Characterisation of the intrinsic dose response of lithium fluoride based thermoluminescent detectors

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***Abstract*—Interpretation of lithium fluoride-based thermoluminescent detectors (LiF TLDs) in mixed high energy radiation fields, such as those encountered in space and accelerator facilities, is challenging due to variations in relative luminescence efficiency with radiation type and energy. This study aims to assess the intrinsic macroscopic dose response of MCP-N, MTS-N, and MTT-7 TLDs with the default protocols used at SCK CEN. The TLDs were subjected to a four-step protocol involving annealing, irradiation, preheating and readout. The irradiation was performed with a ^{60}Co γ-ray source. The dose response data were obtained by analyzing the light intensity signals using the WinREMS software and deconvolution software GlowFit. The glow curves of the TLDs were analyzed from a dose of 0.05 Gy up to 15.6 kGy and the linearity indices of the TLDs were determined. The results show the effect of dose on the glow curves and the emergence of additional peaks at higher doses. The response to radiation dose from the main peak was found to be supralinear for MTS-N and MTT-7 detectors and sublinear for MCP-N detectors. The study provides valuable insights into the dose response characteristics of LiF TLDs and contributes to improving their interpretation in mixed radiation fields.**

***Index Terms*: High doses; LiF; Thermoluminescence; TLD**

I. Introduction

In mixed high energy radiation fields – such as those encountered in space, hadron therapy and other accelerator facilities – interpretation of lithium fluoride based thermoluminescent detectors (LiF TLDs) measurements is not straightforward because of variations of the relative luminescence efficiency with radiation type and energy. The relative luminescence efficiency can be predicted by the Microdosimetric *d(z)* Model [1]. This model requires the intrinsic macroscopic dose response of the LiF TLDs for irradiation with low ionization density, usually Co-60 gamma radiation. Such data are scarce and depend also on the protocols used for thermoluminescent detector (TLD) preparation, readout and analysis. Therefore, the main goal of this work is to assess the intrinsic macroscopic dose response of MCP-N, MTS-N and MTT-7 TLDs with the default protocols used at SCK CEN.

II. Materials and methods

The intrinsic dose response of three different LiF-based thermoluminescent detectors (TLDs) have been tested: high-sensitive LiF:Mg,Cu,P (MCP-N), standard LiF:Mg,Ti (MTS-N) and another version of LiF:Mg,Ti (MTT-7) which has a modified activator composition and an increased high-LET response [2]. The N and 7 in these abbreviations stand for natural Li abundance (N) and enriched in Li-7 (7). In this study the Li abundance does not matter since there is only a difference in interaction between Li-6 and Li-7 for neutrons and not for gamma. The version enriched in Li-7 was used for MTT because that is the only type of MTT detector available. All TLDs were fabricated by RADCARD (former TLD Poland) and had a consistent cylindrical shape with a diameter of 4.5 mm and a thickness of 0.9 mm. The following section explains the four-step protocol used at SCK CEN that every detector must undergo: annealing, irradiation, preheating and readout.

A. Protocol

1) Annealing

Prior to radiation exposure, each TLD was reset by annealing according to the protocol used at SCK CEN. The MTT-7 and MTS-N detectors required one hour at 400°C followed by a two-hour warm-up at 100°C. The final part of the annealing process for MTT-7 and MTS-N included slow cooling at room temperature. Contrary to MTT-7 and MT-N, the MCP-N detectors had to be annealed for ten minutes at 240°C followed by a prompt cooling at -10°C in a freezer.

2) Irradiation

As soon as the annealing was finished, the detectors were inserted into their irradiation holder. This holder consisted of Polymethyl methacrylate (PMMA) and did affect the irradiation process in positive terms. The irradiations were performed in terms of air kerma in Gray (Gy). The 3 mm PMMA for the detectors provided build-up so that there was a charged particle equilibrium and that the absorbed dose in the detectors was equal to the delivered air kerma. Each radiation exposure took place at the laboratory of nuclear calibration (LNK) of the Belgian Nuclear Research Centre SCK CEN in Mol and was performed using a ^{60}Co γ-ray source. The explicit details regarding irradiation depend on the part of the experiment and are further explained in sections B and C of the readout.

