MODELING OF RADIATION AND THERMAL-HYDRAULIC CHARACTERISTICS OF THE CONVERTER-TARGET UNDER IRRADIATION WITH ELECTRONS IN ORDER TO OPTIMIZE ISOTOPE ^{99m}Tc PRODUCTION

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The method of the modeling of radiation and thermal-hydraulic characteristics of the system for ^{99m}Tc isotope production was developed, using the Monte Carlo method for computation of radiation fields and computational fluid dynamics for modeling of thermal-hydraulic characteristics. The three-dimensional solid models of this system were developed, where system geometry was fully described and necessary initial conditions was taking into account. We considered the isotope ^{99}Mo production due to the (γ, n) and (n, γ) reactions and analyzed the efficiency of these reactions. To improve the efficiency of the isotope ^{99}Mo production and to reduce the energy deposition in the target material it is proposed to use the composite converter made of Pb + Al, Pb + Cu, Pb + Be, for which the optimal parameters were defined. Application of this method makes it possible to optimize the performance of the system for ^{99m}Tc isotope production.

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INTRODUCTION

Now the clinical diagnosis of human diseases by introducing to his body the radioisotopes as the indicators is being developed. These substances are called radiopharmaceuticals (RFP). The main objective while the production of radiopharmaceuticals is obtaining the necessary isotopes, for example, ^{99m}Tc – it radiopharmaceuticals are used for the diagnosis of diseases of nearly all the human major organs and are used in 80…85% of diagnostic procedures with labeled atoms. In the U.S. the number of treatments with drugs on the basis of ^{99m}Tc reaches 12 million per year. The annual world market for ^{99m}Tc is estimated at 3.7 billion dollars. High needs in ^{99m}Tc come from its relatively short half-life – 6.02 h and low gamma-radiation energy – 0.1405 MeV. These factors provide low exposure dose (0.5…5% of the allowable level) and at the same high penetration capability for radiometric measurements. Therefore, obtaining ^{99}Mo isotope, which transforms through its beta-decay ($T_{1/2}$ = 2.748 days) to the isotope ^{99m}Tc, plays an important role in nuclear medicine. [1]

The traditional method of producing ^{99}Mo in nuclear reactors is technically and environmentally difficult. The alternative methods of producing ^{99}Mo at the charged particle accelerators was proposed at KIPT, and researches on obtaining ^{99}Mo nuclide from the reactions ^{100}Mo(γ, n)^{99}Mo (bremsstrahlung), ^{98}Mo (n, γ)^{99}Mo (Photonuclear neutrons from bremsstrahlung) were done using a linear high-current electron accelerator. The experimental facility was developed on the basis of the acceleration stand LU-20 (20 MeV, 10 kW), and ^{99}Mo was produced by irradiation for 10 hours of molybdenum target of natural isotopic composition [2].

To obtain high-energy photons at the electron accelerator it is necessary to use converters from materials that have a high conversion rate of electrons in the photona (tantalum, lead). The intensity of the obtained photons depends on electron beam energy, and at the same power the system with greater energy will be more effective. So, it is more preferable to use stand based on the accelerator KUT-30 with electron energy 27…40 MeV [3]

To maximize the amount of the ^{99}Mo isotope it is necessary to ensure optimum converter and target parameters with taking into account the radiation properties of the used materials used and the requirements of the cooling system. The optimization of such a system is considered in this work.

1. CALCULATION METHOD

Such optimization tasks are the multi-objective optimization problems with taking into account the various physical processes. To carry out appropriate calculations we developed principles and three-dimensional models using Monte Carlo methods to calculate the radiation fields, nuclear reactions, and the released energy in the materials of the system, as well as the methods of computational fluid dynamics (Computational fluid dynamics - CFD) calculations of the cooling conditions and system thermal-hydraulic parameters [4].

Bremsstrahlung converter is a lead cylinder in the copper shell with water cooling. The target for production of the ^{99}Mo isotope is set of large and small disks from natural molybdenum sandwiched between copper plates for efficient air cooling (Fig. 1).

