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DOSIMETRY METHODS IN BORON NEUTRON CAPTURE THERAPY

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Abstract

Dosimetry studies have been carried out at thermal and epithermal columns of LVR-15 research reactor for investigating the spatial distribution of gamma dose, fast neutron dose and thermal neutron fluence. Two different dosimetry methods, both based on solid state detectors, have been studied and applied and the accuracy and consistency of the results have been inspected. One method is based on Fricke gel dosimeters that are dilute water solutions and have good tissue equivalence for neutrons and also for all the secondary radiations produced by neutron interactions in tissue or water phantoms. Fricke gel dosimeters give the possibility of separating the various dose contributions, i.e. the gamma dose, the fast neutron dose and the dose due to charged particles generated during thermal neutron reactions by isotopes having high cross section, like 10B. From this last dose, thermal neutron fluence can be obtained by means of the kerma factor. The second method is based on thermoluminescence dosimeters. In particular, the developed method draw advantage from the different heights of the peaks of the glow curve of such phosphors when irradiated with photons or with thermal neutrons. The results show that satisfactory results can be obtained with simple methods, in spite of the complexity of the subject. However, the more suitable dosimeters and principally their utilization and analysis modalities are different for the various neutron beams, mainly depending on the relative intensities of the three components of the neutron field, in particular are different for thermal and epithermal columns.

Keywords: BNCT, Neutron dosimetry; Fricke gel; TLD

1.- INTRODUCTION

Dose measurements in boron neutron capture therapy (BNCT) require special methods, due to the complexity of neutron interactions, to the very high neutron fluxes (the desirable minimum beam intensity would be 10^9 epithermal neutrons cm^{-2} s^{-1}) and finally to the need to separate dose components with different biological efficacy.

In tissue exposed to epithermal neutron beams, the reactions mainly responsible for the absorbed dose in tissue are those with hydrogen and nitrogen: $^{1}H(n,\gamma)^{2}H$ (σ = 0.33 b), whose γ-rays of 2.2 MeV can travel many centimetres through tissue, and $^{14}N(n,p)^{14}C$ (σ = 1.81 b), whose emitted protons of about 0.6 MeV have short range in tissue, giving local dose deposition. The fast neutron component of epithermal neutron beams, gives often a not negligible contribution to the absorbed dose, mainly due to elastic scattering with hydrogen nuclei. If the isotope ^{10}B is selectively accumulated in tumor tissue, the reaction with thermal neutrons $^{10}B(n,\alpha)^{7}Li$ (σ = 3837 b) causes localized energy absorption in cancerous cells; this is exploited by BNCT.

Methods based on laboratory-made Fricke-Xylenol-Orange gel dosimeters having different geometries have shown remarkable potentiality for in-phantom or in-free-beam dose measurements, with separation of the different dose contributions [Bartesaghi *et al.*,2009; Gambarini *et al.*, 2007; Gambarini *et al.*, 2010a; Gambarini *et al.*,2011]. Moreover, at BNCT thermal/epithermal neutron fluence levels, methods based on the analysis of the glow curve of LiF thermoluminescence detectors (TLDs) allow the mapping of both gamma dose and thermal neutron fluence [Gambarini *et al.*, 2010b; Gambarini *et al.*, in print].

The aim of this work was to investigate both the advantages and confidence limits of the methods and infer conditions and suitability of their employment.

2.- MATERIALS AND METHODS

Measurements were carried out at the BNCT facility of the LVR-15 research reactor of the Research Centre Řež (Czech Republic). This epithermal column is now dedicated to the development of appropriate dosimetry methods and also utilized for the study of physical and biological aspects of BNCT [Burian *et al.*, 2009]. The collimator exit has circular shape, with 12 cm of diameter. For biological experiments, in which typically small volumes have to be exposed to thermal neutrons, a polyethylene disk 2 cm thick is frequently inserted at the collimator exit, to moderate neutron energy. Both measurements in water-phantom and in free-beam were performed. The facility is provided with a laser system for centering the samples on the beam axis. In Figure 1, an example of exposure at this column is shown.

