.

3.1. WinREMS glow curve analysis

As the TLD reader temperature continues to rise, the TL intensity increases until such time as the population of trapped electrons in the metastable state is sufficiently depleted at which point the TL intensity decreases with further increase in temperature. This will produce the characteristic of TL glow curve, for which consisting of the luminescence peaks in a plot of luminescence intensity versus temperature, as was illustrated schematically by WinREMS software.

As seen in Figs. 1 and 2, the experimental glow curve of TLD-100 consists of five peaks, whereas, there is only one peak for GFF, which are constructed by WinREMS application originally from the accumulated TL signal by the TLD reader.

The analyzed glow curves by WinREMS revealed that there are some significant changes in the TLD-100 glow-curve shape with increasing dose in the range of 1 kGy to 5 kGy. As shown in Fig. 1, there are only 4 glow peaks (glow peaks 5, 6, 7 and 8) visible in the overall temperature range of 60–400 °C. Glow peak 4 was not visible within the UHD dose range of 1 kGy to 5 kGy. All glow peak height showed high dependency on UHD by a factor of ~2.5 to 3. While, the glow curve of GFF remains constant, with one main peak at the temperature range of 250–350 °C, as the dose increases from 1 to 5 kGy.

As shown in Fig. 2, the GFF glow curve was found to be constant in shape, within this dose range. The main peak is located within the temperature range of 250–350 °C.

The TLD-100 of glow peak 4 starts to be visible at 10 kGy in the temperature range of 150–250 °C. Further increased of doses above 10 kGy, resulted on peak 5 to be diminished at a dose of 40 kGy (see Fig. 2) and clearly observed the appearance of the new high temperature peaks 7 and 8 in the temperature range of 250–400 °C.

Slight reduction in peak height was observed in glow peaks 5 and 6 while glow peaks 7 and 8 showed a significant increase in peak height by a factor of ~2. Interestingly, as the dose was increased to 40 kGy, glow peaks 5 and 6 were diminished while glow peaks 4, 7 and 8, remain visible, with a slight increase in their peak height. As shown in Figs. 1 and 2, glow peak 5 appears within the dose range of 1–10 kGy before being diminished with further increase of dose. The peak height of peak 5 was found to increase with increasing dose from 1 kGy to 4 kGy by 40% and then decrease by 43% with further increase of the dose up to 10 kGy.

This result was found to be in agreement with other research about peak height response with UHD which was done by Montaño-García and Gamboa-deBuen (2006); they have evaluated the response of TLD-100 to high doses of ⁶⁰Co gamma rays up to

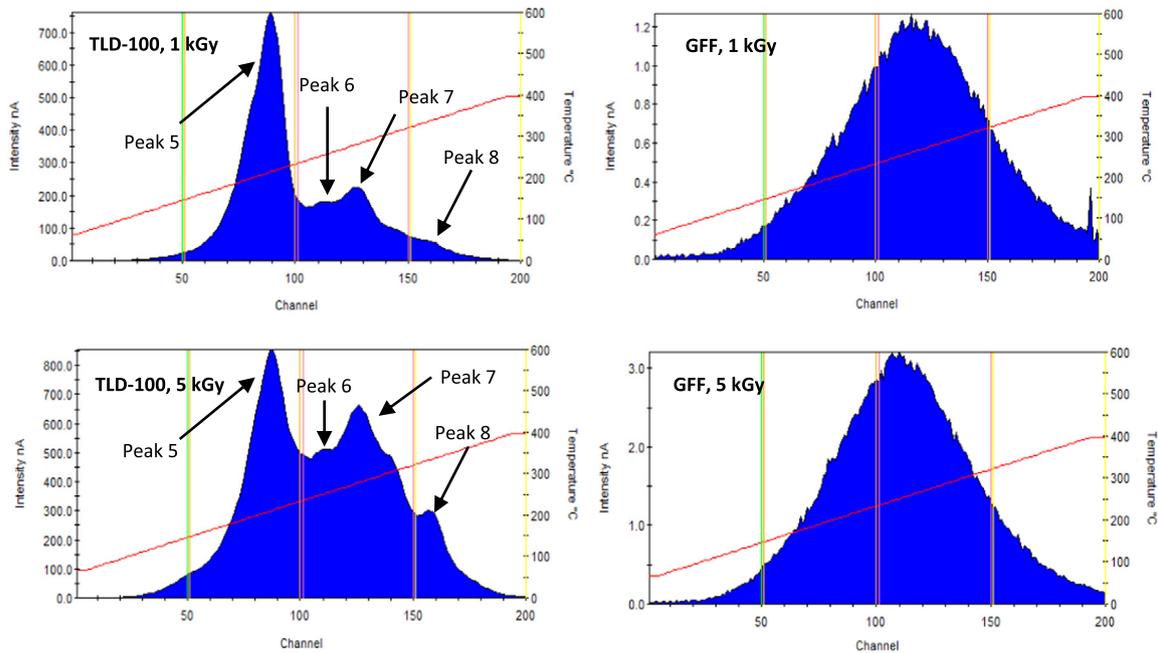


Fig. 1. WinREMS glow curve of TLD-100 and GFF, following 2.5 MeV electron irradiation at doses of 1 kGy and 5 kGy.

