DEVELOPMENT OF TARGETS FOR THE RIC-80 PROJECT

V.N. Panteleev, A.E. Barzakh, D.V. Fedorov, A.M. Filatova, V.S. Ivanov,
**K.A. Mezilev, F.V. Moroz, P.L. Molkanov, S.Yu. Orlov**

1. Introduction

The rapidly growing studies in nuclear physics, developments of experimental methods and implementation of very sensitive and effective detectors have provided a very good possibility for development of a completely new direction in medical diagnostics and therapy of diseases, which is presently known as nuclear medicine. The exceptional ability of nuclear medicine methods is based on the possibility to detect and treat a large set of diseases at a very early stage, saving human lives and reducing expenses for treatment. The technology of nuclear medicine used for diagnostics and therapy is based on employing radioactive isotopes with selected properties. The common methods in nuclear diagnostics use the technology of marking medications by specific radionuclides. By measuring the distribution of radioisotopes in the human body, it is possible to observe the functions of the investigated organs or their parts. Similar methods are used for treatment of malignant tumors. In this case, the compounds containing radioactive isotopes are absorbed by malignant formations, which leads to their local irradiation and subsequent destruction. The main advantage of nuclear radiomedicine methods is that the diagnostics and treatment can be carried out at very early stages of the disease.

This work is the first part of the program of the target development for the new project of RIC-80 (Radioactive Isotopes at the cyclotron C-80) [1], which is presently being carried out at PNPI. The main task of this work is the development of methods of production and separation of the isotope-generator ^{82}Sr, which is used for PET diagnostics of heart diseases, and also of some other isotopes used for diagnostics and therapy. The methods are worked out using a 1 GeV proton beam of the synchrocyclotron with the aim to use the developed target prototypes at the new radioisotope facility RIC-80 with the proton energy of 80 MeV and the beam intensity of up to 200 μA.

2. Description of the experiment and experimental results on production of ^{82}Sr

In the first experiments, niobium foils were used as target material for production and yield measurements of ^{82}Sr. This nuclide with the half-live $T_{1/2} = 25.55$ days is the generator for its daughter isotope ^{82}Rb ($T_{1/2} = 1.25$ min), which is widely used in PET diagnostics.

A stack of niobium foils of 16 g/cm^2 thickness was placed into a Ta-W container which was designed as a prototype of the target device for production of ^{82}Sr at RIC-80. Results of FLUKA [2, 3] calculations of the ^{82}Sr activity produced in the irradiated Nb and Zr targets are presented in Fig. 1.
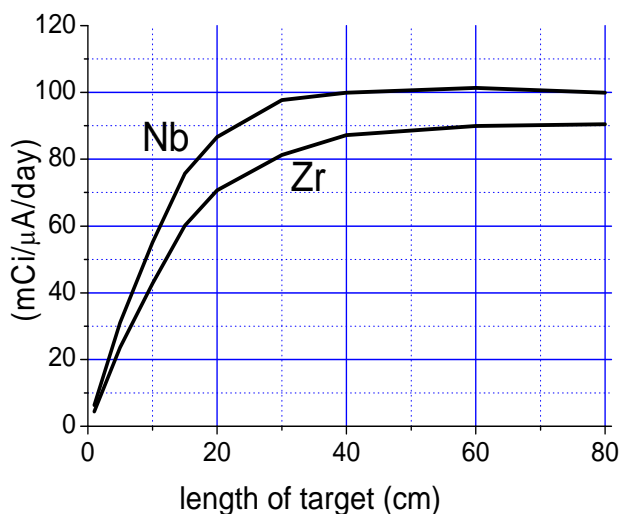
Fig. 1. ^{82}Sr radioactivity produced in the Nb or Zr foil target *vs* target length. The activity value is in mCi; the proton energy is 1 GeV; the proton beam intensity is 1 μA; the irradiation time is 24 hours

One can see in Fig. 1 that even with a rather low current of the PNPI synchrocyclotron (up to 1 μA), it is possible to obtain about 0.5 Ci of ^{82}Sr after 5 day irradiation of a target of the appropriate length of 30 cm.

