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Quantitative evaluation of heat dissipation of a tellurium dioxide target front cooling during deuterons irradiation

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To ensure the maximum possible cyclotron beam current during the production of iodine-based radiopharmaceuticals, a new method of the TeO_2 target forward cooling was developed and tested. Using a mathematical model of irradiation and cooling processes, the intensity of heat dissipation was determined, and assumptions about the criteria of destruction of the TeO_2 target were made.

Keywords: nuclear medicine, radioiodine, radiopharmaceutical, tellurium dioxide, cyclotron, target cooling.

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Introduction

In modern medical practice several isotopes of Iodine are used as diagnostic and therapeutic drugs (Table 1). The production of these isotopes occurs by irradiating a target made of TeO_2 with a beam of accelerated protons or deuterons in a cyclotron. Due to the same production technology of all iodine isotopes, a somewhat „unified“ technical solution is possible, which is represented a system for irradiating targets based on various Tellurium isotopes.

During the process of producing radioisotopes at a cyclotron, in order to intensify the yield of the product, they try to use the maximum possible current of a beam of charged particles. However, high irradiation intensity often leads to emergency situations, especially if it concerns a target material with poor thermal conductivity. A beam of charged particles passing through matter loses its energy, mostly through ionization and excitation of atoms. Moreover, in the case of insufficient heat removal, energy dissipated in the form of heat can lead to melting and sublimation of the substance. To prevent radiation accidents, it is necessary to apply intensive cooling of the target, while trying to maintain a high density of the charged particle beam, which will avoid phase transformations in the target material.

This problematics becomes special actuality when oxide targets used for the production of radioactive Iodine for medical purposes [1]. The target is an TeO_2 layer melted on a Pt substrate. The target preparing involves applying TeO_2 powder with a mass of 200−300 mg into a recess on the surface of a Pt plate, and then it placed in a furnace, where heated to the melting temperature of 733°C. When cooled, moltenTeO_2 solidifies on the Pt substrate, forming a glassy layer.

Cooling is realized using a water flow directed to the back side of the target (Pt substrate), and a gas flow, most often Helium, directed to the front side (TeO_2layer). Due to the low thermal conductivity TeO_2 ($30\,\mathrm{mW\cdot cm^{-1}\cdot K^{-1}}$ [2]), the process of heat transfer from the target substance to the cooled substrate becomes difficult, and the front gas cooling makes a very small contribution to the heat removal. In this regard, in order to increase the capability of radioisotope production, it is necessary to implement more intensive cooling of the front surface of the target. For this purpose, in the laboratory of radioactive substances and technologies of Tomsk Polytechnic University, a front target cooling device based on a flow of liquid droplet using ultrasonic sputtering was developed and tested. During the experiments, various target cooling modes and their effect on the target temperature were studied.

In connection with the empirically proven effectiveness of the new cooling method, there is a need to evaluate its effectiveness by determining the heat transfer coefficient of the cooled surface, taking into account experimentally determined target temperature parameters.

1. Cooling method description

Typically, air or Helium cooling is used to cool the front side of the target during irradiation in the cyclotron beamline [3,4]. This is primarily due to the low gas activation by the beam and the low energy losses of charged particles when passing through the gas layer. At the same time, the gas coolant has low thermal conductivity, which is why even when using high gas flow rates (Fx18xE m/s) , the removed thermal power is at the level of 20−30 W.

An alternative method for cooling the front side of the target was developed and tested at the R7M cyclotron of the Tomsk Polytechnic University [5]. The essence of the method is to use finely dispersed water as a coolant. For this purpose, a cooling device, including an ultrasonic oscillating system (UOS), a peristaltic pump and a tank

Table 1. Iodine isotopes and their applications

Isotope	Decay type	Half-life	Application
^{120g}I	EC*: 100%	1.35 h	SPECT**
^{123}I	EC: 100%	13.2 h	SPECT
^{124}I	β^+: 77.3% EC: 22.7%	4.2 days	PET*** + therapy
^{125}I	EC 100%	59.4 days	Brachytherapy

Note. $*$C — electron capture, $**$ — single photon emission tomography$* * *$ — positron emission tomography.

