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Paper

Initial approach to application of gamma spectroscopy to alpha particles dosimetry in BNCT

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Abstract

The aim of the study is to show a method of real-time determination of the dose deposited in a tissue-like medium by α-particles emitted from the $^{10}B(n,\alpha)^{7}Li$ reaction. The applied research method is to determine the correlation between the measured density of α-particle traces and measured in real time the 478 keV prompt-gamma rays derived from the $^{10}B(n,\alpha)^{7}Li$ reaction. To achieve this aim, an appropriate construction of an experimental set-up is needed. The experimental set-up built for the purpose of the measurements carried out in the MARIA Reactor at the National Centre for Nuclear Research in Świerk, Poland, is presented. The main challenges related to obtaining optimal conditions for the measurement of the 478 keV gamma photons; the preliminary results of spectrometric measurements and further studies are also discussed.

Introduction

Radiotherapy using α-particles is becoming an object of increasing interest. It is extremely important to monitor the dose deposited by the particles in the tissues and therefore the dose rate during the therapy. Measurements of α-particles deposited in tissues in the form of radiopharmaceutical or boron carrier as a source of α-particles in the reaction with neutrons can be only carried out indirectly. Difficulties in the direct *in vivo* measurements of α-particles released in the tissues are associated with their small ranges, of the order of μm. It is therefore necessary to develop appropriate measurement methods. A detection of low-energy γ photons accompanying the emission of these particles can be a suitable method[1]. α-particles are emitted, among the others, in the $^{10}B(n,\alpha)^{7}Li$ reaction. The reaction is the basis of boron neutron capture therapy (BNCT). In this reaction, with 94% probability, γ photons with an energy of ca. 478 keV are emitted[2, 3]. The paper includes a proposed method for the determination of the dose deposited in a tissue-like medium by α-particles derived from the $^{10}B(n,\alpha)^{7}Li$ reaction. The method is based on the real-time measurements of γ photons produced in the same reaction.

The experimental set-up built for the purpose of the measurements carried out in the MARIA Reactor at the National Centre for Nuclear Research in Świerk, Poland, is discussed in the following chapters. The conclusions drawn on the basis of gamma rays spectra measurements data obtained in this part of the research are also presented.

Methods

A special experimental set-up is designed and built to test the measuring method. Measurements were carried out in the field of neutrons emitted from Pu–Be source. Gamma photons were detected by the RayMon10 handle gamma ray spectrometer based on CdZnTe (Cadmium Zinc Telluride) detector and by the BE3830 model of HPGe (High Purity Germanium) detector supplied with the Genie 2000 Basic Spectroscopy Software manufactured by Canberra. PADC (Polyallyl-Diglycol-Carbonate) nuclear track detectors, known as CR-39[4], designed to determine the fluence of α-particles at a given position inside of the boron solution filled phantom irradiated by neutrons, will be used in further research stages. In order to determine the optimal conditions for the $^{10}B(n,\alpha)^{7}Li$ reaction products measurements, the calculations based on the MCNP code are also provided. The initial concept of the experimental set-up is shown in Figure 1. Pu–Be source (1) as a fast neutron emitter is surrounded by the polyethylene and lead shields (9) with the exit of neutron beam

