Radiation Protection Dosimetry
Vol. 49 No. 4 pp. 443–449 (1993)
Nuclear Technology Publishing

ERROR ANALYSIS OF NEUTRON DOSE MEASUREMENT IN MIXED GAMMA–NEUTRON FIELDS BY A TWO PEAK TL METHOD

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Received September 10 1992, Revised May 20 1993, Accepted June 21 1993

Abstract — Expressions for errors of separate measurements of gamma and neutron doses by the two peak TL method were derived. As follows from these expressions, the main factors governing the errors are precision of the TL peaks measurement, neutron–gamma ratio of peaks and gamma dose contribution to the mixed gamma–neutron field. Calculation of determination errors of neutron dose in a mixed gamma–neutron field by single crystal ^{7}LiF:Mg,Ti detectors shows that the detectors are completely suitable for accident dosimetry.

INTRODUCTION

Among different methods of thermoluminescence (TL) dosimetry of mixed fast neutron and gamma radiation fields the two peak method is considered to have the best prospects[1]. The method is based on the simultaneous measurement of two TL peaks of one detector with different dependences of sensitivity on ionising radiation LET. One such TL phosphor, CaF_2:Tm, is used for dosimetry of neutron beams[2] and pions[3]. However, its neutron–gamma ratios (relative neutron sensitivities) are not sufficiently high (0.05 for 150°C peak and 0.18 for 250°C at the neutron energy of 14 MeV[4]) for it to be used for accident and personal neutron dosimetry. Another thermoluminophor of the same type is LiF:Mg,Ti with the 200°C peak used for measurement of gamma radiation dose and a 260°C peak which has increased sensitivity to heavy charged particles and neutrons[5,6]. Because of the high sensitivity of ^{6}Li to thermal neutrons the two peak method with ^{6}LiF:Mg,Ti is successfully employed for dosimetry of mixed fields of gamma radiation and thermal neutrons[7–9]. Fast neutron measurement by the two peak method is used in clinical dosimetry of neutron beams[10,11], when the neutron dose is high and the contribution from accompanying gamma radiation is small. Possible use of LiF:Mg,Ti detectors for accident dosimetry was described previously[1], where the main dosimetric parameters of DTGN-2 gamma–neutron detectors designed in our Institute were presented.

The main difficulty in using LiF:Mg,Ti as a gamma–neutron detector is connected with the measurement of the weak 260°C peak partially overlapped by the intense 200°C peak. At the present time a computer deconvolution of a glow curve is widely used for solving this problem: this allows us to separate needed dosimetric peaks from the total curve[12,13]. Application of this method to TLD-100 detectors allows the lower detection limit of the 200°C peak for gamma irradiation to be decreased by a factor of 14[12] and to the 260°C peak to be isolated at gamma irradiation doses more than 5 mGy[13]. Another way to separate the 200 and 260°C peaks is sequential reading out of the peaks using a special profile of the detector heating, such as a linear stepwise heating[1,14].

Strong supralinearity of the 260°C peak under gamma irradiation is its salient feature[9,11,13], which significantly complicates the detector calibration and the neutron dose measurement. To overcome this difficulty an attempt was made to use the LiF:Mg,Ti, sensitised by gamma irradiation, as a gamma–neutron dosemeter[15,16]. After sensitisation the dose dependences of the 200 and 260°C peaks sensitivities become linear and neutron sensitivity increases. However, simultaneously increasing a high temperature background and decreasing the 260°C peak neutron–gamma ratio depreciates any virtues of the method.

Another way to avoid the difficulties connected with the 260°C peak non-linearity is, in the case of a narrow dose range, linearisation of the detector dose dependence. This method is suitable for neutron beam dosimetry in clinical therapy[11]. Simultaneous sensitivity of both the peaks to gamma radiation and neutrons is an attributive property of the method considered.

Therefore, the measurement precision of gamma radiation and neutron doses is subject not only to the measurement precision of TL peaks but also to the ratio of doses in mixed

gamma–neutron fields. The latter circumstance limits the capabilities of the two peaks method. For clarifying the possible application of the method and its scope for future development, a detailed examination of the sources of measurement errors of gamma and neutron doses is needed. The calibration of the detectors with non-linear gamma dose response and the effect of different factors, especially the dose ratios, on the measurement errors of gamma radiation and neutron doses in mixed gamma–neutron fields have been carefully considered and are here reported.

