In Defense Of Thermoluminescence Dosimeter Zero Dose Readouts

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Abstract: Zero dose readout of thermoluminescence dosimeters is a very important parameter which is considered in all accurate dosimetry procedures in order to correct for the additive doses arising from other sources than irradiation processes, however, in many cases this parameter is neglected. In this paper, effects of zero-dose readings for three different thermoluminescence dosimeters glow curves were investigated. Dosimeters included in this study are: sensitized TLD-700, sensitized TLD-600, and CaF_2:Tm (TLD-300). Deconvolution of glow curves was performed in order to investigate individual behavior of each glow peak using a GCAFIT glow curve analysis software. It was found that readout of zero doses usually accompanied by changes in glow curves quantitatively (i.e. area under the curve increases), and qualitatively (relative changes in glow curve peaks intensities and their maxima positions). It is recommended that, even if the zero dose value is to be neglected as an added value to be subtracted, zero dose readouts should be performed for enhancements arise in thermoluminescence glow curves and hence better performance. This behavior is verified even LiF detectors were sensitized or not. In contrary, for CaF_2:Tm (TLD-300), a little effect is noticed because there is no thermal quenching effect and competing deep trap in this material.

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Key words: Zero dose readout –Pre-irradiation Background – GCAFIT software - TLD –Sensitized LiF – CaF_2.

1. Introduction:

Dosimetry is a fundamental part in quality programs assuring that the irradiation procedure is carried out according to standard regim. To assess the radiation doses precisely, the sources of dose errors should be identified and minimized (Robertson 1981 and Rocha et al., 2003). There are several techniques for radiation dosimetry (Schönbacher H. et al., 2009), among all these techniques, thermoluminescence dosimetry (TLD) is now the most widely used technique in many fields like personal and environmental radiation exposure (Gilvin and Perks, 2010, Sandouqa et al., 2011), geochronology and space dosimetry. In addition, TLDs are extensively used in both of diagnostic and therapeutic medical applications (Abd El-Hafez et. al., a and b, Czopyk et al., 2007, Gual M. et al., 2011, Sharma et al., 2011 and Shousha et. al., 2011), and in other health related aspects (Vandana et al., 2011) as well.

There are many of well-known thermoluminescent (TL) dosimeters used in radiation monitoring due to their high sensitivity, stability and tissue equivalency (Haiyong Jung et al., 2003). Thermoluminescence phenomena results in forming the characteristic glow curves for the phosphor material. The glow curves obtained for each material are different, and each glow peak is ascribed to the recombination centers and is related to traps (Furetta, 2011).

Typically, standard practice procedures for TL dosimetry involve pre-irradiation background readings of TLDs especially at low dose levels, to be subtracted from readings of irradiated dosimeters (ASTM 1998, Izewska, et al., 2007). However, some experimentalists do skip this procedure at different circumstances: (1) if dosimeters are being irradiated to high doses compared to which background values could be neglected, (2) when high level of dosimetry with minimized uncertainties is not within the scope, (3) when dealing with large number of dosimeters, it was thought to be enough to select randomly few dosimeters for pre-irradiation background measurements instead of reading all dosimeters for saving time, and (4) when the glow curves is a subject of an analytical study regardless of doses delivered (Ixquiac-Cabrera, et al., 2011, Harvey, and Kearfott, K. 2011).

Recently, (Abd El-Hafez and Maghraby 2011) noticed that readouts of LiF based TL dosimeters background before irradiation (regardless of values of background) can enhance TL output and may cause changes to the glow curves both qualitatively in terms of peaks positions and peaks relative intensities, and quantitatively in terms of areas under the curve. Hence, it is of some importance to investigate if this enhancement in glow curves is observable in other TL materials and the effect of sensitization.

One of the major problems that afflict TLD in general is the complexity of the glow curve obtained with many TLD materials. Hence, a glow curve analytical toolkit (GCAFIT) was used in this study for assessing individual behavior of each peak in terms of both intensity and position (Abd El-Hafez et al., 2011).

