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DOSIMETRY AT THE PORTUGUESE RESEARCH REACTOR USING THERMOLUMINESCENCE MEASUREMENTS AND MONTE CARLO CALCULATIONS

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This work presents an extensive study on Monte Carlo radiation transport simulation and thermoluminescent (TL) dosimetry for characterising mixed radiation fields (neutrons and photons) occurring in nuclear reactors. The feasibility of these methods is investigated for radiation fields at various locations of the Portuguese Research Reactor (RPI). The performance of the approaches developed in this work is compared with dosimetric techniques already existing at RPI. The Monte Carlo MCNP-4C code was used for a detailed modelling of the reactor core, the fast neutron beam and the thermal column of RPI. Simulations using these models allow to reproduce the energy and spatial distributions of the neutron field very well (agreement better than 80%). In the case of the photon field, the agreement improves with decreasing intensity of the component related to fission and activation products. ^{7}LiF:Mg,Ti, ^{7}LiF:Mg,Cu,P and Al_2O_3:Mg,Y TL detectors (TLDs) with low neutron sensitivity are able to determine photon dose and dose profiles with high spatial resolution. On the other hand, ^{nat}LiF:Mg,Ti TLDs with increased neutron sensitivity show a remarkable loss of sensitivity and a high supralinearity in high-intensity fields hampering their application at nuclear reactors.

INTRODUCTION

Reactor dosimetry presents great difficulties arising from the high-intensity and mixed radiation fields. It is generally required to discriminate various particles (especially neutrons and photons), at dose levels for which most dosemeters will be saturated or damaged. Monte Carlo (MC) codes for simulating radiation transport are an invaluable tool to calculate such radiation fields and are increasingly used as a complement to experimental techniques(1).

The operation of nuclear reactors requires the usage of numerous, small and cheap dosemeters to monitor each irradiation, thereby, justifying the interest in the expansion of thermoluminescent (TL) dosimetry from the conventional low-dose photon measurements to the challenging aspects of reactor dosimetry(2,3). Paired TL detectors (TLDs) having different neutron sensitivities—ideally with one of them insensitive to neutrons(4)—are occasionally used for measuring the photon and neutron doses in such mixed radiation fields(5,6). The accuracy of TL measurements is frequently assessed via a comparison between measured and calculated neutron and photon doses.

This work presents an extensive study on MC radiation transport simulation and TLD for characterising mixed radiation fields occurring in nuclear reactors. The feasibility of these methods is investigated for radiation fields at various locations of the Portuguese Research Reactor (RPI) having distinct dosimetric characteristics—a fast neutron beam with a significant photon dose component(7,8) and a thermal neutron beam having a low photon dose contribution(9). The performance of the approaches developed in this work is compared with the techniques already existing at RPI, namely the usage of activation foils and ionisation chambers.

MATERIALS AND METHODS

The Monte Carlo MCNP-4C(10) code was used for a detailed modelling of the fast neutron beam and the thermal column of RPI, employing a source term previously determined using an MCNP model of the reactor core(11). Beam descriptions and models are reported elsewhere(8,9). For converting spectra of neutron fluence and photon energy fluence in neutron and photon doses, tabled point-wise kerma factors and energy absorption coefficients were used(12,13). Calculated neutron spectra were further adjusted via the multiple-foil activation method(14). Details on the experimental procedure may be found in the papers by Fernandes *et al.*(8,9).

Concerning the photon spectra, MCNP allows to discriminate the prompt components from reactor background (fission and activation reactions in the pool) and neutron interactions with beam materials. Since MCNP does not consider photons emitted in product decay, the reactor history-dependent

