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Improved dosimetry for BNCT by activation foils, modified thermoluminescent detectors and recombination chambers

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Abstract Boron neutron capture therapy (BNCT) is an experimental therapy of selected tumours, based on a nuclear reaction initiated by the capture of thermal neutron by the ^{10}B nucleus. After ^{10}B had been delivered selectively to tumour cells, it can be activated by neutrons to deliver locally lethal high-LET radiation. BNCT beams are complex mixed radiation fields, because of broad neutron energy range, presence of gamma contamination and necessity of precise determination of several dose components. The paper presents some results of the research project on BNCT dosimetry with activation foils, recombination chambers and TL detectors.

Key words BNCT • recombination chambers • TLD • activation foils

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Introduction

Boron neutron capture therapy (BNCT) is a binary bio-targeted therapy, considered as a possible way to cure some kinds of malignant tumours, which up to now cannot be successfully treated in any other way. Physical concept of the method is based on nuclear reaction that occurs when a nucleus of boron (^{10}B) captures thermal neutron, producing an α particle and Li ion:

$$^{1}_{0}n + {}^{10}_{5}B \rightarrow {}^{7}_{3}Li + {}^{4}_{2}\alpha\,.$$

The therapy is based on physiological rather than physical, targeting of radiation. This is achieved by the administration of boron-containing compounds which have the property to accumulate selectively in the tumour tissue and much less in the immediate surrounding.

Following the accumulation of boron, the tumour area is irradiated with low-energy (epithermal) neutrons produced by a nuclear reactor. The neutrons slow down in tissue to thermal energies in the tumour area. Some of the boron atoms absorb thermal neutrons and then, in a very short time, release two heavy charged particles (^{7}Li and ^{4}He) that dissipate most of their energy within the volume of a single cell. This binary process damages the involved cell, while both boron drug and thermal neutrons alone are to some extent innocuous to tumour and normal tissues. Because the boron concentrates in the tumour cells, the cancer can be destroyed, while the normal cells nearby receive the acceptable radiation dose.

At present, BNCT is experimentally used for treatment of glioblastoma. Clinical trials of BNCT were initiated at few reactors in Europe, USA and Japan. Phase II studies

showed [4] that the treatment was tolerated well and that the median survival was better than available best care. Results of these studies were promising enough to promote a decision about construction of a BNCT beam line at the Polish research reactor MARIA at the Institute of Atomic Energy at Świerk.

It has been generally recommended that for the BNCT to be successful, a thermal neutron fluence of about 5×10^{12} n cm^{-2} should be delivered to a tumour with a ^{10}B concentration of 30 μg per gram of tissue. For clinical trials, it is thought that epithermal neutrons (neutron energies between 1 eV and 10 keV) are an optimum for the treatment. Epithermal neutrons thermalize at a depth of about 2.5 cm. Therefore, they can provide a maximum thermal neutron flux density at the tumour site with a minimum damage to normal tissue. Production of sufficient dose of epithermal neutrons, with acceptably low background of fast neutrons and of gamma radiation, requires special nuclear reactor features. As a consequence, the construction of the BNCT facilities is justified only at some of the existing reactors. Some reactors can be adapted for BNCT by the use of fission converters. This is also the case of the Polish research reactor MARIA. The technical concept of the BNCT facility was elaborated at the Institute of Atomic Energy. At present, the facility is under construction [7].

This also created a need for development of dosimetric systems for characterization and monitoring of the beam. This work presents a number of improvements of the techniques of dosimetric measurements, which can be of interest for dosimetry of BNCT beams.

Detectors and methods

Non-dilute activation foils – correction for neutron self-shielding

Activation foils are commonly used for dosimetry of epithermal neutrons, which constitute the most important component of the BNCT beam. The neutrons initiate nuclear reactions in the foils, associated with emission of gamma rays from the activated foils. The use of an appropriate set of foils, having different cross sections for nuclear reactions, depending on neutron energy, $\sigma(E)$, makes it possible to reconstruct the incident neutron spectrum based on the measured activities of individual foils and techniques of mathematical deconvolution. One of the important difficulties in this method is caused by the effect of neutron self-shielding within the foil, especially in the neutron energy region associated with a strong resonance absorption. The effect can be reduced by the use of dilute activation foils which are commonly used for dosimetry within nuclear reactors, but their applicability for BNCT is rather limited. Therapeutic neutron beams have flux densities of the order of $10^8 \div 10^9$ cm^{-2}s^{-1}, that is not enough for dilute detectors, so the non-dilute detectors should be used.