2. Experimental Work

2.1. Radiation source and dose determination:

The irradiation were performed using the ^{137}Cs gamma source model GB-150, was manufactured by Atomic Energy of Canada Limited on April 1970 with original activity of 1000 Ci.

Air kerma (K_{air}) was determined according to the International Atomic Energy Agency (IAEA) code of practice TRS-(381) (IAEA, 1997). K_{air} determination was performed using the secondary standard dosimetry system of the National Institute of Standards (NIS) - Egypt, which is composed of the NPL-2560 electrometer (UK) and its NE2561 ionization chamber (UK),. The secondary standard calibration system was calibrated at the Bureau International des Poids et Mesure (BIPM). The expanded uncertainty associated to K_{air} determination was about 0.9 % at 95% level of confidence (coverage factor = 2).

2.2. Thermoluminescent dosimeters and reader:

Three types of TL dosimeters were incorporated in this study, those types are sensitized TLD-700, sensitized TLD-600, and CaF_2:Tm (TLD-300) in the form of chips with dimensions of 6.4 x 6.4 x 0.9 mm^3. The Harshaw 4500 TLD Reader is equipped with two photomultiplier tubes that can read independently; the reader operates on WinREMS software, which runs under Windows® on a separate computer. All dosimeters and reader were manufactured by Harshaw Chemical Co. (USA).

2.3. Experimental procedures:

The sensitization process was performed through exposing the TL dosimeters to about 20 KGy gamma rays. TL dosimeters were annealed, and kept in dark. All dosimeters were divided into two sets; each set is composed of eight dosimeters. Experimental procedures are composed of two main steps, in the first step: background readings were performed for the first set (S_1), while the second set (S_2) were left without reading of the background. The two sets were grouped together and exposed to a well-defined radiation dose from ^{137}Cs gamma source. Dosimeters readings were acquired using linear heating rate equals to 1 ^oC /s over the range 100 - 400 ^oC. This low heating rate was used to avoid extreme overlapping in glow peaks (Abdel-Hafez, 1999, Yazici. 2004). After readout of dosimeters, annealing was performed as one hour at 400 ^oC, and slowed down at room temperature, followed by two hours at 100 ^oC for LiF and 1 hour for CaF_2.

The second step involves the reverse of the two sets, where pre-irradiation background was evaluated for S_2 while S_1 were left without reading of the background. The two sets were grouped together again and exposed to almost same radiation dose. Dosimeters readings were acquired using the same conditions.

A control set of TLD-100 was used during the two steps to assure that irradiation process in the two steps was similar; annealing and readout conditions were performed in the same way as the investigated dosimeters.

Thus the same dosimeter passed the two cases; the first case (Case A) represents the readout of dosimeters irradiated without experiencing pre-irradiation background measurements, while the second case (Case B) represents the readout after gamma irradiation for dosimeters experienced pre-irradiation background measurements (Abd El-Hafez and Maghraby 2011).

2.4. Deconvolution:

Glow curves were deconvoluted using GCAFIT software which is a computerized glow curve analysis program was written using the MATLAB technical computing language and developed at the National Institute. of Standards (NIS), Egypt (Abd El Hafez et al., 2011). GCAFIT software uses the nonlinear least-square method with the Levenberg–Marquardt algorithm. The results of the fitting process are estimates of the model coefficients, It worth to confirm that deconvolution processes were performed over the experimentally studied range of temperature (100 - 400 ^oC), hence, whole first peak (P_1) was not involved.

3. Results and Discussion:

3.1. Sensitized TLD-700:

Figure (1) represents the two cases (A and B) for sensitized TLD-700 dosimeters where triangles represent case (A) while circles represent case (B). Obviously there are several changes occurred, first of all the averaged total area under the curve has been increased almost twice (211 % ± 1.1 %) when the pre-irradiation background readings were considered. When comparing the two glow curves, it is easy to notice that the increase in sensitivity is not uniform over the heating range; hence a glow curves deconvolution was a necessity for estimating individual behavior of each peak separately.

