FEATURES OF PHOTONUCLEAR RADIONUCLIDE PRODUCTION IN THE THICK TARGETS

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One of the basic requirements for the production technology of radionuclides is to provide a high specific activity of the target nuclide as well as its full activity in target. Traditionally this problem is solved by using a large volume of photonuclear targets with subsequent by radiochemical separation target nuclide. Modeling and experimental study of the spatial distribution of the activity carried out in this work for the various targets. Optimal parameters of irradiation to achieve the maximum yield of isotopes Mo^{99}(Tc^{99m}) and Cu^{67} in targets from molybdenum and zinc of natural composition have been found.

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INTRODUCTION

In recent years the development of the photonuclear nuclide production method made it possible to imply in practice [1]. The advantages of this method include reliability, relatively low cost of electron accelerators and environmental safety. The production of ^{99}Mo isotope is of particular interest (as it is a generator of the most common isotope for diagnostics ^{99m}Tc), as well as the production of ^{67}Cu that is being considered very promising for immune therapy [2, 3].

One of the major tasks for the photonuclear technology is the conversion of the high-intensity electron beam (($\geq$10 kW/cm^2) to the flux of mixed X,n-radiation for target irradiation. The main component of the X,n-radiation is represented by high-energy photons of different origin: bremsstrahlung, Compton scattering and positron annihilation. The next components by their importance are neutrons and electrons. The interaction of all components of the irradiation field with matter leads to the production of the byproduct nuclides and to the activation and heating of the setup construction elements and output devices.

The contribution of neutrons, formed by (γ,Xn) reactions at the converter can be both positive and unwanted during the photonuclear isotope production. For example, while producing the $Tc^{99м}$ by generator method form the natural molybdenum target the Mo^{99} is produced mainly by the reaction Mo^{100}(γ,n)Mo^{99}. The existence of the photoneutrons allows for increase of the Mo^{99} yield due to the additional reaction channel Mo^{98}(n,γ)Mo^{99}. However, for ^{67}Cu production by the reaction ^{68}Zn (γ,p) ^{67}Cu in the case if natural zinc target the contribution of the ^{67}Zn (n,p) ^{67}Cu reaction is not significant because of low natural abundance of ^{67}Zn (4.1%).

1. MODELLING AND CALCULATIONS

The key characteristics of the photonuclear isotope production technology are the specific activity and the product yield. In the cases of, for example, ^{67}Cu and ^{99m}Tc the non-optimal target shape and size may lead to considerable reduction of these quantities. The problem of choosing the target shape and size then reduces to a simpler problem of activation of the cylindrical samples by the particle flux of anisotropic density.

There are several known methods to solve this problem: analytical solution for the interaction of the high-energy bremsstrahlung with the thick (technological) target and the computer simulation of the physical processes using the Monte-Carlo methods. The first was used in the work [4]. The yield values based on this method are overestimated for the materials with the low atomic number (Z<30) for energy range of 50…80 MeV. The isotope yield estimation was done by calculating the micro-yields of the corresponding reactions along the trajectories of all above-threshold photons using the specifically developed programs based on the transport code PENELOPE-2008 [5].

In present work we use the computer simulation of the radiation and matter interaction based on the GEANT4 library to estimate the isotope yields from the finite size targets. We included the multiple scattering, bremsstrahlung generation and photonuclear reactions in the simulations.

One can calculate the activity and take into account the absorption of the high-energy photons in the target by using the formula:

$$\frac{dY}{dt} = \frac{\sigma\rho}{M} N_{Av} \cdot \Phi \int_v e^{-\mu d} dv, \qquad (1)$$

where dY/dt – yield of new nuclei per unit of time; Φ – flux density of the activating radiation; σ – activation cross-section; d – distance from the target center to the integration volume dv; N_{Av} – Avogadro's number; M – molar mass of the target material; ρ – target material density. In the case of irradiation by bremsstrahlung from the linear electron accelerator one has to add a factor that takes into account the anisotropy of the beam of γ-rays. The exact analytical calculation of the angular distribution of the bremsstrahlung from a thick target can be hard to implement, so it is preferable to use the modified Shiff formula for thick radiator that was obtained in [6]:

$$\frac{J(\theta)}{J(0)} = \frac{-E_i\left\{-(E_{\gamma m}\theta)^2 \frac{\ln(183Z^{-1/3})}{1580 \cdot 8t_p}\right\} + E_i\left\{-\frac{(E_{\gamma m}\theta)^2}{1,78m_0}\right\}}{\ln\left\{\frac{1580 \cdot 8t_p}{m_0 \ln(183Z^{-1/3})}\right\} - 0,5772}, \qquad (2)$$

where J(θ) and J(0) are intensities at θ and zero angle respectively, E_i – exponential integral $E_i(y) = -\int_{-y}^{\infty} \frac{e^{-X}}{X} dx$;

$E_{\gamma m}$ – the maximum energy of the γ – quanta; Z – atomic number of the radiator material; t_p – radiator thick-

ness (normalized to the radiation length); m_0 – electron mass. The formula gives a satisfactory agreement with the experiment. In the 0…40 degree angle range of the emitted γ-rays the discrepancy with experiment does not exceed 12%.

The sample can be positioned in two different ways with respect to the converter during the irradiation, as shown on the Fig. 1: the sample axis is parallel to the beam axis (a), and perpendicular to the beam axis (b).

The target is irradiated with the converter of radius **r** and at the distance **a** from the converter. The target radius is R and target height is H.

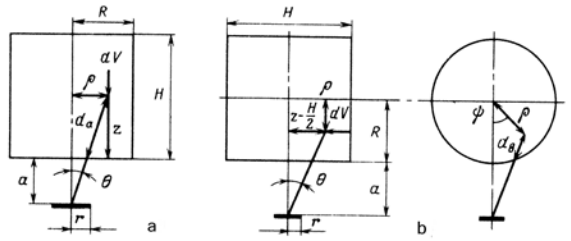

Fig. 1. Schematics of the bremsstrahlung converter and target position. a – irradiation "into cylinder's end"; b – irradiation "into cylinder's generatrix"

$$A_A = \frac{\sigma\rho}{M} N_{Av} \int_0^{2\pi} d\phi \int_0^H dz \int_0^R \Phi_A \exp(-\mu d_A) \frac{J_A(\theta)}{J_A(0)} \rho d\rho, \quad (3)$$

$$A_Б = \frac{\sigma\rho}{M} N_{Av} \int_0^H dz \int_0^R \rho d\rho \int_0^{2\pi} \Phi_Б \exp(-\mu d_Б) \frac{J_Б(\theta)}{J_Б(0)} d\varphi. \quad (4)$$

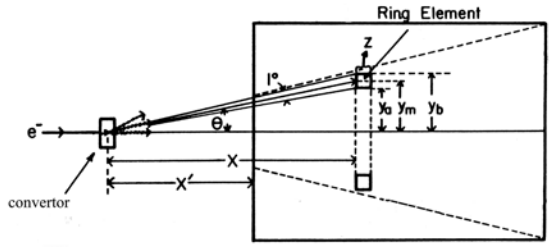

Fig. 2. The model of radiation to determine the accumulated activity

The calculation of the target activity is done by integrating the activity by the volume of the elementary ring (Fig. 2). The activity of the elementary ring is determined with the help of the formula (5), from [7]:

$$A_r = A(E,\theta) C_p \left(\frac{(2\,8)^2}{x^2 + y_m^2} \right) \cdot \exp\left\{ -0.1\mu \left[\sqrt{x^2 + y_m^2} - \sqrt{x'^2 + (x' y_m / x)^2} \right] \right\} \cdot \left[\pi z (y_b^2 - y_a^2) \right], \quad (5)$$

where A_r is a total activity of the ring volume; A – activity generated in 1g of the target material that is located at an angle θ to the electron beam; C_p – the density of the parent nuclide in the target; E – electron energy; μ – attenuation coefficient for the studied target; other symbols can be found on the Fig. 2.

