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Photoneutrons for Radiation Therapy and Radionuclide Production

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Abstract

The possibility of organizing neutron therapy with a photoneutron beam produced by the electron accelerator target, and ensuring the required dose at the tumor at a reasonable exposure time and with minimal impact on patients investigated. Generation of neutrons from the target of electron accelerator takes place in two stages: $e^{-} \rightarrow \gamma \rightarrow n$, and in the selected electron energy range of 20-100 MeV, the bremsstrahlung gamma radiation in many times (~ 3 orders of magnitude) offers more than "useful" neutron yield. This raises the problem of the selective control of the "harmful" for radiotherapy secondary gamma radiation while providing the minimum attenuation of the neutron flux in the output beam. In order to solve the general problem of the formation of a neutron beam with necessary spectral characteristics having sufficient intensity, there has been resolved a number of computational tasks of the selection of the optimal configuration of the output beam unit and its composition. The matter of high importance is to minimize additional irradiation of the patient from the bremsstrahlung (generated by electrons) and secondary gamma radiation (generated by neutrons) from the accelerator target as well as from output unit's materials. On the other hand, at a generation stage $e^{-} \rightarrow$ γ the bremsstrahlung beam could be applied for effective radionuclide production by reactions (γ,n) and (γ,p) due to high leak intensity ~ $1.3 \cdot 10^{17}$ photon/s. By the $Mo^{100}(\gamma,n)^{99}Mo$ reaction the main diagnostic nuclide ^{99}Tc could be produced sufficiently for the clinical needs. The resulting configuration of the output unit provides the required beam quality for the neutron capture therapy (NCT), which commonly assumed to be the only competitive technology of neutron therapy on the background of the massive invasion of proton therapy and other highly selective techniques that ultimately damage the target sparing the surrounding tissues and organs. For the accessible accelerator (average current 4 mA and electron energy 35 MeV) the flux density of epithermal photoneutrons (they required for NCT) in the beam at the output is of the order of magnitude or more higher than the typical yield from existing and planned reactors' beams. The proposed scheme of generation and extraction of photoneutrons for NCT has a number of strong advantages over traditional techniques: a) the applying of electron accelerators for neutron production is much safer and cheaper than conventional reactor beams in use; b) accelerator with the target, the beam output unit with the necessary equipment can be placed

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on the territory of the clinic without any problems of radiation safety; c) the proposed target – liquid gallium, which also serves as a cooler, is an "environmentally friendly" material due to low activation which rapidly (in ~ 4 days) falls to the background level.

Keywords: electron accelerator, epithermal neutrons, neutron capture therapy, bremsstrahlung beam for radionuclide production

1. FOREWORD

A powerful photoneutron source for medicine described in [1]. In [2], the optimized configuration of the photoneutron beam forming unit for neutron capture therapy (NCT) presented, and in [3, 4] the thermohydraulics of the composite flow target (W + Ga) and the beam applicability for radiation therapy investigated. The stationary fragment of the target – a matrix of refractory tungsten, through which gallium flows, allows a sharp increase in the yield of photoneutrons in comparison with the target only from gallium. In order to normalize the calculation results, an average current of 4 mA of the available MEVEX accelerator [5] was used on electron energy of 35 MeV. There is a distinct advantage of the proposed neutron generation scheme for NCT in comparison to reactor-based generation.

First, from the ecological viewpoint, the activity of liquid gallium drops to the background for 4 days, there is no fission products in the unit, and the activation of the equipment is strictly localized.

The natural gallium consists of a mixture of two isotopes: ^{69}Ga (60.1%) + ^{71}Ga (39.9%). It is a low-melting metal (mp = 29.8 ° C) with a density of 5.904 g/cm^3 in the solid state and 6.095 g/cm^3 in the liquid state. When smelted, gallium remains in the liquid phase for a long time at room temperature. The activation of natural gallium occurs due to photoreactions and intrinsic neutrons conversion. The main processes: ^{69,71}Ga(γ, n)^{68,70}Ga, ^{69,71}Ga(n, 2n)^{68,70}Ga, $^{69.71}$Ga(n, γ) $^{70.72}$Ga lead to the short-lived products ^{68}Ga ($T_{1/2}$ = 68.3min), ^{70}Ga ($T_{1/2}$ = 21.2 min) and ^{72}Ga ($T_{1/2}$ = 14.1 hours). The calculations show that while generating the neutron fields acceptable for NCT and when the working medium of the target is circulated, the total activity of gallium (for typical irradiation scenarios and the number of fractions) falls to the level of the natural background in a time not exceeding 4 days (Fig. 1).