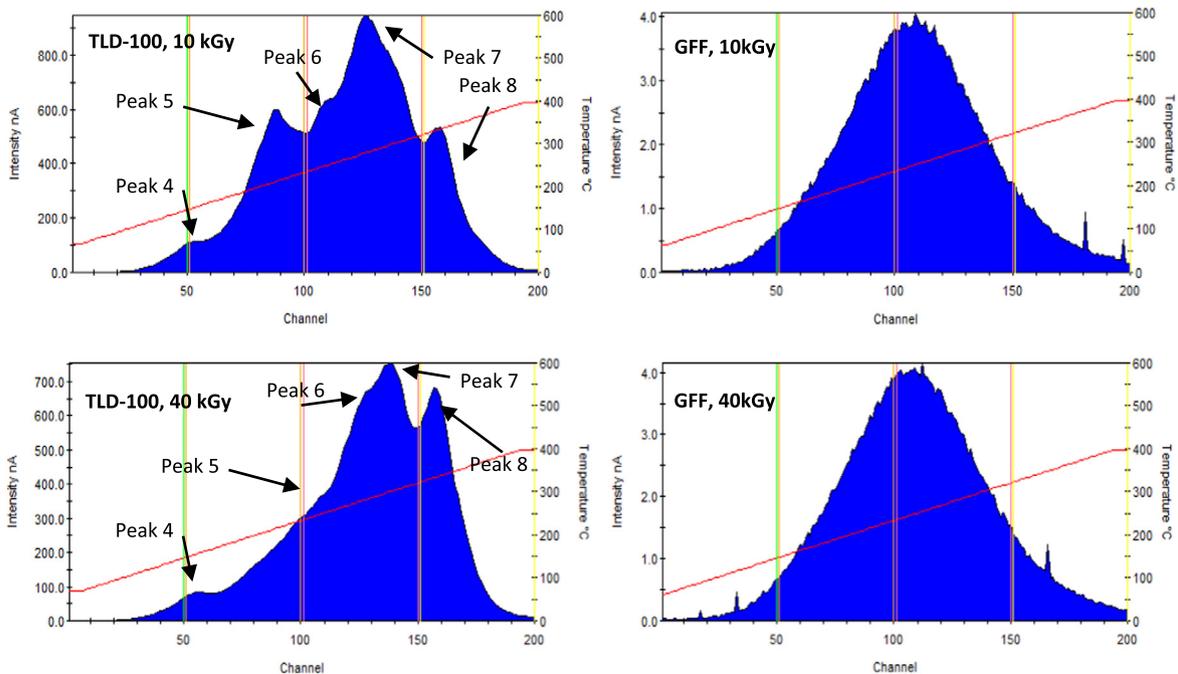


Fig. 2. WinREMS glow curve of TLD-100 and GFF following 2.5 MeV electron irradiation after the doses of 10 kGy and 40 kGy.

10 kGy. They observed a significant increase of the peak height of peaks 5 and 7, at the dose of 2.5 kGy and to 7 kGy, respectively, and then decrease. In addition, [Khoury et al. \(2011\)](#) found that an increase in dose would cause the TL intensity (peak height) of main dosimetric peak at 218 °C, decreases as the dose increases above 1 kGy. Peak 6 was located within the temperature range of 250–350 °C, which is between peaks 5 and 7 in the glow curve structured by WinREMS, as shown in [Fig. 1](#). The appearances of peak 6 can only be visible from the dose range of 1–10 kGy and being diminished following dose > 10 kGy.

Glow peak 7 diminished after 100 kGy of UHD whereas glow peaks 4 and 8 remained in the glow curve for doses up to 500 kGy, as shown in [Fig. 3](#). As the dose increases, peaks 5 and 6, tend to

merge with peaks 7 and 8 (in the temperature range of 250–400 °C), which then producing more widely glow curves with lower value of peak height by 20%. The appearance of peaks 7 and 8 is clearly dominant as the dose further increase above 100 kGy.

As shown in [Fig. 4](#), the glow curve of GFF remains constant in shape, at 1 MGy. The shapes of the glow curves are not significantly changed as a result of increasing dose within the range of UHD. The intensities of all glow curves increase with increasing dose up to 50 kGy and then decreases by 40% of its original value, with further increasing in dose up to 1 MGy. As for TLD-100, peak 4 and peak 8, remain in the glow curve as the dose reached its maximum at 1 MGy.

[Fig. 5](#) shows that the normalized TL intensity of TLD-100 increases with increasing dose starts from 1 kGy to 10 kGy and then decreases