For irradiation and target heating tests, the target prototype was manufactured with the parameters close to those specified for the target of the RIC-80 facility, which were the following: the target material – Nb foils of 25 mm in diameter; the target thickness – 16 g/cm^2; the length of the target container – 200 mm; the cylindrical container of Ta-W alloy – 30 mm in diameter. A target prototype (shown in Fig. 2) was fabricated for irradiation by the proton beam to accumulate ^{82}Sr in the target material.
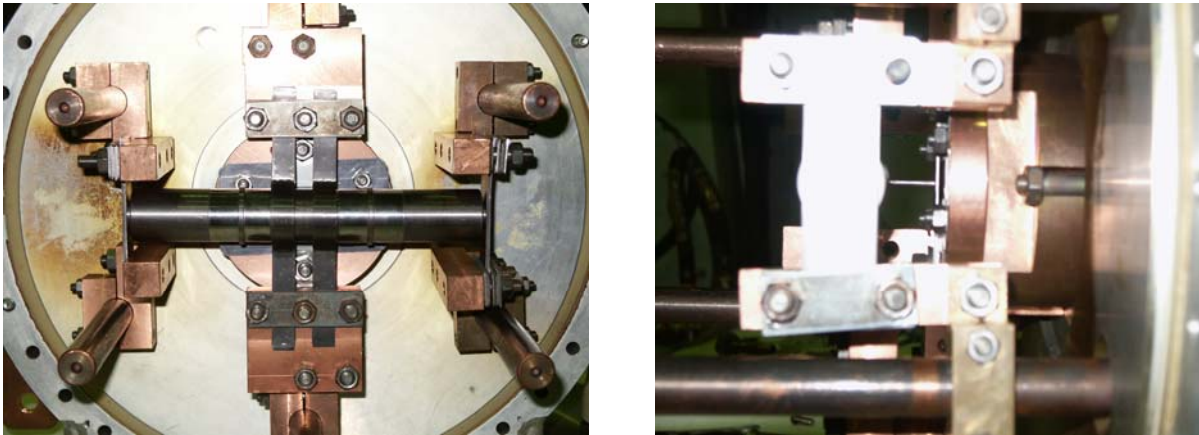
Fig. 2. Target prototype constructed for tests of production and thermal extraction of the generator isotope ^{82}Sr from the irradiated niobium foil target

The proton beam was directed along the target container with a stack of niobium foils. In the central part, the target container was connected to a tungsten tube with a hole of 2 mm in diameter. The radioactive nuclides accumulated in the target material were evaporated through this tube in the course of the target heating up to 2100 °C and were absorbed by the tantalum foil collector, which was placed in contact with the copper flange cooled by water. For the tungsten tube, along which the evaporated nuclides passed through, the same method of heating was used as that for the target container. Before irradiation by the proton beam, the container was installed and tested at a test bench. The target was heated up to its working temperature of 2000–2100 °C and subsequently cooled down, and this procedure was repeated many times under vacuum of 10^{-5} Torr.
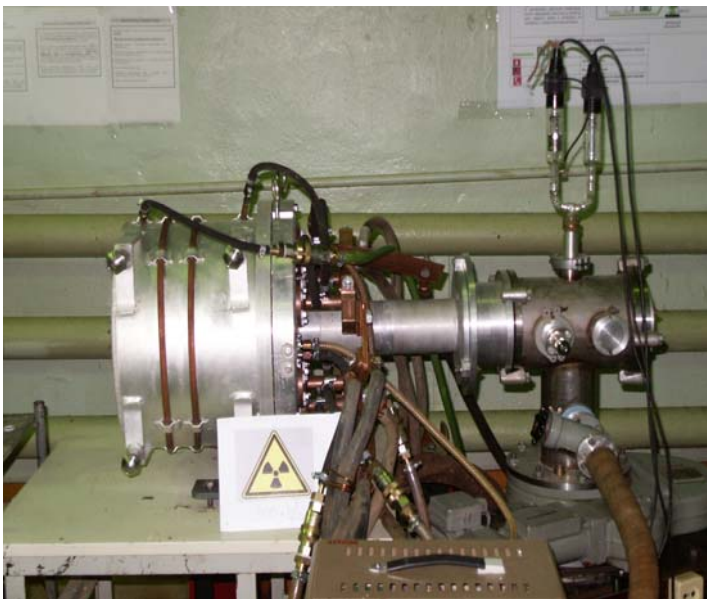
Fig. 3. Vacuum test bench with the investigated target installed. The target itself is inside of the water cooled aluminum vacuum container

The target heating tests demonstrated that the repeated process of target heating and cooling had not caused any noticeable changes of such target parameters as the shape of the target container, its position and electric resistance. The thermal investigations of the target confirmed the results of our earlier tests with high-temperature and high-density uranium carbide target units investigated in the same temperature conditions [4]. Figure 3 shows the vacuum test bench constructed for testing high-temperature targets with the heating power of up to ~ 9 kW. The parameters of the test bench heating system are the following: the current through the target is up to 1100 A; the voltage drop on the target container is up to 8 V; the target heating power is up to 8.8 kW. This target construction provides production and extraction of radionuclides from diverse target materials such as refractory metal foils, refractory metal carbides and liquid metals.

