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Comparison of Radiation Response of LiF:MgCuP and Al₂O₃:C Thermoluminescent Dosimeters

L.S. Erhardt, L. Prud'homme-Lalonde, T. Cousins,
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Abstract

A study was conducted comparing the properties of two types of thermoluminescent dosimeters (TLDs): LiF:MgCuP (TLD-100H) and Al₂O₃:C (TLD-500). This was done in order to determine which is better suited for the dosimetry requirements of the Radiation Effects Group at DREO (RE/DREO). The dose and energy response of the TLDs were measured by exposing them to a wide range of photon doses and energies. In addition, the sensitivity of the TLDs to fast and thermal neutrons was determined with a PuBe source. Fading and background detection were also studied. The TLD-100H dosimeter was found to be slightly more tissue-equivalent in its energy response but the TLD-500 spanned a larger dose range. In addition, TLD-500 had a desirable insensitivity to thermal neutrons. The effect of fading and response to background were similar in the two types of TLD. The TLD-500 dosimeter was found to be better suited for work in the environments studied by RE/DREO.

Résumé

Une étude comparant les propriétés de deux types de dosimètres thermoluminescents (DTL), LiF:MgCuP (DTL-100H) et Al₂O₃:C (DTL-500), a été effectuée. Cette étude a été effectuée afin de déterminer quel type de dosimètre est le plus approprié pour la dosimétrie requise par le Groupe des effets du rayonnement au CRDO. Les dosimètres ont été exposés à une vaste gamme de doses et d'énergies de rayons gamma afin d'en évaluer leurs réponses. De plus, la réponse des dosimètres aux neutrons rapides et thermiques a été évaluée à l'aide d'une source de PuBe. L'évanouissement des signaux ainsi que la détection du rayonnement de fond ont aussi été étudiés. Le dosimètre DTL-100H s'est avéré légèrement plus équivalent au tissu dans sa réponse à diverses énergies, mais le DTL-500 couvrait une plus vaste gamme de doses. De plus, le DTL-500 s'est avéré supérieur dans sa réponse à diverses énergies et doses, et il démontrait une insensibilité souhaitable aux neutrons thermiques. L'effet de l'évanouissement des signaux et la réponse au rayonnement de fond étaient semblables pour les deux types de dosimètres. Le DTL-500 s'est avéré supérieur pour l'utilisation requise dans les laboratoires du Groupe des effets du rayonnement au CRDO.

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Executive summary

Introduction: The Radiation Effects Group at DREO conducts experiments that require accurate dosimetry over a wide gamut of radiation doses and photon energies. Photon dosimetry at DREO is generally conducted using thermoluminescent dosimeters (TLDs), with $\text{Al}_2\text{O}_3:\text{C}$ (TLD-500) being the most commonly used type. There are two types of TLD that are in general (commercial, regulatory and lab) use for low radiation doses, with the TLD-500 being one and the other being TLD-100H ($\text{LiF}:\text{MgCuP}$). TLD-100H is widely used for personnel dosimetry (primarily as a result of its neutron sensitivity) and is more commonly available because of this. This study is intended as a comprehensive comparison between the two different types of TLD to determine which is better suited to the needs of DREO.

Results: The TLD-500 ($\text{Al}_2\text{O}_3:\text{C}$) outperformed the TLD-100H ($\text{LiF}:\text{MgCuP}$) in most of the areas that were studied. The TLD-500 had a more consistent response over a wider range of doses and energies, and had a more desirable (negligible) response to neutron radiation. The dose response of the TLD-500 was flat over the range 1 mrem to 100 rem, compared to 5 mrem to 10 rem for the TLD-100H. The energy response for the TLD-100H was flat from 150 keV to 1250 keV, but fell off at lower energies. The TLD-500 under-responded in the energy range between 150 and 200 keV, but was otherwise similar in performance to the TLD-100H. The TLD-500 is insensitive to thermal neutrons, a possible source of background in laboratory, high-altitude, mine detector or reactor measurements, while the TLD-100H has a strong response to these neutrons. Other properties of the TLDs, such as fading and background detection, were found to be similar. Handling of the TLD-500 is more difficult due to sensitivity to UV light, but this is mitigated with the use of careful lab practices.

Significance and Future Plans: Although the TLD-100H may be better suited to personnel dosimetry, the TLD-500 is better suited to a DREO's needs. DREO should continue to use the TLD-500 as its main dosimeter. The TLD-100H, while also a useful dosimeter, should not replace the TLD-500 for the bulk of the dosimetry performed at DREO. During this study it was noted that the performance of DREO's monoenergetic x-ray production facilities is in need of upgrading.

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Sommaire

Introduction: Le Groupe des effets du rayonnement au CRDO effectue des expériences qui demandent une dosimétrie pour une vaste gamme de doses de radiation à diverses énergies. Au CRDO, la dosimétrie des photons est généralement effectuée en utilisant des dosimètres thermoluminescents (DTL), le DTL-500 ($\text{Al}_2\text{O}_3:\text{C}$) étant celui qui est le plus couramment utilisé. Il y a deux types de dosimètres qui sont généralement utilisés pour mesurer de faibles doses de radiation, le DTL-500 ainsi que le DTL-100H ($\text{LiF}:\text{MgCuP}$). Le DTL-100H est surtout utilisé pour la dosimétrie individuelle (principalement à cause de sa sensibilité aux neutrons), ce qui en fait un DTL plus communément disponible. Une étude de comparaison entre les deux types de DTL a été effectuée afin de déterminer lequel est le plus approprié pour l'utilisation requise au CRDO.

Résultats: Le DTL-500 ($\text{Al}_2\text{O}_3:\text{C}$) a surpassé le DTL-100H ($\text{LiF}:\text{MgCuP}$) dans la plupart des aspects étudiés. Le DTL-500 était plus consistant dans sa réponse à une vaste gamme de doses et d'énergies, et il avait une réponse plus désirable (négligeable) aux neutrons. La réponse du DTL-500 était uniforme sur une portée de 1 mrem à 100 rem, comparativement à une portée de 5 mrem à 10 rem pour le DTL-100H. La réponse énergétique du DTL-500 était uniforme sur une portée de 150 keV à 1250 keV mais chutait aux basses énergies. Le DTL-500 démontrait une réponse inférieure de 150 à 200 keV, mais autrement sa performance était similaire au DTL-100H. Le DTL-500 est insensible aux neutrons thermiques, une source possible de rayonnement de fond dans les mesures effectuées en laboratoire, à haute altitude, dans les détecteurs de mine ou dans les réacteurs, tandis que le DTL-100H répond fortement à ces neutrons. D'autres propriétés, telles l'évanouissement des signaux et la détection du rayonnement de fond, sont semblables pour les deux types de dosimètres. La manipulation du DTL-500 s'avère plus difficile à cause de sa sensibilité à la lumière UV, mais ce point faible peut être contourné avec de bonnes pratiques de laboratoire.

Importance: Même si le DTL-100H convient mieux à la dosimétrie individuelle, le DTL-500 donne une réponse qui convient mieux aux besoins du CRDO. Le CRDO devrait continuer à utiliser le DTL-500 comme son principal dosimètre. Même si le DTL-100H est un dosimètre utile, il ne devrait pas remplacer le DTL-500 pour la plus grande partie de la dosimétrie effectuée au CRDO. Cette étude a de plus démontré qu'il faudrait apporter des améliorations aux installations de rayon X monoénergétiques.

Erhardt, L.S.; Prud'homme-Lalonde, L.; Cousins T.; Estan, D. and B. Hoffarth. 2001. Comparaison de la Réponse de Rayonnement pour les Dosimètres Thermoluminescents $\text{LiF}:\text{MgCuP}$ et $\text{Al}_2\text{O}_3:\text{C}$. DREO TM 2001-074. Centre de recherches pour la défense
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1. Introduction

The Radiation Effects Group at Defence Research Establishment Ottawa (DREO) conducts experiments to study the effects of radiation on materials, devices and personnel relevant to the Canadian Forces. The experiments performed at DREO are quite widely varied; ranging from irradiations of biological samples to advanced electronics for use in space. Thermoluminescent dosimeters (TLDs) are used extensively for photon dosimetry in these experiments. However, by far the most important use of TLDs is as personnel dosimeters for DREO and CF staff in experiments and field operations. The given radiation dose for DREO experiments varies widely, from background levels up to hundreds of kilorem. It is essential that accurate dosimetry be available to measure the dose given in these experiments.