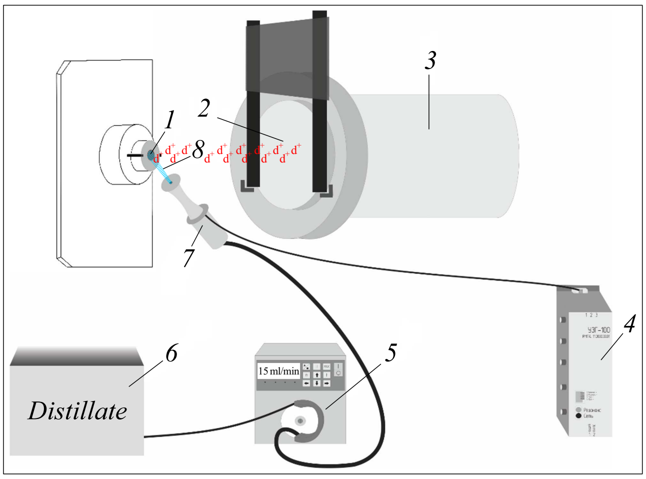

Figure 1. Target cooling scheme: *1* — cooled target, *2* — charged particle beam, *3* — beamline, *4* — ultrasonic oscillator, *5* — peristaltic pump, *6* — tank with distilled water, *7* — cooling system base on UOS , *8* — spray torch.

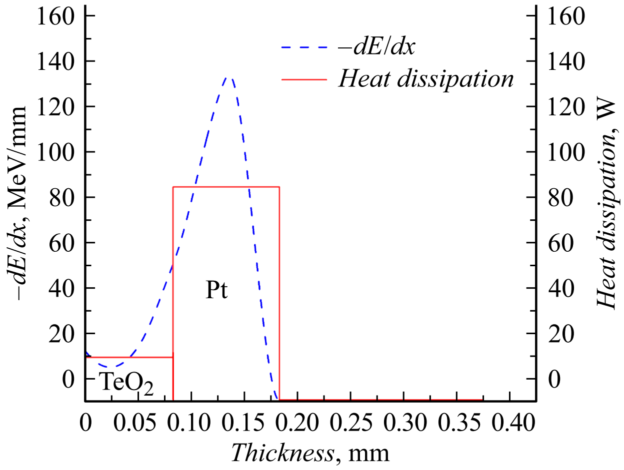

Figure 2. Change in the value of specific energy losses and heat dissipation power over the thickness of a two-layer target (thickness TeO_2 is 0.083 mm, thickness Pt is 0.29 mm).

with distilled water was developed (Fig. 1). The UOS creates a sputtering plume consisting of water droplets with a diameter of 80−100 μm, directed at the front (relative to the beam) side of the irradiated target. TeO_2, deposited on a Pt substrate, was used as a target material.

The main idea of the developed method is that water, falling on the heated surface of the target, partially evaporates and cools the target due to the energy of the phase change. In this regard, it is important to select such a flow of sprayed water so that, on the one hand, the water does not evaporate completely and maintains a minimal liquid film on the target surface; on the other hand, the cooling mode should not turn into the usual convective washing of the target with water. These cooling modes depend on the target surface temperature and can be adjusted by increasing or decreasing sputtering performance using a feedback system planned for implementation in the future.

2. Energy loss and heat generation in the target

The main way of heat generation in the target during the charged particles passing through matter is the process of ionization stopping, which occurs when a charged particle interacts with electrons in atomic shells. In this case, the kinetic energy of a charged particle is spent on excitation and ionization of atoms of the substance through which it transits. Energy losses due to ionization play a dominant role because the cross section of the Coulomb interaction with atoms exceeds the cross section of the interaction with nuclei. [6].

To calculate energy losses in the TeO_2 and Pt layers, the SRIM [7] program was used. The specific ionization losses and the dependence of the range on energy, are given in Table 2.

The target was irradiated with a 13.6 MeV deuteron beam and a beam current of 10 μA. After passing alternately Be−Al metallic foil separating the vacuum from the atmosphere, a layer of air between the foil and the target, and a layer of water, the beam energy decreased to 11.3 MeV. Using the data from Table 2, it was established that the heat dissipation power in theTeO_2 layer is equal to 19 W, in the Pt — 94 W layer (Fig. 2).

From Fig. 2 it can be seen that the beam deposits 19 W of heat in TeO_2 and then loses all the remaining heat in Pt.