Figure 1. Fricke gel dosimeter placed at the exit of the BNCT column collimator. Behind the dosimeter, the polyethylene moderator is dimly seen.

All exposures to photons, aimed at testing dosimeter behavior and achieving calibration, have been performed at the Fondazione IRCCS Istituto Nazionale Tumori of Milano (Italy). The irradiations of gel dosimeters were made with a ^{137}Cs source, at a dose rate of about 0.126 Gy/s. The calibration of TLDs was attained by means of the ^{60}Co source of a

radiotherapy facility, with uniform dose rate of 0.102 Gy/min. For calibration, both dosimeters were placed in a suitable TE phantom.

2.1.-Fricke gel dosimeters

Fricke gel dosimeters in form of layer are laboratory-made radiochromic gels consisting of a Fricke solution incorporated into a gel. For neutron irradiation, the more suitable gelling agent is porcine skin. The standard composition of the prepared dosimeters is: ferrous sulphate [1mM $Fe(NH_4)_2(SO_4)_2 \cdot 6H_2O$]; sulphuric acid [25mM H_2SO_4]; Xylenol Orange [0.165mM$C_{31}H_{27}N_2Na_5O_{13}S$]; porcine skin [3% of the final weight].

Before gelling, the solution is inserted, with a syringe, into suitable supports consisting of a circular frame 3 mm thick, closed between two transparent polystyrene sheets 1 mm thick. The gel dosimeters prepared for the described experiment were of various diameters, from 12 cm to 16 cm.

Fricke gel dosimeters are radiochromic: before irradiation they show light absorption cantered at 430 nm, and after irradiation an absorption peak centered at 585 nm appears. The absorbed dose is proportional (up to saturation) to the light absorbance around 585 nm. The dosimeter analysis consists in detecting, before and after irradiation, the images of optical transmittance at 585 nm. The variation of dosimeter absorbance is obtained evaluating the difference in optical density Δ(OD) that is obtained by pixel-based analysis of such two images. The grey-level (GL) images of light transmittance are acquired with a CCD camera provided with a band-pass filter around 580 nm. A proper software in MATLAB® code has been developed in order to perform a pixel-to-pixel elaboration and obtain a matrix of Δ(OD). By means of calibration with a reference photon source, the Δ(OD) images are converted into dose images. For the analysis, the dosimeters are placed on a plane uniform source of white light. This instrumentation for the dosimeter analysis is transportable and it was located at Research Centre Řež, in order to perform optical analysis just before and after irradiation. In the optimized protocol, dosimeters are imaged 40 minutes after irradiation, in order to reach chemical equilibrium and to avoid sensible effects of ferric ion diffusion in the dosimeter.

When a dosimeter with the described standard composition is exposed to a thermal or epithermal neutron beam, the absorbed dose is due to gamma radiation (from reactor or from the reactions of thermal neutrons with hydrogen in the phantom) and to the fast neutron component, if it is not negligible. In order to separate such two dose contributions, some dosimeters were prepared using heavy water D_2O instead of H_2O. In fact, the fast neutron dose in water is mainly due to recoil protons and in heavy water it is due to recoil deuterons. So, if such two dosimeters are exposed in the same geometry of irradiation, by pixel-to-pixel analysis of the dose images obtained with these dosimeters the fast neutron dose image is obtained. To attain this result, proper algorithms must be used . In fact, it is necessary to evaluate the ratio of the energy released by recoil protons and recoil deuterons and also the sensitivity of the gel dosimeter to these two charged particles. Both protons and deuterons release their energy mainly at the end of their path (Bragg peak) where the linear energy transfer (LET) is very high. The response of gel dosimeters decreases at increasing of LET, and the information regarding this sensitivity reduction are scarce. Concerning the sensitivity to recoil protons, we have taken a reduction value of 0.85 as obtained by Bäck et al. [1999]. In fact, the height of a Bragg peak measured with gel dosimeters (calibrated with photons) has resulted to be 85% lower. No experimental data about Fricke gel sensitivity to deuterons were available in literature. Therefore, some general considerations regarding Fricke dosimeter sensitivity dependence on radiation LET have been performed and a sensitivity to deuterons with respect to photons of 55% has been chosen. A better determination of such sensitivity coefficients could be of great advantage for the precision of the obtained dose values.

