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Validation and optimization of the epithermal neutron flux detector using the ^{71}Ga(n,γ)^{72}Ga reaction

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Abstract: An intense epithermal neutron flux is necessary for boron neutron capture therapy (BNCT), a promising technique for the treatment of malignant tumors. The epithermal neutron flux is an essential characteristic of the BNCT neutron beam and its measurement is directly related to the reliability of the treatment planning system. Such a tool could be a cylindrical activation detector using ^{71}Ga(n,γ)^{72}Ga reaction. This paper describes a detector made in the likeness of the previously proposed one and presents the results of numerical simulation of the detector sensitivity and its experimental use. The paper notes the difference between the simulated sensitivity of the detector and the sensitivity of the previously proposed one and makes assumptions about the reason for this difference. The work also proposes an improvement of the detector, which allows not only to reduce the contribution of fast neutrons to the detector sensitivity, but also to provide a new opportunity to refine the spectrum of neutrons that are most effective for the treatment of deep-seated tumors.

Keywords: Neutron detectors (cold, thermal, fast neutrons); Detector modelling and simulations I (interaction of radiation with matter, interaction of photons with matter, interaction of hadrons with matter, etc); Models and simulations

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1 Introduction

Boron neutron capture therapy (BNCT) is currently considered as a promising technique for treatment of malignant tumors [1, 2]. As a result of the neutron absorption by boron a nuclear reaction ^{10}B(n,α)^{7}Li takes place with a large energy release inside the cell which contains a boron nucleus that leads to the destruction of this cell. An intense epithermal neutron flux (not less than 1×10^9 n/cm^2/s) is necessary to treat deep-seated tumors. The epithermal neutron flux is an essential characteristic of the BNCT neutron beam and its measurement is directly related to the reliability of the treatment planning system. Consequently, it is of great interest to accurately measure the epithermal neutron flux of the BNCT neutron beam. Article [3] presents a description of a cylindrical activation detector using ^{71}Ga(n,γ)^{72}Ga reaction optimized by Monte Carlo simulations; below we call this detector original. In the detector, the activation material, i.e., gallium nitride (GaN), is positioned in the geometrical center of a high-density polyethylene cylinder covered with cadmium foil. The description of the original detector is sufficient for the manufacture of such a detector and for measuring the intensity of the neutron flux.

The aim of this work is to use the activation detector to measure the epithermal neutron flux, analyze the results obtained, and optimize the detector if necessary.

2 Design of the detector

The principle of the epithermal neutron flux monitor is based on the activation method using ^{71}Ga(n,γ)^{72}Ga reaction [3, 4], because the detector sensitivities in epithermal neutron energy range are much more uniform than those of other considered reactions, i.e., ^{197}Au(n,γ)^{198}Au, ^{151}Eu(n,γ)^{152m}Eu, ^{127}I(n,γ)^{128}I, ^{115}In(n,γ)^{116m}In, ^{55}Mn(n,γ)^{56}Mn, ^{37}Cl(n,γ)^{38}Cl and ^{23}Na(n,γ)^{24}Na. The detector is made similar to the original one [5]. The dimensions of the detector are 63.2 mm in height and 65.2 mm in diameter. In the detector, the activation material (gallium foil with a diameter

of approximately 10 mm and a weight of approximately 50 mg) is positioned in the geometrical center of the polymethyl methacrylate (PMMA) cylinder (neutron moderator, 63 mm in height and 65 mm in diameter) covered with 0.1 mm thick Cd foil as thermal neutron absorber. Unlike the original detector, our detector is made of the PMMA, not the high-density polyethylene (HDPE), uses pure gallium in the form of a foil as an activation material, and not a GaN wafer, and the thickness of the cadmium foil is 0.1 mm, not 0.05 mm.

The isomeric view of the detector is shown in figure 1. The detector is made collapsible in order to remove activated gallium foil and measure its activation. Gallium is placed in a recess of a PMMA disk 20 mm in diameter and 7 mm thick, which is wound onto a part of the moderator made in the form of a truncated cone. A cone with a wound disk is screwed into the main body of the detector so that the activation material is positioned in the geometrical center of the PMMA cylinder. The bases and side surface of the resulting cylinder are covered with 0.1 mm thick cadmium foil.

