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Thermoluminescence Kinetic Parameters of TLD-600 and TLD-700 after ^{252}Cf Neutron+Gamma and ^{90}Sr-^{90}Y Beta Radiations *

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The thermoluminescent (TL) properties such as glow curve structure, relative thermoluminescence sensitivity, dose response linearity of lithium fluoride thermoluminescent dosimeters ^{6}LiF:Ti,Mg (TLD-600) and ^{7}LiF:Ti,Mg (TLD-700) are investigated after irradiation ^{252}Cf neutron+gamma and ^{90}Sr-^{90}Y beta sources at room temperature and then the obtained results are compared. The kinetic parameters, namely the order of kinetics b, activation energy E_a and the frequency factor s, are calculated using the computerized glow curve deconvolution (CGCD) program. The effect of heating rate on the glow curves of dosimeters is also investigated. The maximum TL peak intensities and the total area under the glow curves decrease with the increasing heating rate. There is no agreement with the kinetic parameters calculated by the CGCD program for both radiation sources.

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Neutron dosimetry continues to remain as an important and a challenging aspect of radiation protection due to the higher biological effectiveness of neutrons than that of gamma and beta radiations and the intricacy in the responses of the detectors. As the numbers of nuclear facilities, nuclear power plants, medical therapy equipment, accelerators and so on have considerably increased, the need and the importance of personal dosimetry in mixed fields of neutrons and gamma rays has attracted greater attention.[1] LiF-based thermoluminescent dosimeters (TLDs) are the most used passive detectors in industrial and medical applications to determine the absorbed dose in any radiation field due to their small dimensions and their tissue equivalence properties.[2,3] The TL dosimetry in neutron fields is characterized by a particular feature. Such particles are not directly ionizing, thus the neutron response of TLDs depends upon the capture cross section of the constituent elements of the dosimeter and upon the response to the secondary particles produced. Thermoluminescent materials such as LiF:Mg,Cu,P; ^{6}LiF:Mg,Ti and ^{7}LiF:Mg,Ti show a TL signal in which different peaks exhibit different dependences on the LET of the radiation. In fact, the thermoluminescent yield of materials may depend on the LET of the ionizing particles. Thus after exposure to radiation of various LETs, the shape of the glow curves obtained from these dosimeters is expected to change with the LET as well as with the dose.[3–6] Many investigations have been made into the thermal neutron response of TL materials, principally aimed at developing thermal neutron sensitive and also neutron insensitive materials, on the basis of the specific requirements of personal dosimetry. In contrast, few works can be found regarding TL dosimeters exposed to fast neutrons. To the best of our knowledge, there is no such study on the investigation of TL properties and calculating the kinetic parameters of^{6}LiF:Ti,Mg (TLD-600) and ^{7}LiF:Ti,Mg (TLD-700) dosimeters using ^{252}Cf neutron+gamma and ^{90}Sr-^{90}Y sources by the computerized glow curve deconvolution (CGCD) program. The dosimetric characteristics of any TL material mainly depend on the sensitivity, energy response and the kinetic parameters quantitatively describing the trapping–emitting centers responsible for the TL emission. In this study, we investigate and compare the sensitivities, dose response linearity, kinetic parameters and the effects of heating rate on the glow curves of lithium fluoride thermoluminescent dosimeters TLD-600 and TLD-700 after being exposed to ^{252}Cf neutron+gamma and ^{90}Sr-^{90}Y beta-rays. The CGCD method was used to determine the kinetic parameters, namely the order of kinetics b, activation energy E_a and the frequency factor s associated with the dosimetric thermoluminescent (TL) glow peaks of TLD-600 and TLD-700 after different dose levels with neutron+gamma and β-irradiation.