There are two types of thermoluminescent dosimeters (TLDs) that are in general use for low-level (few mrem) radiation measurements [1]. These are LiF:MgCuP and $\text{Al}_2\text{O}_3:\text{C}$ ¹, otherwise known as TLD-100H and TLD-500 respectively. DREO has used both types of TLDs in the past, and based on this experience prefers to use the TLD-500 [2]. The preference for the TLD-500 is due mainly to its superior response to doses in the sub-mrem range. Supporters of the TLD-100H cite as its benefits: increased sensitivity to neutrons and decreased sensitivity to optical radiation. These properties of the TLD-100H may make them desirable as stand-alone personnel dosimeter badges, where neutron exposure must be monitored and ease of handling is desirable, but a different approach is necessary for experimental work at DREO. DREO separates the photon and neutron dose contributions by employing a combination of TLD-500s and bubble detectors [3].

This report is a comprehensive comparison of the two types of TLD in a range of tests that mimic DREO work. The tests included exposure dependence measurements between 10^{-4} and 10^3 rem, energy dependence measurements between 20 and 1250 keV, and response to both thermal and fast neutrons. This report is not intended as a discussion of the theory behind the operation of thermoluminescent dosimeters, but rather as a direct comparison of the performance of two similar types of TLD under conditions typical of operation at DREO. As a result, there will be no general discussion of theory; only that necessary for the interpretation of the results will be discussed. For a basic background on TLD theory one should refer to McKeever, Moscovitch and Townsend [1].

¹ The notation for the TLD composition refers to the main material and the dopants used to produce the TLD. This corresponds to lithium fluoride doped with magnesium, copper and phosphorous for the TLD-100H and aluminum oxide doped with carbon for the TLD-500.

2. Test Procedures

The TLDs were tested to determine their response to a variety of different types, energies and doses/fluences of radiation. The TLDs were all handled according to the instructions supplied by the manufacturer – in some cases augmented with DREO acumen based upon years of field experience. This section details the general procedures that were followed in handling the TLDs and performing the measurements and provides an overview of the tests that were performed.

2.1 General Procedures

Before performing a measurement the TLDs to be used were calibrated by giving them a known dose of radiation and subsequently measuring their glow curve with the appropriate TLD reader. Before each measurement (including the calibration) the TLDs were prepared by annealing them to remove any stored energy (or residual dose from previous exposure) and then wrapping them in 0.018" of tin to flatten their energy response. The tin also helped serve to establish electronic equilibrium within the TLDs. TLDs were always handled using tweezers to avoid deposition of oil from one's hands, which could cause spurious thermoluminescent signals. One other consideration in handling the TLDs is that there is a difference in light sensitivity between the TLD-100H and TLD-500. TLD-100H could be handled in normal room light but the TLD-500 is light sensitive and had to be handled in a darkened room. Indirect light from a desk lamp was used to aid visibility when handling the TLD-500s. A summary of the nominal physical properties of the two types of TLD is given in Table 1.

Table 1. Summary of the physical properties of the TLDs used in this study. Nominal dose ranges are as specified by the manufacturer.

	TLD-100H	TLD-500
Material	LiF:MgCuP	AL ₂ O ₃ :C
Shape	Disc	Disc
Diameter	4.5 mm	5 mm
Thickness	0.60 mm	1 mm
Colour	White opaque	Translucent
Nominal Range (rem)	0.1 mrem – 1000 rem	5 µrem – 100 rem
Light Sensitivity	Not sensitive	Extremely sensitive

2.1.1 TLD Preparation

Five of each type of TLD were used for each test. The TLD-500s were annealed in an oven at 400°C for ten minutes. The TLD-100Hs were

annealed two to three times in the Harshaw Bicron 3500 TLD reader at 240°C. The original plan was to anneal these in an oven as well, but this yielded difficulties which will be detailed in section 3.6. The tin used was in the form of a foil with a thickness of 0.0015". The TLDs were wrapped with 12 turns of the foil for a total thickness of 0.018". They were then numbered and placed in small, clear, plastic bags and then taped to an acrylic stand for irradiation.

2.1.2 Calibration

The TLDs were individually calibrated (as per DREO practice) with the 10 Ci UDM-1 ⁶⁰Co source (see Figure 1). The TLDs were given a dose of 10 mrem at a rate of 100 mrem/h for 6 minutes. The calibrations were repeated three times, after each of which the charge was read using the procedure outlined in section 2.1.3. The readings were averaged to obtain the calibration factor for each TLD (see Appendix A). Individual calibration mitigates the increased errors associated with batch calibration and associated statistical response variation.

2.1.3 Measurements

All of the measurements for this study were performed with the Harshaw Bicron 3500 manual TLD reader. Each measurement involves three phases: preheating, acquisition and annealing. Measurements are done on individual TLDs in a dry nitrogen atmosphere within the TLD reader. The temperature and data acquisition are controlled automatically. The time-temperature profiles for the TLDs are summarized in Table 2.

Table 2. Time-temperature profiles for TLD-100H and TLD-500.

		TLD-100H	TLD-500
Preheat	Temperature	135 °C	55 °C
	Time	10 s	10 s
Acquire	Rate of Change	10 °C/s	10 °C/s
	Max. Temperature	240 °C	265 °C
	Time	23 s	40 s
Anneal	Temperature	240 °C	400 °C
	Time	10 s	10 s