The boron dose is obtained with the aid of dosimeters containing a suitable amount of ^{10}B. This was obtained by adding, to the Fricke solution, a suitable amount of the compound Sodium Tetraborate Decahydrate [$B_4Na_2O_7 \cdot 10H_2O$], to obtain a concentration of 40 μG/g of ^{10}B. In such a dosimeter, the absorbed dose is the gamma dose, as in standard gel dosimeter, plus a dose contribution due to the α and ^{7}Li particles generated by the reaction with ^{10}B. Therefore the boron dose can be evaluated by pixel-to-pixel elaboration of the dose images obtained with a standard and a borated gel dosimeter, exposed in the same configuration. Owing to the high LET of such charged particles, the reduction of gel sensitivity to α and ^{7}Li particles must be introduced in the algorithms. For the relative

response to such charged particles with respect to photons the coefficient 0.4 has been assumed, obtained in a previous experiment carried out, at another research reactor, with the same kind of dosimeters.

2.1.-Thermoluminescence detectors

The utilized thermoluminescence dosimeters were LiF:Mg,Ti: TLD-700 (chips of 3.1 · 3.1 · 0.9 mm^3 from the Harshaw Chemical Co). In these dosimeters the contribution of fast neutrons is negligible. On the opposite, both gamma rays and thermal neutrons give a measurable contribution to the dosimeter response. A method was developed to achieve both photon dose and thermal neutron fluence from the glow curve (GC) of such dosimeters, exploiting the different ratio of the heights of the two dosimetric peaks. The advantage of utilizing TLD-700 in thermal neutron beams of very high flux, as is the case of reactors for which the power is fixed and cannot be lowered, comes from the fact that TLD-700 contains only a very low amount of ^{6}LiF (≈ 0.01%) and then, in spite of the high cross section of ^{6}Li for the reaction $^6Li(n,\alpha)^3H$, it does not undergo radiation damage, as happens for TLD-600 and TLD-100 that show an irreversible reduction of sensitivity [Piesch*et al*., 1978]. Moreover, the TLD-700 response does not depend on the chip orientation, differently from the response of TLD-600 and TLD-100.This property is of great importance in epithermal neutron dosimetry, because neutron anisotropy continuously increases with their thermalization. For BNCT dosimetry, very often TLD-700 detectors are utilized to measure the gamma dose. In this case, it is necessary to subtract the contribution of thermal neutrons to their response [Aschan *et al*., 1999; Burgkhardt *et al*., 2006]. In order to evaluate such a contribution, other measurements are necessary, usually performed with activation foils. The method that has been developed to map both photon dose and thermal neutron fluence using only TLDs-700, has the advantage of requiring only dosimeter calibration and no other specific measurements.

Before each irradiation, the TLDs were annealed (heated at 400°C for 1 h, then left at 100°C for 2 h and finally quenched, with gradual cooling, down to room temperature). All readouts were performed waiting at least 5 days after irradiation to allow the decay of low temperature peaks. The GCs were acquired with a Model 3500 TLD reader from

Harshaw/Bicron. The adopted reader modality was: preheating at 120 °C for 2 seconds, acquisition from 120 °C up to 350 °C for 40 seconds, with a heating rate of 7 °C per second. To achieve higher precision, each dosimeter was characterized with its own calibration coefficient. In order to avoid problems due to superlinearity effects, dosimeter calibrations with ^{60}Co were performed by irradiating the TLDs in a dose interval giving TL emissions, after gamma irradiation, with about the same intensities obtained in the experiment with neutrons.

