Monte Carlo simulation of the MTS-N (LiF:Mg,Ti) relative response in function of the photon energy

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Abstract. We performed MCNPX simulations and experimental measurements to determine the relative energy response of MTS-N detectors. The energy spectra simulated were monoenergetic beams (from 20 keV to 250 keV), X-rays beams (N and RQR), 137Cs and 60Co gamma radiation. Experimental measurements were made with groups of MTS-N detectors irradiated with standardized 137Cs gamma radiation beams and continuous X-ray beams (N and RQR quality). Relative intrinsic efficiencies were calculated combining the MCNPX simulations and experimental measurements. Our results showed a good agreement with other studies using LiF detectors and the same radiation qualities.

Keywords. Monte Carlo modelling; TL dosimeter; Thermoluminescence; Detector response; MCNP

1. Introduction
The knowledge of the energy response of thermoluminescent detectors (TLDs) is of practical importance as they can be used in radiation fields with different energy spectra. In personal or environmental monitoring applications, crystals are often kept in holders behind filters, not only to provide build-up conditions but also to provide spectral information [1]-[2]. Crystals with different energy responses can also be used to determine the effective energy of incident radiation (Tandem method) [3][4].

A large number of TL materials are commercially available. The most popular of them are based on lithium fluoride (LiF). The addition of magnesium (Mg) and titanium (Ti) impurities (activators) to LiF led to the development of the well-known TLD-100, TLD-700 and TLD-600 (manufacturer: Thermo Fisher Scientific Inc.), whose different nomenclatures are due to the use of different isotopic lithium compositions (natural lithium, or enriched with isotopes $^7$Li and $^6$Li, respectively). LiF:Mg,Ti detectors produced by TLD Poland are designated: MTS-N, MTS-6 and MTS-7, also depending on isotopic lithium composition [2].

The response, R, of a dosimeter is given by the ratio, under specified conditions, of the indicated value of the quantity measured by the dosimeter under test by the conventional value of this quantity.
The response can be in terms of air kerma (Ka), ambient dose equivalent (H*(10)), personal dose equivalent (Hp(10)), etc. The response in a given condition can be compared with the response obtained in a reference condition. In this case, it is called relative response, r. For the influence quantity radiation energy, E, the reference condition is the quality S-Cs, which corresponds to the gamma radiation emitted by the $^{137}$Cs radionuclide [2].

The International Organization for Standardization (ISO), in ISO 4037-1 [5], specifies the characteristics and production methods of reference radiation for dosimeter calibration (for radiological protection levels) and dose rate meters, with respect to operational quantities defined by the International Commission on Radiation Units and Measures (ICRU) [6][7]. The International Electrotechnical Commission (IEC), in the standard IEC 61267 [8], deals with the methods of generation of standardized radiation beams used in test conditions typically found in testing laboratories or manufacturer facilities to determine the characteristics of medical diagnostic x-ray equipment.

Several computational codes based on Monte Carlo methods (MMC) have been used in dosimetry applications, including: MCNP (Monte Carlo N-Particle) [9], EGSnrc (Electron-Gamma-Shower) [10], Penelope (Penetration and Energy Loss of Positrons and Electrons) [11], GEANT4 (Geometry and tracking) [12]. The MCNP code was developed by Los Alamos National Laboratory - LANL (USA) and is internationally accepted for neutron, photon and electron transport analysis from the Monte Carlo Method.

The computational model of a thermoluminescent dosimeter has to take into account the transport of radiation through and/or around the thermoluminescent crystal, including backscattered radiation from the surrounding materials, such as: filters, phantoms, holders, etc. There are some papers that show the calculation of the relative response of TLDs using Monte Carlo codes [2],[13]-[15].

The aim of this work was to determine the relative air kerma response of the detector MTS-N, in function of the photon energy, using the Monte Carlo code MCNPX. Our data were compared with the values reported in the literature and measurements performed in the Dosimeter Calibration Laboratory of the CDTN.

