

# IN-PHANTOM DISCRIMINATION OF THE DOSE CONTRIBUTIONS BY MEANS OF FRIXY-GEL AND THERMOLUMINESCENT DOSIMETERS

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## ABSTRACT

In order to answer to the dosimetric requirements of BNCT concerning experimental determination of absorbed dose in phantoms, two different kinds of detectors have been used: gel and thermoluminescent dosimeters. By means of the developed method, it is possible to discriminate the various dose components in phantoms exposed to a neutron field characteristic of BNCT. As known, experimental information, and not only calculations, is of fundamental importance to evaluate the feasibility of the therapy. Four kinds of hand-made gel have been used: standard gel for  $\gamma$ -dose measurements; standard gel added with nitrogen for the proton dose due to the  $^{14}\text{N}(n,p)^{14}\text{C}$  reaction; standard gel added with the proper amount of boron to measure the therapeutic dose and standard gel made with heavy water ( $\text{D}_2\text{O}$ ) instead of light water to measure the fast neutron dose. Among thermoluminescent dosimeters commercially available, calcium and lithium fluorides have been chosen:  $\text{CaF}_2:\text{Tm}$  (TLD-300) and  $\text{LiF}:\text{Mg,Ti}$  with different percentage of  $^6\text{Li}$  (TLD-100, TLD-600, TLD-700). Calcium fluoride has been used for  $\gamma$ -dose evaluations, and lithium fluorides have been used to determine thermal neutron fluxes. The absorbed dose profiles have been measured in a polyethylene phantom, exposed both in the thermal and in the epithermal column available at the RVS-TAPIRO reactor, at the ENEA Casaccia Research Centre (Rome). Experimental results with gel, thermoluminescent dosimeters and standard activation techniques have shown good consistency.

## Introduction

The dosimetry for BNCT involves measurements in high fluxes of thermal and epithermal neutrons and therefore it is very complex. This difficulty comes from the multiplicity of energy release and absorption mechanisms in neutron fields. In fact the various secondary radiations are characterised by different interaction modalities, different LET (Linear Energy Transfer) and RBE (Relative Biological Effectiveness); consequently, it is mandatory to perform precise evaluations of the absorbed dose in tissue, separating the dose contributions whose biological effects are different. Furthermore, for an evaluation of feasibility of treatments, the dose distributions in polyethylene phantoms have to be measured, because the maximum thermal neutron fluence admitted for treatments is established on the basis of the dose delivered by thermal and epithermal neutrons to the health tissue surrounding the tumour.

Absorbed dose profiles have been measured in a cylindrical polyethylene phantom (14 cm high, 16 cm in diameter), exposed in the thermal or epithermal column of TAPIRO Reactor. The phantom is shown in Fig.1 and in Fig.2. In the thermal column, the irradiations have been performed with the aim of developing the method and of calibrating the dosimeters



Fig.2. Phantom during set up for irradiation in the thermal column.

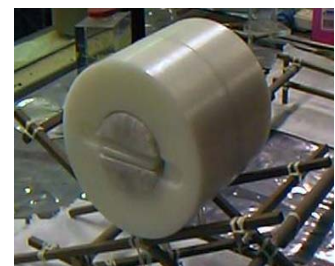


Fig.1. Phantom settled on the support for irradiation in the epithermal column.

in a thermal neutron field; for such calibration, the neutron fluxes have been measured by means of activation techniques, utilising Au foils, bare and cadmium-shielded, and unfolding technique analysis. In the epithermal column, measurements have been essentially finalized to study in-phantom dose distributions also in presence of deep brain tumour simulations (that is, volumes containing the proper amount of  $^{10}\text{B}$ ). In these configurations, the dose contributions to be discriminated are: the  $\gamma$ -dose arising from

$^1\text{H}(n,\gamma)^2\text{H}$ , thermal neutron reaction on the hydrogen nuclei in the polyethylene phantom, and from the reactor background; proton dose due to the  $^{14}\text{N}(n,p)^{14}\text{C}$  reaction; therapeutic dose coming from  $^{10}\text{B}(n,\alpha)^7\text{Li}$  and fast neutron dose mainly due to recoil protons.

