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Improvement of ESR dosimetry for thermal neutron beams through the addition of gadolinium

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Abstract

In this paper, the addition of gadolinium is proposed as a useful tool to enhance the electron spin resonance (ESR) sensitivity of organic compounds to thermal neutrons. The target of this work is the detection, through the ESR technique, of the thermal neutron fluence in a mixed field of photons and neutrons. Gadolinium was chosen because it has a very high capture cross section to thermal neutrons; its nuclear reaction with thermal neutrons induces complex inner shell transitions that generate, besides other particles, Auger electrons, which in turn release their energy in the neighborhood (only several nanometers) of the place of reaction. Gadolinium was added to two organic molecules: alanine and ammonium tartrate. The main result obtained was a greater neutron sensitivity for dosimeters with gadolinium than for those without gadolinium for both organic molecules used. Since a dosimeter pair is required to discriminate between the two components of a mixed field, we studied the response of each dosimeter pair irradiated in a mixed field. Through a blind test we verified the usefulness of this dosimetric system and we obtained an estimate of the fluence in the mixed field with a relative uncertainty of 3%, when the pair composed of an alanine dosimeter and a dosimeter with alanine and gadolinium is used.

1. Introduction

Neutron applications to tumor radiotherapy are of great clinical and research interest since the discovery of the neutron by Chadwick in 1962. In fact, fast neutron radiotherapy was

first used in 1938 in the national laboratories of Lawrence Berkeley (Stone 1940). Nowadays, neutron radiotherapy is adopted for particular non-operable tumors and in the neoplastic stage.

Recently, special attention was paid to thermal neutrons since these particles permit us to selectively hit the treatment volume (Hawthorne 1998, Nigg *et al* 1997). Neutron capture therapy (NCT), first proposed by Locher (1936), is based on the ability of marking tumor cells with molecules containing nuclei with a high cross section for thermal neutrons. After the injection of these compounds, the exposure to a thermal neutron beam induces nuclear reactions that produce particles with high linear energy transfer (LET), either heavy charged particles (alpha, protons and/or light ions) or electrons and/or x-rays with very low energy. These particles release their energy locally, inside the 'marked' tumor cells that are killed, whereas the dose absorbed by the healthy tissues is minimized.

Nowadays, the most used nucleus for NCT is ^{10}B, because of both its high neutron capture cross section (about 3800 barn) and the high LET of the secondary particles produced (^{7}Li and alpha). Recently, the ^{157}Gd nucleus was also utilized in combined therapy (Oyewumi and Mumper 2003, Culbertson and Jevremovic 2003). This nucleus has a capture cross section to thermal neutrons (255 000 barn) greater (more than 60 times) than that of ^{10}B. The nuclear reaction with neutrons induces complex inner shell transitions that involve the emission of prompt γ photons, of internal conversion electrons and of Auger electrons, together with soft x-ray and photon emissions. The Auger electrons are the high LET particles which release energy locally in tumor cells at a distance of a few nanometers from the reaction place (Salt *et al* 2004).

The efficacy of the radiotherapy with thermal neutrons (Nigg *et al* 1997, Alvarez-Estrada and Calvo 2004) strongly depends on the injection of high concentrations of ^{10}B or ^{157}Gd inside the tumor cells, and on the choice of the neutron fluence suitable for providing the prescribed dose. The dosimetry in thermal neutron therapy is complicated by the presence of a photon component which also releases dose to tissues, often in regions far from the tumor's place.

Since any dosimeter for neutrons is also sensitive to photons, neutron dosimetry in mixed beams is usually performed through two detectors, whose sensitivities to photons and to neutrons are known (ICRU 1977).

Electron spin resonance (ESR) dosimetry has been studied since the 1960s (Bradshaw *et al* 1962, Rotblat and Simmons 1963), and in the last 20 years ESR dosimetry has made great progress in various fields such as radio-sterilization and radiotherapy. The ESR technique is based on the detection of free radicals produced by ionizing radiation in organic and/or inorganic molecules. The most used molecule for ESR dosimetry is alanine which constitutes an international standard (recognized by the International Atomic Energy Agency). This amino acid is nowadays widely adopted for photon and electron dosimetry.

