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PHYSICAL INSTRUMENTS FOR ECOLOGY, MEDICINE, BIOLOGY

A Polyethylene Moderator with Volumetric Bismuth Inclusions for Boron Neutron Capture Therapy

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Abstract—Boron neutron capture therapy, a promising treatment for malignant tumors, requires intense neutron beams in the epithermal energy range. Nuclear reactors or particle accelerators generate higher-energy neutrons and slow them to the required energy through elastic and inelastic scattering of neutrons on the atomic nuclei of the moderator material. Neutrons with energies around 10 keV are optimal for treating deeply localized tumors, and a beam shaping assembly with a moderator made of crystalline or pressed magnesium fluoride is used to generate such a neutron beam. For research with cell cultures and laboratory animals, a hydrogen-containing moderator (plexiglass, polyethylene) is used; in this case, the neutron energy spectrum is shifted toward the thermal region. The authors have proposed using a moderator made of polyethylene with a volumetric inclusion of bismuth. This paper presents the results of measurements of the spatial distribution of the intensity of nuclear neutron absorption reactions by boron and the intensity of γ-radiation in a water phantom for three similar moderators made of plexiglass, polyethylene, and polyethylene with volumetric bismuth inclusions. The measurements were conducted using a compact sensor with a pair of injection-molded polystyrene scintillators, one of which is enriched with boron. It is shown that the use of polyethylene with volumetric bismuth inclusions, compared to plexiglass or polyethylene, reduces the unwanted γ-radiation dose while maintaining the neutron flux intensity.

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1. INTRODUCTION

Boron neutron capture therapy (BNCT) is considered a promising method for treating malignant tumors [1, 2], which ensures the selective destruction of tumor cells by accumulating boron in them and subsequent irradiation with neutrons. As a result of the absorption of a neutron by a boron nucleus, a nuclear reaction occurs with a large release of energy in the tumor cell, which leads to its death.

Currently, the BNCT technique is beginning to enter clinical practice: treatment is being carried out in Japan, clinical trials are being conducted in Japan, China, South Korea, Finland, and will soon begin in Russia. All clinics use charged particle accelerators with a beryllium or lithium target to generate neutrons. A therapeutic neutron beam in the epithermal energy range is produced using a beam shaping assembly with a magnesium fluoride moderator.

Thermal energy neutrons can and are used to conduct scientific research on organic samples and laboratory animals. In this case, it is more efficient to use a hydrogen-containing moderator (plexiglass, polyethylene) to produce them: due to the rapid deceleration of neutrons, the thickness of the moderator is smaller and, as a result, the intensity of the neutron flux is higher. To reduce the γ-radiation dose, we propose using a moderator made of polyethylene with a volumetric inclusion of bismuth.

2. EXPERIMENTAL SETUP

The research was carried out at the Budker Institute of Nuclear Physics, Siberian Branch, Russian Academy of Sciences, with the VITA accelerator neutron source [3]. The facility diagram is given on Fig. 1. The facility consists of an electrostatic tandem charged particle accelerator of an original design (a tandem accelerator with vacuum insulation) for producing a stationary monoenergetic beam of protons or deuterons with an energy of up to 2.3 MeV, a current of up to 10 mA, a lithium target for generating neutrons with an output of up to 2×10^{12} s^{-1}, and a set of neutron

