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Study of suitability of Fricke-gel-layer dosimeters for in-air measurements to characterise epithermal/thermal neutron beams for NCT

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HIGHLIGHTS

- Fricke gel dosimeters in form of layers were deeply inspected.
- Suitability of gel dosimeters for characterization of epithermal beams was determined.
- Proper dosimeter shape and dimension were identified.
- Good consistency between measurements and MC simulations was found.

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ABSTRACT

The reliability of Fricke gel dosimeters in form of layers for measurements aimed at the characterization of epithermal neutron beams has been studied. By means of dosimeters of different isotopic composition (standard, containing ^{10}B or prepared with heavy water) placed against the collimator exit, the spatial distribution of gamma and fast neutron doses and of thermal neutron fluence are attained. In order to investigate the accuracy of the results obtained with in-air measurements, suitable MC simulations have been developed and experimental measurements have been performed utilizing Fricke gel dosimeters, thermoluminescence detectors and activation foils. The studies were related to the epithermal beam designed for BNCT irradiations at the research reactor LVR-15 (Řež). The results of calculation and measurements have revealed good consistency of gamma dose and fast neutron 2D distributions obtained with gel dosimeters in form of layers. In contrast, noticeable modification of thermal neutron fluence is caused by the neutron moderation produced by the dosimeter material. Fricke gel dosimeters in thin cylinders, with diameter not greater than 3 mm, have proved to give good results for thermal neutron profiling. For greater accuracy of all results, a better knowledge of the dependence of gel dosimeter sensitivity on radiation LET is needed.

1. Fundamentals

Dose measurements in thermal/epithermal neutron fields are challenging due to the complexity of neutron interactions, the very high neutron fluence rates (frequently around 10^9 cm^{-2} s^{-1}) and, finally, the requirement of separating dose components with different radiobiological effectiveness (RBE).

In tissue exposed to an epithermal neutron field, the absorbed dose components are: (i) the gamma dose, due to the capture reaction $^1H(n, \gamma)^2H$ and to background from the reactor structural materials, (ii) the 'fast neutron' dose, mainly due to recoil protons produced by elastic scattering of fast and epithermal neutrons with Hydrogen nuclei and (iii) the dose due to protons emitted in the reaction $^{14}N(n, p)^{14}C$. The photons emitted in capture reactions

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with Hydrogen nuclei have energy of 2.2 MeV and release this energy in the irradiated volume and in normal tissue even in positions far from where the interaction occurs. Then, the spatial distribution of the gamma dose depends on both shape and size of the irradiated volume. The fast neutron dose, in each point, is almost independent of the shape of the irradiated volume and decreases in depth with a distribution depending on the energy spectrum of the beam. The dose due to the protons (about 630 keV in energy) generated from capture reaction with Nitrogen, shows a spatial distribution similar to that of the thermal neutron fluence. The more the epithermal neutrons slow down, the more the direction of their motion becomes isotropic. Therefore, thermalized neutrons diffuse in any direction, also backwards in the volume already passed through. As a consequence, the thermal neutron spatial distribution is dependent on both shape and size of the irradiated volume. Thus the determination of the dose distribution in tissue irradiated in epithermal fields is complex, because all the components of the radiation field should be known, since their RBE is different. In measurements, great attention has to be paid to the composition and dimension of the volumes exposed to the neutron field, in order to avoid altering the distribution of thermal neutrons and the resulting doses.

Currently, the thermal neutron fluence is mostly measured with activation foils or wires, while gamma and fast neutron doses are estimated with paired ionization chambers (Kosunen et al., 1999). Thermoluminescence dosimeters (TLDs) are also utilized for gamma dose mapping, coupled to another detector in order to subtract the contribution of thermal neutrons to their response (Aschan et al., 1999; Burgkhardt et al., 2006). Suitable algorithms to attain gamma dose, with a satisfactory accuracy, from the TL emission curve, avoiding coupled measurements or Monte Carlo (MC) calculations, have been developed too.

In recent years, gel dosimetry has created considerable interest in the neutron capture therapy (NCT) community in particular by exploiting the isotope ^{10}B (BNCT). Both Fricke and polymer gels have been studied and tested to investigate their suitability for BNCT dosimetry. Polymer gel dosimeters are often preferred, for the higher stability of the dose distribution after irradiation. In fact, in Fricke gel dosimeters ferric ion diffusion causes a loss of resolution of the acquired images and therefore the acquisition of the images must be performed within a relatively short time. In the case of Fricke–Xylenol-Orange gel dosimeters, the diffusion coefficient is dependent on the amount of Xylenol Orange, and is around 1 mm^2/h (Gambarini et al. 2004a). Even though in neutron fields there are not high dose gradients, as instead occurs in the case of radiotherapy with photons or with charged particles, however it is appropriate to perform the dosimeter analysis within a few hours after irradiation. In order to avoid the trouble of setting up the analysis instrumentation sufficiently near to the irradiation facility, polymer dosimeters have been taken into consideration. Some studies on the suitability of polymer gel dosimeters for BNCT have been carried out, concerning both their tissue equivalence (Wojnecki and Green, 2001) and the potential for dose distribution measurements in NCT neutron fields (Uusi-Simola et al., 2003, 2007). Such dosimeters have revealed good tissue equivalence, but in epithermal neutron beams they allow only relative dose determinations, because the absorbed dose is the sum of contributions due to different secondary radiation with different linear energy transfer (LET), for which the sensitivity of the dosimeter is different.