Reaction rates α_i, evaluated from the measured activity of non-dilute foils of nuclide i, can be expressed as:

$$\alpha_i = \int_0^\infty \sigma_i(E) G(E) \varphi(E) dE \tag{1}$$

where: $\sigma_i(E)$ – microscopic cross section for the reaction of nuclide i with a neutron of energy E; $\varphi(E)$ – neutron flux density per unit energy; $G(E)$ – self-shielding coefficient. Integration covers the whole neutron energy range. The self-shielding coefficient $G(E)$ falls within the limits $0 \div 1$. When the self-shielding effect does not appear or can be neglected, the coefficient $G \approx 1$.

For typical surface densities of foils of $20 \div 100$ mg/cm^2, the self-shielding effects are important for cross sections greater than 10 barns. In the case of threshold reactions with fast neutrons (e.g. ^{27}Al(n,α)^{24}Na or ^{58}Ni(n,p)^{58}Co), the cross sections are relatively low and self-shielding is not-measurable.

Standard computer codes applied for deconvolution of neutron spectra (e.g. SAND-II [2]) do not take into account any effects of self-shielding. The self-shielding effect refers to the neutron field within the foil, but from the point of view of reaction rate (1) it does not matter whether the factor $G(E)$ modifies the flux density or the respective cross section. Therefore, replacing the original library cross sections with the modified data seems to be the simplest method to include the self-shielding effect to the SAND-II code. The point of such modification is that pointwise SAND-II library cross section is replaced by:

$$\sigma_i^{\mathrm{mod}}(E) \equiv \sigma_i(E) G(E). \tag{2}$$

The energy dependence of self-shielding coefficients $G(E)$ was calculated using a simple, one interaction event model [1]. The respective self-shielding coefficients for monoenergetic are given by eqs. (3) and (4) for isotropic radiation field and parallel beam, respectively.

$$G = \frac{1 + x\, Ei(-x) - \exp(-x) \cdot (1 - x)}{2x} \tag{3}$$

$$G = \frac{1 - \exp(-x)}{x} \tag{4}$$

where: x – optical thickness of the foil; $Ei(x)$ – exponential integral function.

Corrected cross sections have been included to SAND-II library and used for the evaluation of neutron spectra.

Modified thermoluminescent detectors

Thermoluminescent detectors (TLD's) are widely used in dosimetry of radiotherapy beams due to such properties like high sensitivity, wide range of measured doses, no need of power supply and small dimensions enabling locating of TLD's inside phantoms. The main problem in application of TLD's in dose measurements for BNCT is a difficulty in separating between thermoluminescent (TL) signals caused by neutron and gamma components. This is usually attempted through application of TLD's with different enrichment in ^{6}Li and ^{7}Li, these isotopes showing extremely different cross sections for reactions with low energy neutrons. The ^{6}Li cross section for the n(^{6}Li, T)α reaction is equal to about 950 barns for thermal neutrons. For higher energy, it decreases with increasing energy, proportionally to $E^{-1/2}$.

The ^{7}Li total cross section is of the order of 1 barn. Above about 100 eV, cross sections for both isotopes are approximately equal, therefore subtracting the TL readings of ^{7}LiF from those of ^{6}LiF, results in obtaining a signal due to neutrons with $E < 100$ eV.

Most commonly, the TLD's are applied only for the determination of gamma component of the BNCT field. This work shows how to use the TLD's also for characterization of the slow neutron component in the BNCT neutron beams. Two methods are proposed:
– by use of ^{6}LiF TL detectors with various thickness and
– by use of miniature TLD's with shielding made of ^{6}LiF.

