

## SECTION 4

# DIAGNOSTICS AND RESEARCH METHODS

UDC 621.039.8.002

## NEW TARGET ASSEMBLY FOR SOLID SAMPLES IRRADIATION IN 11-MeV MEDICAL CYCLOTRON

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In this work we present the new target assembly developed for solid samples irradiation by protons with different energies at maximum of 11 MeV and 40  $\mu$ A for current. The new target was designed and manufactured for conventional target changer (carousel) of 11 MeV Eclipse RD medical cyclotron. The MCNP simulation of the beam profile after degradation was performed and testing irradiation of the target assembly was successfully carried out showing the target suitability for such type of cyclotron facilities.

### INTRODUCTION

There are a lot of medical isotopes that can be produced by solid samples irradiation via  $(p, n)$ ,  $(p, 2n)$  nuclear reactions. The bigger part these reactions has the thresholds above 10 MeV, but some of them can also occur at lower energies, producing commonly used isotopes for nuclear medicine. In particular, the  $^{99m}\text{Tc}$  for SPECT and  $^{64}\text{Cu}$ ,  $^{68}\text{Ga}$  [1] for PET imaging can be produced by low energy protons (Table 1) using small medical cyclotrons. Despite low thresholds, the high current of protons is still required for sufficient isotope yield leading to high energy release. In typical production conditions the temperature of the samples and surroundings can reach the values up to 400...800 K [2]. Therefore special design of the target and geometry of irradiation must be realized for safety production performance.

Table 1

Possible reactions for production of nuclear medicine isotopes

Isotope	Reaction	Q-value, MeV
$^{99m}\text{Tc}$	$^{100}\text{Mo}(p,2n)^{99m}\text{Tc}$	-7.7
$^{68}\text{Ga}$	$^{68}\text{Zn}(p,n)^{68}\text{Ga}$	-3.7
$^{64}\text{Cu}$	$^{64}\text{Ni}(p,n)^{64}\text{Cu}$	-2.5

Along with production issues, the low incident energies within the high currents allow investigating rare nuclear reactions near and below the thresholds (in particular with dineutron in the outgoing channel [3], sub-barrier proton-induced reactions [4–5], proton radiative capture etc. [6]), that are important for correct cross sections measurements and can give valuable information about the nuclear structure. In this connection small cyclotrons (below 20 MeV) are very convenient to get the protons with lower than designed energies using degraders, in which small energy release is expected relative to the high energy machines.

To cover the tasks described above, the targets containing degraders and with good heat dissipation must be applied for conventional medical cyclotrons. In

this study we present the new target assembly for 11-MeV Eclipse RD cyclotron, installed in All-Ukrainian Center for Radiosurgery of the Clinical Hospital "Feofaniya" [7, 8]. Nowadays this cyclotron is exploited only for  $^{18}\text{F}$  production using silver targets for  $^{18}\text{O}$ -water irradiation, but the analysis in [9] showed sufficient yield of  $^{99m}\text{Tc}$  in such facilities using the solid  $^{100}\text{Mo}$  sample (other tracers from Table 1 can also be produced). Hence the development of new target assembly for solid samples irradiation offers additional opportunities for isotopes production and scientific investigations of  $(p, x)$  reactions using the activation technique.

### DESIGN AND TEMPERATURE PROFILE CALCULATIONS

The new target assembly was designed according to the following requirements: suitability for conventional Eclipse RD cyclotron, safe and effective usage of expensive enriched samples (for instance, the 99% enrichment of  $^{100}\text{Mo}$  costs 2000...4000 \$/g [10]) and flexibility for scientific researches with different protons energies. The structural concept and scheme of target assembly under the beam is presented in Fig. 1. The main components of the assembly are Al body, Cu sample's holder and degraders. The incident 11-MeV proton beam enters the assembly through the vacuum window and degraders, and then strikes the solid sample (Fig. 1 shows the case of Molybdenum pellet sample with diameter of  $d_{\text{Mo}} = 15$  mm and thickness of  $l_{\text{Mo}} = 3$  mm), placed in the copper sample's holder. This holder is housed in cylindrical aluminum body with external dimensions of 99 x  $\varnothing$ 27 mm and with the orifice for cooling water with diameter of  $d_w = 3$  mm located at 5 mm from holder's side. This assembly is installed in a chamber of cyclotron's target changer (carousel).

