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Neutron-gamma mixed field measurements in Hp(10) by means of a TLD600-TLD700 dosemeter pair

SOSA VERA, Cristian[1,2*]; GUARIN CABRERA, Luis[3]; ANDRES, Pablo[1,2]

[1] *Instituto Balseiro, Universidad Nacional de Cuyo*
[2] *Div. Protección Radiológica, Gerencia Ingeniería Nuclear, Comisión Nacional de Energía Atómica*
[3] *Dpto. Reactores de Investigación, Gerencia Ingeniería Nuclear, Comisión Nacional de Energía Atómica*
*Corresponding autor: cristian.sosavera@ib.edu.ar

Abstract. Mixed neutron-gamma field dosimetry still stands a challenge because of the difficulty to experimentally discriminate the dose from each field component. As explained in the bibliography, the use of a suitable pair of dosemeters is an option to discriminate the contributions of gamma photons and neutrons in the mixed field. The thermoluminescence dosemeters ^{7}LiF: Mg, Ti (TLD-700) and ^{6}LiF: Mg, Ti (TLD-600) are usually chosen for measurements in a neutron-gamma mixed field. The TLD-600 is much more sensitive to thermal neutrons than the TLD-700. On the other hand, the sensitivity to gamma photons of both types of dosemeters is approximately equal. In this work, the method applied for neutron-gamma personal dosimetry used in the Radiological Protection Division of the Bariloche Atomic Center was studied. The dosemeter response in terms of the personal equivalent dose Hp(10) was analysed from different measurements. The irradiations were carried out with the dosemeters on the ISO slab phantom. The reference radiations were from radionuclide sources, for gamma field ^{137}Cs and ^{60}Co were used, while for mixed neutron-gamma field, ^{241}AmBe^{9}. The latter´s activity was previously verified by an absolute method of activation of an indium foil and high-resolution gamma spectrometry. Doses delivered were measured empirically and the neutron flux linked to these doses were verified analytically by Monte Carlo simulations. The TLD-700 reading could be linked to the gamma dose by applying the gamma calibration factor obtained. Regarding the neutron dose, with the tested dosemeter, the presence of neutron doses can be detected, but not quantified. In order to obtain a neutron calibration factor, it would be necessary to modify the dosemeter to differentiate the incident neutrons from those of albedo (backscattered in the phantom). This new albedo dosemeter design will be based on the recommendations of ICRU-66 and to meet the ISO-21909.

KEYWORDS: Dosimetry, Thermoluminescence, Neutron-gamma field.

1 INTRODUCTION

The exposure to radiation of workers is controlled by individual monitoring measurements. Neutron dosimetry shows more operational difficulties than photon dosimetry. One reason is that the quality factor at the point of interest depends on the secondary charged particle spectrum and hence the neutron energy [1]. Therefore, dose equivalent cannot be measured directly by conventional dosimetry. No neutron detector can measure, without additional information, the whole neutron energy spectrum ranging from thermal (0.025 eV) to 20 MeV [2]. This is the range of energies found in occupational neutron fields [3]. Furthermore, reference neutron fields are not easily available and all individual neutron dosemeter responses present energy dependence [4].

Around the world, individual monitoring is performed with a wide variety of active and passive detectors. The passive individual dosemeters are still the most commonly used technique. Among these, thermoluminescence dosemeters (TLD) are the most widely used for neutron individual monitoring, which have a low detection limit and angle dependence [1]. In gamma-neutron mixed fields ^{6}LiF: Mg, Ti and ^{7}LiF: Mg, Ti are usually used as a thermoluminescent detector pair (TLD-600 and TLD-700). TLD-600 containing ^{6}Li are used to detect low-energy (slow) neutrons. This is due to the large cross-section reactions ^{6}Li(n,α)^{3}H. The photon sensitivity is the same for both thermoluminescent materials. Consequently, the difference in the readings of both detectors is the slow-neutron reading.

Owing to the strong energy dependence of the albedo dosemeter response, a single calibration factor cannot be used in different neutron fields with widely varying spectra if accurate dose results are to be obtained. Instead, location specific calibration factors must be established based on a characterization of neutron energy spectrum at each location. It is necessary to keep a record of the locations in which the dosemeter was used in order to apply the appropriate calibration factor to the reading [1].

