MATHEMATICAL MODELING AND MULTICRITERION OPTIMIZATION FOR PHOTONUCLEAR PRODUCTION OF THE ^{67}Cu ISOTOPE

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This paper considers a method for ^{67}Cu isotope production using electron bremsstrahlung by the ^{68}Zn(γ, p)^{67}Cu reaction. The facility for ^{67}Cu isotope production contains an electron accelerator, electron-gamma converter and zinc target. To optimize this facility we developed three-dimensional model of the converter and the target. Using this model, we performed the mathematical modeling of zinc target irradiation and thermal-hydraulic processes inside the target for various parameters of the electron beam and converter configurations. For mathematical modeling of radiation processes we used the MCNPX software. Thermal-hydraulic simulation utilized the commercial SolidWorks software with Flow Simulation module. Mathematical modeling revealed that efficient ^{67}Cu isotope production needs smaller beam diameter and higher electron energy. Under these conditions target heat power also increases, thus additional cooling is necessary. If the beam diameter and the electron energy are fixed the most effective method to satisfy the operating parameters and retain an efficient isotope yield is to optimize photonuclear spectra of the target by variation of converter thickness. We developed an algorithm for multicriterion optimization and performed the optimization of the facility with account to coupled radiation and heat transfer processes.

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1. INTRODUCTION

During the recent years, technique of the radiopharmaceuticals production (RPP) using particle accelerators has gained widespread acceptance in medicine and applied physics [1, 2]. Various designs of converters and targets were developed to optimize the RPP. The main attention was paid to the radiation characteristics of bremsstrahlung and also to beam parameters of the primary particles in order to increase the isotope yield. However, the real-world operation of such facilities faces the bunch of problems related to the thermohydraulic characteristics of the target. In some cases overheating caused melting of the target and no reliable operation RPP facility was possible. In this case beside the radiation characteristics one needs to account for the thermal-hydraulic parameters. Therefore, comprehensive investigation of radiation and temperature fields using mathematical modeling to ensure adequate performance is an urgent task.

Within the framework of this paper we consider the multicriterion optimization of radiation and thermal-hydraulic characteristics of the facility used in KIPT for ^{67}Cu isotope production.

2. INITIAL DATA

Our main goal is the isotope yield maximization under the appropriate target temperature regime for reliable operation of RPP facility. The KIPT RPP facility utilizes the linear electron accelerator KUT-30 for isotope production. This facility has the electron beam energy of 40 MeV with beam current of 250 mkA. The beam was assumed to be Gaussian with FWHM 1 cm [3]. Long exposure period leads to the reasonably high converter induced activity. Thus converter was made of copper because of its environment-friendly characteristics under high irradiation dozes. Fig.1 shows the modeling scheme of the target and the converter.

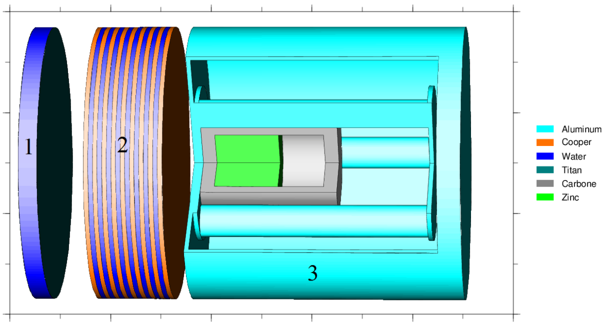

Fig.1. *Modeling scheme of the target and a converter*

The target is 20 g zinc cylinder placed into the graphite container sized ⌀20 mm×30 mm, and they both are placed into aluminum holder consisting of three pillars ⌀7 mm×50 mm and fixed between

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two disks ⌀34 mm×1 mm. This holder is placed inside the air cooled aluminum shell ⌀60 mm×6 mm and 1 mm thick. (see Fig.1. Block 3). This assembly is irradiated by bremsstrahlung radiation from the converter that consists of 9 copper disks ⌀60 mm×1 mm with 1 mm water cooling channels. Converter is irradiated by electron beam passing through the linac output window made of titanium foil 2×(⌀60 mm×0.05 mm) with 4 mm water cooling channel(Fig.1. Blocks 2 and 1).