The temperature profile at maximum heat transfer of 440 W (11 MeV and 40  $\mu$ A current) was calculated using one-dimensional equation for heat flow  $Q_x$ :

$$Q_x = -\frac{\lambda}{l} S \frac{\partial T}{\partial x}, \quad (1)$$

where  $\lambda$  – thermal conductivity of corresponding material;  $l$  – thickness of the material;  $S$  – area of heat input normal to the  $x$  direction, and  $T$  – temperature.

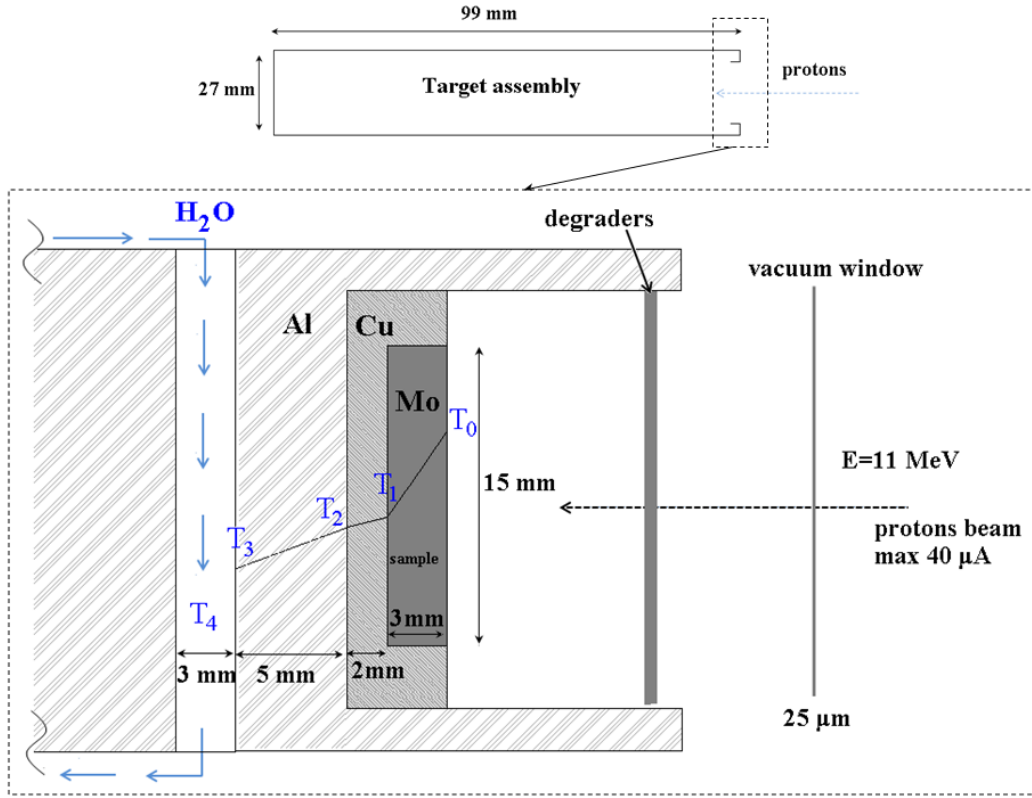


Fig. 1. The scheme of target assembly: external dimensions and zoomed view of the front part of the assembly (in dotted box)

The  $x$ -axis coincides with beam direction. Since  $\lambda$ ,  $l$ ,  $S$  are assumed to be constant within considered range of temperatures (400...800 K) and heat transmission  $Q_x$  is steady, the temperature of the assembly's components and sample were calculated as

$$\begin{aligned} T_2 &= T_3 + \frac{l_{Al} \cdot Q_x}{\lambda_{Al} \cdot S}; & T_1 &= T_2 + \frac{l_{Cu} \cdot Q_x}{\lambda_{Cu} \cdot S}; \\ T_0 &= T_1 + \frac{l_{Mo} \cdot Q_x}{\lambda_{Mo} \cdot S}, \end{aligned} \quad (2)$$

with  $S = \pi \cdot d_s^2 / 4$  for area of the heat input,  $l_{Al} = 5$  mm for Al body thickness from the holder to cooling water channel, and  $l_{Cu} = 2.6$  mm for Cu holder thickness. The temperature of the Al body side  $T_3$  was calculated using convection equation

$$\begin{aligned} T_3 &= T_4 + \frac{Q_x}{h \cdot S_w}; & h &= \frac{Nu \cdot \lambda_w}{d_w}; \\ Nu &= 0.023 \cdot Re^{0.8} \cdot Pr^{0.4}. \end{aligned} \quad (3)$$