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Table 1: Overview of the TLD types and the protocol used at SCK CEN

TLD type	Size	Dopants	Annealing	Preheat	Integration
MCP	Cylindrical Thickness: 0.9 mm Diameter: 4.5 mm	Mg, Cu, P	10 min 240°C Cooling -10°C	30 min 120°C	Whole range
MTT MTS		Mg, Ti	1 h 400°C 2 h 100°C Cooling 21°C		150°C – 248°C

3) Preheat

After irradiation and before readout, it was necessary to preheat the detectors. This was done to prevent the occurrence of low-temperature deviations in the glow curve structure of the detectors and to avoid signal decay over time [3]. All TLD types required heating at 120°C for half an hour. Afterwards, the TLDs cooled gradually at room temperature.

4) Readout

The readout of the irradiated detectors was done using the Thermo Fisher Scientific Harshaw 5500 reader. The device could lift each detector one by one by using a needle that created a vacuum. From the moment the detector was raised, it was heated by a gaseous nitrogen stream. The MTTs and MTSs were heated up to 340°C while the MCPs were heated up to 255°C. All detectors worked on the luminescence principle. When the detector experienced radiation, some of its radiant energy was absorbed by the detector. During readout, this absorbed energy was re-emitted in the form of light [1]. A lateral photomultiplier tube (PMT) collected this luminescent light and converted it into a signal of light intensity. In this way, a glow curve was created in which the light intensity was shown as a function of temperature. The reader was monitored through the computer software WinREMS. The software WinREMS was able to determine the total intensity delivered by the irradiated detector. The software did this by taking the integral under the glow curve. In the case of MCP detectors, this method was used. Since the MTT and MTS detectors have more peaks at higher temperature with a dose and LET response different from the main peak, it is not reliable to look at the total light intensity of these detectors [4]. The WinREMS software was programmed to consider only the light intensity of the main peak by taking the integral or region of interest (ROI) between 150°C and 248°C (ROI 1). The rate of readout was also important, but again depended on the part of the experiment. Further explanation regarding the readout rate is discussed in sections B and C.

The complete protocol and characteristics of the detectors are summarized and shown in table 1.

B. *Determination of individual sensitivity factors (IF)*

Each TLD has its own sensitivity. The individual sensitivity factors of all TLDs were determined by irradiating all TLDs used in the experiment together with a fixed group of reference detectors. The irradiation was accomplished with the Co-60 source and with a dose of 50 mGy. These radiations were performed with the LNK's Panoramic DS-20 irradiator at a distance of 100 cm. The calibrations were based on the ISO 4037: 2019 standard and fell under the ISO 17025:2017 accreditation scope of LNK [5].

First, the reference detectors and other detectors were read out. The reading of all TLDs was performed at a temp of 10°C per second. The software WinREMS provided the light intensity for each detector within the range shown in Table 1. Through an Excel sheet, the average amount of light was determined from all the reference detectors. Based on this average, the individual correction factor of all detectors used in the measurements was determined. This was obtained by dividing the total amount of light from any TLD by this calculated average. The same method is applied for all detector types.

C. *Irradiation and readout of TLDs*

The actual experiment required irradiating groups of three TLDs of each type (MCP-N, MTT-7 and MTS-N) with different doses. Since this study focuses on intrinsic dose response, it was necessary to work with a diaphragm. Non-linearity effects of the reader were eliminated by using this technique. Therefore, the doses were divided into two different sets where a distinction was made in the method of readout. Set A was read with a 5 mm diameter aperture. Set B, on the other hand, used a 0.4 mm diaphragm for MCP-N detectors and a 1.2 mm diaphragm for the MTT-7 and MTS-N detectors. Both set A and B were read at a rate of 1°C per second. Table 2 shows the doses of the two sets. The overlapping doses between set A and B were to allow the calculation of the rescaling factor between readings with two diaphragm sizes in order to get dose response data from 0.05 Gy up to 15.6 kGy.