Of course, gallium can not be removed from the detector and measured without disassembling the detector. However, in this case, in addition to the gallium emission lines, there will be a cadmium emission line, which although different in energy is more intense. Additional loading leads to an increase in the dead time of the spectrometer and to a systematic error in the measurement.

The intensity of γ-radiation is measured by a high purity germanium γ-ray spectrometer (SEG-1KP-IPTP 12 produced by Institute of Physical and Technical Problems, Dubna, Russia).

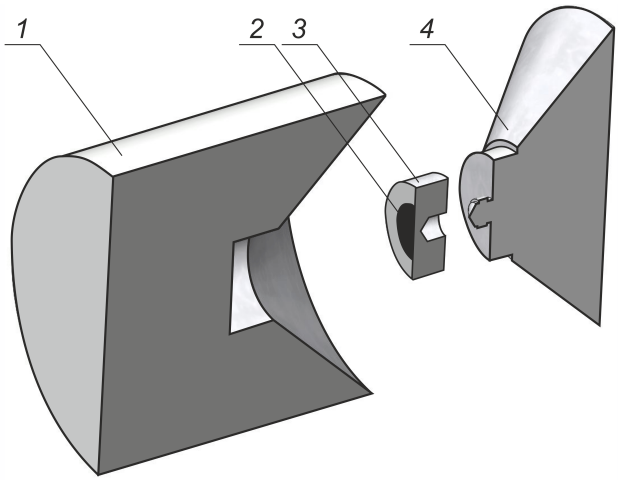

Figure 1. Schematic view of the epithermal neutron flux detector: 1 — main part of the PMMA cylinder, 2 — gallium foil, 3 — foil placement disk, 4 — screw-in part of the PMMA cylinde.

3 Detector sensitivity simulation

The ^{71}Ga(n,γ)^{72}Ga reaction cross section is shown in figure 2. It can be seen that in the epithermal energy range there are a number of resonances with a high value of the reaction cross section, which ensures the selective efficiency of the detector to epithermal neutrons. The use of HDPE as a moderator makes it possible, as shown in [5], to achieve a flat sensitivity curve in the epithermal neutron energy range, while its sensitivities to thermal and fast neutrons are low.

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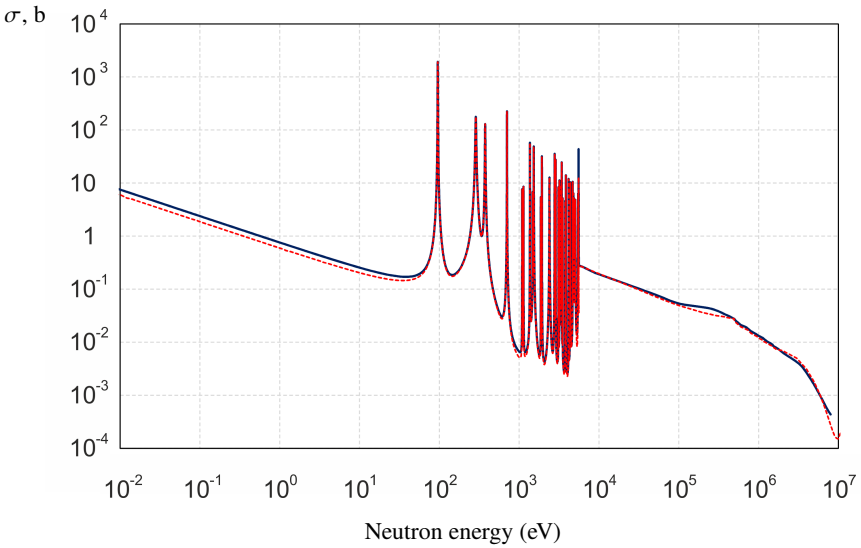

Figure 2. ^{71}Ga(n,γ)^{72}Ga reaction cross-section: in the ENDF-VII library (blue solid line), in the JENDL-4.0 library (red dashed line) [6].