^{99}Mo isotope produced due to the fact that the primary electron beam generates bremsstrahlung photons in the lead converter, and if the converter thickness is not sufficient to absorb electrons – and then also in the molybdenum. Photon with energies greater than the threshold value of the converter material participate in various reactions, such ^{208}Pb(γ, n)^{207}Pb, and in the target area – in the reaction ^{100}Mo(γ, n)^{99}Mo with probabilities which are proportional to the reaction cross sections. As a result, both the converter and the target generates photonuclear neutrons that may also take part in the production of the desired isotope by reaction ^{98}Mo(n, γ)^{99}Mo.

Calculation of neutron and gamma radiation fields was carried out by Monte Carlo method using MCNPX software, with taking into account all the possible nuclear reactions that may result in the ^{99}Mo isotope production. However, the main contribution to the isotope production for the considered electron energy comes from the reactions ^{100}Mo(γ, n)^{99}Mo and ^{98}Mo(n, γ)^{99}Mo.

The irradiation of electrons with 34.7 MeV energy (beam diameter 10 mm, the current 250 μA) was simulated. The simulation of RZ symmetry RZ is shown at Fig. 2.

To estimate the efficiency of channels and optimization of system for ^{99}Mo isotope production it is necessary to estimate the neutron and gamma leakage from the system, for this purposes cylindrical surface 1, 2 and plane 3 was introduced in this model (see Fig. 2,b).

Some simulation results for the neutron fluxes are shown below. The integrated neutron flux outside the assembly was $6.54 \cdot 10^{12}$ n/s.

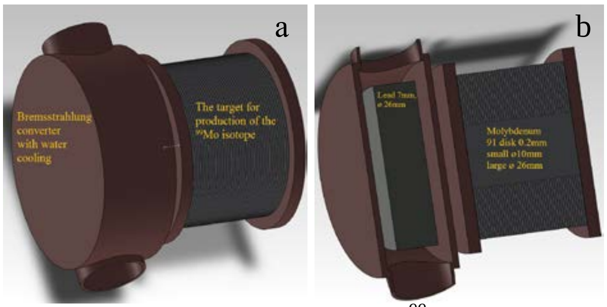

Fig. 1. Solid model of the facility for ^{99}Mo production: a) general view; b) cross-cut

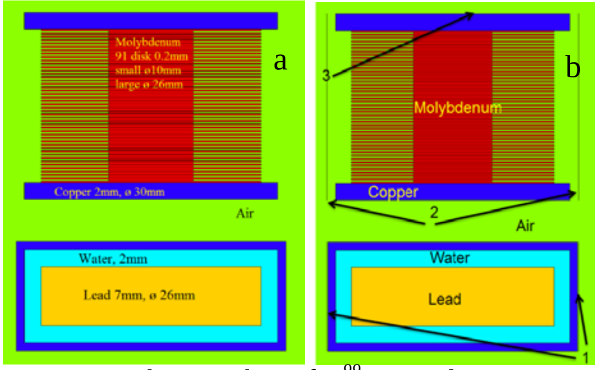

Fig. 2. Simulation scheme for ^{99}Mo production using the Monte-Carlo method a) general view; б) scheme with additional cylinder surfaces 1, 2 and plane 3