The constructed target device was installed in the PNPI synchrocyclotron proton beam for irradiation with the beam intensity of 0.1 μA during 24 hours. After two weeks of radiation cooling (the time interval for decay of short-lived isotopes), the target unit was transported to the experimental hall of the IRIS facility and placed on the test bench for ^{82}Sr extraction by target heating. Figure 4 presents the gamma-spectrum measured by an ultrapure germanium detector placed at a three-meter distance from the irradiated target before its heating started.
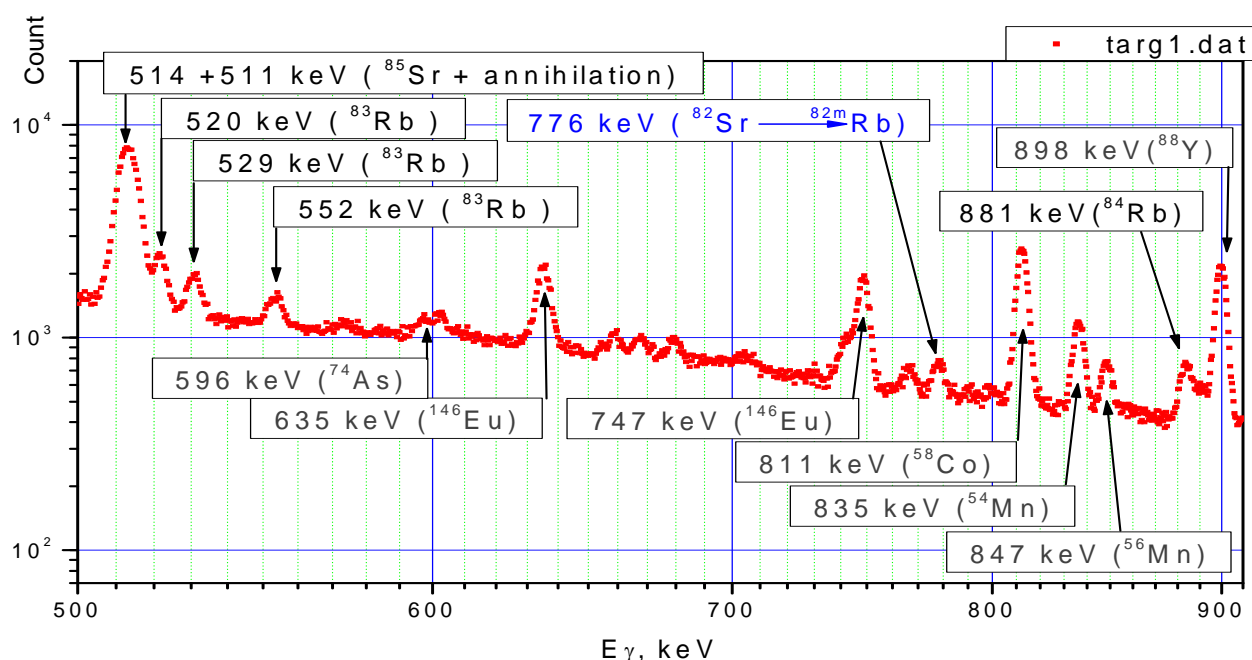
Fig. 4. Gamma-spectrum of the irradiated niobium foil target. The gamma-line of 776 keV belongs to the decay of ^{82}Sr daughter isotope ^{82}Rb used for PET diagnostics

To extract the radioactive species produced in the target material, there were several stages in the process of target heating in high vacuum. Since strontium is released from the target by relatively slow diffusion and effusion processes, the target heating was started at low temperatures of 1500–1800 °C to evaporate Rb, Mn and other radionuclides with low values of diffusion-effusion parameters. The gamma-spectrum of the radioactive species extracted from the target during two hours of heating at 1500 °C and deposited on a tantalum collector is shown in Fig. 5.

