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Thermoluminescence and storage stability of TLD-100 dosimeter irradiated with ^{60}Co-gamma rays

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Thermoluminescence glow curves of TLD-100 revealed three peaks at 373, 460 and 518 K for all samples irradiated with gamma ray doses of 0.5 to 700 Gy. The total thermoluminescence response and the height of the main peak at 460 K showed similar characteristics to radiation dose. On the other hand, the total area under the glow curve increases continuously with radiation dose up to 1000 Gy. All irradiated samples investigated showed no significant fading over 28 d. Activation energy, *E*, and escape frequency factor, *s*, for the main glow peak were calculated by the modified empirical equation, as well as by methods depending on the shape of the glow peak. It was found that *E* has a value of 1.33 to 1.83 eV and *s* falls between 5.8×10^{13} and 3.06×10^{19} sec^{-1}, depending on the method used.

1. Introduction
In the early 1950s most of the thermoluminescent phosphors were discovered and used for dosimetric purposes, such as LiF [1], $CaSO_4$ [2] and CaF [3]. LiF was extensively used in the technique of radiation dosimetry because of its near tissue-equivalent response. Studies were resumed by Cameron and colleagues [4, 5] who conceived the systematic regeneration of LiF and encouraged the commercial production by Harshaw Chemical Company, of products known as TLD-100, TLD-600 and TLD-700 depending on their preparation, from natural lithium of lithium enriched with ^6Li or ^7Li. A similar quality of LiF, stabilized with sodium was studied in France by Portal and colleagues [6–8] and commercialized by Desmarquest and CEC, South African, under the names PTL-710, PTL-716 and PTL-717.

Pure LiF was found to have poor thermoluminescent (TL) emission [9]. The necessity of the presence of some impurities such as magnesium and titanium in LiF for a high TL response was determined [10–12]. Numerous investigations of LiF:Mg, Ti revealed ambiguous results concerning the magnesium and titanium contents necessary for maximum TL sensitivity. Several authors found the contents for optimum TL to be in the range 8 to 15 p.p.m. Ti and 100 to 300 p.p.m. Mg [13–19]. Wachter and colleagues [20, 21] presented a correlation of the characteristic features of the glow curve structure with various levels of magnesium and titanium. It was also shown that the hydroxyl ion which eliminates Mg^{2+} ions as electron traps by forming magnesium–hydroxyl complexes has to be considered as a third important constituent in the TL optimization procedure.

2. Experimental procedure
Lithium fluoride (TLD-100) was used in a powder form (Harshaw Chemical Company, USA). Cobalt units (Siemens, West Germany) had been used for LiF irradiation, the activity of a ^{60}Co source being $\sim 10^{14}$ Bq. The output of the source was calibrated by a Farmer sub-standard dosimeter (type 2502/3) and a water phantom type (2528/30) (both manufactured and supplied by Nuclear Enterprises Ltd, UK).

TLD-100 samples were dosed by gamma radiation in the range 0.5 to 700 Gy and then kept in light-tight containers ready for experimental measurements. Thermoluminescence of dosed LiF samples was measured by a thermoluminescence analyser (Harshaw Chemical Company, USA, Model 2000 A and B). Glow curves were recorded using an *x–y* recorder (Kipp and Zonen model B041).

3. Results and discussion
The crystal structure of both irradiated and non-irradiated lithium fluoride (TLD-100) was examined using a Shimadzu X-ray diffractometer with Cu$K\alpha$ radiation of 0.1542 nm. The X-ray patterns obtained proved a high degree of crystallinity and no change in crystal structure of the material due to gamma irradiation with doses up to 700 Gy which is desirable in materials used for radiation dosimetry. As these materials are usually used several times in dosimetric applications, any changes in their crystal structure during irradiation could lead to their deterioration, causing serious changes in their physical properties.

The thermoluminescence curves recorded for TLD-100 irradiated with gamma radiation doses of 0.5, 1, 2.5, 5, 10, 20, 50, 75, 100 and 700 Gy are shown in