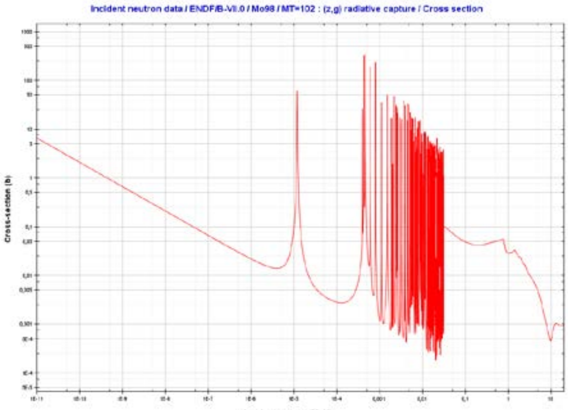

Fig. 3. Cross-section of ^{98}Mo(n, γ) ^{99}Mo reaction

The neutron flux crossing the surface 1 is equal to $1.646 \cdot 10^{11}$ n/cm^2/s. The neutron flux crossing the surface 2 is $1.258 \cdot 10^{11}$ n/cm^2/s. The neutron flux crossing the surface 3 – $1.264 \cdot 10^{11}$ n/cm^2/s. The neutron yield for a given electron energy and converter + target geometry is $8.3 \cdot 10^{-3}$ n/e$^-$. The neutron flux in the target is $2.5 \cdot 10^{-4}$ n/e$^-$/cm^2. In addition, the cross-section of ^{98}Mo(n, γ)^{99}Mo reaction has resonances (Fig. 3), therefore, ^{99}Mo is effectively produced from irradiation with neutrons in the energy range 10 eV…1 keV. However, since the spectrum corresponds to the photo-nuclear neutron spectrum, the effective cross section of the reaction is less than 1 Barn (Figs. 3, 4). Hence, the efficiency of the reaction ^{100}Mo(γ, n)^{99}Mo will not high. This assumption is fully agreed with the simulation results.

In the simulation of this system the following ratio of ^{99}Mo production was found: 99.66 % as a result of the ^{100}Mo(γ, n)^{99}Mo reaction and 0.34 % from the ^{98}Mo(n, γ)^{99}Mo reaction.

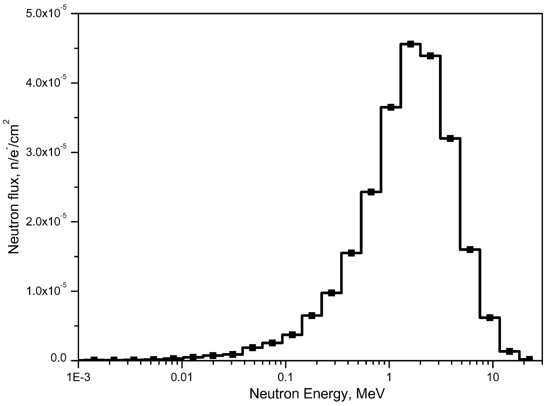

Fig. 4. Neutron spectrum in the target

2. EFFICIENCY OF THE ^{98}Mo(n, γ)^{99}Mo CHANNEL

The maximum intensity of the photonuclear neutron flux is restricted with primary electron energy. The neutron spatial distribution in the RZ symmetry is shown at Fig. 5. The system can be optimized by adding a moderator and reflector to increase the neutron flux density and providing in the target area the resonance energy in the range 1 eV…1 keV.

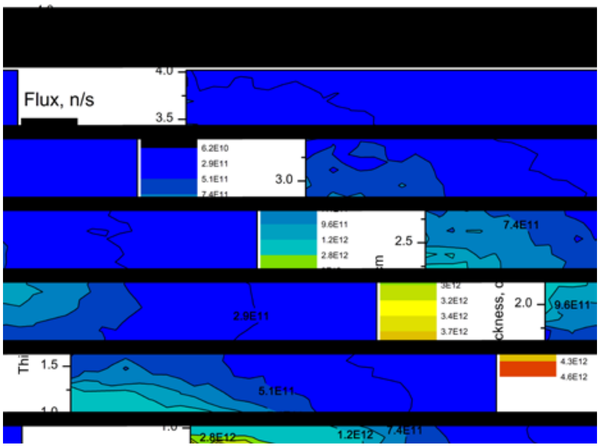

Fig. 5. Spatial distribution of the neutron fluxes in the converter-target system

The system can be modified by adding polyethylene moderator and graphite reflector with thickness 10 cm or more.

The modeling results showed that it is possible to increase the reaction contribution up to 1%. However, even if we moderate all leakage photonuclear neutrons to resonance energies and redirect them back to the target, the efficiency of ^{98}Mo(n, γ)^{99}Mo reaction will be less than 50% compared to ^{100}Mo(γ, n)^{99}Mo reaction. Taking into account the significant neutron loss due moderating and scattering, the maximum contribution will not exceed 1…2%.

Taking into account the cooling efficiency reducing when moderator and reflector are used we can conclude that for the given primary electron energy costs for increasing the reaction ^{98}Mo(n, γ)^{99}Mo efficiency exceeds benefit from light increasing the ^{99}Mo isotope producing.