In accordance with the recommendations of ICRU Report 39, all instruments are to be calibrated in terms of the operational dose equivalent quantities. Hp(d) is the dose equivalent in ICRU standard tissue at a depth d, in the body of an exposed person, with the recommended depth, d, being 10 mm for strongly penetrating radiation. Calibration of these quantities requires the dosemeter to be placed on a phantom that provides a reasonable approximation to the backscatter properties of that part of the body on which it is worn. For measurements of Hp(10), a phantom of outer dimensions 30 cm x 30 cm x 15 cm with PMMA walls filled with water should be used. Besides, reference radiation sources should be used for dosemeter calibration. The ISO 8529-1 describes the characteristics and methods of production of the reference neutron radiations. While the ISO 4037-1 specifies the characteristics and production methods of X and gamma reference radiation for calibrating protection-level dosemeters [5], [6].

ISO 21909 provides performance and test requirements for determining the acceptability of personal neutron dosemeters. These are to be used for the measurement of Hp(10), including neutron fields energies ranging from thermal to 20 MeV. This standard covers five classes of passive neutron detectors that can be used as personal dosemeters including TLD [3].

2 MATERIALS

2.1 Dosemeters and reading equipment

In this work, TLD-700 and TLD-600 were used, whose characteristics are shown in Table 1.

Table 1. Characteristics of the TLDs used

	Brand	Composition	Dimensions [mm]	Mass [mg]
TLD-600	Rados	^{6}LiF: Mg, Ti; 95.62% ^{6}Li	3.2 x 3.2 x 0.9 ± $1x10^{-1}$	23.03 ± $1x10^{-3}$
TLD-700	Harshaw	^{7}LiF: Mg, Ti; 99.99% ^{7}Li	3.2 x 3.2 x 0.9 ± $1x10^{-1}$	23.84 ± $1x10^{-3}$

The TLDs were measured in a 3500 Harshaw manual TLD reader (see Figure 1.). The time and temperature profiles (TTP) configured for the TLD readings with the Winrems software are shown in Table 2.

Figure 1. 3500 Harshaw TLD reader and Winrems software used.

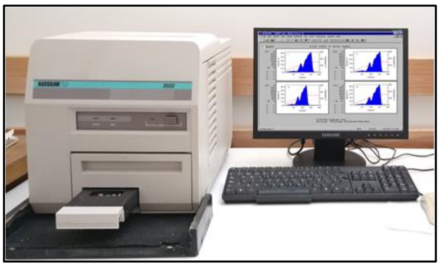


Table 2. Time and temperature profiles used for reading TLDs

TTP	TLD-700	TLD-600
Preheating	16 [s] at 110 [°C]	16 [s] at 110 [°C]
Acquisition: temp. rate	12 [°C/s]	12 [°C/s]
Acquisition: max. temp.	255 [°C]	280 [°C]
Acquisition: total time	23.33 [s]	23.33 [s]

2.2 Irradiation sources

The irradiation sources used are described in Table 3.

Table 3. Gamma calibration sources. [a]Average emission energy. [b]Calculated on the basis of the neutron spectra given in annex A of ISO 8529-1 and the conversion coefficients given in ICRU Report 57. [c]Exposure rate at 1 m from source of ^{137}Cs.

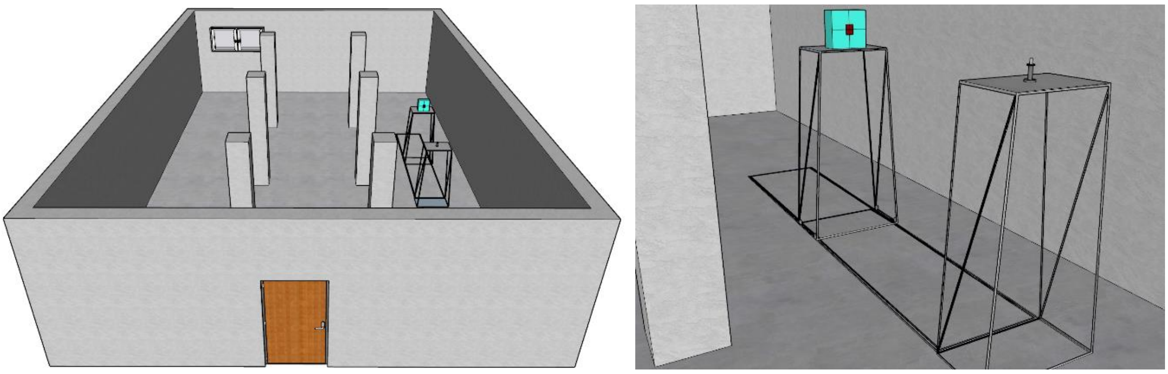
Radionuclide	^{60}Co	^{137}Cs	^{241}Am-Be[9]
$T_{1/2}$ [y]	5.27	30.08	432.6
E [MeV] [a]	1.252	0.662	4.4 [b]
Calibration date	7/12/2018	7/5/1981	20/9/1978
Calibrated intensity	21.5 ± 5% [mGy.m^2.h^{-1}]	327 ± 5% [c] [mR.h^{-1}]	7.4 x10^6 ± 10% [n.s^{-1}]
A_0 [Ci]	1.9 ± 5%	1 ± 5%	3 ± 10%