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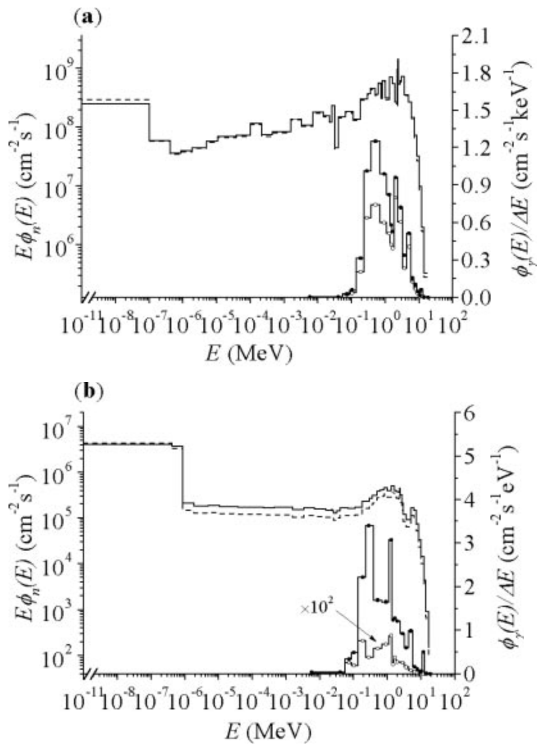

Figure 1. Spectra of neutron fluence rate per unit lethargy and calculated spectra of photon fluence rate per unit energy in the (a) fast and (b) thermal neutron beams (dashed line (- - -) calculated neutron spectra; straight line (—) adjusted neutron spectra; lines joining open circles: photon reactor background; lines joining closed circles: total photon) in the irradiation facilities.

component of photon dose is not simulated. A neutron-insensitive Mg ionisation chamber flushed with argon[15] (type M2 Exradin, Lisle, IL) was used for total photon dose measurements. The chamber was calibrated at the Metrology Laboratory of Ionising Radiations and Radioactivity, Nuclear and Technological Institute.

Paired $^{nat/7}$LiF:Mg,Ti TLDs from different producers (TLD-100/700, Harshaw, OH and GR-100/107, Solid Dosimetric Detector and Methods Laboratory, SDDML, People's Republic of China) were used as neutron and photon detectors. In addition, TLD-700H (^{7}LiF:Mg,Cu,P, Harshaw) and Al_2O_3:Mg,Y[16,17] (D-2 and D-3, Institute of Isotope and Surface Chemistry, IKI, Hungary) were applied for photon dose measurements. D-2 and D-3 aluminium oxide TLDs contain different activator concentrations, which result in different sensitivity ranges (10^{-1}–10^3 Gy for D-2 and 10^{-4}–10 Gy for D-3). The applicability of this material to high dose measurements may be expanded further by considering a high temperature peak (~450°C) rather than the

Table 1. Main dosimetric parameters of the irradiation facilities.

	Fast neutron beam	Thermal column
Neutron fluence rate (cm^{-2} s^{-1})	1.0×10^9 (E > 1 MeV)	4.0×10^7 (E < 0.5 eV)
Average neutron kerma factor (Gy cm^2)		
In air	2.6×10^{-12}	6.7×10^{-12}
In water	1.5×10^{-12}	6.7×10^{-13}
Average photon energy absorption coefficient (cm^2 kg^{-1})		
In air	22	21
In water	24	23
Neutron dose rate in air (Gy h^{-1})	30	1.2
Photon dose rate in air (Gy h^{-1})	50	0.1

Data at 1 MW reactor power. Data concerning the neutron field refer to the adjusted spectrum. The photon dose rate was measured using an ion chamber and the average photon energy absorption coefficients were determined in the calculated spectra

main dosimetric one (~250°C). The TLDs were evaluated with a Harshaw 3500 reader. Detector dimensions, usage and evaluation conditions are described elsewhere[9].

RESULTS AND DISCUSSION

The calculated and adjusted neutron spectra and the calculated photon spectra in the fast and thermal neutron beams are shown in Figure 1. The agreement between calculated and measured foil responses was generally better than 90%, except for an overestimation of the thermal component of 20 and 40% for the fast and thermal neutron beams, respectively. After the adjustment, the agreement improved to 95%. The most important dosimetric parameters in each beam, at full reactor power (1 MW) are summarised in Table 1.

Important dosimetric properties for the application of TLDs in nuclear reactors were investigated. The glow curves after an irradiation in the thermal column are shown in Figure 2. In the case of LiF:Mg,Ti, TLD-100 and TLD-700 curves are compared to demonstrate the increase of high temperature peaks after the mixed-field irradiation. Furthermore, the glow curve of TLD-100 irradiated with photons (1 Gy ^{60}Co) is presented. In contrast, the glow curves of TLD-700H and D-3 were not modified by the mixed-field irradiation.