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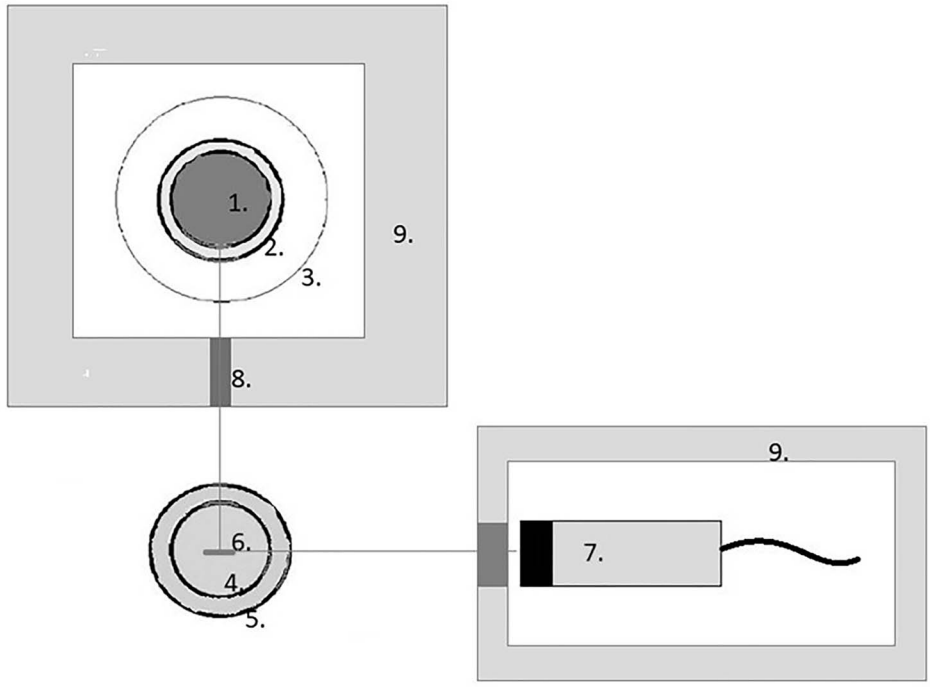

Figure 1. The initial set-up top scheme view. 1. Pu–Be source, 2. Steel cover, 3. Aluminium tube, 4. Inner vessel filled with boric acid, 5. Outer vessel filled with water (moderator), 6. CR-39 trace detector, 7. Gamma-ray spectrometer (CdZnTe, HPGe as a target detector), 8. Exit of the neutron beam collimator hole, 9. Radiation shields.

collimator hole (8). The vessels (inner vessel placed inside of the outer vessel) irradiated with neutrons are placed behind the shields and opposite the neutron beam collimator. The inner vessel (4) is filled with boric acid solution. Inside of the boron solution the CR-39 track detector (6) is placed. The outer vessel (5) is filled with water used as a moderator for fast neutrons emitted from Pu–Be source. Slowed down to thermal energies the neutrons reach the inner vessel and react with boron ^{10}B. The CR-39 track detector is designed to determine the α-particles emitted in the $^{10}B(n,\alpha)^{7}Li$ reactions. The gamma ray spectra emitted from the irradiated vessels are measured by the CdZnTe and HPGe detectors (7). In further studies HPGe spectrometer will be used. The detector is also properly shielded (9). In the gamma ray spectra measured on the experimental set-up, there are present lines corresponding to the energies of the prompt and the delayed gamma-ray photons from the proceedings reactions and decays of nuclei with short half-life, coming from the materials of the environment and the experimental set-up itself. The purpose of the construction of this stand, including the selection of appropriate neutron and gamma radiation shields, is:

(1) to improve the conditions for the 478 keV gamma photons measurements,
(2) to reduce the radioactive background, which affects the detection limit of measuring photons with an energy 478 keV emitted from irradiated sample,
(3) protection against excessive exposure to the absorbed dose during the measurements of the personnel and the detection system itself.

Selection of the detection system with suitable parameters and appropriate chemical composition is also crucial. It is important to select the appropriate shields against neutron and gamma radiation—especially the selection of gamma shields should be carefully made (the materials irradiated with neutrons themselves become a gamma radiation sources). Also, neutron shields should be carefully selected because of the nuclear reactions that take place in a material[5]. These phenomena cause the creation of undesirable radiation, which has an impact on the quality of the target experiment, namely on the detection limit of the 478 keV gamma photons measurement.