Some experimental studies have been realized to investigate the applicability of alanine dosimeters to high stopping power radiation such as fast neutrons, protons and heavy charged particles (Hansen *et al* 1987, Katsumura *et al* 1985, Simmons 1987).

Along with the NCT development and with the use of thermal neutrons for radiotherapeutic purposes, ESR dosimetry was applied to measure dose and fluence of these radiation fields. A limiting feature of alanine for measuring neutron fluence must be underlined, i.e. this molecule ($C_3H_7O_2N$) is constituted by nuclei with a relatively low neutron capture cross section; therefore, the probability of interaction with thermal neutrons is low and so the energy released inside the dosimeter is small. Consequently, the number of free radicals produced is very small. Thus, it is very difficult to measure quantitatively the neutron beam properties, since the ESR signal of irradiated samples is very weak.

Table 1. Percentage in mass of the elements present in the dosimeters.

	Element					
Dosimeter	H	C	N	O	Others	ρ (g × cm^{-3})
Alanine	8.24	43.03	14.79	33.90	0.04	1.04 ± 0.06
$B(OH)_3$–alanine	7.11	27.57	8.77	50.24	6.31	0.88 ± 0.05
Gd_2O_3–alanine	4.54	24.02	7.39	23.31	40.74	1.35 ± 0.08
Ammonium tartrate	6.98	29.53	14.30	49.15	0.04	1.20 ± 0.07
Gd_2O_3–ammonium tartrate	3.91	17.27	7.15	30.93	40.74	1.31 ± 0.08

A method for improving the interactions with thermal neutrons is the addition of nuclei with a high neutron capture cross section that increase the interaction probability with neutrons. In particular, the sensitivity of alanine to thermal neutrons was increased by the addition of ^{10}B (Bartolotta *et al* 2004, Galindo and Ureña-Nuñez 1993), which after nuclear reaction with thermal neutrons produces heavy charged particles (Li ions and alpha particles) which release a great amount of energy inside the dosimeter.

In the last few years, substances different from alanine were studied in ESR dosimetry (Bartolotta *et al* 1999, Ikeya *et al* 2000, Lund *et al* 2002). Among these substances, ammonium tartrate ($C_4H_{12}O_6N_2$) is a very promising molecule for the dosimetry of photon and electron beams (Olsson *et al* 2000, Bartolotta *et al* 2001, Yordanov and Gancheva 2004, Marrale *et al* 2006); however, it also consists of nuclei with a relatively low neutron capture cross section. Consequently, the addition of nuclei with a higher cross section is needed to increase the ammonium tartrate dosimeters' sensitivity to thermal neutrons.

The innovative feature of this work is the choice of gadolinium as additive for maximizing the interaction probability with thermal neutrons. We analyzed the ESR response of five blends (alanine, alanine with ^{10}B-boric acid, alanine with gadolinium oxide, ammonium tartrate and ammonium tartrate with gadolinium oxide) exposed to a mixed field of radiation (neutrons and photons).

2. Materials and methods

2.1. Dosimeters' preparation

Solid state pellets for ESR dosimetry were made by using a blend of L-α-alanine (Fluka, Buchs, Switzerland), ammonium tartrate (Carlo Erba, Italy), gadolinium-oxide (Gd_2O_3, Aldrich Chem. Co) and boric acid (BO_3H_3, Aldrich Chem. Co) enriched with ^{10}B (99%) in known proportions by mass. The gadolinium oxide is composed of the various gadolinium isotopes in their natural isotopic composition. In particular, the atomic fractions of the isotopes ^{157}Gd and ^{155}Gd, which have the highest cross sections for thermal neutron capture (respectively ∼250 000 and ∼75 000 barn), are 0.156 and 0.148, respectively.