The Fig. 2 shows the total and specific activities of ^{99}Mo in the natural molybdenum target that were calculated according to the above procedure using different distances in the case A where the target diameter is equal to the target height. In the case B the target irradiation leads to minor increase of the specific activity when the converter-target distance is larger than the target diameter.

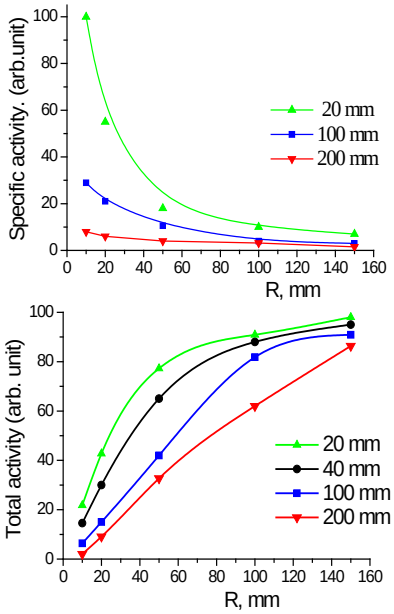

Fig. 3. The dependence of the total and specific activity generated by the radius of the target at distances from the converter to the target 20, 40, 100, 200 mm

2. EXPERIMENTAL METHODS

2.1. In order to determine the yield of the ^{99}Mo by the photonuclear reaction channel the device shown on the Fig. 3 was used. It is made of an aluminum tube that incorporates the converter C and target assembly T. The tube is aligned along the electron beam axis. In order to measure the total yield of the ^{99}Mo in (γ,n) and (n,γ)-channels at increased flux of moderated neutrons the tube with the converter and targets was placed inside the neutron moderator device (Fig. 4).

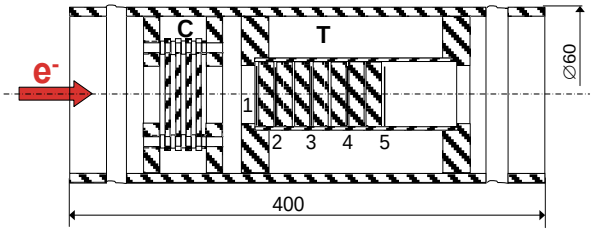

Fig. 4. The target device without a neutron moderator

The converter is made of 4 tantalum plates, each 1mm thick and separated by 1mm air gaps for cooling. The converter thickness is deliberately made more than optimum to lower the thermal stress of the isotope target. The target includes 7 discs of molybdenum, 19 mm in diameter and 3 mm thick each. There are 5 foils of molybdenum, 0.09 mm each, placed between discs, as shown on the Fig. 4. The foils are used for spectrometric analysis of the isotope yield without the influence of the self-attenuation of γ-rays in the target.

The neutron moderator is represented by the cylindrical graphite case with the wall thickness of 15 mm, internal diameter of 27 cm and height of 30 cm, filled with paraffin, that has an axial opening for incorporation of the converter and target assembly.

2.2. In order to measure the activities of the samples and identify the produced radionuclides we used the γ-spectrometer InSpector-2000 (made by Canberra) based on the HPGe detector. The detector energy range is

50…2000 keV, resolving power at 1332 keV is 1.75 keV, the relative uncertainty edges for point-like geometry at P=0.95 are 6 %.

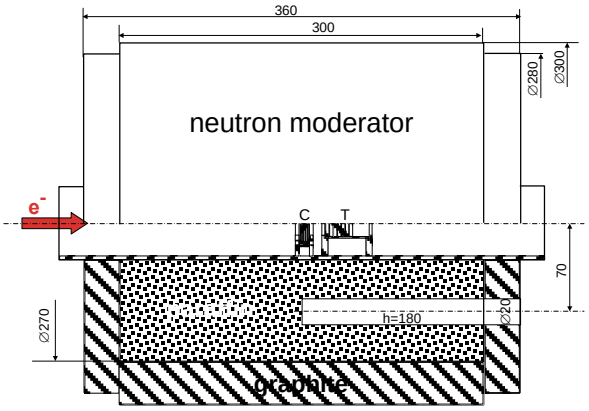

Fig. 5. The target device with a neutron moderator

2.3. The target irradiation is done as shown on the Fig. 5. The preference is given to obtaining of the maximum specific activity of the target isotope. The total activity produced in the target is no more than 20% of possible maximum that could be obtained with the same parameters of the bremsstrahlung beam.