Neutron irradiations have been carried out at the RVS-TAPIRO reactor, at the ENEA Casaccia Research Centre (Rome). Thermal and epithermal columns have been properly designed and constructed for BNCT experimentation. The nominal power is 5 kW and the maximum neutron flux at core is  $4 \cdot 10^{12} \text{cm}^{-2} \text{s}^{-1}$ . Dosimeter exposures have been performed at suitably chosen reactor powers and irradiation times, depending on the dosimeter kind, in order to optimise their response.

### Gel dosimetry

The utilised FriXy-Gel dosimeters are aqueous dilute solutions incorporated in gel matrixes that maintain the spatial distribution of the absorbed dose firm in time. To image and profile dose distributions, gels in form of layers (5 cm wide, 11 cm long and 3 mm thick) have been utilised. This is possible because the absorbed dose is correlable to the variation of the gel optical density ( $\Delta(\text{OD})$ ) around 580 nm<sup>(1)</sup>. Gel dosimeters are placed on a plane illuminator and light absorbance images, before and after irradiation, are detected using a CCD camera equipped with an optical band pass filter centred around 580 nm<sup>(2)</sup>. Then a pixel-to-pixel elaboration of the grey levels (GL) values of the images before and after exposure are performed to obtain the variation of the optical density:

$$\Delta(\text{OD}) = \text{GL}_{\text{after}} - \text{GL}_{\text{before}} = \text{Log}_{10} (\text{GL}_{\text{before}} / \text{GL}_{\text{after}}).$$

For the gels utilised in this experiment, the absorbed dose is linearly proportional to the  $\Delta(\text{OD})$  at least until 20 Gy, and the slightest detectable dose is about 0.5 Gy.

The standard FriXy-Gel composition is: Fricke solution (1 mM  $\text{Fe}(\text{NH}_4)_2(\text{SO}_4)_2 \cdot 6\text{H}_2\text{O}$ , 25 mM  $\text{H}_2\text{SO}_4$ ), Agarose (1% of the final weight  $\text{C}_{12}\text{H}_{14}\text{O}_5(\text{OH})_4$ ) as gelling agent and Xylenol-Orange (0.165 mM  $\text{C}_{31}\text{H}_{27}\text{N}_2\text{Na}_5\text{O}_{13}\text{S}$ ) that shifts the light absorbtion band from UV to visible and moreover forms a complex with ferric ions giving absorbtion around 580 nm. This gel allows to measure the photon dose component of the radiation field.

If to the standard gel composition is added an isotope that reacts with neutrons producing charged particles, whose energy is locally realised and absorbed, then it is possible to measure the dose contribution due to such component. To do this, two gel dosimeters, one made with standard gel and the other made with gel added with a chemical compound containing this isotope, are irradiated one near the other in the phantom. Because the photon field is substantially the same for both dosimeters, the dose coming from the added isotope can be obtained from the comparison between the images of the two gel dosimeters. A standard gel added with urea (to correctly simulate the adult brain tissue it is necessary 2.2% of nitrogen) allows to discriminate the dose contribution due to the  $^{14}\text{N}(\text{n,p})^{14}\text{C}$  reaction. A standard gel added with sodium tetraborate decahydrate ( $\text{Na}_2\text{B}_4\text{O}_7 \cdot 10\text{H}_2\text{O}$  in the proper amount to have 35 or 40 ppm of  $^{10}\text{B}$ ) permits to determine the radiotherapeutic dose. Furthermore, thermal neutron fluxes can be calculated utilising the  $^{10}\text{B}$  dose and the corresponding kerma factors.

