HIGH TEMPERATURE PEAK CHARACTERISTICS OF THE READER-ANNEALED TLD-600 AND ITS APPLICATION TO PERSONNEL PROTECTION DOSIMETRY IN MIXED NEUTRON-PHOTON FIELD

James C. Liu
Stanford Linear Accelerator Center, Stanford, CA 94309, USA

C. S. Sims
Oak Ridge National Laboratory, Oak Ridge, TN 37831-6379, USA

ABSTRACT

The high temperature peaks 6-7 of TLD-600 have higher responses to high LET radiation than to low LET radiation. This characteristic was studied for the automatic reader-annealed Harshaw albedo neutron TLD. The high temperature peaks response is linear for neutrons, but is supralinear above 20 mSv of ^{137}Cs photons. The peaks ratio (peaks 6-7/peaks 3-5) of TLD-600 is 0.15 for neutrons of any energy, 0.01 for ^{137}Cs gammas and 0.02 for M-150 x-rays. Based on the results, a personnel dosimetry using a single TLD-600 was developed and evaluated in mixed neutron-photon fields. The estimations for neutron, photon and total equivalent are better than 20% except in one case. However, an error analysis shows that the estimations are sensitive to the neutron and photon peaks ratios, depending on the neutron-photon dose equivalent ratio and the neutron source.

INTRODUCTION

The TL glow curve of the LiF-TLD (TLD-100, TLD-700, TLD-600) has several peaks, among which peaks 3-5 are main dosimetric peaks (peak 5 at ~200°C), and peaks 6-7 are high temperature peaks (peak 7 at ~260°C). Other peaks are usually not important for dosimetric purposes. The high temperature peaks have higher responses to high LET radiation than to low LET radiation and this characteristics can be influenced by many factors, e.g., TLD material, annealing, cooling, readout method, etc. Conventional long and high temperature oven annealing for LiF-TLD is usually not used for automatic TLD systems. This paper presents the high temperature peak characterization results for the Harshaw automatic reader-annealed TLD-600. Based on the results, a mixed field neutron-photon dosimetry using a single TLD-600 element was developed and evaluated in mixed fields. A few factors which may affect the accuracy of the dosimetric method are discussed.

MATERIALS AND METHODS

Harshaw albedo neutron TLDs (two pair of TLD-600/TLD-700; one pair is shielded in front by a 28×13×0.46 mm^3 cadmium sheet) were used in this study. The sensitivities of all TLD chips (3.2×3.2×0.9 mm^3) were individually calibrated with free-in-air ^{137}Cs irradiations. The TL signals were normalized to a constant ^{137}Cs exposure and were in units of mGy. The Harshaw 8800 automatic TLD reader was used to readout and anneal the TLDs. The digitized 200-channel TL glow curves of the TLD-600 exposed to neutrons or photons from the Harshaw 8800 reader are shown in Figure 1. Neutrons produce much higher peaks 6-7 and slightly lower peaks 3-4 than photons. The linear heating profile (no preheat, a heating rate of 25°C s^{-1} from 50°C to 300°C, and a hold time of 6.7 s at 300°C) using hot N_2 gas in this study is also shown in Figure 1. The Computerized Glow Curve Deconvolution (CGCD) program[1] was used to separate the glow curve into individual peaks, but the deconvolution result was not satisfactory. This might be due to the first-order TL kinetic model in the CGCD program being inappropriate to describe the TL response of a neutron-exposed TLD-600.

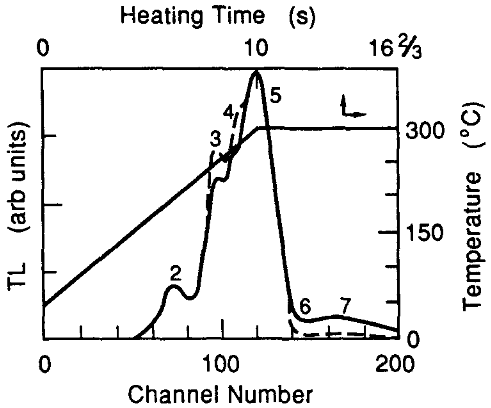

Figure 1. TL glow curves induced by neutrons (—) or photons (- - -) for TLD-600. Peaks 3-5 cover channels 96-145 and peaks 6-7 cover channels 146-200. The heating profile used is also shown.