EXPERIMENTAL

Six gamma–neutron DTGN-1 detectors were used for all measurements. The detectors were ^{7}LiF:Mg,Ti single crystal discs 5 mm diameter and 1 mm thick. Irradiation of the detectors by gamma and neutron radiation was carried out using ^{60}Co and ^{252}Cf sources, respectively. TL curves were read out using the UNIC-02 TL complex[17] which provides the possibility for flexible control of the detector heating profile depending on the glow curve behaviour. For separation of the 200°C peak from the 260°C peak double linear heating was used. The second heating was made after the 200°C peak maximum had passed and following some seconds cooling of the detector with the heater switched of. The heating profiles and corresponding TL curves of the DTGN-1 detector irradiated with ^{252}Cf gamma–neutron radiation are shown in Figure 1. It is clear that the heating profile used allows one to separate the 200 and 260°C peaks well. TL yields of the 200 and 260°C peaks were determined as their intensities (I_1 and I_2, respectively).

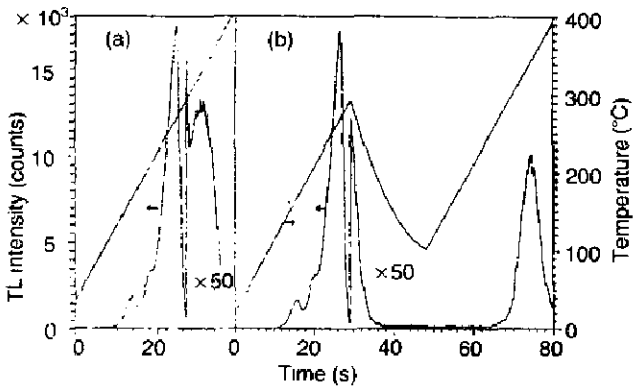

Figure 1. Glow curves of the LiF:Mg,Ti (DTGN-1 No 3) detector irradiated by the ^{252}Cf source (neutron dose 0.202 Gy, gamma dose 0.106 Gy) for (a) linear and (b) double linear heating. Temperature shift of the peaks maxima is caused by raised (10°C.s^{-1}) heating rate used.

BASIC RELATIONS

Under the detector irradiation in the mixed gamma–neutron field a storage of energy in both the TL peaks occurs. Therefore, dependences of TL yields on doses or dose equivalents of gamma radiation (D_γ) and neutrons (D_n) can be expressed as follows:

$$I_1 = a_{1\gamma}(D_\gamma)D_\gamma + a_{1n}D_n \quad (1)$$

$$I_2 = a_{2\gamma}(D_\gamma)D_\gamma + a_{2n}D_n \quad (2)$$

Coefficients $a_{1\gamma}$, $a_{2\gamma}$, a_{1n} and a_{2n} are gamma and neutron sensitivities of corresponding TL peaks. Generally speaking, these coefficients have a complicated dependence on an energetic spectrum and dose of ionising radiation and design of the dosemeter. In order to avoid these difficulties we only consider a case when dose is measured in the same spectrum of ionising radiation as the spectrum used for the calibration. In this work the calibration and measurement are considered for the ^{252}Cf source widely used in practical dosimetry. When the radiation spectrum is fixed the coefficients of sensitivity will be dependent only on the dose of radiation. Considering strong supralinearity under gamma irradiation the dependences of $a_{1\gamma}$ and $a_{2\gamma}$ on D_γ in Equations 1 and 2 are represented in an explicit form. Information being available about linearity of dependence of a_{1n} and a_{2n} on a neutron dose under ^{252}Cf irradiation at doses less than 10 Gy[1] but taking into account all the data on the dose dependences of the 200 and 260°C peaks under alpha[18] and neutron[8] irradiation, it was assumed that the coefficients a_{1n} and a_{2n} are independent of the neutron dose at least up to 100 Gy.

It follows from Equations 1 and 2 that for determination of gamma and neutron doses in mixed gamma–neutron fields by LiF:Mg,Ti detectors it is necessary during preliminary calibration to determine the gamma dose dependences $a_{1\gamma}(D_\gamma)$ and $a_{2\gamma}(D_\gamma)$ and values of the coefficients a_{1n} and a_{2n}. It is then necessary to measure the intensities of the 200 and 260°C peaks, substitute their values in Equations 1 and 2 and solve this system for D_γ and D_n.