Therefore, the optimization of the system based on the ^{98}Mo(n, γ)^{99}Mo channel performance is not feasible.

3. EFFICIENCY OF THE ^{100}Mo(γ, n) ^{99}Mo CHANNEL

Bremsstrahlung converter is the lead disc of 7 mm thickness and 26 mm diameter.

Molybdenum target is the assembly of 91 disks of 0.2 mm thickness and 10 mm and 26 mm diameters and which is sandwiched between copper disks of 2 mm thick and 30 mm diameter. The energy electron beam energy is 34.7 MeV, the beam current is 250 μA.

The modeling results showed that:

- energy release in the converter is 4,124.75 Watts. The water cooling is used to ensure the operation;
- energy release in the molybdenum assembly is 725 watts. Here air cooling is possible.

4. STUDYING OF THE ASSEMBLY PARAMETERS CONCERNING THE ^{99}Mo PRODUCING

The efficiency of ^{99}Mo producing is defined by the gamma flux energy distribution, as ^{100}Mo(γ, n)^{99}Mo reaction has photon threshold at 8.29 MeV. The photon distribution in the target for energies above 8 MeV is shown at Fig. 6.

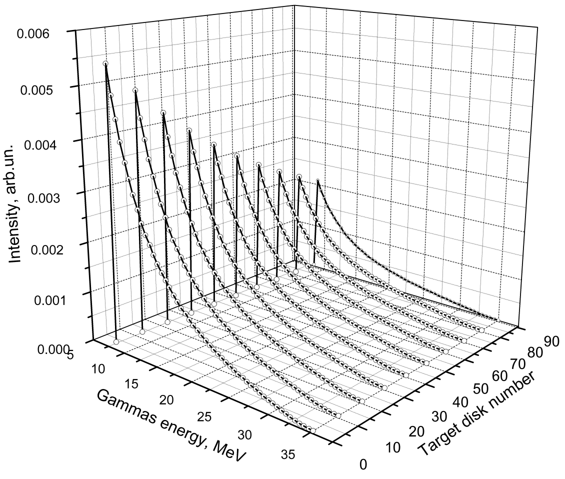

Fig. 6. Photon flux distribution in the target (91 disk)

From the analysis of the obtained distribution it can be seen that the flux in the last molybdenum disk is more than 35 % in the first. Therefore, to increase the efficiency of isotope producing it is possible to increase the target volume by increasing the number of disks, or placing the second target after the primary one for preliminary isotope production. Calculations showed that after increasing the number of disks from 91 to 127 the efficiency of ^{99}Mo producing is increased by 18 %. The photon distribution for 127 discs is shown in Fig. 7.

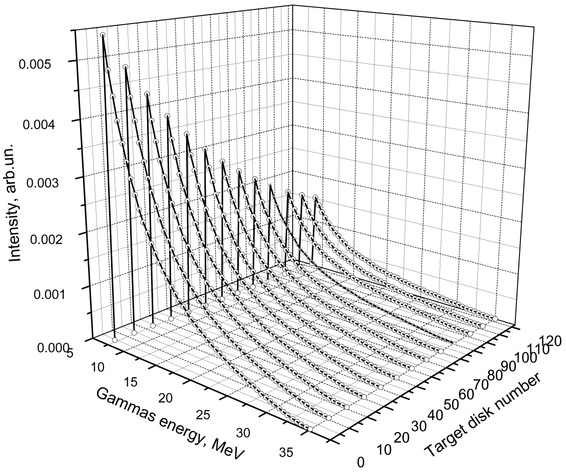

Fig. 7. Photon distribution in the target (127 disks)

5. STUDYING OF THE CONVERTER PARAMETERS CONCERNING THE ^{99}Mo PRODUCING

In our case, ^{100}Mo(γ, n)^{99}Mo is the main channel of ^{99}Mo producing and hence we must ensure the maximum high-energy photon flux in the target.