Therefore, in this study (see Figure 1), the TL signal between channels 96-145 is regarded as peaks 3-5 and the TL signal between channels 146-200 is regarded as peaks 6-7. It has been found[(2)] that these channel settings can achieve a satisfactory sensitivity and response stability over reuse, and minimize the fading influence from peaks 2 and 3. The TLDs were reader-annealed using the same heating profile just prior to irradiation. However, repeated annealings or annealing using a longer hold time (20 s) at 300°C was used sometimes for highly dosed TLDs in order to reduce the residual TL signal to an acceptable level. No pre-irradiation or post-irradiation, low temperature annealing was used.

Four radioisotopic neutron sources [^{252}Cf(D_2O), ^{252}Cf(PE), ^{252}Cf and ^{238}PuBe], eight monoenergetic neutrons (0.1, 0.25, 0.565, 1.2, 2.6, 3.2, 5.0 and 14.8 MeV), ^{137}Cs and the M150 x-rays were used for the TLD irradiations[(3)]. The ^{252}Cf(PE) source is a ^{252}Cf moderated by a 15 cm radius polyethylene sphere. The errors associated with neutron fluences were 10-15% for monoenergetic neutrons and 5-10% for radioisotopic neutron sources. All TLDs were irradiated perpendicularly with dosimeters mounted on the front face of a 40×40×15 cm^3 Lucite phantom, except the M150 x-rays irradiations which were made using a 30×30×15 cm^3 phantom. The dose equivalent quantity used is the ICRP 21 neutron dose equivalent quantity[(4)] for neutrons and is the deep dose equivalent quantity[(4)] for photons. The photon contribution from the neutron source to the TLD-600 signal was estimated by the paired TLD-700 element.

CHARACTERIZATION RESULTS

LINEARITY

The TL response for the Cd-covered TLD-600 exposed to ^{252}Cf(PE) are shown in Figure 2. The linear response level for peaks 6-7 is up to ~2.56 mGy at 3 mSv neutron exposure. Since both peaks 6-7 and peaks 3-5 have linear responses over the range of 0.05-3 mSv, the total TL response (peaks 3-7 = the sum of peaks 3-5 and peaks 6-7) is also linear. The peaks ratio, peaks 6-7/peaks 3-5, is equal to the slope ratio in Figure 2. Therefore, the neutron peaks ratio for ^{252}Cf(PE) is a constant of (0.854/5.921)=0.144 (1σ=2%) over the dose range. The neutron sensitivity of the Cd-covered TLD-600, defined as the peaks 3-7 response, for ^{252}Cf(PE) is (0.854+5.921)= 6.78 mGy mSv^{-1}. Figure 3 shows the linear response curves of the Cd-covered

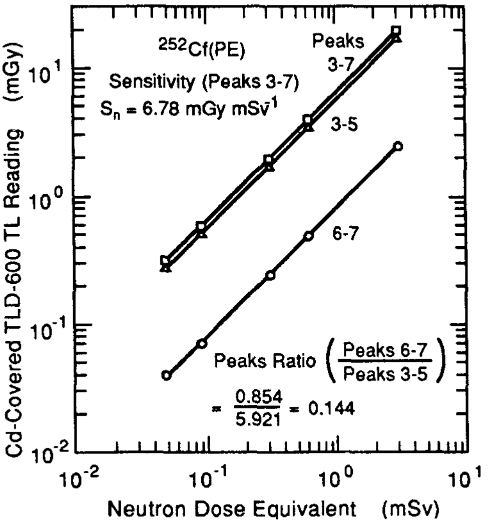

Figure 2. TL response linearities of the peaks 3-5, peaks 6-7, and total peaks 3-7 for the Cd-covered TLD-600 exposed to the ^{252}Cf(PE) neutrons.