The pellets were realized according to a procedure previously optimized (Bartolotta *et al* 1999) by pressing a blend of 94% of the appropriate active material (either $B(OH)_3$-alanine or Gd_2O_3–alanine or pure alanine or Gd_2O_3–ammonium tartrate or pure ammonium tartrate), 5% of polyethylene (Polysciences, MW = 700) as binder and 1% of magnesium stearate (Carlo Erba, Milano, Italy) as lubricant. The pellets of alanine (or ammonium tartrate) with Gd_2O_3 (or $B(OH)_3$) were realized by pressing a blend in which alanine (or ammonium tartrate) and Gd_2O_3 (or $B(OH)_3$) were present in equal proportion by mass (47%). Table 1 reports the percentages in mass of the elements present in the dosimeters and the mean mass density

Table 2. Values of neutron fluence and photon dose provided by the irradiation facility (TAPIRO reactor, ENEA, Casaccia).

Fluence (10^{12} cm^{-2})	Gamma dose (Gy)
2.40	0.205
3.92	0.325
19.2	1.64
39.2	3.25

values ρ (± 1 S.D.) obtained as the ratio of the measured mass to the measured volume of a representative set of the dosimeter batch.

Pellets of about 4 mm in diameter and 2.2 mm, 1.7 mm, 2.5 mm, 2.5 mm and 1.9 mm in thickness for ammonium tartrate, Gd_2O_3–ammonium tartrate, alanine, $B(OH)_3$–alanine and Gd_2O_3–alanine, respectively, were obtained.

2.2. γ irradiations

Pellets were irradiated at room temperature in a Perspex phantom with the 1.25 MeV photons of a ^{60}Co source (Alcyon II, General Electric, France) used for radiotherapy treatments at the Radiotherapy Department of the Oncology Hospital 'M.Ascoli' in Palermo. Irradiations were performed in a Perspex phantom $30 \times 30 \times 30$ cm^3, with a field size of 15×15 cm^2, at the water equivalent depth of 5 cm, and with a source–detector distance of 80 cm. The dose rate at the effective dosimeter location was evaluated with the ENEA (Ente per le nuove tecnologie, l'energia e l'ambiente, Italy) secondary standard ionization chamber, with an overall uncertainty of 2% (95% confidence level).

2.3. Thermal neutron irradiations

Thermal neutron irradiations were carried out in the TAPIRO reactor, at the ENEA Casaccia Center, near Rome. Since TAPIRO is a source of fast neutrons, a suitable removable beam-shaping assembly (BSA) is needed for obtaining a beam of thermal neutrons. The facility used is HYTHOR (HYbrid Thermal spectrum sHifter TapirO Reactor) which is based on a hybrid Pb-CF2-RGGraphite neutron spectrum shifter configuration (Esposito *et al* 2007). It provides an eminently thermal neutron beam (about 92% of the total neutron flux) inside the irradiation cavity with a very low gamma background. More details are given in Esposito *et al* (2007).

Cylindrical dosimeter holders (24 mm diameter, 7 mm height) were made with Teflon and utilized for irradiations. Four dosimeters were placed in suitable small cavities realized inside the holders, and were irradiated inside the thermal column of the TAPIRO nuclear reactor at the neutron flux and gamma dose reported in table 2.

Since the neutron flux used consists mainly of thermal neutrons, conventionally in the following text we will refer to it as a thermal neutron flux.

2.4. ESR measurements

The ESR measurements were taken with a Bruker ECS 106 spectrometer equipped with a TE_{102} rectangular cavity and operating in the X band at approximately 9.70 GHz. A quartz holder and quartz spacers were used to place the dosimeters inside the cavity in the

position, where the strongest signal was obtained. The first derivative of the ESR absorption spectrum was recorded at room temperature with the following parameters that maximize the S/N ratio:

- center field: 348.5 mT
- time constant: 655 ms
- sweep time: 42 s
- modulation amplitude: 1 mT.

Moreover, for the dosimeters containing alanine the additional recording parameters were

- field sweep: 20 mT,
- microwave power: 4 mW,

whereas for the dosimeters containing ammonium tartrate,

- field sweep: 10 mT
- microwave power: 1.59 mW.

The peak-to-peak signal height h_{pp} of the central line was measured and used as the dosimetric parameter (Regulla and Deffner 1982, ASTM 1995).