Non-linear Equations 1 and 2 are conveniently solved by an iteration procedure, rewriting them as follows:

$$D_\gamma = \frac{I_1 (1 - C_n I_2/I_1)}{a_{1\gamma}(D_\gamma)(1 - C_n/C_\gamma)} \quad (3)$$

$$D_n = \frac{I_2 (C_\gamma - I_1/I_2)}{a_{2n}(C_\gamma - C_n)} \quad (4)$$

where $C_\gamma = a_{1\gamma}(D_\gamma)/a_{2\gamma}(D_\gamma)$ and $C_n = a_{1n}/a_{2n}$ are

Table 1. Calibration parameters.

Detector number	Peak 200°C			Peak 260°C			
	A (Gy^{-1})	B	D_1 (Gy)	A (Gy^{-1})	B	D_1 (Gy)	a_{2n} (Gy^{-1})
3	149000	6.19	59.3	577	44.2	45.3	550 ± 140
6	157000	6.09	66.8	607	41.7	44.4	560 ± 160
25	123000	6.55	63.5	503	45.9	51.0	570 ± 220
102	161000	5.14	40.2	365	60.3	27.3	580 ± 230
313	165000	5.66	66.1	546	45.0	38.2	630 ± 160
361	170000	6.21	73.8	522	44.9	34.3	800 ± 380

ratios of gamma and neutron sensitivities of the 200 and 260°C peaks.

It follows from Equation 3 that for determination of gamma dose in a mixed gamma–neutron field we must have not only the neutron sensitivity ratio but the sensitivities. It should be noted also that in the case of ^{252}Cf irradiation of the LiF:Mg,Ti, C_n is much less than C_γ. Therefore, in Equations 3 and 4, C_n may be neglected. Physically this means neglecting the neutron contribution to the 200°C peak yield in comparison with the gamma one.

CALIBRATION OF THE DETECTOR UNDER GAMMA IRRADIATION

A detector for routine dosimetry usually has a linear dose dependence in dose range used; therefore, for calibration, it is enough to determine the sensitivity of the detector at some dose. In the non-linear case sensitivity of the detector depends on dose and calibration proves to be a sufficiently complicated problem. There are two approaches to calibration of a detector with non-linear dose dependence. The first one consists of measuring sensitivities at some doses and determination of their intermediate values by interpolation. The second way is to define an analytical function describing the dose dependence to an arbitrary precision in the dose ranged used.

At the present time there are a number of analytical expressions in the literature describing dependences of the 200 and 260°C peak yields on the dose of gamma irradiation. When the dose range described is narrow, a combination of linear and power (as a rule quadratic) functions is commonly used[11,19,20]. For a wide dose range a number of types of dependences have been proposed[21,22]. Some of them are empirical, others have a theoretical basis. The question of the dependence of TL yield upon gamma irradiation dose is complicated by a circumstance that data obtained by different authors differ significantly. This concerns especially the 260°C peak for which different dose dependences have been obtained for different batches of the same detectors[11,19]. Furthermore, there is at present no consensus on a region of linearity for this peak[10,13]. Our own operating experience with LiF:Mg,Ti detectors testifies that sensitivity and supralinearity of the 260°C peak are not closely connected with sensitivity of the 200°C peak and vary from detector to detector; thus, at present individual calibration of the detectors is really necessary.

For a description of the peaks' dose dependences under gamma irradiations expressions obtained in previous work will be used[23]; these describe the experimental data up to 10^6 Gy well. For the dose range used (0–100 Gy) this expression can be rewritten in the form:

$$I(D) = A\, D_\gamma (B + (1-B) \exp(-D_\gamma/D_1)) \qquad (5)$$

where A, B and D_1 are parameters (A governs sensitivity in the region of linearity, B governs the value of supralinearity and D_1 is a dose at which supralinearity appears).

It is considered that the main merit of this expression is that it was derived on the basis of fundamental relations between distribution of the absorbed energy density in ionising particle tracks and dose distribution in an irradiated thermoluminophor[23,24]. Moreover, at $D_\gamma \ll D_1$ Equation 5 reduces to linear quadratic dependence; the applicability of this at small doses was noted above. For obtaining the parameters A, B and D_1 dose dependences of the 200 and 260°C peaks for each of six detectors used have been measured and approximated by Equation 5. Fitting was made by the weighted non-linear least squares method. As a weight an inverse value of error mean square of the peak intensity was used which was represented as

$$\Delta I^2 = I_b^2 + \sigma_o^2 I^2 \qquad (6)$$

where σ_o is the error of the TL yield (I) measurement in the dose range, where its value is much more than background value (I_b).