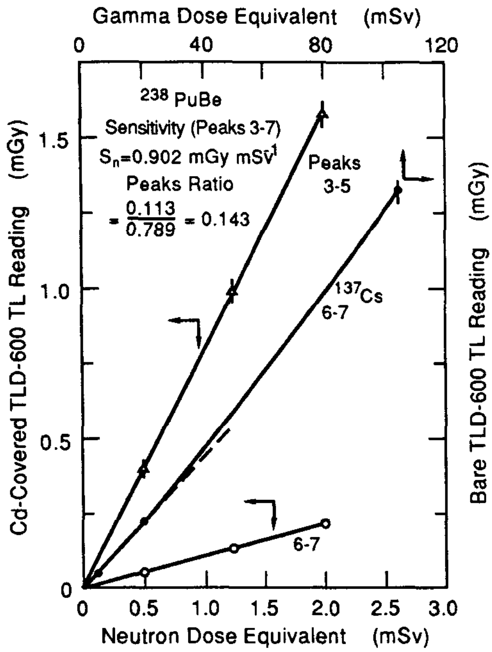

Figure 3. TL response linearities of the peaks 3-5 and peaks 6-7 for the Cd-covered TLD-600 exposed to ^{238}PuBe neutrons. The supralinearity of the peaks 6-7 response for bare TLD-600 exposed to ^{137}Cs photons is also shown.

TLD-600 for ^{238}PuBe. The neutron peaks ratio is (0.113/0.789)=0.143 (1σ=3%), which is very close to that of ^{252}Cf(PE), but the neutron sensitivity is only (0.113+0.789)=0.902 mGy mSv^{-1}, due to the albedo neutron detection principle.

Figure 3 also shows the peaks 6-7 response of the bare TLD-600 exposed to ^{137}Cs free-in-air. The linear response level for peaks 6-7 is up to only 0.23 mGy at 20 mSv gamma exposure and the linear region has a slope of 0.012 mGy mSv^{-1}. The deviation from linearity (see dashed line in Figure 3) is ~15% overreponse at 103 mSv. The peaks 3-5 response of the TLD-600 exposed to ^{137}Cs, which is not shown in Figure 3, is linear up to 100 mSv with a slope of 1.045 mGy mSv^{-1}. Therefore, the photon peaks ratio of the TLD-600 for ^{137}Cs is a constant of (0.012/1.045)=0.01 (1σ=10%) only up to the level of 20 mSv, due to the supralinear response of peaks 6-7.

Since the peaks 6-7 response is only ~1% of the peaks 3-5 response for a gamma-exposed TLD-600, the supralinearity of peaks 6-7 response is masked by the linearity of peaks 3-5 response. Therefore, the total peaks 3-7 response for a gamma-exposed TLD-600 is treated as linear in most personnel protection dosimetric practices.

The finding that the supralinearity of peaks 6-7 is LET-dependent (the lower the LET, the lower the TL response level at which supralinearity occurs) is consistent with results previously reported in Refs. 5 and 6. However, the gamma dose levels at which the supralinearity occurs are different between our results and others (~100 mGy for TLD-100 in Ref. 5, 2.5 mGy for TLD-700 in Ref. 6, and ~20 mGy for TLD-600 in this work). It is also demonstrated that the peaks 6-7 have a higher response to neutrons than to photons; a factor of (0.113/0.012)=9.4 between ^{238}PuBe and ^{137}Cs, and a factor of (0.854/0.012)=71 between ^{252}Cf(PE) and ^{137}Cs.