Table 2 presents the relative sensitivity of the various TLDs (normalised to the low temperature peaks

Table 2. Relative sensitivity[a] of the dosemeters investigated.

	TLD-100		TLD-700		GR-100		GR-107		TLD-700H	D-3
	LT	HT	LT	HT	LT	HT	LT	HT		
γ	1	0.05	1.2	0.05	1.6	0.04	1.3	0.05	7.7	0.3
n+γ	1	5.5	0.018	0.17	0.57	4.1	0.0065	0.018	0.0050	0.0047

γ: irradiations with 1 Gy ^{60}Co photons; n + γ: irradiations in a mixed field of 3×10^{11} cm^{-2} thermal neutron fluence and 100 mGy photon air kerma; LT: low temperature peaks; HT: high temperature peaks
[a]Sensitivity normalised to the low temperature peaks of TLD-100

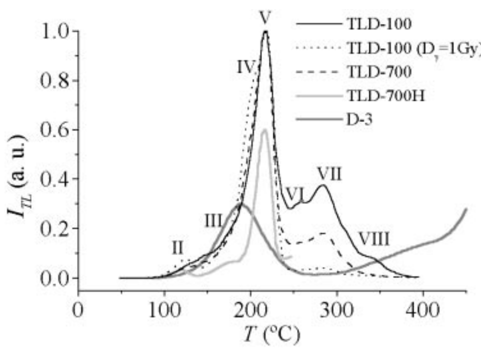

Figure 2. Glow curves (not to scale) of TL materials irradiated in a mixed radiation field of thermal neutrons (thermal neutron fluence: 3×10^{11} cm^{-2}) and photons (air kerma: 200 mGy). The glow curve peaks of LiF:Mg,Ti are identified by roman numerals.

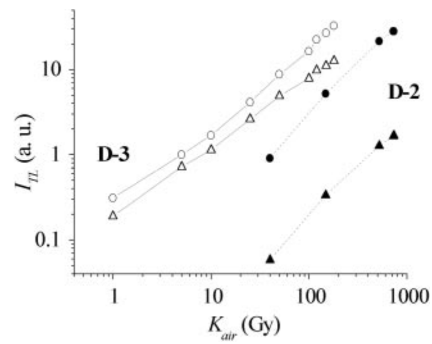

Figure 3. Relative TL intensity of Al_2O_3:Mg,Y detectors D-2 and D-3 as a function of photon kerma in air (circles, low temperature peak; triangles, high temperature peak).

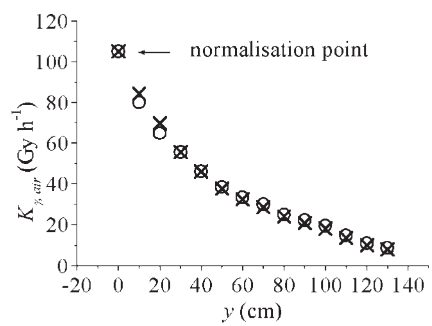

Figure 4. Axial photon dose profile in the fast neutron beam (open circle, calculated values; cross, measured values). The measurements were performed using D-3 TLDs and the calculated values were normalised to a measurement point.

of TLD-100), irradiated in a pure photon field and in the mixed field of the thermal column. The measurements indicate that the SDDML detectors have lower relative sensitivities to thermal neutrons compared with Harshaw's. Therefore GR-107 should be used for measuring photon doses in mixed fields, while TLD-100 should be chosen for detecting thermal neutrons at low-dose levels (e.g. albedo neutrons in individual monitoring). It is worth noting that the high temperature ratios of the various detectors are different and have the lowest value for GR-107. TLD-700H and Al_2O_3:Mg,Y are the detectors with the lowest thermal neutron sensitivity and, therefore, should be preferred for measuring photon doses in thermal neutron beams in adequate dose levels.