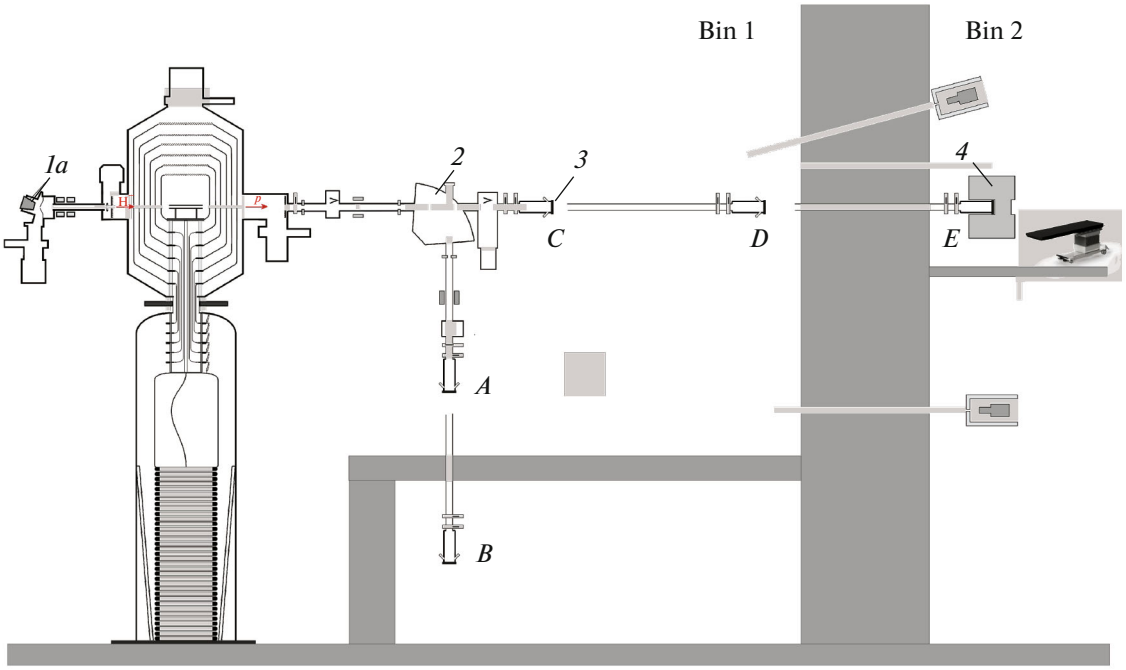

Fig. 1. Schematic diagram of the VITA accelerator neutron source: (*1*) tandem accelerator with vacuum insulation, (*2*) rotating magnet, (*3*) lithium neutron-generating target, (*4*) system for forming a neutron beam with a magnesium fluoride moderator. The lithium target is placed in positions *A*, *B*, *C*, *D,* or *E*.

beam shaping assemblies for producing a beam of cold, thermal, epithermal or fast neutrons.

In this study, we used a proton beam with an energy of 2.1 MeV and a current of 1 mA. The lithium target was placed in the horizontal tract in the position *C*. One of three manufactured moderators was placed close to the target: plexiglass, polyethylene, or polyethylene with a volumetric inclusion of bismuth. Next, a water phantom was placed with a gap of 5 mm to the moderator. Two sensors were attached to the moving platform of the phantom to measure the intensity of the neutron flux and γ-radiation.

All moderators were made in the form of disks with a diameter of 155 mm. The thickness of the disks made of organic glass and polyethylene with volumetric inclusion of bismuth was 72 mm, and the thickness of the polyethylene disk was 52 mm.

The disk from polyethylene with volumetric inclusions of bismuth was made from polyethylene powder and high-purity bismuth powder VI-00 using the plastic injection molding method at the Institute of Solid State Chemistry and Mechanochemistry (Siberian Branch, Russian Academy of Sciences). This moderator is similar to the material Poly-Biz [4], produced by Shieldwerx (United States) and is used to protect against γ-radiation; the density of hydrogen atomic nuclei in it is considered to be equal to 5.39×10^{22} cm^{-3}, and the density of bismuth atomic nuclei is 6.6×10^{21} cm^{-3}. The thickness of the moderator disks was chosen so that all moderators form a neutron beam with a similar energy spectrum. Since the slowing down of neutrons is achieved primarily by their elastic scattering on hydrogen atomic nuclei, the thickness of the moderator was selected in such a way as to obtain approximately the same linear density of hydrogen atomic nuclei in them. In the moderator made of organic glass with a density of 1.18 g/cm^3, the linear density of hydrogen atomic nuclei was 4.08×10^{23} cm^{-2}, that in a moderator made of polyethylene with volumetric inclusions of bismuth was 5% less (3.88×10^{23} cm^{-2}), and that in a moderator made of polyethylene with a density of 0.94 g/cm^3 was 3% more (4.20×10^{22} cm^{-2}).