PEAKS RATIO AND SENSITIVITY

Since the Cd-covered TLD-600 responds mainly to the albedo thermal neutrons, the peaks ratio is expected to be the same for all incident neutron energies. The peaks ratios of the Cd-covered TLD-600 for the monoenergetic and radioisotopic neutron sources ranged from 0.143 to 0.163 and the mean neutron peaks ratio was 0.15 (1σ=7%). The neutron sensitivity (S_n) follows the typical energy-dependent curve of an albedo-type TLD (the response is high at low energies and is low at high energies).

Contrary to the neutron results, the photon peaks ratio is energy-dependent; 0.01 (1σ=10%) for ^{137}Cs 662 keV gammas and 0.02 (1σ=3%) for M150 x-rays. The photon sensitivity (S_p) of the Cd-covered TLD-600 is slightly energy-dependent (0.872 mGy mSv^{-1} for ^{137}Cs gammas and 0.916 mGy mSv^{-1} for M150 x-rays), due to the tissue-equivalence of the TLD-600 to photons.

Budd et al.[7] studied the peaks ratios of the TLD-600 to x-rays and their peaks ratio values are a factor of five higher than ours, probably due to the slow cooling they used. However, a comparison on the relative peaks ratio as a function of photon energy (the peaks ratio at a given energy divided by the peaks ratio for ^{137}Cs gammas) between their results and ours shows good agreement (see Figure 4). The peaks ratios for neutrons and photons in this work are close to those of Doles et al.[8] who used TLD-100 with different readout and annealing techniques.

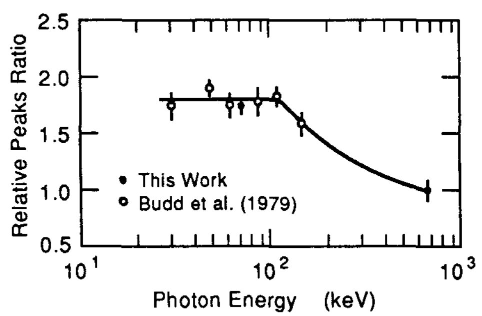

Figure 4. Relative peaks ratios as a function of photon energy (peaks ratio at a given energy divided by that of ^{137}Cs photons) for the Cd-covered TLD-600.

MIXED NEUTRON-PHOTON FIELD DOSIMETRY

A neutron-photon dosimetry using a single Cd-covered TLD-600 element in mixed fields can be developed by using the peaks ratio and sensitivity values. A Cd-covered TLD-600, irradiated to a neutron dose equivalent (H_n mSv) and a photon dose equivalent (H_p mSv), has a total peaks 3-7 signal of T mGy. Let the peaks ratio be PR (i.e., PR = peaks 6-7/peaks 3-5) and $K = PR/(1+PR)$ (i.e., K = peaks 6-7/peaks 3-7). The following two equations can be established.

$$T_h = H_n S_n K_n + H_p S_p K_p \quad (1)$$

$$T_l = H_n S_n (1 - K_n) + H_p S_p (1 - K_p) \quad (2)$$

T_h, T_l = measured peaks 6-7 and peaks 3-5 TL signals in units of mGy, respectively, and $T = T_h + T_l$

K_n, K_p = K values for neutron and photon radiations, respectively

S_n, S_p = neutron and photon sensitivities (peaks 3-7), respectively, of the Cd-covered TLD-600 in units of mGy mSv^{-1}

$H_n S_n K_n$ = TL signal component of peaks 6-7 contributed by neutrons.

Since *PR* is 0.15 for all neutrons, K_n is 0.13 for all neutrons. The value K_p is dependent on photon energy and can be determined from Figure 4. If neutron and photon energies are known, there are only two unknowns, H_n and H_p, to be solved in equations 1 and 2.