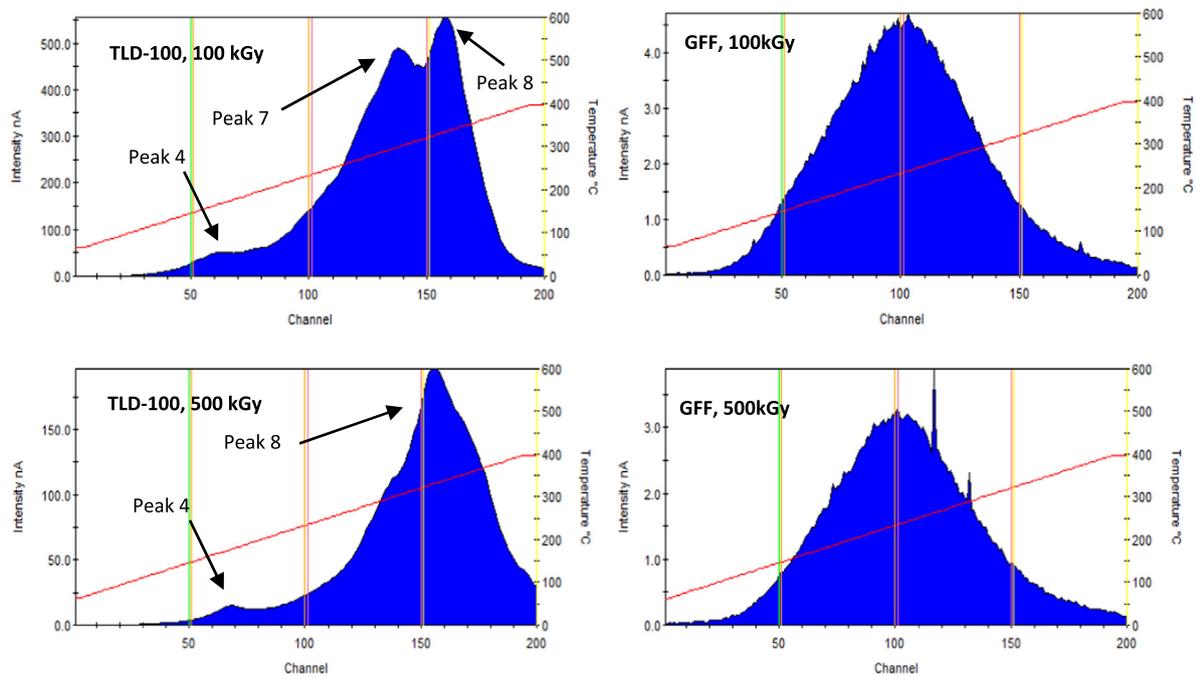


Fig. 3. WinREMS glow curve of TLD-100 and GFF, following 2.5 MeV electron irradiation at doses of 100 kGy and 500 kGy.

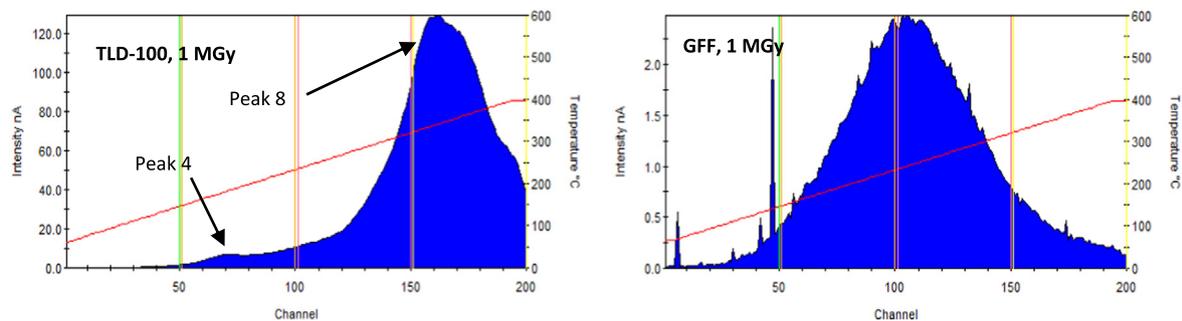


Fig. 4. WinREMS glow curve of TLD-100 and GFF, following 2.5 MeV electron irradiation at doses of 1 MGy.

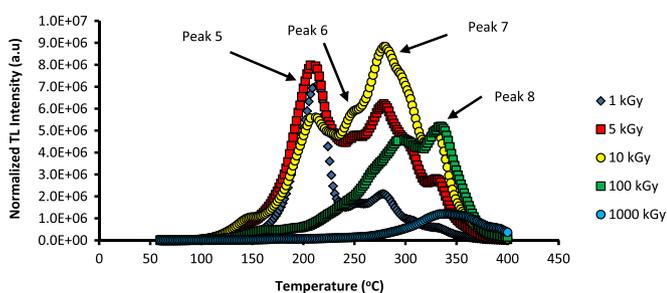


Fig. 5. The variation of TL glow curve of TLD-100 as a function of temperature, following electron irradiation from 1 kGy to 1 MGy.

with dose starts from 100 kGy to 1 MGy. Peaks 7 and 8, tend to be shifted to a higher temperature region, within the range of 280 °C to 305 °C for peak 7 and 330 °C to 340 °C for peak 8. This result indicates that trapping of electrons at the deep seated traps with higher range of the activation energy of traps. The higher activation energy results in shifting of TL peaks to higher temperatures. These electrons remain trapped until enough thermal energy is available to help them escape and recombine at a luminescent center.

It is also notable, that at lower doses no peak is present in this high temperature range, 270–350 °C. These findings are consistent with the previous results for gamma-ray exposures (Bilski et al., 2008), as well as for protons (Obryk et al., 2010).

Our results were also in agreement with Horowitz et al. (2008), stated that the high temperature peak (HTP) of TLD-100, appears at temperatures above the main dosimetric peak located at ~ 205 °C at heating rates of ~ 1.2 K s $^{-1}$. The maximum intensity of HTP appears at ~ 270 °C and is referred as peak 7 (Horowitz et al., 2008).

Horowitz et al. (2008) also found that at a very high dose of gamma/electron irradiation, there is a continuum of TL between peaks 5 and 7 which indicates the presence of at least one additional peak, which is referred to as peak 6. Above peak 7 in temperature, additional peaks appear which have been labeled as peaks 8–10.

It is usually assumed that there is no glow peak beyond 400 °C since it has also been assumed in the literature that annealing at 400 °C for 1 h totally resets the material, emptying all the traps and recombination centers and returns the material to its original/initial status.

Bilski et al. (2008) found a new peak of temperature exceeds 400 °C when the TLD 100H was exposed to the electron irradiation above 50 kGy. The glow curve shape was found to be constant at doses ranging from μ Gy to kGy. They also found that the sensitivity loss as the dose increased.

Horowitz et al. (2008) found that at high dose irradiation, they observed the appearance of TL peaks beyond 400 °C. In addition, a small peak was observed near 500 °C starting at 20 kGy. Its

presence was also reported by Bilski et al. (2010) after irradiation with electrons and by Obryk et al. (2010) after proton irradiation. Khoury et al. (2011) also observed the appearance of a new peak at 420 °C for the dose range of 700 kGy to 1200 kGy of gamma irradiation.

These findings are found to be contradicted with this study. It is found that, as the dose increases, the high temperature peak (peak 7 and peak 8) appeared and become dominant peaks at the temperature range of 280 °C to 340 °C which was not exceeded 400 °C, as shown in Fig. 7. The peak height of peaks 7 and 8 show a significant decrease with increasing dose. The peak 4 of TLD-100 starts to be noticeable at 10 kGy and TL signal tends to decrease with dose up to 1 MGy.