3. MODELING OF THE ^{67}Cu PRODUCTION FACILITY

The RPP facilities[1, 2] have tantalum bremsstrahlung converter. Other widely used materials for electron converters are copper, lead and tungsten. During our simulation we compared the yield of ^{67}Cu for all the above materials. For the correct comparison of ^{67}Cu isotope yield in all cases mass thickness was 8.064 g/cm^2, which corresponds to 9 mm thick copper converter. Fig.2 represents the ^{67}Cu isotope yield for copper, lead, tungsten and tantalum converters. Copper has a predictable lower yield compared to tantalum (up to 30%) due to lower atomic number. However, for the converters with increased thickness one can expect the smaller yield difference due to the self-absorption effect. In this case copper converter is preferable because of environmental safety and easier manufacturing.

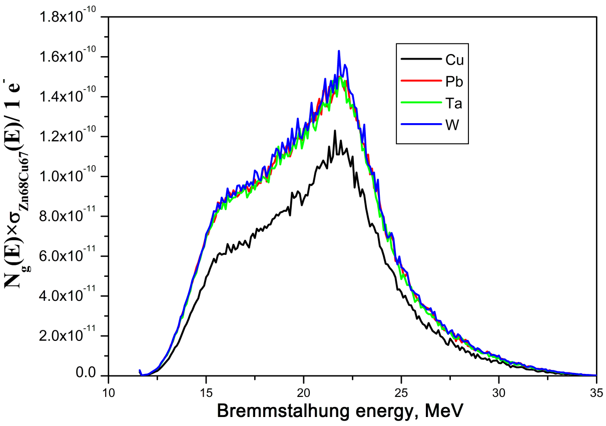


Fig.2. *Yields of production of ^{67}Cu for various converter materials*

Efficiency of the ^{67}Cu production depends on cross section of ^{68}Zn(γ,p)^{67}Cu reaction. Thus the main radiation characteristics affecting the isotope yield are the energy of primary electrons and flux density, which in turn depends on the average current and the beam profile. We have calculated the corresponding dependencies of the target activity for various electron energies and the beam profiles of the electron beam by Monte-Carlo simulation using MCNP software. Fig.3 shows the calculated zinc target activity versus the the incident electron beam energy. The reaction ^{68}Zn(γ,p)^{67}Cu has a 9.99 MeV threshold. Thus the eventual yield of ^{67}Cu isotope depends on the bremsstrahlung spectra, which in turn is defined by the incident electron energy spectra.

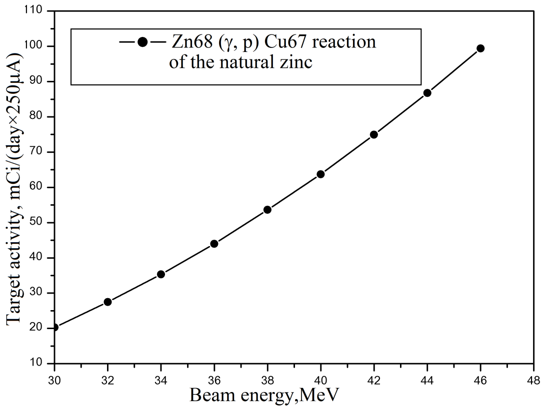


Fig.3. *Activity of the zinc target*

For the beam profile with larger FWHM we have smaller ratio of high energy electrons and the isotope yield decreases. Fig.4 shows the calculated target activity for 40 MeV beam with gaussian profile versus beam diameter (defined at FWHM).