Since the neutron source calibration was 41 years old, prior to its use, it had been decided to carry out the verification of the neutron intensity. For this, an absolute method of indirect determination of the neutron intensity had been applied. This method consisted of measuring the neutron flux through activation on an indium foil with the reaction $^{115}In\ (n, n'\gamma)^{115m}In$, which was sensitive to the energy range issued by the source. After irradiation, the activity of the foil was measured with a calibrated high-resolution gamma spectrometer, with which the number of counts of the gamma photopeak in question was integrated. It was concluded that the neutron intensity had been reduced to 6.9x10^6 ± 11% [n/s] and the activity to 2.8 ± 11% [Ci], at the measurement date [7].

2.3 Irradiation room

The place where the irradiations were carried out was a room with dimensions of 9 m x 13.5 m with a height of 3 m and 6 columns of 40 cm x 40 cm distributed equally. Its walls, floor, ceiling and columns were made of concrete. Due to the occupation of the room for other activities, the irradiations were carried out on one side of the room, 1.65 m from one of its walls. The source and the phantom were placed on the supports of the irradiation system (made of light materials), standing 1.35 m above the ground (see Figure 2).

Figure 2. Irradiation room (left) and the irradiation set (right).

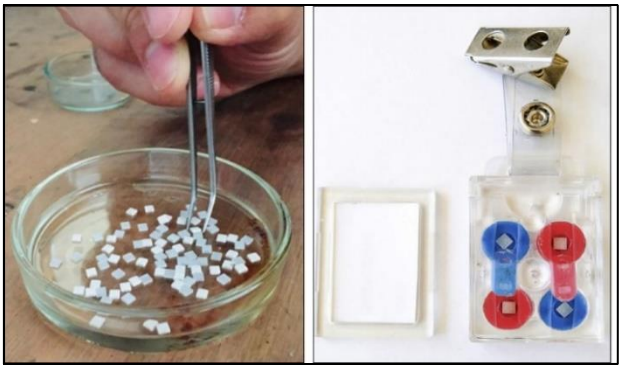


3 METHODOLOGY

3.1 Dosemeter preparation

Before each irradiation, the TLDs were placed in a Petri dish and annealed at 400 °C for one hour. After this, they were rapidly cooled and then placed in another oven at 75 ºC for 24 hours. The rapid cooling consisted in transferring the TLDs from the Petri dish to another one at room temperature. After 24 hours, they were removed from the oven and the dosemeters were assembled as shown in the Figure 3. The assembly of the dosemeters consisted of placing two pairs of TLD-600 and TLD-700 inside plastic cups and these in the acrylic dosemeter holder.

Figure 3. Assembly of the dosemeter.