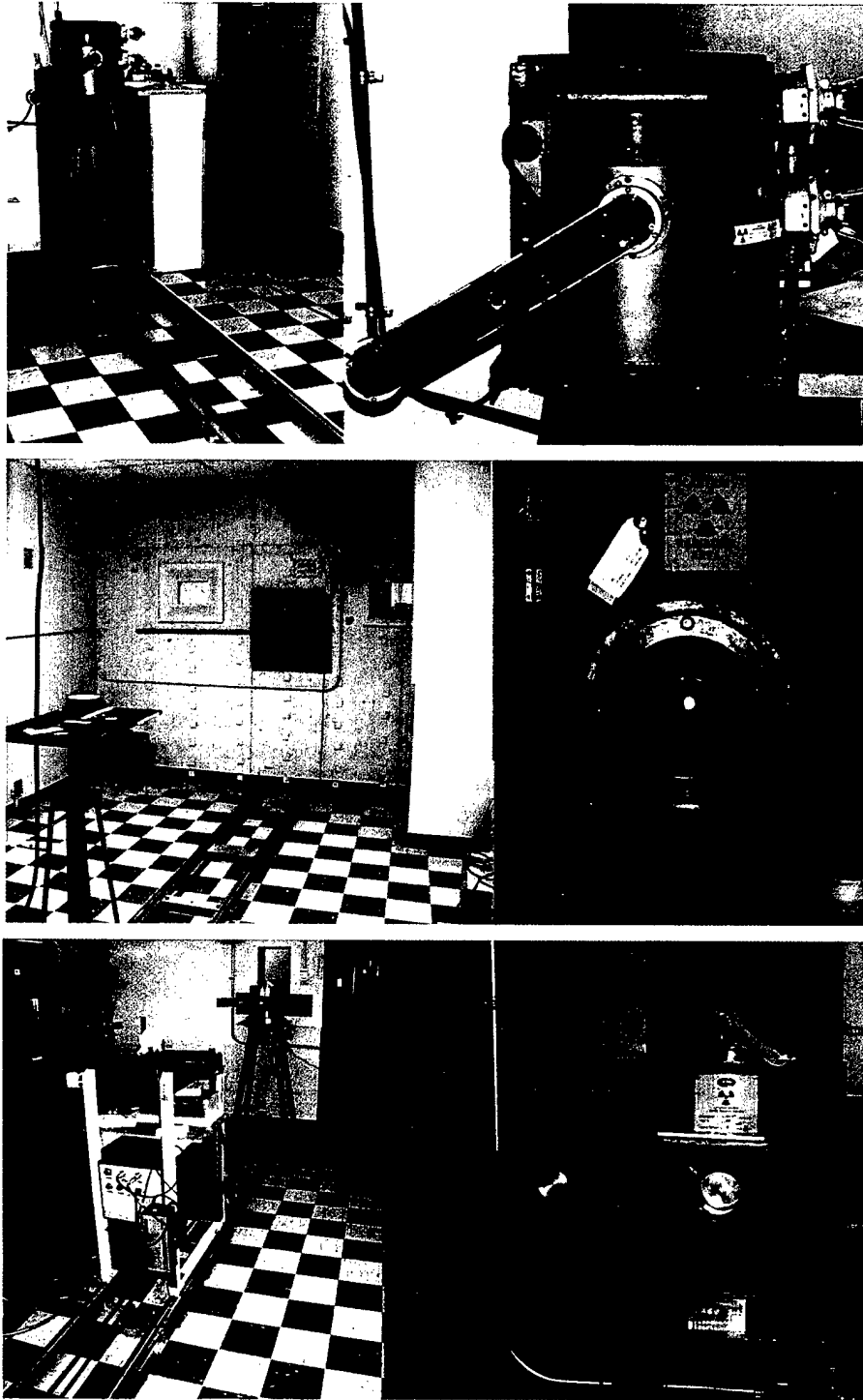


Figure 1. The DREO gamma-ray sources used for performing TLD irradiations. (Top) The UDM-1 source is a calibrated 10 Ci ^{60}Co source. (Middle) The GRM-750 is a calibrated 500 Ci ^{60}Co source. (Bottom) The AN/UDM-1A is a calibrated 120 Ci ^{137}Cs source. All photographs show a close-up of the source on the right side and a wide view of the source room on the left.

2.2 Individual Tests

The TLDs were subjected to a variety of different tests in order to determine their efficacy for DREO work. These tests were designed to measure the TLD response to a variety of photon energies and doses, as well as to test sensitivity to neutrons. Testing was also done on fading and ambient radiation background measurement capabilities.

2.2.1 Dose Dependence

After calibration, the TLDs were exposed to a series of doses from ^{60}Co gamma rays in order to determine their response. Measurements for this section were done in two groups: low doses (10 mrem and below) and high doses (10 mrem and above). Each group used a different set of TLDs consisting of five of each of the TLD-100H and the TLD-500. Exposures were done at various dose rates in order to achieve the given dose in a convenient period of time.

The first set of experiments tested the TLD response at low doses. These were done in a series of exposures starting at 10 mrem with subsequent exposures of 5, 2, 1, 0.5 and 0.1 mrem in that order. These irradiations were all performed with the UDM-1, which is a calibrated 10 Ci ^{60}Co source (see Figure 1). Following each irradiation, the TLDs were read according to the procedure outlined in section 2.1.3.

TLD response at high doses was measured in the second set of experiments with doses starting at 10 mrem followed by 0.1, 1, 10, 100 and 1000 rem. These irradiations were mainly performed with the GRM-750, which is a calibrated 500 Ci ^{60}Co source (see Figure 1). The gamma-ray sources are all calibrated in units of R, with conversion to rem done according to ICRU Report 47 [4]. The lower dose irradiations (10 and 100 mrem) were done with the UDM-1. TLD response was also measured according to the procedure outlined in section 2.1.3. After this set of exposures, the TLDs were again exposed to 10 mrem to determine their ability to recover from large exposures.

2.2.2 Energy Dependence

TLD response was measured for a variety of energies of x-rays and γ -rays. X-rays with energies ranging from 20 keV to 200 keV were supplied by two x-ray generators: the Muller MG300 and the Lorad LPX200 (see Figure 2). X-rays of 20, 40 and 60 keV were supplied by the LPX200 and the MG300 supplied energies of 80, 100, 150 and 200 keV. Nearly monoenergetic x-ray spectra were obtained through the use of filters made of various materials, according to standard DREO practice [7, 5]. The filters used to produce these energies are summarized in Table 3. Gamma rays of energy 662 keV were supplied by the AN/UDM-1A, which is a calibrated (traceable to NRC) 120

Ci ^{137}Cs source (see Figure 1). The UDM-1 was used to deliver 1250 keV (average) ^{60}Co gammas. The dose delivered during the x-ray experiments was measured directly using a Microspec-2 spectrometer (see Figure 2). The Microspec E-probe was used for x-ray energies of 150 and 200 keV and the X-probe was used for all of the lower energies [6].

Table 3. Summary of the filters used to produce the nearly monoenergetic x-ray spectra used in this study. The filters that have letter labels are from a standard set used at DREO [5]. All of the labelled filters have a 0.020" thick aluminum layer as well as the listed materials.

Peak Energy (keV)	Machine Energy (kV)	Filter	Material	
			Lead	Copper
21.37	25	--	--	0.0625"
40.45	46	--	--	0.0625"
61.63	68	--	--	0.125"
80.58	100	A	--	0.20"
100.54	120	C	--	0.50"
150.41	180	G	0.0625"	0.50"
198.57	240	H	0.0937"	0.50"

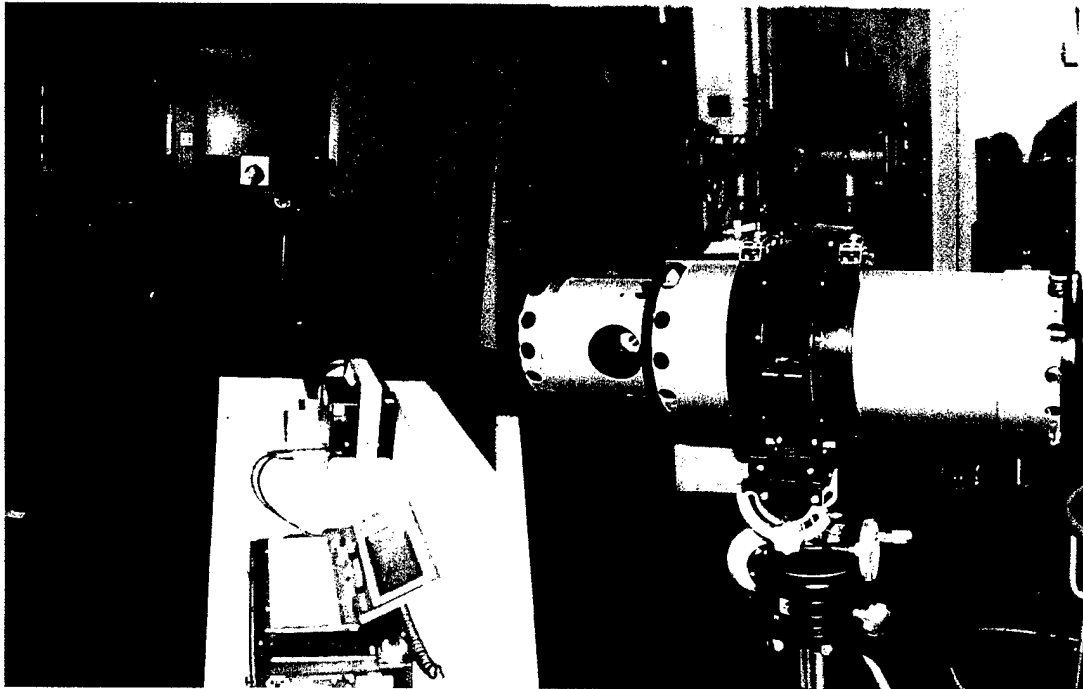


Figure 2. The DREO x-ray sources used for performing TLD irradiations. (Left) The x-ray source room. The sources are in the background, in front of the door. The Microspec-2 and probe are in the foreground on the table. (Right) The x-ray sources. The Lorad LPX200 is the foreground, and the Muller MG300 is in the background, marked with the letter 'A'.