Values of σ_o for the 200 and 260°C peaks were assumed to be 5 and 10% respectively (the same

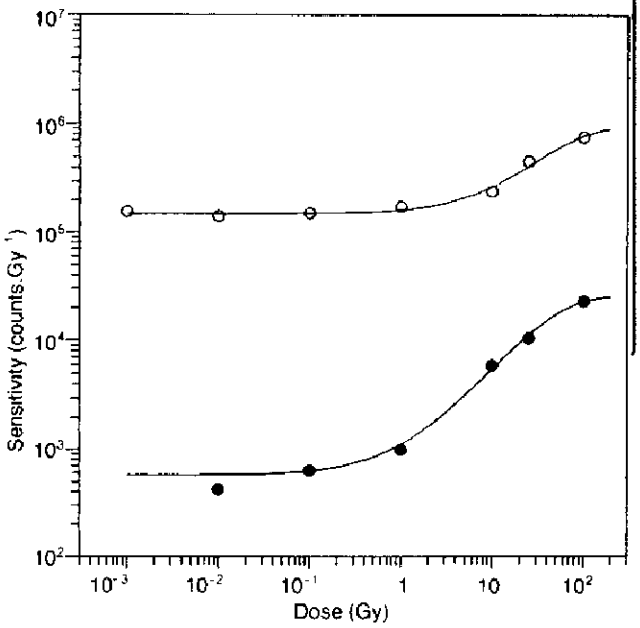

Figure 2. Experimental (points) and calculated (lines) dose dependences of the 200 (o) and 260°C (•) peaks gamma sensitivities.

as for DTGN-2 detectors[1]. The mean value of the TL curve intensity at the beginning of the second heating (see Figure 1(b)) was used as the background value. The parameters obtained in this way are listed in Table 1.

Dose dependences of $a_{1\gamma}$ and $a_{2\gamma}$ for detector No 3, experimentally determined and calculated by Equation 5, are shown in Figure 2. A quality of the fitting is illustrated by Figure 3 where mean errors of sensitivity approximation and the error standard deviations are shown. It is evident that for the 200°C peak Equation 5 provides a good fit (the error is less than 10%) to experimental dose dependence over all the dose range investigated. Error variance of the approximation for the 260°C peak increases with dose decrease because of the difficulty of measuring this peak at low doses. Deviations of the experimental data (small but exceeding experimental error) from values calculated by Equation 5 should be noted; they seem to be caused by simplifying assumptions used during the Equation 5 derivation.

CALIBRATION OF THE DETECTOR UNDER NEUTRON IRRADIATION

Determination of the detector neutron sensitivity is complicated by the fact that all neutron sources have accompanying gamma radiation. As follows from Equation 2:

$$a_{2n} = (I_2 - I_{2\gamma})/D_n \qquad (7)$$

where $I_{2\gamma} = a_{2\gamma}(D_\gamma)D_\gamma$ is the contribution of gamma irradiation to the 260°C peak.

Measurement error ΔY of a certain value Y dependent on some other statistically independent values X_i (i=1,...,n) may be found by formula:

$$(\Delta Y)^2 = \sum_{i=1}^{n} \left(\frac{\partial Y}{\partial X_i}\Delta X_i\right)^2 \qquad (8)$$

where ΔX_i is the measurement error of value X_i. Whence

$$\Delta a_{2n}^2 = (\Delta I_2^2 + \Delta I_{2\gamma}^2)/D_n^2 \qquad (9)$$

Neutron sensitivities of the detectors were determined by Equation 7 as a result of measuring the 260°C peak intensity after irradiation by the ^{252}Cf source with neutron doses of 0.024, 0.202, 0.899 and 8.62 Gy. The dose of accompanying gamma radiation was $D_\gamma = 0.53\ D_n$. The gamma contribution to the 260°C peak was calculated using Equation 5 with parameters listed in Table 1. Correction was made for regular deviation of Equation 5 from the experimental dose dependences.