Moreover, irradiating a couple of standard gel, one made with light water and the other with heavy water, it is possible to deduce the fast neutron dose. Such a dose is obtained by means of algorithms that take into account the dose owing to neutron elastic scattering on protons in light water and on deuterons in heavy water<sup>(3),(4)</sup>.

Each gel preparation is preliminarily calibrated with  $^{137}\text{Cs}$   $\gamma$ -rays, firstly because gel dosimeters are hand-made and little weighing errors during preparation are unavoidable, and secondly because each kind of gel has a different composition which influences its own sensitivity.

The gel-response dependence on the LET of the charged particles emitted in a reaction has been studied, too. As in the most part of dosimeters, gel sensitivity decreases when LET increases. In particular, measurements have been done to determine the gel sensitivity to  $\alpha$  particles and  $^7\text{Li}$  ions, emitted in the  $^{10}\text{B}(\text{n}, \alpha)^7\text{Li}$  reaction, reported to  $\gamma$ -rays sensitivity: a value included between 0.40 and 0.45 has been found. Values found in literature have been utilised for protons and deuterons.

## TL dosimetry

Beside the procedures which concern gel dosimeters, an other and independent method has been studied and experimented<sup>(3)</sup>. The method is uniquely based on TL detectors and, also by this approach, separation of dose contributions due to secondary radiation has been obtained.

When irradiated in polyethylene phantom, the TLDs absorb the  $\gamma$ -dose coming from  $^1\text{H}(n,\gamma)^2\text{H}$  in-phantom reactions and reactor background; moreover, they absorb doses from the charged particles emitted in the neutron reactions occurring in the dosimeters themselves. This energy release is dependent on the dosimeter isotopic composition and proportional to thermal neutron fluence. As the consequence, if the TLDs have been previously calibrated, the thermal neutron fluence can be evaluated, after subtraction of the  $\gamma$ -dose contribution, if necessary. Regarding the fast neutron component of therapy beams, the energy is released mainly by elastic scattering with nuclei, more probable for light nuclei that is for hydrogen. Therefore, in non-hydrogenous materials, the released energy is very low; and then, the absorbed dose is not measurable, because negligible compared to the other doses. In conclusion, with TLDs it is possible to determine the  $\gamma$ -dose and the thermal neutron fluence. From this fluence, knowing the kerma factors, the doses, due to all the charged particles generated in nuclear reactions of thermal neutrons, can be easily calculated.

The proposed method takes advantage of the combination of two types of TL materials, which have a different sensitivity regarding thermal neutrons and  $\gamma$ -component of radiation field:  $\text{CaF}_2:\text{Tm}$  has very poor sensitivity to low energy neutrons;  $\text{LiF}:\text{Mg,Ti}$  has thermal neutron sensitivity which is based on reactions  $^6\text{Li}(n, \alpha)^3\text{H}$ ,  $\sigma = 940$  barns<sup>(5)</sup>; the ratio thermal neutron to gamma radiation sensitivity of lithium fluorides depends on different  $^6\text{Li}$  percentage whose these dosimeters are formed<sup>(6)</sup>.

In this study, the following dosimeters have been utilized: TLD-300 ( $\text{CaF}_2:\text{Tm}$ ); TLD-600 ( $^6\text{LiF}:\text{Mg,Ti}$  -95.6% $^6\text{Li}$ ); TLD-700 ( $^7\text{LiF}:\text{Mg,Ti}$  -99.9% $^7\text{Li}$ ); TLD-100 ( $\text{LiF}:\text{Mg,Ti}$  -7.5% $^6\text{Li}$ ), produced by Harshaw Chemical Company, all in form of chips of 3.1 3.1 0.9 mm<sup>3</sup>. The TLD small size allows to obtain punctual measurements and a mapping both of  $\gamma$ -dose distribution and thermal-neutron fluence in phantom, without affecting neutron transport. Before each irradiation, all dosimeters have been annealed following the recommendation of the producer: TLD-300 at 400°C for 1 h; lithium fluorides at 400°C for 1 h, and then at 100°C for 2 h.

