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EPR DOSIMETRY IN A MIXED NEUTRON AND GAMMA RADIATION FIELD

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Suitability of Electron Paramagnetic Resonance (EPR) spectroscopy for criticality dosimetry was evaluated for tooth enamel, mannose and alanine pellets during the 'international intercomparison of criticality dosimetry techniques' at the SILENE reactor held in Valduc in June 2002, France. These three materials were irradiated in neutron and gamma-ray fields of various relative intensities and spectral distributions in order to evaluate their neutron sensitivity. The neutron response was found to be around 10% for tooth enamel, 45% for mannose and between 40 and 90% for alanine pellets according their type. According to the IAEA recommendations on the early estimate of criticality accident absorbed dose, analyzed results show the EPR potentiality and complementarity with regular criticality techniques.

INTRODUCTION

Electron Paramagnetic Resonance (EPR) was proposed many years ago as a method to assess radiation dose[1], based on the measurements of the concentration of free radicals induced by ionizing radiation in irradiated materials. Stability of radio-induced free radicals in solid materials allows long-term retrospective dosimetry. In the last 20 y, EPR retrospective dosimetry performed on available natural or artificial materials (bone and tooth tissues, polymers, sucrose, etc.), has been helpful for individual dose reconstruction after radiological accidents, especially in case of lack of personal dosemeters, orphan sources found by public or malfunctioning of radiotherapy facility[2–7]. In particular, tooth enamel dosimetry has been established as a very effective tool for photon dose assessment in populated areas contaminated after bomb explosion[8] or nuclear power plant accident[9,10]. It is worth noting that in most of the accidents only photon exposure was involved, nevertheless, in some cases such as criticality accidents, as for example Tokai Mura, the neutron dose component may not be ignored. The only studies which dealt with fast neutron dosimetry using EPR, mainly concerned alanine and sucrose[11–14]. In particular, these materials have been tested during the criticality accident intercomparison performed in Valduc, France in 1993[14]. The results showed that alanine pellets estimate, in addition to the photon dose, almost half of the neutron dose. Sucrose responds similarly, but a dose-dependent time instability of the EPR signal limits its use[14]. To our knowledge, only one study was published on tooth enamel response for neutrons of energy comparable to fission spectrum[15].

The aim of this present work is to study the influence of the radiation quality factor on the EPR material sensitivity in order to estimate the neutron contribution to the total dose for a mixed neutron gamma field. During the 'international intercomparison of criticality dosimetry techniques' held in Valduc, France, in June 2002, three types of materials were exposed to the SILENE reactor mixed fields: tooth enamel, alanine pellets of different types and mannose. Despite of medical restrictions concerning enamel removal from irradiated persons, neutron dependence of tooth enamel was investigated, because, on one hand, it can be useful for a later dose reconstruction and on the other hand, the technique is on fast development for *in vivo* application. According to the promising results concerning alanine in the previous studies[11–14], alanine pellets with different compositions were compared: ^{10}B doped and conventional alanine pellets. Mannose, whose neutron response was already studied by lyoluminescence[16,17], was also investigated as an EPR material for criticality dosimetry.

MATERIALS AND METHODS

Description of the reactor

Samples were irradiated at the SILENE reactor during the 'international intercomparison of criticality dosimetry techniques' held in Valduc (France) in June 2002. This facility, dedicated to experimental research, provides mixed gamma and neutron field as those encountered in criticality accidents[18,19].

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Sample preparation

The samples were independently prepared and measured in three different laboratories: at the Institute for Radiological Protection and Nuclear Safety (IRSN), at the Istituto Superiore di Sanità (ISS) and at the University of Palermo (UNIPA).

Tooth enamel

Tooth enamel samples were prepared from molar teeth extracted for medical indications. At IRSN, whole teeth were irradiated without specific preparation. After irradiation, they were prepared with the use of hard alloy dental drill and crashed with the use of nippers to particle with average size of ~1.5 mm. Samples were cleaned with acetic acid and acetone. At ISS, six molar teeth were cut along the sagittal plane. Half of each tooth was irradiated in a head-like phantom, and, after the irradiation, they were prepared following the ISS enamel powder preparation[20]. The other half of each tooth was first prepared as enamel powder samples following the ISS procedure[20] and then individually measured with EPR. The measured spectra were taken as the control spectra for the half teeth irradiated in the head phantom. After the EPR measurement, the powder samples were pooled together and then divided in seven aliquots of ~120 mg mass. One aliquot was not irradiated in the neutron beam and was used as a control sample of the pool. The other six aliquots were used for neutron irradiation in a miniphantom.

