Using the different LET response of high and low temperature peaks in LiF:Mg,Ti for mixed neutron-gamma dosimetry – a controversial subject

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Abstract. The main high temperature peaks in the glow curves of LiF:Mg,Ti (assigned as 6 and 7), which appear at about 260°C-270°C, are known to have an enhanced response to higher LET radiation (as α particles) than to lower LET radiation (as photons). This effect can be used to discriminate between neutron and gamma radiations. In most works in the past a slow heating rate of up to \( \sim 10^0 \text{C/s} \) was employed for readout. However, in routine dosimetry a higher heating rate, normally about 25°C/s, is used. The high rate heating causes a shift of the high temperature peaks towards 300°C, which is also the limit when using TLD cards with Teflon coating of the chips. Thus, marginal conditions are present, which caused controversy on the justification of the experimental procedures. Besides, there are claims that the method may suffer from the non-constancy of the crystals impurity content and non-linearity of the peak 7 response to gamma dose. Published data is presented, along with new experimental results, leading to the conclusion that in spite of the technical constraints, the method has potential for mixed neutron-gamma dosimetry and further research efforts should be invested.

KEYWORDS: peaks; HT; thermoluminescence; neutron; dosimetry.

1. Introduction

LiF:Mg,Ti is the most widespread thermoluminescent dosimetry (TLD) material, and its glow curve exhibits about 14 peaks at different temperatures. The main dosimetry peaks (in common nomenclature peaks 4 and 5) appear at about 210°C, but there are also higher temperature peaks (HT), the most prominent being peaks 6 and 7, which appear at about 260°C-270°C. Almost from the beginning of studying the thermoluminescence and its application to dosimetry, it was observed that the HT peaks respond differently when irradiated by low and high LET radiation, pointing to the possibility to use this effect for mixed thermal neutron-gamma dosimetry. Already in 1970, Busuoli et al. [1] presented results on evaluation of the light output from low temperature peaks (dosimetry peaks) and high temperature peaks by a dual heating cycle. The authors concluded that the method is valid in practice for mixed field evaluations in a defined range and is reliable for normal personal dosimetry. Also Marshall et al. [2] published similar results based on dual temperatures readout in one cycle. Nash and Johnson [3] published in 1977 their results on the subject. They concluded that the accuracy of the method based on HTTL (HT thermoluminescence) was reasonable and comparable to that of the pair \(^{6}\text{Li}^+\text{Li}^\text{+} \) method. The two peaks method was applied routinely in 1976 at Naval Research Labs./USA. Uray [4] performed experiments with similar crystals, applying a slow heating rate of 3°C/s. The light intensity was measured in two separate regions: from 100°C to 225°C and from 225°C to 400°C. Empirical constants were defined for the two regions for gamma and neutron radiations, which can be used for mathematical evaluation of the doses in a mixed field. Uray [4] found evidence of a supra-linear component in the HTTL region also for the lower dose range, which seems not to be significant due to the statistical uncertainty. Additional experiments to resolve High Temperature and Low Temperature peaks information in a mixed field are presented by the same author [5], this time by using an adaptation of an unfolding program developed for gamma spectroscopy. Horiuchi at al. [6] published in 1992 a work on simultaneous evaluation of neutron and gamma doses with a single \(^{6}\text{Li} \) TLD by deconvoluting the glow curves into 7 peaks, with shapes approximated by gaussian functions. A TLD reader using a heating plate with a 5°C/s heating rate was employed. The different peak area ratios were studied for different combinations of gamma and neutron doses, and the method was found satisfactory for mixed field neutron-gamma dosimetry.

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Most other investigations that followed, including the latest publications [7,8] employed a slow heating rate of about 5°C/s as well. The slow heating rate is the most accurate because the TLD crystal temperature follows the controllable heating element temperature without significant lag. Besides, the shape of the glow curves is more distinct, enabling visual observation of different glow curve features.