Mean neutron sensitivities of the 260°C peak, $\bar{a}_{2n}$, and their variances σ_{2n} were calculated by formulae for measurements with different precisions[25]:

$$\bar{a}_{2n} = \sum_{i=1}^{n} (a_{2n})_i w_i / \sum_{i=1}^{n} w_i \qquad (10)$$

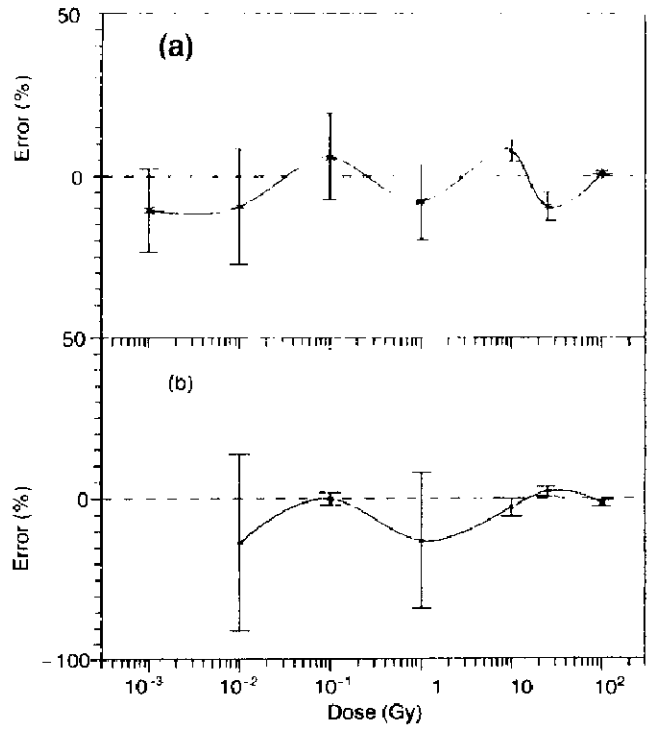

Figure 3. Error means and standard deviations of the (a) 200 and (b) 260°C peaks dose dependences approximation by Equation 5.

$$\sigma^2_{2n} = \sum_{i=1}^{n} (a_{2n} - \bar{a}_{2n})^2_i \, w_i/(n-1) \qquad (11)$$

where $w_i = 1/\Delta a^2_{2n}$ is a weight of the ith measurement.

Values of ΔI^2_2 were calculated by Equation 6. Values of ΔI^2_γ were assumed equal to variances of fitting of the 260°C peak gamma dose dependence shown in Figure 3(b). The mean values of the 260°C peak neutron sensitivity obtained in this way and their variances are listed in Table 1.

For the 200°C peak contribution of accompanying gamma radiation $I_{1\gamma} = a_{1\gamma}(D_\gamma)D_\gamma$ agrees with the intensity of this peak under gamma–neutron irradiation I_1 to within the measurement error. It indicates that the neutron contribution to the 200°C peak under the ^{252}Cf source irradiation is negligibly small, comparing the contribution of accompanying gamma radiation. Such a result is in a good agreement with data of the work[26] where the neutron–gamma ratio of the 200°C peak under the ^{252}Cf source irradiation was shown to be equal to 0.026 ± 0.014. A low value of the 200°C peak neutron–gamma ratio allows one to neglect the neutron contribution to this peak in real gamma–neutron fields (except when the contribution of accompanying gamma radiation is rather small) and to determine the dose of gamma radiation using the relation $I_1 = a_{1\gamma}(D_\gamma)D_\gamma$.

DOSE MEASUREMENT ERRORS IN MIXED GAMMA–NEUTRON FIELDS

The general Equation 8 and Equations 3 and 4 (with C_n=0 for simplification) were used to derive an expression for measurement errors of gamma and neutron doses. The measurement error of gamma dose is determined by the expression:

$$(\Delta D_\gamma/D_\gamma)^2 = (\Delta I_1/I_1)^2 + (\Delta a_{1\gamma}/a_{1\gamma})^2 \qquad (12)$$

The measurement error of neutron dose can be expressed as

$$(\Delta D_n/D_n)^2 = K^2 [(\Delta I_1/I_1)^2 + (\Delta a_{1\gamma}/a_{1\gamma})^2 + (\Delta D_\gamma/D_\gamma)^2] + (1+K)^2 (\Delta I_2/I_2)^2 \qquad (13)$$

where $K = a_{2\gamma}D_\gamma/a_{2n}D_n$.