Earlier, the modeling results showed that the most effective thickness of the converter is 4 mm, in which high-energy photons from converter bremsstrahlung interact with the assembly (for 7 mm thickness they are absorbed in the converter) and transmitted electrons generate additional bremsstrahlung directly to the molybdenum. For converter of 4 mm thickness efficiency of ^{99}Mo isotope producing is increased by 24%. However, the use of converter with such thickness is difficult because of problems of target air-cooling. Energy, released on the target, was 1.687 watts, of which 901.5 watts was due to the electrons.

To reduce the energy released it was proposed to put an absorber of electrons after the target and to use the material with atomic number small enough to prevent excessive absorption of high-energy photons.

We modeled converters of different configurations, such as Pb + Al, Pb + Cu, Pb + Be. As there are restrictions due to assembly air cooling optimal converter is 4 mm Pb + 5 mm Cu.

In this case, the efficiency of ^{99}Mo isotope producing is 10% higher than for 7 mm converter and energy released in the target was ≈771 W, that corresponds to the value of the released energy for 7 mm converter.

6. MODELING OF THE COOLING CONDITIONS AND STUDYING THE TEMPERATURE GRADIENTS OF THE SYSTEM

An important problem in the design of this class is the choice of optimal cooling conditions and related thermo-mechanical parameters that provide secure mode and a long life of the designed system.

The study of temperature gradients during irradiation and cooling is an important task, since the presence of significant temperature gradients can lead to thermo-mechanical stress and performance degradation of the target, right up to the destruction of the structure. During irradiation temperature gradients occur not only because of the unevenness in the energy release, but also due to uneven cooling of the target, caused by the geometrical characteristics of cooling system.

Modeling of radiation using Monte Carlo method provides information about the energy released in irradiated object of research and using the data for thermal-hydraulic parameters of the system.

To calculate the thermal-hydraulic parameters and cooling conditions the three-dimensional solid model of the optimized system was developed. The heat source is modeled on the basis of data on the energy release obtained by the Monte Carlo simulation (Fig. 8).

Calculation was done for molybdenum assembly consisting from 91 disks as the most energy-intensive event.

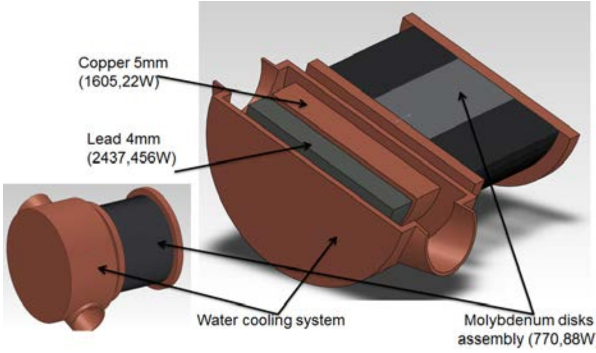

Fig. 8. Solid model of the optimized converter for calculation the target thermal-hydraulic parameters and cooling conditions

The thermohydraulic parameters and cooling conditions system can be calculated by solving the thermodynamics and hydrodynamics adjoint problem. In the three-dimensional geometry of the most accurate solution can be obtained by using computational fluid dynamics (CFD) method and the finite volume method. The modeling was done using the SolidWorks Flow Simulation software package [5, 6].

SolidWorks – a versatile tool for analysis in hydraulic gas dynamics and heat transfer. SolidWorks Flow Simulation is hydraulic gas dynamics analysis module in the SolidWorks.

The input parameters can be: velocity, pressure, mass and volume flow of liquid or gas. Heat sources may be volume and surface. In our case we consider anisotropic volume sources.

In the software package SolidWorks Flow Simulation motion and heat transfer of the flow medium is modeled by the Navier-Stokes equations, which give the laws of conservation of mass, momentum and energy in the non-stationary problems. In addition, the equation of state of environment components, and the empirical dependences of the viscosity and thermal conductivity of these components on the temperature are used. Non-Newtonian fluids are set as the dependence of the viscosity on the rate of shear deformation and temperature; compressible fluids are given as density dependence on the pressure. These equations simulate turbulent, laminar and transitional flows. The transition between laminar and turbulent flow is determined by the critical value of the Reynolds number. For simulation of turbulent flows, the Navier-Stokes equation are averaged by Reynolds, i.e. we use the average for small time interval the turbulence influence on the flow parameters (pressure, speed, temperature), which take into account for by applying appropriate time derivatives. As a result, the equations have additional terms of the Reynolds stress and to close this system of equations SolidWorks Flow Simulation uses the transport equation of turbulent kinetic energy and its dissipation within a -ε turbulence model.

