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Studies of recombination chambers filled with nitrogen for BNCT dosimetry

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Abstract. Dosimetric characterization of therapy beams for boron neutron capture therapy (BNCT) involves determination of dose components and among them the "nitrogen" dose due to protons generated by neutron capture on ^{14}N. In this work, investigations were carried out using a graphite recombination chamber in order to determine the ^{14}N capture, gamma, and fast neutron dose components. The separation of the dose components is based on differences in the shape of the saturation curve, depending on the LET spectrum of the investigated radiation. The measurements were performed in reference radiation fields at the Institute of Atomic Energy at Świerk and at a reactor beam of the INP Řež (the Czech Republic). The gamma component was determined with an accuracy of about 5%, while the variations in its value could be monitored with an accuracy of about 0.5%. Relative changes in the beam components (thermal/fast neutrons) could be detected on line with an accuracy of about 5%. It was shown that the chamber with tissue-equivalent cups could be used for the determination of the ^{14}N capture dose at different depths in tissue.

Key words: recombination chamber • BNCT • dose components

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Introduction

Boron neutron capture therapy (BNCT) is a binary therapy, considered as a possible way of curing some kinds of malignant tumours (especially of those the brain), which up to now cannot be successfully treated in any other way. The physical concept underlying the method is based on the reaction of thermal neutron capture by a boron nucleus:

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

A boron compound is administered intravenously to the patient, in whom it accumulates preferentially in malignant tumour tissues. The tumour is then irradiated with low-energy neutrons produced by a nuclear reactor. The neutrons are moderated in the patient's sculp and tissues, so mostly they have energy in tumour. Some of the boron atoms absorb 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 cell involved, while both the boron drug and thermal neutrons alone are to some extent innocuous to tumour and normal tissues. Since the boron concentrates in tumour cells, the cancer can be destroyed, while the adjacent normal brain cells receive an acceptable radiation dose.

Until now, clinical trials of BNCT were initiated at a small number of reactors in Europe, the USA and Japan. Advanced studies on BNCT are carried out in

numerous research centres, where suitable or convertible reactors are available [1, 2, 9, 10].

The radiation effects of BNCT are associated with a radiation field which results in four components of the absorbed dose: a boron dose (from the $^{10}B(n,\alpha)^{7}Li$ reaction), a proton dose from the $^{14}N(n,p)^{14}C$ reaction, a neutron dose (mainly fast and epithermal neutrons) and a gamma-ray dose (practically only external dose and that from the capture reaction $^{1}H(n,\gamma)^{2}D$). The absorbed dose resulting from the neutron $^{14}N(n,p)^{14}C$ capture is usually considered to be calculable only based on the measured thermal neutron fluence. This paper presents the possibility of determining proton dose using a recombination chamber filled with nitrogen and a recombination microdosimetric method (RMM).

Materials and methods

Recombination chambers [12] are the high-pressure ionization chambers operating in the conditions of initial recombination of ions in the filling gas. Usually, the chambers are tissue-equivalent and then, they are used for the determination both of the absorbed dose in tissue and of the radiation quality factor [11] or LET spectrum.

The RMM was used for the determination of absorbed dose components. Originally, the method has been developed for tissue-equivalent parallel-plate recombination chambers [4]. In this work, it was extended to a cylindrical chamber filled with nitrogen.

The RMM is based on the dependence of the initial recombination of ions on the restricted linear energy transfer (LET, L_Δ) and on the comparison of the measured saturation curve $f_{mix}(U)$ with the saturation curve determined earlier in a reference gamma radiation field. The relationship between the ion collection efficiencies for these two kinds of radiation can be expressed as [3, 4]:

$$f_{mix}(U) = \frac{1}{D}\int \frac{D(L)}{1+\frac{L_\Delta}{L_0}\frac{1-f_\gamma(U)}{f_\gamma(U)}}\cdot dL \tag{1}$$

where: $D(L_\Delta)$ is the absorbed dose distribution versus restricted LET, cut-off Δ is of about 500 eV, $f_{mix}(U)$ and $f_\gamma(U)$ are the ion collection efficiencies measured at the polarized voltage U for mixed and reference gamma radiation, respectively; $L_0 = 3.5$ keV/μm.