In the proposed method, the gamma dose is evaluated from the TLD-700 GC with the following formula:

$$D_g = \frac{h_1 - h_2 \times R_n}{h_1^* - h_2^* \times R_n} \quad (1)$$

where h_1 and h_2 are the heights of the first and second peaks of the measured GC, h_1^* and h_2^* are the heights of the first and second peaks of the gamma calibration GC, normalised to dose unit and R_n is the ratio between the heights of the first and second peaks in a LiF:Mg,Ti TLD exposed to thermal neutrons. Some observations on this ratio R_n are given in the previous article [Gambarini *et al.*, in press]. In the present work, the value R_n = 1.7 was used.

This algorithm (1) could be used also for separation of gamma dose contribution in the response of TLD-100. As will be evident from the reported results, the gamma contribution, in the case of BNCT beams, is a very low percentage of the total response in such dosimeters and therefore the error in gamma dose determination would be unacceptably high. If exposures at fluences not so high to get close to the dosimeter damage are possible, the second dosimetric peak of TLD-100 GC can give good information about thermal neutron fluence, without any subtraction of gamma dose contribution. Some results are reported below.

In addition to the measures at the BNCT column, some measurements were carried out at the horizontal thermal column HK1 of the same reactor. Here the relative contributions of photons and thermal neutrons in the beam are very different with respect to those in the moderated beam of the BNCT column. This peculiarity gives way to draw useful deductions about the conditions of reliability of the proposed and applied method.

In Figure 2, the collimator exit, with an array of TLD chips, is shown.

Figure 2. TLD dosimeters placed at the exit of the HK1 collimator for irradiation.

In order to avoid dirtying TLDs, for every exposure the chips were wrapped in Teflon film and then covered with Parafilm®M.

3.- RESULTS

Various measurements were carried out at the BNCT column, with Fricke gel dosimeters having the composition above described: standard, borated and made with heavy water. Dosimeters were placed in contact with the polyethylene moderator. From the light transmittance images acquired, before and after irradiation, with the CCD camera, the Δ(OD) images were obtained and, utilising the calibration factor for each dosimeter, dose images have been attained. By mean of pixel-to-pixel elaboration of such images, the images of gamma, fast neutron and boron doses have been achieved and finally central dose profiles have been extracted in order to compare the results with those obtained with TLDs. TLD-700 and TLD-100 chips, kept between two polystyrene sheets of 15 mm x 130 mm with a thickness of 1 mm, were fixed to the polyethylene moderator along a diameter. The

gamma dose was attained with the formula (1). The dose rates were then compared to those obtained with gel dosimeters. An example of results is reported in Figure 3.