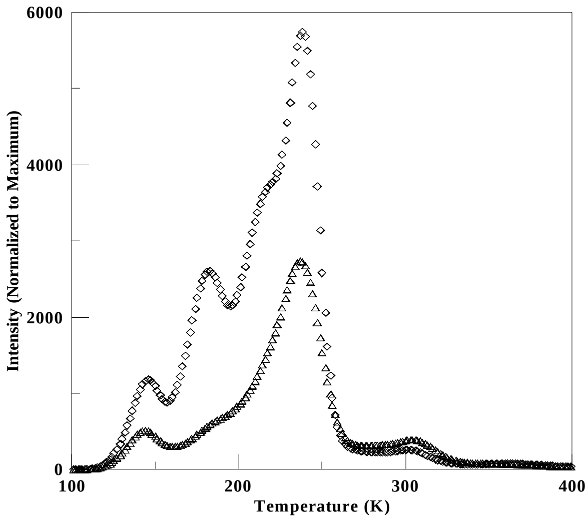
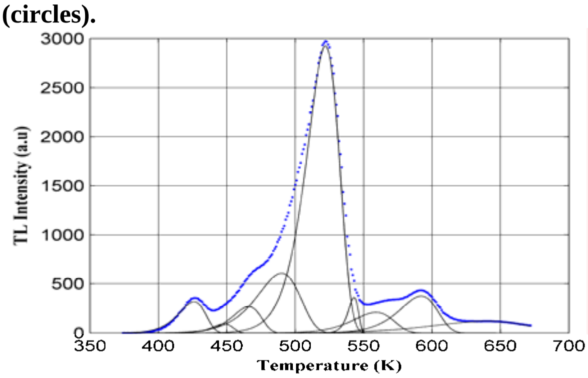


Figure (1): LiF (TLD-700) glow curve with heating rate 1 ^{o}C/s, Case A (triangles), and Case B (circles).



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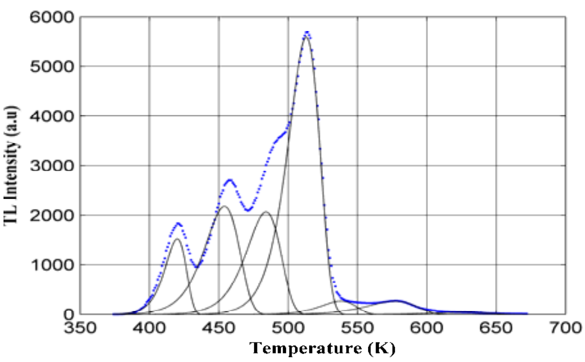


b

Figure (2): Computerized Glow Curve Analysis (GCAFIT) of one of TLD-700 samples, a- Case A, b- Case B.

Figure (2) represent the deconvoluted glow curves of gamma irradiated TLD-700 sensitized dosimeters: Figure (2-a) represents the deconvoluted glow curve of dosimeters irradiated without performing pre-irradiation background readouts, it has been deconvoluted into nine peaks (P_2 , P_{2a}, P_3, P_4, P_5, P_6, P_{7a}, P_7, and P_8). The regression coefficients of fitting (R^2) were ranged from 0.898 to 0.998. On the other hand, case (B) dosimeters glow curves were deconvoluted into seven peaks (P_2: P_8) as shown in Figure (2-b), and The regression coefficients of fitting (R^2) were ranged from 0.910 to 0.999.

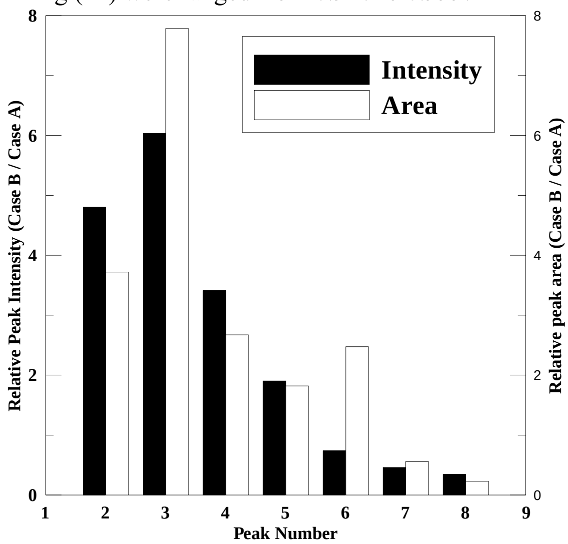


Figure (3): Changes in intensities and areas of resolved peaks in one of TLD-700 dosimeters, Solid denotes to intensity, and striped denotes to area.