TL emission has been detected with laboratory-made instrumentation. Each detector has been characterised with its intrinsic sensitivity, by  $^{137}\text{Cs}$   $\gamma$ -radiation and the glow curves have been stored. Such calibrations have been employed during all the experiments for normalising each result, in order to reduce dispersion. Some subsequent calibrations have been performed, aimed to enquire possible radiation damage effects induced by neutrons.

TLD-300 has been chosen in this work, because it has shown to be an interesting candidate for gamma dosimetry in thermal or epithermal neutron fields, because its sensitivity to neutrons decreases with energy decreasing<sup>(7)</sup> and its sensitivity to  $\gamma$ -radiation is very high, almost one order higher than that of  $\text{LiF}:\text{Mg,Ti}$ . Therefore, the couple of this calcium fluoride, for gamma dose measurements, and lithium fluorides, for thermal neutron fluence determination, allows to determine the separation doses.

To deduce thermal neutron fluence from TLD-100 and TLD-700, the  $\gamma$ -dose contribution has been subtracted utilising the gamma calibration of the dosimeter itself and the dose value measured with a TLD-300 in the same position of the phantom. For TLD-600,  $\gamma$ -dose subtraction is not necessary, because in such detector this dose is negligible in comparison with thermal neutron dose. So, the lithium fluorides response, depleted of  $\gamma$ -contribution, is due only to the thermal neutrons whose fluence can be evaluated by means of calibration coefficient.

Care has been taken in the choice of exposition time and reactor power, in order to avoid radiation damage effects<sup>(8)</sup> (loose linearity) in lithium fluorides, and also to keep the  $\gamma$ -dose level in the region of  $\text{CaF}_2\text{:Tm}$  linearity response.

## Results

In figures 3 and 4, some experimental results, obtained by the two mentioned methods, are shown. The absorbed dose in the central axis of the phantom, exposed in the epithermal column, is reported in figure 3: dose profiles obtained with gel dosimeters have been compared with TL measurements. In figure 4, the thermal flux, measured with gel and TL dosimeters in the central axis of the phantom exposed in the thermal column, are shown. In the figure, also the results obtained by means of activation foils are reported, in order to check the validity of the obtained results. The consistency of all results is a confirmation of the validity of both the proposed methods for absorbed dose separation.

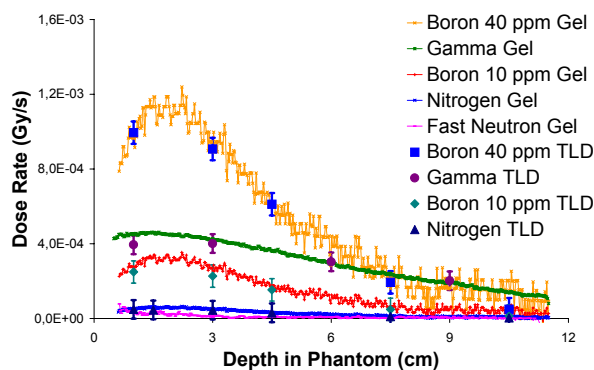


Fig.3. In-phantom absorbed dose profiles.

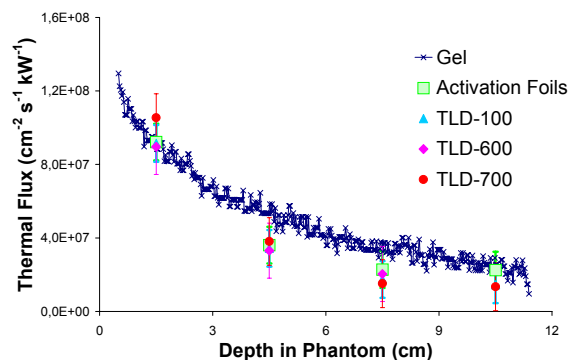


Fig.4. In-phantom thermal flux.

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