Table 2: Doses per set

Set	Doses
A	0.05 Gy, 0.1 Gy, 0.2 Gy, 0.5 Gy, 1 Gy, 2 Gy, 5 Gy, 10 Gy
B	1 Gy, 2 Gy, 5 Gy, 10 Gy, 20 Gy, 50 Gy, 100 Gy, 200 Gy, 500 Gy, 1 kGy, 2 kGy, 5 kGy, 10 kGy, 15.6 kGy

Sets A and B were further divided into five series because it was impossible to read out an entire set on one day. Per set, three TLDs of each kind with a reference dose of 50 mGy were read together with the other TLDs to correct for potential sensitivity variations of the reader from day to day. Thus, anomalies were corrected. Irradiations up to 1 Gy were performed using the LNK's Panoramic DS-20 irradiator at a distance of 100 cm. These calibrations were based on the ISO 4037:2019 standard and fell under the ISO 17025:2017 accreditation scope of LNK. The G-100 irradiator was utilized for the remaining irradiations ranging from 2 Gy to 15.6 kGy.

Irradiations between 2 Gy and 200 Gy were performed at a distance of 150 cm. These calibrations were also based on the ISO 4037:2019 standard and fell under the ISO 17025:2017 accreditation scope of LNK. In the case of irradiations from 500 Gy to 15.6 kGy, the dose rate had to be increased, as the irradiation would otherwise take too high a time. Therefore, the stands were placed 50 cm from the source. These irradiations were performed with reference standards and calibration equipment for which the traceability to national and international standards was not demonstrated or some corrections were not under control. The irradiation is not within the ISO 17025: 2017 accreditation scope of LNK. However, a reference measurement was taken at this distance and based on this obtained dose rate, it was possible to precisely irradiate at the TLDs with the correct dose.

D. Analysis of the data

1) Analysis of data obtained by the software WinREMS

For series 1 MCP-N detectors, all total light values were corrected using the individual sensitivity factors. In other words, the total light value of a detector was divided by the individual factor of that particular detector. The formula for these individual-sensitivity-corrected signals (ISCS) are shown in equation (1). Wherein S is the total light value for MCP-N and the ROI for MTT-7 and MTS-N.

$$ISCS = \frac{S}{IF} \quad (1)$$

Since three detectors were read for one particular dose, the average of these three individual-sensitivity-corrected signals was calculated. This average of the individual-sensitivity-corrected signals was then divided by the dose. These results give the signal per dose. The signal per dose of a given measurement was divided by the signal per dose value of the dose of 50 mGy of series 1. In this way, the linearity index was obtained. The same procedure was applied for series 2 of the MCPs. This operation is visualized in equation (2).

$$Linearity\ index_{1,2} = \frac{(S/IF)_{av,dose\ x}/dose\ x}{(S/IF)_{av,dose\ 50mGy}/50\ mGy} \quad (2)$$

In this equation, $(S/IF)_{av,\ dose\ x}$ is the average individual-sensitivity-corrected signal for the considered dose x.