Detector sensitivity for a range of neutron energies is simulated by the Monte Carlo method using the NMC code [7] and ENDF-VII library of evaluated incident-neutron data. Noted that the neutron flux determined by the detector is a cell flux. The neutron source is set in a circle with a diameter of 6.52 cm, located at a distance of 10 cm from the PMMA cylinder. The mass of gallium is taken equal to 51.2 mg, the content of ^{69}Ga isotone is taken equal to 60.2%, ^{71}Ga — 39.8%. The detector temperature is set to 20°C. The results of the simulation are shown in figure 3. The results of calculating the sensitivity of the original detector with the same mass of gallium in a gallium nitride plate for comparison are also presented there.

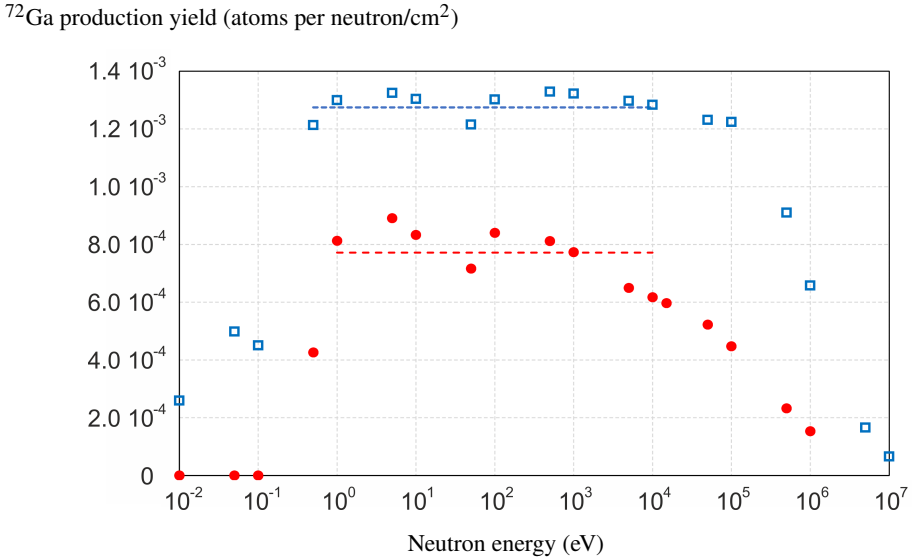

Figure 3. Calculated sensitivities of the epithermal neutron flux detector: solid circles — calculated by us, squares — from [5]. The sensitivity averaged over the neutron energy range from 1 eV to 10 keV is shown by the dashed lines.

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The first thing you immediately pay attention to is the significant difference between these two results. The calculated sensitivity of our detector averaged over the energy range from 1 eV to 10 keV is $\eta = (7.7 \pm 0.9)10^{-4}$ count/(n/cm^2), which is 1.6 times less than the similar value of the original detector.

A third of this difference is explained by the use of different materials. The concentration of hydrogen atomic nuclei in PMMA is lower than in HDPE and neutron moderation occurs over a longer distance. As a result, the detection efficiency of the PMMA detector is 20% lower according to comparative calculations.

Part of this discrepancy is explained by the difference in the used values of the ^{71}Ga(n,γ)^{72}Ga reaction cross section. We used the ENDF-VII library in simulations, and the authors of [5] used the JENDL-4.0 library. In figure 2 shows that if in the resonance region the reaction cross sections are practically the same, then in the region of energies below the resonances, the cross section in ENDF-VII library approximately 1.3 times larger than in JENDL-4.0 library. There is nothing surprising. Sometimes the data in different libraries can differ, such as the values of the ^{7}Li(p,α)^{4}He reaction cross section [8].

If in the epithermal energy range the sensitivity of our detector and the sensitivity of the original detector, although they differ by 1.6 times, are similar, then in the region of thermal neutrons or fast neutrons the difference in behavior will be significant.