The samples used in this study were commercial TLD-600 (with 95.6% ^{6}LiF) and TLD-700 (with 99.9% ^{7}LiF) from the Harshaw Chemical Co., in the shape of chips measuring $3 \times 3 \times 0.9\,\text{mm}^3$. The samples were firstly annealed at 400±1°C for 1 h prior to irradiation with a specially designed microprocessor-controlled electrical oven followed by a fast cool on an aluminum block. For beta and mixed neutron+gamma irradiations, ^{90}Sr-^{90}Y ($D \approx 0$, 04 Gy/min) and ^{252}Cf (the average neutron energy of the fission spectrum is ∼2.3 MeV and $D \approx 0.08\,\text{Gy/min}$, $D_n/D\gamma = 1$ and 2) sources were used, respectively. At each experimental measurement, four chips were read out. Each chip was read out twice and the second readout is considered to be the background of the reader plus chip. This was subtracted from the first one and all of the analyses have been carried out after the subtraction. The beta source was purchased from Little More Scientific Engineering in the UK and calibrated by the manufacturer on 10 March 1994. On the other hand, the neutron source was purchased from Eckert & Ziegler Isotope Products GmbH and its calibration was also carried out by the manufacturer

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on 1 March 2015. The glow curves were obtained using a Harshaw QS-3500 manual-type TL reader interfaced to a PC where the TL signals were analyzed. Glow curve readout was carried out on a platinum planchet at a linear heating rate of 1℃/s up to 400℃ except for the heating rate experiments. In the variable heating rate (VHR) method, the heating rates were changed between 1℃/s and 10℃/s. The thermoluminescent dosimeters TLD-600 and TLD-700 were irradiated both ^{252}Cf mixed (neutron (n)+gamma (γ)) and ^{90}Sr-^{90}Y beta (β)-sources at different doses between ≈15 and ≈500 Gy for mixed (n+γ) and β-irradiations to observe the changes in the glow curve structures and to check the glow peak intensities and dose dependence effects of the peak positions as a function of exposed dose levels. Some of the selected glow curves of the TLD-600 after irradiating with different dose levels using mixed (n+γ) and β-sources are shown in Figs. 1(a) and 1(b), respectively. The experimental observations have clearly shown that there were no significant changes in the glow curve structures and peak temperatures of glow peaks of TLD-600 with increasing dose levels after different types of radiation sources and levels. This result shows that all the glow peaks of TLD-600 should have first-order kinetics. The measured peak temperature of the highest peak intensity is obtained at around 245±2℃ after both types of irradiations and dose levels. The obtained results of peak temperature for TLD-600 dosimeter are in agreement with the recent studies.[7–14] After being exposed to mixed (n+γ) and β-sources, the total area under the glow curves of TLD-600 is measured for a variety of exposed dose levels. As seen in Figs. 2(a) and 2(b), the total glow curve area under the glow curve of TLD-600 increases linearly with the exposed dose levels in the studied dose levels after both types of irradiations ((n+γ) and β). Some of the recorded glow curves of the TLD-700 after irradiating with different dose levels using mixed (n+γ) and β-sources are shown in Figs. 3(a) and 3(b). The experimental observations have clearly shown that there were no significant changes in the peak temperatures of highest glow peak of TLD-700 with increasing dose level. This result clearly shows that all the glow peaks of TLD-700 should have first-order kinetics. As is seen, the measured maximum peak temperatures of this peak are obtained at around 245±2℃ for both types of sources. On the other hand, the structures of glow curves are considerably different from each other after both types of sources. In contrast, the variations of total areas under the glow curves of TLD-700 after being exposed to mixed (n+γ) and β-sources increase linearly as a function of the exposed dose levels (see Figs. 4(a) and 4(b)).

When the peak intensities of glow curves of TLD-600 and TLD-700 were compared after mixed neutron+gamma and beta sources, it was observed that the glow curve intensities of both dosimeters after irradiation by mixed neutron+gamma are always lower than beta-irradiations. The total glow curve areas and at the same time the maximum peak intensities of TLD-600 and TLD-700 after β-irradiations are approximately 10^4 and 10^3 times greater than after mixed n+γ-irradiations, respectively. It is well known that the heating rate is an important experimental parameter on the glow peak intensities and peak temperatures of many TL dosimetric materials.[15–17] In general, the peak intensity decreases with the increasing heating rate due to thermal quenching.[18] Therefore, the effects of heating rate on the glow curves of the thermoluminescent dosimeters TLD-600 and TLD-700 are also investigated using ^{252}Cf mixed neutron+gamma and ^{90}Sr-^{90}Y beta sources in this study. Some of the recorded glow curves after various heating rates are shown in Figs. 5 and 6 for TLD-600 and TLD-700, respectively.