The water phantom was a parallelepiped-shaped vessel measuring 330 × 330 × 315 mm^3, filled with water, with a platform that can be moved in three directions. The vessel was made of monolithic polycarbonate. The front wall of the vessel, which is placed against the moderator, was made of a thin polyethylene terephthalate membrane with a thickness of 0.5 mm. The thickness of all other polycarbonate walls was 8 mm, and the thickness of the bottom was 16 mm.

To measure the radiation intensity, we used a small-sized sensor developed with a pair of injection-molded polystyrene scintillators, one of which was enriched with boron [5]. The size of the sensor's sensitive element was approximately 1 mm^3. The light signal from the sensor was transmitted via a fiber optic cable to a micropixel avalanche photodiode, and the recording electronics incremented the event counter

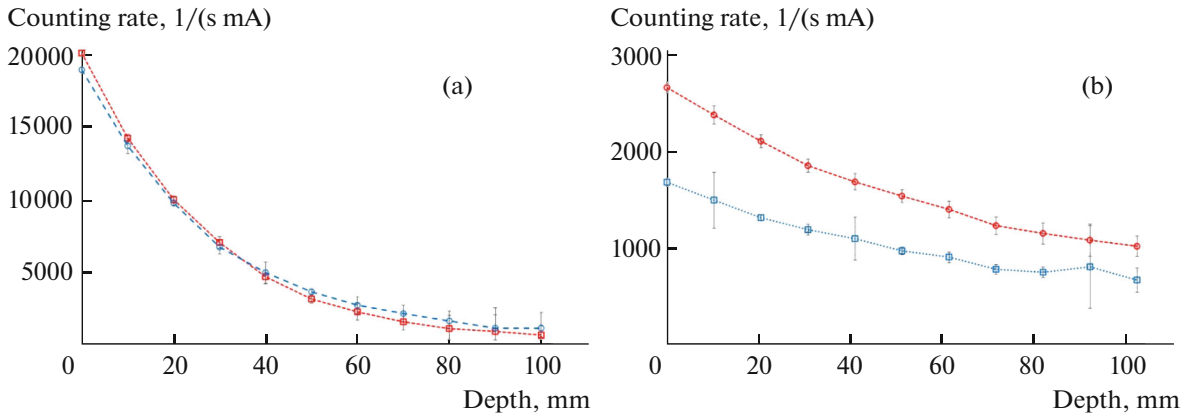

Fig. 2. Count rate distributions by depth for the (a) neutron sensor and (b) γ-radiation sensor, normalized to a proton beam current of 1 mA.

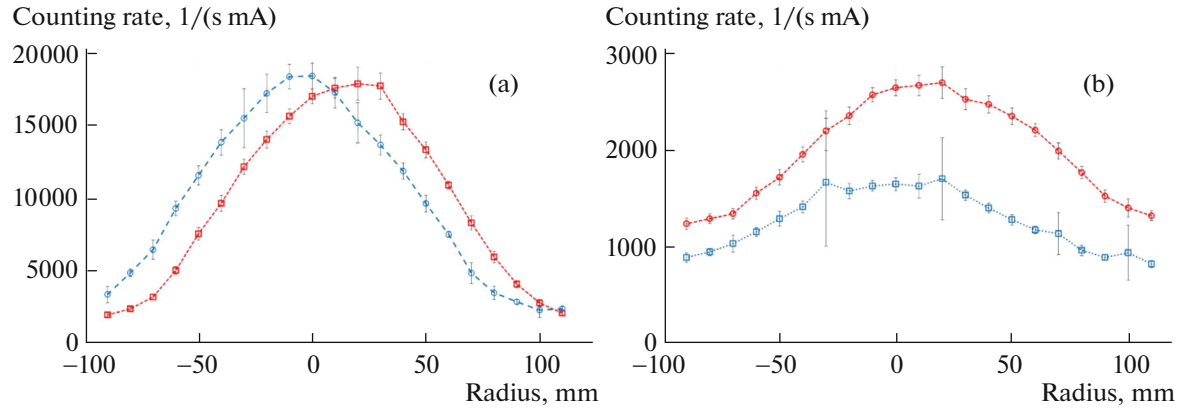

Fig. 3. Radial distributions of the (a) neutron sensor count rate and (b) γ-radiation sensor, normalized to a proton beam current of 1 mA.