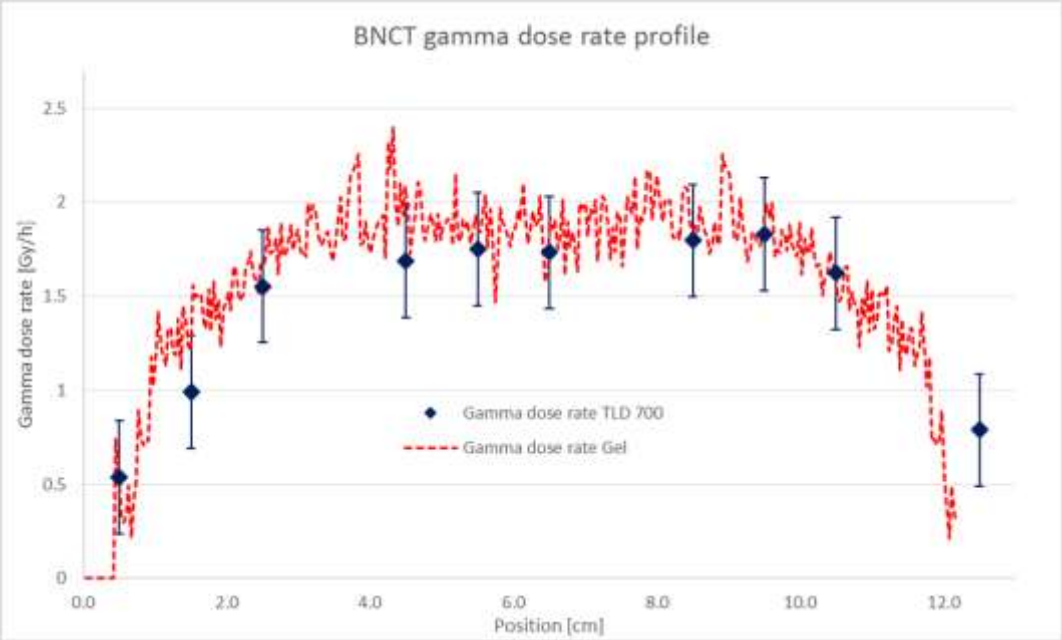

Figure 3. Gamma dose rate profile extracted from a dose image obtained with gel dosimeters and gamma dose rate values obtained from the TLD-700 GC with formula (1).

From the boron dose images, thermal neutron fluence images were obtained by means of the kerma factor.

In order to better understand the TLD behavior in neutron fields like those of these reactor columns, thermal neutron fluence rates along a beam diameter were extracted from the images and compared with parameters got from TLD results. To clarify these considerations, the results concerning 4 TLDs are here reported: a couple of TLD-700 and TLD-100 chips (named A8 and b7 respectively) exposed, near each other, in a central region of the polyethylene moderator of the BNCT beam and a similar couple (named C3 and a6) exposed in the central region of the HK1 collimator exit. In Figure 4, the GCs of TLDs-700 and TLDs-100 are reported. In the figure is inserted a curve obtained from the γ-calibration curve per Gy multiplied by the γ-dose value obtained by (1). This curve represents the contribution of photons to the dosimeter GC. This curve was subtracted from

the initial GC, giving so a curve, shown in the figures, that represent the contribution of thermal neutrons in the dosimeter response. If the gamma dose obtained with the formula (1) is correct, in the curve representing the thermal neutron contribution, the ratio R_n of the heights of the two dosimetric peaks has to be near to the value 1.7.