Since the target is cooled only from the front side (from the beam side), the heat released in the TeO_2 and Pt layers goes towards the cooled surface, and the temperature

Table 2. Specific energy losses and range of high-energy deuterons in $^{122}TeO_2$and Pt

Energy, MeV	$^{122}TeO_2$		Pt	
	$-dE/dx$, MeV/mm	Range, mm	$-dE/dx$, MeV/mm	Range, mm
14	18.87	0.459	47.52	0.185
13	19.88	0.406	49.74	0.165
12	21.02	0.355	52.23	0.146
11	22.33	0.308	55.04	0.128
10	23.84	0.263	58.25	0.111
9	25.62	0.222	61.19	0.094
8	27.74	0.183	66.28	0.079

gradient is also directed there. From this it follows that the experimentally measured temperature [5] on the Pt surface $(129 \pm 0.51^\circ C)$ is the maximum, which is taken into account in the further calculation of the heat transfer efficiency.

3. Geometry and boundary conditions

The simulation was carried out using the Heat Transfer in Solids and Fluids module of the COMSOL Multiphysics [8] software package. This module allows to simulate the process of heat transfer in solids using the finite element method (FEM). The simulating process consists of the following steps: defining the geometry, selecting the material to model, selecting the heat transfer type, defining boundary and initial conditions, defining the finite element mesh, selecting a solver, and visualizing the results.

To solve the problem, the geometry of a Pt disk with a cavity on which theTeO_2 layer is located was created (Fig. 3). The properties of the simulated materials are presented in Table 3. When creating the target geometry, it was assumed that the TeO_2 layer is distributed uniformly over the Pt substrate cavity and its thickness is the same over the entire area.

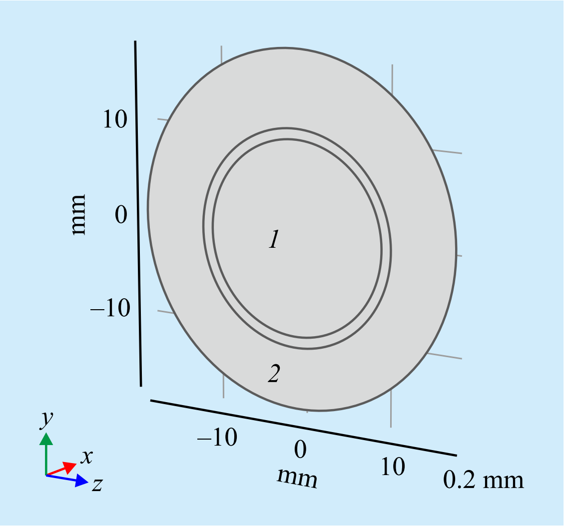

Figure 3. TeO_2 (*1*)-Pt (*2*) target geometry Target dimensions: diameter TeO_2 — 20 mm, thickness — 0.083 mm; diameter Pt — 36 mm, thickness — 0.29 mm. The TeO_2 layer is located in a special Pt substrate crucible with a diameter of 22 mm.

To simulate heat transfer in solids, the heat conduction equation is used, which is a differential form of Fourier's law:

$$\rho C_p u \cdot \nabla T + \nabla q = Q, \tag{1}$$

where ρ — density of the solid, [kg/m^3]; C_p — heat capacity of a solid at constant pressure; u — velocity field, [m/s]; Q — source of volumetric heat release, [W/m^3]; q — heat flux, [W/m^2], which is determined based on the Fourier equation:

$$q = -k\nabla T, \tag{2}$$

where k — thermal conductivity of the material, [W/(m·K)]; ∇T — temperature gradient, [K/m].

Since the simulation is carried out for a stationary case, the temperature field is considered steady and independent of time.

Due to the fact that the cyclotron beam has a Gaussian density distribution, the volumetric heat release was specified using the Deposited Beam Power and the Top-hat density distribution type. Based on the beam dynamics of the R7M cyclotron, the beam radius R was chosen equal to 11 mm and the size of the transition smoothing zone $\Delta R = 11$ mm, which corresponds to an rounded profile (discontinuous cylindrical beam). It is important to note that the target has a 90° geometry to the beam, the axis x is directed perpendicular to the irradiated surface of the target, i.e. in the direction of movement of the ion beam.

Assessment of the intensity of heat removal on the cooled surface of the target is determined through the heat transfer coefficient from Newton's–Richmann's equation:

$$q_0 = h\Delta T, \tag{3}$$

where q_0 — heat flux across the heat exchange boundary, [W·m^{-2}]; h — heat transfer coefficient,[W·m^{-2}·K^{-1}]; ΔT — difference between hot wall temperature and coolant temperature.