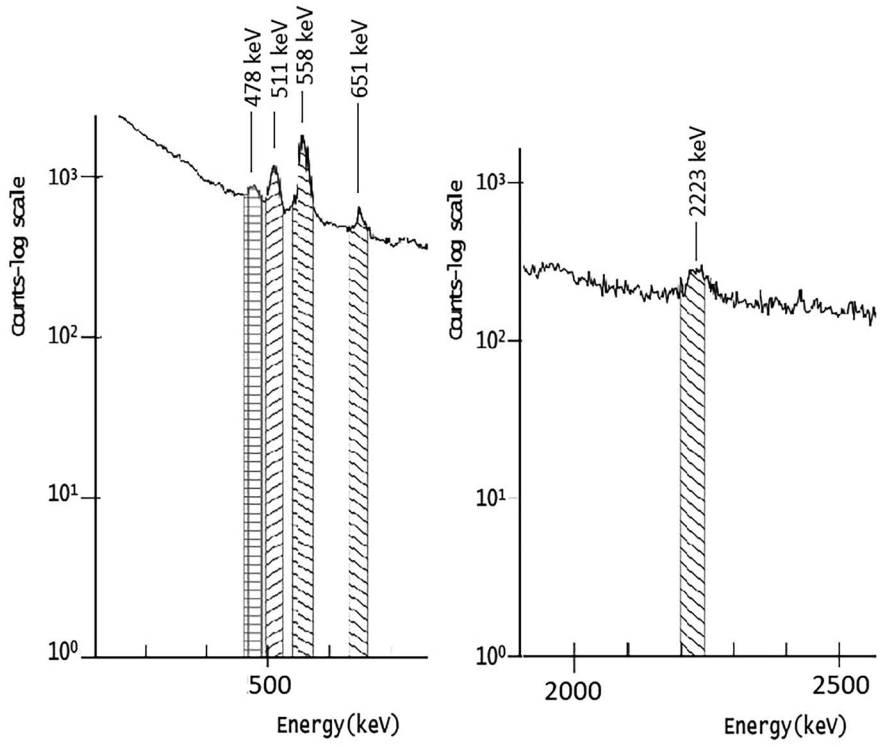

Figure 2. The representative fragments of the gamma rays spectrum with the identified spectral lines emitted from irradiated boron solution measured by the CdZnTe detector.

Results

In this part of the research the initial gamma rays spectra measurements and analysis were performed. In measured spectra—see the Figure 2—the spectral lines corresponding to the energies of 558 and 651 keV are observed. The prompt gamma rays are produced in ^{113}Cd(n,γ)^{114}Cd reaction[3, 5]. Cadmium is a component of the CdZnTe detector[6] used in the studies. The spectral lines coming from cadmium are located close to the line corresponding to energy photons of 478 keV from the investigated reaction. Cadmium is also commonly used as a component of neutron shields due to the high neutron capture cross section[7]. The 511 keV annihilation peak is also observed in the spectra measured by the CdZnTe detector due to interaction of high-energy photons with the matter, e.g. 2223 keV prompt gamma photons produced in the ^{1}H(n,γ)^{2}H reaction[3, 8]. This reaction occurs in polyethylene shields, water or other construction elements containing hydrogen. The high-energy gamma photons are also emitted from Pu–Be source[9] and from nuclei decays process taking place in experimental set-up construction elements. As an example ^{60}Co beta minus decay process can be indicated. ^{60}Co decay process most likely results from neutron activation of ^{59}Co nuclei[10], which are components of steel construction elements[11] of the experimental set-up. In ^{60}Co decay process the 1173 and 1332 keV delayed gamma photons[8] observed in spectra, measured by the HPGe detector, are produced. The spectral lines corresponding to the energies of 846 and 1778 keV prompt gamma photons are also observed. The 846 and 1778 keV gamma photons are produced in ^{50}V(n,γ)^{51}V reaction[3] occurring in steel. The 1408 keV prompt gamma photons occurring in the ^{27}Al neutron capture reaction is also observed. However, the probability that the 1779 keV delayed gamma photons created in ^{28}Al nuclei decays process[8] are present in registered spectra cannot be excluded. ^{28}Al radioactive nuclei are products of neutron activation of ^{27}Al stable isotopes[10] contained in construction elements of the set-up. Moreover, the 1201 and 1711 keV prompt gamma photons from ^{115}Sn(n,γ)^{116}Sn reaction were also registered in the spectra. In the measured gamma rays spectra emitted from the irradiated vessels with boron solution the 478 keV prompt gamma photons and 472 keV delayed gamma photons from gamma background shall be expected. The appearance of the

Table 1. the identified and the most likely predicted factors affecting the gamma spectra measurements([3,8,10])