In practical routine dosimetry a high number of dosimeters must be evaluated at an acceptable rate. Besides, the TL crystals are mounted in cards being covered mostly by a thin Teflon layer which may be damaged by temperatures over 300°C. Therefore, a high heating rate (mostly 25°C/s) up to a temperature of 300°C is normally employed. In an extensive work on the subject [9], a heating rate of 25°C/s to 300°C with a hold time of 6.7 s at 300°C was applied. An automated Harshaw 8800 hot gas reader was used and the Region of Interest (ROI) method was employed for glow curve analysis. Based on successful tests with different combinations of gamma and thermal neutron doses, a four-element neutron-photon-beta TL dosimeter was developed [10]. In a later article [11], additional tests are presented, including successful comparison with an algorithm which did not utilize the high temperature peaks. This mixed field radiation dosimeter, was also registered as US Patent nr. 5,340,985 in 1994. In two recent publications [12,13] we also investigated the possibility to use the HTTL for mixed thermal neutron-gamma dosimetry when using a high heating rate. It was found that for low dose levels, characteristic to normal radiation protection situations, and within a defined neutron to gamma doses ratio range, the thermal neutron and gamma ray doses may be evaluated with acceptable accuracy. However, although being in accordance with other works published on the subject, our works attracted significant criticism [14], questioning the validity of the experimental methods employed and the application of HTTL to dosimetry in general. The subject became controversial [14-17], and it is the purpose of this presentation to enlighten the main issues involved.

2. Possible problems in routine application

2.1 General characteristics

Using a method in practical routine dosimetry requires that it fulfils several basic requirements. The most important among them are reproducibility and linearity, or at least known dose dependence. These issues were addressed in various past publications, but it seems that there are still disagreements about their characteristics and significance to the HTTL issue.

In several publications, Horowitz et al. questions the universality of the dosimetric characteristics of the HTTL [14,16,18]. They claim that the relative TL production efficiency is not uniquely dependent on LET, and is batch-dependent. They attribute this behavior to the variation in the concentrations of various impurities at the PPM level. Moreover, they claim that it appears not to be correlated with the addition of Mg and Ti dopants to LiF, but to uncontrollable impurities of unknown origin, presenting several examples of abnormal glow curves, in which the HTTL region seems to be of unexpected form and intensity [16].

The question of linearity is addressed in almost every publication on HTTL. It is accepted that the dosimetric peaks 4 and 5 are linear with dose. However, there are disagreements concerning the dose response of the HT peaks. At doses above about 250 mGy there seems to be a general consent that the TL response of the HT peaks to gamma radiation becomes supra-linear, but there are different attitudes towards the dose response in the low dose region, which is the useful range for routine radiation protection applications.

2.2 Experimental procedures

Due to their technical superiority, mostly gas heating TLD readers are used nowadays. Despite the non-linear temperature profile in the TLD crystals, these systems are extensively used in routine dosimetry laboratories for readout of TLD cards. The crystals in the widespread TLD cards, as used also by us, are coated by a thin layer of Teflon, which may be damaged at temperatures above 300°C.
When high heating rates are used, the peak maxima ($T_{\text{max}}$) shifts to higher temperatures. It is evaluated that for a heating rate of 25°C/s, $T_{\text{max}}$ of peak 5 appears at ~250°C, whereas $T_{\text{max}}$ of peak 7 appears at ~300°C. A possible problem for HT peaks evaluation lies in the limitation of the temperature applicable for TLD cards readout to 300°C (as $T_{\text{max}}$ of peak 7 for high heating rates is expected to be also near 300°C). This temperature limitation may lead to an incomplete readout of the high temperature region, as claimed by Horowitz et al. [14].

The shape of the glow curve is influenced by the heating profile, among other factors. Horowitz et al. [14,16] defined the shape of the glow curve obtained in our works (using a 25°C/s heating rate) as "anomalous", as there is no peak indicating the maximum TL intensity of the HTTL. Because the shape could hint to a monotonic decay in the HT region, it was presented by them as a definite proof that the HT region consists mainly of isothermal decay associated with a probably incomplete decay of peak 7, pointing (in their view) to a faulty experimental method by using a high heating rate.

All issues mentioned above will be addressed in the next sections.