when the signal exceeded a threshold of several optical photons. We determined the magnitude of the neutron flux through the difference in counts between the two sensors. Since the neutron flux signal is caused by the absorption of neutrons by boron nuclei, similar to the process in BNCT, we will consider this signal as a "boron" dose: one of the four dose components considered in BNCT. Dose of γ-radiation was determined through the difference in the count in the detector channel with a scintillator without boron and in the channel without a scintillator but with a fiber optic cable since this makes it possible to exclude the contribution of Cherenkov radiation generated in the fiber optic cable.

3. RESULTS

During the studies, the proton beam energy was 2.100 ± 0.002 MeV, the proton beam current was maintained at a level from 0.98 mA to 1.1 mA with a stability of 0.3% to 1.5%. The characteristic depth and radial distributions of the count rate of the sensors, normalized to a unit of proton beam current of 1 mA, using a moderator made of polyethylene with volumetric inclusions of bismuth are shown in Figs. 2, 3. When using other moderators, the distributions are similar.

It is evident that the radiation intensity decreases with depth: thus, the recorded intensity of the neutron flux decreases by two times at a depth of 2 cm, the γ-radiation intensity decreases by two times at a depth of 7 cm. The width at half-height of the radial distribution of the neutron flux intensity is 11 cm and γ-radiation is approximately 15 cm.

It was established during the research that the implemented fastening of the sensors to the moving carriage of the water phantom did not ensure the desired accuracy of their position during the movement of the carriage, namely: when the sensor returned to its original position, its actual position could differ by a distance of approximately 1 mm, and this led to different results during the measurement. The reason is that we placed all three fiber optic cables

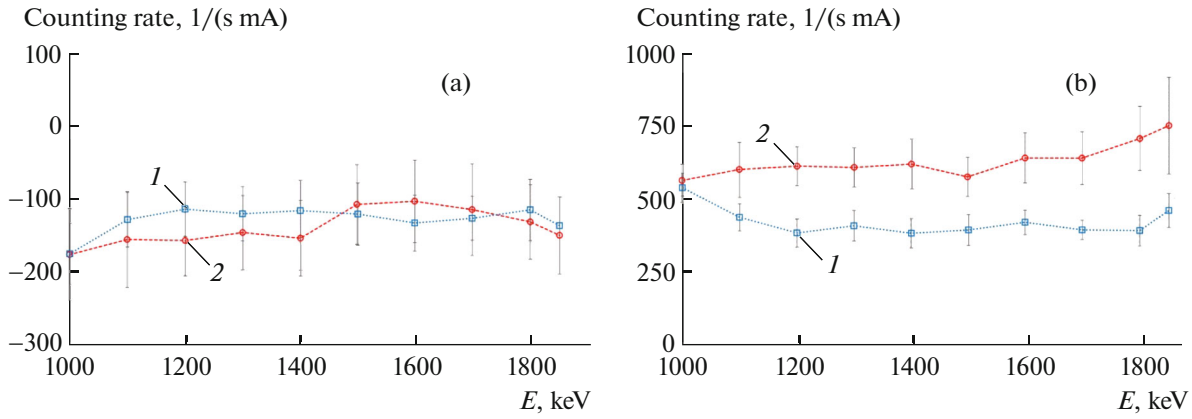

Fig. 4. Dependences of the (a) neutron sensor count rate and (b) γ-radiation sensor from the energy of the proton beam E when using retarders made of polyethylene with volumetric inclusions of (*1*) bismuth and (*2*) from plexiglass.

of each sensor in a light-tight plastic tube, and all three tubes were pressed together with screws, not very tightly, trying not to damage the fiber optic cables, to the moving carriage of the water phantom, directing the tubes upward and holding them above the water surface of the phantom with the hook of the crane beam.