One can see in Fig. 5 that only the isotopes of easily diffusing and volatile elements (Rb, Mn) escape the target at the temperature of 1500 °C. The gamma-spectrum of radioactive species collected at the tantalum foil is shown in black; the red points show the gamma-spectrum of these radioactive species washed out of the foil-collector by alcohol. The efficiency of washing out Rb and Mn from the surface of the tantalum foil collector by alcohol was about 30 %. In the collected radioactive matter, no traces of ^{82}Sr are seen. It means that at this temperature all volatile species can be removed from the target in a rather short time (6–8 hours) without losses of strontium. After that, strontium can be selectively extracted by heating the target to higher temperatures.
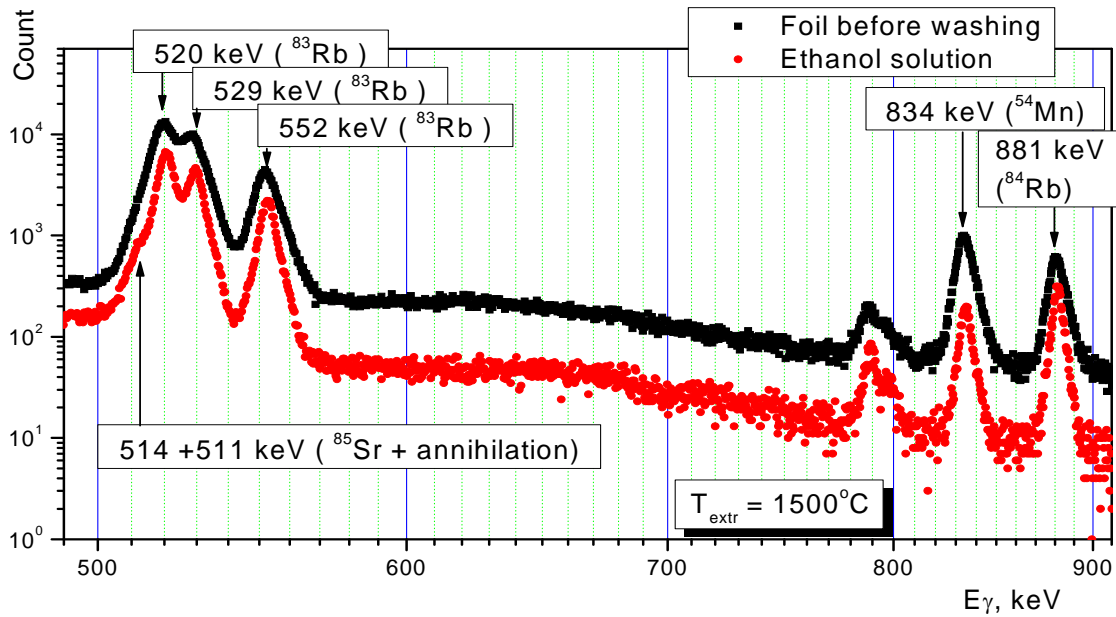
Fig. 5. Gamma-spectra of the radioactive species extracted from the target during two hours of heating at 1500 °C and deposited on a tantalum collector