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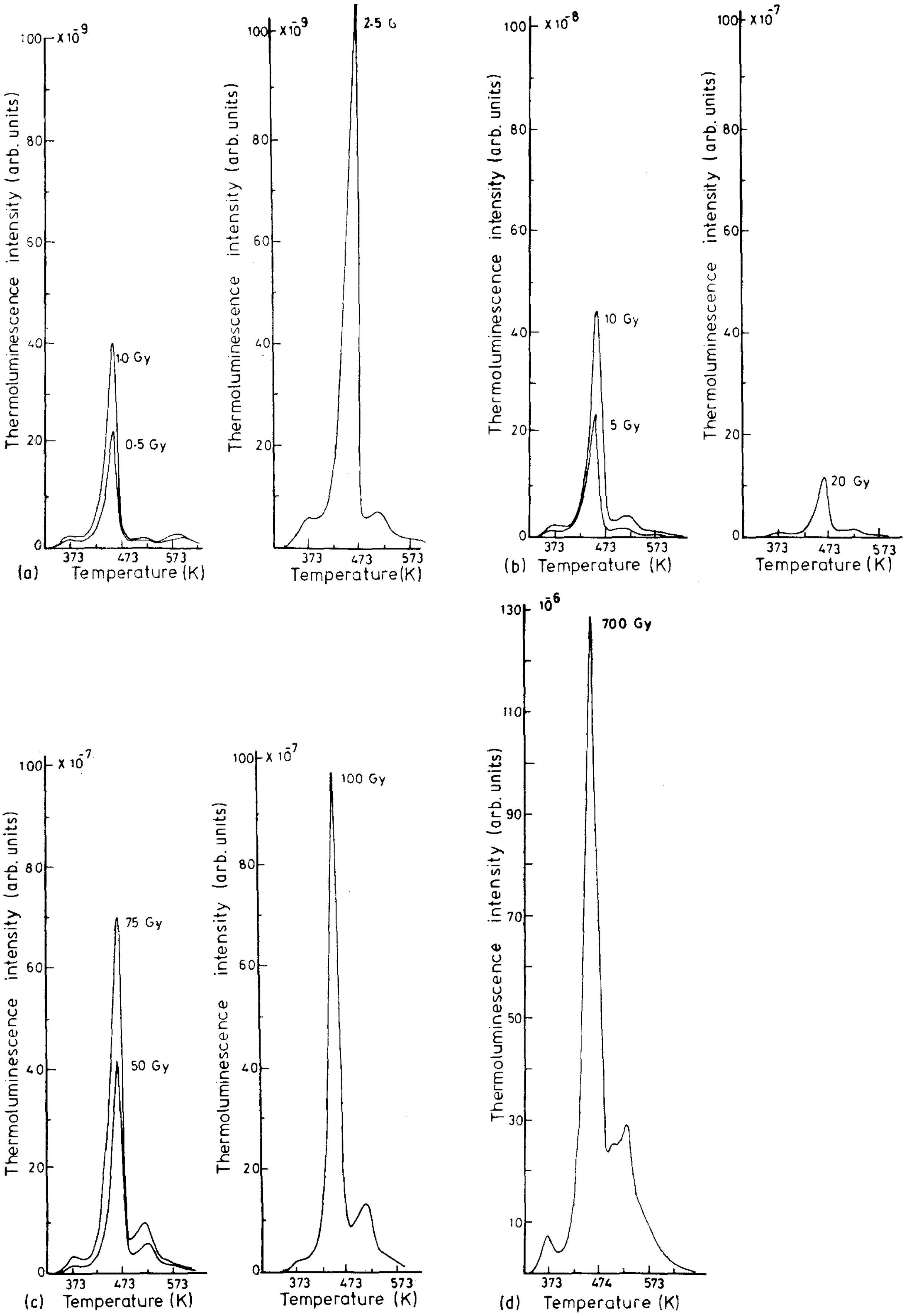

Figure 1 (a to d) Thermoluminescence glow curves of TLD-100 at various gamma radiation dose.

Fig. 1a to d. For each dose level investigated, the glow curves showed three peaks at 373, 460 and 518 K. The dominant glow peak for all irradiated samples appeared around 460 K. The shape of the glow curves did not change significantly with increasing gamma radiation dose. The only observed effect was an increase in the main peak intensity at 460 K as the radiation dose increased; this is due to the increase population of the electron traps. The glow peak temperatures of all measured glow curves are summarized in Table I. It is evident from the table that the temperatures of the glow peaks vary little with gamma