More accurate results for comparing radiation parameters when using different moderators were obtained when we did not move the sensor but replaced the moderators. It was for this procedure that a 5-mm gap was left between the surface of the moderator and the wall of the water phantom. Using a thinner polyethylene moderator, we carefully moved the water phantom 20 mm towards the lithium target. Measurements for comparing moderators of the same thickness were taken at several sensor locations near the maximum signal; measurements for comparing a thin moderator with thicker moderators were taken at one point of maximum signal.

Of the two sensors used, one gave more stable readings, and it was this reading that we analyzed further.

The neutron sensor is set up so that, in the absence of neutrons, it produces a negative signal at 100 s^{-1}. Although such a shift in the zero signal is small (at the level of 0.5% of the analyzed signal), we did not neglect it but took it into account. Figure 4a shows a graph of the dependence of the neutron sensor count rate on the proton beam energy in the range of 1.0–1.85 MeV using moderators made of organic glass and polyethylene with volumetric inclusions of bismuth. It should be noted that neutron generation did not occur at these values of proton energy; the reaction threshold ^{7}Li(p,n)^{7}Be is equal to 1.882 MeV. It can be seen that the count rate of the neutron sensor in the absence of neutrons does not depend on the energy of the protons and the moderator used. By averaging all these values, we obtain a value equal to -130 ± 20, which we will subtract from the readings of the neutron sensor when generating neutrons to determine the boron dose.

Determining the zero signal of the sensor reading of γ-radiation is a bit more difficult. We were interested in γ-radiation produced by neutrons as a result of their interaction with the atomic nuclei of the structural materials of the target unit, moderator, and water phantom, for example, in the form of photons with an energy of 2.2 MeV, formed in the reaction ^{1}H(n,γ)^{2}H, and a significantly lower intensity emission of photons with an energy of 478 keV generated as a result of inelastic scattering of protons on lithium atomic nuclei in the reaction ^{7}Li(p,p'γ)^{7}Li [6]. However, in the conducted studies, the sensor recorded not only the radiation of interest to us but also the accelerator's bremsstrahlung. To measure the contribution of accelerator bremsstrahlung, we performed measurements excluding neutron generation. Figure 4b shows the dependence of the sensor count rate of γ-radiation on a proton beam energy in the range of 1.0–1.85 MeV using moderators made of organic glass and polyethylene with volumetric inclusions of bismuth. It can be seen that the counting rate depends weakly on energy and depends on the moderator used. The bremsstrahlung radiation of an accelerator is caused primarily by the deceleration of electrons generated in the accelerator's accelerator channel in the structural materials of the gas stripping target placed inside the high-voltage electrode of the accelerator. With an increase in the voltage of the high-voltage electrode, the energy of the electrons increases and, as a consequence, the intensity of the bremsstrahlung radiation; however, the electron flux decreases at the same time due to the smaller cross-section of ionization of the residual gas by accelerated ions. This effect may explain the weak dependence of the detector count rate of γ-radiation on the accelerator voltage (the voltage of the high-voltage electrode is half the energy of the protons minus the initial energy of the beam of negative hydrogen ions injected into the accelerator). This weak

Table 1. Neutron sensor count rate Y, normalized to the proton beam current, and a certain boron dose rate D_B

Moderator	Y, 1/(s mA)	D_B, rel. units	D_B, %
Plexiglass	14280 ± 400	14400 ± 400	100
Polyethylene with volumetric inclusions of bismuth	13740 ± 300	13870 ± 300	96 ± 4
Polyethylene	22500 ± 1000	22630 ± 1000	157 ± 7

Table 2. Count rate of the gamma-ray detector Y, normalized to the proton beam current, and a certain dose rate of γ-radiation D_γ