3. The variability in the HTTL characteristics

Batch variability is a common encountered problem when using TLD, in general. Small variations in the impurities concentrations during the production process may influence significantly the TLD light emission characteristics. The change of crystal characteristics due to different treatments is a possible occurrence as well. The TLD crystals must be firstly selected to fit to a defined characteristic range (e.g. sensitivity), but they must be also checked periodically. HTTL characteristics make no exception. Abnormal responding TLD crystals should be discarded, and the characteristics variation must be checked regularly. If a careful selection of standard responding cards is performed, no unexpected variability should be encountered. In our publication on using the HTTL to mixed neutron-gamma dosimetry [19] an algorithm was developed, which uses two parameters - $R_{\gamma}$ and $R_n$ (assigned in [19] as $P_1$ and $P_2$), which are the ratio between the HT peaks area (peaks 6+7) and the dosimetry peaks area (peaks 4+5) for gamma and neutron radiations. These are the key parameters, and they should remain constant for an accurate dose evaluation. Their values, when using a 25°C/s heating rate, were determined as [19]:

$$R_{\gamma} = 0.0180 \pm 0.0006 \quad R_n = 215 \pm 0.001$$

In another experiment performed with different TLD cards about 2 years earlier [20], similar ratios were obtained:

$$R_{\gamma} = 0.0195 \pm 0.0010 \quad R_n = 0.215 \pm 0.004$$

The maximal uncertainty of the parameters in both cases was ~5%.

In a separate experiment, the standard deviation of $R_n$ for a group of randomly chosen TLD cards from a certain batch was determined by irradiating them to a fixed neutron dose. The readouts were performed by high and low heating rates. An example of the statistical distribution of the ratio values for 60 TLD-600 ($^6\text{LiF:Mg,Ti}$) crystals from 30 TLD cards read by applying a 3°C/s heating rate is presented in Fig. 1. It should be mentioned, that by using a slow heating rate, the separation between the HT peaks and the dosimetry peaks improves (see Fig. 2) and a better ratio of the HT peaks area to the dosimetry peaks area can be achieved. The average ratio for the slow heating rate data presented in Fig. 1 is ~0.33 compared to 0.215 (see above) obtained by a high heating rate.

The ratios are normally distributed with a standard deviation of 6.5%. No abnormal TL response was observed. The results support our view that rigorous calibration and quality assurance procedures are expected to produce reliable results, also when dealing with high temperature peaks.
Figure 1: The statistical distribution of the ratios between the high temperature and dosimetry peaks regions in TLD-600 glow curves, when applying a 3°C/s heating rate after thermal neutron irradiation.

4. The linearity issue

The linearity characteristic is one of the most controversial issues. It is claimed that peak 7 is supra-linear for gamma rays, but almost all published results are for high doses, over the usual radiation protection levels. However, some works claim supra-linearity down to the lowest doses, but a conclusive proof cannot be obtained due to the high uncertainty encountered in low dose experiments. Different supra-linearity detection limits were reported in the literature. Busuoli et al. [1] presented graphs on the dose response of "peak II", and mention that supra-linearity can be observed for ⁶⁰Co rays "for doses as low as (10 rads)". They concluded that "this situation does not prevent interesting results in the applied field from being obtained". Nash and Johnson [3] mention that "no change in linearity of response was observed at least up to 100R (RE)". On the other hand, Uray [4] brings evidence that the supra-linear component in the high temperature peaks exists also in the very low range, but "it seems to be lost in the statistical error". Also in his opinion, the proposed method has potential to be used in mixed field dosimetry. The results of Horiuchi et al. [6] do not indicate any non linearity up to "a ⁶⁰Co equivalent gamma dose of about 8R". A supra-linearity for ¹³⁷Cs gamma rays response from ~20 mSv (reaching ~15% at 100 mSv) is mentioned by Liu and Sims [9], but the authors conclude that the high temperature peak methodology "is applicable to most protection dosimetry situations, but not suitable for accident dosimetry".

The results of our work [20], which indicate no observed supra-linearity for the dose range of up to 50 mSv, are not in conflict with the observations in other works. This practical result should not be mixed with "a definitive and universal answer to the question of existence or non-existence of a linear dose-response region" [14], which is merely an academic consideration. It seems that there is a general agreement that in the low dose range the supra-linearity of peak 7 will probably not be distinguished as its influence will be in the range of the experimental error, thus it poses no practical obstacle in applying HTTL to mixed field neutron-gamma routine dosimetry for radiation protection application.
5. Materials, equipment and method