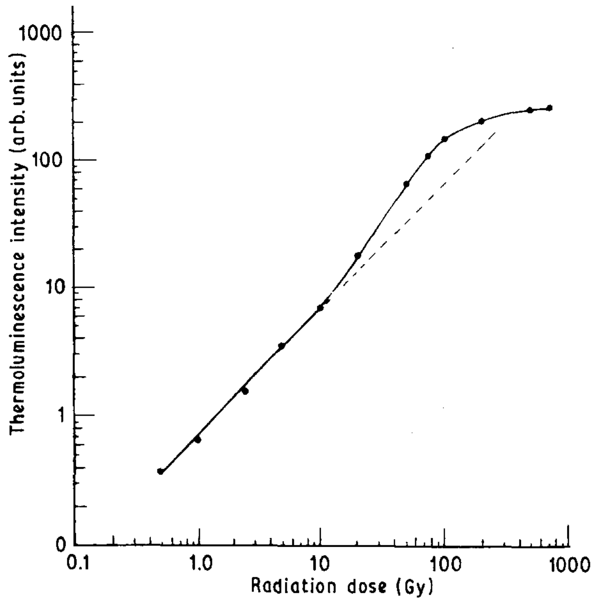

Figure 2 Thermoluminescence dose–response curve of TLD-100.

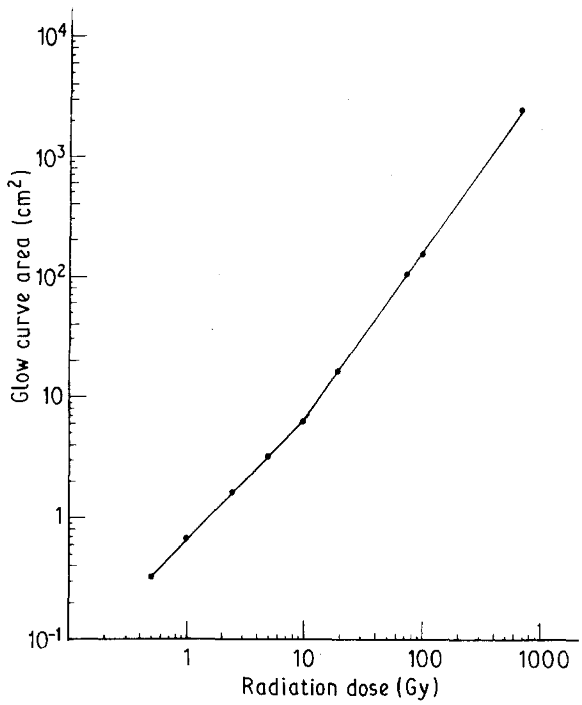

Figure 3 Variation of TLD-100 glow curve area with gamma radiation dose. (●) Peak 1, (○) Peak 3.

radiation doses of 0.5 to 700 Gy. This reveals that the TL of each peak is caused by the same kind of trapping centres. This observation is consistent with that reported by many authors [9, 22–24].

In recent papers [25, 26] the determination of activation energy from the shape of thermoluminescence peak was reviewed. The recorded glow curves can be analysed to obtain activation energy values for the various electron trap states present in the sample and the escape frequency factor. The activation energy, E, and escape frequency factor, s, were calculated according to the formulae of Grosswiener [27], Lushchik [28, 29] and Chen [30]. These formulae depend on the geometrical shape of the TL peak, i.e. the total width or half-widths of the peak, and also assumed that the kinetics involved in the TL glow peaks are first-order kinetics. The various formulae used assumed that the main peak obeys first-order kinetics. The calculated E and s values for the main glow peak using these methods are summarized in Table II. Apparently, the three methods used gave different values for the activation energy and the escape frequency factor. The activation energy and the escape frequency factor determined by Chen [30] agree reasonably with values reported in the literature for the main peak, and are also close to that estimated by the empirical formula, given by Urbach [31] which had the formula. $E = T_{max}/500$ and modified in this work to the formula

$$E\ (\text{eV}) = \frac{T_{max}(\text{K})}{331} \tag{1}$$

which is applicable for a heating rate of $2.5\ \text{K sec}^{-1}$ and escape frequency factor, s, of 10^{14} to $10^{15}\ \text{sec}^{-1}$ [32]. This formula gives an activation energy, E, of 1.39 eV. For these reasons, it seems reasonable to assign the activation energy value of 1.45 ± 0.04 eV for the trap depth responsible for the main glow peak with the corresponding frequency factor of $2.02 \times 10^{15}\ \text{sec}^{-1}$ in TLD-100. These values are compatible with that reported by Kathuria and Moharil [33].