The relationship (1) cannot be used directly for the determination of $D(L_\Delta)$ by deconvolution because the saturation curve is too smooth. Instead, it can be converted to the sum of n compartments of L_Δ:

$$f_{mix}(U) = \sum_{i=1}^{n} d_i s_i \tag{2}$$

where

$$s_i(U) = \frac{1}{\mu_{i+1}-\mu_i}\int_{\mu_i}^{\mu_{i+1}} \frac{1}{1+\mu\frac{1-f_\gamma(U)}{f_\gamma(U)}}d\mu \tag{3}$$

$$s_1(U) = f_\gamma(U) \tag{4}$$

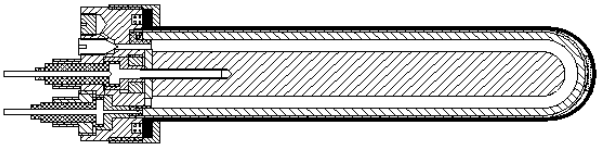

Fig. 1. Recombination chamber of the G5.1 type.

where: $\mu_i = (L_\Delta)_i/L_0$ are the arbitrarily chosen compartments of LET and d_i are the fitted parameters interpreted to be the dose fractions associated with certain compartments μ_i.

The reference saturation curve $f_\Delta(U)$ is measured during calibration, and then the $f_{mix}(U)$ values have to be determined at the same voltages which were applied for the reference curve. Practically, it is very difficult to determine $f_{mix}(U)$ in mixed radiation fields because the chamber is rather far from saturation, even at a maximum polarizing voltage, U_{max}. It was shown earlier [4] that what was sufficient to determine was only the relative ion collection efficiency $f'_{mix}(U) = i(U)/i(U_{max})$. Then, the measured curve is fitted with the equation:

$$f'_{mix}(U) = \sum_{i=1}^{n} d_i s_i \tag{5}$$

The fitting procedure is performed on the condition that d_i cannot be negative. The saturation current can then be determined from the sum of d_i values.

The $f_\gamma(U)$ function has to be determined as precisely as possible. For this purpose, the extrapolation of $1/i$ vs. $1/U$ was used.

A cylindrical graphite chamber of G5.1 type (Fig. 1), manufactured at the Institute of Atomic Energy, Świerk [12] was used for the measurements. This is a pen-like ionization chamber designed for the determination of absorbed dose at high dose rates (up to 500 Gy/h). The chamber is 115 mm long and 18 mm in diameter. The distance between the electrodes is 2 mm and the active volume is 7.1 cm^3. The chamber is enclosed in a 0.3 mm thick aluminium envelope. For this work, the chamber was filled with nitrogen up to 2 MPa. A small amount (below 1%) of air was mixed with the gas, in order to ensure proper recombination of ions.

For comparison, the measurements were performed also with a cylindrical graphite chamber of KG2 type, with a gas volume of 150 cm^3 and a distance between electrodes of 2 cm. The chamber was filled with nitrogen up to 1.5 MPa with about 1% of air. The chamber was irradiated in a mixed neutron-gamma radiation field of a ^{239}Pu-Be source, exposed in the iron filter. Additionally, the chamber was shielded with 3 cm of lead, in order to decrease the gamma component of the radiation field.

The ionization current measured by the KG2 chamber was normalized by the ratio of the gas mass in this chamber to the gas mass in the G5.1 chamber. The ratio was derived from the calibration in a ^{137}Cs gamma radiation field.

Development of a method for the determination of proton dose

An important step in the RMM procedure is the choice of the compartment borders. Most of the earlier works concerning RMM were aimed at determining the ambi-

ent dose equivalent in fast- and high-energy neutron radiation fields. Therefore, the tissue equivalent recombination chambers were employed, and the compartment borders were chosen in such a way that five components of the radiation dose could be distinguished, including the gamma component, the component with LET of about 100 keV/μm and the high-LET component, above 200 keV/μm [4]. In the present work, the compartments should be adjusted preferably to the expected dose components in nitrogen, in order to distinguish between the gamma dose, the proton dose (from neutron capture) and the dose due to nitrogen recoils. All the experimental curves were fitted by Eq. (5) with several different sets of L compartments, so as to find the most stable solution. Finally, a set of four compartments was Chojen. Taking into account that the LET of protons from the $^{14}N(n,p)^{14}C$ reaction is of about 75 keV/μm, it was proposed to divide the whole LET range into four compartments with the following borders:

1. Compartment I – below 20 keV/μm – associated with the gamma dose.
2. Compartment II – from 20 keV/μm to 80 keV/μm – expected to be associated with the proton dose due to the $^{14}N(n,p)^{14}C$ reaction.
3. Compartment III – from 80 keV/μm to 400 keV/μm – expected to be associated with the dose from fast neutrons (mostly alpha particles from reactions with carbon in a graphite wall).
4. Compartment IV – above 400 keV/μm – associated with heavy recoils.