Figure (3) represents the relative changes both in peak intensity and peak area (Case B / Case A). From the Figure, it is clear that maximum change either of peak intensity or peak area point of view was for the third peak (P_3), where peak intensity was increased six times and peak area was increased by a factor of 7.79 as a result of zero dose readout. Other peaks show remarkable increments in both of peak intensity and peak area. Those peaks are P_2 with relative change in peak intensity and peak area factors of 4.8 and 3.7 respectively, followed by P_4 whose intensity has been increased by a factor of 3.4 and 2.7 for peak intensity and peak area increment. P_5 was increased in peak intensity by a factor of 1.9 and in peak area by a factor of 1.8. P6 was increased in peak area by a factor of 2.5 while its intensity decreased a little and the ratio is 0.74. Remaining peaks (P_7 and P_8) showed a reverse behavior where both have been decreased in their intensities (ratios are 0.46 and 0.36) and their areas (ratios are 0.56 and

0.23) respectively, noting that P_7 in case (B) has been compared to the sum of P_7 and P_{7a} in case (A).

Other changes in glow curves arose after switching to case (B) were changes in peak maxima positions (T_{Max}), as represented by Figure (4) which clarifies the shift of two peaks (P_3 and P_7) toward higher temperature direction, which has been shifted by 6 ^{o}C and 18 ^{o}C respectively. Remaining peaks showed a shift toward lower temperature direction: 6 ^{o}C for P_2 and P_4, 5 ^{o}C for P_6, and 13 ^{o}C for P_8.

This shift in the temperature was held responsible for the increase in the contribution of non-radiative transitions due to the presence of the competing traps. It was inferred that the glow peaks occurring at higher temperatures must exhibits higher thermal quenching. This is attributed to that the energy dissipated in an indirect transition, however, is much less than the band-gap energy and may thus be dissipated either radiatively (via photons) and non-radiatively (via phonons). Non-radiative capture of free carriers takes place because the lattice vibrations cause the energy level to change its position in the forbidden gap. For large enough vibrations, the energy level crosses into the conduction band and captures a free electron. The lattice relaxation which follows the capture lowers the position of the level back into the energy gap, the excess energy being propagated as lattice phonons (McKeever, 1985). However, the glow curve is controlled by the release of the charge carriers from traps and not by the properties of the luminescence centers (Horowitz, 1984).

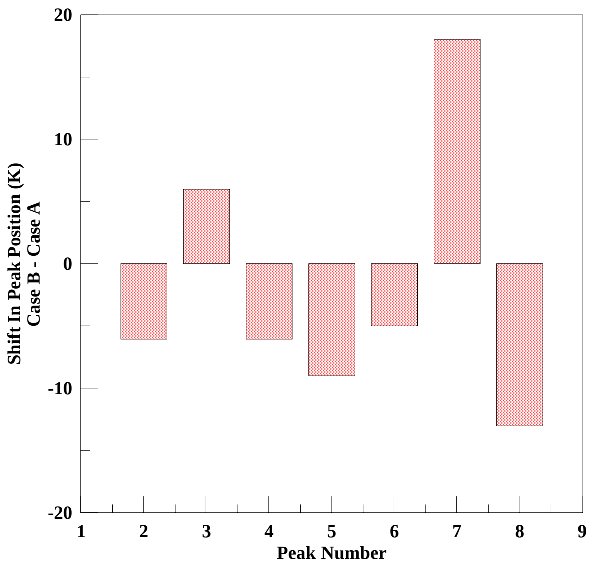


Figure (4): Possible shift in peak position (T_{Max}) of resolved peaks in one of TLD-700 dosimeters.