FGLDs give the possibility of measuring the spatial distribution of each dose component. In fact, a method for achieving the separation of the different dose contributions has been developed, based on Fricke–Xylenol-Orange gel dosimeters in form of layers, 5 mm in total thickness and 3 mm in gel thickness. The separation of the dose components is achieved by analytical comparison of the doses measured with gel dosimeters prepared with or without an isotope whose reaction with neutrons produces charged particles. Images and profiles of boron dose (and also of thermal neutron fluence), are attained by means of two FGLDs, one with the standard composition and the other containing a suitable amount of ^{10}B (Gambarini et al., 2002, 2004b). Usually, 40 μg/g of ^{10}B are introduced into the gel composition. In a gel dosimeter containing ^{10}B, the absorbed dose is due to gamma rays, to epithermal and fast neutrons (mainly through recoil protons) and to the charged particles generated by the $^{10}B(n,\alpha)^7Li$ reaction. The boron dose is then obtained by pixel-to-pixel elaboration of the dose images measured with a standard dosimeter and a dosimeter containing ^{10}B. The sensitivity of gel dosimeters to the dose due to charged particles is lower than that due to gamma dose. The dependence of the gel dosimeter sensitivity on linear energy transfer (LET) is not well known and needs further research. The coefficient of relative sensitivity to boron dose with respect to gamma dose was recently found to be 0.55 (Gambarini et al., in press), but the uncertainty on this value is still high. This uncertainty is the main source of error in the determination of the boron dose in BNCT and of the thermal neutron fluence that can be evaluated from the boron dose by means of kerma factors. Due to this low sensitivity to charged particles, the boron dose is often a low contribution to the total dose measured with gel dosimeters. With the aim of imaging thermal neutron fluence, therefore, a higher amount of ^{10}B in the dosimeter composition may seem advantageous, but a higher amount of the chemical compound containing boron gives a great decrease of the yield of ferrous ion oxidation in the dosimeter. For the preparation of the dosimeters with boron, sodium tetraboratetecahydrate [$B_4Na_2O_7 \cdot 10H_2O$] was added to the chemical solution. A compound containing only the isotope ^{10}B (that is 19.9% of natural B) would be more advantageous for thermal fluence measurements, but it was not yet used because too expensive.

Fast-neutron and photon doses can be separated by means of a pair of FGLDs, one made with water and the other with heavy water. Fast neutron dose is mainly due to recoil protons in the first gel and to recoil deuterons in the second one. In the algorithms that allow separating gamma and fast neutron doses (Gambarini et al., 2010) the relative sensitivity of gel dosimeters to slow-down protons and deuterons have to be considered. Satisfactory results have been obtained with the present uncertainty on the dependence on LET of the gel dosimeter sensitivity. Further knowledge on this topic will increase the achieved precision.

Concerning the consistency of the results, a better reliability is attained for measurement in-phantom than in-air. For in-phantom measurements, Fricke-gel-layer-dosimeters (FGLDs) are suitable, because of their water equivalence. Moreover, the layer geometry avoids significant variation of neutron transport due to variations of the isotopic composition of the gel matrix.

FGLDs have been utilized many times for in-phantoms measurements, initially at the thermal and epithermal columns of the ENEA's research reactor TAPIRO in Casaccia (It) (Gambarini et al., 2001, 2002) and subsequently at the BNCT epithermal column of LVR-15 research reactor, at the Research Center Řež (Czech Republic) (Burian et al., 2009, Bartesaghi et al., 2009; Gambarini et al., 2010).

Some in-air measurements have also been carried out for gamma and fast neutron dose imaging at the collimator exit of the BNCT epithermal column of the LVR-15 reactor (Gambarini et al., 2011). Other measurements have been performed with moderated neutrons. In fact, for biological experiments on small animals or cell cultures carried out at epithermal columns, the epithermal neutrons are suitably thermalized by means of a layer of moderating material. The moderator chosen for biological experiments at the LVR-15 research reactor consists in a polyethylene disk placed

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