In addition, these results are to have good agreement with Khoury et al. (2011) findings on high temperature peak. Khoury et al. (2011) stated that the peaks located at higher temperatures of 317 °C and 350 °C of peaks 8 and 9 respectively, which become dominant peaks of the glow curve as the dose increasing. Using gamma irradiation of high dose on TLD-100, Khoury et al. (2011) found that peak height near 315 °C decreases as the dose increases, and the peak at 350 °C is dominant up to 300 kGy (Khoury et al., 2011).

It is apparent from this study that increasing the dose results in the growth of the high temperature peaks (HTP). The HTP were peaks 6, 7 and 8 with the temperature region at 250 °C, 280 °C and 330 °C, respectively. As the dose increases up to 20 kGy, peak 6 merged with peak 7 and peak 8, which then producing the maximum TL intensity of peaks 7 and 8 at that dose region. Further increase of dose up to 1 MGy caused a significant decrease in TL intensity of 5 times.

Bilski et al. (2010) observed a significant change in the color of the TLDs which at the dose of 46 kGy, the TLDs became brownish while at 1 MGy the color changed to dark brown. After readouts, the TLDs returned to their original initial colors. In this research, we also observed a significant color changed from opaque white to light yellow, as the dose increases from 1 kGy to 10 kGy, and then the color changed to darker yellow as the dose increases from 10 kGy to 100 kGy. Further increases in dose up to 1 MGy resulted in brown color TLDs. We also found that our TLDs showed permanent changes in its color when the dose of irradiation exceeded 10 kGy. For that reason, within this work all TLDs were used only once.

It is also found that these results are consistent with Bilski et al. (2010) who stated that the position of the high temperature peak shifts with increasing dose towards higher temperatures, which is about 50 °C from 75 kGy to 1 MGy (Bilski et al., 2010). The shape of the peak was very well fitted with a first order kinetics function, with the activation energy, $E \sim 4$ eV. These results, which were found consistently in all the studied batches, are in a good agreement with those described and discussed in the previous report (Bilski et al., 2008).

The finding seems to be contradicted with Bilski et al. (2010) who used 10 MeV electron exposures and found that TLD-100 exhibited a visible peak at 450 °C and the shape of glow curves is very similar in all cases. Since, the maximum temperature for peak 8 was found around the temperature range of 330 °C to 340 °C, which is far from their findings at 450 °C of the high temperature peak. Another remarkable finding from this research is that the shape of the glow curve showed significant changes with increasing dose, which again contradict with Bilski et al. (2010) who said about similar glow curve shape in all cases.

In case of TLD-100 the glow-curve after dose of 1 MGy is different since it is dominated by two peaks located at about 340 °C and 370 °C. There is also visible a small peak located at 480 °C (Bilski et al., 2010). These previous findings are not consistent with the observation on the glow curve of dose 1 MGy, showed in this

study as one single dominant peak at 340 °C and no other peak visible after that temperature.

A much smaller shift between 15 and 20 °C also noticed for the TLD-100 ultra-high temperature peak for doses above 200 kGy (Bilski et al., 2010). This result was consistent with our findings; peak 8 was shifted to smaller temperature difference of 8 °C for the doses above 250 kGy. This is due to the effect of thermally disconnected traps at the dose higher than 200 kGy.

For doses lower than 10 kGy, almost the whole glow curve appears below 250 °C. While above 10 kGy, the peak temperature of a region between 270 and 350 °C dominates. It is found that peak 7 (280 °C) can be used for dosimetry starting from 1 kGy and before it diminished above 250 kGy. Peak 8 (330 °C) starts to be useful at about 2 kGy and it will not be saturated for the dose up to 1 MGy. We also observed a strange behavior of peak 4 (150 °C) which starts to be visible at about 10 kGy and will remain visible, of the dose up to 1 MGy. In this way the whole range from 1 kGy to 1 MGy is covered.

3.2. WinGCF deconvoluted peak analysis

As seen in Fig. 6, there are 5 deconvoluted peaks, which denoted as peaks 4, 5, 6, 7 and 8. It is observed, as the dose increases from 1 kGy to 10 kGy, E_a decreases within 15–48% (except for peaks 6 and 8). All peaks were shifted to the lower temperature region by 1.4–12% and the peak integral of all peaks increased up to 10 times of its initial value. As the dose was further increased from 100 kGy to 1 MGy, E_a is observed to increase by $\sim 70\%$ with dose (except for peaks 6 and 8), T_{max} is increased by ~ 2.9 –4.4% (except for peak 5) and peak integral shows a significant decrease by 10 times, as shown in Figs. 7 and 8.

Considering the simple model with and without the effect of thermally disconnected traps, the discussion is based on TL glow curve shape and behavior of the TLD-100 samples. WinREMS provided the experimental data of glow curve and then extracted and analyzed by WinGCF for determining the kinetic parameters associated with the charge transfer process in the samples. These parameters include the activation energy, E_a , maximum peak temperature, T_{max} and peak integral, PI .

The two assumptions are normally employed includes: (1) that the free carrier concentration in the conduction band is always very much less than the trapped carrier concentration, and (2) that the rate of change of the free carrier concentration is always very much less than the rate of change of the trapped carrier concentration. According to the criteria of Randall and Wilkins (1945) the TL models yield the first order kinetics Eq. (1) if the probability of retrapping is negligible compared with the probability of recombination.

$$I(T) = -\frac{1}{\beta} \frac{dn}{dt} = n_0 \frac{s}{\beta} \exp\left\{-\frac{E}{kT}\right\} \times \exp\left\{-\frac{s}{\beta} \int_{T_0}^T \exp\left\{-\frac{E}{kT'}\right\} dT'\right\} \quad (1)$$

Eq. (1) is the Randall and Wilkins first-order kinetic expression of a single glow peak.