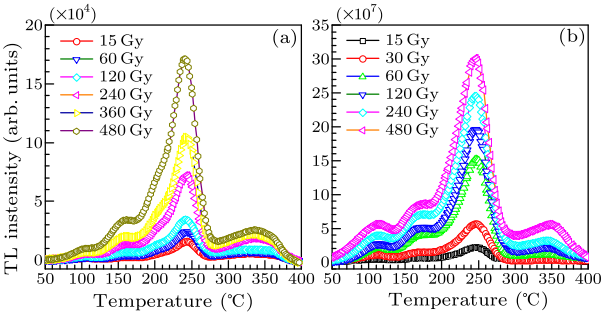

Fig. 1. The glow curves of TLD-600 corresponding to various radiation doses using (a) ^{252}Cf mixed (n+γ)) and (b) ^{90}Sr-^{90}Y β-sources with a heating rate of 1℃/s.

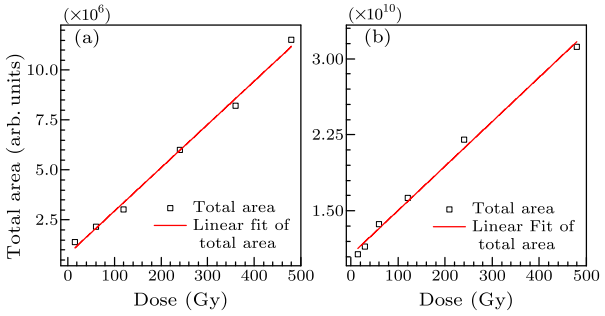

Fig. 2. The total area under glow curves of TLD-600 as a function of the exposed dose levels using mixed (a) (n+γ) and (b) ^{90}Sr-^{90}Y β-sources.

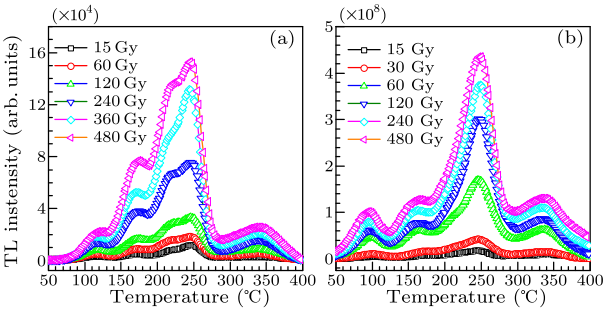

Fig. 3. The glow curves of TLD-700 corresponding to various radiation doses using (a) ^{252}Cf mixed (n+γ) and (b) ^{90}Sr-^{90}Y β-sources with a heating rate of 1℃/s.

It is clearly seen from Figs. 5 and 6 that the peak

temperatures of all glow peaks increase while the corresponding TL intensities and the total areas under the glow curves of both dosimeters decrease with the increasing heating rates (Figs. 7 and 8). On the other hand, as seen from Figs. 5–8, the effects of heating rates on the peak intensities and total area under the glow curves of both dosimeters after β-irradiations are more profound than mixed n+γ irradiations. The CGCD method has become a very popular way to obtain the kinetic parameters from the glow curves of TL materials.[19,20] Therefore, the kinetic parameters such as number of glow peaks, activation energies (E_a), frequency factor (s) and kinetic order (b) for the dosimetric peaks of TLD-600 and TLD-700 were obtained by the CGCD method. In this study, the following general order kinetic analytical equation is used by the approximation for $b \approx 1.1$,[21]

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

where $\Delta = \frac{2kT}{E}$, $\Delta_m = \frac{2kT_m}{E}$, and $Z_m = 1+(b-1)\cdot\Delta_m$.

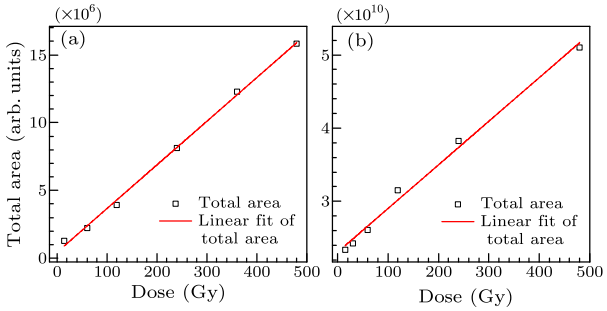

Fig. 4. The total area under glow curves of TLD-700 as a function of the exposed dose levels using (a) mixed (n+γ) and (b) ^{90}Sr-^{90}Y β-sources.