2. Materials and Methods

2.1. Monte Carlo simulations

We performed simulations with the computational code based on the Monte Carlo method, MCNPX, version 2.7, with the ENDF/B-VI cross section library (MCPLIB04 and EL03) [16], to determine the air kerma response, as a function of photon energy, of the MTS-N (LiF:Mg,Ti) detector. Circular disk sources of photons, with 1.0 cm diameter, were positioned at 1.0 cm from the surface of the LiF:Mg;Ti pellet of 4.5 mm diameter and 0.9 mm thickness. A thickness of PMMA (polymethyl methacrylate) layer was simulated and, when necessary, added to the crystal (in contact with the front surface), aiming to provide charged particle equilibrium. The crystals were irradiated with monoenergetic beams (from 20 keV to 300 keV), narrow-spectrum series [5], RQR [8]. $^{137}$Cs and $^{60}$Co gamma radiation. For ISO and IEC qualities, we used as source term of radiation the spectrum data measured by Ankerhold (2000) at 1.0 m distance between the focus of the X-ray tube and the spectrometer [17].

Composition and density of the crystal and PMMA were taken from McConn et al. (2011). Composition by weight of the crystal: 73.2415% Li, 26.7585% F (Mg and Ti was neglected), with a density of 2.64 g/cm$^3$. The environment between the crystal and source was treated as dry air [18].

The air kerma response, R, i.e. the Dose to the TLD ($D_{\text{TLD}}$) per unit air kerma ($K_a$), for each energy or quality, E, was calculated performing two simulations for each photon beam. In the first simulation run, we calculated the dose to the crystal using the tally F6:P (MODE P E) and tally *F8:P (MODE P). In the second simulation run, the air kerma was calculated, changing the TLD volume by air and using the same tallies [15][19][20]. Sufficient particle histories were generated so that the tally errors were less than 1%. Kerma and dose values obtained with both tallies and the fluence, obtained with F4:P
tally, were used to calculate mass energy-absorption coefficient \( (\mu_{en}/\rho) \) for air and LiF and compared with data published by [16]. Data obtained with the tally which presented better agreement of the \( (\mu_{en}/\rho) \) with [17] were used to calculate the energy responses.

The energy response, relative to \(^{137}\)Cs gamma radiation, for a beam of energy or quality, \( E \), \( ((r_{Cs}(E))_{sim}) \) has been calculated as shown in equation 1. In simulations, we also calculated the response relative to \(^{60}\)Co \( ((r_{Co}(E))_{sim}) \).

\[
(r_{Cs}(E))_{sim} = \left( \frac{\frac{\partial TL}{\partial A}}{\frac{\partial TL}{\partial A}} \right)_{E} \left( \frac{\partial TL}{\partial A} \right)_{Cs}
\]  

(1)

Uncertainties, for all qualities, were calculated as the root sum square of the statistical components (relative error in the MCNPX) and type B uncertainty components, estimated through the maximum deviation on energy responses, considering a \( \pm 5\% \) variation in the following parameters: (a) densities of the crystal and air; (b) diameter and thickness of the crystal; (c) ratio Li/F and; (d) mean energy of the beams. A rectangular probability distribution was assumed with the range, 2\( a \), equals to the maximum deviation on energy responses, considering the \( \pm 5\% \) variation in the mentioned parameters. Uncertainties are expressed with a coverage factor of one (\( k = 1 \)).

2.2. Experimental

A group of 68 MTS-N detectors, manufactured by RadPRO International GmbH (Wermelskirchen, Germany), former TLD Poland, were selected and individual correction factors were calculated. Prior to their first use, the TL detectors were annealed for 1 h at 400º C, followed by cool-down to ambient temperature and next annealed for 2 h at 100º C. After that, in each irradiation-readout cycle, TLDs were annealed and readout using the RADOS RE2000 hot-gas reader with the software WinTLD Light. The heat treatment for low temperature peaks was 150º C (with no pre-heating and 15 s readout time). The routine readout procedure was applied, i.e. readout temperature of 300º C (1.0 s pre-heat and 11 s readout time). The annealing procedure was applied with the same reading parameter. One set of five unexposed detectors was used for subtracting residual signals and the background influence from the TL readings. Another set of 5 detectors was used for quality control purposes.