To take into account the dependence of the amplitude of the ESR signal on the orientation of each pellet inside the resonating cavity (Kojima *et al* 1995), we read out all samples at four orientations; each measured peak-to-peak amplitude h_{pp} was divided by the fractional weight content of alanine or ammonium tartrate in the sample. For each gamma dose and neutron fluence value, we irradiated three dosimeters and we used the mean value of all the 12 h_{pp} measurements (four orientation $\times$ three dosimeters). The h_{pp} standard deviation was always within $\pm 3\%$ of the average value.

The dosimeters of Gd_2O_3–alanine (Brai *et al* 2007a) and Gd_2O_3–ammonium tartrate (Brai *et al* 2007b) show an ESR signal due to Gd_2O_3 which is very wide ($\sim$270 mT) and centered in the $g \sim 2$ region field. This background signal is also present in the unirradiated samples and it does not depend on dose. The contribution of the Gd_2O_3 signal in the magnetic field range swept (338.5–358.5 mT for alanine and 343.5–353.5 mT for ammonium tartrate) is linear with a negative slope. To eliminate this background signal, a linear baseline was subtracted from each spectrum of Gd_2O_3–alanine and Gd_2O_3–ammonium tartrate. h_{pp} for these dosimeters was measured after this baseline correction.

3. Results and discussion

To perform measurements capable of discriminating the two components (photons and neutrons) of a mixed radiation field, it is desirable to use a dosimetric system consisting of a pair of detectors, one sensitive solely to neutrons and the other one solely to photons.

However, the most commonly used dosimeters, such as ionization chambers, Geiger–Müller counters, photographic emulsion and thermoluminescent materials, detect both photon and neutron components without distinguishing one contribution from the other.

If the values of neutron dose and photon dose in a mixed radiation field must be determined, it is necessary to select an adequate pair of dosimeters with different and known sensitivities to both radiation beams (ICRU 1977). Furthermore, if the neutron energy spectrum is known, the neutron kerma can be easily calculated through simple multiplication of the neutron fluence by the appropriate neutron kerma factor of the irradiated matter; the absorbed dose, in turn, can be calculated from the kerma value (ICRU 1977). Therefore, we analyzed the response of

our ESR pellets exposed to a mixed radiation field as a function of the neutron fluence Φ and of the gamma dose D_γ through the following equations:

$$\begin{cases} R_A^{n+\gamma} = f_A^n \Phi + f_A^\gamma D_\gamma \\ R_B^{n+\gamma} = f_B^n \Phi + f_B^\gamma D_\gamma, \end{cases} \tag{1}$$

where $R_A^{n+\gamma}$ is the response of the A dosimeter to the mixed field. The f_A^γ and f_A^n values are, respectively, the sensitivity factors to photons and neutrons. In an analogous way, the quantities with the subscript B refer to the B dosimeter.

The expressions of D_γ and Φ can be deduced from these equations:

$$\begin{cases} D_\gamma = \dfrac{f_B^n R_A^{n+\gamma} - f_A^n R_B^{n+\gamma}}{f_A^\gamma f_B^n - f_B^\gamma f_A^n} \\ \Phi = \dfrac{f_A^\gamma R_B^{n+\gamma} - f_B^\gamma R_A^{n+\gamma}}{f_A^\gamma f_B^n - f_B^\gamma f_A^n}. \end{cases} \tag{2}$$

To obtain the gamma dose and the neutron fluence we must know, for each dosimeter type, its sensitivity factor to each mixed field component. Therefore, we performed the calibrations of the ESR signal versus the gamma dose and the calibration of the ESR signal versus the neutron fluence.

3.1. Response to ^{60}Co

Calibration to γ photons was carried out in the range 1–50 Gy.

A linear behavior was found for all dosimeter types, and the following function was used for calibration:

$$h_\gamma = h_{\gamma 0} + f^\gamma D_\gamma. \tag{3}$$

The lowest detectable dose (LDD), defined as the dose that produces an ESR signal equal to the mean value of the zero-dose signal plus three standard deviations (Bartolotta *et al* 1993), was also calculated for all blends. The zero-dose signals were obtained from background measurements of six unirradiated dosimeters for each type in the magnetic field range, wherein the ESR signal of the radiation-induced free radicals was expected to appear. The LDD values for each dosimeter were found to be about 3 Gy for alanine and ammonium tartrate, and about 1 Gy for dosimeters with $B(OH)_3$ and Gd_2O_3 (Brai *et al* 2007a, 2007b). The reduction of the LDD in the dosimeters with Gd_2O_3 can be correlated with the increase of the dosimeter effective atomic number because of the presence of gadolinium. This increase is found to enhance the sensitivity to gamma photons and to cause a reduction of the LDD.