For series 3, 4 and 5, the diaphragm correction factor had to be determined first (because different diaphragm sizes were used here). Four doses were read with both the 5 mm aperture and the 0.4 mm aperture. By taking a ratio of the average individual-sensitivity-corrected signals of the 5 mm diaphragm to the average individual-sensitivity-corrected signals of the 0.4 mm diaphragm, this diaphragm correction factor was obtained for each of the four doses. From these four different ratios, an average was taken which is used as the diaphragm correction factor. The average individual-sensitivity-corrected signals were multiplied by the diaphragm correction factor (*DF*). This resulting value was in turn divided by the dose so that the signal per dose is obtained. These values are then divided by the signal per dose value of the 50 mGy dose of that series. As a result, the linearity index is derived. This operation is visualized in equation (3).

$$Linearity\ index_{3,4,5} = \frac{\langle (S/IF)_{av,dose\ x} * DF \rangle / dose\ x}{(S/IF)_{av,dose\ 50mGy}/50\ mGy} \quad (3)$$

Similar to MCP-N, the values of the MTT-7 and MTS-N detectors were analyzed in a similar way. Only here one should not use the total light values, but the ROI 1 values.

2) Analysis of data obtained by the software GlowFit

As the dose on the detectors increased, more peaks appeared in the glow curve. In the presence of increasing contribution of the other peaks, integrating over a given ROI is no longer sufficiently accurate. Hence the more accurate analysis by deconvolution with GlowFit. In the next step of the analysis, the glow curves were re-examined to look at structural changes due to the degree of irradiation in this way. The glow curve was split into several peaks in order to examine the contribution of each peak separately. The deconvolution of the glow curves was done by the software GlowFit, developed at Institute of Nuclear Physics (IFJ) in Kraków, Poland. Three parameters had to be determined per peak for this deconvolution to be successful. These parameters included the maximum light intensity, the activation energy (*E*) and the temperature at the maximum of the annealing curve (T_{max}). Previous research [1] examined the number of peaks in the deconvolution of the glow curves of MCP-N, MTT-7 and MTS-N. In parallel, this study examined what values the activation energy and temperature at the maximum are for each deconvolution peak for the different types of detectors. In the case of MCP-N detectors, the glow curve could be split into four sub-peaks. For MTT-7 and MTS-N detectors, there were six. The values of activation energy and temperature at maximum intensity of the peaks are shown in table 3.

Table 3: Deconvolution parameters [3]

Parameter	Peak 1	Peak 2	Peak 3	Peak 4	Peak 5	Peak 6
LiF:Mg,Cu,P						
T_{max} [°C]	168.4	208.4	222.3	241.7		
E [eV]	1.37	2.09	2.66	2.11		
LiF:Mg,Ti						
T_{max} [°C]	163.2	190.5	219.4	249.0	282.9	315.3
E [eV]	0.77	2.18	1.70	1.48	1.62	2.71

Deconvolution in GlowFit yielded the integrated light intensity of each peak. However, only the light intensity of the main peak was considered. This peak was peak 3 for MTT-7 and MTS-N and peak 2 for MCP-N. Based on these obtained light intensities of the main peak, the same analysis was performed as in part one of the analysis 'Analysis of data obtained by the software WinREMS'.

III. Results

Figures 1, 2 and 3 present glow curves at 50 mGy, 2,000 Gy and 15,600 Gy for MCP-N, MTT-7 and MTS-N, respectively. The TLD signals are normalized to the maximum height of the main peak.

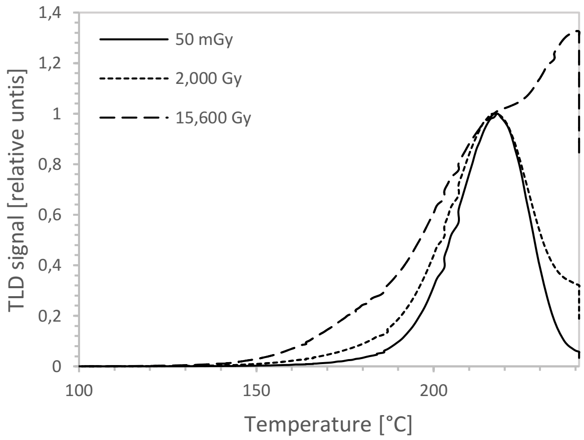

Figure 1: Glow curves of MCP at various dose levels normalized to maximum height of main peak