Groups of 10 detectors, previously selected, were irradiated with a standardized \(^{137}\)Cs gamma beam and series N (ISO quality S-Cs) [5] and some continuous X ray beams, quality RQR (IEC 61267) [8], established with an industrial X-ray system Pantak/Seifert, model ISOVOLT Titan E. Figure 1 shows experimental setup for \(^{137}\)Cs and X-ray beams irradiations of the TLDs. The irradiations were performed at Laboratório de Calibração de Dosímetros (LCD/CDTN), which is accredited by the National Institute of Metrology, Standardization and Industrial Quality (INMETRO, Brazil). For \(^{137}\)Cs, a buildup layer of 3 mm PMMA was used to provide charged particle equilibrium. For other X-ray qualities (average energy lower than 70 keV), it was not necessary to add any buildup layer. Figure 2 show details of the holders used for TLD irradiations at \(^{137}\)Cs and X-ray beams. TL detectors were positioned at 1.5 m distance from the gamma irradiator and 1.0 m distance from the X-ray system. Constant air kerma values of 2.0 mGy were adopted in all irradiations.
Figure 1. Experimental setup for $^{137}\text{Cs}$ (a) and X-ray beams (b) irradiations of the TLDs.

The measured air kerma energy response, normalized to $^{137}\text{Cs}$ gamma radiation $(r_{Cs}(E))_{meas}$, was calculated accordingly equation 2, where $TL(E)$ and $TL(Cs)$ are the thermoluminescent (TL) readings for a beam of energy or quality, E or S-Cs.

$$
(r_{Cs}(E))_{meas} = \frac{(TL(E))}{K_{ref}(E)} \frac{(TL(Cs))}{K_{a(Cs)}}
$$

(2)

Uncertainties in measured responses, for all qualities, were calculated combining uncertainty in dosimetry of the beams, repeatability of TLD readings and uncertainties on individual correction factors. Uncertainties are also expressed with a coverage factor of one (k = 1).

Figure 2. Details of the holders used for TLD irradiations at $^{137}\text{Cs}$ (a) and X-ray beams (b).
2.3. Relative intrinsic efficiency

The relative intrinsic efficiencies, $\eta_i(E)$, were calculated combining the MCNPX simulations and experimental measurements, using the equation 3 [19][20]. Results were compared with published data. Uncertainties in $\eta_i(E)$ were calculated combining uncertainties in both responses

$$\eta_i(E) = \frac{(r_{cs}(E))_{\text{meas}}}{(r_{cs}(E))_{\text{sim}}}$$

(3)

3. Results and Discussions

Table 1 and 2 show, respectively, the percentage deviations of the energy absorption coefficients simulated with and without build-up layer, using tallies F6:P (MODE P E) and *F8:P (MODE P), were compared to the values reported in the literature [16].

Table 1 shows that for tally F6:P (MODE P E), the percentage deviations between the simulations performed with and without build-up and the values reported in the literature were less than 1.5%. The simulations performed without build-up showed the best agreement with the values reported in the literature. The interquartile ranges (I IQs), given by the differences between the values of the 3rd and 1st quartile, were less than 0.66% for both cases. For air, the I IQs were less than 0.23%. Table 1 shows that for simulations performed without build-up, the largest percentage deviation for LiF, 0.73%, was for the 300 keV energy and that only for the 150 keV and 200 keV and $^{137}$Cs and $^{60}$Co, the percentage deviations were greater than 0.50%. For air, the largest percentage deviation was for $^{137}$Cs energy, 0.94%. For the other energies, the percentage deviations were less than 0.51%. Table 1 shows that for the simulations performed with build-up, the highest percentage deviation for LiF, 1.45%, was for the $^{60}$Co energy and that, for the 150 keV, 200 keV, and 300 keV energies, the deviations were greater than 0.78%. For air, the highest percentage deviation was also for $^{60}$Co, 0.67%. For the other energies, the percentage deviations were less than 0.38%.

Table 1. Percentage differences between the values obtained with the tally F6:P (MODE P E), with and without a build-up layer, and the values reported in the literature [16].