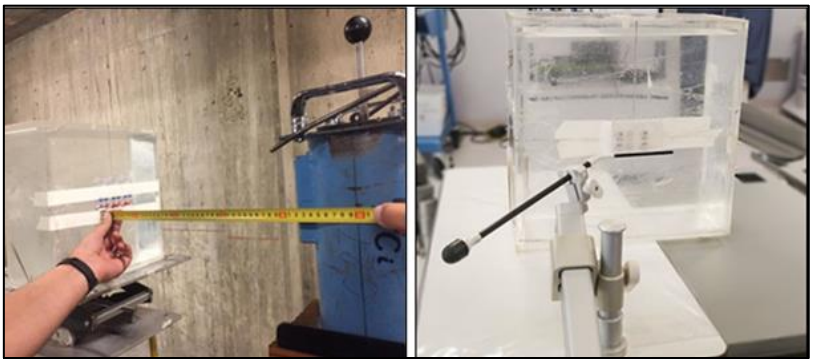
3.2 Gamma irradiations

For the ^{137}Cs and ^{60}Co irradiations, the dosemeters were separated into groups, each containing between 8 and 10 TLD-600 and TLD-700. In all cases, one group was left as "zeros" (background measurement), and the remaining groups were irradiated at a distance (dose rate) and time calculated according to the activity of the source on the day of irradiation (see Table 4). In the case of the ^{137}Cs source, the magnitude used for the calculations is the exposure, since it is the magnitude reported by the manufacturer of the source. Therefore, it is converted by calculation into personal equivalent dose, Hp(10), to express the results. Irradiations with the ^{137}Cs source were carried out in the irradiation room. While those with ^{60}Co were performed in the brachytherapy room, where it is usually used (see Figure 4). In both cases, the ISO slab phantom was used.

Table 4. Gamma irradiations parameters.

Gamma source	Distance [m]	Hp(10) [mSv]
^{137}Cs	0.5	0; 5.1; 10
^{137}137	1	0; 1; 10
^{60}Co	0.1	0; 100; 500; 1000
^{60}Co	0.2	0; 10; 50

Figure 4. Gamma irradiation set. Irradiations with ^{137}Cs (left). Irradiations with ^{60}Co (right)



3.3 Neutron irradiations

Neutron irradiations were performed with the ^{241}Am-Be[9] reference source in the irradiation room. They were separated into groups of four dosemeters, always leaving one group as "zeros" (background measurement), and the remaining groups were irradiated at the determined distance (dose rate) for a time calculated according to the activity of the source on the day of irradiation (see Table 5). The irradiations were performed with and without phantom. In the latter case, it was evaluated the contribution to the dose caused by the dispersion of neutrons in the irradiation room. Neutron fluence conversion factors to personal equivalent dose were used to calculate the Hp(10) [8]. The neutron fluence of the source was calculated analytically and verified by simulation in MCNP. In addition, these calculations were compared with measurements from a Thermo model FHT-752 neutron detector (see Figure 5).

Table 5. Neutron irradiations parameters

Distance [m]	Time [h]				Hp(10) [mSv]			
0.316	3	9.5	30	95	2.4	7.6	24	76
1	-	9.5	30	95	-	0.76	2.4	7.6
3.06	3	9.5	30	95	0.027	0.085	0.27	0.85

Figure 5. Neutron irradiation set. Irradiations with and without phantom (left). Measurements of the dose rates with the FHT-752 neutron detector (right).

3.4 Gamma calibration factor

The gamma calibration factor ($CF\gamma$) is the slope of the calibration line, which was obtained by linear regression of the points measured in the irradiations. These points relate the calculated Hp(10) (depending on the distance and irradiation time) with the average net readings obtained from the TLD-700 group. Equation 1 shows how the net gamma reading ($R\gamma$) of the TLDs is calculated.

$$R\gamma\ [nC] = \frac{\sum_1^n (R_1^7 - R_2^7)}{n} - \frac{\sum_1^n (R_{0,1}^7 - R_{0,2}^7)}{n} \qquad (1)$$

Where:
$(R_1^7 - R_2^7)$: Difference of the first and second reading of the TLDs 700 irradiated [nC].
$(R_{0,1}^7 - R_{0,2}^7)$: Difference of the first and second reading of the TLDs 700 not irradiated [nC].
n: Number of TLDs in the same dose group.

The lower detection limit (LDL) was obtained by taking three times the standard deviation of the measurements of not irradiated detectors ($\varDelta R_{0,7}$) and multiplying by the gamma calibration factor (see Equation 2).

$$LDL\ [mSv] = 3\ .\varDelta R_{0,7}.CF\gamma \qquad (2)$$

3.5 Neutron calibration factor

The neutron calibration factor ($CF\eta$) is also the slope of the fitted line of the calibration curve. However, in this case the net neutron reading ($R\eta$) was calculated as the difference in average net readings of the TLD-600 and the TLD-700 (see Equation 3).

$$R\eta\ [nC] = \left(\frac{\sum_1^n (R_1^6 - R_2^6)}{n} - \frac{\sum_1^n (R_{0,1}^6 - R_{0,2}^6)}{n}\right) - \left(\frac{\sum_1^n (R_1^7 - R_2^7)}{n} - \frac{\sum_1^n (R_{0,1}^7 - R_{0,2}^7)}{n}\right) \qquad (3)$$

Where:
$(R_1^6 - R_2^6)$: Difference of the first and second reading of the TLDs 600 irradiated [nC].
$(R_{0,1}^6 - R_{0,2}^6)$: Difference of the first and second reading of the TLDs 600 not irradiated [nC].
$(R_1^7 - R_2^7)$: Difference of the first and second reading of the TLDs 700 irradiated [nC].
$(R_{0,1}^7 - R_{0,2}^7)$: Difference of the first and second reading of the TLDs 700 not irradiated [nC].