The first method is based on the fact that neutrons with low energies are strongly absorbed in ^{6}LiF due to the high cross section for n(^{6}Li,T)α reaction. The number of absorbed neutrons increases with increasing thickness of ^{6}LiF pellet, but for thick detectors this effect saturates. The TL signal is approximately proportional to the number of absorbed neutrons, so the application of a set of detectors with various thickness and the analysis of the shape of the relationship between TL signal and detector thickness make it possible to estimate the average energy of slow neutrons (i.e. neutrons with $E < 100$ eV). The effect, which also has to be taken into account in these considerations, is self-attenuation of TL light within a TL pellet [3]. Assuming that both absorption of neutrons and self-attenuation of TL light undergo an exponential law (and that the neutron beam is perpendicular to the detector surface), one can write the following equation describing dependence of the intensity of the TL signal, I, on the detector thickness, d.

$$(5) \qquad I = \text{const} \cdot \left(\begin{array}{l} \dfrac{1}{\Sigma+\mu}\left(1-\exp\left(-(\Sigma+\mu)d\right)\right) \\ +\dfrac{\varepsilon\exp(-2\mu d)}{\Sigma-\mu}\left(1-\exp\left(-(\Sigma-\mu)d\right)\right) \end{array} \right)$$

where: Σ – macroscopic cross section of neutron absorption, cm^{-1}; μ – light attenuation coefficient, cm^{-1}; ε – coefficient describing reflection of TL light from the heater, dimensionless. Values of μ (10.2 cm^{-1}) and ε (0.19) were found with the method described in details elsewhere [3].

Fitting of the eq. (5) to the measured dependence $I(d)$ makes it possible to determine the effective value of Σ and then the corresponding neutron energy.

The method described above may work correctly only when used in a directional neutron field. For the in-phantom measurements, where neutrons are strongly scattered, another method was proposed. It is based on the use of ultra-miniature ^{6}LiF:Mg,Ti TL detectors (2 mm diameter and 0.5 mm thickness) located inside a miniature shielding container made of non thermoluminescent ^{6}LiF (^{6}Li enrichment 95.6%). The shape and dimensions of such set are illustrated in Fig. 1. Thermal neutrons are absorbed within the shielding, so the signal of the miniature TLD inside the container is caused only by the epithermal component and gamma radiation. The signal due to gamma radiation should be subtracted by application ^{7}LiF:Mg,Ti detectors and then the difference in response of ^{6}LiF:Mg,Ti detectors inside and outside of the container can be used for estimation of doses due to thermal and epithermal components of the neutron field. The advantage of this method

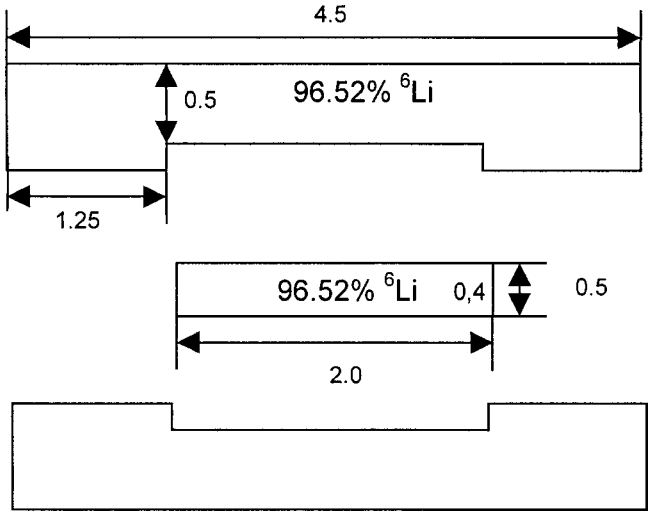

Fig. 1. Illustration of the shape and dimensions of a miniature ^{6}LiF container and TL detector.

is that the outer dimensions of the whole set (4.5 mm diameter and 1.5 mm thickness) are close to standard detectors enabling their application in typical phantoms.