The model was solved with the following boundary conditions:

— heat deposition in the TeO_2 layer: 19 W;

Table 3. Basic properties of materialsTeO_2 [9] and Pt [10]

Layer	Density (ρ), $g \cdot cm^{-3}$	Thermal Conductivity (k), $W \cdot m^{-1} \cdot K^{-1}$	Specific Heat Capacity (lC_p), $J \cdot kg^{-1} \cdot K^{-1}$	Melting Point (T_{melt}), °C
TeO_2	5.67	3	398	733
Pt	21.47	71.7	132.6	1769

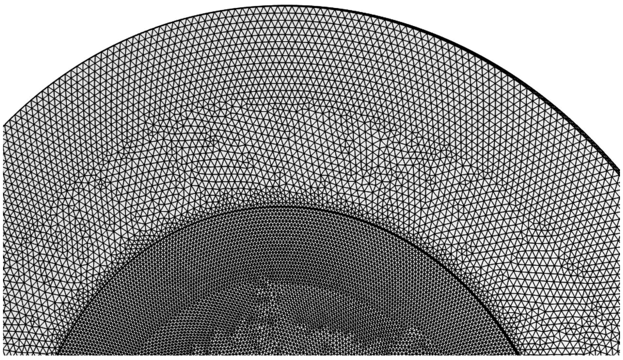

Figure 4. Graphical representation of the object under study along with the mesh used for computational simulation.

— heat deposition in the Pt layer: 94 W;

— heat transfer coefficient on the front surface of the target: from 9000 to 10000 $W \cdot m^{-2} \cdot K^{-1}$;

— all other surfaces of the target are adiabatic.

The mesh for solving the thermal problem was specified using a built-in physics-controlled mesh node with extremely fine element sizes (Fig. 4). To study the stability of the solution, calculations were carried out with a gradual increase in the number of mesh nodes; in all cases, the convergence of the solution results reached the level of 10^{-3}.

4. Results and discussion

Compositionally, tellurium dioxide is a ceramic with poor thermal conductivity ($3\,W \cdot m^{-1} \cdot K^{-1}$), resistant to fairly high thermal loads without damage. This is evidenced by the method of preparing the target itself, which consists of melting the TeO_2 powder located in the recess of the Pt substrate and further cooling to form a glassy layer. Evenly heated or cooled, Tellurium dioxide does not deteriorate or crack. However, the thermal effect of charged particles beam when irradiated in a cyclotron is very different from heating in a vacuum furnace and has the following features.

1. The center of the target always heats up more than the periphery, since the flux density of charged particles in the beam volume is uneven and obeys a Gaussian distribution.

2. The specific energy losses of particles when passing through a substance are nonlinear because they increase as the beam energy decreases, and the target layers located in the middle and closer to the substrate heat up more than the surface ones. It follows from this that the thicker the oxide layer, the greater the temperature gradient in the substance. These features of the dynamics of thermal losses of the beam, coupled with the poor thermal conductivity of tellurium dioxide, lead to the formation of „hot spots“, i.e. places of local overheating of the target substance, leading to its destruction.

Table 4. Calculated value of the maximum temperature Pt depending on the heat transfer coefficient on the front surface of the target

Heat transfer coefficient, $W \cdot m^{-2} \cdot K^{-1}$	Temperature Pt, °C
9000	136.20
9500	131.39
10 000	127.05

To determine the intensity of heat removal, the temperature field was calculated at various heat transfer coefficients on the front cooled side of the target (Table 4). From the results it is clear that the temperature value closest to the experiment is calculated at a heat transfer coefficient equal to 9500 $W \cdot m^{-2} \cdot K^{-1}$ (131.39°C — calculated temperature and 129 ± 0.51°C — experimental one).

The temperature distribution is shown in Fig. 5. For a heat transfer coefficient of 9500 $W \cdot m^{-2} \cdot K^{-1}$, the maximum temperature is at uncooled Pt (131.4°C), while the TeO_2 layer has a maximum temperature of 108°C. Comparison of calculated values with experimental data suggests that the intensity of heat removal on the front surface of the target using finely dispersed water spraying is 9000–10 000 $W \cdot m^{-2} \cdot K^{-1}$. This value significantly exceeds the heat transfer coefficients that can be achieved using a multi-jet gas cooling system (200–300 $W \cdot m^{-2} \cdot K^{-1}$ at gas speed ~ 60 m/s [11]).

The temperature gradient presented in Fig. 6 shows the temperature change across the thickness of the TeO_2 and Pt layers. Due to poor thermal conductivity properties, the temperature of the thinner TeO_2 layer changes more than that of the thicker Pt layer, which once again proves the need for intensive front cooling of the two-layer target.