The following sections deal with experimental details. As it was intended to study the practicality of the HTTL method to routine dosimetry, a 6600 TLD reader (Bicron/Harshaw - now Thermo Inc.) employing hot nitrogen heating was operated for our investigations. LiF:Mg,Ti Bicron/Harshaw standard cards containing four crystals of dimensions $3 \times 3 \times 0.38$ mm were used. Two of the chips (3 and 4) contain $^6$LiF, and the other two chips (1 and 2) contain $^7$LiF. Only the results of the $^6$LiF crystals were analyzed. The pre-heat conditions, heating rate and maximum temperature are determined through a PC computer, which controls the reader. The glow curve of each chip is digitized to a 200 channel spectrum and stored on the computer. The heating profiles consisted of a preheat to 50°C (less than 0.5 s) and a linear heating rate up to a pre-determined temperature (up to 300°C), then keeping at that temperature for a total pre-determined time. After each readout, the crystals were let to cool to room temperature in the nitrogen stream of the reader. Before each experiment the TLD cards were read to check their background. Normally, cards with a background lower than about 3% from the expected readout were selected. The TLD cards were irradiated to either gamma rays or thermal neutrons. Gamma irradiations were carried out using a $^{137}$Cs source. The neutron irradiations were carried out using thermalized neutrons from a 74 MBq (at 1.5.2008) $^{252}$Cf shielded source. The source was placed in a borated paraffin shielding equipped with collimator holes. The neutrons were thermalized by layers of polypropylene. The thermal neutron dose rate at the irradiation location was ~0.27 mSv/h (at 1.5.2008). The gamma contribution was negligible (about 2%). Different neutron doses were obtained by changing the irradiation time.

6. The shape of the LiF:Mg,Ti glow curves obtained via hot gas heating

The shape of a glow curve is influenced by a combination of different factors, which determine the heat transfer conditions. These include the heating method, the presence of Teflon coating, the thickness of the chip and the heating rate. Fig. 2 presents characteristic glow curves obtained from $3\times3\times0.38$ mm $^6$LiF:Mg,Ti chips with different heating rates, after irradiation by thermal neutrons. The obvious change of the separation between the dosimetry peaks and the high temperature peaks region can be clearly observed. Increasing the heating rate causes a process of merging of the two regions, and for the $25^\circ C/s$ rate no separation can be distinguished anymore.

Figure 2: Typical glow curves of 0.38mm TLD-600 crystals in cards irradiated by thermal neutrons and read with different heating rates in a 6600 Harshaw TLD reader.
It seems that the loss of separation is a gradual process, and can be no proof that HT peaks have vanished or that another mechanism took place. This claim is supported also by the results of Burgkhardt and Piesch [21], who present two glow curves: one obtained with a heating rate of 10^3°C/s and another with a hot gas instant heating to 400°C (made possible by using bare crystals). The good separation between the low and high temperature peaks obtained with the low heating rate was lost for the fast heating. This occurred although the heating was to 400°C (which leads to a certain readout of peak 7) proving that not an incomplete decay is the reason for a featureless glow curve shape when using high heating rates. Thus, the shape of the HT region in the glow curve may be characterized as "featureless" but not "abnormal", as it is expected from the merging process.

7. HTTL high heating rate readout under the 300°C limitation

7.1 Readout with a heating rate of 30^3°C/s

Due to the shifting of the HT peaks towards 300°C for high heating rates, the question of complete readout under the 300°C limitation becomes critical. Due to the complex heat transfer process from the heating gas to the TL material, it is difficult to evaluate the true time dependant temperature of the crystals, which is non-linear. Stadtmann et al. [22] published calculations of the temperature profiles in TLD chips for conditions very close to ours (Harshaw/Bicron TLD cards containing 0.38mm LiF:Mg,Ti chips, heated by gas to 300°C). Their heating rate was 30^3°C/s, thus some readouts were performed at this rate (and not the usual 25°C/s) and for the same cycle time (20 s) with TLD cards irritated by thermal neutrons. Fig. 3 presents a typical glow curve obtained on the same time scale with the calculated temperature profile [22]. Based on it, an approximate correlation between the time/crystal temperature and the corresponding region in the glow curve can be made.