A test of the above mixed field dosimetry was made by irradiating eight groups of albedo TLDs to two neutron dose equivalents (0.5 and 1.5 mSv) with four H_n/H_p ratios (2.6/1, 1/1, 1/3 and 1/10), using both ^{238}PuBe and ^{137}Cs sources. The small 4.43 MeV gamma dose equivalent component (~4%) of the ^{238}PuBe source[3] was included in the photon dose equivalent. The test results presented in Table 1 show the bias (*B*), precision (*P*) and accuracy (*A*) values in percentage for the neutron, photon and total (neutron + photon) dose equivalent estimations in eight mixed fields by using the four Cd-covered TLD-600 elements per exposure group.

Table 1. Dose equivalent measurement performance of the Cd-covered TLD-600 in mixed ^{238}PuBe + ^{137}Cs fields, using the high temperature peaks method.[a] The values for *B*, *P*, and *A* are in percentage.

			Neutron			Photon			Total[f]		
H_n[b]	H_n/H_p	H_p[b]	*B*[c]	*P*[d]	*A*[e]	*B*	*P*	*A*	*B*	*P*	*A*
0.5	2.6/1	0.19	-22.0	6.6	28.6	28.9	4.1	33.0	-7.2	3.3	10.5
	1/1	0.52	-14.0	3.3	17.3	7.7	3.0	10.7	-2.9	1.0	3.9
	1/3	1.52	-10.0	3.1	13.1	2.6	1.1	3.7	-0.5	1.3	1.8
	1/10	5.02	0	9.8	9.8	1.4	1.3	2.7	1.3	1.8	3.1
1.5	2.6/1	0.57	-12.0	5.9	17.9	7.0	6.2	13.2	-6.8	4.8	11.6
	1/1	1.57	-10.0	4.0	14.0	1.9	3.2	5.1	-3.9	2.7	6.6
	1/3	4.57	-7.3	3.5	10.8	1.3	1.6	2.9	-0.8	2.0	2.8
	1/10	15.07	4.0	4.3	8.3	-1.3	0.9	2.2	-0.9	1.1	2.0

[a] Peaks ratio is 0.15 for neutrons and 0.01 for ^{137}Cs photons.

[b] H_n is the neutron dose equivalent from ^{238}PuBe, H_p is the photon dose equivalent from both ^{137}Cs and the 4.43 MeV gamma component of ^{238}PuBe (~4% of its neutron dose equivalent)[3] in mSv units.

[c] Bias (*B*) is $(H - H_o)/H_o$, where H is the mean dose equivalent estimated from the four Cd-covered TLD-600 elements per exposure group and H_o is the reference value.

[d] Precision (*P*) is one relative standard deviation per group.

[e] Accuracy (*A*) is the sum of the absolute value of bias and precision.

[f] Total dose equivalent (neutron + photon) estimation.

The neutron or photon bias is small when the H_n/H_p is small, and the bias is also smaller at the higher neutron dose equivalent level. The largest bias (-22% for neutrons and 29% for photons) occurs in the mixed field with H_n = 0.5 mSv and H_n/H_p = 2.6/1. The photon precision is better when the H_n/H_p is smaller (<7% in all cases). The neutron precision is better in the fields with H_n/H_p = 1/1 or 1/3 (<10% in all cases).

Since the precision values are smaller than the corresponding bias values in most fields, the accuracy values show the same trend as the bias values. The worst accuracy is 29% for neutrons and 33% for photons in the field of H_n=0.5 mSv and H_n/H_p=2.6/1, while in the other fields the accuracy values are better than 18%. The total dose equivalent estimation is very good (accuracy is better than 12% in all cases) due to the opposite bias in the neutron and photon dose equivalent estimations. The opposite bias result is expected due to the use of a single TLD element to estimate both neutron and photon dose equivalents in our methodology.

ERROR ANALYSIS

The good dose equivalent measurement performance shown in Table 1 is an ideal case in which the neutron and photon sources are known (so peaks ratios and sensitivities are both known with small errors). In real fields, the photon and neutron spectra may be known only to a limited extent. In that case, although photon sensitivity has a small error due to the small energy-dependence of TLD-600 to photons, the photon peaks ratio may have a large error (peaks ratio varies from 0.01 to 0.02). In contrast to the case for photons, the neutron sensitivity may have a large error if the neutron energy is not well known, while the neutron peaks ratio is still a constant of 0.15. In either case, the uncertainty in photon or neutron energy can result in error to the neutron and photon dose equivalent estimations.