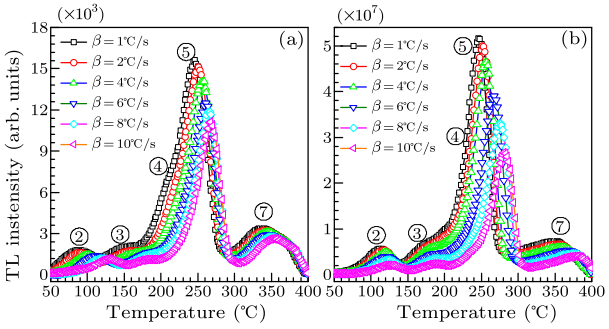

Fig. 5. The glow curves of TLD-600 measured after various heating rates between 1 and 10°C/s. Measurements are taken after irradiation of 15 Gy using (a) ^{252}Cf mixed n+γ and (b) β-sources.

The number of glow peaks is not a free fitting parameter during the deconvolution of the measured glow curve into individual components in the CGCD programs. If the number of peaks is unknown, it can be found by fitting the glow curve several times with a different number of components to obtain the best-fit result. In this study, after many tries with different numbers of glow peaks, it is observed that the glow curve structures of TLD-600 and TLD-700 are well described by a linear combination of at least seven glow peaks. In this case, a reasonably good fit is always obtained. Some of the analyzed and deconvoluted glow curves of both dosimeters after mixed (n+γ) and β-irradiations are shown in Figs. 9 and 10. The obtained results of E_a, s and b using the CGCD methods are listed in Tables 1 and 2. The first column in the table shows the result of neutron+gamma irradiation and the second column beta irradiation of the dosimeters. It is clear that the obtained results are in agreement with each other. These results are also in agreement with Refs. [7,11,22].

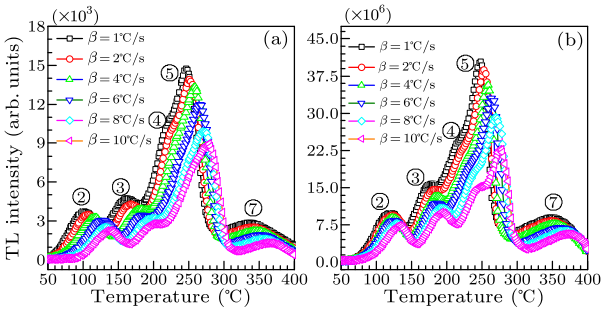

Fig. 6. The glow curves of TLD-700 measured after various heating rates between 1 and 10°C/s. Measurements are taken after irradiation of 15 Gy using (a) ^{252}Cf mixed n+γ and (b) β-sources.

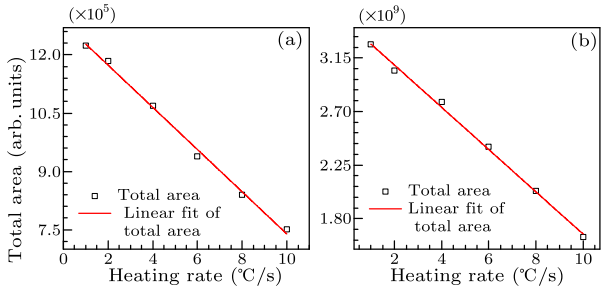

Fig. 7. The total area under the glow curves of TLD-600 as a function of the heating rates after irradiation of 15 Gy using (a) ^{252}Cf mixed n+γ and (b) ^{90}Sr-^{90}Y β-sources.

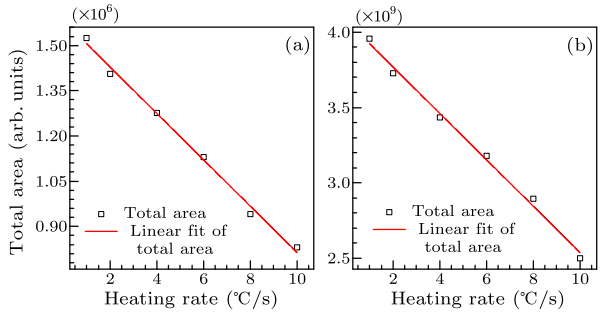

Fig. 8. The total area under the glow curves of TLD-700 as a function of the heating rates after irradiation of 15 Gy using (a) ^{252}Cf mixed n+γ and (b) ^{90}Sr-^{90}Y β-sources.