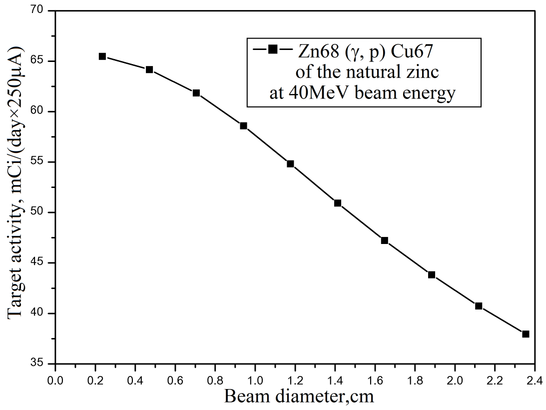


Fig.4. *The target activity at 40 MeV*

4. OPTIMIZATION OF THE ^{67}Cu PRODUCTION FACILITY

The results of modeling enable us to formulate the basic optimization criteria for efficient production of ^{67}Cu isotope. From one hand, one should provide the maximum energy of the electron beam and maximum flux density of bremsstrahlung. The later could be performed by tuning the thickness of the bremsstrahlung converter, the average current and the electron beam diameter. From the other hand under these conditions we obtain considerably high heat production in the converter, which puts forward the problem of efficient cooling. To solve the problem we need multivariate optimization procedure that accounts for coupled physical processes of radiation transport, heat transfer and hydrodynamics. In the previous paper [4] we have developed an approach to cope with such a problem. Fig.5 presents the conceptual diagram of the proposed approach.

The starting point of our consideration was Monte-Carlo simulation of radiation processes in the converter and target. This simulation used the previously developed model (see Fig.1), but with variable number of copper disks.

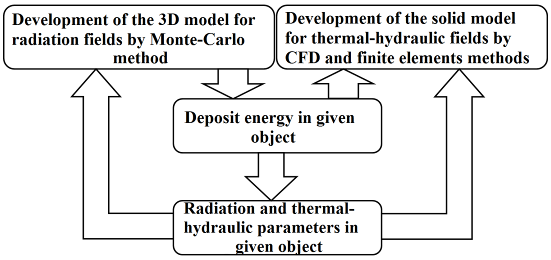


Fig.5. *Conceptual diagram of the multivariate optimization*

This way we studied the influence of converter thickness on the ^{67}Cu isotope production and energy deposition in the target. The number of disks varied from zero (no converter) to fifteen disks.

Speaking about the energy deposition in the zinc target one can distinguish two main components of this effect: energy deposition due to bremsstrahlung radiation from converter and deposition by the electrons that passed through converter. These electrons have high energy and produce bremsstrahlung radiation that also contribute to the ^{67}Cu isotope production.

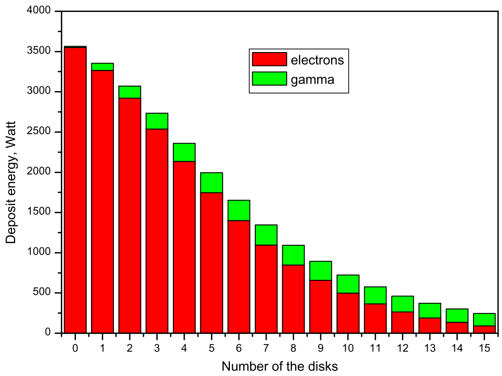


Fig.6. *Target energy deposition*

Our calculations showed that for limiting case of zero-thickness converter about 36% of beam power is deposited in the target. In this case zinc target in fact acts both as converter and isotope production target. Due to almost similar atomic numbers for copper and zinc (29 and 30, respectively) the bremsstrahlung conversion in the target provides high gamma flux. Self-absorption of the radiation in the target results in increased ^{67}Cu yield, but at the same time it results in high target heat deposition of 3600 W and consequent target overheating. Fig.6 shows the ratio of target energy deposition due to bremsstrahlung gamma radiation and beam electrons, and Fig.7 shows the ^{67}Cu yield ratio due to these two factors. The small amount of bremsstrahlung photons for zero-thickness converter emerges through the electron conversion on the target construction elements. From the Fig.7 one can see that thick converter reduces ^{67}Cu yield from the gamma radiation generated inside the target and for nine disks it becomes negligible while the energy deposition from the beam electrons is still noticeable.