Here  $T_4 = 290$  K is cooling water temperature;  $h$  – heat transfer coefficient;  $S_w$  – the effective area of heat transfer between water and Al side;  $\lambda_w$  – thermal conductivity of water;  $Nu$ ,  $Re$ ,  $Pr$  – Nusselt, Reynolds, and Prandtl number respectively. The last ones were calculated from formulas

$$Re = \nu \cdot \frac{d_w \cdot \rho}{\eta}, \quad Pr = \frac{\eta \cdot C}{\lambda_w}, \quad (4)$$

where  $\nu$  stands for velocity of water, and  $\eta$ ,  $\rho$ ,  $C$  are viscosity, density, and specific heat of water. These parameters and calculated values for convection heat transfer are presented in Table 2.

Table 2

Parameters of temperature profile calculations

$\eta$ , kg/(m·s)	$\rho$ , kg/m <sup>3</sup>	$C$ , J/(kg·K)	$\nu$ , m/s	$Pr$	$Re$	$Nu$	$h$ , W/(m <sup>2</sup> ·K)	$\lambda_w$	$\lambda_{Al}$	$\lambda_{Cu}$	$\lambda_{Mo}$
								W/(m·K)*			
10 <sup>-3</sup>	10 <sup>3</sup>	4183	5.34	7.2	1604	117	22670	0.58	217	350	120

\*The values of thermal conductivities are presented for  $T = 800$  K for solids and  $T = 290$  K for water.

The results of the temperature profile calculations are presented in Table 3. It can be seen that proposed scheme of irradiation ensures temperature profile within

34...70% safety factors relative to the critical values, which were chosen as melting points of assembly's components materials.

Table 3  
Calculated temperatures of the target assembly components under the beam

Temperature	Critical value, K	Calculated value, K	Safety factor, %
$T_0$	2380 (Mo)	702	70
$T_1$	1356 (Cu)	640	53
$T_2$	933 (Al)	622	34
$T_3$	933 (Al)	565	40

### TESTING IRRADIATION AND MCNP SIMULATIONS

The new target assembly was manufactured at Hospital's workshop and it is shown in Fig. 2. Apart from the parts described above, it also includes degrader's holder and clamp. The degraders are cooled by helium cooling system designed for heat dissipation from 25  $\mu\text{m}$  Al vacuum window.

The assembly was installed in cyclotron's target chamber and then tested under the proton beam with the energy of 10.79 MeV (after vacuum window) and current of 5  $\mu\text{A}$  during 10 s (the total number of protons on holder was  $3 \cdot 10^{14}$ ). The testing irradiation was carried out without sample and with 50  $\mu\text{m}$  Al degrader. To check the beam position the paper target was placed in Cu sample's holder. It can be seen from Fig. 2,b that the beam is fully positioned on the sample's holder and that is of special importance for correct calculations of protons hitting the sample, while the measured current of protons is the total incident current to the assembly.

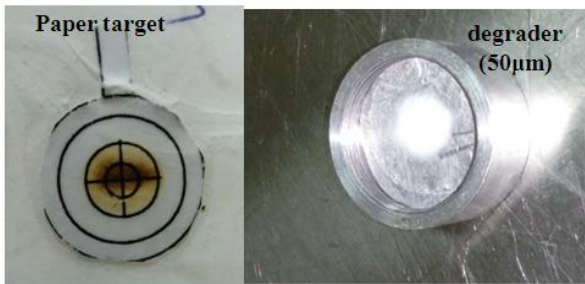
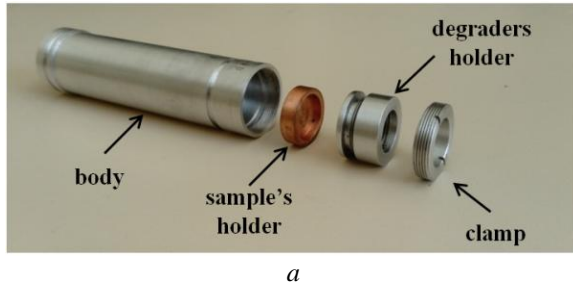