Furthermore, from the radiation and nuclear safety viewpoint, it is ultimately safer than reactor generation. Radiation safety, as well as the relatively small dimensions and weight of the unit, allows the unit to be placed directly at the clinic. Finally, the flux

Table 1: Flux density, spectrum and average energy of neutrons at the outlet of the reference (FCB MIT), designed (MARS) and existing (TAPIRO) reactor beams in comparison to characteristics of the photoneutron beams.

		Φ_{tot}, cm^{-2}s^{-1}, 10^9	Φ_{epi}/Φ_{tot}, %	Φ_{fast}/Φ_{tot}, %	Φ_{therm}/Φ_{tot}, %	$E^{\Phi}{}_{aver}$, MeV
Values, desirable for the NCT		> 1	~100	→ 0	→ 0	–
FCB MIT		4.2	?	?	?	?
MAPC		1.24	81.6	13.4	5.0	0.0337
TAPIRO		1.07	73.6	6.5	20.0	0.00857
Photo-nuclear beams	“best” version [4]	18.5	74.9	25.1	0.014	0.0345
	current beam	27.8	73.3	21.6	5.11	0.0325

Table 2: NCT characteristics at the outlet of reactor and photonuclear beams: epithermal neutron flux density, “poisoning” of a beam with gamma radiation and fast neutrons, directivity.

		Φ_{epi}, cm^{-2}s^{-1}, 10^9	D_{γ}/Φ_{epi}, sGy·cm^2, 10^{-11}	D_{fast}/Φ_{epi}, sGy·cm^2, 10^{-11}	J_{epi}/Φ_{epi} (“flux-to-current»)
Values, desirable for the NCT		≥ 1	< 2–5	< 2–5	≥ 0,7
FCB MIT		?	1.3	4.3	0.8
MAPC		1.01	5.38	11.8	0.8
TAPIRO		0.788	6.77	8.49	0.8
Photo-nuclear beams	“best” version [4]	13.9	0.0407	15.9	0.8
	current beam	20.4	0,0262	13.4	0,8

Table 3: The target characteristics for Model 3 mode of the radionuclide production.

Target material	Tl	Pb	Bi	^{238}U	Pb + Bi (45% +55%)
R, cm	1.0	0.75	0.75	0.50	0.75
H, cm	1.0	0.75	1.0	1.0	1.5
Density, g/cm^3	11.843	11.342	9.79	19.05	10.6
Melting point, ^{o}C	304	324	271	1133	124
Bremsstrahlung yield, s^{-1}	1.29.10^{17}	1.32.10^{17}	1.34.10^{17}	1.25.10^{17}	1.33.10^{17}
Average energy, MeV	14.7	15.9	15.6	15.5	13.8

density of epithermal neutrons (required for NCT) at the output of the beam is at least

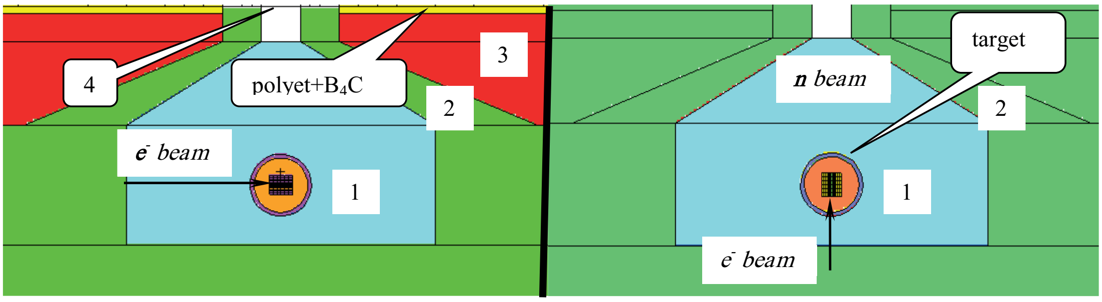

Figure 1: Axial sections of the axisymmetric beam extraction unit for NCT: “best” version from [4] to the left and modernized version to the right (MCNP5 [6] input visualization). The fragments of the removal unit with a collimation system are presented: a channel filled with a spectrum shifter (1, lead fluoride PbF_2, also performs the function of a gamma filter); the channel is surrounded by the cone collimator (2, Pb, the main function is the slowing down and canalization of neutrons); zirconium hydride $ZrH_{1.8}$ (3) in the collimating system is a light shielding; the borated polyethylene and Cd plate 1 mm thick (4) at the outlet of the channel are a thermal neutron filter.