Thus, in our simulations, the detector is not sensitive to thermal neutrons (< 0.4 eV), and this result seems natural. First, the detector is wrapped in cadmium, which absorbs thermal neutrons (note that we are using twice the thickness of the cadmium foil). Second, even if neutrons reach gallium, the probability of their absorption by gallium will be small, since the reaction cross section will be only a few units of barn, as can be seen in figure 2. And third, thermal neutrons are efficiently absorbed by hydrogen. The second and third arguments can be combined and presented as follows. Although the cross section for radiative capture of a thermal neutron (0.025 eV) by gallium is 3.5 times larger than the cross section for radiative capture by an atomic nucleus of hydrogen (1.16 b and 0.33 b, respectively), hydrogen atomic nuclei in the PMMA cylinder weighing 240 g in 2.7×10^4 times more than gallium atomic nuclei weighing 52 mg. Therefore, a thermal neutron is much more likely to be absorbed by hydrogen than by gallium. So the high value of the thermal neutron detection efficiency of the original detector seems strange. It is possible that the authors of [5] did not take into account the absorption of neutrons by hydrogen and cadmium, or did not set the temperature of the detector, or both.

So, summing up the above: 1) Despite the differences in the efficiency of neutron detection by our detector and the original detector, they have a general similarity: the detector is sensitive to epithermal neutrons and it has a flat sensitivity curve in epithermal neutron range, while its sensitivities to thermal (< 0.4 eV) and fast (> 10 keV) neutrons are low. This property allows the detector to be used to determine the epithermal neutron flux of the BNCT neutron beam. 2) To use the detector in practice, it is necessary to eliminate the discrepancy in the detection efficiency, which can be achieved both by joint research and by independent calculations, or by both.

4 Experimental validation

Experimental verification of the detector was carried out at the VITA accelerator based neutron source at the Budker Institute of Nuclear Physics, Novosibirsk, Russia [9]. The detector is placed along the proton beam axis at a distance of 100 mm from the lithium layer of the neutron generating target. PMMA disks 200 mm in diameter and 12 mm thick are placed between the target and the detector close to the target, from 0 to 6 pcs. The target is irradiated with a 2 MeV proton beam with a fluence of 2.5 C for 1 hour. After that, it is disassembled, the HPGe spectrometer measures the activation of the gallium foil, corrects for the mass of gallium, and restores the epithermal neutron flux density.

The epithermal neutron flux density normalized to 1 mA proton beam current is shown in figure 4. Although the stability of the proton energy and the accuracy of its measurement are high (0.1% and 0.3%), however, the uncertainty of the neutron yield is much higher — about 7%, due to the closeness of the energy of 2 MeV to the threshold of the ^{7}Li(p,n)^{7}Be reaction [10]. The absolute sensitivity of the HPGe-spectrometer for the 834 keV γ-radiation line is determined with an accuracy of 5% [11]. The statistical measurement error was 1%. As a result, the total measurement error was 9%.