Recombination chambers and high pressure C-CO_2 ionization chambers

It was expected that ionization chambers and recombination methods could be useful for the dosimetry of fast neutrons and of the external gamma radiation, which contaminates the BNCT beam. Tissue equivalent ionization chambers are commonly used for determination of total absorbed dose, or kerma in tissue. Recombination chambers additionally provide information on photon and neutron components of the absorbed dose (or kerma). In this work, the possibility of application of a recombination chamber for determination of neutron and photon kerma in BNCT beams was investigated. Two measuring methods were compared – a recombination microdosimetric method, RMM (a simplified approach) [5, 6, 9] and a twin-detectors method [8], with the C-CO_2 chamber as a neutron insensitive detector [5].

Recombination chambers are high-pressure, tissue-equivalent ionization chambers operated in unsaturated mode, under conditions of initial recombination of ions. Usually, such chambers contain several parallel-plate tissue-equivalent electrodes spaced by few millimeter gaps. Mostly, the chambers are filled with tissue-equivalent gas mixtures up to the pressure of some hundreds of kilopascals. The electrical charge created between the electrodes is proportional to the absorbed dose, while the shape of the saturation curve of the chamber provides information on radiation quality [5, 9, 10].

Determination of kerma components by RMM method is based on measurements of ion collection efficiency for several values of the polarizing voltage U_j, in the investigated radiation field, $f(U_j)$, and in a reference field of ^{137}Cs gamma radiation source, $f_\gamma(U_j)$. Then, the photon component, Γ, (ratio of the photon kerma to total kerma) can be determined by fitting the set of equations (6) to the experimental data.

$$f(U) = \Gamma f_\gamma(U) + \frac{1-\Gamma}{1+\dfrac{1-f_\gamma(U)}{f_\gamma(U)}\nu_{ef}} \tag{6}$$

where ν_{ef} is the effective local ionization density of the neutron component.

On the other hand, the neutron and photon components of the tissue kerma can also be determined by the twin-detector method [8] using two detectors of different neutron sensitivity. Commonly, two ionization chambers are used, for example, a tissue-equivalent (TE) ionization chamber combined with a C-CO_2, "neutron-insensitive" chamber, operated at atmospheric gas pressure. Because the C-CO_2 chambers have also some sensitivity to neutrons, the classical method requires information about the neutron spectral fluence of the radiation field, the sensitivity of the detectors as a function of neutron energy, and the kerma factors (fluence-to-kerma conversion coefficients) in materials of both detectors. In our method, the high-pressure C-CO_2 chamber is operated at low polarizing voltages. Under these conditions, the recombination of the ions generated in tracks of secondary particles of high linear energy transfer (LET) is considerably greater than for low-LET tracks. As a result, the relative neutron sensitivity k_U of such chambers is much lower than those operated at saturation and depends on the voltage applied. The earlier calculations and measurements [5] showed that, for chambers filled with CO_2 under the pressure of few MPa, the value of k_U is below 0.03 for neutron energies of up to 15 MeV supposing that the collecting electric field strength in the chamber cavity does not exceed 200 V/cm.

Measurements

The self-shielding effect was investigated experimentally in isotropic neutron fields and parallel neutron beams at the MARIA research reactor. In the first case, two sets of dilute (Al-0.1%Au; Al-0.1%Co; Al-0.1%W; Al-1%In; Al-2%Sc) and corresponding non-dilute activation foils, both covered with cadmium, were irradiated in carefully selected, homogeneous neutron field. In the parallel beam, only bare (without cadmium cover) gold and cobalt foils were used, both dilute and non-dilute.

Measurements with the recombination chambers were also performed at the reactor MARIA. The beam from the horizontal channel H8 was used. The neutron spectrum of the beam is similar to the expected spectrum at the therapeutic channel H2. The important difference was that the H8 beam was not filtered, so the radiation beam contained much more photons of gamma radiation than BNCT beams.

Two recombination chambers of different types were used. The first one was a tissue-equivalent, an in-phantom chamber of F-1 type [6]. This is a 3.8 cm^3 parallel-plate chamber, filled with methane up to a pressure of 1.1 MPa. The chamber has three ∅ 34 mm electrodes, a wall thickness of 0.6 g/cm^2 and the distance between electrodes of 1.75 mm.