Fig. 2. Target assembly components: general view (a) and after testing irradiation (b)

The output energy of protons after degradation (the incident energy to the sample's holder) can be calculated using the Bethe-Bloch formula for energy losses of protons inside aluminum:

$$\frac{dE}{dx} = \frac{2 \pi z z_p e^4 \rho N_A m_p}{m_e E M} \times \left( \ln 2 \left( m_e \frac{2E}{I m_p} \right) - \ln \left( 1 - \frac{2E}{c^2 m_p} \right) - \frac{2E}{c^2 m_p} \right), \quad (5)$$

where  $z, M$  – atomic number and mass of Al isotope;  $z_p, m_p$  – charge and mass of proton;  $e, m_e$  – elementary charge and electron mass;  $E$  – the energy of protons;  $\rho$  – density of aluminum;  $c$  – the speed of light;  $N_A$  – Avogadro constant;  $I$  – the mean excitation potential for Al, that can be calculated with the help of semi-empirical expression from [11]:

$$I = (9.76 + 58.8 \cdot z^{-1.19}) \cdot z = 162.9 \text{ eV}. \quad (6)$$

The beam degradation leads to the energy spread near the specified output energy  $E_{out}$  and can also result to the low-energy "tail" of the protons. The low-energy spectrum usually has intensities about 5...10% of the intensity maximum [12, 13]. The spread of the main peak also results to significant flux of protons within the energy range  $E_{out} \pm \Delta E$ . The spread of the beam energy  $\Delta E$  after degradation can be estimated with the help of Gaussian distribution for the number of pairs produced by protons inside the vacuum window  $N_w$  and degrader  $N_d$ , in assumption that energy losses go only to ionization:

$$\begin{aligned} E_{out} \pm \Delta E &= E_{in} - E_d - E_w \pm \Delta E = \\ &= E_{in} - (N_d + N_w)I \pm \sqrt{(N_d + N_w)I^2}. \end{aligned} \quad (7)$$

Here  $E_{in} = 11 \text{ MeV}$  is the incident energy of protons;  $E_w, E_d$  – energies, lost by protons inside vacuum window and degrader. According to above considerations, the output energy of protons after 25  $\mu\text{m}$  vacuum window and 50  $\mu\text{m}$  degrader equals  $(10.35 \pm 0.0) \text{ MeV}$  within the  $2\sigma$  confidence level.

The beam spread was also simulated using MCNPX code [14]. The model of mono-energetic protons with initial energy of 11-MeV going through the vacuum window, He-cooling channel and degrader of 50  $\mu\text{m}$  Al was implemented. The  $10^8$  protons were generated and the energy bin width of 0.077 MeV was used. The results of simulations compared with calculations performed in accordance with (5)–(7) are presented in Fig. 3 and Table 4.

It can be seen from Fig. 3 that the output energy in case of MCNPX simulations is almost the same (10.36 MeV) as for Bethe-Bloch formula calculations, but the spread of energy is about twice bigger. The spectrum of protons (Fig. 4) obtained in MCNPX simulations has the low-energy tail (below 10.2 MeV), caused by inelastic multiple scattering and nuclear interactions. In such condition the beam core and tail are considered with corresponding number of particles  $S_{core}$  and  $S_{tail}$ .