From Fig. 3 it can be seen that T_max of peak 5 appears at about ~8.5 s. At this moment, the calculated crystal temperature is ~250°C (see right scale). It is difficult to situate peak 7 due to the uniform shape of the HT region, but it seems to appear between 12s and 15s, corresponding to about 280°C-300°C crystal temperature, still during the temperature rise in the crystal. The area of the glow curve region after 15 s (when isothermal emission begins) is relatively small, and is expected to be even smaller for 25°C/s due to less shift of T_m, thus the speculation of Horowitz et al. [14,16] that when using 25°C/s heating rate most of the contribution in the HT region is due to isothermal decay, leading to an incomplete readout of peak 7, seems unfounded.

Figure 3: The glow curve of a thermal neutron irritated TLD-600 card, read with a heating rate of 30°C/s to 300°C. The crystal calculated time dependant temperature [22] was added.
7.2 Residual dose considerations

The results of additional experiments are described below. TLD cards containing TLD-600 crystals were irradiated by thermal neutrons, as in the previous experiments. Some cards were read with a fast heating rate profile (a heating rate of 25°C/s up to 300°C for 20 s), while the others were read by applying a slow heating rate of 3°C/sec up to 300°C for a total readout time of 93 seconds (a hold time of 10 s at 300°C in both cases). Afterwards, the cards were read again to measure the residual glow curves, by using slow or fast heating rates. Similar residual glow curves were obtained in all cases, like the example given in Fig. 4. The peak observed in the lower part of the spectrum is due to the Teflon coating, as it is obtained also with no TLD crystal present. For all crystals and for both evaluation processes, the residual doses in the region of the HT peaks were of the same order of magnitude, in the range 0.8%-2% from the glow curve areas of the first reading. No major additional contribution to the residual curve was observed, after applying a high heating rate. These results support the evidence that no significant difference in the emptying of the HT traps is expected when applying the high or low heating rate profiles.

Figure 4: The glow curve of the residual readout of a TLD-600 crystal irradiated by thermal neutrons and read by a 25°C/s heating rate for a total time of 30 s.

7.3 The role of the isothermal decay

The HTTL response of LiF:Mg,Ti when using a fast gas heating to 300°C is a mixture of the HTTL signal, supplemented by some isothermal decay (depending on the time-temperature profile). The linearity of the HT peaks region and of the isothermal decay component was checked by analyzing glow curves of 6LiF crystals (TLD-600) irradiated by different doses of thermal neutrons, by a two steps temperature profile method [1,2]. For the readout conditions defined for Fig. 3, it can be seen from the crystal temperature profile shown in the figure that the isothermal decay begins after ~15 s. The region between 10 s and 13 s corresponds surely to the rising temperature time, and contains mainly HT peaks information, while after 20 s only isothermal decay occurs. In Fig. 5 it can be seen that both regions are linear as a function of dose / irradiation time in the investigated range. The ratio between the 10 s to 13 s region and 20 s to 30 s region remains constant with an average value of 3.92 ± 0.71. The actual proportions between the two contributions, although being a subject of dispute [14-17], are actually of no practical interest, as both effects add to the light emitting efficiency linearly. The isothermal decay enhances the light output, increasing the total efficiency, thus it is not to be regarded as a disturbing effect, but as a complimentary one. It is interesting to mention, that there were even proposals to use the isothermal decay as a sole readout method [23]. The authors proposed this method for LiF:Mg,Cu,P crystals which may be damaged irreversibly by a too high temperature.
Figure 5: The glow curve region integrals of a TLD-600 crystal irradiated by different doses of thermal neutrons and read by a 30°C/s heating rate.

8. Conclusions

The works published on the dosimetric applications of HTTL to mixed field dosimetry did not report any erratic or abnormal response; they present results within acceptable accuracy ranges. The experimental procedures, also when using fast rate gas heating, produce reliable and expected results, similar to those obtained by applying a slow rate heating. The batch differences and scattered abnormal glow curves are a known reality to any TLD dosimetrist, and apply to the use of TLD in general. Adequate quality assurance programs must be implemented when employing TL dosimetry for any application. For low dose values, normally encountered in radiation protection situations, the non-linearity poses no real problem, as it is not apparent on the background of the normal experimental uncertainties.

Therefore, the HTTL has potential to be employed in mixed neutron-gamma dosimetry. Further research is still needed, especially in improving experimental procedures, statistical characterizations and data analysis methods.
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