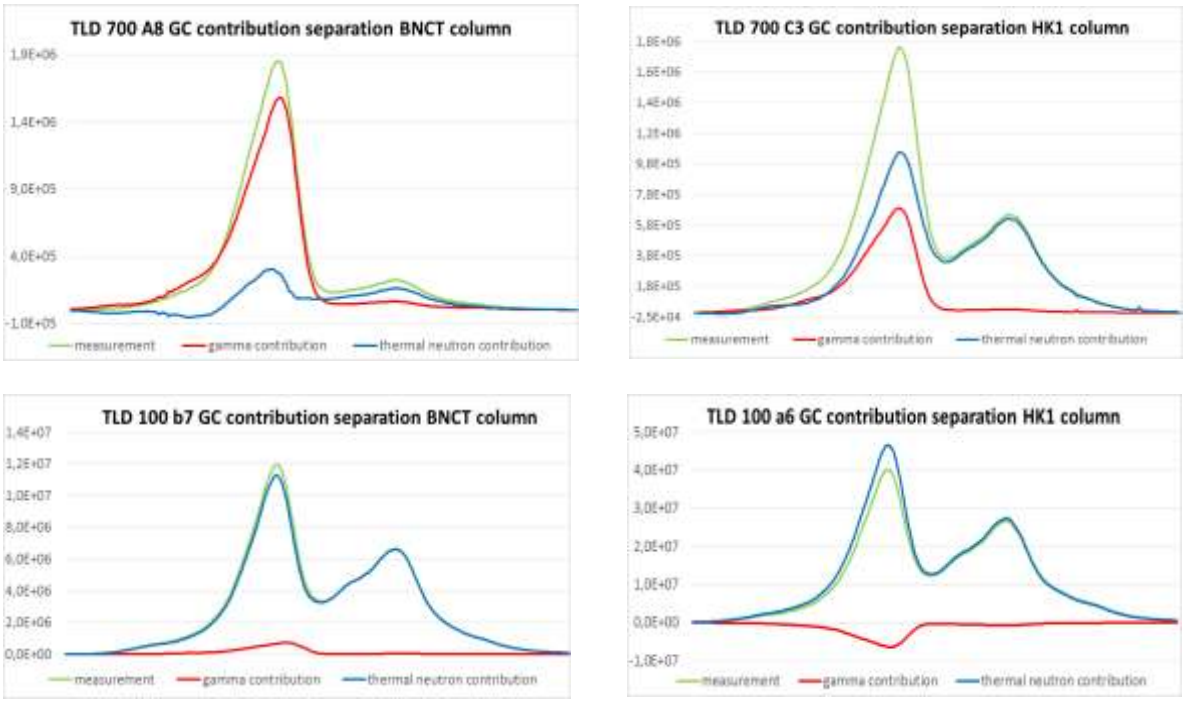

Figure 4. GCs of TLD-700 and TLD-100 irradiated at the two LVR-15 columns.

From Figure 4 it is evident that in the two columns the relative contributions of γ-dose and thermal neutron fluence are very different: in the moderated beam of BNCT column, the γ-dose contribution is predominant and therefore the shape of the curve concerning thermal neutrons is not very well shaped, even if acceptable. Better shapes of all the TLD-700 curved are obtained with the HK1 beam, where the thermal neutron contribution in the GC is predominant and is obtained with good values of the ratio R_n. Coherently, if we make in the same way the separation of contributions in the GCs of TLDs-100, where the thermal neutron contribution is predominant, the gamma dose cannot be reliably obtained: in HK1 beam the γ-dose is lower than the measurement error and therefore from formula (1) a negative value can result. This is no surprising if we take into account that in some cases, in particular for TLD-100, the numerator in formula (1) results to be the difference of two

very similar values. To get a wider panorama of the situation, some significant parameters concerning the same TLDs of Figure 4 are reported in Table. 1.

Table 1.- Peculiar parameters of TLD-700 and TLD-100 irradiated at the two LVR-15 columns.

TLD		h_1	$h_2 \cdot Rn$	Relative difference $(h_1-h_2 \cdot Rn)/h_1$	h_1/h_2
100	b7 BNCT column	11 999 279	11 333 650	0.0555	1.800
	a6 HK1 column	40 142 968	45 413 703	-0.1313	1.503
700	A8 BNCT column	1 855 892	380 309	0.7951	8.296
	C3 HK1 column	1 740 427	1 096 063	0.3702	2.699

In order to evaluate the thermal neutron profile in BNCT column using TLD dosimeters, it is useful to consider the height of the second peak of the neutron contribution in the GC. From the data reported in Figure 4, it is evident that the second peak is more sensitive to thermal neutrons rather than to γ-rays and it is also less sensitive to possible shift in peak position general due to instability of heating rate during reading. Therefore, the evaluation of the thermal neutron contribution using second peak is more reliable than using the first one.