We shall use the parameters, such as γ sensitivities (f^γ) and lower detectable dose, obtained in the above-cited works, to perform the analyses on neutron data.

3.2. Response to thermal neutrons

At this stage we will describe the results obtained after irradiation of the various blends to the thermal neutron beam. The amplitude of the ESR signal as a function of the thermal neutron fluence was studied by exposing the dosimeters of each blend to four fluences between 2.40×10^{12} and 3.92×10^{13} cm^{-2}.

The ESR spectra of the solid state dosimeters, realized with alanine, $^{10}B(OH)_3$–alanine, Gd_2O_3–alanine, and exposed to a thermal neutron fluence $\Phi = 3.92 \times 10^{12}$ cm^{-2}, are reported in figure 1; figure 2 shows the ESR spectra of ammonium tartrate and Gd_2O_3–ammonium

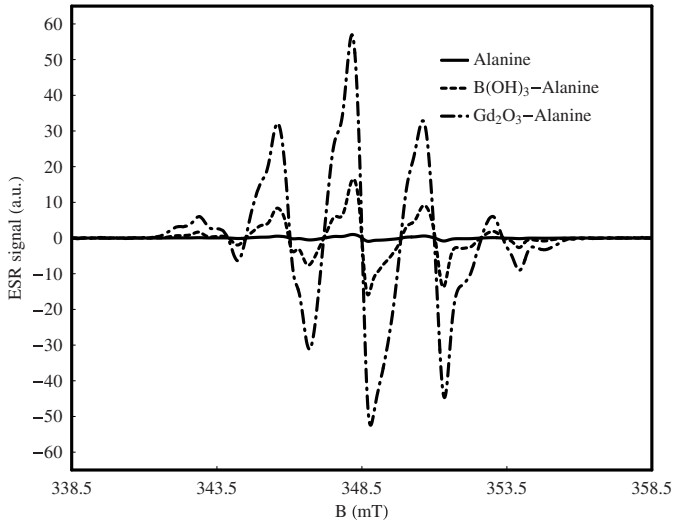

Figure 1. ESR spectra of alanine, $B(OH)_3$–alanine and Gd_2O_3–alanine dosimeters exposed to the mixed field (thermal neutrons and photons) at the same fluence value ($\Phi = 3.92 \times 10^{12}$ cm^{-2}).

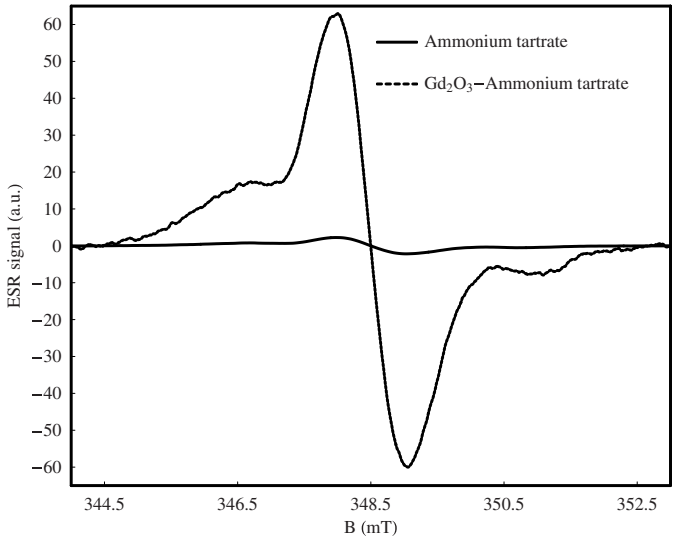

Figure 2. ESR spectra of ammonium tartrate and Gd_2O_3–ammonium tartrate dosimeters exposed to the mixed field (thermal neutrons and photons) at the same fluence value ($\Phi = 3.92 \times 10^{12}$ cm^{-2}).

tartrate irradiated with the same neutron fluence. The spectra of Gd_2O_3–alanine and Gd_2O_3–ammonium tartrate were obtained by subtraction of the Gd_2O_3 signal. The qualitative features of the EPR spectra obtained after irradiation with the mixed photon+neutron field are similar to those obtained after pure gamma irradiation.