The lower detection limit was obtained by taking three times the mean standard deviation of the background average readings of TLD-600 and TLD-700 ($\Delta R_{0,6-7}$) and multiplying by the neutron calibration factor (see Equation 4).

$$LDL\ [mSv] = 3\ .\Delta R_{0,6-7}.CF\eta \tag{4}$$

4 RESULTS

4.1 Gamma irradiations

Tables 6 and 7 show the average net readings obtained for the dose points measured with the gamma sources. Likewise, the coefficients of variation (CV) and the lower detection limits are shown.

Table 6. Irradiations with the ^{137}Cs source. The dose error (10%) includes the uncertainty of the source, the positioning, the time measurement and the conversion factor of the exposure rate (reported by the manufacturer at 1 meter) to the personal equivalent dose rate. The error of the readings (~ 3%) includes the random error of the different measurements for the same dose (standard deviation of the mean) and the error of the reading equipment (evaluated from the reference reading that it performs). In all cases, they were added in squares since they were random and independent errors.

Distance [cm]	Hp(10) [mSv]	Net Reading TLD-600 [nC]	CV [%]	Net Reading TLD-700 [nC]	CV [%]	LDL [mSv]
50	5.1 ± 0.5	28.6 ± 0.7	2%	30.9 ± 0.9	3%	0.05
50	10 ± 1	56.3 ± 2.2	4%	61 ± 2	3%	
100	1 ± 0.1	5.01 ± 0.16	3%	5.8 ± 0.3	5%	0.03
100	10 ± 1	50.8 ± 1.5	3%	59 ± 3	5%	

Table 7. Irradiations with the ^{60}Co source. Due to the high specific activity of this source, it could be measured in a wider dose range. Verifying the linearity of the dosimetric factor for doses up to 1 Sv. The reported uncertainties were obtained in the same way as explained for the irradiations with ^{137}Cs.

Distance [cm]	Hp(10) [mSv]	Net Reading TLD-600 [nC]	CV [%]	Net Reading TLD-700 [nC]	CV [%]	LDL [mSv]
10	1000 ± 100	5592 ± 125	2%	6252 ± 178	3%	0.03
10	500 ± 50	2750 ± 81	3%	3114 ± 88	3%	
10	10 ± 1	559 ± 17	3%	630 ± 17	3%	
20	5.0 ± 0.5	287 ± 7	2%	314 ± 7	2%	
20	1.0 ± 0.1	56 ± 1	2%	62 ± 2	3%	

4.2 Gamma Calibration Factor

The gamma calibration factors obtained for the TLD-600 and TLD-700 with the two calibration sources used are presented in Table 8. The calibration factor to calculate the Hp(10) is **0.16 ± 0.01 [mSv/nC]**. It was obtained from the weighted average by its uncertainties of the factors calculated for the TLD-700. It was verified that the TLD-600 have the same sensitivity to gamma radiation as the TLD-700. However, the readings of the TLD-700 are used when evaluating the gamma doses in mixed fields.

Table 8. Gamma calibration factors.

TLD	CFγ [^{137}Cs at 0.5 m] [mSv/nC]	CFγ [^{137}Cs at 1 m] [mSv/nC]	CFγ [^{60}Cs at 0.1 and 0.2 m] [mSv/nC]
600	0.18 ± 0.01	0.19 ± 0.01	0.18 ± 0.01
700	0.17 ± 0.01	0.17 ± 0.01	0.16 ± 0.01

4.3 Neutron irradiations

The average net readings obtained for the dose points measured with the neutron source are shown in Table 9. Likewise, the coefficients of variation, the lower detection limits and the neutron calibration factors of each irradiation are shown.

Table 9. Irradiations with the ^{241}Am-Be[9] source at different source-detector distances. The reported uncertainties were obtained in the same way as explained for the gamma irradiations.