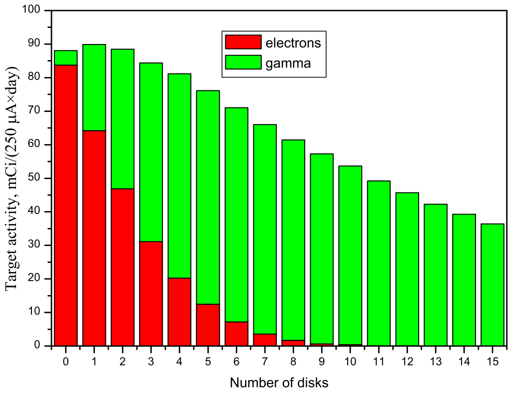


Fig.7. *^{67}Cu isotope production rate*

The physical reason for this situation is presence of 9.99 MeV threshold for ^{68}Zn(γ,p)^{67}Cu reaction. Thick converter reduces the ratio of electrons with energies above the threshold that reach the target, while low energy electrons do not contribute to ^{67}Cu production. This consideration shows that optimal balance between isotope yield and heat deposition corresponds to the converter containing nine copper disks. Thus we used this configuration for the further analysis. Fig.8 shows the distribution of the deposition energy among the disks of the converter with 9 disks during the irradiation. This distribution provides the initial data for the simulation of heat transfer processes of the cooling system. From the figure one can see that disk 8 has the maximum heal load and thus define the upper bound of the converter working temperature.

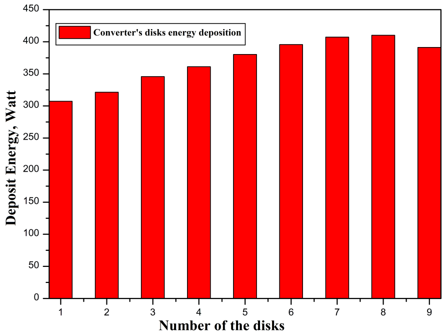
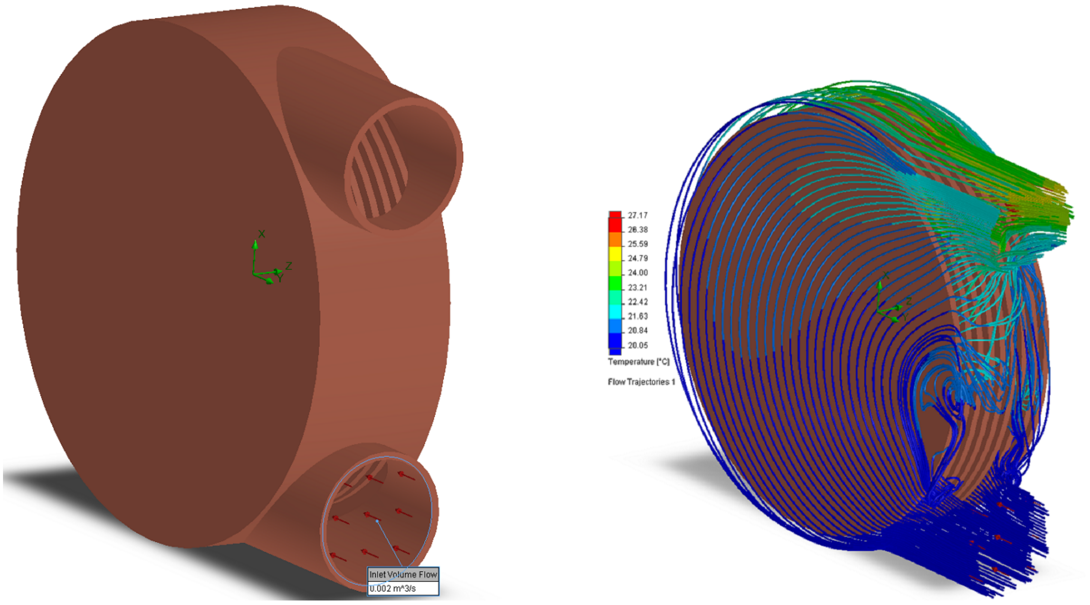