Since the measured total (h_{tot}) ESR signal is due to both neutron and photon radiations, the ESR signal due to the gamma component must be subtracted to take into account only the neutron contribution. Therefore, the gamma dose measured by the ENEA laboratories was

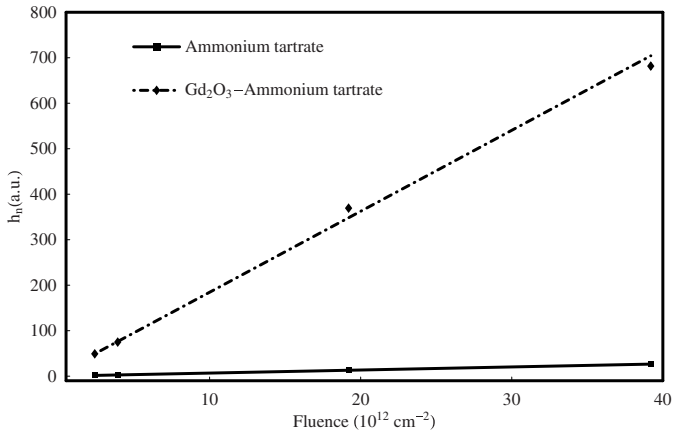

Figure 3. Trend of the h_n amplitude of the ESR signal of dosimeters of ammonium tartrate and Gd_2O_3–ammonium tartrate as a function of thermal neutron fluence. In the figure, the best fit curves are also reported.

Table 3. Results (sensitivity to γ photons f^{γ} and thermal neutron f^{n} parameters ± 1 S.D.) of the fitting procedure for the three typologies of samples.

Blend	Sensitivity to γ photons f^{γ}	Sensitivity to neutrons f^{n}
Alanine (A)	0.362 ± 0.004	$(3.88 \pm 0.04) \times 10^{-13}$
Ammonium tartrate (AT)	0.403 ± 0.005	$(6.75 \pm 0.17) \times 10^{-13}$
$B(OH)_3$–alanine (AB)	0.434 ± 0.010	$(49.1 \pm 0.9) \times 10^{-13}$
Gd_2O_3–alanine (AG)	0.670 ± 0.009	$(148.0 \pm 0.6) \times 10^{-13}$
Gd_2O_3–ammonium tartrate (ATG)	0.724 ± 0.009	$(178.0 \pm 1.5) \times 10^{-13}$

converted to the corresponding ESR signal intensity (h_γ), using the ^{60}Co calibration function of ESR dosimeters, and h_γ was subtracted from the total signal, h_{tot}. The 'residual' ESR signal ($h_n = h_{\mathrm{tot}} - h_\gamma$) is an estimate of the ESR signal attributable to thermal neutrons.

We analyzed the experimental data of the various typologies of dosimeters. A fit with a linear function was performed:

$$h_n = h_{n0} + f^n \Phi, \tag{4}$$

where Φ represents the fluence value provided by the irradiation center (which also provided the fluence uncertainties) and f^n is the sensitivity of the blend to the used neutron beam.

In table 3, the values of the gamma sensitivity f^{γ} and the neutron sensitivity f^{n} for each blend (± 1 S.D.) are shown.

As an example figure 3 shows the 'residual' ESR signal, h_n (with ± 1 S.D.), as a function of the neutron fluence for all blends with ammonium tartrate. In this figure, the best-fit curves are also shown. After thermal neutron irradiation, the dosimeters with Gd_2O_3–alanine and with $^{10}B(OH)_3$–alanine showed ESR signals were more intense (respectively, about 35 times and about 12 times) than alanine dosimeters. The dosimeters with Gd_2O_3–ammonium tartrate showed an ESR signal, about 26 times stronger in intensity with respect to the ammonium tartrate dosimeters. Our results show that the addition of gadolinium in both alanine and ammonium tartrate enhances very much the neutron sensitivity of the pellets. This is due to the very high thermal neutron capture cross section of the gadolinium and to the Auger

electrons released in the reaction with the thermal neutrons. Moreover, the sensitivity of alanine dosimeters with Gd_2O_3 is greater (about three times) than that of $B(OH)_3$–alanine.