The system of equations of conservation of mass, momentum and energy of non-stationary spatial flow looks like the approach of Euler in the Cartesian coordinate system (xi, i = 1, 2, 3), rotated with angular velocity Ω about an axis passing through its beginning:

$$\frac{\partial \rho}{\partial t}+\frac{\partial}{\partial x_k}(\rho u_k)=0;$$

$$\frac{\partial(\rho u_i)}{\partial t}+\frac{\partial}{\partial x_k}(\rho u_i u_k-\tau u_k)+\frac{\partial P}{\partial x_i}=S_i;$$

$$\frac{\partial(\rho(E))}{\partial t}+\frac{\partial}{\partial x_k}((\rho E+P)u_k+q_k-\tau_{ik}u_i)=S_k u_k+Q_H,$$

where t – time; u – velocity of the flow medium; ρ – density of the flow medium; P – pressure of the flow medium; S_i – external mass forces, acting on the mass unit of the flow medium ($S_{porous\ bodies}$ – effect of resistance of the porous body, $S_{gravitation}$ – effect of gravitation, $S_{rotation}$ – effect of coordinate system, rotation, i.e. $S_i = S_{porous\ bodies} + S_{gravitation} + S_{rotation}$); E – full energy of mass unit; Q_H – heat from the heat source per unit volume of the flowing medium; τ – tensor of viscous shear stress; q_i – diffusive heat flux. Lower indices indicate summation over the three coordinate directions.

As used differential and integral equations have no analytical solutions, they are reduced to a discrete form and solved on a computational grid (can be digitized in space or in time).

When digitizing in space:

- estimated area is covered by the computational grid, the edges of cells of which are parallel to the coordinate planes;
- values of the independent variables are calculated at the centers of the cells (finite volume method);
- computational grid cells have the shape of the box;
- the area in which this net is constructed has a parallelepiped shape;
- on the border with the solid body, the procedures of local cell-division are used.

When digitizing in time:

- For each cell of the computational grid the maximum allowable time step is determined;
- step depends on the physical values and the sampling step in space in the cell.

Three separate classes of finite volume mesh are used for solving problems of heat transfer.

1. Creating the computation area and generation of the grid in the flow area for calculating the diffusion flow in liquid or gas

$$q_k=-\left(\frac{\mu_l}{P_r}+\frac{\mu_t}{\sigma_c}\right)c_p\frac{\partial T}{\partial x_k},$$

where P_r – Prandtl number; c_p – specific heat at constant pressure; μ_l – coefficient of dynamic viscosity, μ_t – coefficient of turbulent viscosity.

2. Creating the computation area and generation of the grid in the solid body area for calculating the heat transfer

$$\frac{\partial pe}{\partial t} = \frac{\partial}{\partial x_i}\left(\lambda \frac{\partial T}{\partial x_i}\right) + Q_H ,$$

where $e = cT$ (c – specific heat; T – temperature); λ – thermal conductivity; Q_H – heat from the heat source per unit volume.

3. Creating the computation area and generation of the grid in the bound of flow area solid area. Convective heat transfer between the solid surface and the fluid is modeled in the modeling of the boundary layer fluid flow.

An important advantage of used system is the possibility to generate a computational grid directly on the SolidWorks model, the creation of the computational area and mesh generation in the solid and the flow region.

After generation of the computational area and computing grid it is necessary to determine the volume heat sources. To take into account the anisotropy of the heat distribution in the converter lead and copper discs were uniformly divided into regions in RZ symmetry with dimensions 10×10 for lead and copper disc respectively. For each of the 200 areas, using the results of radiation calculations by Monte Carlo method, it was defined the appropriate heat source, volume-averaged in the corresponding area. The simulation results are shown below.