The gamma-spectrum of the species collected at the temperature of 2000 °C after the target heating during 8 hours at the temperature of 1500–1600 °C is shown in Fig. 6. As one can see, all the radioactive species were finally evaporated from the irradiated target, except for strontium. The efficiency of washing out strontium from the tantalum foil collector by a solution of HNO_3 acid was 70 %. During the target heating at the temperature of 2000 °C, 93 % of ^{82}Sr was removed from the target in ten hours, with 65 % collected on the tantalum foil and 70 % transferred to the acid solution. Thus, we can estimate that the integral efficiency of our first experiment on ^{82}Sr production was 42%.
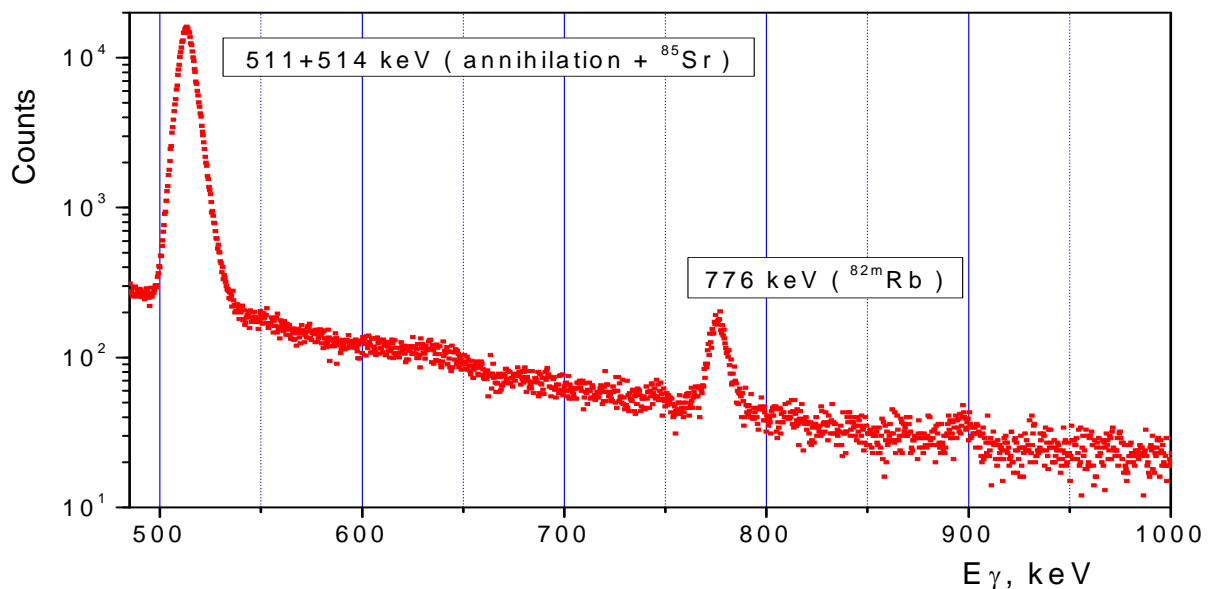
Fig. 6. Gamma-spectrum of the species collected at 2000 °C after target heating for 8 hours at 1500–1600 °C to evaporate the volatile species. During this heating process, the gamma spectrum of the collected active matter was measured every 2 hours to control that strontium did not escape

3. Production of thallium isotopes from a Pb target

The radionuclide ^{201}Tl ($T_{1/2}$ = 3.04 d) is used for diagnostics of myocardial diseases. It can be effectively produced from lead targets of natural or enriched abundance. The cross section for production of ^{201}Tl from a lead target is presented in Fig. 7.
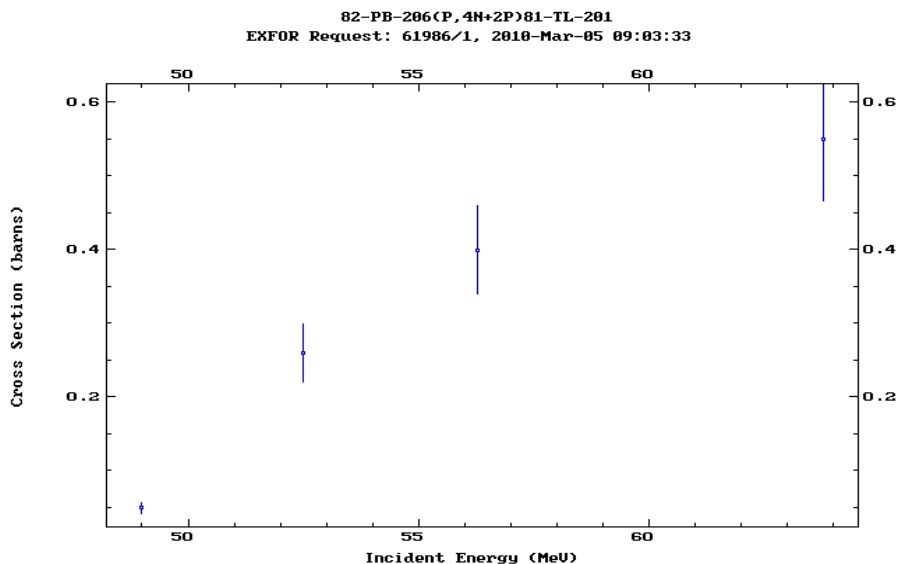

Fig. 7. Dependence of the cross section for production of ^{201}Tl from ^{206}Pb on the incident proton beam energy

One can see in Fig. 7 that the production cross sections are very high and, hence, this method can be used for effective production of ^{201}Tl. The only problem is the admixture of the neighbour isotopes with similar half-lives, ^{200}Tl ($T_{1/2}$ = 1.09 d) and ^{202}Tl ($T_{1/2}$ = 12.23 d). In this case, the mass-separator method of production of high purity ^{201}Tl can be used.