The total charge in microcoulombs (μC) collected and displayed on the TLD analyser was recorded, this charge represents the total integrated area under the full glow curves of the irradiated thermoluminescent sample. When this charge was plotted as a function of the gamma-ray dose: the dose–response curve obtained in Fig. 2, represents the relation between the total charge displayed and gamma-ray dose. It is evident that the linear response extends over a dose range of 0.5 to 10 Gy, supralinearity was observed between 10 and 100 Gy, then a plateau or approximately saturation extends over high doses. On the other hand, when we tried to measure the area (in cm^2) under the recorded glow curves using a planimeter and plotted these values as function of gamma dose, we obtain Fig. 3 which represents a straight-line

TABLE I Glow peak temperatures for TLD-100 irradiated with ^{60}Co gamma rays

Dose (Gy)	Glow peak temperature (K)		
	Peak I	Peak 2	Peak 3
0.5	373	460	518
1.0	373	460	518
2.5	368	460	518
5.0	373	460	518
10.0	373	468	523
20.0	373	468	523
50.0	368	460	518
75.0	373	460	518
100.0	368	460	518
700.0	373	460	518

TABLE II Activation energy, E, and escape frequency factor, s, for the main glow peak of TLD-100

Reference	E (eV)	s (sec^{-1})
Chen [30]	1.45 ± 0.04	2.02×10^{15}
Grosswiener [27]	1.33 ± 0.019	5.80×10^{13}
Lushchik [28]	1.83 ± 0.025	3.06×10^{19}

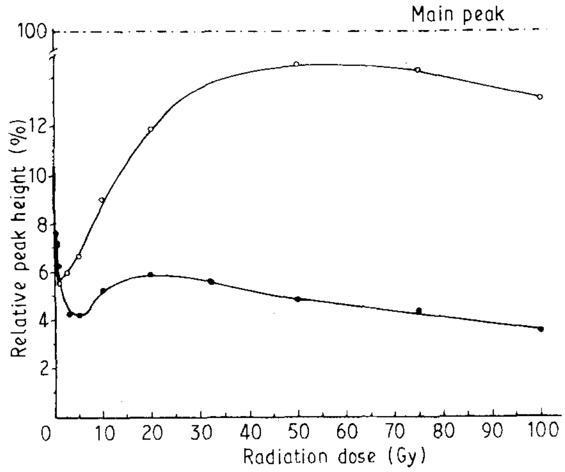

Figure 4 Relative peak heights for TLD-100 as a function of gamma radiation dose.

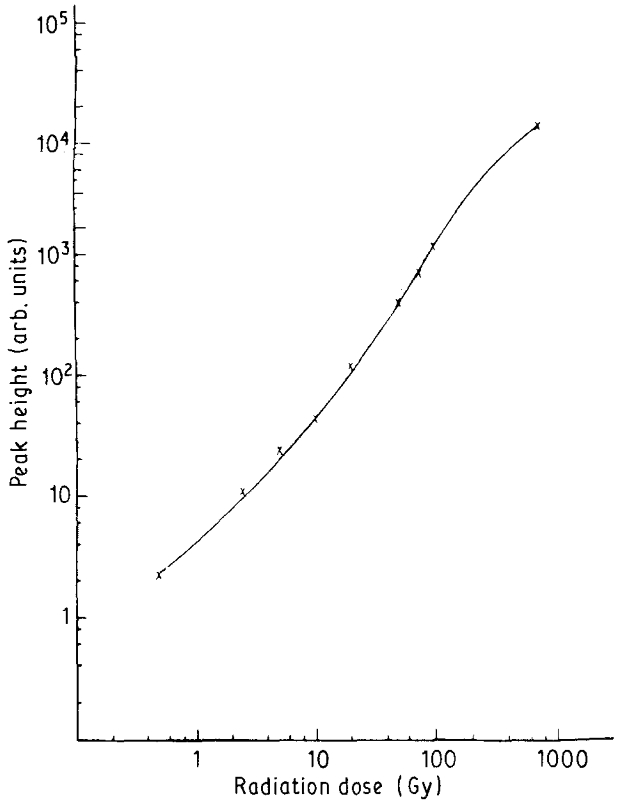

Figure 5 Changes in the main peak height of TLD-100 with gamma-ray dose.

relationship of two phases, 0.5 to 10 Gy and 10 to 700 Gy ranges. Therefore, high-dose measurement could be achieved using the second segment. It is interesting to mention that the linearity of the dose response curve observed agrees reasonably with observation of other authors [12]. Also, some authors [34] reported that the TL response of LiF goes to saturation at 1 kGy but supralinear behaviour appears at much lower doses.