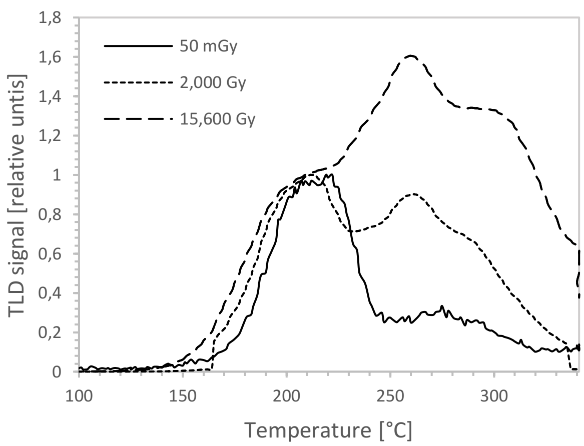

Figure 2: Glow curves of MTT at various dose levels normalized to maximum height of main peak

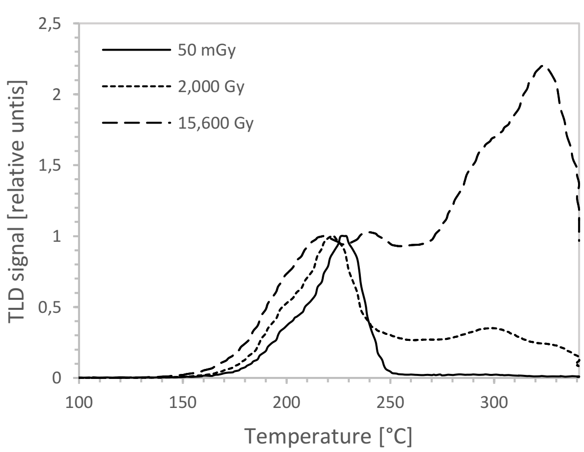

Figure 3: Glow curves of MTS at various dose levels normalized to maximum height of main peak

Figures 4, 5 and 6 show the linearity index of the main peaks as a function of delivered dose for MCP-N, MTT-7 and MTS-N detectors, respectively. Both the analysis based on the results obtained in WinREMS and Glowfit are shown in the same graph.