Moderator	Y, 1/(s mA)	D_γ, rel. units	D_γ, %
Plexiglass	2860 ± 150	2230 ± 160	100
Polyethylene with volumetric inclusions of bismuth	2140 ± 120	1720 ± 130	77 ± 9
Polyethylene	4110 ± 190	3480 ± 200	156 ± 10

dependence allows us to extrapolate the values to an energy of 2.1 MeV and obtain the following values of bremsstrahlung intensity: 630 ± 20 when using a moderator made of organic glass and 420 ± 30 when using a moderator made of polyethylene with volumetric inclusion of bismuth. The lower value of the recorded intensity of bremsstrahlung when using a moderator made of polyethylene with a volumetric inclusion of bismuth is naturally explained by the presence of bismuth, which reduces the γ-radiation intensity. A polyethylene moderator, due to its higher content of oxygen and carbon atomic nuclei, should reduce the recorded intensity of bremsstrahlung a little better than plexiglass, but this reduction will be small since these atomic nuclei are light, so we will consider the magnitude of the bremsstrahlung intensity when using a polyethylene moderator to be the same as when using a plexiglass moderator.

Let us compare the measurement results when the sensor remained stationary and the moderators were replaced. First, the organic glass moderator and the polyethylene moderator with volumetric bismuth inclusions were replaced; measurements were carried out three times, each time at energies of 2.1 and 1.85 MeV. The water phantom was then moved 20 mm towards the lithium target and measurements were also carried out three times with a polyethylene moderator. The measurement results are shown in Tables 1 and 2. We determined here the boron dose rate and γ-radiation dose, subtracting from the measured count rate of the corresponding detector the zero signal shift value determined above.

We have previously used a plexiglass moderator and conducted a large number of studies with organic samples, laboratory animals, and also irradiated domestic animals (cats and dogs) with spontaneous tumors. The list of publications of these studies can be found in separate chapters of monographs [1–3] and in [7]. For this reason, we will compare the results obtained with the use of a moderator made of plexiglass.

It is evident that the use of a moderator made of polyethylene with volumetric inclusions of bismuth allowed for a significant (by 23%) reduction in the dose rate of γ-radiation with virtually no reduction in the power of the boron dose. This effect is explained by the fact that the sizes and retarding properties of the moderators are the same and, therefore, there is no difference in the boron dose, but the presence of a heavy metal inside the moderator reduces the γ-radiation dose.

It is also evident that the use of thinner polyethylene instead of plexiglass resulted in an equal increase in the power of both the boron dose and the γ-radiation dose. This increase is explained by the proximity of the detector to the radiation source. If, when using a moderator made of organic glass, the detector is placed at a distance of 108 mm from the lithium layer of the target, then it is placed at a distance of 88 mm in the case of using a moderator made of polyethylene. Assuming that the dependence of radiation intensity on distance is quadratic, we find that the use of a thinner moderator increases the dose rate by 1.51 times, which is in good agreement with the measured values.

Thus, as a result of the conducted study, it was established that the use of a moderator made of polyethylene with volumetric inclusions of bismuth improves the characteristics of the neutron beam for conducting research in the field of boron neutron capture therapy of malignant tumors.

4. CONCLUSIONS

In boron neutron capture therapy (BNT) research, a hydrogen-containing moderator is used to form the neutron beam, providing a higher dose rate than the magnesium fluoride moderator used to treat patients. The paper compares three moderators made of organic glass, polyethylene, and polyethylene with a

volumetric inclusion of bismuth. Using a small-sized detector with a pair of injection-molded polystyrene scintillators, one of which is enriched in boron, the spatial distributions of the boron dose and the γ-radiation dose were measured in a water phantom. It has been established that a moderator made of polyethylene, compared to a moderator made of organic glass, due to its smaller thickness, provides a greater power of both the boron dose and the γ-radiation dose. A moderator made of polyethylene with volumetric inclusions of bismuth, compared to a moderator made of plexiglass, significantly reduces the γ-radiation dose rate while maintaining the power of the boron dose. The use of a bismuth-doped polyethylene moderator is recommended for BNCT research using organic samples, laboratory animals, and for irradiating domestic animals with spontaneous tumors to develop dosimetry tools and test new boron delivery drugs.

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CONFLICT OF INTEREST

The authors of this work declare that they have no conflicts of interest.

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