Alanine

Two types of alanine dosemeters were studied: commercial alanine pellets (Bruker), conventional and ^{10}B doped alanine pellets, both manufactured at UNIPA according to the preparation procedure described by Bartolotta *et al.*[21].

Mannose

D-Mannose purchased from Prolabo, France, was used without further preparation. Samples were placed in a cylindrical plastic container.

Sample irradiation

Three experiments were performed at the SILENE reactor, with various neutron to gamma ratios and different spectral distributions. The characteristics of each experiment are given in Table 1. The samples were circularly placed at 4 m from the reactor core at a height of 1.2 m above the floor corresponding to the centre of the SILENE reactor tank. For each experiment, the samples were irradiated in the following conditions: a pair of whole teeth, mannose and alanine in free air, two half teeth (belonging to two different molar teeth) in a PMMA (polymethyl methacrylate) cylindrical phantom simulating a head, two enamel powder samples in PMMA miniphantoms[20].

Table 1. Characteristics of the SILENE experiments.

	Mode	Number of fissions	Shield
Experiment 1	Free evolution	$1.25\ 10^{17}$	Bare
Experiment 2	Steady state	$7.25\ 10^{16}$	Lead
Experiment 3	Free evolution	$1.54\ 10^{17}$	Lead

Reference dosimetry

The neutron and gamma tissue kerma, were separately estimated. For samples irradiated in free air, the gamma dosimetry was carried out with alumina powder. Standards in terms of tissue kerma have been established with a ^{60}Co reference source. This thermoluminescent detector (TLD) has a weak response to neutrons[22]. For tooth samples irradiated in the head-like phantom, five TLD-300 ($CaF_2:Tm$) were placed on the top of the tooth, in order to measure the gamma dose inside the cylinder. TLD measurements were performed with a TOLEDO reader. The uncertainties were respectively estimated at 5% for alumina and 30% for TLD 300 at 2σ.

The neutron tissue kerma was given by silicon diodes used as passive detectors[23]. These silicon diodes produced in Czech Republic has negligible response to photons[23]. The uncertainties were given at 15% at 2σ.

EPR measurements

EPR measurements were performed at room temperature with spectrometers operating in X-band. The IRSN and the ISS spectrometers were equipped with high-Q cavities (ER4"122SHQ at IRSN and SHQ at ISS), provided with an internal standard of Mn^{2+}/MgO. The UNIPA spectrometer was equipped with a TE_{102} rectangular cavity. Unless stated otherwise, the spectra recording conditions and parameters were those described by Ivannikov *et al.*[24], by Fattibene *et al.*[20] and by Bartolotta *et al.*[21] for the measurements carried out respectively at IRSN, ISS and UNIPA.

Dose estimation procedure

IRSN procedure

For tooth enamel, alanine and mannose, the radio-induced signal intensity was determined as its maximum amplitude and normalized to the mass and to

the internal standard intensity line. For tooth enamel, the native background signal was subtracted using a spectrum deconvolution software provided by MRRC, Russia[25]. For alanine and mannose no initial signal was observed. In the case of mannose, since a short life radical overlapped the main radio-induced signal[26], measurements were performed at least 2 months after irradiation[26]. The total absorbed dose was then determined from a ^{60}Co calibration curve in term of tissue kerma respecting conditions of electronic equilibrium.

ISS procedure

For enamel, the signal intensity was determined with a EPR powder spectrum simulation software. The signal intensity was corrected by a number of factors, which take into account the sample mass, the manganese line intensity, the microwave cavity filling factor and the systematic contribution from the microwave cavity signal. For each sample, the corrected signal intensity of the respective control sample was subtracted. The total dose was evaluated from a calibration curve established at ISS with a ^{60}Co source.