The gamma sources are all calibrated in units of Roentgens, while the Microspec determines the dose in rem. The dose from the gamma sources is converted to rem according to ICRU Report 47 [4]. A complicating factor with the x-ray irradiations is the fact that the x-ray beams are not truly monoenergetic. In general, the x-ray spectrum contains a broad peak at the desired energy as well as a flat background over part of the energy range and possibly extraneous peaks at lower energies. Individual x-ray spectra are tailored through the use of filters of various materials to remove as much off-peak dose contribution as possible. Examples of x-ray spectra are shown in Figure 5. Corrections were applied to the data to remove the off-peak contributions to the dose.

2.2.3 Fast and Thermal Neutron Responses

Tests were performed to determine the TLD response to both thermal and fast neutrons. A set of 10 TLDs, 5 of each type, was exposed to a mixed field consisting of fast and thermal neutrons as well as gamma rays, supplied by a PuBe source. Two separate measurements were done, one with the PuBe source moderated by a paraffin sphere and the other with it bare. The paraffin moderator has the effect of thermalizing a fraction of the fast neutrons, changing the fast-to-thermal ratio at the exposure point.

2.2.4 Fading

Twenty TLDs of each type were used for this measurement; all were prepared according to the procedure outlined in section 2.1.1. Ten TLDs of each group were irradiated to 10 mrem in the UDM-1 ^{60}Co source, while the remaining ten were left unirradiated. Following the irradiation one irradiated and one unirradiated TLD of each type were read out daily, for a total of ten days. The dose in the unirradiated TLD was used as a measure of the background radiation and subtracted from that of the irradiated TLD. The results were examined for signs that the stored energy from the radiation exposure decreased over time.

2.2.5 Background Measurement

Twenty TLDs of each type were also used for this measurement. All of the TLDs were annealed, read out immediately and then stored for 21 days away from any radioactive sources. After 21 days they were again read out to determine the background dose that they recorded. This recorded dose was compared to the actual DREO background dose.

3. Results

The results of the measurements outlined in section 2.2 are presented here along with a discussion of some other issues that arose during the experiments. In Appendix A there is a list of calibration factors for the individual TLDs that were used in this study.

3.1 Dose Dependence

The results for both the high and low dose measurements are presented in Figure 3. The results are shown as a ratio of TLD-measured dose to the actual delivered dose as a function of the actual dose. The uncertainty in the measurements is the standard deviation of the measurements from five TLDs of each type. The important factor here is not the uncertainty in the data points, but rather the relative response of the two different types of TLD. Both types of TLD were irradiated at the same time to a given dose level, so if the value of the delivered dose is skewed, it is skewed equally for both types.

From the graph in Figure 3 it can be clearly seen that the TLD-500 has a flat response over a wider range of doses than does the TLD-100H. The response of the TLD-500 is essentially flat (to within 5 %) over the entire range from 1 mrem up to 100 rem, while the TLD-100H is flat from 5 mrem to 10 rem. Deviations from a flat response are greater for the TLD-100H at low doses and increasingly more significant with lowering dose. Although the TLD-500 has a flat response to a higher dose than the TLD-100H, when it does deviate from flat, at 1000 rem, it does so to a significantly greater extent than the TLD-100H. From this it is clear that the TLD-500 is able to accurately measure a wider range of dose levels than the TLD-100H.

After the TLDs were exposed to the 1000 rem irradiation, they were prepared, given a 10 mrem dose, and read out. Both types of TLD produced results that were larger than the 10 mrem of applied dose, but by different amounts. The TLD-100H produced a result of 8.7 rem, 870 times larger than the given dose. The TLD-500 reported a dose of 17.7 mrem or 1.77 times larger than the given dose. The glow curves of both TLDs were abnormal, suggesting radiation damage. The TLDs that received these large doses were not used for any subsequent measurements. Clearly for personnel dosimetry purposes, this is not a big concern. However, for electronics irradiations this should be noted and separate records should be kept.

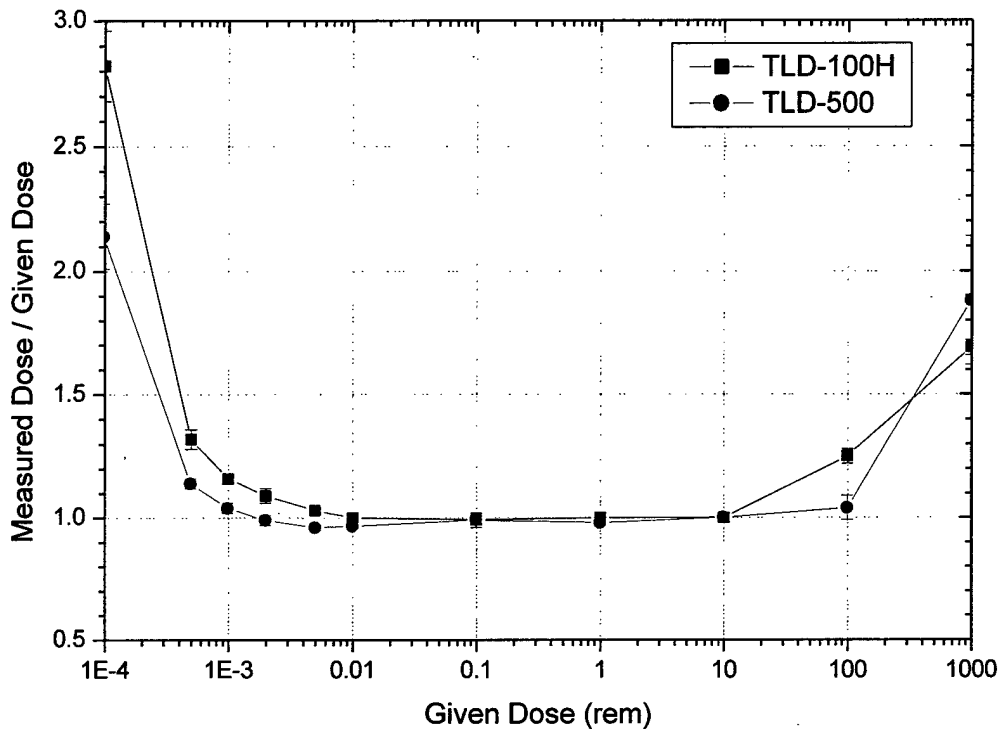


Figure 3. The Ratio of dose as measured by the TLD to that given to the TLD as a function of given dose. Doses were given to the TLDs in two segments: starting at 0.01 rem and going higher and lower respectively. The response of the TLD-500 is flat in the range from 0.001 rem up to 100 rem, while the TLD-100H has a flat response from 0.005 up to 10 rem. The error bars are the standard deviation of measurements with five TLDs.