3.2. Sensitized LiF (TLD-600):

Figure (5) represents glow curves of irradiated TLD-600 sensitized dosimeters, where triangles denote to case (A) and circles denote to case (B). As was the case in sensitized TLD-700, averaged total area under the glow curve has been increased dramatically by a factor of 2.06 ± 0.06 when performing pre-irradiation background readouts (Case B). As shown in Figure (6-a), best fit of case (A) glow curve results in eight peaks (P_2, P_3, P_4, P_5, P_6, P_{7a}, P_7, and P_8). Coefficients of determination (R^2) of fitting were in the range 0.878:0.998, while case (B) glow curve deconvolution results in seven peaks (P_2: P_8) with (R^2) in the range of 0.847: 0.999 as represented in Figure (6-b) in which peak P_{7a} is not seen.

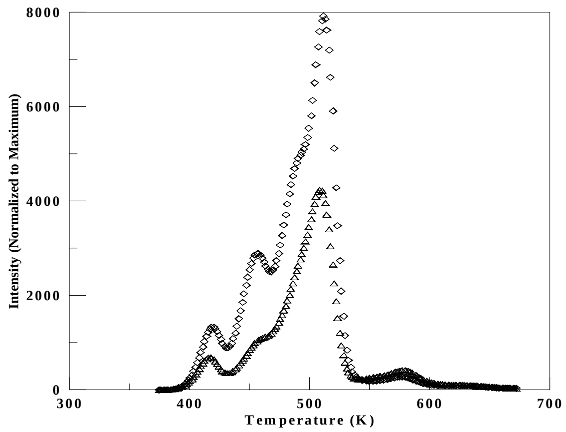
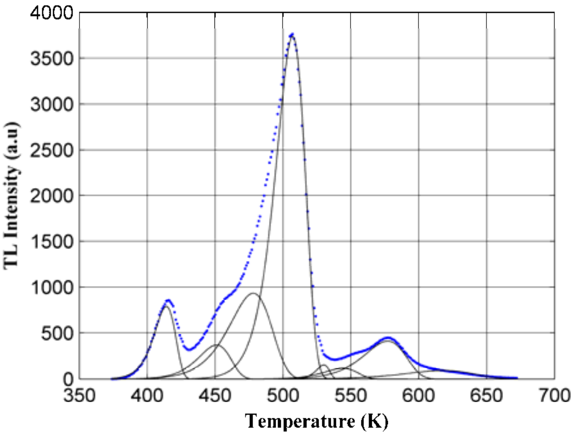


Figure (5): TLD-600 glow curve with heating rate 1 K/s, Case A (triangles), and Case B (circles).

Figure (7) represents the changes in glow curves of LiF (TLD-600) sensitized dosimeters in terms of peak intensities and areas. Maximum increase belongs to P_3, where peak intensity has been increased by a factor of 5.39 and peak area has been increased by a factor of 4.60 after switching to case (B), followed by P_4 which has been increased in intensity by a factor of 3.45 and in area by a factor of 2.87. P_5 has been almost doubled in both of peak intensity (factor of 2.12) and peak area (factor of 2.04). Also, a slight increase was noticed in P_8 intensity and area by factors of (1.17and1.39) respectively. P_2 and P_6 shows a reasonable stability where P_2 intensity has been changed only by a factor of 0.82 and its area by a factor of 0.91, while P_6 intensity has been changed by a factor of 0.91 and a factor of 1.26 for its area. On the other hand, P_7 was the only peak which exhibited a decrease in intensity and the factor is 0.58 and in area the factor is 0.50 after switching to case (B).

The change in resolved peaks maxima positions (T_{Max}) as a result of considering pre-irradiation background reading is represented by Figure (8). All peaks maxima positions have been shifted toward higher temperature direction except for P_6 which shows a shift (3 ^oC) toward lower temperature direction. P_8 showed maximum shift (10 ^oC), followed by both of P_4 and P_5 (9 ^oC), P_2 (7 ^oC), P_3 (6 ^oC), and a slight shift was in case of P_7 (2 ^oC).