It seems that the nitrogen chamber is not a proper instrument for the determination of dose components associated with compartments III and IV, thus they are outside the scope of this work.

Measurements

A series of calibration measurements were performed in the reference radiation fields of ^{137}Cs and ^{252}Cf sources in the Laboratory of Dosimetric Measurements at the Institute of Atomic Energy at Świerk. The neutron source was exposed either free in air or in 10 cm thick spherical filters made of paraffin or iron. The measurements were also performed with a chamber covered with a 3 cm thick polyethylene cup. The aim of the measurements was: (a) to establish whether it is possible to select the L compartment specific for protons generated in the neutron capture reaction, (b) to determine the calibration factor as a ratio of the ionization charge associated with the specified compartment to the thermal neutron fluence.

The same chamber was then used for the measurements in a BNCT therapeutic beam at the Institute of Nuclear Research at Řež in the Czech Republic [8].

Saturation curves were determined by sequential application of 15 polarizing voltages, ranging from 10 to 600 V, of both polarities. The ionization current was measured using a Keithley 642 electrometer, connected to a data acquisition system. The measured values were averaged for each voltage, normalized to the ionization current at 600 V for each polarity of the applied voltage, and finally, the mean value for the two polarities was calculated. The relative accuracy of so determined ionization current $i(U)$ was always better than 0.2%.

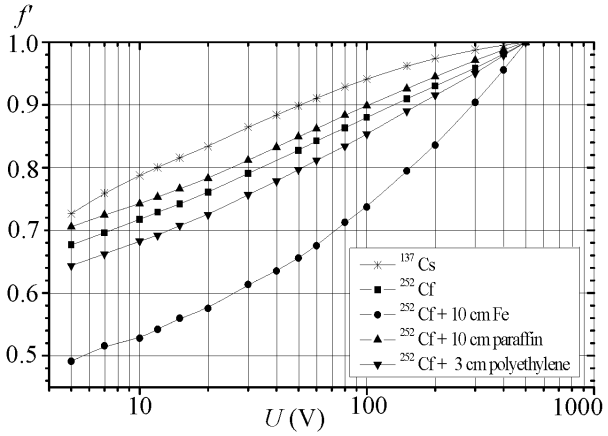

Fig. 2. Saturation curves determined by the recombination chamber G5.1, free in air, in reference gamma radiation field of ^{137}Cs source, and in radiation fields of ^{252}Cf neutron source exposed free in air or in the indicated filters. The curve marked as ^{252}Cf + 3 cm polyethylene was determined for the source free in air and the chamber placed in a 3 cm polyethylene cup.

Results and discussion

Figure 2 shows the saturation curves determined in a reference gamma radiation field (^{137}Cs) and in neutron radiation fields of a ^{252}Cf source. As described above, the measured values of the ionization current were normalized for each curve to the ionization current at a maximum polarizing voltage and displayed as f', relative ion collection efficiency.

The upper curve represents ^{137}Cs gamma radiation, while the others characterize mixed radiation. The lower values of ion collection efficiency for the mixed radiation are caused by differences in the gamma dose component and in neutron LET spectra. The higher the neutron LET and the lower the gamma component of the dose, the lower the ion collection efficiency. The use of paraffin and polyethylene filters moderates neutrons, so the flux of thermal neutrons and proton dose increase. The iron filter changes the LET spectra of the fast neutrons only slightly, but it strongly attenuates the gamma radiation from the californium source. The thick paraffin filter causes considerable changes in the LET spectrum and increases the gamma radiation flux, because of the neutron capture in hydrogen nuclei, which is accompanied by the emission of gamma radiation.