| Energy (keV) | ($\mu_{en}/\rho$)$_{LiF}$ | LiF - F6:P (MODE P E) | LiF | Ar | LiF | Ar |
|-------------|-----------------------------|------------------------|-----|----|-----|----|
|             | with Build-up |                      |     |    |     |    |
| 20          | 0.20           | 0.05                   | 0.20| 0.05|
| 30          | 0.17           | 0.11                   | 0.18| 0.11|
| 40          | 0.48           | 0.01                   | 0.14| 0.00|
| 50          | 0.46           | 0.02                   | 0.47| 0.03|
| 60          | 0.45           | 0.02                   | 0.46| 0.03|
| 80          | 0.01           | 0.02                   | 0.01| 0.00|
| 100         | 0.30           | 0.02                   | 0.34| 0.02|
| 150         | 0.67           | 0.08                   | 0.79| 0.20|
| 200         | 0.67           | 0.03                   | 0.86| 0.17|
| 250         | 0.22           | 0.50                   | 0.49| 0.22|
| 300         | 0.73           | 0.03                   | 1.07| 0.37|
| $^{137}$Cs  | 0.52           | 0.94                   | 0.16| 0.26|
| $^{60}$Co   | 0.55           | 0.22                   | 1.45| 0.67|
Table 2. Percentage differences between the values obtained with the tally *F8:P (MODE P), with and without a build-up layer, and the values reported in the literature [16].

| Energy (keV) | (\frac{\mu_{en}}{\rho})_{LiF} LiF - tally *F8:P (MODE P) with Build-up | LiF | Ar | LiF | Ar |
|--------------|-------------------------------------------------------------------|-----|----|-----|----|
| 20           | 0.19                                                               | 0.20| 0.08| 0.08|
| 30           | 0.40                                                               | 0.62| 0.31| 1.22|
| 40           | 0.56                                                               | 0.21| 0.47| 2.15|
| 50           | 0.53                                                               | 0.64| 0.42| 1.04|
| 60           | 0.42                                                               | 0.88| 0.29| 0.22|
| 80           | 0.06                                                               | 1.19| 0.08| 0.45|
| 100          | 0.29                                                               | 1.06| 0.35| 0.62|
| 150          | 0.61                                                               | 0.21| 0.77| 1.19|
| 200          | 0.66                                                               | 0.28| 0.89| 1.20|
| 250          | 0.21                                                               | 0.25| 0.55| 1.19|
| 300          | 0.74                                                               | 0.92| 1.15| 1.55|
| 137Cs        | 0.40                                                               | 0.60| 0.29| 1.70|
| 60Co         | 0.88                                                               | 0.20| 1.75| 0.23|

Table 2 shows that for tally *F8:P (MODE P), the percentage deviations between the simulations performed with and without build-up and the values reported in the literature were less than 2.2%. Again, the simulations performed without build-up layer showed the best agreement with the values reported in the literature. Interquartile ranges were less than 0.55% for LiF and less than 1.05% for air. Table 2 shows that for simulations carried out without build-up, the largest percentage deviation for LiF, 0.88%, was for $^{60}$Co energy. For air, the highest percentage deviation was for the energy of 80 keV, 1.19%, and 1.06% for 100 keV. For the other energies, the percentage deviations were less than 0.93%. Table 2 shows that for the simulations performed with build-up, the highest percentage deviation for LiF, 1.75%, was for the $^{60}$Co energy and that for the 300 keV energy was 1.15%. For air, the highest percentage deviation was also for 40 keV, 2.15%. For the other energies, the percentage deviations were less than 1.7%.

We concluded that the tally F6:P (both in MODE P E and MODE P) without build-up, presented calculated values closer to the literature for the entire energy range, both for air, as for LiF. For the aforementioned tally, the values found for the two modes of transport show agreement within the statistical uncertainties. Thus, in the present work, the tally F6:P (MODE P E) was adopted to determine the values of \(r(E)\). Table 3 shows the simulated and measured air kerma relative responses to $^{137}$Cs and simulated energy responses relative to $^{60}$Co with the corresponding uncertainties for beam qualities N and RQR.
Table 3. Simulated energy responses relative to $^{137}$Cs ($r_{Cs}(E)_{sim}$) and $^{60}$Co ($r_{Co}(E)_{sim}$) and measured relative responses ($r_{Cs}(E)_{meas}$) for qualities N and RQR.