The chamber is designed in such a way that it can be placed inside a water phantom and directly used for measurements at relatively high dose rates. Therefore, it can be applied for the determination of dosimetric parameters in different points in the phantom. Broad operational dose rate range of the chamber – from 10^{-5} up to 100 Gy/min makes it useful for characterization of both the radiotherapy fields and the fields used for calibration of radiation protection instrument.

The second chamber was a high-pressure graphite ionization chamber of G-5 type [11]. The cylindrical chamber is 115 mm in length and 19 mm in diameter. The distance between the electrodes is equal to 2 mm. The chamber is enclosed in a 0.3 mm thick aluminium container and can be filled with CO_2 gas up to a pressure of several MPa. The gas pressures of 2.1 MPa was used for this work. The chamber was operated with low collecting voltages (30 V) in order to provide the conditions of strong initial recombination of ions in the high-LET particle tracks. The chamber can be used for measurements of photon dose rates from 0.5 mGy/h up to 500 Gy/h.

The thermoluminescent detectors were investigated with neutrons from a ^{238}Pu-Be source placed inside the moderating polyethylene cylinder of 20 cm in diameter. A set of ^{6}LiF detectors with a thickness ranging from 0.2 mm to 3 mm and diameter 4.5 mm were manufactured at the Institute of Nuclear Physics in Kraków.

Results and discussion

Energy dependence of self-shielding coefficients $G(E)$, calculated by a simple, one interaction event model [1], is shown in Fig. 2 for activation foils of gold (having density of 24 mg/cm^2), scandium (30 mg/cm^2), cobalt (67 mg/cm^2) and tungsten (193 mg/cm^2) placed in a double sided 1 mm thick cadmium cover.

Corrected cross sections have been included to SAND-II library and used for the evaluation of neutron spectra. The resulting spectra were integrated (eq. (1)) with reaction cross sections for dilute foils.

The calculated reaction rates were compared with those obtained from the activation of dilute foils both in isotropic field and parallel beam. The discrepancies observed were rather low, especially for the most popular detector in BNCT dosimetry with a strong resonance absorption, i.e. the gold foil (below 1%).

Successful reconstruction of reaction rates of dilute foils proved indirectly that the method of cross section library modification by self-shielding coefficients and use of such modified library for the de-convolution of neutron spectra leads to correct results.

The next group of detectors, being of interest for this work, were ^{6}LiF detectors of different thickness. As mentioned above, a set of such detectors has been manufactured at the INP-Kraków. The TLD's were then irradiated with neutrons from the ^{238}Pu-Be source placed inside the moderating polyethylene cylinder of 20 cm in diameter. The results are presented in Fig. 3, for the detectors with the thickness ranging from 0.2 mm to 3 mm and diameter 4.5 mm. The solid line represents fitted equation (5). The fit resulted in a value of $\Sigma = 13.4 \pm 1.8$ cm^{-1}. For 95.6% enrichment of lithium in ^{6}Li, this value corresponds to the slow neutron energy of 0.44 ± 0.12 eV. This result rather overestimates the average energy of slow neutron component of the radiation field, because of the geometry of exposure.