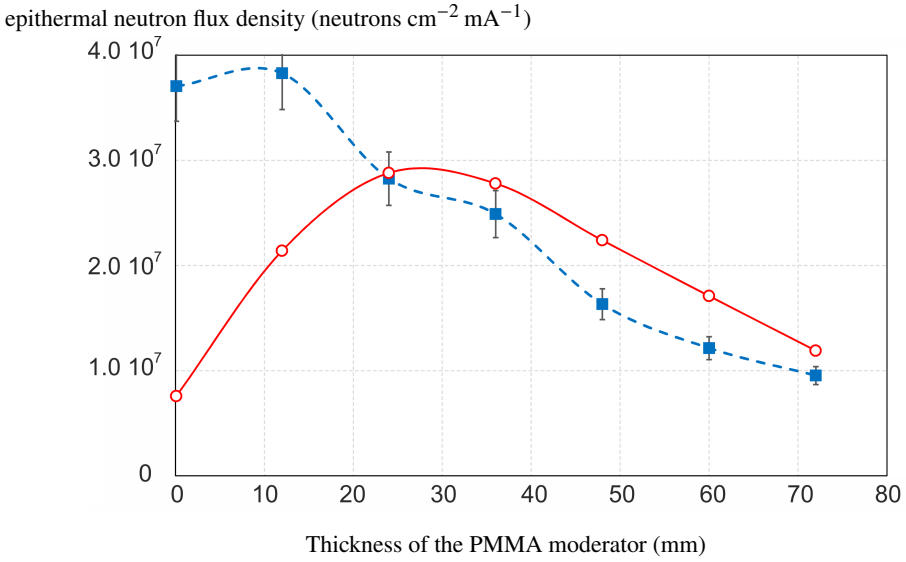

Figure 4. Dependence of the epithermal neutron flux density on the thickness of the PMMA moderator at a proton beam energy of 2 MeV: blue dashed line — measured, red solid line — simulated.

It can be seen that the results of the experiment are consistent with the results of simulations for a PMMA thickness of more than 24 mm. At a smaller thickness, the results differ greatly due to the significant number of fast neutrons. The neutron energy spectrum for several PMMA thicknesses is shown in figure 5. It can be seen that without the use of a moderator most of the neutrons are fast and they make a significant contribution to the activation of gallium although the detector's sensitivity to them is less than to epithermal neutrons. As the moderator thickness increases the spectrum shifts towards the epithermal energy range and the measurement results become closer to the simulation results.

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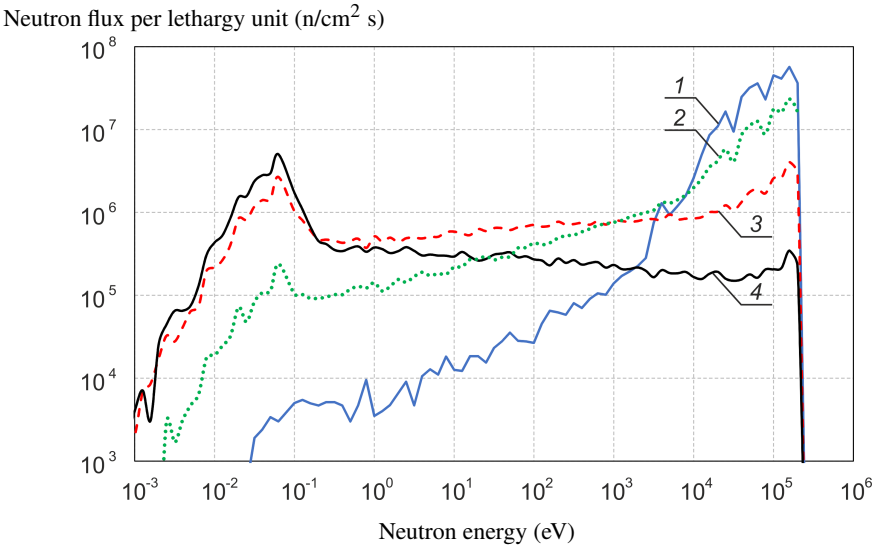

Figure 5. The neutron energy spectrum for several PMMA thicknesses: 1 — 0 mm, 2 — 12 mm, 3 — 36 mm, 4 — 72 mm.

5 Detector improvement

The relatively high sensitivity of the detector to fast neutrons can be reduced by using filters such as titanium. The cross section of neutron elastic scattering on the ^{48}Ti atomic nucleus is in figure 6. It can be seen that there is a noticeable resonance in the region of 10 keV — just at the border of the division of the neutron spectrum into epithermal and fast ones. We use this property to improve the detector: we add a titanium disk 65 mm in diameter and 6 mm thick to the base of the cylinder facing the neutron flux. The addition of this titanium disk scatters the neutrons and reduces the sensitivity of the detector.