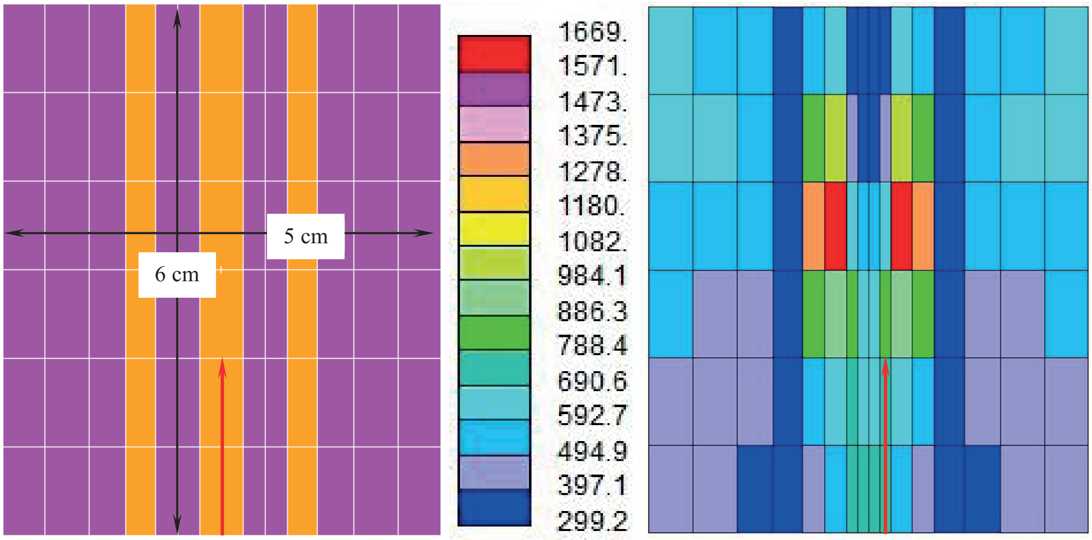

Figure 2: Axial sections of two cylindrical axisymmetric models of the target: on the left for calculating the energy release (MCNP5 code), on the right for calculating the temperature (STAR-CD® code [7]); in the center – temperature scale (°K). The arrow shows the input point of the electrons, the temperature data correspond to the coolant velocity of 2 m/s.

an order of magnitude higher than the neutron flux density of existing and projected reactor-based beams.

In this paper, a multifunctional application of the electron beam in radiation medicine is described: a) the beam output unit for NCT has been renovated in order to increase the photoneutron flux density at the beam output (for the same target and the distance ”target-beam output”) by 1.5 times in comparison with [4]; the modernization of the unit consisted in replacing the hydrogen-containing shielding of the block and removing the cadmium plate at the beam output (the rationale provided below in the text of the paper); b) the possibilities of radioisotope generation by the reaction (n,γ) are investigated while maintaining the configuration of the output block; c) radioisotope