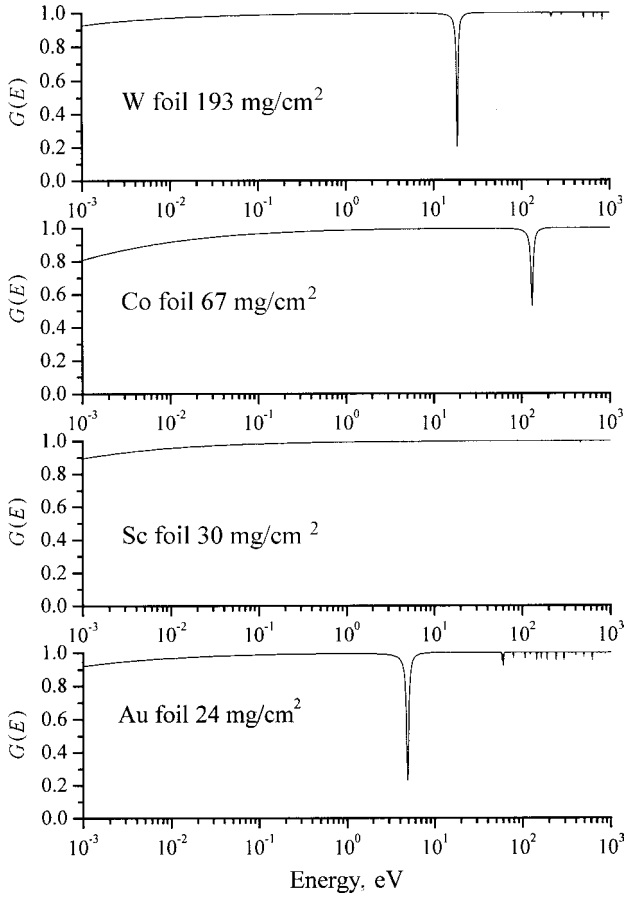

Fig. 2. Self-shielding coefficients for activation foils in a cadmium shield.

For the 3-mm thick pellet, the fraction of neutrons entering the detector from side surfaces was not negligible, what could explain the outlying of the respective data point in Fig. 3 from the trend. The fit performed without this point resulted in $\Sigma = 16.7 \pm 1.1$ cm^{-1}, what corresponds to slow neutron energy of 0.28 ± 0.04 eV. The result is in the range of realistic values for this kind of neutron field.

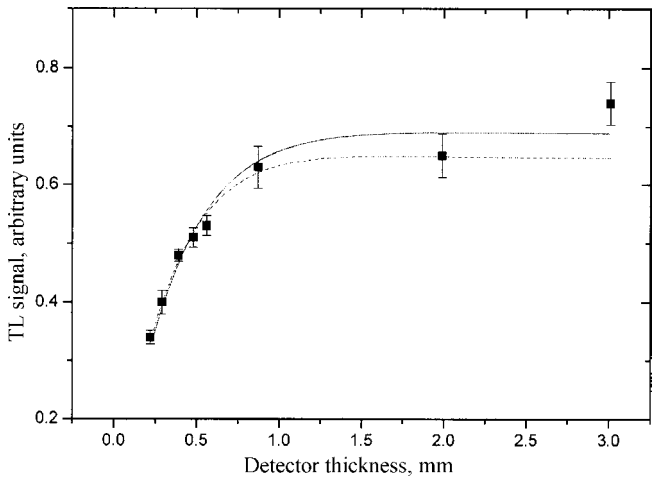

Fig. 3. Signal of the ^{6}LiF detectors of different thickness after exposure to moderated Pu-Be neutrons. Lines represent the fit with equation (5): solid line refers to all data points and broken line with the exception of data point for 3 mm thickness.

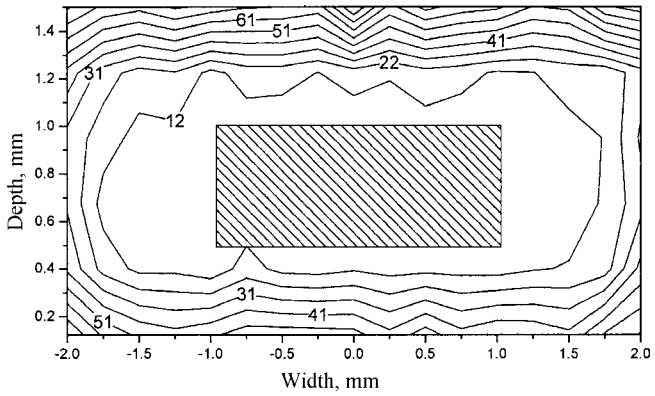

Fig. 4. Relative neutron dose distribution inside the ^{6}LiF pellet of 1.5 mm in diameter, located at 7.5 cm depth in-phantom in BNCT neutron field, calculated with MCNP code. The shaded rectangular represents the area of the 2-mm detector.