An error analysis can be performed by calculating the variations of the neutron and photon dose equivalent estimations as a function of the variation of the neutron or photon peaks ratio. Figure 5 shows the fractional changes in the neutron and photon dose equivalent estimations, if the photon peaks ratio is changed from 0.01 to 0.02. For this increase in the photon peaks ratio, the fractional change in the photon dose equivalent estimation is increased, but the fractional change in the neutron dose equivalent estimation is decreased. The fraction increase in the photon dose equivalent estimation is 9-11% in any field, regardless of neutron source type and the H_n/H_p ratio. The fractional decrease in the neutron dose equivalent estimation, however, shows strong dependence on both the neutron source type and the H_n/H_p ratio. Extreme results in the ^{137}Cs fields mixed with $^{252}Cf(D_2O)$ or $^{238}PuBe$ are shown in Figure 5. The fractional decrease is higher when H_n/H_p is lower; it is <10% in the mixed fields with $^{252}Cf(D_2O)$ in any H_n/H_p ratio; the decrease can be as high as 90% in a $^{238}PuBe$ mixed field with a H_n/H_p of 0.1. Fortunately, the fractional change of total dose equivalent, which can also be estimated from Figure 5, is less than 8% in any mixed field with $^{252}Cf(D_2O)$ and less than 1% in any mixed field with $^{238}PuBe$.

Figure 6 shows the fractional changes in the neutron and photon dose equivalent estimations, if the neutron peaks ratio is changed from 0.15 to 0.14 (i.e., changed by 1σ). The situation in Figure 6 is reversed from that in Figure 5. The fractional increase in the neutron dose equivalent estimation is 9-10% in any mixed field, regardless of neutron source type and the H_n/H_p ratio. The fractional decrease in the photon dose equivalent estimation has strong dependence on both the neutron source and the H_n/H_p ratio. The fractional decrease is higher when H_n/H_p is higher; it is as high as 95% in a $^{238}PuBe$ mixed field with a H_n/H_p of 10. The fractional decrease in the photon dose equivalent estimation in a $^{252}Cf(D_2O)$ mixed

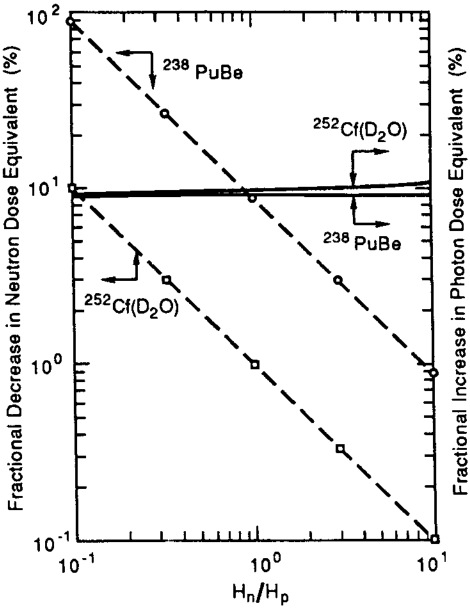

Figure 5. Fractional changes in the neutron and photon dose equivalent estimations in mixed neutron-photon fields, if the peaks ratio for photons is changed from 0.01 to 0.02. Extreme results in the mixed fields with two different neutron sources are shown.