| Radiation Quality | Energy (keV) | $r_{Cs}(E)_{sim}$ | U | $r_{Co}(E)_{sim}$ | U | $r_{Cs}(E)_{meas}$ | U |
|------------------|-------------|------------------|---|------------------|---|-------------------|---|
| $^{137}$Cs       | 661.70      | 1.000            |   | 1.001            | 0.017 | 1.000            |   |
| $^{60}$Co        | 1253.00     | 0.999            | 0.017 | 1.000            |   |
| N-30             | 24.62       | 1.252            | 0.061 | 1.253            | 0.060 | 1.358            | 0.036 |
| N-40             | 33.27       | 1.275            | 0.017 | 1.276            | 0.014 | 1.350            | 0.038 |
| N-60             | 47.88       | 1.231            | 0.026 | 1.232            | 0.024 | 1.282            | 0.035 |
| N-80             | 65.19       | 1.141            | 0.022 | 1.142            | 0.02 | 1.177            | 0.032 |
| N-100            | 83.26       | 1.078            | 0.017 | 1.079            | 0.015 | 1.114            | 0.032 |
| N-150            | 118.2       | 1.030            | 0.021 | 1.031            | 0.019 | 1.072            | 0.029 |
| N-200            | 164.8       | 1.012            | 0.022 | 1.013            | 0.021 |               |   |
| N-250            | 207.3       | 1.007            | 0.022 | 1.008            | 0.021 |               |   |
| N-300            | 248.4       | 1.004            | 0.021 | 1.005            | 0.020 |               |   |
| RQR-2            | 28.25       | 1.252            | 0.081 | 1.253            | 0.080 |               |   |
| RQR-3            | 32.35       | 1.254            | 0.017 | 1.255            | 0.014 | 1.326            | 0.043 |
| RQR-4            | 36.01       | 1.252            | 0.048 | 1.253            | 0.048 |               |   |
| RQR-5            | 39.36       | 1.247            | 0.036 | 1.248            | 0.035 | 1.302            | 0.042 |
| RQR-6            | 42.78       | 1.238            | 0.031 | 1.239            | 0.029 |               |   |
| RQR-7            | 45.96       | 1.234            | 0.027 | 1.235            | 0.026 | 1.244            | 0.040 |
| RQR-8            | 48.84       | 1.224            | 0.025 | 1.225            | 0.024 |               |   |
| RQR-9            | 53.87       | 1.220            | 0.025 | 1.221            | 0.023 | 1.230            | 0.041 |
| RQR-10           | 60.97       | 1.188            | 0.025 | 1.189            | 0.024 |               |   |

Air kerma responses relative to $^{137}$Cs and $^{60}$Co were lower than 1.29. The lowest calculated energy responses were 0.999 and 1.001 for $^{137}$Cs and $^{60}$Co, respectively. The divergences between the responses for $^{137}$Cs and $^{60}$Co are small (minor than 0.1%). The uncertainties in the relative responses ranged from 1.1% to 8.4%. Measured air kerma responses relative to $^{137}$Cs were lower than 1.36. The uncertainties in the relative responses ranged from 2.2% to 3.3%.

Figure 3 shows a comparison of the data obtained from the simulation of X-ray beams (qualities N), $^{137}$Cs and $^{60}$Co, with the data from Davis et al. [2]. Figure 4 shows a comparison of the relative intrinsic efficiency ($\eta_i(E)$) obtained in this work and data published elsewhere [2][21][22].
Figure 3. Comparison of this work with Davis et al [2] for air kerma response for X-ray beams, $^{137}$Cs, $^{60}$Co, obtained from simulation.

Figure 4. Comparison of the relative intrinsic efficiency ($\eta_i(E)$) obtained in this work and data published elsewhere [2][22][23].
In Figure 3 we can observe that, in general, our data agree well (within 6%) with data of Davis et al. [2]. In our paper the relative responses are systematically smaller than those reported by Davis et al. [2], which used the code EGSnrc to perform the simulations.

Figure 4 shows a relatively good agreement of the intrinsic efficiency obtained in this work with the published data. The maximum deviation was lower than 8.6%. Data published by Davis et al. [2] presented the better agreement (within 3.0%).

4. Conclusions
The relative air kerma response of the detector MTS-N, in function of the photon energy, was calculated using the Monte Carlo code MCNPX and compared with experimental measurements and other published data. Experimental measurements, performed in the Dosimeter Calibration Laboratory of the CDTN, were made with groups of MTS-N detectors irradiated with standardized $^{137}$Cs gamma radiation beams and continuous X-ray beams (N and RQR quality). Relative intrinsic efficiencies were calculated combining the MCNPX simulations and experimental measurements and also compared with published data. Our results showed a good agreement with other studies using LiF detectors and the same radiation qualities.

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