3.2 Energy Dependence

The measurements for this section were carried out following the procedure outlined in section 2.2.2, with the results summarized in Figure 4. The uncertainty on the two highest energy data points is given by the standard deviation of the TLD readings. The lower energy points were all done using x-ray generators, the actual delivered dose was measured with a Microspec-2 spectrometer. This combination leads to some difficulties in performing and interpreting the results of the irradiations. The spectra produced by the x-ray machines are not monoenergetic, but have broad peaks with low energy tails or secondary peaks. In order to properly measure the spectra, the dose rate must be low enough for the Microspec to handle, but this leads to long exposure times for the full irradiation. Uncertainty on the lower energy points was taken to be 12.5 % for the spectra produced with the Muller MG300 and 7.5 % for those produced with the Lorad LPX200, based on measurements of the stability of the x-ray generator over the time necessary to achieve the required total dose. A problem arose during the 200

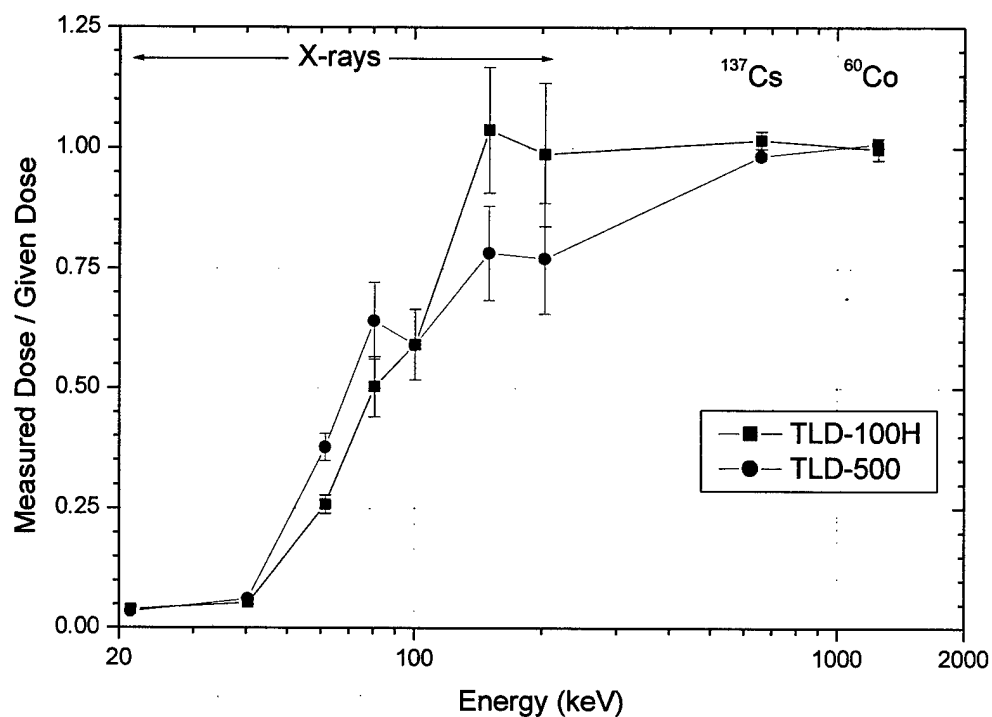


Figure 4. The Ratio of dose as measured by the TLD to that given to the TLD as a function of energy. X-ray sources were used between 20 and 200 keV; ¹³⁷Cs and ⁶⁰Co were used for 662 keV and 1250 keV respectively.

keV irradiation, which made the exposure time uncertain and cut the irradiation short. The uncertainty on that data point has been increased to 15 % to reflect this.

The off-peak portions of the spectra add a complicating factor to the data analysis. In order to correctly determine the TLD response at a given energy, this background contribution to the total dose as measured by the TLD must be ascertained. In order to do this, the TLD response as a function of energy must be used to correct the background dose contribution. The background dose contribution was weighted by a factor that convolves the shape of the background spectrum with the TLD energy response. This effect was significant in measurements where the background dose was large as in the 150 and 200 keV measurements (and to a lesser extent the 100 keV measurement). A summary of the x-ray irradiations is presented in Table 4, and the x-ray spectra are shown in Figure 5.

Table 4. Summary of the x-ray irradiations performed. The peak energy and the both the peak and off-peak doses contributions are given. All values were measured using the Microspec-2. The 200 keV irradiation was only done to a dose of 3.7 mrem peak due to problems with the Muller MG300.

Peak Energy (keV)	X-ray Generator	Microspec Probe	Given Dose (mrem)	
			Peak	Off-peak
21.37	LPX200	X-probe	10.0	0
40.45	LPX200	X-probe	10.0	0.57
61.63	LPX200	X-probe	10.0	0.04
80.58	GM300	X-probe	10.0	0.10
100.54	MG300	X-probe	10.0	0.7
150.41	MG300	E-probe	10.0	6.8
198.57	MG300	E-probe	3.7	2.2

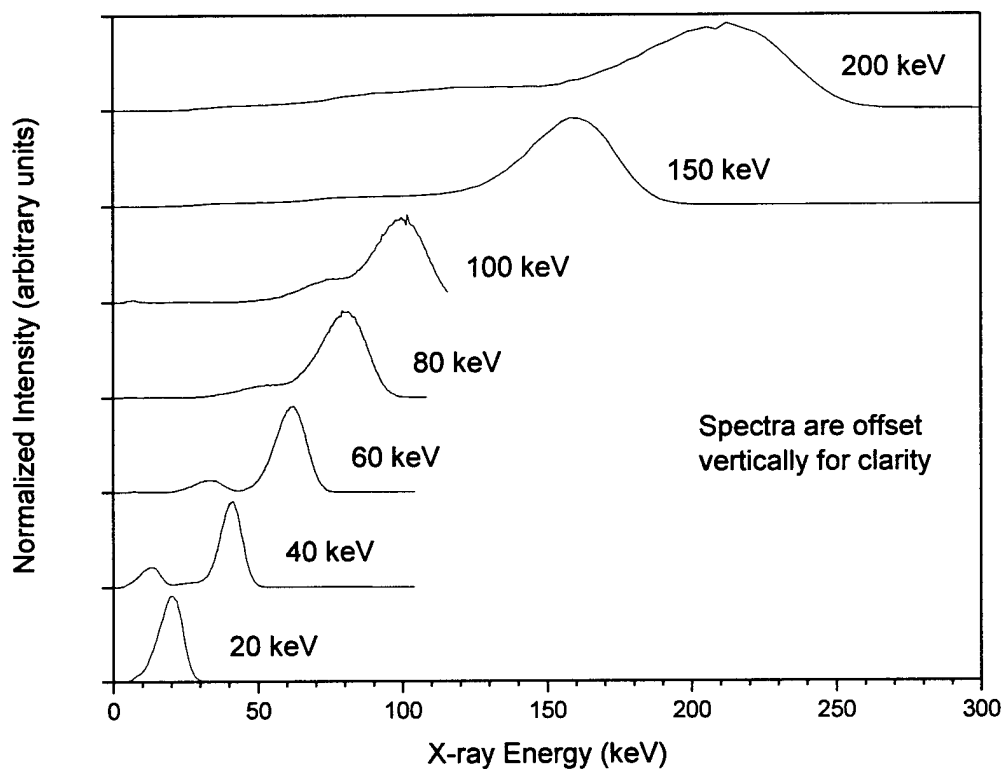


Figure 5. X-ray spectra for the irradiations performed. The spectra have been normalized and offset vertically in order to accentuate the shape of the spectra. A summary of the peak and off-peak doses is given in Table 4. The secondary peaks on the 40 and 60 keV spectra are escape peaks, artifacts of the NaI detector used in the Microspec-2. There is no correction to the dose contribution necessary for these peaks.

Figure 4 shows a plot of the ratio of measured dose to given dose as a function of energy. This ratio was calculated as follows:

$$R_E = \frac{D_M - w_E D_B}{D_P}$$

where R_E is the response ratio at energy E , D_M is the dose measured with the TLD, D_B is the background dose as measured with the Microspec-2, D_P is the peak dose measured with the Microspec-2 and w_E is the weighting factor that takes into account the shape of the background spectrum and the TLD response. The weighting factor for a spectrum with the peak at energy E , is a convolution of the TLD energy response, $R(E')$, with the background energy spectrum $B_E(E')$. This can be expressed as:

$$w_E = \int_0^{\infty} R(E') B_E(E') dE'$$

The weighting factors that were used in the data analysis were estimated based on the available data. The estimated weighting factors are summarized in Table 5.

Table 5. The weighting factors used to correct for the background contribution to the applied dose. The weighting factor depends on the TLD response at the energies where most of the background occurs. For example, the weighting factor for 150 keV is the average of the TLD response ratio at 80 and 100 keV, due to the fact that background occurs in this region. This factor is calculated for each TLD type.