The abovementioned γ-spectrometer was used to measure the activity of the irradiated samples when they were placed at 250 mm distance from the detector input window along the detector axis. The samples were hold for "cooling" in the safety container for a certain time in order not to overload the spectrometer and keep its dead time below 2%.

3. RESULTS AND DISCUSSION

The computer simulation of the ^{99}Mo production on the electron accelerator with maximum energy of 90 MeV was done. The converter and targets were placed inside the neutron moderator, represented by paraffin layer. The physical processes for electrons, positrons and γ-rays were described with the help of the class library GEANT4. Further calculations used the bremsstrahlung spectra calculated in the low-energy model. The simulation results (solid curves) as well as experimental data (points) for the ^{99}Mo production with and without neutron moderator for 60MeV incident energy of electrons are shown on the Fig. 6.

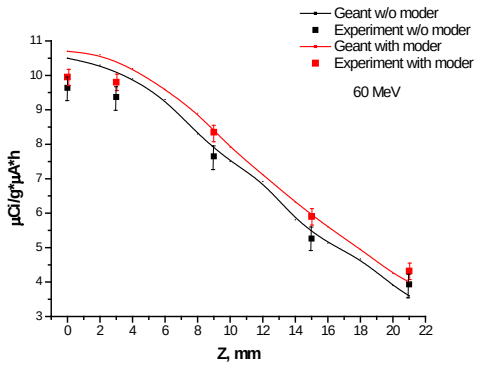

Fig. 6. Modeled yield ^{99}Mo at the electron energy 60 MeV

The measurements of the ^{99}Mo yield on the LU-40m accelerator were carried out with and without the neutron moderator for the 19 mm diameter and 21 mm high natural molybdenum targets. The target assembly was irradiated with mixed γ;n flux from tantalum converter during 1 hour with average electron beam current of ≈4 μA. After the target post-cooling the specific activities of the ^{99}Mo and ^{90}Mo in the monitoring foils were measured. The dependency of the ^{99}Mo production on the depth was obtained from the measurement results. The data normalized to the ^{90}Mo yield is shown on the Figs. 7, 8 [8, 9].

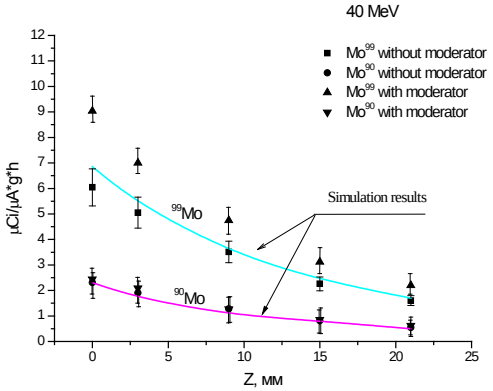

Fig. 7. The dependence activity of ^{99}Mo on depth at Ee =60 MeV

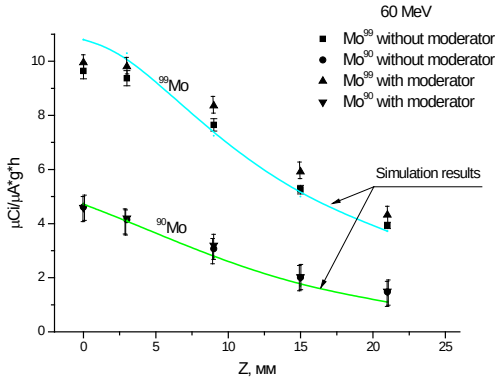

Fig. 8. The dependence activity of ^{99}Mo on depth at Ee =60 MeV

The comparative analysis of the ^{99}Mo activity distribution along the target depth shows that with increasing the incident electron energy up to 60 MeV the distribution profile somewhat changes. This can be explained by the fact that the part of the electron beam escapes the converter and generates bremsstrahlung directly at the molybdenum target, which, in its turn, alters the spatial distribution of the mixed γ,n radiation. Nevertheless, the increase of the ^{99}Mo yield of up to 30% takes place when using the neutron moderator, for electron energy of 40 MeV.