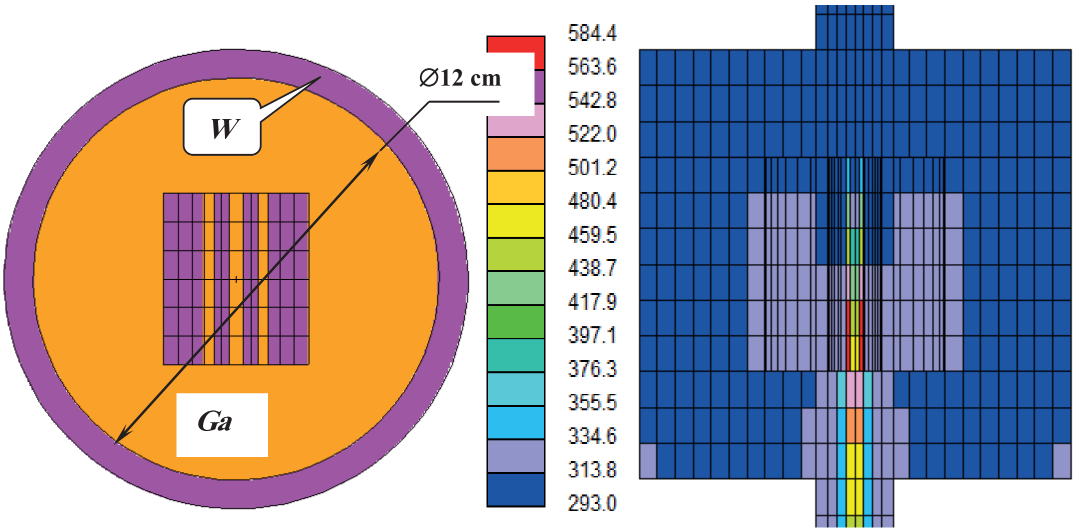

Figure 3: Models of the modernized target: on the left to calculate the energy release, on the right to calculate the temperature field. Coolant velocity ~ 2 m/s, maximum temperature 310 °C.

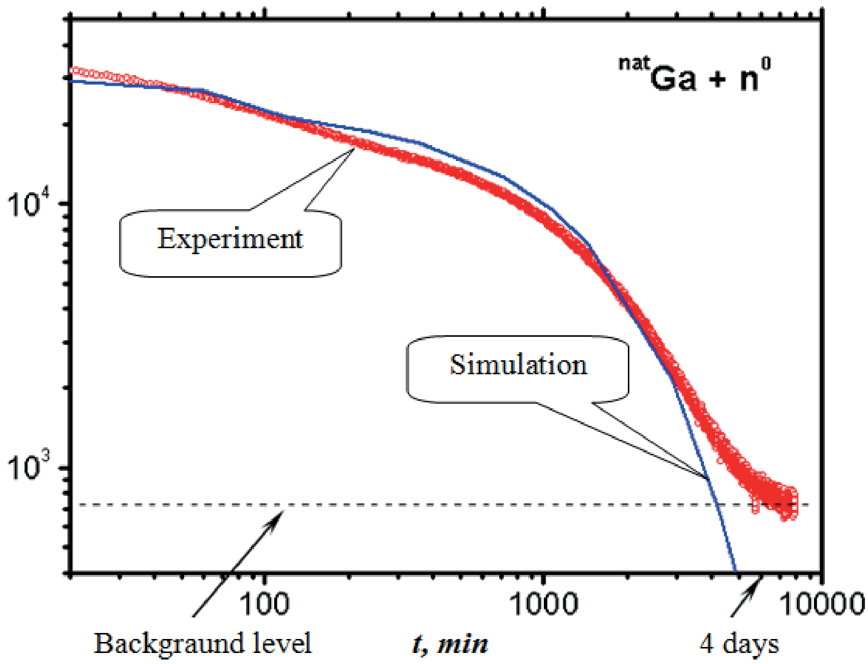

Figure 4: Activity decay of Gallium after typical irradiation scenario (rel. units).

generation for an output unit with a "booster" (converter of thermal neutrons CTN with keff ≤ 0.90) is studied; d) finally, the possibilities of radioisotope generation by the (γ, n) and (γ, p) reactions are investigated. The most promising was the latest approach.

One of the most important reaction is ^{100}Mo (γ,n)^{99}Mo → ^{99}Tc, which allows getting Technetium-99, that is used almost in 90% of the procedures in radionuclide diagnostics. The calculations of ^{99}Mo generation rate by the gamma radiation of the target from the lead-bismuth eutectic are performed. A high yield of bremsstrahlung from