Energy (keV)	Weighting Factor
20	$w_{20} = 0$
40	$w_{40} = 0$
60	$w_{60} = 0$
80	$w_{80} = R_{60}$
100	$w_{100} = (R_{60} + R_{80}) / 2$
150	$w_{150} = (R_{80} + R_{100}) / 2$
200	$w_{200} = (R_{100} + R_{150}) / 2$

The response of the TLD-100H (see Figure 4) is quite flat in the region from 150 keV to 1250 keV with the ratio of measured to given dose over this region essentially one. The response does fall off to just over 50 % at 100 keV, 25 % at 60 keV and is zero by 40 keV. This TLD is certainly usable in the range from approximately 80 keV and above, but for energies below 150 keV, it is necessary to have knowledge of the energy spectrum of the absorbed radiation in order to have confidence in the results.

The TLD-500 energy response is at 662 and 1250 keV, but drops below a flat response somewhere between 200 keV and 662 keV. The energy region from 80 keV to 200 keV is relatively flat, with the response between 60 and 75 %. The TLD-500 has no response at 40 keV and below, similar to the TLD-100H.

From these data, it can be concluded that the TLD-100H has a superior energy response to that of the TLD-500. This result is based mainly on the x-ray data at 150 keV and 200 keV. It should be noted that there is quite a lot of uncertainty in these data points due to inadequacies in the x-ray production system at DREO. This will be discussed further in section 3.6.2. It is likely that the under-response of the TLD-500 can be corrected by a reduction in the thickness of the tin used to wrap the TLD. A reduction in the thickness of tin would result in less attenuation of low-energy gamma rays but would have minimal effect on the higher-energy gamma rays.

3.3 Fast and Thermal Neutron Response

The response of TLDs to both thermal and fast neutrons was measured according to the procedure outlined in section 2.2.3. Two irradiations were performed with the relative amounts of fast and thermal neutrons changed by the presence or absence of a moderator. The parameters of the two irradiations are presented in Table 6. Each irradiation was done for a duration designed to give a gamma ray dose of 10 mrem as measured with a BGO detector. Fast neutron flux was measured with a Microspec-2 N-probe, and thermal flux was measured with a BF₃ detector.

Table 6. Summary of the neutron fluences and gamma ray doses for the two irradiations that were performed. The duration of each irradiation was set to give 10.0 mrem of gamma dose.

Source	Neutron Fluence (n/cm ²)		Gamma Dose (mrem)
	Fast	Thermal	
Bare	7.32×10^6	1.55×10^5	10.0
Moderated	4.78×10^6	7.13×10^5	10.0

For the irradiations, the response of the TLDs to the total given field was determined as usual and corrected to remove the contribution from the gamma rays. Assuming that the TLD response to the high-energy gamma rays from the PuBe source is consistent with that to ⁶⁰Co gamma rays, then the dose measured by the TLDs in excess of 10 mrem was due to neutrons. In a simplistic approach, the TLD response to the fast and thermal neutrons can be separated by performing analysis of two irradiations with different relative fluences of each type. The responses can be determined by solving the set of simultaneous equations:

$$D = A\Phi_f + B\Phi_{th}$$

$$D' = A\Phi'_f + B\Phi'_{th}$$

Table 7. Summary of the measured doses in the TLDs after exposure to the bare and moderated PuBe neutron source. The response of each TLD to fast (*A*) and thermal (*B*) neutrons is also shown. The simple approach that was used was adequate to describe the response of the TLD-100H, but the negative coefficient for the TLD-500 implies that it does not work for the TLD-500.

TLD	Source	Measured Dose (mrem)	<i>A</i> (mrem cm ² /n)	<i>B</i> (mrem cm ² /n)
TLD-100H	Bare	7.66	8.43×10^{-7}	9.61×10^{-6}
	Moderated	10.89		
TLD-500	Bare	7.14	1.04×10^{-6}	-3.15×10^{-6}
	Moderated	2.73		

where *D* is the measured response in excess of the 10 mrem due to the gamma radiation, Φ_f and Φ_{th} are the fast and thermal fluence respectively. The two equations represent the bare and moderated source, with the moderated source being denoted by the primes in this case. The coefficients *A* and *B* represent the strength of the TLD response to fast and thermal neutrons respectively. A summary of the measured neutron dose along with the derived *A* and *B* coefficients is shown in Table 7.

Both TLDs had a very similar response to the bare source irradiation where fast neutrons are the dominant type. The TLD responses to the moderated source were, however, very different. The response of the TLD-100H was greater with the moderated source than with the bare one while the TLD-500 response was greatly reduced for the moderated source. This suggests that thermal neutrons dominate the response of the TLD 100-H and fast neutrons dominate for the TLD-500.

When one solves the above set of equations for the response of the TLD-100H, the fast (*A*) and thermal (*B*) neutron coefficients can be determined; these are given in Table 7. The contribution from thermal neutrons is approximately ten times greater than that from the fast neutrons. The TLD-100H is quite sensitive to thermal neutrons and would be of great use in an environment where the monitoring of thermal neutrons is essential.

The response of the TLD-500 to thermal neutrons is quite different. When the simultaneous equations are solved for the TLD-500 the coefficient, *B*, for the thermal neutron response has a negative value. This result is physically meaningless and suggests that our simple model for the neutron response is inadequate for the interpretation of the TLD-500 results. The negative coefficient comes from the fact that when the fluence of fast neutrons is reduced by approximately 1/3, the total dose as measured with the TLD-500 is reduced by approximately 2/3. In order for this to occur in our simple model the increased fluence of thermal neutrons would have to remove dose from the TLD-500; hence the negative coefficient.

These results for the TLD-500 are, however, consistent with a neutron response that is significant for higher energy neutrons, but falls off rapidly at lower energies. The effect of the moderator is not just to decrease the fast neutron flux in favour of thermal, but it also modifies the energy spectrum of the fast neutrons, lowering the average energy. The response of the TLD-500 is consistent with it being totally

insensitive to thermal neutrons, and having a non-trivial energy response function for fast neutrons. These results are consistent with the neutron cross sections for the two different TLD materials. The thermal neutron absorption cross section for lithium is 70.5 ± 1.9 b [8], approximately 300 times larger than that for aluminum and many times larger still than that for any other material in either TLD². The lithium absorption cross section for fast neutrons is on the order of 1 b, approximately 70 times smaller than at thermal energies, and comparable to those of the other materials in the TLDs. Lithium is the only component of the TLDs that has a cross section that is significantly greater for thermal neutrons than for fast neutrons. The disadvantage that the TLD-100H has because of this is that there is no separation between the photon and neutron contributions to the TLD response: both components are ultimately measured together. The use of a combination of TLD-500s and bubble detectors allows these two components to be separated [3], which is of great importance when irradiating electronics.

3.4 Fading

As outlined in section 2.2.4, the effect of fading was measured over a two-week period by irradiating a set of TLDs to 10 mrem, and storing them with an equal number of unirradiated TLDs. Each day one each of the irradiated TLD-100H and TLD-500s were read-out and corrected for background with unirradiated TLDs. The dose measured as a function of time for the two different TLD types is shown in Figure 6. There is no evidence for fading in either the TLD-100H or the TLD-500 over two weeks.