Material	Reaction	Gamma emission	Comments
Containing boron and/or cadmium: neutron shields, vessels, set-up structural elements, CdZnTe detector	$^{10}B(n,\alpha)^{7}Li$	γ 478 keV (p)	Spectrum overlap with analysed spectrum.
	$^{113}Cd(n,\gamma)^{114}Cd$	γ 558 and 651 keV (p)	Spectral lines located close to the analysed line.
Containing hydrogen (water, polyethylene shields, others)	$^{1}H(n,\gamma)^{2}H$	γ 2223 keV (p)	Interaction with the matter causing the production of the 511 keV annihilation peak located close to the analysed line
Steel structural elements—Co neutron activation	$^{59}Co(n,\gamma)^{60}Co\ (\beta^{-}) \rightarrow {}^{60}Ni$	γ 1173 and 1332 keV (d)	Emission from ^{60}Co decay, significantly increases the radiation background level
Vessels containing Na, Aluminium structural elements	$^{23}Na(n,\gamma)^{24m}Na$ $^{27}Al(n,\alpha)^{24m}Na$	γ 472 keV (d)	Emission from ^{24m}Na decay, located relatively close to the analysed line
Set-up construction elements—Al neutron activation	$^{27}Al(n,\gamma)^{28}Al\ (\beta^{-}) \rightarrow {}^{28}Si$	γ 1779 keV (d) and 1408 keV (p)	(d) Emission from ^{28}Al decay, significantly increases the radiation background level
Sn neutron activation	$^{115}Sn(n,\gamma)^{116}Sn$	γ 1201 keV and 1711 keV (p)	
Steel structural elements—V neutron activation	$^{50}V(n,\gamma)^{51}V$	γ 1778 and 846 keV (p)	Significantly increases the radiation background level

(p)—prompt gamma photons (d)—delayed gamma photons

478 keV gamma photons in the background spectra can be caused by boron ^{10}B contents in the elements of the experimental set-up such as measuring vessels, while the 472 keV photons are emitted from the decay of ^{24m}Na nuclei[3, 8] and most likely coming from the neutron capture reaction on sodium in the measured vessels. Creation of the ^{24m}Na nucleus from the $^{27}Al(n,\alpha)^{24m}Na$ reaction induced by neutrons of energy greater than 7 MeV[10] is also very likely. This nuclear reaction occurs in the aluminium construction elements located relatively close to the Pu–Be source. The 472 keV spectral line is located very close to the analysed 478 keV spectral line. It is important to note that the measuring vessels used in the studies are made of silicon glass. It was found that there are different types of the silicon glass, which are commercially available in the market. The types of the glass differ from each other with chemical and isotopic composition, e.g. with sodium ^{23}Na and boron ^{10}B content. Moreover, there was found only one type of the silicon glass, that does not contain boron, therefore, it is most appropriate for the experiments. Comments on the measured spectra are present in Table 1[3, 8, 10]. In Figures 2 and 3 some examples of gamma rays spectra fragments with registered spectral lines are also shown.

Discussion

On the basis of the performed spectrometric measurements the following conclusions were drawn:

(1) Neutron and gamma shields and other elements of the experimental set-up including detection system, measurement vessels etc. become a source of undesirable gamma radiation.
(2) Selection of the irradiation vessels made of an appropriate chemical composition is also important, e.g. sodium is a source of undesirable gamma radiation. Other materials of set-up construction elements should be also carefully selected due to neutron activation, e.g. steel construction elements (cobalt isotopes) or lead gamma shields covered by lacquer (chemical composition and impurities are crucial).
(3) All materials containing boron (measurement vessels, shields or other structural elements of the experimental set-up) should be eliminated.
(4) The most appropriate type of silicon glass for making measuring vessels was selected. The selected material is boron free. According to the manufacturer's specification the material is characterized by the lowest content of impurities with other chemical elements.
(5) Problems with the separation of the 478, 511, 558 keV spectral lines may be solved by selecting alternative neutron shields, e.g. polyethylene, or by using a detector with appropriate parameters (energy resolution and detection efficiency, chemical composition of detection material—without cadmium, e.g. HPGe).
(6) The 511 keV annihilation peak was always present in spectra due to the interaction of high-energy