In Fig. 3, the same data are presented in a form which is more convenient for the RMM, i.e. the values $f'_{mix}(U)$ are plotted against the values of ion collection efficiency for the reference gamma radiation $f_\gamma(U)$. The curves are fitted with the theoretical relationship (5) with four LET compartments, as described above. The results of the fitting procedure are shown in Table 1.

As mentioned above, the ionization current, J_{th}, associated with compartment II was expected to be caused by protons from the neutron capture reaction in nitrogen. In order to prove this expectation, the ratio of the J_{th} to thermal neutron kerma in nitrogen, K_{th} was determined (a constant value was expected).

The energy spectra of the neutron fields were measured by the Bonner sphere method [5–7] and calculated by the Monte Carlo method. Figure 4 shows an example of the calculated spectrum.

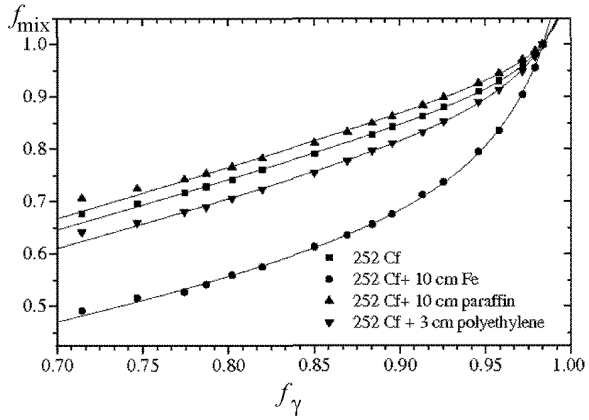

Fig. 3. Saturation curves from Fig. 2 plotted as the relative values of ion collection efficiency $f_{mix}(U)$ against $f_\gamma(U)$. Solid lines show the results of fitting with the relationship (5).

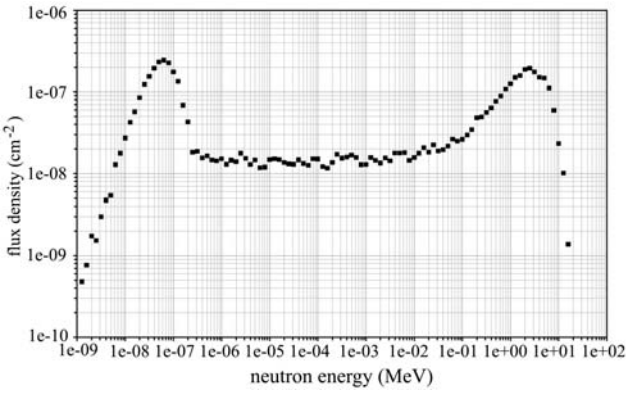

Fig. 4. Neutron flux density at a 1.0 m distance from the ^{252}Cf source in a paraffin filter, normalized to the number of emitted neutrons (notation 1e-09 indicates 1×10^{-9}).

Table 1. Ionization currents associated with the compartments of LET as they were derived by the RMM procedure. Compartment I – below 20 keV/μm – associated with the gamma dose; compartment II – from 20 keV/μm to 80 keV/μm associated with the proton dose due to the ^{14}N(n,p)^{14}C reaction; compartment III – from 80 keV/μm to 400 keV/μm – no signal expected for the graphite chamber filled with nitrogen; compartment IV – above 400 keV/μm – associated with heavy recoils. The last column shows the ratio of the ionization current associated with the compartment II to the kerma of thermal neutrons in nitrogen.

Radiation sources and filters	Chamber	Ionization current associated with the compartments of L_Δ (pA)				J_{th} / K_{th} (μC/Gy)	
		< 20 (keV/μm)	20–80 (keV/μm)	80–400 (keV/μm)	> 400 (keV/μm)		
^{252}Cf	G51	0.39	0.0126	0	0.1324	0.153	
^{252}Cf	G51 + cup	0.38	0.085	0.005	0.09	–	
^{252}Cf in iron filter	G51	0.044	0.014	0.002	0.05	0.147	± 4%
^{252}Cf in paraffin	G51	0.38	0.028	0	0.015	0.150	
^{239}Pu-Be in iron filter	KG2	0.27	0.235	0	0.495	0.146	

Then, the values of the thermal neutron flux density and thermal neutron kerma in nitrogen were determined from these data. The data were not available for the chamber in the polyethylene cup. The experimentally obtained values of J_{th}/K_{th} are shown in the last column of Table 1.