² Recall that: TLD-100H is LiF:MgCuP and TLD-500 is Al₂O₃:C

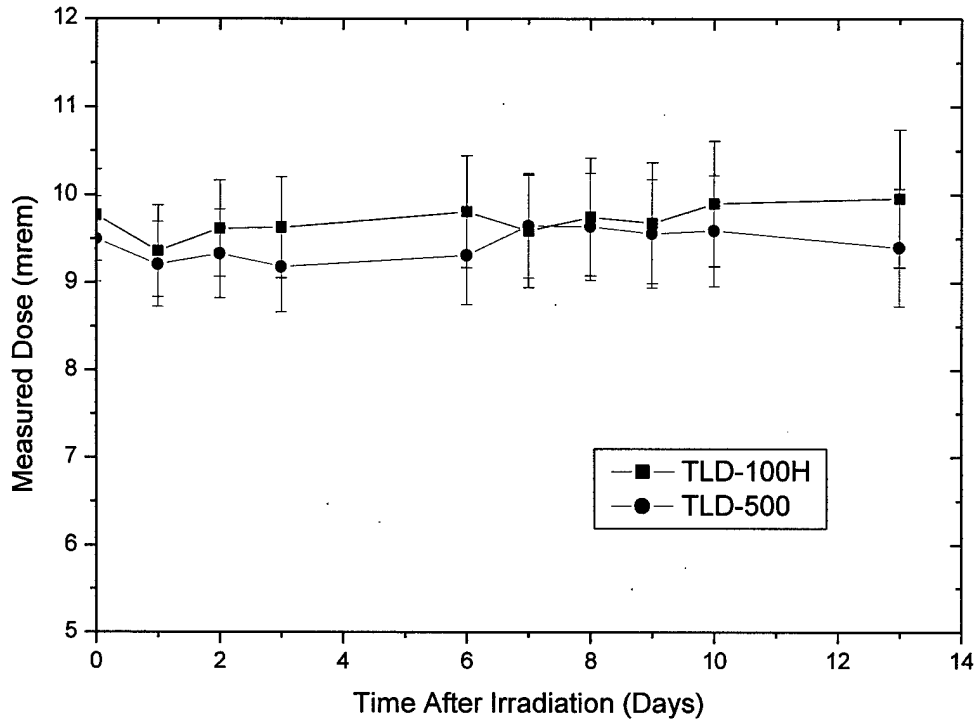


Figure 6. Dose measured in the TLD-100H and TLD-500 as a function of time for two weeks following irradiation to 10 mrem. There is no evidence for fading in either type of TLD.

3.5 Background Measurement

Background was measured by storing annealed TLDs away from radiation sources for a period of 21 days, after which the TLDs were read out. The measured background dose was 4.53 mrem for the TLD-100H and 3.27 mrem for the TLD-500. The background dose rate at the location where the TLDs was stored was measured with the Microspec-2. The measured background was 4.3 $\mu\text{rem/h}$, which translates to 2.17 mrem dose in 21 days. Both types of TLDs returned results that were higher than the background as measured with the Microspec, but the TLD-500 was closer to the correct result.

3.6 Other Issues

3.6.1 Annealing the TLD-100H

The initial annealing of TLDs is generally done in an oven rather than in the TLD reader. This procedure works well for the TLD-500, but some difficulties were encountered with the TLD-100H. After the initial anneal in the oven, the TLDs were exposed to 10 mrem for calibration. Upon reading the TLDs it was noticed that the glow curves for the TLD-100H were unusual in shape making the results suspect. Subsequent measurements with the TLD-100H without another oven anneal (just the normal post-read anneal in the reader) showed normal glow curves. The glow curves for the two cases are shown in Figure 7.

Correspondence with the TLD supplier highlighted the sensitivity of the TLD-100H to temperature during the annealing process. It was concluded that the metal plate on which the TLDs were placed for the oven anneal process kept the temperature of the TLDs below the optimum anneal temperature, resulting in the strange glow curves. For the remainder of the study, all anneal steps for the TLD-100H were performed in the reader only.

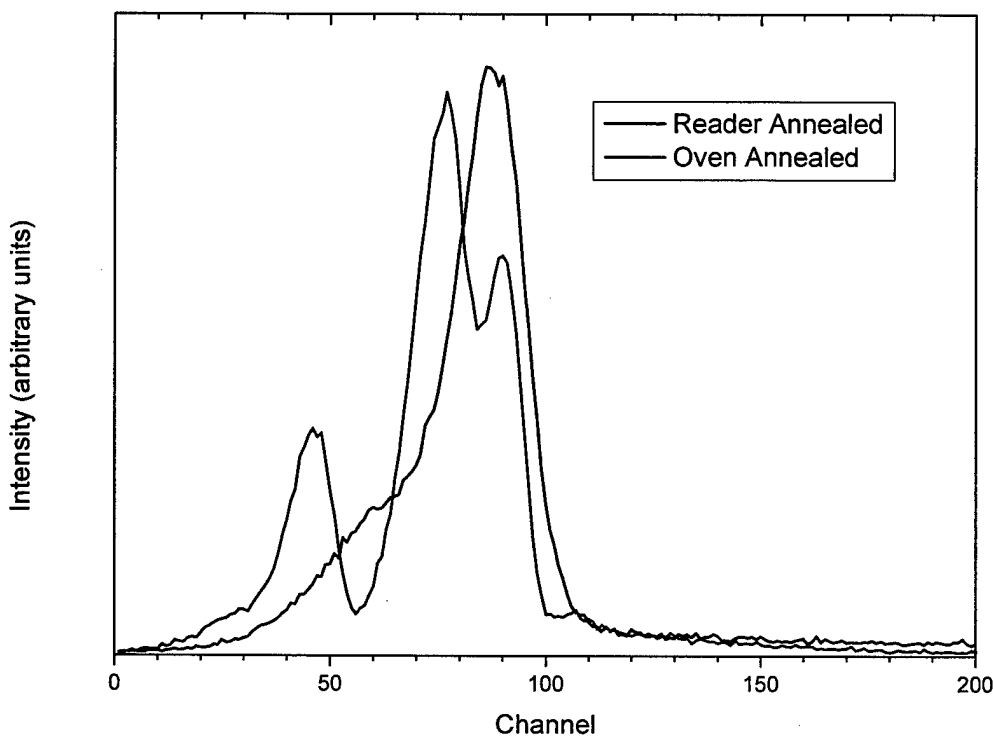


Figure 7. Typical glow curves for the reader-annealed and oven-annealed TLD-100H. The curves have been normalized in order to have the same integrated intensity. The reader-annealed curve matches what is expected (according to the manufacturer) for the TLD-100H.

3.6.2 X-ray sources

There are a few issues regarding the DREO x-ray sources that have been identified over the course of this study that should be addressed. These are in regards to both the quality of the x-ray spectra produced with, and the ease of use of, the sources.

DREO needs to have a standardized set of filters and settings for the x-ray sources in order to produce high quality, reproducible x-ray spectra. The spectra used in this report were obtained with quite a lot of trial and error, using different materials as filters both at the x-ray source and around the target. Filter combinations were found that resulted in reasonable spectra for most of the energies, but there is a lot of room for optimization. A systematic study of the x-ray system should be done to determine optimal filter combinations for a variety of x-ray energies. This would result in reproducible, high quality spectra that would be readily available for future studies requiring x-rays.

DREO has two x-ray sources, the Muller MG300 and the Lorad LPX200. Both of these machines have quirks of operation that need to be worked around in order to get the best results. The MG300 is an older machine that has a maximum energy of 300 keV. The machine works well, but the power supply has a few limitations. There are separate adjustments for voltage and current, but increasing the current has an effect on the output voltage, and therefore the shape of the produced spectrum. Optimizing the x-ray spectrum from the MG300 requires an iterative process, with adjustments to both voltage and current. Once the output is optimized, the dose rate from the machine cannot be increased by simply increasing the current because this changes the shape of the spectrum.

The LPX200 power supply is simpler to operate in that the current and voltage settings are more stable and much easier to reproduce. There are, however, two major limitations to the LPX200. The first is that it has a maximum operating voltage of 200 keV, limiting its maximum x-ray energy output to about 175 keV. If x-rays of energy 200 keV or higher are required, the MG300 must be used. The second limitation is that the power supply cannot be operated for an arbitrary period of time. There is a 99.9 minute time limit to the operation, which was inadequate for the exposures done in this study. All of the exposures at 10 mrem had to be done in 5-10 segments, which was inconvenient.

The use of the Microspec to measure the x-ray spectrum limited the maximum dose rate for the irradiations. In order to obtain reliable information on the shape of the x-ray spectrum the dose rate had to be low enough to avoid dead-time effects in the Microspec. Because this was the only dose rate measurement and spectrographic information available, the dose rate for the irradiation could not be increased without introducing unknown and unmeasurable effects into the shape of the spectrum. This led to inconveniently long exposure times.