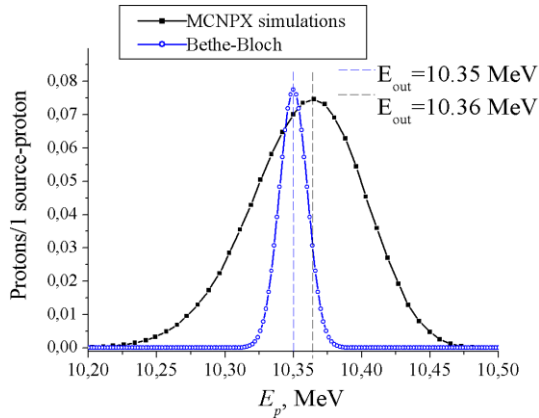


Fig. 3. The spread of the beam energy near the output energy  $E_{out}$

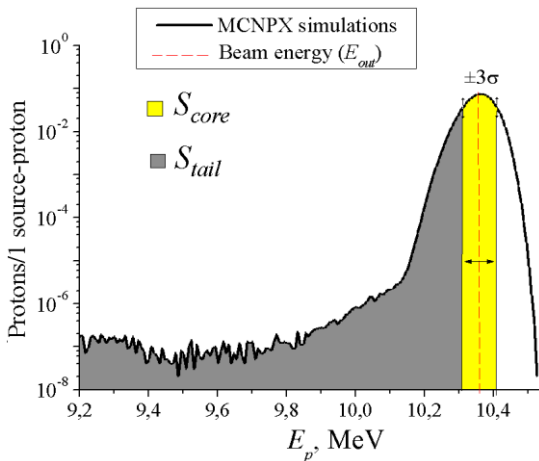


Fig. 4. The spectrum of protons on sample's holder

Table 4

The values of the beam energy spread

	$E_{out}$	$\sigma$ , MeV	FWHM, MeV
MCNPX	10.36	0.017	0.082
Bethe-Bloch with Gaussian distribution	10.35	0.010	0.048

The proton belongs to the core if it has the energy within  $3\sigma$  of the output energy. This requirement ensures more than 99% of particles in the main peak to populate the core, and then output energy equals

$$E_{out} = (10.36 \pm 0.05) \text{ MeV}. \quad (8)$$

The contribution of the low-energy tail  $C_{tail}$  to the full energy spectrum is calculated by the expression

$$C_{tail} = \frac{S_{tail}}{S_{tot}} = \frac{S_{tail}}{S_{tail} + S_{core}} = 11.2\%, \quad (9)$$

where  $S_{tot}$  is the total number of protons in the spectrum. This contribution must be taken into account for correct yield estimation. According to this result, the part of protons that strike the sample within the energy range (8) is about 89% of incident protons current. The low-energy contribution can be reduced by the usage of materials with minimum cross sections for nuclear interactions and low ionization potentials. Besides, the reduction of nuclear interactions minimizes the emissions of secondary particles, which are undesirable background in both cases – tracer production and research irradiation. To keep the beam on the sample the multiple scattering angles also must be minimized, which depends on radiation length of materials. All these facts indicate that the degraders with low atomic numbers are well suited for protons degradation. Along with aluminum other materials are also going to be considered in our further investigations.

## CONCLUSIONS

The new target assembly for solid samples irradiation in 11-MeV cyclotron was designed and manufactured. The option of degrader installation was realized to vary the energies of protons for different studies of proton-induced reactions. The assembly was tested under the 10.79 MeV protons and the beam was fully positioned on the sample's holder. The output energy and spread of the beam after vacuum window and degrader was calculated using Bethe-Bloch formula with statistical fluctuations and then simulated in MCNPX code. The results have good agreement for the output energies, but the energy spread of the beam is almost twice bigger for MCNPX, where inelastic scattering and nuclear interactions are also taken into account. According to simulations, in case of 50  $\mu\text{m}$  Al degrader usage the 89% of the protons have the output energy of  $(10.36 \pm 0.05)$  MeV.

The testing irradiation was carried out and it showed the suitability of developed target assembly for low-energy cyclotrons, allowing irradiations of solid samples in harsh conditions. There are about 950 small medical cyclotrons installed all over the world, and the implementation of such assembly will increase the efficiency of cyclotron usage for isotopes production for nuclear medicine purposes as well as for scientific investigations of proton-induced reactions.

## REFERENCES

1. D. Mueller et al. First Results of Cyclotron Produced  $^{68}\text{Ga}$  Using Solid Targets // *Journal of Nuclear Medicine*. 2019, v. 60, p. 1163.
2. Technical reports series No.465 "Cyclotron produced radionuclides: principles and practice". Vienna, IAEA, 2008, p. 215.
3. I.M. Kadenko. A new type nuclear reaction on  $^{159}\text{Tb}$  in the outgoing channel considering observation of a bound dineutron // *Acta Physica Polonica B*. 2019, v. 50, p. 55-64.
4. I. Gheorghe et al. Absolute Cross Sections for Proton Induced Reactions on  $^{147,149}\text{Sm}$  Below the

Coulomb Barrier // *Nuclear Data Sheets*. 2014, v. 119, p. 245-248.