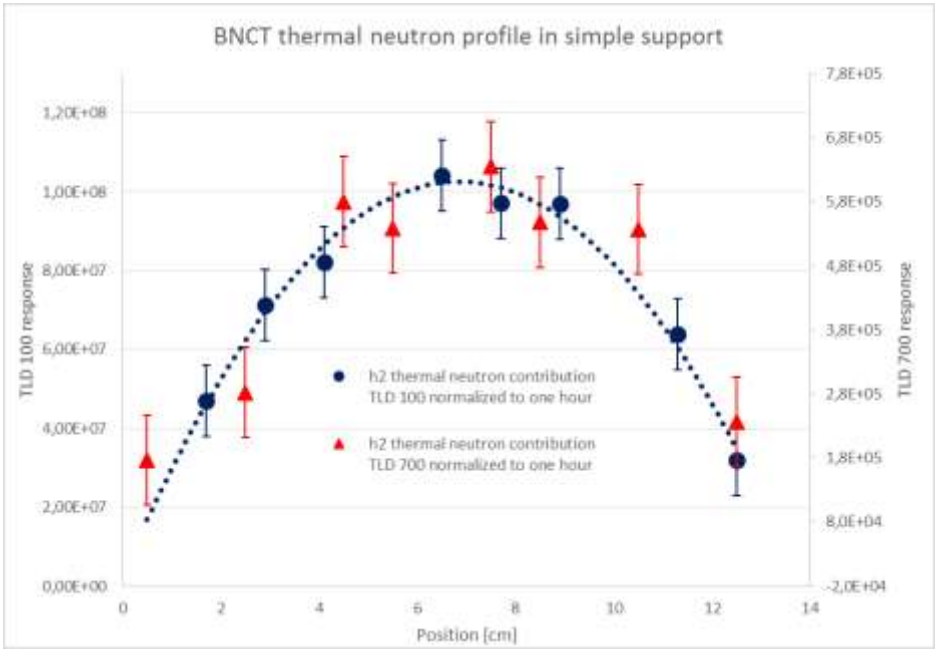

Figure 5. Thermal neutron profiles obtained using the height of the second peak of neutron contribution curve for TLD-700 and TLD-100.

In Figure 5, thermal neutron profiles, obtained using the second peak of neutron contribution curves for TLD-100 and TLD-700, are reported. While both profiles follow a similar trend, it could be noticed that the TLD-700 profile spread is higher than the TLD-100 one. This behavior can be explained by observing that the second peak of γ-contribution in TLD-100 is negligible thanks to the very high sensitivity of TLD-100 to thermal neutrons while in TLD-700 the thermal neutron contribution is very low in a mixed γ-neutron beam like that of the moderated BNCT beam, where the γ-component is relatively high.

The profile attained with TLD-100 can be considered satisfactory also because it is very similar to the thermal neutron profile got with Fricke gel, as shown in Figure 6.