4. Conclusions and Recommendations

The TLD-500 ($\text{Al}_2\text{O}_3:\text{C}$) outperformed the TLD-100H ($\text{LiF}:\text{MgCuP}$) in most of the areas that were studied. The TLD-500 had a more consistent response over a wider range of doses and energies, and had a desirable insensitivity to neutron radiation.

The TLD-500 response was flat for total doses ranging from 1 mrem up to 100 rem, while the TLD-100H response was flat over the range from 5 mrem to 10 rem. It should be noted that the manufacturer suggested dose range extends up to 100 rem for the TLD-500, but up to 1000 rem for the TLD-100H, much higher than where the response was found to be flat. The energy dependence of the TLD-100H response was, however, superior to that of the TLD-500. The TLD-100H response was flat between 150 keV and 1250 keV, and fell off at lower energies. The TLD-500 under-responded in the 150 to 200 keV range, but was otherwise similar in performance to the TLD-100H. This under-response of the TLD-500 can likely be corrected through the reduction in the thickness of tin used to wrap the TLD.

One major difference between the two types of TLD is in their response to neutrons. The TLD-100H responds to both fast and thermal neutrons, with the contribution from thermal neutrons being approximately ten times greater than for fast neutrons. The TLD-500 responds to fast neutrons, but does not seem to respond significantly to thermal neutrons. Response to thermal neutrons is an asset if the dosimeter is used for monitoring personal radiation dose to workers but is an unwanted source of background when the dosimeter is being used for scientific purposes.

In other areas there were no significant differences between the two types of TLD. There is no noticeable difference between the TLD-100H and the TLD-500 when it comes to fading, neither TLD showed signs of fading over a two-week period. Both TLDs measured background radiation within the range expected, although the value measured by the TLD-100H was higher than that measured by the TLD-500.

There are a few issues in handling the TLDs that are important. The TLD-500 is sensitive to UV light, while the TLD-100H is not. When handling the TLDs it is important to keep the TLD-500 in a dark environment to avoid exposure. This can be inconvenient, but with proper preparation it is not difficult to do. Proper annealing of the TLD-100H is essential for their correct operation. This can be done by annealing them in the reader, where the temperature is controlled precisely.

The ease of handling and good response to thermal neutrons make the TLD-100H an attractive dosimeter for monitoring radiation exposure of personnel. However, the TLD-500's insensitivity to thermal gives many more advantages, when it is combined with an accurate low-dose neutron detector such as the bubble detector [3].

DREO requires accurate dosimetry for a wide range of gamma and x-ray sources. Dose measurements are taken at DREO in a laboratory setting, where accuracy is essential and where any special handling requirements of the dosimeters can be

supplied. The TLD that is best suited to the requirements and working environment at DREO is the TLD-500 ($\text{Al}_2\text{O}_3:\text{C}$).

A further recommendation, not directly related to the DREO TLD dosimetry system, is that the procedures for the use of the DREO x-ray sources need to be improved. Clean, reproducible spectra from these sources are difficult to obtain at present. A comprehensive study into the DREO x-ray sources needs to be done in order to rectify this situation.

5. References

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Appendix A: TLD Calibration Factors

The calibration factors for individual TLD-100H samples are shown in Table 8 and those for the TLD-500s are shown in Table 9. The average calibration factor for the TLD-100H samples is 2.14 nC/mrem, and that for the TLD-500 is 4.30 nC/mrem. The average standard deviation for individual samples is 1.2% for the TLD-100H and 1.7% for TLD-500, while the standard deviation amongst all samples of TLD-100H is 3.4% and 5.4% for TLD-500.

Table 8. The calibration factors for the 25 samples of the TLD-100H used in this study. All values given in the table are in nC. There are three measurements for each TLD, all of which are for 10 mrem exposures to ⁶⁰Co gamma rays from the UDM-1 source. Also reported are the mean and standard deviations of the three measurements.

TLD#	#1	#2	#3	MEAN	St. Dev.
1	21.72	21.55	20.89	21.39	0.44
2	19.71	19.98	19.54	19.74	0.22
3	21.77	21.87	21.58	21.74	0.15
4	21.98	21.97	21.90	21.95	0.04
5	20.72	20.97	20.29	20.66	0.34
6	21.39	21.03	21.14	21.19	0.18
7	20.42	20.84	20.53	20.60	0.22
8	21.55	21.79	21.36	21.57	0.22
9	21.03	21.15	21.80	21.33	0.41
10	21.98	21.40	21.86	21.75	0.31
11	22.04	21.50	21.45	21.66	0.33
12	21.91	21.79	21.78	21.83	0.07
13	20.61	20.07	20.23	20.30	0.28
14	22.78	21.89	22.28	22.32	0.45
15	20.37	20.42	20.46	20.42	0.05
16	21.55	21.41	20.91	21.29	0.34
17	22.88	22.23	22.44	22.52	0.33
18	21.94	21.46	21.77	21.72	0.24
19	22.50	22.07	22.00	22.19	0.27
20	20.65	20.93	20.90	20.83	0.15
21	21.73	21.89	21.57	21.73	0.16
22	21.94	21.90	21.80	21.88	0.07
23	21.34	20.21	20.14	20.56	0.67
24	22.46	21.85	21.92	22.08	0.33
25	21.53	21.59	21.04	21.39	0.30

Table 9. The calibration factors for the 25 samples of the TLD-500 used in this study. All values given in the table are in nC. There are three measurements for each TLD, all of which are for 10 mrem exposures to ⁶⁰Co gamma rays from the UDM-1 source. Also reported are the mean and standard deviations of the three measurements.

TLD#	#1	#2	#3	MEAN	St. Dev.
1	44.31	43.92	43.12	43.78	0.61
2	46.84	45.80	44.69	45.78	1.08
3	44.91	44.36	44.07	44.45	0.43
4	43.05	42.32	41.88	42.42	0.59
5	42.85	42.58	41.93	42.45	0.47
6	41.73	40.95	41.15	41.28	0.41
7	38.51	38.22	37.81	38.18	0.35
8	40.11	39.17	39.34	39.54	0.50
9	39.66	39.12	39.03	39.27	0.34
10	39.93	39.24	39.05	39.41	0.46
11	46.88	46.03	45.62	46.18	0.64
12	43.14	42.57	41.66	42.46	0.75
13	43.04	41.99	41.02	42.02	1.01
14	44.32	43.87	43.09	43.76	0.62
15	43.77	43.14	41.89	42.93	0.96
16	43.74	42.08	42.18	42.67	0.93
17	46.36	45.87	44.10	45.44	1.19
18	45.66	45.22	44.84	45.24	0.41
19	44.63	43.07	42.28	43.33	1.20
20	45.04	44.11	43.40	44.18	0.82
21	43.35	42.84	42.32	42.84	0.52
22	44.48	44.49	43.50	44.16	0.57
23	42.65	41.96	41.44	42.02	0.61
24	46.43	44.29	44.34	45.02	1.22
25	47.90	47.56	44.72	46.73	1.75

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A study was conducted comparing the properties of two types of thermoluminescent dosimeters (TLDs): LiF:MgCuP (TLD-100H) and Al₂O₃:C (TLD-500). This was done in order to determine which is better suited for the dosimetry requirements of the Radiation Effects Group at DREO (RE/DREO). The dose and energy response of the TLDs were measured by exposing them to a wide range of photon doses and energies. In addition, the sensitivity of the TLDs to fast and thermal neutrons was determined with a PuBe source. Fading and background detection were also studied. The TLD-100H dosimeter was found to be slightly more tissue-equivalent in its energy response but the TLD-500 spanned a larger dose range. In addition, TLD-500 had a desirable insensitivity to thermal neutrons. The effect of fading and response to background were similar in the two types of TLD. The TLD-500 dosimeter was found to be better suited to the environments employed by the RE/DREO.

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Thermoluminescent dosimeter, dosimetry, TLD, TLD-100H, TLD-500, dose dependence, energy dependence, neutron response, fading, background, x-ray, gamma-ray

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