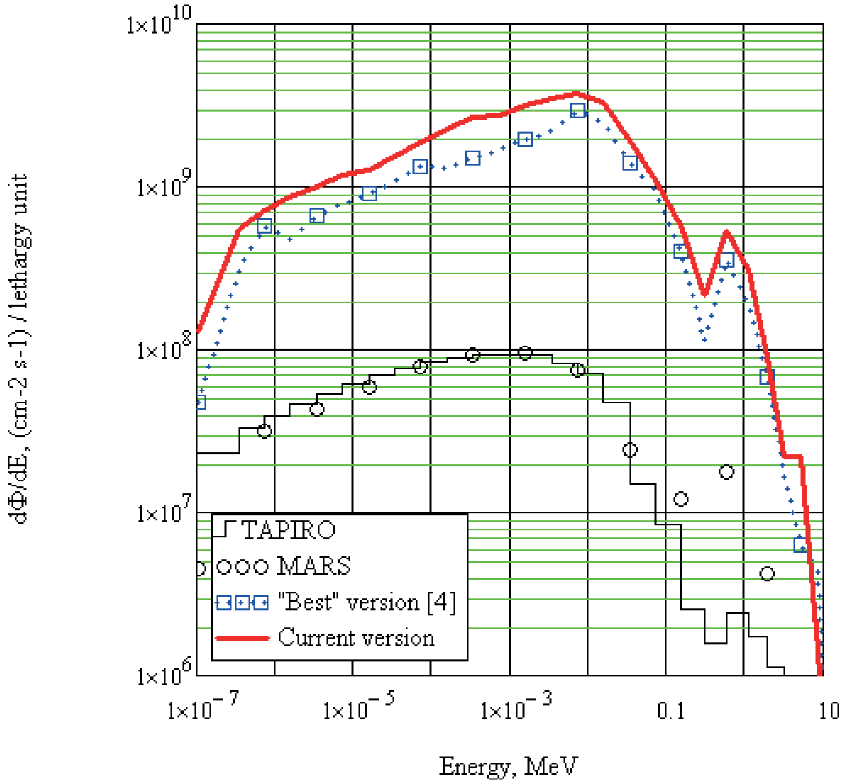

Figure 5: Neutron spectra at the outlet of the beam channel for NGT.

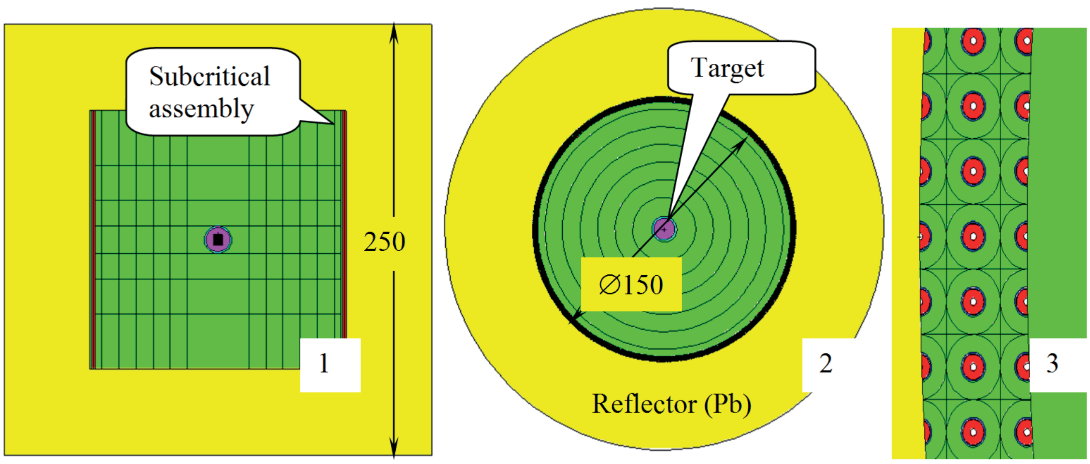

Figure 6: Radial (1) and axial (2) sections of Model 2 composition; 3 – fragment of radial section with subcritical assembly. All dimensions are in *cm*.

the target (~1.3·10^{17} photon/s) for the accepted electron beam characteristics (average current 4 mA, electron energy 35 MeV) provides a high efficiency of ^{99}Mo production. The generation of other radioisotopes has also been studied, and the characteristics of the heat dissipation of the radiation energy released by the lead-bismuth eutectic were estimated. Further, calculations of beam characteristics for a combined target

(W + Ga) with a hollow channel of beam extraction, which can be used for combined neutron-gamma therapy and for radiobiological studies, were performed.

2. MATERIALS AND METHODS

The beam removal unit version with the maximal output flux density was selected [4] to proceed with an optimization. In Fig. 1 the axial sections of the optimal version of the beam output unit [4] and the updated version proposed in this paper are compared.