In the experiment on production and extraction of thallium isotopes, the target of 1 g lead of natural abundance was irradiated by the 1 GeV proton beam of the PNPI synchrocyclotron for 24 hours. After 10 days of cooling, it was transported to the experimental hall of the IRIS facility and was placed on the vacuum test bench into a tungsten container to be heated. The gamma-spectrum of the irradiated lead sample before its heating is shown in Fig. 8.
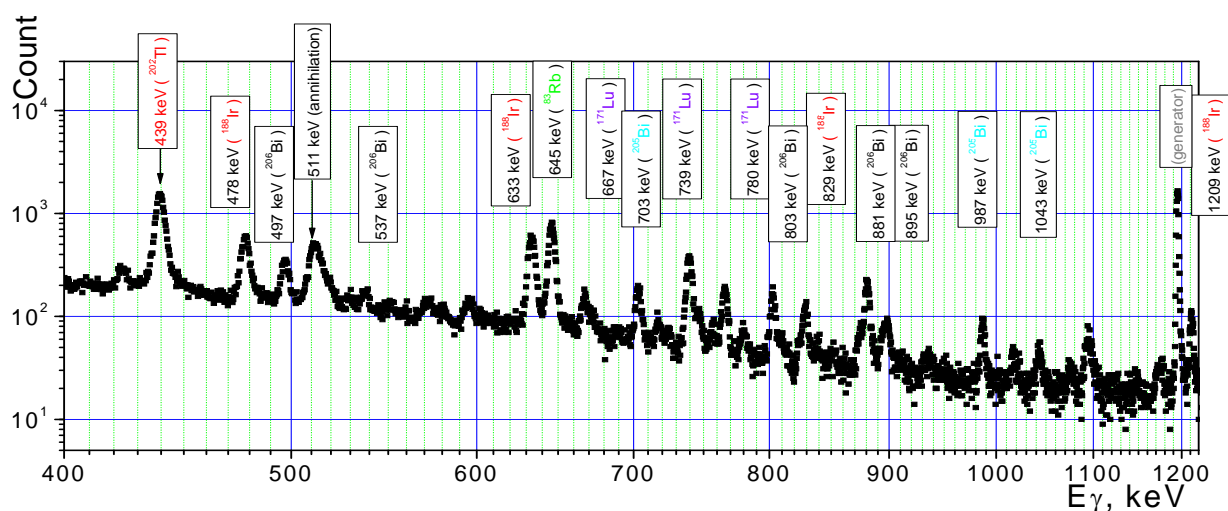

Fig. 8. Gamma-spectrum of the irradiated 1 g lead sample of natural abundance after 10 days of radiation cooling. The gamma-line of 439 keV is from the decay of ^{202}Tl ($T_{1/2}$ = 12.23 d)