Irradiation at 306 cm distance			
Time [h]	Hp(10) [mSv]	TLD-600 – TLD-700 [nC]	CFη [mSv/nC]
3.0	0.03	0.500 ± 0.053	**0.05 ± 0.01**
9.5	0.08	1.622 ± 0.068	LDL [mSv]
30.0	0.3	5.198 ± 0.154	0.01
94.9	0.8	15.734 ± 0.405	
Irradiation at 100 cm distance			
Time [h]	Hp(10) [mSv]	TLD-600 – TLD-700 [nC]	CFη [mSv/nC]
9.5	0.8	4.011 ± 0.695	**0.21 ± 0.01**
30.0	2.4	10.780 ± 0.923	LDL [mSv]
94.9	7.6	34.374 ± 2.751	0.02
Irradiation at 31.6 cm distance			
Time [h]	Hp(10) [mSv]	TLD-600 – TLD-700 [nC]	CFη [mSv/nC]
3.0	2.4	4.816 ± 0.623	**0.49 ± 0.02**
9.5	7.6	16.906 ± 2.460	LDL [mSv]
30.0	24	48.321 ± 5.394	0.08
94.9	75.9	144.937 ± 11.299	

Regarding the irradiations without phantom, Table 10 shows the results obtained to evaluate the dispersion effect of the calibration room.

Table 10. Irradiations without phantom. The reported uncertainties were obtained in the same way as explained for the gamma irradiations.

Distance [m]	Time [h]	Net Reading TLD-600 [nC]	Net Reading TLD-700 [nC]	TLD-600 – TLD-700 [nC]	TLD-600/TLD-700
0.316	3	3.817 ± 0.223	3.677 ± 0.133	0.140 ± 0.260	1.04
0.316	9.5	12.341 ± 0.515	11.946 ± 0.304	0.395 ± 0.598	1.03
1	30	7.443 ± 0.169	4.150 ± 0.195	3.294 ± 0.258	1.79
1	94.9	23.175 ± 0.865	13.149 ± 0.361	10.026 ± 0.937	1.76

5 DISCUSSION & CONCLUSION

From the gamma irradiations results (Table 6, 7 and 8) it can be observed that the gamma sensitivity is the same for the TLD-600 and TLD-700, as expected. This means that the detectors have the same response for the gamma energies used (^{60}Co and ^{137}Cs), even evaluating at different dose rates (source-detector distances). Regarding the repeatability, it can be observed that the coefficients of variation are in all cases less than or equal to 5%. This means that a set of dosemeters responds relatively the same if they are irradiated with the same dose. Furthermore, it can be analysed that within the range of measured doses, reaching up to 1 Sv, the relationship between the dosemeter response and the calculated personal equivalent doses is linear. It can be stated that the gamma calibration factor for Hp(10) has the same value, within the error, in all the cases measured in this work.

On the other hand, the results of the tests carried out with the neutron source (^{241}Am-Be^{9}) highlighted the need to carry out modifications in the albedo dosemeter and in the irradiation room. It was found that for different irradiation distances the calibration factor decreased its value as the source-detector distance increased. Assuming that for irradiations of dosemeters on phantoms at source-detector distances close to one meter, it is not necessary to apply geometric correction factors [9]. This means that the observed effect is due to the fact that when this distance increases, so does the scattering of neutrons on the walls, ceiling and floor of the irradiation room. This could be observed in the irradiations without the phantom (dispersion object). In this way, it was possible to detect the contribution to neutron scattering made by the calibration room. Resulting at the distance of one meter, the ratio between the reading of the TLD-600 and the TLD-700 was approximately 77%. While the standard states that the increase in instrument reading due to room dispersion should be a maximum of 40% [9].

In conclusion, it is necessary to change the irradiation room for one that generates less neutron scattering. In addition, the dosemeter needs to be modified to discriminate the neutrons that are moderated in the room from the albedo neutrons. The new albedo dosemeter will be designed based on the recommendations of ICRU 66 and to meet the ISO 21909. It will consist of two pairs of ^{6}Li-^{7}Li detectors: one pair on the outside of a thermal neutron absorber (e.g., boron loaded plastic), and the other pair on the inside (relative to the body). The difference between the readings of the first pair provides a measure of the incident slow-neutron fluence, and the difference between the readings of the second pair provides a measure of the albedo slow-neutron fluence [1]. The strong albedo energy dependence of this dosemeter is partially compensated by the use of a correction of the calibration factor based on the ratio of the incident neutron component to the albedo neutron component. However, some information on the workplace environment is still necessary [4].

6 REFERENCES

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