UNIPA procedure

The peak-to-peak height of the ESR central line of each irradiated dosemeter was measured and normalised to the mass of the dosemeter and to the spectrometer reference standard; the mean value of normalised signal intensity of non-irradiated samples was also subtracted. The total absorbed dose was obtained by means of a ^{60}Co calibration factor; irradiations of alanine and ^{10}B–alanine dosemeters for calibration purposes were carried out at ISS with the same facility used for the tooth enamel calibration.

RESULTS AND DISCUSSION

For all the samples, no significant qualitative differences were found between the EPR spectra following the ^{60}Co irradiations performed for calibration curves and neutron–gamma irradiations at the SILENE reactor. Therefore, it was not possible to separate neutron and photon components. The reference dosimetry was then used to estimate the relative neutron sensitivity, which was defined as the difference between EPR total dose and photon dose divided by neutron kerma, all terms expressed in tissue kerma.

The photon and neutron doses measured by TLD and silicon diodes are given in Table 2. The total mean doses estimated by EPR are given in Tables 3 and 4. The relative sensitivity to neutrons is given for each material in Table 5.

For all experiments, EPR results underestimate total dose reference. For the lead configurations, results for the different materials are consistent with previous studies. For the bare configuration, neutron sensitivity for all exposed material is found lower by roughly a factor 2 and with higher uncertainties compared to lead configuration results. The lead reactor configuration, with roughly a factor 10 between neutron and photon in term of kerma, is then the most propitious configuration to evaluate it

Table 2. Reference dosimetry results (tissue kerma).

	Photon dose (Gy)		Neutron dose (Gy) Silicon diode
	Alumina	TLD 300	
Experiment 1	2.50 ± 0.13	2.13 ± 0.64	1.74 ± 0.26
Experiment 2	0.14 ± 0.02	0.17 ± 0.06	0.79 ± 0.12
Experiment 3	0.30 ± 0.03	0.37 ± 0.12	1.74 ± 0.26

Table 3. Average of total doses estimated by EPR measurements on tooth enamel irradiated in free air, in miniphantom and in a head-like phantom expressed in tissue kerma (Gy).

	Average of EPR total dose in enamel (Gy)		
	Free air (IRSN)	Miniphantom (ISS)	Head phantom (ISS)
Experiment 1	2.56 ± 0.09	2.04 ± 0.09	2.17 ± 0.09
Experiment 2	0.33 ± 0.12	0.24 ± 0.09	0.26 ± 0.09
Experiment 3	0.38 ± 0.12	0.36 ± 0.09	0.45 ± 0.09

Table 4. Average of total doses estimated by EPR measurements on alanine and mannose expressed in tissue kerma (Gy).

	Average of EPR total dose (Gy)			
	Commercial alanine pellets (IRSN)	Alanine pellets (UNIPA)	^{10}B doped alanine (UNIPA)	Mannose (IRSN)
Experiment 1	2.83 ± 0.28	3.45 ± 0.35	3.29 ± 0.33	2.92 ± 0.23
Experiment 2	Not exposed	<DL	<DL	<DL
Experiment 3	1.00 ± 0.10	1.83 ± 0.18	1.54 ± 0.15	1.05 ± 0.12

Table 5. Relative sensitivity to SILENE neutrons of the EPR materials studied.

	Mannose	Alanine (IRSN)	Alanine (UNIPA)	^{10}B doped Alanine (UNIPA)	Whole tooth (IRSN)	Enamel powder (ISS)	Half tooth (ISS)
Experiment 1	0.24 ± 0.15	0.20 ± 0.18	0.55 ± 0.23	0.47 ± 0.21	0.03 ± 0.09	-0.26 ± 0.10	0.02 ± 0.09
Experiment 2	<DL	Not exposed	<DL	<DL	0.24 ± 0.12	0.13 ± 0.12	0.11 ± 0.12
Experiment 3	0.43 ± 0.09	0.42 ± 0.08	0.88 ± 0.17	0.71 ± 0.14	0.05 ± 0.07	0.04 ± 0.05	0.05 ± 0.09

with minimal uncertainties. Thus, only results in lead configuration should be taken into account for neutron sensitivity estimation and compared with previous work.