Water flow and temperature distribution in the bremsstrahlung converter and cross-section at the plane of maximum temperature of the lead disk are shown at Figs. 9 and 10, respectively.

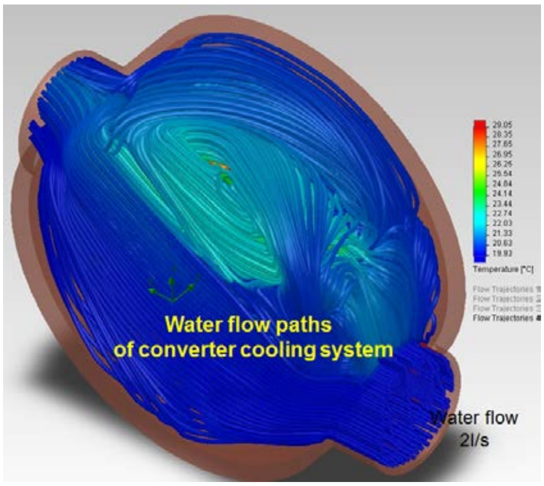

Fig. 9. Water flow paths with the temperature distribution in the converter cooling system

Analysis of the temperature distribution for the target shows that, while air cooling (flow rate 15 m/s) during irradiation the target is substantially heated and the maximum temperature in the first disks reaches values of more than 700 °C. Therefore, in the model calculations we take into account additional heat loss by radiation surfaces. The temperature distribution in the cross-cut corresponding to the maximum temperature in the assembly of molybdenum disks is shown in Fig. 11 (red and blue points in the figure correspond to the maximum and minimum temperatures).

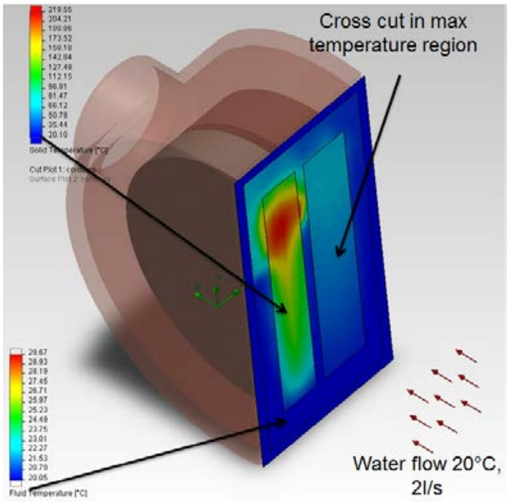

Fig. 10. Temperature distribution in the converter. Cross-cut in the region of temperature maximum

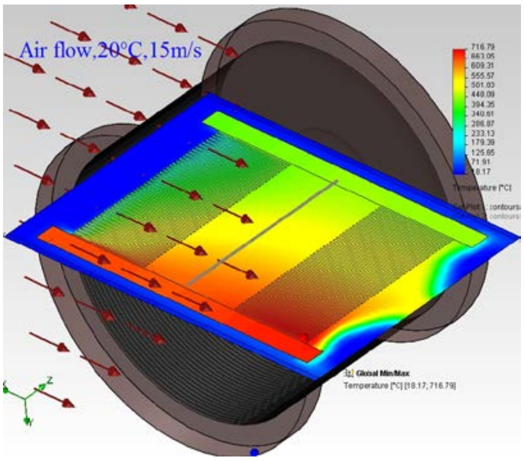

Fig. 11. Temperature distribution in the converter. Cross-cut in the region of temperature maximum in the molybdenum disks

The temperature gradient between the hottest and coldest molybdenum discs is 300 °C. The temperature gradient in the bremsstrahlung converter reaches about 200 °C (maximum temperature of the lead disc corresponds to turbulence zone in the cooling system liquid). When operating similar systems it is necessary to take into account the temperature gradients, so the relevance of this calculation method is not in doubt.