3.3. Dosimetric system realization

When a dosimeter pair (e.g. *A* and *B* dosimeters) is exposed to a mixed radiation field, through the equation system (2), the values of D_γ and Φ can be obtained once the sensitivity factors to both photons and neutrons are known.

For the next analysis, we rewrite equation (2) in the following way:

$$\begin{cases} D_\gamma = \dfrac{f_B R_A^{*n+\gamma} - f_A R_B^{*n+\gamma}}{f_B - f_A} \\ \Phi = \dfrac{R_B^{*n+\gamma} - R_A^{*n+\gamma}}{f_B - f_A}, \end{cases} \tag{5}$$

where $R_A^{*n+\gamma} = R_A^{n+\gamma}/f_A^\gamma$ is the response to the mixed radiation field of the *A* detector compared to the sensitivity factor for gamma photons used for calibration and $f_A = f_A^n/f_A^\gamma$ is the ratio of the neutron sensitivity f_A^n to the photon sensitivity f_A^γ. The quantities with the subscript *B* are related to the *B* dosimeter. Equation (5) is valid if the detector response is linear with respect to the gamma dose and to the neutron fluence with a zero intercept. Since the calibration curve we utilized for both photons and neutrons is a straight line with non-zero intercepts, we evaluated the quantities $R_A^{*n+\gamma}$ as $R_A^{*n+\gamma} = \left(h_{\text{tot}}^A - h_{\gamma 0}^A - h_{n0}^A\right)/f_A^\gamma$ where $h_{\gamma 0}^A$ and h_{n0}^A indicate the intercepts of the calibration lines for photons and neutrons, respectively, h_{tot}^A is the peak-to-peak amplitude of the ESR signal in the mixed radiation field and f_A^γ is the slope of the calibration curve to the ^{60}Co photons. An analogous expression was used for the *B* detector.

The next step in our study was the determination of the most accurate pair of detectors for the measurement of thermal neutron fluence. As can be derived from equation (5) through the uncertainty propagation law, the indeterminates in the fluence and in the dose depend inversely upon the absolute value of the difference $(f_B - f_A)$. If this $|f_B - f_A|$ value is high, i.e. one dosimeter is more sensitive to photons than to neutrons and the other is more sensitive to neutrons than to photons, the dosimetric system can distinguish the two components of the mixed field and the indeterminates are smaller than for low $|f_B - f_A|$ values. Actually, for low $|f_B - f_A|$ values both detector *A* and detector *B* have almost the same sensitivity to neutrons and to photons; therefore $f_B \approx f_A \approx 1$ and the dosimetric system is not capable of distinguishing the contribution of each beam to the total signal. Consequently, the uncertainty under these conditions is greater than that in the first situation. Finally, we expect that the indeterminates on fluence measurements would be smaller when we use two dosimeters, one with and the other without additive nuclei, than when the dosimetric pair is constituted by two dosimeters, both with or without the additive nuclei.

3.4. The blind test

To verify this expectation and, in general, the usefulness of the dosimetric system under analysis, we performed a blind test. We irradiated at the nuclear reactor TAPIRO four dosimeters of each blend at unknown values of neutron fluence and photon dose using the same experimental setup used for the calibration procedure, as discussed in section 2.

The aim of this irradiation was to determine the fluence value through experimental measurements with a dosimeter pair in the mixed field and by using equation (5) and the corresponding sensitivity factors to photons and to neutrons.

Table 4. Values of neutron fluence and photon dose (±1 S.D.) obtained using various dosimeters pairs. In the table, the values of the sensitivity ratio $f_A = f_A^n / f_A^\gamma$ are also reported.