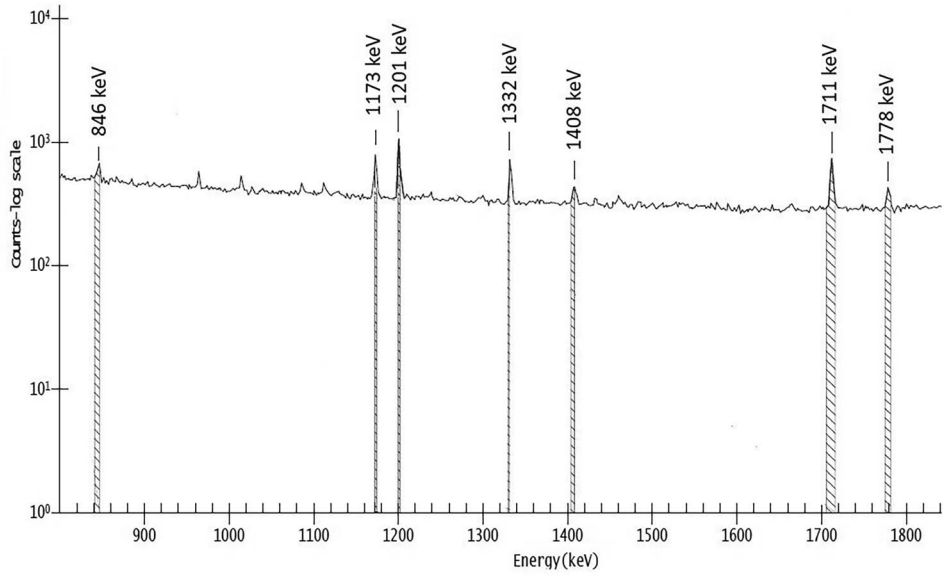

Figure 3. The representative fragment of the background gamma rays spectrum with the identified spectral lines measured by the HPGe detector.

gamma rays with the matter—for that selection of a detector with better energy resolution e.g. HPGe can be a good solution for the separation of the 511 keV from the 478 keV spectral line.

(7) In the spectra the gamma photons being a result of the neutron capture reaction on the CdZnTe detector are also observed.

(8) Selection of detector with suitable parameters and appropriate chemical composition is also crucial—in further studies the HPGe detector shall be used.

Conclusions

Based on the conclusions drawn, works on further improvements of the experimental set-up shall be carried out. This will allow to create more optimal conditions for the 478 keV gamma photons measurements. It is necessary to choose appropriate materials, not only for the radiation shields, but also for the measuring vessels and other construction elements of the experimental set-up and according to the gained experience—see Table 1—to use a spectrometer with a better energy resolution and another chemical composition, e.g. the HPGe detector. In the further research stage the CR-39 detectors will be used to determine the fluence of α-particles derived from $^{10}B(n,\alpha)^{7}Li$ reaction. The number of α-particles from the $^{10}B(n,\alpha)^{7}Li$ reaction will be determined on the basis of the intensity of the measured 478 keV γ photons. Distribution of the $^{10}B(n,\alpha)^{7}Li$ reaction rate in the total boron solution volume will be calculated using numerical calculations based on the MCNP. The number of registered gamma photons emitted from the total solution volume will be related to the reaction rates determined on the basis of the measured traces of $^{10}B(n, \alpha)^{7}Li$ reaction products on the CR-39 detector, and to the calculated rates of reactions taking place in the total solution volume. On that basis the dose deposited by the α-particles in a tissue-like medium will be estimated. The dose deposited by the α-particles from this reaction will be determined on the basis of the analysis of the interaction of the α-particles with the matter using the Bethe–Bloch formula[12] (computational concept of deposited dose determining is developed). In order to verify the measurements, numerical calculations based on the MCNP code will be performed. In the case of the experimental set-up the neutrons emitted from the Pu–Be source are used, whereas in the case of the target set-up the neutron beam from the horizontal channel at MARIA Reactor will be applied[13, 14].

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Conflict of interest

No conflicts of interest were notified.

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