Based on this model regarding the variation of n_0 , it can be noted that as the initial concentration, n_0 , increases, the peak height increases but the maximum peak temperature remain constant (Chen and McKeever, 1997). The peak position stays fixed, while the height of peak is directly proportional with n_0 . This is an important characteristic of all first-order TL glow curves. In the application of dosimetry, n_0 is the parameter that is proportional to the absorbed dose. And the area under the glow peak is equal to n_0 since n_∞ is zero for $t \rightarrow \infty$, see Eq. (2) (Bos et al., 2001).

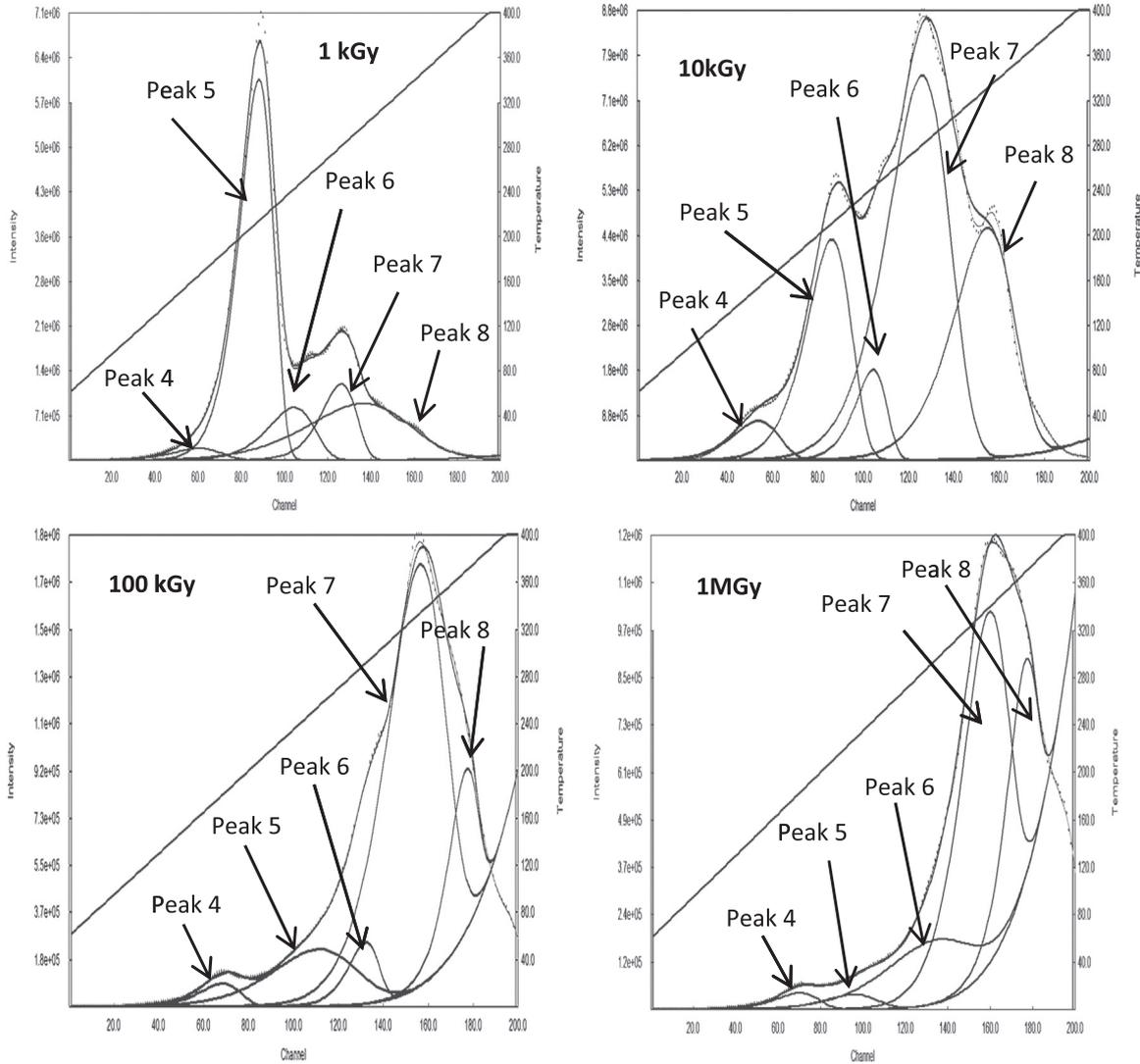


Fig. 6. A WinGCF deconvolved glow peaks of TLD-100 for the ultra-high dose of 1 kGy, 10 kGy, 100 kGy and 1 MGy, electron radiation 2.5 MeV.

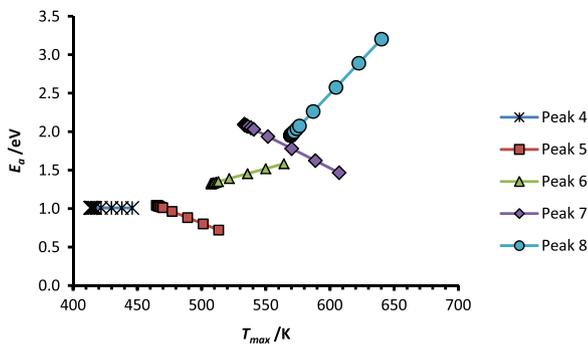


Fig. 7. The kinetic parameter variation of E_a on T_{max} of TLD-100.