When fast neutrons are involved, the fast neutron sensitivity of Al_2O_3:Mg,Y detectors(18) may influence its applicability. For example, the relative neutron sensitivity of D-3 TLDs irradiated in the fast neutron beam is 8% higher than that of GR-107, in contrast to a 27% lower sensitivity to thermal neutrons. The use of aluminium oxide is, nevertheless, justified in case high dose levels are involved (Figure 3). Figure 4 shows the measured (using D-3) and calculated photon dose profiles in the fast neutron beam. An agreement better than 15% was obtained, which demonstrates the special ability of low neutron sensitivity TLDs for photon dose profile measurements in mixed fields.

Figure 5 presents the photon and neutron dose responses of TLD-100, normalised to the lowest-dose irradiation. In the dose ranges considered, the high temperature peaks (peaks VI–VIII) exhibited a supralinear response both for photon and neutron irradiations. The low temperature peaks

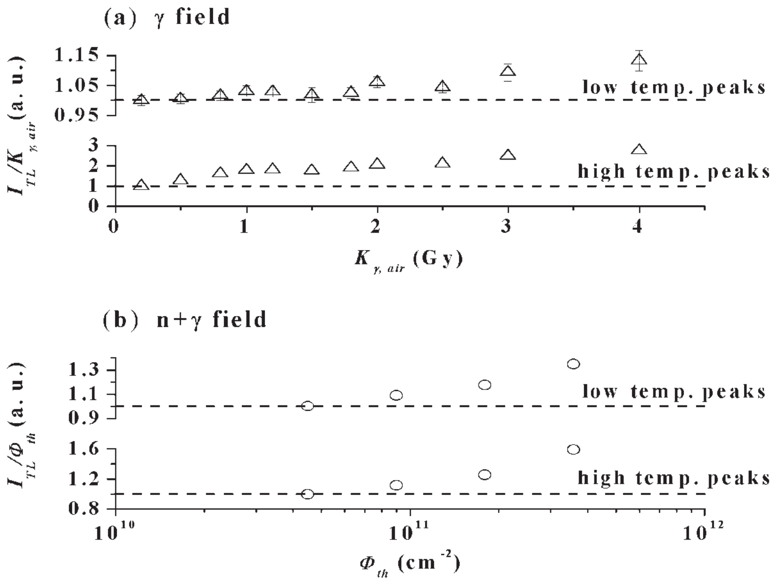

Figure 5. Sensitivity of TLD-100 as a function of photon kerma in air and thermal neutron fluence. Except for the low temperature peaks in the pure photon field, the uncertainties were omitted since they are smaller than the symbol size.

(peaks III–V) show the normal linear response for photon doses up to ~2 Gy, but presented a strongly supralinear response in the mixed-field irradiations. No modification was observed in the dose response of materials with low sensitivity to thermal neutrons.

The discrepancy between photon doses measured using TLDs (TLD-700H, GR-107 and D-3) and ionisation chambers is within 10%, therefore the photon dose measurements are consistent. When the calculated photon doses are normalised to a measurement of neutron fluence, the ratio between calculated and measured photon doses is 50 and 110% in the fast neutron beam and the thermal column, respectively. According to calculations, the contribution from the reactor background to the total photon dose is >60% in the fast neutron beam, while being negligible (<1%) in the thermal column. These results point to the need of a simulation code that can consider the reactor background, in case its contribution to the total photon dose becomes important.

CONCLUSIONS

Simulations of reactor-based neutron beams using the MCNP code allow to reproduce neutron spectra very well (agreement better than 20%). In the case of the photon field, the agreement improves with decreasing intensity of the component related to fission and activation products.

TLDs with low neutron sensitivity (^{7}LiF:Mg,Ti, ^{7}LiF:Mg,Cu,P and Al_2O_3:Mg,Y) are able to determine photon dose and dose profiles in mixed fields with high spatial resolution. Among the materials investigated, ^{7}LiF:Mg,Cu,P and Al_2O_3:Mg,Y are those with the lowest (and similar) relative thermal neutron sensitivities and will, therefore, be the most suitable for photon dose measurements in adequate dose ranges.

On the other hand, TLDs with elevated neutron sensitivity (LiF:Mg,Ti) exhibit a supralinear response and show a remarkable loss of sensitivity in high-intensity neutron fields[19,20]. Careful (and time-consuming) calibration procedures may partly overcome these limitations[9]. Nevertheless, the application of TLDs for neutron dosimetry at high-intensity reactor neutron beams is precluded relatively to the standard activation procedure.

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