Dosimeter A	$f_A = f_A^n / f_A^\gamma$	Dosimeter B	$f_B = f_B^n / f_B^\gamma$	Φ (10^{12} cm^{-2})	D_γ (Gy)
A	1.15	AG	22.1	4.81 ± 0.14	1.2 ± 0.4
A	1.15	ATG	26.5	4.7 ± 0.5	1.3 ± 0.7
AT	1.71	AG	22.1	4.76 ± 0.15	2.2 ± 1.2
AT	1.71	ATG	26.5	4.6 ± 0.6	2.4 ± 1.5

As reported in table 4, the best results were obtained when gadolinium was used. The uncertainties on the fluence and on the dose were calculated by using the uncertainties of the experimental measurements $R_A^{*n+\gamma}$ and $R_B^{*n+\gamma}$ and of the calibration parameters.

The nominal values of neutron fluence and photon dose, measured at the TAPIRO laboratories of the ENEA center, Casaccia (Rome), with different measurement systems (ionization chambers and activation gold foils), are $\Phi = (4.80 \pm 0.14) \times 10^{12}$ cm^{-2} and $D_\gamma = 0.410 \pm 0.012$ Gy, respectively.

As can be observed from table 4, all the neutron fluence values obtained through experimental measurements of the chosen dosimeter pairs are in good agreement with the nominal value (highest deviation about 4%). Moreover, for the dosimeter pairs of alanine and Gd_2O_3–alanine and ammonium tartrate and Gd_2O_3–alanine, the relative uncertainty is about 3%, well within the limiting value (5%) for application in the radiotherapeutic field.

Regarding the photon dose, the values obtained by using the various detector pairs (table 4) are all consistent with the zero value. This is observed because the absorbed photon dose is for all the dosimeter pairs smaller than the lowest detectable dose of at least one dosimeter of the pair. Consequently, this value cannot be distinguished from the background signal.

Our results clearly show that the addition of gadolinium significantly increases, and more than boron, the response of the ESR dosimeters to thermal neutrons.

It is however necessary to underline that alanine and ammonium tartrate dosimeters with such high amounts of gadolinium oxide lose their tissue equivalence characteristics, particularly in the low energy range, due to the high atomic number of gadolinium ($Z_{Gd} = 64$). This can be a serious drawback, since the material used for dosimetry in radiation therapy should be tissue equivalent, i.e. its atomic composition and density should be close to those of soft tissue (ICRU 1989). Therefore, under the same irradiation conditions, the photon response of alanine and ammonium tartrate dosimeters with gadolinium will be different than that of soft tissue. However, it could be possible to take these differences into account and to correct the measured dose if the energy spectrum of the photons is well known. It is necessary to underline that, even though the dosimeter with gadolinium is not tissue equivalent, the neutron fluence value is determined with high accuracy because the gamma component contributes much less to the total ESR signal than the neutron component. Also, it should be noted that in this work we mainly wanted to extend the applicability of ESR dosimetry to characterize the thermal neutron fluence in the mixed field. New experiments are planned to find the optimized percentage of gadolinium inside the ESR dosimeter, so that the dosimeter sensitivity to thermal neutrons can be further improved without significantly modifying its tissue equivalence.

4. Conclusion

We conclude from this study that the employed dosimetric system (a dosimeter pair of alanine and Gd_2O_3–alanine and a dosimeter pair of ammonium tartrate and Gd_2O_3–alanine) is able

to determine the neutron fluence in a mixed field (thermal neutrons and photons) with an uncertainty of about 3%. This result holds good under irradiation conditions similar to the calibration ones, that is, when the thermal neutrons are the main component of the neutron flux.

From calibration curves, we observed that the introduction of Gd in these organic compounds significantly increases (about a factor of 30) the sensitivity of dosimeters to neutron radiation. This result is due to the high gadolinium cross section for neutron capture, that greatly increases the probability of thermal neutrons' interaction, and also to the secondary particles and, in particular, the Auger electrons that release their energy entirely in the dosimeter.

We performed a blind test to analyze the usefulness of the dosimetric system constituted by two dosimeters, and we obtained that the alanine and Gd–alanine pair and ammonium tartrate and Gd–alanine furnish a neutron fluence value with a percentage error of 3%, which is lower than the threshold of 5% for application in radiotherapy.

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