For ultra-miniature, ^{6}LiF:Mg,Ti TL detectors in a shielding container, the neutron dose distribution was determined by Monte Carlo calculations (Fig. 4). The calculations were performed with the MCNP code for typical BNCT neutron spectrum and depth 7.5 cm inside a phantom. A strong dose gradient is observed in the LiF shielding, what corresponds to absorption of thermal neutrons. Within the miniature TLD, the dose distribution is almost uniform, what corresponds to absorption of epithermal neutrons. The prototypes of such miniature detectors were manufactured at the INP-Kraków and first tests with the moderated Pu-Be source have been performed. In this field, the ratio of response of unshielded and shielded detectors was equal to 6.7.

Tests of the recombination chamber were focused on determination of photon component of the beam and on comparison with the C-CO_2 chamber. In such measurements, one has to remember that in BNCT beams, the value of k_U of the C-CO_2 chamber can be considerably influenced by activation of the chamber elements by thermal neutrons.

In case of the measurements at the H8 channel of the MARIA reactor, the ionization current of the F1 chamber, measured immediately after closing the reactor channel constituted $\beta_T = 0.037$ of the current at the open channel and decreased to the half of its initial value after 160 s. This corresponds with the decay of activated aluminium ($T_{1/2} = 2.3$ min, $E_\beta = 2.87$ MeV).

This activation was taken into account by an appropriate modification of the classic equations of twin detectors method. The following set of equations was proposed:

$$\begin{aligned} i_T\left(1-\beta_T\right) &= A_T\left(h_T\dot{K} + k_T\dot{K}_{\mathrm{n}}\right) \\ i_U\left(1-\beta_U\right) &= A_U\left(h_U\dot{K}_{\gamma} + k_U\dot{K}_{\mathrm{n}}\right) \end{aligned} \tag{7}$$

where: indices T and U concern the tissue equivalent and hydrogen free ionization chambers, respectively; h is the relative sensitivity of a detector to gamma radiation; k is the relative sensitivity of a detector to neutron radiation (relatively to the sensitivity to gamma radiation of the reference ^{137}Cs radiation source); A is the calibration factor of the chamber. β_T and β_U denote the contributions of β particles to the ionization current of the chambers F1 and G5, respectively.

The values of the above coefficient for the chambers F1 and G5 are equal to: $h_T = h_U = 1.0 \pm 0.015$; $k_T = 1.03 \pm 0.05$; $k_U = 0.01 \pm 0.002$; $\beta_T = 0.037 \pm 0.005$; $\beta_U = 0.015 \pm 0.01$.

The value of the gamma component of the total kerma $\dot{K}$ obtained from the measurements by the twin detector technique was equal to $\dot{K}_\gamma/\dot{K} = 0.878 \pm 0.007$. The uncertainty of the results is mostly due to uncertainty of the relative neutron sensitivity k_T of the chamber F1.

Measurements with the F1 chamber and application of the recombination method (eq. (6)) resulted in values of $\nu_{ef} = 16$ and $\Gamma = 0.877 \pm 0.009$.

Comparison of the obtained values of Γ and $\dot{K}_\gamma/\dot{K}$ shows that both methods gave the same result, within 0.1% of the measured value, while the uncertainty of each of the methods was estimated as being of about 1%.

Conclusions

From the dosimetry point of view, BNCT beams are the complex mixed radiation fields, because of broad neutron energy range, presence of gamma contamination and necessity of precise determination of several dose components. Despite of a rather long time experience, the dosimetric characterization of such beams is still a challenging task. Therefore, the development of measuring methods is of interest in all BNCT centres, both clinical and research.

The aim of the work was to improve the accuracy of the beam parameters determination (activation foils, ionization chambers) and to get more information about the dose contributions from some radiation components (TLD's, recombination chambers). The methods concerning nondilute activation foils and determination of photon kerma by a recombination chamber are ready for use. The ultraminiature TLD needs only calibration in real therapeutic beam. The parameters ν and Σ can provide additional information for monitoring of the beam quality. All the methods will be used for characterization of the BNCT beam at the MARIA reactor at Świerk.

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