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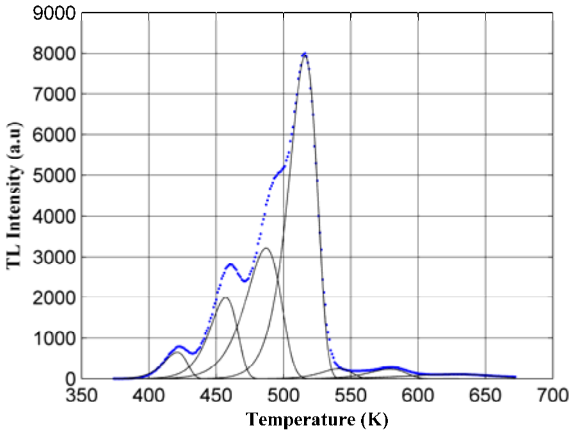


b

Figure (6): Computerized Glow Curve Deconvolution (GCAFIT) of one of TLD-600 samples, a- Case A, b- Case B.

3.3. CaF_2 (TLD-300)

Figure (9) represents the two cases: A, and B, triangles curve represents case A, and the circles one represents case B. The average of the total area under the glow curve after considering pre irradiation readouts has been increased by a factor of about 1.13 with percentage standard deviation σ equals to ± 7.8 %. As TLD models can be classified as models in which the critical processes occur during the absorption of radiation, and models in which the critical processes occur during heating (i.e. during TL readout). The evidence, at least for LiF-based materials, clearly shows that the critical mechanism is that of competition-during the stage of TL readout. During heating, electrons released from traps, may either recombine with trapped holes to produce TL or be retrapped in deeper traps, which act as competing centers. The competition – during heating process was first suggested by Rodine and Land 1971 and was later analysed mathematically by Kristianpoller et. al. 1974.

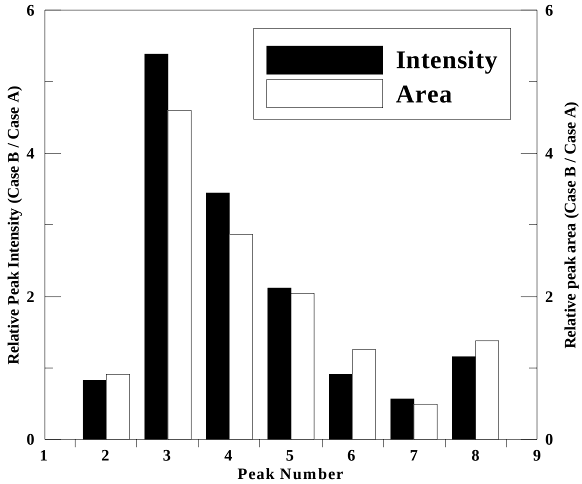


Figure (7): Changes in intensities and areas of resolved peaks in one of TLD-600 dosimeters, Solid denotes to Intensity, and striped denotes to Area.

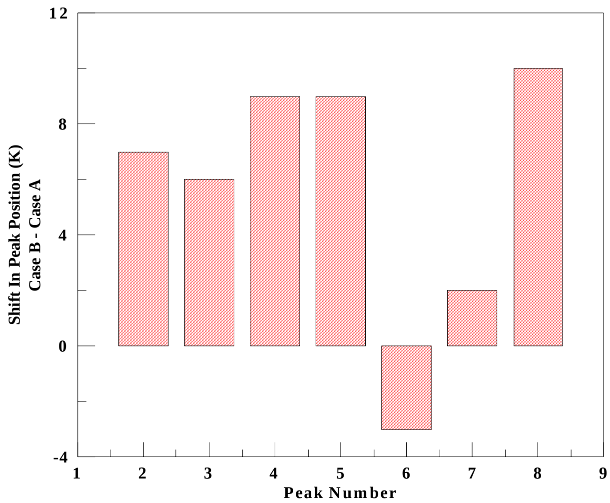


Figure (8): Possible shift in peak position (T_{Max}) of resolved peaks in one of TLD-600 dosimeters.