The beam extraction unit is an axisymmetric assembly of cylindrical and conical layers and carries protective and collimating functions (a conic layer of lead), as well as the function of the spectrum shifter destined to form epithermal spectrum required for NCT.

The combined flow target (Fig. 2, [4]) is a tungsten cylinder with cylindrical channels for the coolant (liquid gallium). In this paper, the target improved in the following way: the cylinder is enclosed in spherical tungsten casing filled with gallium to improve both heat removal and neutron production. The thermohydraulic calculation for the new target configuration is illustrated below (see Fig. 3).

In the interaction of accelerated electrons with a massive metal target W&Ga, the main energy loss channel is bremsstrahlung. At electron energies above ~14 MeV, the bremsstrahlung absorbed by the W&Ga nuclei generates neutrons in the (γ, n) reactions in the area of the so-called giant dipole resonance. Gallium is chosen as an accelerator target, because of its small induced activity which falls down quickly enough; herein the neutron yield is sufficient for the NCT providing. Thus, for characteristic irradiation at NCT, the target's activity decay up to background will occur practically during four days (Fig. 4). Besides, liquid gallium has necessary thermohydraulic characteristics as the coolant: a) low fusion temperature 29.8 ^{o}C, and b) a wide range of liquid-phase temperature. It means that radiation heat released in the target could be easily carried away.

3. RESULTS & DISCUSSION

One of the modes of heat removal illustrated in Fig. 2. Note that of 140 kW beam power the amount that directly releases in the target is from 65 to 70 kW.

3.1. The NCT beam modernization

Additional calculations made it possible to do justifiable changes in the configuration and material composition of the beam output unit, which led to safely increase the main function, i.e. the density of the epithermal neutron flux at the beam exit. The changes are the following:

- the Cd plate at the outlet of the channel was removed, and the zirconium hydride was replaced by lead. The role of these elements in reducing the thermal neutron flux is negligible: epithermal neutrons entering the tissue generate the backscattered thermal neutrons right close to the entrance, which intensity considerably exceeds the thermal neutron flux out of the channel;
- the combined flow target was deployed along the axis of the neutron beam exit and enclosed in a spherical tungsten body filled with gallium. This improvement leading to the higher heat sink as well as increasing the generation of neutrons and reducing the yield of "harmful" bremsstrahlung;

3.2. Beam quality for NCT

The "in air" functionals characterize the radiation field at the output of a beam without an irradiated phantom and simplify the task of selecting the optimal configuration and composition of the output unit materials (without the laborious calculations of the "in phantom" functionals). It is assumed that if the "in the air" characteristics satisfy specific criteria worked out by the world community, then it is to be expected that the "in phantom" functionals [mono] will also satisfy the requirements of NCT.

For comparison of the computed beams from a target of the electronic accelerator, the neutron beams' characteristics of existing and projected reactors were used:

- the FCB MIT beam, that is "reference" for the NCT (measurements, [8, 9]), is decommissioned at the time;
- the epithermal column beam of the fast TAPIRO ([([10-14])]) reactor which is intended for application in the NCT (the calculation confirmed with measurements; the beam is decommissioned);
- the beam of specialized medical MARS reactor (calculation, [15, 16]).

Here are the generally accepted meanings of the basic "in air" criteria (see, for example, [17-19]):

- flux density of epithermal (0.4 eV <E <10 keV) neutrons

$$\Phi_{epi} \geq 10^9\ cm^{-2}s^{-1}; \tag{1}$$

- the ratio of the absorbed gamma radiation dose rate to the epithermal neutron flux density

$$D_\gamma/\Phi_{epi} < (2 \div 5)\cdot 10^{-11}\ sGy\cdot cm^2; \tag{2}$$

- the ratio of the absorbed dose rate of fast (E> 10 keV) neutrons to the epithermal neutron flux density

$$D_{fast}/\Phi_{epi} < (2 \div 5)\cdot 10^{-11}\ sGy\cdot cm^2; \tag{3}$$

- the ratio of the epithermal neutron axial current of to the flux

$$J_{epi}/\Phi_{epi} > 0.7. \tag{4}$$

Moreover, for a decrease of “harmful” dose at an entrance to the tissue region, the following requirements are important: $\Phi_{\rm epi}\to\Phi_{\rm tot}$; $\Phi_{\rm fast}\to 0$; $\Phi_{th}\to 0$(Φ_{th}– thermal neutron flux density).