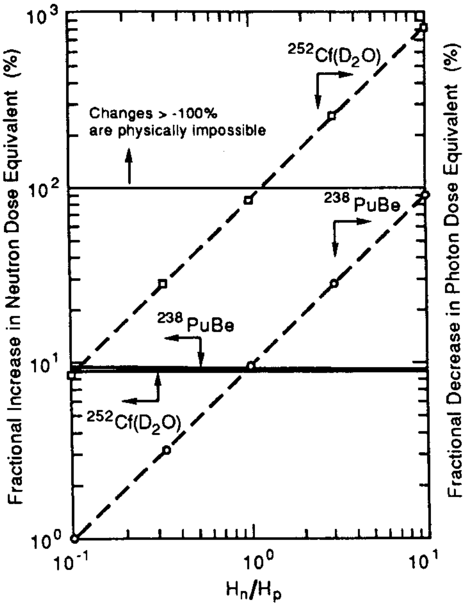

Figure 6 Fractional changes in the neutron and photon dose equivalent estimations in mixed neutron-photon fields, if the peaks ratio for neutrons is changed from 0.15 to 0.14. Extreme results in the mixed fields with two different neutron sources are shown.

field could be larger than -100%, but this is only a calculated value and is physically impossible. The fractional change in the total dose equivalent estimation in a mixed field with ^{238}PuBe is <1% in all cases. The fractional decrease in the total dose equivalent estimation in a $^{252}Cf(D_2O)$ mixed field with a $H_n/H_p = 1$ could be as high as 50%.

DISCUSSION

The upper linear response level of the peaks 6-7 in our study is ~3 mSv for neutrons and ~20 mSv for gammas. Using a three-month dosimeter exchange period, the maximum neutron and gamma dose equivalent limits per year with no supralinear peaks 6-7 response are 12 mSv and 80 mSv, respectively. Therefore, the high temperature peak dosimetry is suitable for most protection dosimetry situations, but not for accident dosimetry.

Low sensitivity of peaks 6-7 to photons (only 0.012 mGy mSv^{-1}) might lead to an impression of insufficient photon sensitivity of this method for use in protection dosimetry. Key points in the high temperature peak methodology are: using the high peaks 3-7 sensitivities of the TLD-600 for photons and neutrons to detect both photons and neutrons, and using the very different peaks 6-7 sensitivities for photons and neutrons to differentiate the photons and neutrons. Therefore, the photon and neutron peaks ratios should be accurately determined, so that the photon and neutron signals can be well separated. The good test results in Table 1 also prove that the method is appropriate for protection dosimetry.

Other concerns are the reproducibility of the peaks ratio during reuse and the variation of the peaks ratios within a group of TLDs. For example, the fading of peaks 2-3 would affect the peaks ratio value if the fading effect is not properly accounted for. A more stable peaks ratio value can be obtained, at the expense of total sensitivity, by using a narrower region of interest (e.g., covering only peaks 4-5). Our experience shows that the current settings of the two regions of interest and the heating profile can achieve a satisfactory result for at least a one-month fading period. The peaks ratio may not be chip-dependent, but it can be batch-dependent. A simpler solution is to use the mean peaks ratio for a batch, if the variation of the peaks ratios within a batch is acceptable. A more complicated solution is to generate individual neutron and photon peaks ratio values for every TLD-600 element. This is a tedious but not difficult procedure, and the mass data manipulation associated with it is easy in a computer-aided TLD system.

CONCLUSION

The high temperature peaks characteristics of the reader-annealed TLD-600 have been studied. The high temperature peaks have linear responses for neutrons, but supralinearity starts at about 20 mSv for gammas. The peaks ratio is 0.15 for neutrons of any energy, and is energy-dependent for photons (0.01 for ^{137}Cs, and up to 0.02 for x-rays below ~100 keV). A mixed field neutron-photon protection dosimetry using a single Cd-covered TLD-600 element was developed and evaluated in different mixed field conditions. The results and an error analysis show that such mixed-field dosimetry would work well if both the neutron and photon sources are known. Otherwise, the neutron and photon dose equivalent estimations may have large errors, depending on the peaks ratio error, the neutron source type, and the neutron/photon dose equivalent ratio in the mixed field.

ACKNOWLEDGEMENTS

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