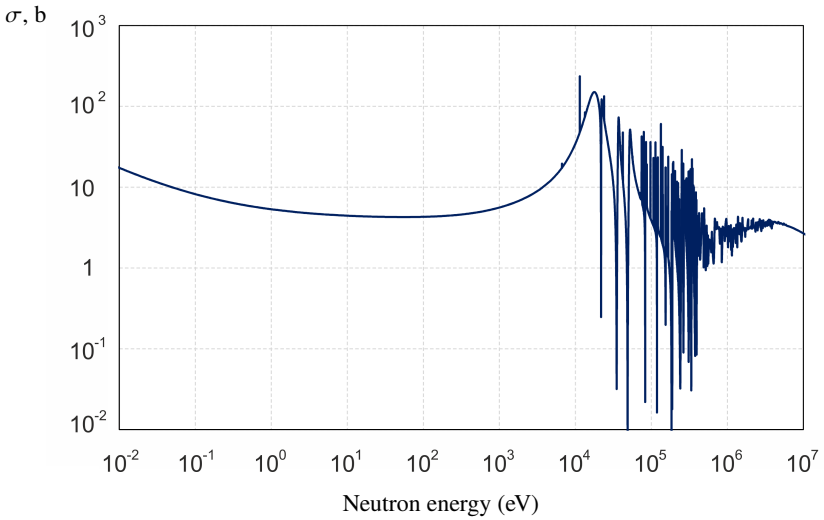

Figure 6. The cross section for elastic scattering of neutron on the ^{48}Ti atomic nucleus.

Figure 7 shows a graph of how much the addition of disk reduces the sensitivity of the detector. It can be seen from this graph that if in the epithermal energy range the detector sensitivity decreases by approximately of 10%, then for 15 keV neutrons it will decrease by 56%.

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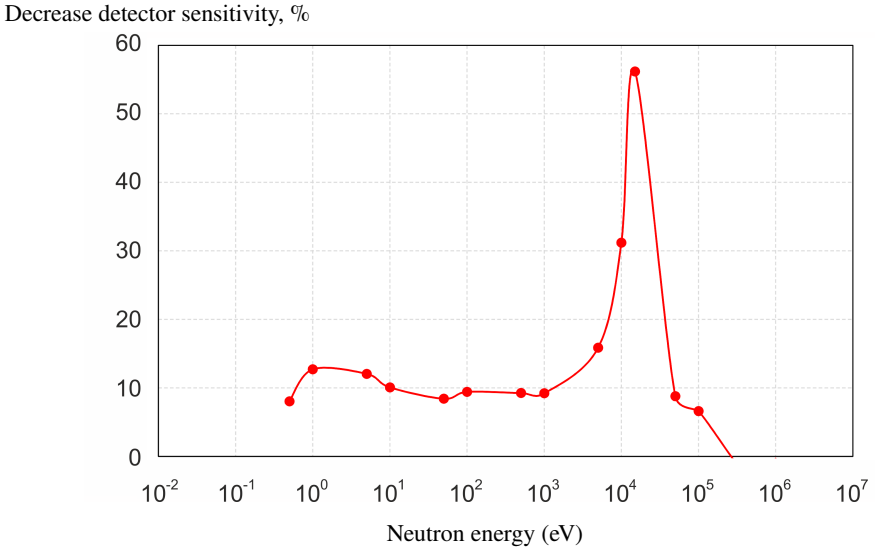

Figure 7. Decreased detector sensitivity due to the addition of a titanium disc.

This improvement reduces the sensitivity of the detector to neutrons in the region of 10–30 keV and makes it more selective for the epithermal energy range. At the same time, it is believed that neutrons with energies of 1 to 30 keV are optimal for deep-seated tumor treatment (see figure 4.11 in [1]). Also in the same book [1] on page 43 Prof. A. Kreiner states that "These neutrons have to moderated . . . to epithermal energies (energies in the interval 0.5 eV–10 keV, ideally centered near the upper end of this range) for deep-seated tumors". It follows that the information on the flux of neutrons with energies from 1 to 30 keV is important for planning the therapy of deep-seated tumors. Such information is obtained by comparing the values measured by identical detectors, one of which is additionally equipped with a titanium disk. If only epithermal neutrons are present in the neutron spectrum, then the use of a titanium disk will reduce the signal by 10%. In our experiments (figure 4) the detector signal when using a titanium disk decreases more than 10%, for example, by 15% with 52 mm moderator. This fact indicates in the neutron spectrum a noticeable content of a component with an about 10 keV, which is optimal for the treatment of deep-seated tumors.