CONCLUSIONS

To simulate radiation and thermal-hydraulic characteristics of the system for ^{99m}Tc isotope production special procedure was developed, using the Monte Carlo method and computational fluid dynamics for modeling of thermal-hydraulic characteristics. To simulate and optimize the converter – target system three-dimensional solid models of the system were developed, which completely describe the geometry of the system and take into account all the necessary initial conditions.

The simulation of the system was done in order to improve the efficiency of obtaining the ^{99m}Tc isotope.

It is shown that for specified electron energy it is not appropriate to use additional reflectors and moderators to increase the ^{98}Mo(n, γ)^{99}Mo reaction efficiency.

Analysis of the obtained data showed that to increase the efficiency of ^{99m}Tc producing it is necessary:

• to replacement the 7 mm lead converter with combined converter (4 mm lead + 5 mm copper);

• to increase the target thickness by increasing the number of molybdenum plates.

To ensure the system operation it is necessary to provide the water cooling of the converter with water volume flow of 2 l/s and the air cooling of the target with air flow of 15 m/s.

The modeling results show the presence of high temperature gradients, which can cause large thermo-mechanical stresses.

The method developed by us have to be used to minimize the potential risks of system functioning, with taking into account the technical parameter of the equipment used in the cooling system to change the values of water volumetric flow and air flow rate as well as the modification of the geometrical parameters of the system to minimize temperature gradients in the process of the complex operation.

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МОДЕЛИРОВАНИЕ РАДИАЦИОННЫХ И ТЕРМОГИДРАВЛИЧЕСКИХ ХАРАКТЕРИСТИК СИСТЕМЫ КОНВЕРТЕР-МИШЕНЬ ПРИ ОБЛУЧЕНИИ ЕЕ ЭЛЕКТРОНАМИ ДЛЯ ОПТИМИЗАЦИИ НАРАБОТКИ ИЗОТОПА ^{99m}Tc

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Разработана методика расчетов моделирования радиационных и термогидравлических характеристик системы получения изотопа ^{99m}Tc, использующая метод Монте-Карло для радиационных полей системы и метод вычислительной гидродинамики для термогидравлических характеристик системы. Созданы трехмерные твердотельные модели системы, полностью описывающие геометрию системы и учитывающие необходимые начальные условия. Рассмотрена наработка изотопа ^{99}Mo за счет (γ, n)- и (n, γ)-реакций и проведен анализ их эффективности. Для повышения эффективности наработки изотопа ^{99}Mo и уменьшения энерговыделения в материале мишени предложено использовать композитный конвертер типа Pb+Al, Pb+Cu, Pb+Be, для которого определены оптимальные параметры. Применение методики дает возможность оптимизировать эксплуатационные характеристики системы получения изотопа ^{99m}Tc.

МОДЕЛЮВАННЯ РАДІАЦІЙНИХ ТА ТЕРМОГІДРАВЛІЧНИХ ХАРАКТЕРИСТИК СИСТЕМИ КОНВЕРТЕР-МІШЕНЬ ПРИ ОПРОМІНЕННІ ЇЇ ЕЛЕКТРОНАМИ ДЛЯ ОПТИМІЗАЦІЇ НАПРАЦЮВАННЯ ІЗОТОПУ ^{99m}Tc

Є.В. Рудичев, С.І. Прохорець, Д.В. Федорченко, М.А. Хажмурадов

Розроблено методику розрахунків моделювання радіаційних та термогідравлічних характеристик системи отримання ізотопу ^{99m}Tc, яка використовує метод Монте-Карло для радіаційних полів системи і метод обчислювальної гідродинаміки для термогідравлічних характеристик системи. Створено тривимірні твердотільні моделі системи, які повністю описують геометрію системи та враховують необхідні початкові умови. Розглянуто напрацювання ізотопу ^{99}Mo за рахунок (γ, n)- і (n, γ)-реакцій та проведено аналіз їх ефективності. Для підвищення ефективності напрацювання ізотопу ^{99}Mo та зменшення енерговиділення в матеріалі мішені запропоновано використовувати композитний конвертер типу Pb+Al, Pb+Cu, Pb+Be, для якого визначено оптимальні параметри. Застосування методики дає можливість оптимізувати експлуатаційні характеристики системи отримання ізотопу ^{99m}Tc.