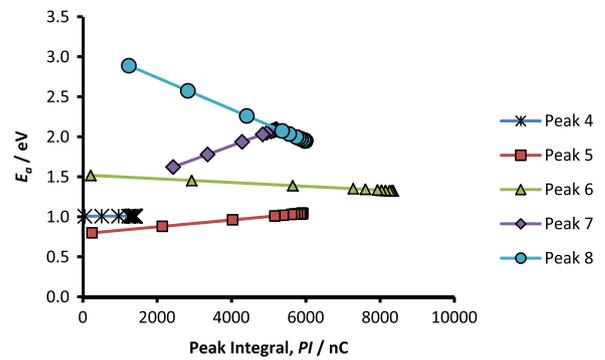


Fig. 8. The kinetic parameter variation of E_a on PI of TLD-100.

$$\int_0^\infty I(t)dt = - \int_0^\infty \frac{dn}{dt} dt = - \int_{n_0}^{n_\infty} dn = n_0 - n_\infty \quad (2)$$

Also stated in the first order kinetic, that as the activation energy increases, the peak position shifts to much higher temperature region with decreasing peak height and increase in peak width while keeping the area constant.

On the other hand, Garlick and Gibson (1948) considered the possibility of retrapping, given by Eq. (3).

$$I_{TL} = \left(\frac{n_0^2}{N} \right) \exp\left\{ -\frac{E_t}{kT} \right\} \left[1 + \left(\frac{n_0 S}{\beta N} \right) \int_{T_0}^T \exp\left\{ -\frac{E_t}{k\theta} \right\} d\theta \right]^{-2} \quad (3)$$

Based on second-order kinetic, as the n_0 increases, the peak height shows a significant increase and peak position shifts to the much lower temperature region, with additional characteristics of wider (slightly broader) and more symmetric peak compared to the first-order peak. This is due to the significant concentrations of

released electrons are retrapped before recombine, thus giving rise to a delay in the luminescence emission and a spreading out of the emission over a wider temperature range.

It is observed that Peaks 6 and 8 were exhibited the first-order kinetic peak response, as the E_a increases with increasing T_{max} , as shown in Fig. 7. In contrast, peaks 5 and peak 7 exhibiting the opposite response of E_a with T_{max} . It is also observed not much change of E_a for Peak 4 as T_{max} increasing.

To confirm the first-order peak kinetic behavior, the peak integral of all peaks is compared. Fig. 8 shows the variation in E_a with changes in PI . It is observed that Peaks 6 and 8 were exhibited the first-order kinetic peak response, as the E_a decreases with increasing PI , whereas Peaks 5 and 7 exhibiting the opposite response of E_a with PI and thus are not showing the behavior of first-order kinetic peaks. It is concluded that Peak 6 and 8 are the first-order kinetic peaks and can be used as the main dosimetric peak at both low and high temperatures. The average T_{max} of Peak 6 and 8 is 250 °C and 340 °C, respectively. Thus, Peak 6 is the dominant peak at lower temperature region, while Peak 8 is the dominant peak at higher temperature.

In variation with dose, the observations on WinREMS glow curves (LTP) are best described by the second-order kinetic Garlick and Gibson TL model. It is noted earlier that n_o is proportional to the absorbed dose. As the dose increases from 1 kGy to 10 kGy, Peak 6 position shifts to much lower temperature region from 254 to 250 °C with increasing peak height, exactly as predicted by second-order kinetic model. Peak 5 also exhibits the same behavior, as the glow peak shifts from 214 to 210 °C as the dose increases from 1 kGy to 5 kGy.

In contrast, for the HTP behavior in response to the dose variation, are successfully explained by first-order kinetic Randall and Wilkins TL model. As stated earlier, it is classified that the following peak 7(280 °C) and 8(330 °C) as HTP according to its temperature. As the dose increases from 1 kGy to 10 kGy, the positions of peaks are constant and the peak height shows a significant increase with dose. This finding is as expected in first-order kinetic model.

This combination of order of kinetic which occurred in the same glow curve, where Peaks 5 and 6 are exhibited the second-order behavior while Peaks 7 and 8 are more prone to the first-order kinetics, with the LTP shifts to the lower temperature region and drags parts of the glow curve and then producing wider and broader glow curve, as the dose increases from 1 kGy to 10 kGy.

Mostly all peaks showed the maximum TL intensity at the dose of 10 kGy and further increased in dose after that point, exhibited a significant decreased in TL intensity. It is called the critical dose limit of this TL material. Since Peaks 5 and 6 were diminished after 10 kGy (refer to the TL glow curve illustrated in blue), so the discussion on higher dose region that is mostly greater than the critical dose limit (> 10 kGy) will be based on Peaks 4, 7 and 8. It is noticed that peak 4 starts to be visible after the dose of 10 kGy.

As the dose increases from 10 kGy to 1000 kGy, Peak 8 position shifts to a much higher temperature region within the range of 330–340 °C and peak height decreasing with dose by 80%. It is observed the same behavior in Peak 7 and Peak 4. It is found that none of kinetics model can explain this interesting behavior at extremely high doses especially after the critical dose limit of 10 kGy. It is also noticed the strange behavior of Peak 4, which tends to be invisible at the lower dose (< 10 kGy) and starts to be appeared at the critical dose limit of 10 kGy.