6 Conclusion

To measure the epithermal neutron flux which is important for planning boron neutron capture therapy a cylindrical activation detector using ^{71}Ga(n,γ)^{72}Ga reaction similar to the previously proposed one by a team of Chinese and Japanese scientists was made. Numerical neutron transport simulation shows that the detector is sensitive to epithermal neutrons and it has a flat sensitivity curve in epithermal neutron range, while its sensitivities to thermal and fast neutrons are low. It is noted that the calculated efficiency of the detector differs from the calculated efficiency of the original detector. Assumptions are made about the possible reasons for the difference in efficiency values and ways to eliminate them are proposed. It is also proposed to equip the detector with a titanium disk, which further reduces the detector's sensitivity to fast neutrons. It is shown that a comparison of the signals from a detector with and without titanium disk makes it possible to

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estimate the component of neutrons with energies in the region of 10 keV, which is optimal for the treatment of deep-seated tumors.

Thus, the cylindrical activation detector using ^{71}Ga(n,γ)^{72}Ga reaction can become a reliable tool in planning boron neutron capture therapy after its further experimental and computational validation.

Acknowledgments

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References

[1] W. Sauerwein, A. Wittig, R. Moss and Y. Nakagawa, eds., *Neutron Capture Therapy*, Springer Berlin Heidelberg (2012), 10.1007/978-3-642-31334-9.

[2] M.A. Dymova, S.Y. Taskaev, V.A. Richter and E.V. Kuligina, *Boron neutron capture therapy: Current status and future perspectives*, *Cancer Commun.* **40** (2020) 406.

[3] X. Guan, M. Manabe, I. Murata and T. Wang, *Design of an epi-thermal neutron flux intensity monitor with gan wafer for boron neutron capture therapy*, *J. Nucl. Sci. Technol.* **52** (2015) 503.

[4] X. Guan, M. Manabe, S. Tamaki, S. Liu, F. Sato, I. Murata et al., *Experimental study on the performance of an epithermal neutron flux monitor for BNCT*, *Appl. Radiat. Isot.* **113** (2016) 28.

[5] X. Guan, Y. Gong, I. Murata and T. Wang, *The new design and validation of an epithermal neutron flux detector using ^{71}Ga(n,γ)^{72}Ga reaction for BNCT*, 2019 *JINST* **14** P06016.

[6] Java-based nuclear information software JANIS, https://www.oecd-nea.org/jcms/pl_39910/janis.

[7] D. Yurov, A. Anikeev, P. Bagryansky, S. Brednikhin, S. Frolov, S. Lezhnin et al., *Parameters optimization in a hybrid system with a gas dynamic trap based neutron source*, *Fusion Eng. Des.* **87** (2012) 1684.

[8] S. Taskaev, M. Bikchurina, T. Bykov, D. Kasatov, I. Kolesnikov, A. Makarov et al., *Cross-section measurement for the ^{7}Li(p,α)^{4}He reaction at proton energies 0.6–2 MeV*, *Nucl. Instrum. Meth. B* **525** (2022) 55.

[9] S. Taskaev, E. Berendeev, M. Bikchurina, T. Bykov, D. Kasatov, I. Kolesnikov et al., *Neutron source based on vacuum insulated tandem accelerator and lithium target*, *Biology* **10** (2021) 350.

[10] C. Lee and X.-L. Zhou, *Thick target neutron yields for the ^{7}Li(p,n)^{7}Be reaction near threshold*, *Nucl. Instrum. Meth. B* **152** (1999) 1.

[11] M. Bikchurina, T. Bykov, D. Kasatov, I. Kolesnikov, A. Makarov, I. Shchudlo et al., *The measurement of the neutron yield of the ^{7}Li(p,n)^{7}Be reaction in lithium targets*, *Biology* **10** (2021) 824.

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