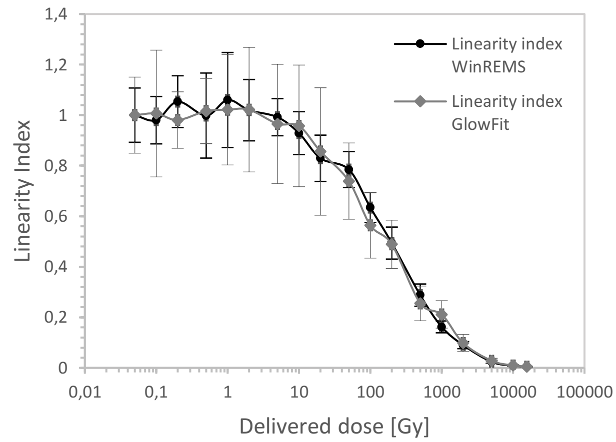

Figure 4: Linearity of the main peaks of MCP detectors

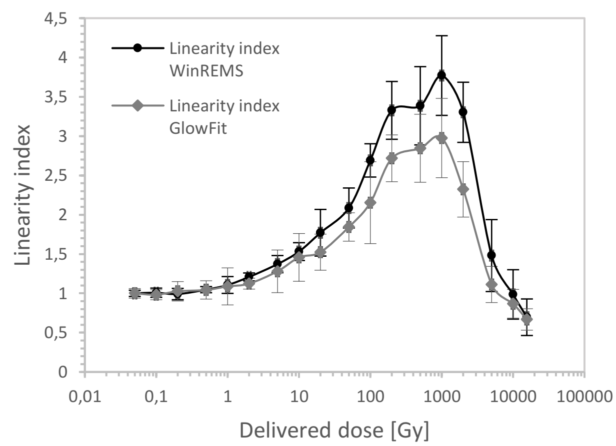

Figure 5: Linearity of the main peaks of MTT detectors

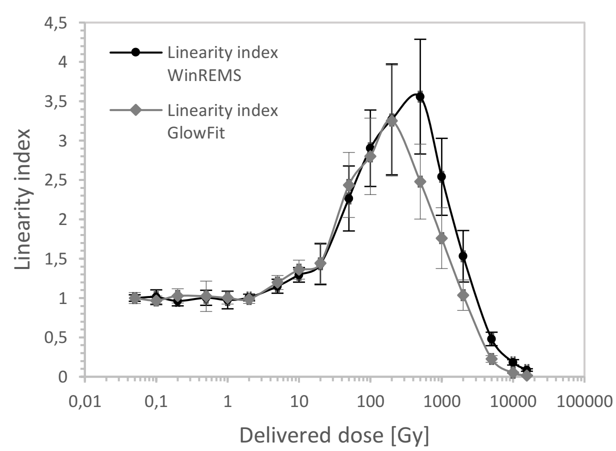

Figure 6: Linearity of the main peaks of MTS detectors

IV. Discussion

A. Glow curve structure

Figure 1 presents a comparison of the glow curves for MCP-N at 50 mGy, 2,000 Gy and 15,600 Gy. Due to the normalization of the glow curves, the relative increase of the different peaks is visualized. The glow curve of 50 mGy serves as a reference. The shape of the glow curves between 50 mGy and 1,000 Gy is appeared to be unchanged. Previous research [6] has shown this to be the similar situation. Starting at doses higher than 1,000 Gy, a widening of the peak occurs indicating an increase in peaks that emerge at doses.

As shown in Table 3, four peaks are represented in the glow curve of MCP-N that includes peak 2 as the main peak. The widening of the glow curve at a dose of 2,000 Gy compared to 50 mGy indicates that peaks 1, 2 and 3, which respectively appear at a temperature of 168.4°C, 208.4°C and 241.7°C, are relatively increased. This increase in peaks is more pronounced at the dose of 15,600 Gy. The general trend is that the higher the dose, the higher the temperature peaks.

A corresponding tendency is evident in Figure 2 where a comparison of glow curves is given for MTT-7 to doses of 50 mGy, 2,000 Gy and 15,600 Gy. Also in this case, the glow curve of 50 mGy serves as a reference. Table 3 indicates that the annealing curve of MTT-7 consists of six peaks. The main peak is peak 3 and is positioned at a temperature of 219.4°C. This peak describes the highest light intensity on the 50 mGy glow curve, and therefore takes the value of 1. If the same temperature is considered on the 2,000 Gy glow curve, this light intensity is no longer maximum. The maximum light intensity is obtained at a lower temperature. The height of the main peak is reduced compared to peak 2. A parallel study obtained a similar result [2]. In parallel, peaks 1, 4, 5 and 6, respectively located at temperatures of 163.2°C, 249.0°C, 282.9°C and 315.3°C, are also increased in magnitude compared to the corresponding peaks at the glow curve of 50 mGy.

A prominent observation of the change in the glow (Figure 3) curve for MTS-N is that the main peak shifts to lower temperatures as the dose increases. Contrary to MTT-7, the height of peak 3 does not become smaller than that of peak 2. For MTS detectors the obtained shapes are similar to MTT-7, with growth of several high-temperature peaks for high doses. Peaks 1, 4, 5 and 6, located at temperatures of 163.2 °C, 249.0 °C, 282.9 °C, and 315.3 °C, respectively, increase in size compared to the corresponding peaks in the 50 mGy glow curve. In the case of MTS-N, the high-temperature peaks are all more prominent than for MTT-7. A previously conducted study, obtained similar results [2].

Since the annealing curve changes shape significantly as the dose increases, it is essential to also calculate the linearity indices with Glowfit as described earlier in the method section.