It follows from these equations that the main factors that govern the errors are precision of TL peaks measurement, their neutron–gamma ratio and the ratio of neutron and gamma doses in mixed fields. When neutron dose is measured in a pure neutron field with a small addition of gamma radiation ($D_\gamma << a_{2n}D_n/a_{2\gamma}$), determination error of neutron dose is the same as the measurement error of the 260°C peak. With a D_γ increase, the determination error of neutron dose becomes dependent on the 200°C peak measurement error which causes the measurement error of accompanying gamma radiation dose and, as a result, determination error of gamma radiation contribution to the 260°C peak. Therefore, with an increase in the fraction of accompanying gamma radiation the determination error of neutron dose will increase and it can reach a significant value.

Equations 12 and 13 were used for calculation of the determination error of neutron dose in mixed gamma–neutron fields. Errors of the detector calibration were neglected because they depend on calibration procedure and may be reduced to a negligible value by increasing the number of calibration measurements. Values of measurement errors of the TL peaks intensities needed for the calculation were determined by Equation 6 with the coefficients from Table 1. Results of the calculation for DTGN-1 detector (No 3) are shown in Figure 4. As is obvious from the calculation, detectors DTGN-1 can be used for measuring a neutron dose of more than 0.04 Gy in a neutron field with a small contribution of accompanying gamma radiation. If this contribution increases then the measurement range becomes limited at the upper end because of the 260°C peak neutron–gamma ratio decrease. For a gamma–neutron field of a ^{252}Cf source with a gamma contribution of 50%, the neutron dose range which can be measured by DTGN-1 detectors (with error less than 50%) is 0.04–10 Gy.

The abilities for measuring neutron dose in

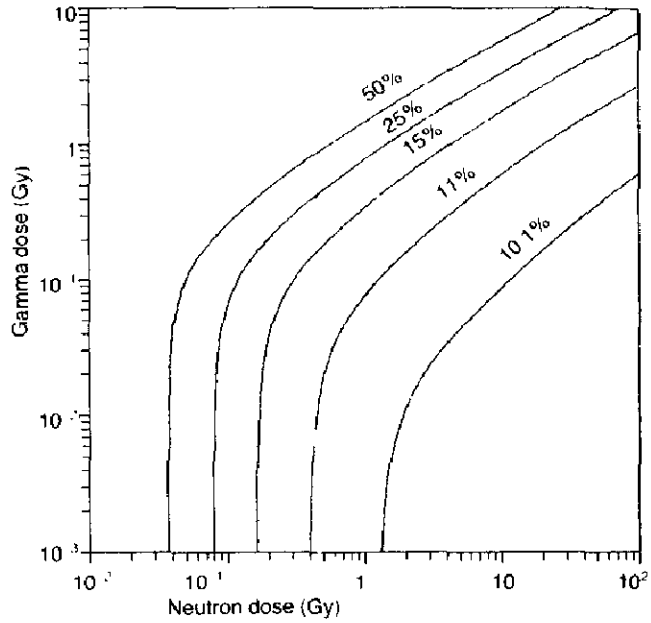

Figure 4. Dependence of neutron dose measurement errors (indicated on the curves) on gamma and neutron doses in mixed fields.

mixed gamma–neutron fields by the DTGN detector achieved at the present time are shown in Figure 4. There are two ways to improve the detector performance. The first way, connected with decrease of the measurement error of the peaks' intensity, is illustrated by Figure 5(a) where values of σ_0 for the 200 and 260°C peaks were put equal to 1 and 2%, respectively. Other parameters were the same as for curves shown in Figure 4. The second way is to increase a neutron sensitivity of the 260°C peak. Expected results are shown in Figure 5(b). The curves are calculated as those shown in Figure 4 but a_{2n} was increased 10 times.