Tooth enamel

The tooth enamel sensitivity to neutrons is estimated to be ~0.1 for the lead configurations, which is, considering associated uncertainties, in agreement with Bochvar *et al.*[15] results. Bochvar *et al.* found a response function of tooth enamel to neutrons equal to 0.03, comparing the response to neutrons of 1 keV – 1 MeV with gamma rays response (^{60}Co, ^{137}Cs). For the bare configuration, results are more scattered, the neutron sensitivity ranges between −0.27 and 0.04.

Total mean doses are slightly higher in the head-like phantom than in the miniphantom. The difference is very likely due to the secondary gamma generated by neutron interactions in the phantom. This additional dose in phantom does not exceed theoretically 10% of the neutron dose under 1 cm of tissue[27]. However, considering the uncertainties, the phantom influence can be considered as negligible in this range of dose. For whole teeth irradiated in free air, as the enamel background dose was not taken into account and may not be negligible, then dose results are slightly higher than in scattering medium.

Alanine

The relative neutron response of commercial pellets from Bruker was found to be 0.42 and 0.20 respectively for lead and bare configurations, which is in good agreement with previous works. Relative response to ^{60}Co gamma rays measured by Schraube *et al.* was found to be 0.36–0.5 for neutron energies between 0.5 and 2 MeV with 20% paraffin mixture[12]. Bermann found 0.54 for 2.0 MeV and 0.57 for 1.8 MeV[11,13]. Katsumura *et al.* reported a relative response of 0.40 and 0.54 for fission neutron[11] and D'Errico *et al.* for the previous SILENE intercomparison (1993) found 0.45[14].

The response of the UNIPA alanine dosemeters was found to be 0.47 for the borated alanine and 0.55 for conventional alanine in bare configuration and respectively 0.71 and 0.88 for lead configuration. The sensitivity for UNIPA conventional dosemeter is twice higher than Brucker or ISS pellets irradiated during the last SILENE intercomparison in 1993[14].

Considering the weak dose component of thermal neutron in the SILENE field, the effect of ^{10}B only partially compensates the smaller amount of alanine in the dosemeter (~50% less than in conventional dosemeters). The detection limits of both UNIPA pellets estimated around 1 Gy did not allow estimating dose for the steady state experiment.

Mannose

Mannose was found slightly less sensitive to neutron than commercial sugar exposed in the 1993 intercomparison. For lead and bare configurations, the relative response to neutron was estimated to be 0.43 and 0.24, respectively. Detection limit estimated around 0.5 Gy did not allow estimating dose in the steady state experiment. Mannose results are consistent with the literature; for instance, with lyoluminescence techniques; Puite *et al.*[16] and Ettinger *et al.*[17]

found respectively a response relatively to ^{60}Co gamma rays of 0.34 for fission neutrons and 0.2 for 1 MeV neutrons. As for commercial sugar, mannose showed a time dependence shape line spectrum modification, which was already observed by Bartlett *et al.*[28]. The signal stabilization occurred within 2 months after irradiation. However, an annealing treatment at low temperature seems allowing similar signal stabilization[26] and could be studied as post-irradiation treatment to solve this time dependence signal problem.

CONCLUSION

Tooth enamel neutron sensitivity was found to be weak for the SILENE neutron energies. Nevertheless, tooth enamel can be an accurate estimator of the photon component in a mixed neutron–gamma field. Alanine pellets allow to measure at least 90% of the total dose delivered in a mixed neutron–gamma field. Mannose results show potentiality of this material for mixed field dose assessment, especially considering a possible neutron sensitivity improvement as done for alanine by adding a binder.

This study shows the potentiality and the advantage of the EPR technique. Two different axes for the use of EPR technique in the field of criticality accident dose reconstruction can be distinguished. The first one concerned development and study of EPR materials for a specific dosemeter dedicated to criticality accident using tissue equivalent materials such as alanine or mannose for example. The second one, based on the materials exposed from the victims (enamel or bone) or in their vicinity (sugar) that could be used for dose reconstruction, could be complementary of regular activation measurement technique, especially when no dosemeter is worn or for lower level exposition. The EPR technique seems fully answer to accident dose reconstruction requirement, either in term of detection limit, uncertainties or dose estimation delay.

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