The main goal of this work was to investigate more thoroughly the G5.1 chamber properties in microdosimetric measurements in a BNCT therapeutic beam. The results of the measurements performed at the Institute of Nuclear Research at Řež in the Czech Republic are shown in Fig. 5. The dose distributions obtained from the RMM procedure are presented in Fig. 6 and in Table 2.

In Fig. 6, the contribution from nitrogen recoils is clearly seen in the high-LET region, but the contribution from 580 keV protons is not so visibly separated. As described above, it may be expected that the ionization current due to protons is associated with compartment II of the distribution. This is well supported by the observed influence of the polyethylene cup, which thermalizes epithermal neutrons and causes a large increase in the total saturation current (over 5 times) and a corresponding change in the shape of the saturation curve (shown in Fig. 5). This results in a decrease in the gamma dose in the chamber and in a very strong increase in the ionization current associated with compartments II and IV.

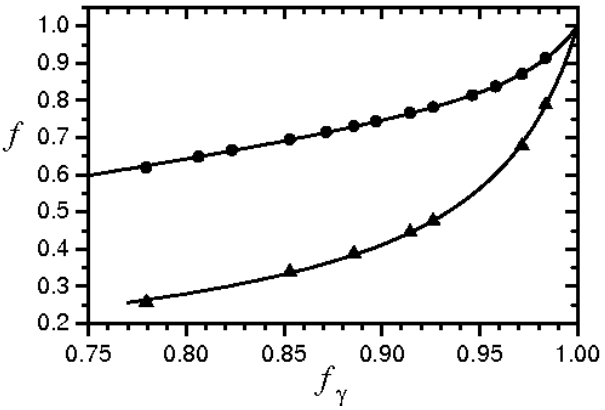

Fig. 5. Ion collection efficiency, f, determined in the BNCT beam with G5.1 chamber irradiated free in air (circles) and under the 3 cm polyethylene cup (triangles), vs. ion collection efficiency, f_γ, measured with the same chamber in the reference field of ^{137}Cs. Both f and f_γ were determined at the same polarizing voltages.

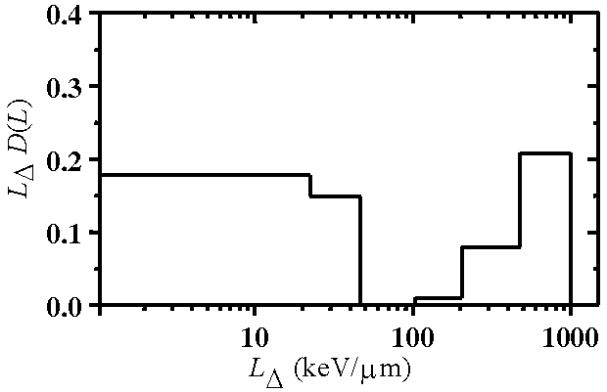

Fig. 6. Relative dose distribution $D(L_\Delta)$ vs. restricted LET measured in the BNCT beam at NRI Řež with a graphite chamber G5.1 filled with N_2.

Table 2. Relative dose distribution $D(L_\Delta)$ vs. restricted LET measured in the BNCT beam in INR at Řež with a graphite chamber G5.1 filled with N_2

Radiation field and filters	Saturation current (pA)	Ionization current associated with the compartments of L_Δ (pA)			
		< 20 (keV/μm)	20–80 (keV/μm)	80–400 (keV/μm)	> 400 (keV/μm)
BNCT beam (LVR-15)	353	234	37.8	0	81.2
BNCT beam (LVR-15) + cup	1990	305	727	0	958
Ratio: cup/free air	5.6	1.3	19.2	–	11.8

Conclusions

The results of the measurements performed with a set of cylindrical chambers showed that the recombination microdosimetric method can be used also for cylindrical geometry of the chambers.

The first measurements performed with the chamber filled with nitrogen clearly indicated that the recombination chamber filled with nitrogen makes it possible to determine the thermal neutron kerma rate in nitrogen and the absorbed dose due to $^{14}N(n,p)$ reactions.

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