Base values of the ”in air” characteristics for the compared beams are given in Table 1. For photoneutrons, the data on “best” previous version [4] and updated current version of the removal unit are submitted (see Fig. 1, the picture on the right). Actual NCT criteria presented in Table 2. According to the data, one might conclude that for ”in air” criteria (or ”for free beam”) the offered photo-neutron beams don’t concede and even partly surpass reactor beams for the NCT. This conclusion confirmed as it is shown in the Fig. 5 in which spectral characteristics of neutrons at the beam outlet presented.

3.3. Radioisotope production

Simplest model 1

For radioisotope production on the first model in (n,γ) reactions the conic moderator from the lead difluoride (PbF_2) has been replaced with heavy water (see Fig. 1). The general configuration of the removal block did not change when the irradiated samples placed at the outlet of the channel.

It has turned out that essential thermalization of a beam neutrons with such small moderator depth of (∼ 0.5 m) could not be achieved: at $\Phi_{\rm tot}$ = 3.10 · 10^{10} $\rm cm^{-2}s^{-1}$

thermal neutron flux density only Φ_{th} = 1.24 · 10^{10} $cm^{-2}s^{-1}$. At the same time in close proximity to a target, the thermal neutrons flux density reaches ∼ 2.5 · 10^{10}. In comparison to the density of thermal neutron flux in the reactor core, it is clear that the first model for radioisotope production is out of use.

Model 2: target with subcritical booster

The model consisting of a cylindrical tank filled with heavy water presented at Fig. 6. In the center of a tank there is a model, and on the periphery – subcritical assembly of k_{eff} < 0.90. Assembly consists of shortened fuel elements of the BN-600 reactor, cooled by heavy water. The moderator is also D_2O.

The performed calculation show that required neutron field in a tank with heavy water could be achieved. The maximum value of total flux density Φ_{tot}= 6.19.10^{11} cm^2s^{-1} is in close proximity to a target, maximum of thermal neutron flux density Φ_{th} = 3.09 · 10^{11} $см^{-2}с^{-1}$ is spaced from a target on ∼ 21 cm.

Neutron flux density has multiplied more than 10 times compared with the results received for the first model. Perhaps, for some conditions the radioisotope production in (n, γ) reaction on model 2 is reasonable, but it cannot compete with reactor production.

Model 3: (γ, n) and (γ, p) reactions

It has turned out that the Model 3 is the most perspective, as the yield of bremsstrahlung from a target is reasonably high. The studied cylindrical targets have been optimized to get maximum yield of bremsstrahlung when electron beam (radius 0.5 cm) falls on an end face of the cylinder (Table 1). At the chosen parameters of an electron beam, the bremsstrahlung yield from optimal targets is almost identical for all heavy materials. The average energy of the bremsstrahlung is in the area of a giant dipole resonance. For technological reasons as a target the eutectic lead-bismuth is preferable; in this case this alloy will be as well the coolant.

The efficiency of the Model 3 is demonstrated by a numerical experiment. For a target from the eutectic "lead-bismuth" (Table 3) which is placed in the "thin" sphere from 100Mo the reaction rate 100Mo(γ, n) 99Mo is counted. Internal radius of the sphere is 1 cm, the thickness of a spherical layer is 1 cm. Under these conditions the reaction rate ∼ 1014 s-1 cm-3, that is quite satisfactory for radioisotope production.

4. CONCLUSIONS

The photoneutron beam from the target of the electron accelerator, intended for neutron capture therapy, renovated by means of precise calculations of radiation transport and thermohydraulics of the target. As a result, the intensity of the neutron beam at the channel outlet increased for 1.5 times.

The opportunities of the radioisotope production on a photoneutron beam were investigated. It has been established that the use of the reaction (n, γ) is not very promising (even with the use of a converter of the thermal neutron); the use of bremsstrahlung in reactions (γ, n) and (γ, p) is quite possible and even advisable (in particular, the production of technetium-99 according to the ^{100}Mo (γ, n) 99Mo → ^{99}Tc scheme).

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