Deconvolution of TLD-300 dosimeters glow curves results at best fit in five peaks namely (P_2 : P_6) in both cases: Case (A) in Figure (10- a), and case (B) in Figure (10-b).

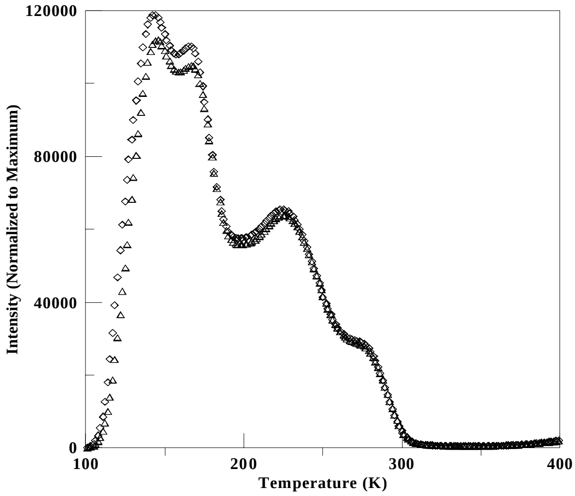
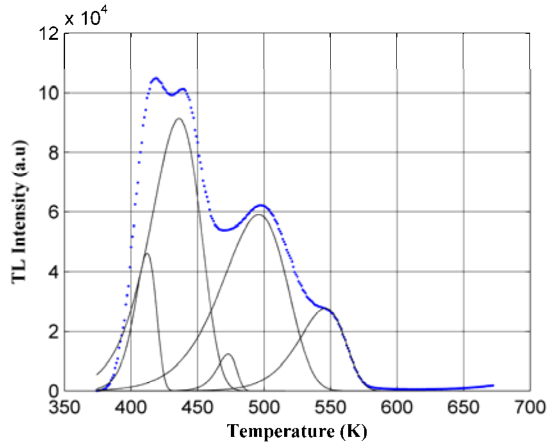


Figure (9): TLD-300 glow curve with heating rate 1 K/s, Case A (triangles), and Case B (circles).



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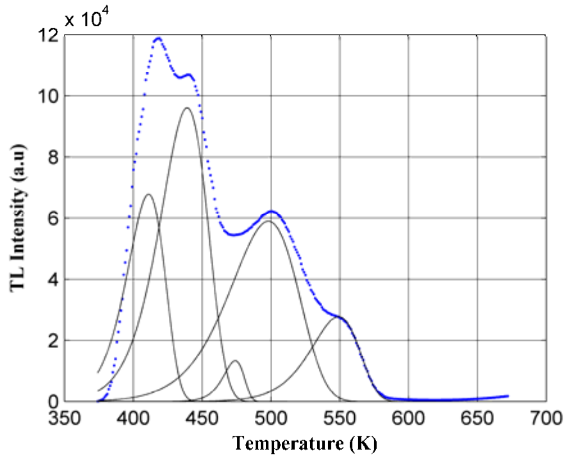

b

Figure (10): Computerized Glow Curve Deconvolution (GCAFIT) of one of TLD-300samples, a- Case A, b- Case B.

As shown in Figure (11), the relative changes in averaged intensities of different peaks were as the following: P_2 intensity has increased by a factor of 2.45 in case B compared to its value in case A, and its area has been increased by a factor of 1.86, Also, P_3 intensity has increased by a factor of 1.63, and its area has been increased by a factor of 1.57, while P_4 shows a slight increase as a result of zero dose readout in intensity and area by factors of 1.08 and 1.13 respectively. On the other hand P_5 exhibited a decrease in intensity and the factor is 0.65 and a slight decrease in Area and the factor is 0.8. P_6 intensity has been decreased and the factor is 0.66 while its area almost was not changed (1.08 factor of change).