Table 1. The obtained kinetic parameters of TL glow peaks of TLD-600 and TLD-700 by the CGCD method for 15 Gy for $\beta = 1$℃/s.

Peak		Neutron+gamma irradiated glow peaks				β-irradiated glow peaks			
	TLD-600	T_{max} (K)	E (eV)	s (s^{-1})	b	T_{max} (K)	E (eV)	s (s^{-1})	b
2		418	0.63	1×10^{6}	1.00	455	0.72	3×10^{6}	1.00
3		472	1.06	1×10^{10}	1.00	504	1.062	1×10^{15}	1.00
4		518	1.22	3×10^{10}	1.00	523	2.10	1×10^{19}	1.06
5		557	2.00	8×10^{16}	2.00	564	2.03	1×10^{17}	2.00
	TLD-700	T_{max} (K)	E (eV)	s(s^{-1})	b	T_{max} (K)	E (eV)	s (s^{-1})	b
2		440	0.88	6×10^{8}	1.25	445	1.00	1×10^{10}	1.27
3		487	1.43	4×10^{13}	1.00	486	1.56	1×10^{15}	1.30
4		518	1.75	6×10^{15}	1.24	519	2.13	4×10^{19}	1.21
5		533	2.09	5×10^{18}	1.37	549	2.00	1×10^{17}	2.00

Table 2. The obtained kinetic parameters of TL glow peaks of TLD-600 and TLD-700 by the CGCD method for 480 Gy for $\beta = 1$℃/s.

Peak		Neutron+gamma irradiated glow peaks				β-irradiated glow peaks			
	TLD-600	T_{max} (K)	E (eV)	s (s^{-1})	b	T_{max} (K)	E (eV)	s (s^{-1})	b
2		429	0.95	1×10^{10}	1.25	437	0.90	1×10^{9}	1.30
3		478	1.34	8×10^{12}	1.25	492	1.04	2×10^{9}	1.00
4		515	1.73	6×10^{15}	1.25	522	1.91	2×10^{17}	1.30
5		558	1.39	1×10^{11}	1.04	547	2.10	1×10^{18}	1.30
	TLD-700	T_{max} (K)	E (eV)	s (s^{-1})	b	T_{max} (K)	E (eV)	s (s^{-1})	b
2		441	1.03	3×10^{10}	1.16	428	0.78	8×10^{7}	1.30
3		485	1.24	4×10^{11}	1.07	491	0.88	4×10^{7}	1.00
4		519	1.51	2×10^{13}	1.14	524	1.82	2×10^{16}	1.23
5		564	1.16	1×10^{9}	1.13	547	2.29	1×10^{20}	1.92

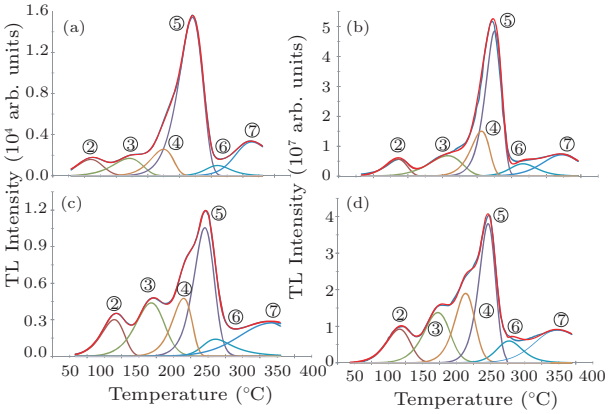

Fig. 9. Computerized glow curve deconvolution results of TLD-600 and TLD-700 dosimeters irradiated with 15 Gy by (a) mixed (n+γ), (b) beta, (c) mixed (n+γ), and (d) beta for 1℃/s.