3.3. Supralinearity

The property of supralinearity is related to the application of TL in dosimetry and is presented quantitatively by the supralinearity index, $f(D)$. Supralinearity index, $f(D)$, is given by Eq. (4), where D

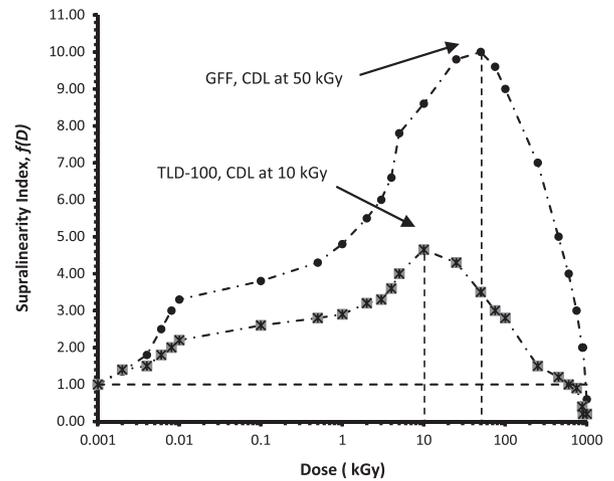


Fig. 9. The supralinearity index as a function of ultra-high dose of TLD-100 and GFF, following 2.5 MeV electron for doses up 1 MGy.

is the actual dose, D_o is the calibration dose level and I is the intensity of TL signal. Generally, the supralinear dose response is a characteristic of TLD-100, over 1 Gy of dose.

$$f(D) = \left[\frac{I(D)}{D} \right] \left/ \left[\frac{I(D_o)}{D_o} \right] \right. \quad (4)$$

Supralinearity is one of the nonlinear dose dependence effects, which grows to much higher values at a certain dose range, before reducing to low values when saturation effects become dominant (Chen and Hag-Yahya, 1996). Many models explaining the nonlinear effects with different perspective, such as; (1) that related to excitation stage, or (2) that related to heating phase and (3) types of radiation (UV, gamma rays, x-rays, α particle, β particle, electron and many more) (Chen and McKeever, 1997). The competing trap model by Kristianpoller et al. (1974), stated that the supralinear effect is mainly due to the competition during excitation and heating, which created the quadratic total dose dependence. The supralinear response was found to be proportional to the concentration of charge carrier in both traps and luminescence centers in the presence of a strong competitor. Instead, Banerjee (2001) suggested that supralinearity is dependent on the recombination cross-section, the thermal velocity of the carriers and the concentration of the recombination centers. The Unified Interaction Model (UNIM) by Horowitz (2001), explained that gamma-ray induced supralinearity at high doses arises from the decreasing efficiency of the competitive non-radiative processes relative to the luminescence recombination process.

It is clearly visible in Fig. 9, that the linearity index, $f(D) > 1$, which indicates that the TLD-100 and GFF exhibit the supralinearity response over the wide range of doses, up to 1 MGy. The supralinearity index $f(D)$ of TLD-100, increases by a factor of 3 as the dose increases from 1 kGy to 10 kGy. It is found that the supralinearity index reached its maximum at 10 kGy. In this study, this point is denoted as the Critical Dose Limit (CDL) of the dosimeter. Further increases in dose after the CDL of TLD-100, resulted in a significant decrease of $f(D)$ by a factor of 9 within the dose range of 10 kGy–1 MGy. It is also found that when the dose increases over the CDL, the peak height of TLD-100 decreases by a factor of 4, and the whole glow curve was slightly shifted to the higher temperature region by 10 °C. The glow curve was also observed to be increased in width.

On the other hand, the supralinearity index of GFF increases rapidly, as the dose increases from 1 kGy to 50 kGy. The CDL of

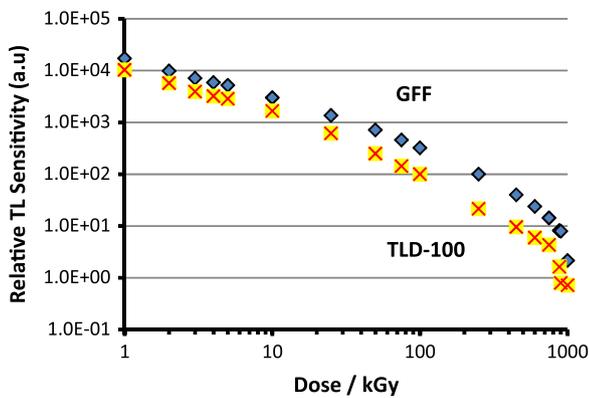


Fig. 10. Relative TL sensitivity as a function of ultra-high dose of TLD-100 and GFF following 2.5 MeV electron irradiation.

GFF, occurs at the dose of 50 kGy. After the CDL is achieved, the width of the glow curve increases, the peak height decreases by 40% and the whole glow curve is slightly shifted to the lower temperature region by 4%.

In this research, it was observed that no saturation effects for the TLD-100 and GFF samples within the studied dose range. This finding is contradicted to [Chen and Hag-Yahya \(1996\)](#), who stated that the saturation effects become dominant at much higher values at a certain dose range. It is also found that, further increase in dose, after the CDL point, causes a significant decrease in the supralinearity index, $f(D)$ of the TL material. However, [Horowitz \(2001\)](#), found that the decrease in TL after it reached the maximum value, was attributed to the saturation or damage effects. We also found that annealing the TLD-100 and GFF with temperature more than 400 °C is required to reset it back to its original condition, following radiation doses up to 1 MGy.

3.4. TL Sensitivity

The TL sensitivity (amount of TL signal per unit of absorbed dose) depends on several experimental and material variables. These variables include the batch of material, the annealing procedure, the heating rate, the glow curve region of integration, and the spectral response of the photomultiplier tube used in the TLD reader. It is customary to use TLD-100 as the standard reference by its relative sensitivity ([Moscovitch and Horowitz, 2007](#)).

The relative sensitivity of the FF samples is presented here in [Fig. 10](#). The GFF shows a good sensitivity at ultra-high dose of electron irradiation.