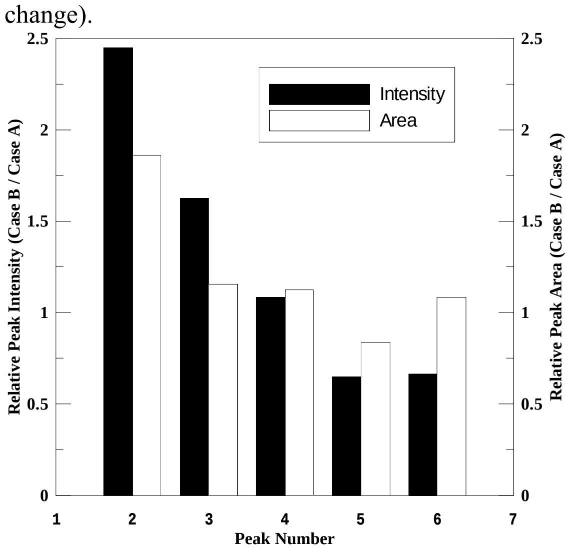

Figure (11): Changes in intensities and areas of resolved peaks in one of TLD-300 dosimeters, Solid denotes to Intensity, and striped denotes to Area.

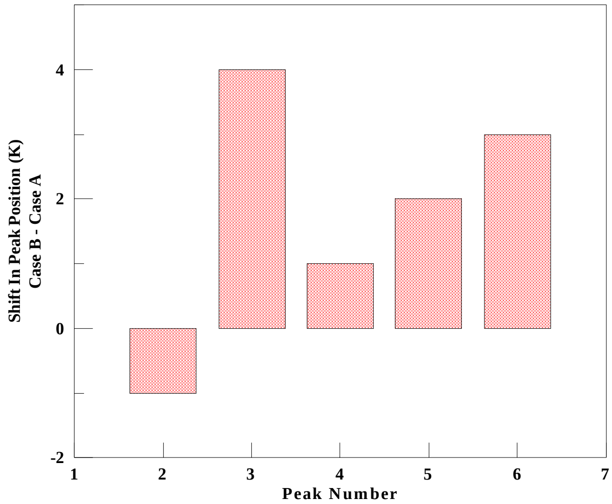

Figure (12): Possible shift in peak position (T_{Max}) of resolved peaks in one of TLD-300 dosimeters.

Figure (12) represent possible shift in peaks maxima positions (T_{Max}) after performing zero-dose readings for TLD-300 dosimeters. Maximum shift toward higher temperature side was in P_3 (4 ^oC), followed by P_6 (2 ^oC), then P_5 (2 ^oC), and P_4 (T_{Max}) has been shifted by 1 ^oC only. Toward lower temperatures, a slight decrease in (T_{Max}) of P_2 has been occurred (1 ^oC). Kafader et. al., 2009 showed that the linearity of glow peaks of TLD-300 crystal is not affected with the change of heating rate. i. e. there is no thermal quenching. Thermal quenching was understood to be due to the increased probability of nonradiative transitions competing with the radiative transitions.

4. Conclusion

It may be concluded that pre-irradiation background measurements for the studied TLDs is not only important for evaluation of residual doses when considering high level of accuracy, but its most importance (as a form of heat treatment) in the enhancement of their sensitivities and adjustment of glow curves peaks distribution and relative ratios. Impacts on glow curves enhancement were dramatically in cases of sensitized TLD-700 and sensitized TLD-600, and were minor in case of TLD-300 dosimeters.

As TLD models can be classified as models in which the critical processes occur during the absorption of radiation, and models in which the critical processes occur during heating (i.e. during TL readout). The evidence, at least for LiF-based materials, clearly shows that the critical mechanism is that of competition-during the stage of TL readout. CaF_2:Tm (TLD-300) clearly shows that the critical mechanism is that of competition-during the stage of absorption of radiation. Hence, it is much recommended to perform background measurements before irradiation regardless how high the radiation dose is, and even the background value will be neglected either if the thermoluminescent dosimeters were sensitized or unsensitized. The condition of better enhancement is that the TL material should have the critical mechanism is that is that of competition during the stage of readout. Further studies should be performed soon regarding behavior of each resolved peak as a function of radiation quality and dose level effects.

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