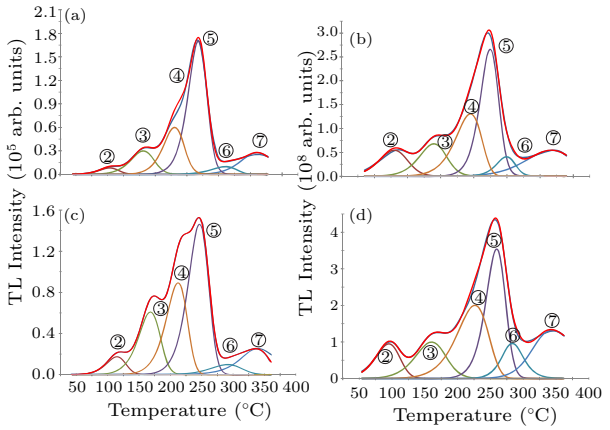

Fig. 10. Computerized glow curve deconvolution results for TLD-600 and TLD-700 dosimeters irradiated with 480 Gy by (a) mixed (n+γ), (b) beta, (c) mixed (n+γ), and (d) beta for 1℃/s.

TL dosimeters are now being increasingly used to monitor neutron doses. There is a need to monitor neutron doses in mixed n, γ fields. The sensitivity to neutrons of TL phosphors depends on their thickness, isotropic composition, irradiation condition and, to some extent, on the activators.[23] Fast neutron detectors take advantage of the fact that an important fraction of the neutron's kinetic energy can be transferred to the target nucleus producing an energetic recoil nucleus. This will behave in a similar way to a heavy charged particle, slowly losing energy while passing through the moderator. Using TL detectors for the dosimetry in mixed neutron-gamma fields requires knowledge of their response to neutrons over a broad energy range. For the calculation of the neutron fluence response, the conversion of the energy deposited by the secondary heavy charged particles to the TL reading effect must be considered.[24] TLDs' readings upon heating come from the light conversion of the energy stored in the material after electronic excitations by the neutron-induced charged recoils. Here ^{6}Li capture and ^{6}Li or ^{7}Li elastic scattering events are the most important contributions to the kerma factor, the dominant recoils being triton and alpha particles for thermal and epithermal energies and lithium recoils for the fast portion of the spectrum.[25] Several researchers have studied the glow curves of LiF dosimeter with various types and dose of irradiation and found several peaks present.[26,27] It has pointed out that the differences could arise by different manners of excitation in thermoluminescent materials for the kinds of radiation when the irradiation dose is not so high. The difference in glow curves of ^{6}LiF and ^{7}LiF dosimeters irradiated by neutron may be caused by the ^{6}LiF (n, α)^{3}H reaction in the LiF dosimeter.[28] Lithium fluoride thermoluminescent

dosimeters TLD-700 are widely used for the measurement of gamma ray dose. Unlike TLD-600 the thermoluminescence in the TLD chip is primarily generated by densely ionizing secondary alpha particles and tritons produced in the dosimeter mass through ^{6}Li(^{1}n, ^{4}He)^{3}H reaction, the thermoluminescence in a TLD-700 chip is created by the ^{7}Li(^{1}n, γ)^{8}Li reaction as well as the energy transferred to dosimeter material via elastic and inelastic scattering of the impinging neutrons.[29] In this work, we have investigated the change in the shape of the glow curves and the dose response properties of TLD-600 and TLD-700 dosimeters using mixed neutron+gamma and beta sources. The dose ranges between 15 and 500 Gy for mixed (n+γ) and β-irradiations are investigated. As can be seen in Figs. 1 and 2, there is no significant change in the shape of the glow curves of TLD-600 for exposure to beta and neutron fields, while the difference in the relative intensity of TLD-600 is remarkable for exposure to beta rays. The glow curves of the TLD-700 when exposed to beta rays and neutron field shows some small variations at the low temperature side due to gamma contributions.[7] The glow curves obtained using mixed (n+γ) and β-sources for TLD-600 and TLD-700 exhibit linear dose response curve between 15 and 500 Gy. The TL glow curves of TLD-600 and TLD-700 are also measured after various heating rates between 1 and 10°Cs^{-1}. It is observed that with the increasing heating rates, the TL intensities of both the samples are reducing and as well its peak temperatures are shifting into the higher temperature side and the total area under the glow curves may be decreasing. The kinetic parameters are obtained by the CGCD method and the results are tabulated in Tables 1 and 2. As seen from the tables there is no agreement with the kinetic parameters calculated by the CGCD program for low and high doses for both radiation sources. The reason for this LiF based dosimeters have very complex glow curves and consist of many glow peaks which are close to each other. Therefore, even small changes in the shape of the glow curves significantly affect the calculated kinetic parameters of all glow peaks.

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