[Fig. 10](#) shows the dose dependency of TL sensitivity in GFF studied with TLD-100 as the reference material. It is observed a significant decrease in TL sensitivity as the dose increases within the range of 1 Gy to 1 MGy, for all samples. The observation was in agreement with the TL studies using ^{60}Co gamma ray irradiation at 0.1 MGy that were performed by [Bilski et al. \(2008\)](#), who found sensitivity loss as the dose increased. In this study, it is also found that the GFF showed a substantially greater TL sensitivity than that of TLD-100 by 4.8 times. The decrease in sensitivity is related to the effects of the deep traps ([McKeever et al., 1997](#)). In this study, we propose the loss in sensitivity at UHD might be contributed with the loss of electron energy when interacting with the inner shell atomic electron, which has a stronger nuclear force compared with the outer shell electrons.

4. Conclusion

UHD radiation allowed us to determine the maximum limit of radiation of each dosimeter. The TL model is valid to explain the changes in TL kinetic parameters of TLD-100 at UHD. The TL response at UHD revealed the abnormal behavior of peak 4 of TLD-100, and might be an important clue to understand the behavior of TLD-100 at extremely high dose range. The supralinearity response at UHD showed the limitations of the TL kinetic models when dealing with the behavior of kinetic parameters of the studied glow peaks. One notable finding which showed that the TL kinetic model can be used and valid to explain the behavior of glow peak before the CDL is achieved, whereas none of the TL models can explain the strange changes in TL glow peak at the dose more than 10 kGy up to 1 MGy.

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References

- Alawiah, A., Bauk, S., Abdul-Rashid, H.A., Gieszczyk, W., Hashim, S., Mahdiraji, G.A., Tamchek, N., Bradley, D.A., 2015. Potential application of pure silica optical flat fibers for radiation therapy dosimetry. *Radiat. Phys. Chem.* 106, 73–76.
- Banerjee, D., 2001. Supralinearity and sensitivity changes in optically stimulated luminescence of annealed quartz. *Radiat. Meas.* 33, 47–57.
- Bilski, P., Obyrk, B., Stuglik, Z., 2010. Behaviour of LiF: Mg,Cu,P and LiF: Mg,Ti thermoluminescent detectors for electron doses up to 1 MGy. *Radiat. Meas.* 45, 576–578.
- Bilski, P., Obyrk, B., Olko, P., Mandowska, E., Mandowski, A., Kim, J.L., 2008. Characteristics of LiF: Mg,Cu,P thermoluminescence at ultra-high dose range. *Radiat. Meas.* 43, 315–318.
- Bos, A.J.J., 2001. High sensitivity thermoluminescence dosimetry. *Nucl. Instrum. Methods B* 184, 3–28.
- Chen, R., McKeever, S.W.S., 1997. *Theory of Thermoluminescence and Related Phenomena*. World Scientific, London.
- Chen, R., Hag-Yahya, A., 1996. Interpretation of very high activation energies and frequency factors in TL as being due to competition between centres. *Radiat. Prot. Dosim.* 65 (1–4), 17–20.
- Garlick, G.F.J., Gibson, A.F., 1948. The electrons trap mechanism of luminescence in sulphide and silicate phosphors. *Proc. Phys. Soc.* 60, 574–590.
- Horowitz, Y.S., Horowitz, A., Oster, L., Marino, S., Datz, H., Margaliot, M., 2008. Investigation of the ionisation density dependence of the glow curve characteristics of LiF: Mg, Ti (TLD-100). *Radiat. Prot. Dosim.* 131 (4), 406–413.
- Horowitz, Y.S., 2001. Theory of thermoluminescence gamma dose response: the unified interaction model. *Nucl. Instrum. Methods Phys. Res. Sect. B* 184, 68–84.
- IAEA, 2000. *Absorbed Dose Determination in External Beam Radiotherapy: An International Code of Practice for Dosimetry Based on Standards of Absorbed Dose to Water*. Technical Report Series, vol. 398. Vienna-Austria.
- ICRU report No. 24., 1976. *Determination of Absorbed Dose in a Patient Irradiated by Beams of X or Gamma Rays in Radiotherapy Procedures*. International Commission on Radiation Units and Measurements (ICRU), Washington, D.C.
- Khoury, H.J., Obyrk, B., Barros, V.S., Guzzo, P.L., Ferreira, C.G., Bilski, P., Olko, P., 2011. Response of TL lithium fluoride detectors (MTS) to high gamma radiation doses. *Radiat. Meas.* 46, 1778–1881.
- Kristianpoller, N., Chen, R., Israeli, M., 1974. Dose dependence of thermoluminescence peaks. *J. Phys. D: Appl. Phys.* 7, 1063–1072.

- McKeever, S.W.S., Agersnap Larsen, N., Bütter-Jensen, L., Mejdahl, V., 1997. OSL sensitivity changes during single aliquot procedures: computer simulations. *Radiat. Meas.* 27, 75–82.
- Montaño-García, C., Gamboa-deBuen, I., 2006. Measurements of the optical density and thermoluminescent response of LiF: Mg,Ti exposed to high doses of ^{60}Co gamma rays. *Radiat. Prot. Dosim.* 119 (1–4), 230–232.
- Moscovitch, M., Horowitz, Y.S., 2007. Thermoluminescence materials for medical applications: LiF: Mg,Ti and LiF: Mg, Cu, P. *Radiat. Meas.* 41, S71–S77.
- Obryk, B., Bilski, P., Glaser, M., Fuerstner, M., Budzanowski, M., Olko, P., Pajor, A., 2010. The response of TL lithium fluoride detectors to 24 GeV/c protons for doses ranging up to 1 MGy. *Radiat. Meas.* 45, 643–645.
- Puchalska, M., Bilski, P., 2006. GlowFit: a new tool for thermoluminescence glow-curve deconvolution. *Radiat. Meas.* 41, 659–664.
- Randall, J.T., Wilkins, M.H.F., 1945. Phosphorescence and electron traps. I. The study of trap distributions. *Proc. R. Soc. Sci. Lond. A* 184, 366–389.