Fig.8. *The energy deposition in the disks of copper converter during irradiation*

According to the algorithm described above (see Fig.5), the next step was simulation of the thermal processes for converter with a water cooling system and target with an air cooling system. Using the SolidWorks software we developed a solid model of the both systems which was used for further simulation using SolidWorks Flow Simulation module. During simulation we assumed that electron accelerator operates in pulse mode with 3.5 ms pulse length and 150 Hz frequency. Using the results of the previous simulation of radiation processes we calculated the corresponding heat powers that are released in converter and target.

Fig.9,a represents a solid model of converter and cooling system, and Fig.9,b shows the results of the flow simulation for this system.

As we have have stated above, the disk 8 has the most critical temperature regime. Fig.10 shows the temperature distribution for this disk. One can see that water cooling provides the working temperatures in the central zone of the converter below the melting temperature.



a) *Solid model* b) *Temperature distribution and flow trajectory*

Fig.9. *Solid model of a water cooling system of the converter and the temperature distribution of water under cooling*

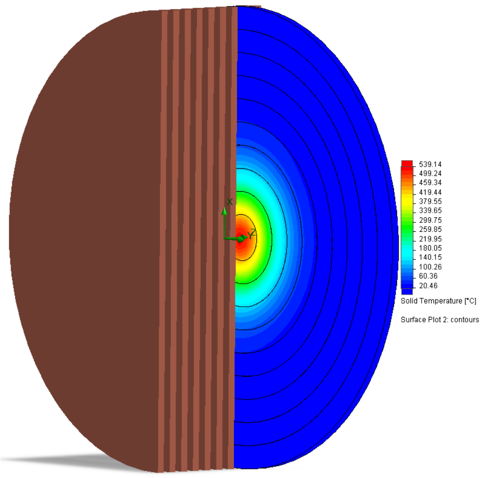


Fig.10. *Temperature distribution for disk 8*

The problem of appropriate cooling system arises for the zinc target. During the irradiation the target gains the high induced activity, and thus the necessary radiation safety must be provided. The present RPP facility uses the air tube for irradiated target transportation to the hot chamber for the subsequent decontamination. Implementation of the water cooling compatible with this transportation system is extremely difficult, so air cooling is the only acceptable option. At present time air cooling system utilizes the air compressor with about 300 l/min output. This output is insufficient because we had observed zinc target melting and aluminum shell burn-out. Thus we are planning to improve the cooling system by the powerful compressor with 1200 l/min output. To appraise the cooling efficiency of such compressor we performed the numerical simulation of heat exchange processes in the target cooling system. Fig.11 shows the solid model used during the simulation.

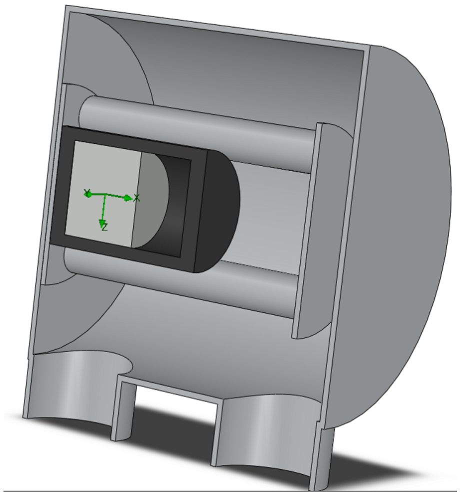


Fig.11. *Solid model of the target cooling system*

As we have stated above 9-disk converter provides the optimal balance between isotope yield and the target energy deposition. From the Fig.6 one can see that for 250 mkA current and 9-disk converter heat deposition in the target is 895 W. It follows from the simulation that the maximum target temperature is about 705 °C, while the zinc melting temperature is 419.58 °C. Obviously, under this conditions one gets target overheating with consequent melting and burning-out. The simplest way to mitigate the heat load is to reduce the average beam current. For 100 mkA the target energy deposition is only 360 W, and the simulation gives the maximum temperature about 410 °C, which is below the zinc melting temperature. But this simple approach has a significant drawback: ^{67}Cu yield considerably decreases to the value of about 23 mCi/day/20g.