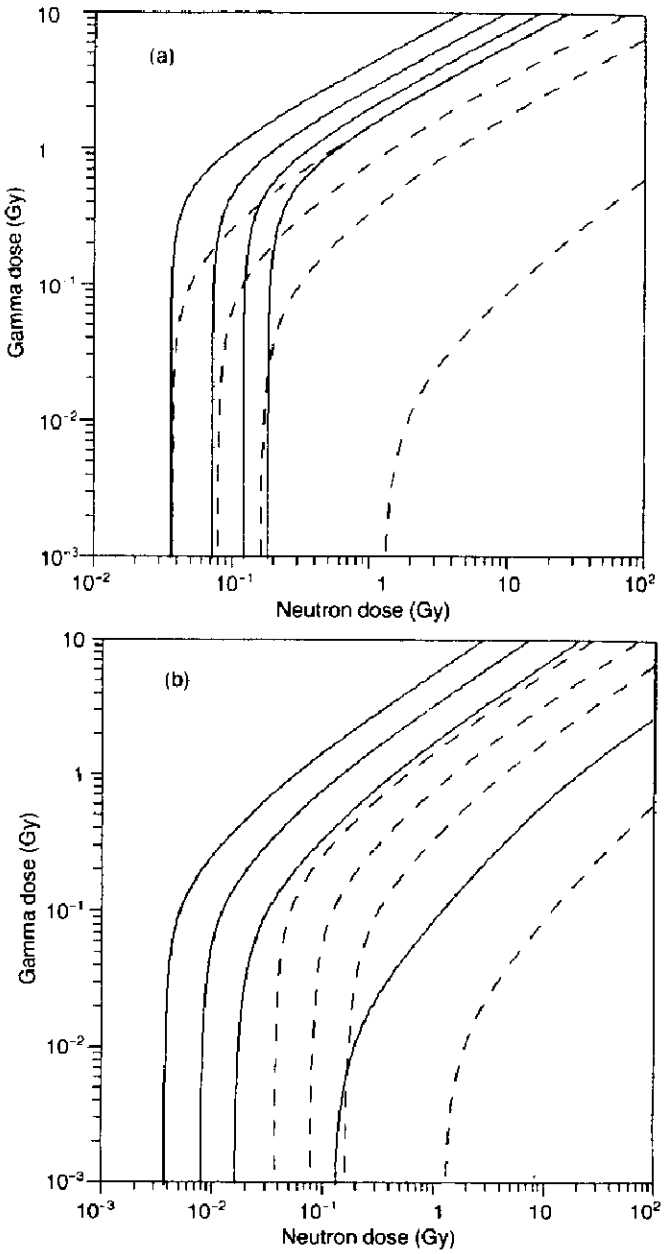

Figure 5. Curves borrowed from Figure 4 (dashed lines) and these calculated (a) at the peak measurement errors reduced to one-fifth and (b) at a neutron–gamma ratio of the 260°C peak increased 10 times (solid lines). Values of neutron dose error for both the sets are 10.1, 15, 25, and 50% from right to left.

CONCLUSION

Error analysis carried out above shows that the two peak method of separate measurement of gamma and neutron doses is sufficiently complicated and needs accurate measuring. The main source of errors (besides measurement error of TL yield) is determination error of the gamma contribution to the 260°C peak. Evaluation of this contribution is complicated by the strong supralinearity of the gamma dose dependence of this peak yield which involves the need of a great number of calibration measurements. It should be noted that high sensitivity of this peak to heavy charged particles and neutrons is closely allied with its strong supralinearity[24]. Therefore, difficulties caused by supralinearity of the 260°C peak are inevitable for the LiF:Mg,Ti based two peak method. An attempt to decrease the supralinearity by sensitisation of LiF:Mg,Ti[15,16] or by using CaF_2:Tm produces a decrease of neutron–gamma ratios and an increase of neutron dose measurable error if accompanying gamma irradiation exists.

According to the requirements for accident dosimetry of gamma–neutron radiation it is necessary to measure separately gamma and neutron doses in the range of 0.1–10 Gy with errors less than 25% SD (see Ref. 27). Comparing this requirement with the data shown in Figure 4 it can be seen that DTGN-1 detectors may find possible application in accident gamma–neutron dosimetry under the condition that the dose of accompanying gamma radiation is less than the dose of neutrons.

Comparing this requirement with data shown in Figure 5 one can see that having decreased the measurement error of the peaks' intensities and having increased the neutron–gamma ratio of the 260°C peak, gamma and neutron doses in mixed fields can be measured separately with any relation of the components in the dose range considered.

Other possible applications of the DTGN-1detector may be in clinical neutron dosimetry, where neutron beams are used which generate doses in the range of tens of Gy with accompanying gamma radiation of some per cents. Under such conditions dose measurement presents no difficulty and the possibility of simultaneous measurement of accompanying gamma radiation makes the method more attractive for clinical dosimetry.

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