The ratios of the individual heights of the glow peaks, appearing in the recorded glow curves, relative to that of the 460 K main peak were plotted as a function of the gamma dose. The results are shown in Fig. 4. With increasing radiation dose, the relative peak height ratios increase slightly (about 5%); this increase remains practically constant over the dose range 20 to 700 Gy, beyond which it decreases. These results indicate that over the full dosage range investigated, the peak which appeared at 460 K is the

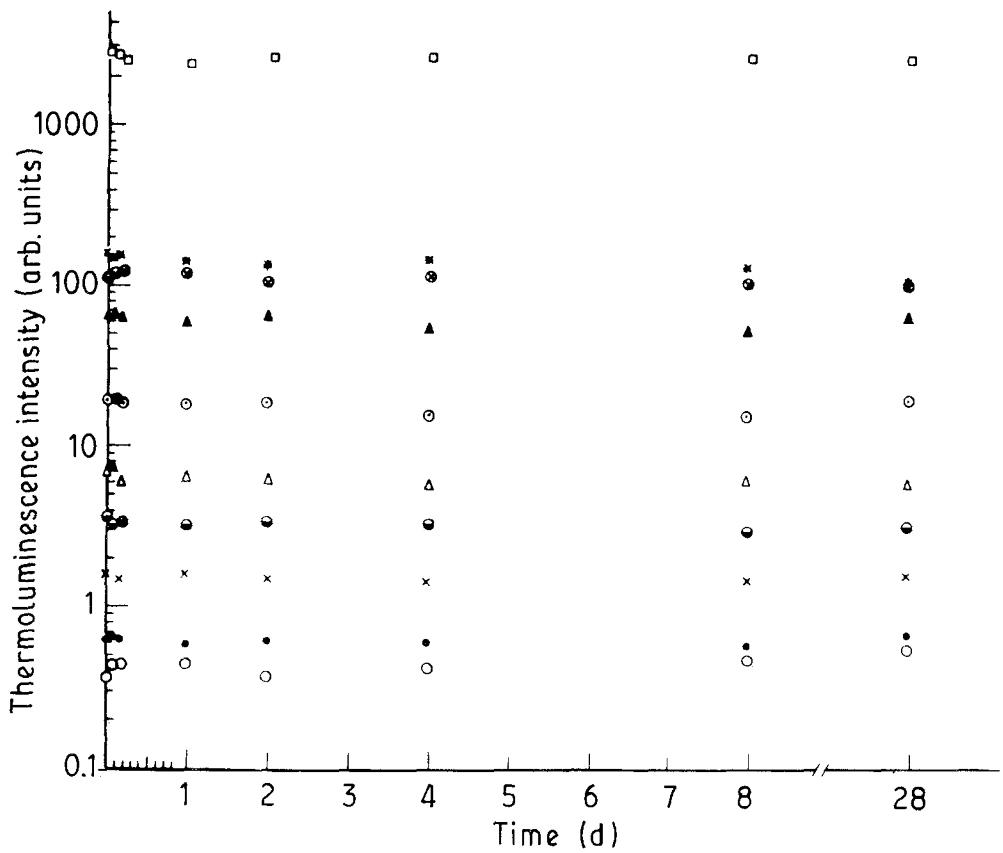

Figure 6 Fading characteristics of TLD-100 irradiated with different doses (Gy): (○) 0.5, (●) 1.0, (×) 2.5, (◒) 5.0, (△) 10.0, (⊙) 20.0, (▲) 50.0, (⊗) 75.0, (∗) 100.0, (□) 700.0.

dominant one, playing the main role in dosimetry work. Results of attempts to investigate the changes in height of the main glow peak at 460 K with radiation dose up to 1000 Gy are shown in Fig. 5. Apparently, the peak height increases linearly with dose up to 10 Gy, above which supralinearity exists showing the same trend observed in the dose–response curve. Supralinearity has been attributed to three main factors [12]: (a) the creation of additional trapping site; (b) the creation of new recombination centres; (c) an increase in the thermoluminescence efficiency of the phosphor.

The results of fading measurements are shown in Fig. 6 for samples irradiated within the dose range 0.5 to 700 Gy. Obviously all samples investigated show practically no significant fading over 28 d. Thus, TLD-100 is suitable for clinical dosimetry from the fading point of view. However, it is worth mentioning that fading in LiF has been reported to reach, in some cases, about 10% over 1 mon [35] this difference may be due to sample preparation conditions.

In conclusion, irradiation of TLD-100 with ^{60}Co gamma-rays does not change the shape of glow curve. A main glow peak at 460 K was observed, its height showed three phases, linearity, supralinearity, and saturation with radiation dose up to 10^3 Gy. Fading in TLD-100 used in this work is negligible over 28 d. The total TL response–dose linear relation can be used over the dose range up to 100 Gy. This linear response could be extended to cover higher doses up to 1000 Gy if the area of the glow curve is plotted against dose to represent the dose–response curve.

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