5. G.G. Kiss. *Proton induced reactions and the astrophysical p process*: Ph.D. thesis. University of Debrecen and Institute of Nuclear Research of the Hungarian Academy of Sciences (ATOMKI), Debrecen, 2008, p. 105.

6. F.R. Chloupek et al. Measurements of proton radiative capture cross sections relevant to the astrophysical rp- and  $\gamma$ -processes // *Nuclear Physics A*. 1999, v. 652(4), p. 391-405.

7. B.M. Bondar et al. F-18 production for PET imaging // *Proc. of 2-d Int. Workshop Medical physics – the current status, problems, the ways of development, innovation technologies*, Kyiv, Ukraine, 2012, p. 70-73.

8. B.M. Bondar et al. Radiation safety aspects during 11-MeV medical cyclotron operation and maintenance // *J. Kyiv Univ. News. Ser. Radiophysics and electronics*. 2014, v. 1/2(21/22), p. 16-18.

9. B.M. Bondar et al. The Study of  $^{99m}\text{Tc}$  Production Using Medical Cyclotrons in Ukraine // *Research bulletin of the National Technical University*

*of Ukraine “Kyiv Polytechnic Institute”*. Series *Engineering*. 2017, v. 6, p. 53-58.

10. Report on the 1-st Research coordinating meeting on “Accelerator based Alternatives to non-HEU Production of  $^{99}\text{Mo}/^{99m}\text{Tc}$ ”, Vancouver, Canada, April 16–20, 2012, p. 12.

11. W.R. Leo. *Techniques for Nuclear and Particle Physics Experiments: A How-To Approach*. Springer-Verlag Berlin Hiedelberg, 1994, p. 25.

12. A. Gerbershagen et al. Simulations and measurements of proton beam energy spectrum after energy degradation // *Proceedings of IPAC2017*, Copenhagen, Denmark, 2017, p. 4740-4744.

13. V. Rizzoglio et al. On the accuracy of Monte Carlo based beam dynamics models for the degrader in proton therapy facilities // *Nuclear Instruments and Methods*. 2018. <https://arxiv.org/abs/1712.00406v2>.

14. D.B. Pelowitz. *MCNPX User's Manual, Version 2.6.0*. Los Alamos National Laboratory report LA-CP-07-1473 (April 2008).

Article received 03.06.2020

## **НОВАЯ МИШЕННАЯ СБОРКА ДЛЯ ОБЛУЧЕНИЯ ТВЕРДЫХ ОБРАЗЦОВ НА 11-МЕГАЭЛЕКТРОНВОЛЬТНОМ МЕДИЦИНСКОМ ЦИКЛОТРОНЕ**

***Б.М. Бондар, А.В. Кравченко, И.Б. Михницкий, Я.В. Кметюк***

Представлена новая мишенная сборка, которая была разработана для облучения твердых образцов протонами различных энергий с максимальным значением 11 МэВ и током до 40 мкА. Новая сборка была сконструирована и изготовлена для типичного мишенного модуля медицинского циклотрона Eclipse RD. Выполнена MCNP-симуляция прохождения протонным пучком вакуумного окна и деградера. Проведено тестовое облучение, результаты которого показали применимость изготовленной сборки в низкоэнергетических медицинских циклотронах.

## **НОВА МІШЕННА ЗБОРКА ДЛЯ ОПРОМІНЕННЯ ТВЕРДИХ ЗРАЗКІВ НА 11-МЕГАЕЛЕКТРОНВОЛЬТНОМУ МЕДИЧНОМУ ЦИКЛОТРОНІ**

***Б.М. Бондар, А.В. Кравченко, І.Б. Міхницький, Я.В. Кметюк***

Представлено нову мішенну зборку, розроблену для опромінення твердих зразків протонами різних енергій з максимальним значенням 11 МеВ та струмом до 40 мкА. Нова зборка була сконструйована та виготовлена для типового мішенного модуля медичного циклотрону Eclipse RD. Виконано MCNP-симуляцію проходження протонного пучка крізь вакуумне вікно і деградер. Проведено тестове опромінення, результати якого показали застосовність виготовленої зборки для низькоенергетичних медичних циклотронів.