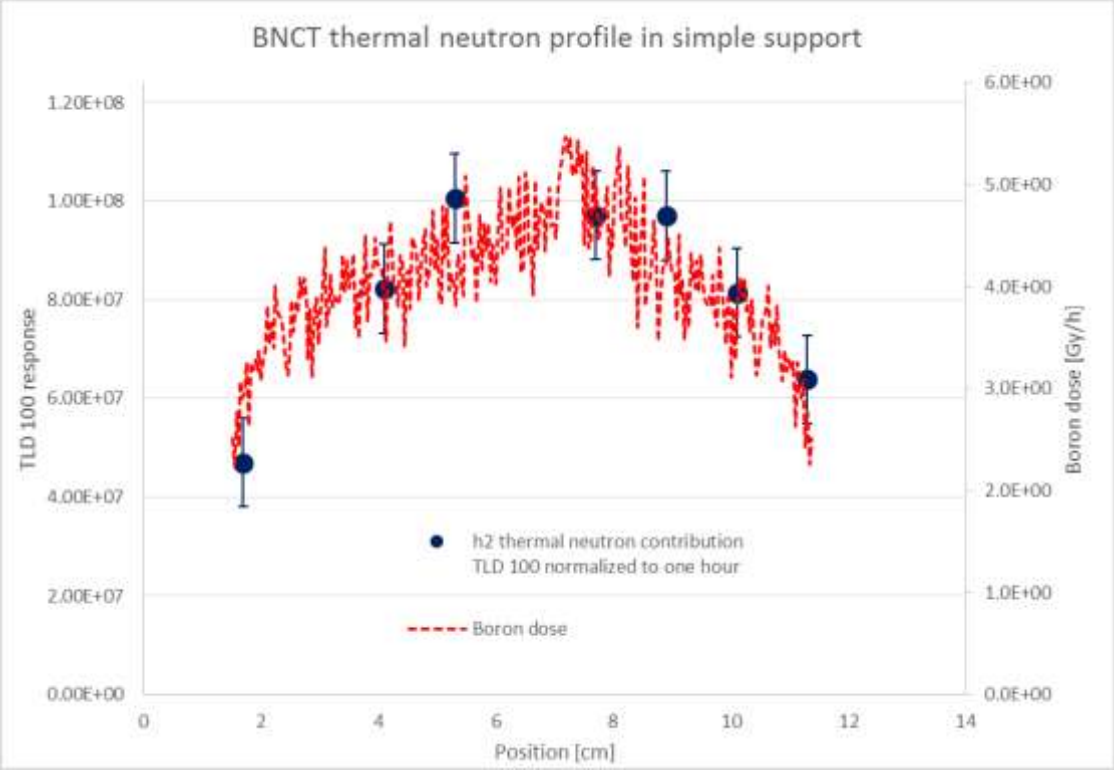

Figure 6. Thermal neutron profiles obtained with Fricke gel dosimeter and with second peak of neutron contribution curve for TLD-100.

4.- DISCUSSION

Gel dosimeters have shown to be a valid tool to get spatial information of absorbed dose in BNCT separating various dose components. Good values of γ-dose can be attained with good dosimeter calibration, mainly if the fast neutron dose contribution is not much high with respect to photons, as usually happened in BNCT beams. The determination of fast neutron dose component could be bettered with an improved knowledge of gel sensitivity dependence on radiation LET. Concerning boron dose and thermal neutron fluence, some considerations could be significant about the error of the obtained values. In experimental situation like this experiment (gel dosimeter transported and used in different days after preparation) the dosimeter response error was estimated of about 10%. This leads to an error of about 30% in the determination of the γ-equivalent boron dose. Assuming a probable error of 50% of the coefficient 0.4, the relative uncertainty on the boron dose results about 58%. So, the main contribution of the boron dose error is due to the uncertainty that is assigned to this coefficient 0.4, therefore it is important to determine this coefficient with better accuracy to improve the precision of the obtained dose values. Through the boron dose is possible calculate the thermal neutron fluence, using the kerma factor. In this last step is introduced an additional error due to the uncertainty the weighted amount of the boron compound added to the solution for the preparation of boron-containing dosimeters. The boron content of gel dosimeter could be tested by means of dosimeter calibration with thermal neutrons, practically impossible.

Concerning TLDs, it is evident form the reported results that good γ-dose values can be achieved from TLD-700 using formula (1).

In order to make some considerations on how to get thermal neutron profiles, it is necessary to pay attention to the thermal neutron fluences of TLD irradiation. In the case of TLDs of Figure 4, the fluence was of about 5.52 10^{10} cm^{-2} for a6, 8.6 10^{10} cm^{-2} for b7, 4.14 10^{11} cm^{-2} for C3 and 3.25 10^{11} cm^{-2} for A8.

While for TLD-700 these values are far below the damage limit (which is over than 10^{13} cm^{-2}) and also well contained in the linearity region, for TLD-100 the above reported values are very near to the damage limit and possibly out of the linearity region of the first peak height. This aspect is also evident in the data reported in Table 1, where one ratio h_1/h_2 is 1.503: this value is too low and indicates a reduction of the first peak height; consequently, the separation of contributions results in a negative gamma dose. Despite this, the second peak seems to maintain linearity to thermal neutron fluence, so they can be used to evaluate a thermal neutron profile also without proceeding with the separation method.

5.- CONCLUSIONS

Considering the consistency of the results obtained with Fricke gel dosimeters and TLDs, the goodness of the gamma dose measurements with whichever method is confirmed. Concerning thermal neutron fluences, also TLD-700 and/or TLD-100, if calibrated with thermal neutron, could be a good method to evaluate the thermal neutron fluences.

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