The average yield of the ^{99}Mo isotope for the whole target (diameter 19 mm, height 21 mm and mass 61 g) was ≈300 μCi/100μA·hour. If the electron energy increases the relative gain in the activity due to ^{98}Mo(n,γ)^{99}Mo reaction decreases. This happens mainly because of reduction of the relative share of the slow neutrons in the total flux of the mixed γ,n radiation. However, the average total yield of the ^{99}Mo isotope from the whole target increases to ≈ 530 μCi/100 μA·hour.

CONCLUSIONS

Thus, the main feature of the isotope production in the thick targets is the significant depth inhomogeneity of the production in the target. The $^{98}Mo(n,\gamma)^{99}Mo$ reaction contribution increases the total produced activity, but does not eliminate the decline in specific activity along the target depth. Such a decline can significantly hinder the mass-production as the increase of the initial target mass may be required. In the end it will cause the increase in highly active waste after radiochemical target treatment.

The work [5] shows the detailed description of the isotope ^{67}Cu yield investigation. It is shown that the optimum converter thickness corresponding to the maximum isotope yield is 1…2 mm for electron energy 40 MeV. The optimum size of the natural zinc target is found to be 20x20 mm. This size offers good ratio between the total and specific activities.

The isotopically rich in ^{68}Zn targets are considered for ^{67}Cu production by the reaction $^{68}Zn(\gamma,p)^{67}Cu$ in order to increase the yield of the target isotope. In this case the ^{67}Cu yield may increase up to 5 times. Taking into account the high cost of such target material one finds the question of the target recycling and reusing very actual. The choice of the optimum geometry in this case will be defined by the running cost of the accelerator, the target recycling cost as well as the useful isotope extraction cost.

The comparison of the values of the photonuclear yield of the ^{67}Cu isotope with another production methods shows much better production conditions in terms of total product yield, relative level and quantity of the side nuclides.

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ОСОБЕННОСТИ ФОТОЯДЕРНОГО ПРОИЗВОДСТВА РАДИОНУКЛИДОВ В ОБЪЕМНЫХ МИШЕНЯХ

А.В. Торговкин

Одним из важных требований к процессу фотоядерного производства радионуклидов является обеспечение как высокой удельной активности целевого нуклида, так и его полной активности в мишени. Традиционно эта задача решается применением технологических мишеней большого объема (до 50 $см^3$) с последующим радиохимическим выделением целевого нуклида. Методом моделирования и экспериментально исследованы условия, определяющие распределение активности в толстых фотоядерных мишенях. Найдены оптимальные параметры облучения для достижения максимальных значений удельной и полной активностей изотопного продукта.

ОСОБЛИВОСТІ ФОТОЯДЕРНОГО ВИРОБНИЦТВА РАДІОНУКЛІДІВ У ОБ'ЄМНИХ МІШЕНЯХ

О.В. Торговкін

Однією з важливих вимог до процесу виробництва радіонуклідів є отримання як високої питомої активності цільового нукліда, так і повної його активності в мішені. Традиційно ця задача вирішується застосуванням фотоядерних мішеней великого об'єму (до 50 $см^3$) із подальшим радіохімічним виділенням цільового нукліда. Методом моделювання та експериментально досліджено об'ємний розподіл активності для різних мішеней. Знайдено оптимальні параметри опромінення для досягнення максимуму виходу ізотопів $Mo^{99}(Tc^{99m})$ та Cu^{67} у мішенях з молібдену і цинку природного складу.