Another way to reduce the energy deposition maintaining the acceptable ^{67}Cu production rate is spectrum moderation. The aim of spectrum moderation is to considerably decrease the ratio of electron energy deposition in the target. As we have discussed in the section 4, the low energy electrons does not contribute to ^{67}Cu production but result only in additional target heating. From the Figure 6 one can see that the efficient way to achieve this is to increase the number of disks in converter. Moreover, if we consider the ^{67}Cu yield versus the number of disks, we can see that energy deposition descends faster than isotope yield (Fig.12). Thus, increasing number of disks could provide the appropriate temperature regime even for 250 mkA beam current. The results of simulation for the system with 14-disk converter prove this suggestion. Table contains the results of simulation for 9-disk converter and and 14-disk converter.

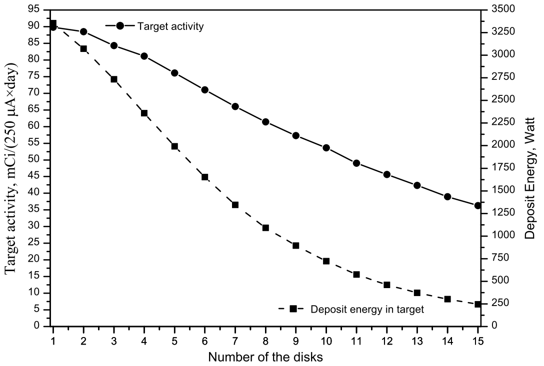


Fig.12. *Activity and deposit energy of the target*

For 14-disk converter we have target temperature 369 °C compared to 705 °C for 9-disk system, while ^{67}Cu yield is significantly higher: 40 mCi/day versus 23 mCi/day. Fig.13 shows simulation results for temperature distribution of the target cooling system for 14-disk converter.

Activity and temperature of the target

Beam current, mkA	Number of disks	Target activity, mCi/day	Maximum temperature of the target, °C
250	9	58	705
100	9	23	410
250	14	40	369

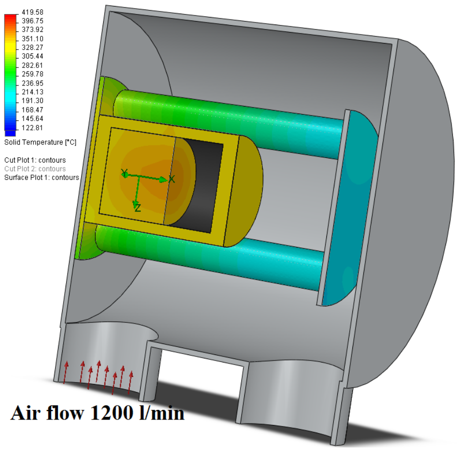


Fig.13. *Temperature distribution of the target cooling system for 14-disk converter*

5.CONCLUSIONS

During our comprehensive analysis of the ^{67}Cu isotope production we used coupled simulation of radiation processes and heat transfer processes to optimize the parameters of isotope production facility. We have shown copper to be the appropriate material for electron-gamma converter with somewhat lower ^{67}Cu yield compared to lead or tantalum but with significantly better thermal-hydraulic parameters and lower values of the radiation-induced activity.

Our analysis also revealed that only thick 14-disk converter provide acceptable temperature conditions for the isotope production target. In this case we achieve the reasonable balance between the isotope yield rate and heat output in the target, and conventional air compressor with 1200 l/s is capable to ensure the reliable air cooling. The further increasing of the air pumping output does not considerably improve the cooling efficiency, thus prohibiting the usage of thick converter with higher isotope yield or higher beam current. One can improve the temperature conditions by implementing the additional cooling elements for the target system such as air radiators, wings, etc.