In the original experiment, when the target was irradiated with beam current $10\,\mu$A , a thermal balance was established, as a result of which the target temperature was constant, however, after increasing the beam current $15\,\mu$, the target temperature began to continuously increase and

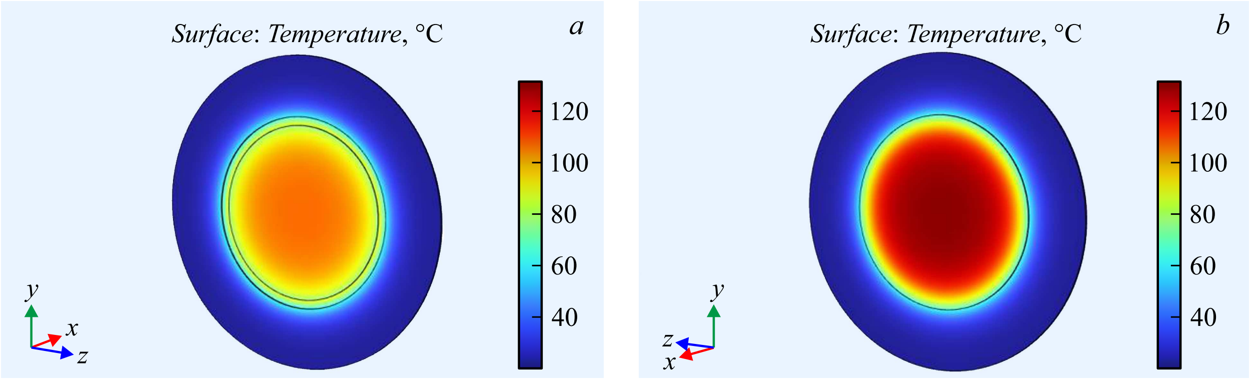

Figure 5. Temperature field of the target at a heat removal intensity of 9500 W·m^{-2}·K^{-1}; *a* — front view, *b* — rear view. The maximum temperature is observed in the Pt substrate (*b*).

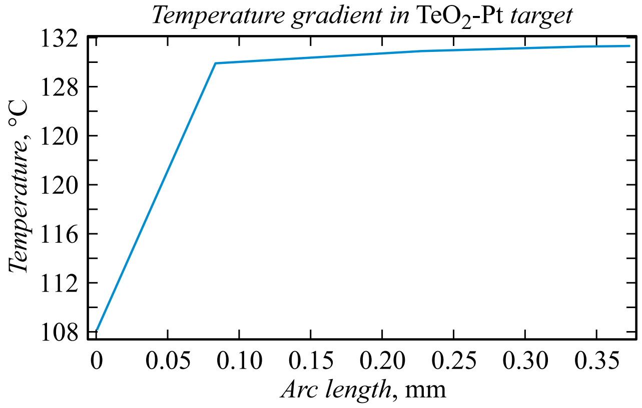

Figure 6. Temperature change along the depth of the target for a heat removal intensity of 9500 W·m^{-2}·K^{-1}. The temperature gradient inTeO$_2$ (the left quarter of the figure Arc length < 0.083 mm) is much greater than in Pt, which is explained by the difference in the thermal conductivity of the materials (about 20 times).

reached values above 200°C, which ultimately led to its destruction. Since the TeO$_2$ layer is under the influence of the beam, there is no possibility of directly measuring the temperature, but we can draw an indirect conclusion about the target destruction criterion, which is as follows. At the end of the experiment, the target material did not peel off completely, but only in places of local overheating (hot spots), where the temperature exceeded the melting point TeO$_2$. This was due to unequal heating, since, despite all attempts to change the beam profile in the direction of reducing the heat release density, it is not possible to make it ideal throughout the entire volume. It is also impossible to predict such overheating using mathematical simulating tools. However, the average temperature of the TeO$_2$ layer were estimated, at which the target remains intact. Based on the calculation results, it can be assumed that the temperature 130–150°C is the maximum at which the target is not destroyed.

Conclusion

Using the COMSOL Multiphysics software package, the simulating of irradiation of a TeO$_2$+Pt target with a 13.6 MeV deuteron beam and front target surface cooling with a flow of finely dispersed water with a flow rate of 15 ml/min was carried out. Based on the simulation results, it was established that the intensity of heat removal on the cooled surface using the specified method is 9000–10 000 W·m^{-2}·K^{-1}, while the temperature TeO$_2$ is 108°C. The results of the study will be used to optimize the technology of radio-Iodine production at a cyclotron.

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Conflict of interest

The authors declare that they have no conflict of interest.

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