B. Linearity index WinREMS and GlowFit

Figure 4 shows the linearity index as a function of delivered dose for the MCP-N detectors. These detectors become sublinear after the dose of 5 Gy. This indicates that the detectors enter saturation. The detectors saturate immediately without reaching supralinearity. The previously cited study [2] came out with similar findings. An identical result is obtained from both WinREMS and Glowfit.

A completely opposite curve is obtained for MTT-7 and MTS-N. Figure 6 shows the linearity index as a function of delivered dose for the MTT-7 detectors. This curve is linear up to a dose of 0.5 Gy and then shows supralinearity. Saturation occurs at 1,000 Gy. The linearity index subsequently decreases and then loses a strong sensitivity to become sublinear from 10 kGy.

There is a significant deviation between the curves of WinREMS and GlowFit. At lower doses, these are similar because the high-temperature peaks are not yet as pronounced. From 10 Gy, all values of the linearity index based on GlowFit are lower than WinREMS. This indicates that the high-temperature peaks interfere with the main peak. GlowFit takes into account several peaks and only considers the area below the main peak after deconvolution has occurred. WinREMS takes only the integral between the ROI. This is the reason GlowFit's values are lower.

A similar trend is shown in Figure 6 where the linearity indices of MTS-N detectors are presented. The difference observed between the MTT-7 detectors is that the linearity index remains 1 up to a dose of 2 Gy for both the WinREMS and GlowFit curves. From this dose, the curve becomes supralinear until saturation occurs at 500 Gy on the WinREMS curve. In the case of the GlowFit curve, saturation already occurs at 200 Gy. This indicates the effect of the high temperature peaks at this point again. After this, the curve will decrease and develop even sublinear from a dose of 5,000 Gy for the WinREMS curve. For the GlowFit curve, this is from 2,000 Gy. Similarly, the WinREMS curve is lower for MTS-N than for Glowfit because of the identical reason as for MTT-7.

It is therefore required to consider mainly the analysis of GlowFit because it observes only the principal peak. Based on these data, the MTS-N detectors are more supralinear than the MTT-7 detectors. In contrast to this study, previous research [2] shows precisely the opposite result.

V. Conclusion

Some conclusions can be drawn from the results obtained. For the MCP-N detectors, it was found that the shape of the glow curves remains unchanged between 50 mGy and 1,000 Gy. However, above a dose of 1,000 Gy, an increase in peaks occurs at higher doses. The temperature peaks on the glow curves increase as the dose increases.

For the MTT-7 detectors, it was observed that the main peak, peak 3, on the glow curve at 50 mGy exhibits the highest light intensity. At higher doses, the height of this peak decreases and other peaks (peaks 1, 4, 5 and 6) increase in magnitude.

With the MTS-N detectors, it was observed that the main peak shifts to lower temperatures as the dose increases. Again, several high-temperature peaks increased in magnitude compared to the glow curve of 50 mGy.

Regarding the linearity indices, for the MCP-N detectors it was observed that they become sublinear after a dose of 5 Gy, indicating saturation without reaching supralinearity.

For the MTT-7 detectors, the linearity index was found to be linear up to a dose of 0.5 Gy and then becomes supralinear until saturation at 1,000 Gy. At doses above 10 kGy, the linearity index becomes sublinear and sensitivity decreases.

For the MTS-N detectors, the linearity index remained 1 up to a dose of 2 Gy, after which it became supralinear until saturation at 500 Gy (WinREMS) or 200 Gy (GlowFit). After saturation, the curve decreased and became sublinear from doses of 5,000 Gy (WinREMS) or 2,000 Gy (GlowFit). Again, GlowFit values were lower due to the influence of the high-temperature spikes.

Based on these findings, GlowFit provides more accurate results because it focuses on the main peak. In addition, the MTS-N detectors are found to be more supralinear than the MTT-7 detectors.

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