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МАТЕМАТИЧЕСКОЕ МОДЕЛИРОВАНИЕ И МНОГОКРИТЕРИАЛЬНАЯ ОПТИМИЗАЦИЯ УСТАНОВКИ ДЛЯ ФОТОЯДЕРНОЙ НАРАБОТКИ ИЗОТОПА ^{67}Cu

Н. П. Дикий, Е. В. Рудычев, Д. В. Федорченко, М. А. Хажмурадов

Рассматривается способ получения изотопа ^{67}Cu с использованием тормозного излучения электронов, где изотоп ^{67}Cu образуется вследствие реакции ^{68}Zn(γ,p)^{67}Cu . Установка для наработки изотопа ^{67}Cu содержит ускоритель электронов, конвертер для получения тормозного излучения и цинковую мишень. Для оптимизации параметров установки разработана трехмерная модель конвертера и мишени. С использованием данной модели проведено математическое моделирование процесса облучения и термогидравлических процессов цинковой мишени для различных параметров пучка электронов и конфигурации конвертера. Математическое моделирование радиационных характеристик осуществлялось с помощью программного комплекса MCNPX. Моделирование термогидравлических параметров осуществлялось при помощи методов вычислительной гидродинамики и методов конечных элементов. При помощи математического моделирования показано, что для повышения эффективности образования изотопа ^{67}Cu следует уменьшать диаметр пучка и увеличивать его энергию. При этом также увеличивается энерговыделение в мишени, что требует дополнительных мер по ее охлаждению. При фиксированных значениях диаметра пучка и энергии электронов показано, что для сохранения эксплуатационных параметров и получения эффективной наработки изотопа, наиболее эффективным методом является оптимизация фотоядерного спектра на мишени за счет изменения толщины конвертера. Разработан алгоритм многокритериальной оптимизации и проведена оптимизация установки с учетом радиационных и термогидравлических параметров.

МАТЕМАТИЧНЕ МОДЕЛЮВАННЯ ТА БАГАТОКРИТЕРІАЛЬНА ОПТИМІЗАЦІЯ УСТАНОВКИ ДЛЯ ФОТОЯДЕРНОГО НАПРАЦЮВАННЯ ІЗОТОПУ ^{67}Cu

М. П. Дикий, Є. В. Рудичев, Д. В. Федорченко, М. А. Хажмурадов

Розглядається спосіб отримання ізотопу ^{67}Cu з використанням гальмівного випромінювання електронів, де ізотоп ^{67}Cu утворюється внаслідок реакції ^{68}Zn (γ,p) ^{67}Cu. Установка для напрацювання ізотопу ^{67}Cu містить прискорювач електронів, конвертер для отримання гальмівного випромінювання та цинкову мішень. Для оптимізації параметрів установки розроблена тривимірна модель конвертера і мішені. З використанням даної моделі проведено математичне моделювання процесу опромінення і термогідравлічних процесів цинкової мішені для різних параметрів пучка електронів та конфігурації конвертера. Математичне моделювання радіаційних характеристик здійснювалося за допомогою програмного комплексу MCNPX. Моделювання термогідравлічних параметрів здійснювалося за допомогою методів обчислювальної гідродинаміки та методів кінцевих елементів. За допомогою математичного моделювання показано, що для підвищення ефективності утворення ізотопу ^{67} Cu потрібно зменшувати діаметр пучка та збільшувати його енергію. При цьому також збільшується енерговиділення у мішені, що вимагає додаткових заходів по її охолодженню. При фіксованих значеннях діаметра пучка та енергії електронів показано, що для збереження експлуатаційних параметрів та одержання ефективного напрацювання ізотопу найбільш ефективним методом є оптимізація фотоядерного спектру на мішені за рахунок змінення товщини конвертера. Розроблено алгоритм багатокритеріальної оптимізації та проведена оптимізація установки з урахуванням радіаційних і термогідравлічних параметрів.