The main goal of the first experiment on Tl isotopes production from a lead target was to figure out the heating conditions for selective Tl extraction from the irradiated lead sample. It was expected that thallium, which is a very volatile element, has the rate of evaporation from the melted lead target material exceeding that of other radioactive species. Since the melting point of lead is 327 °C, the extraction was carried out at 400 °C to make sure that the lead sample was completely melted. The gamma-spectrum of the species evaporated at 400 °C from the lead sample and deposited onto the cooled tantalum collector is shown in Fig. 9.
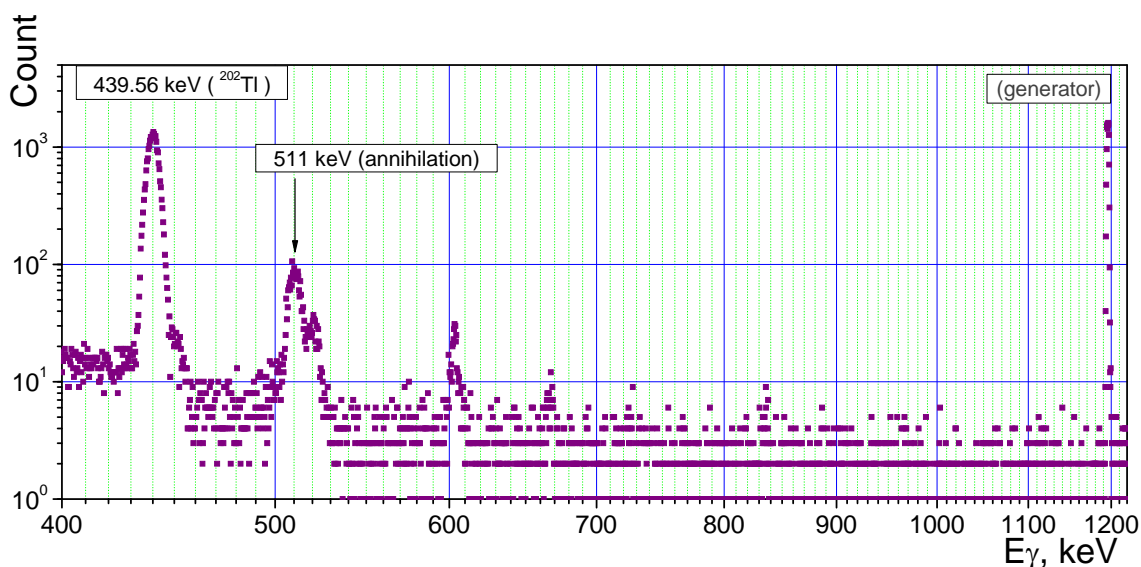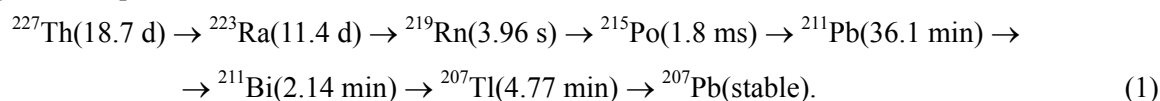

Fig. 9. Gamma-spectrum of the species evaporated at 400°C from the lead sample and condensed on a cooled tantalum collector. The time of evaporation was 2 hours

Comparing the gamma-spectra in Figs. 8 and 9, we see that the selective evaporation of thallium from the melted lead target is very fast. In two hours of heating, about 80 % of ^{202}Tl escaped from the target and was absorbed by the collector. In this case, the situation is very different from the case of strontium extraction, where the target material (niobium foils or yttrium carbide for RIC-80) had to be heated up to 2000 °C.

4. Production of alpha decaying radium isotopes from a uranium carbide target

Radionuclides decaying by alpha particle emission can be a very effective tool for therapy of malignant tumors at very early stages of their formation. The main advantage of alpha particles (the so called "alpha-knife") is their very short range (60–80 µm) in biological tissue, which is very important for treatment of small malignant formations. The action of radionuclides emitting alpha particles is very effective, as it is very local and does not destroy the surrounding tissues. Among the alpha decaying radionuclides that can be used for therapy, there are two isotopes – ^{223}Ra ($T_{1/2} = 11.4$ d) and ^{224}Ra ($T_{1/2} = 3.66$ d) – that can be effectively produced by proton irradiation of uranium or thorium targets. The cross section for ^{227}Th production from a thorium target [5] is presented in Fig. 10.

This nuclide has a rather high value of the cross section for production in the spallation reaction for the case of a natural ^{232}Th (100 %) target and the proton beam with the energy close to 80 MeV. The subsequent alpha decay of ^{227}Th produces ^{223}Ra:

$$^{227}\text{Th}(18.7 \text{ d}) \rightarrow {}^{223}\text{Ra}(11.4 \text{ d}) \rightarrow {}^{219}\text{Rn}(3.96 \text{ s}) \rightarrow {}^{215}\text{Po}(1.8 \text{ ms}) \rightarrow {}^{211}\text{Pb}(36.1 \text{ min}) \rightarrow$$

$$\rightarrow {}^{211}\text{Bi}(2.14 \text{ min}) \rightarrow {}^{207}\text{Tl}(4.77 \text{ min}) \rightarrow {}^{207}\text{Pb}(\text{stable}). \tag{1}$$

In the decay chain presented above, the decay of the selected ^{223}Ra produces three alpha particles and considerably increases the efficiency of therapy.
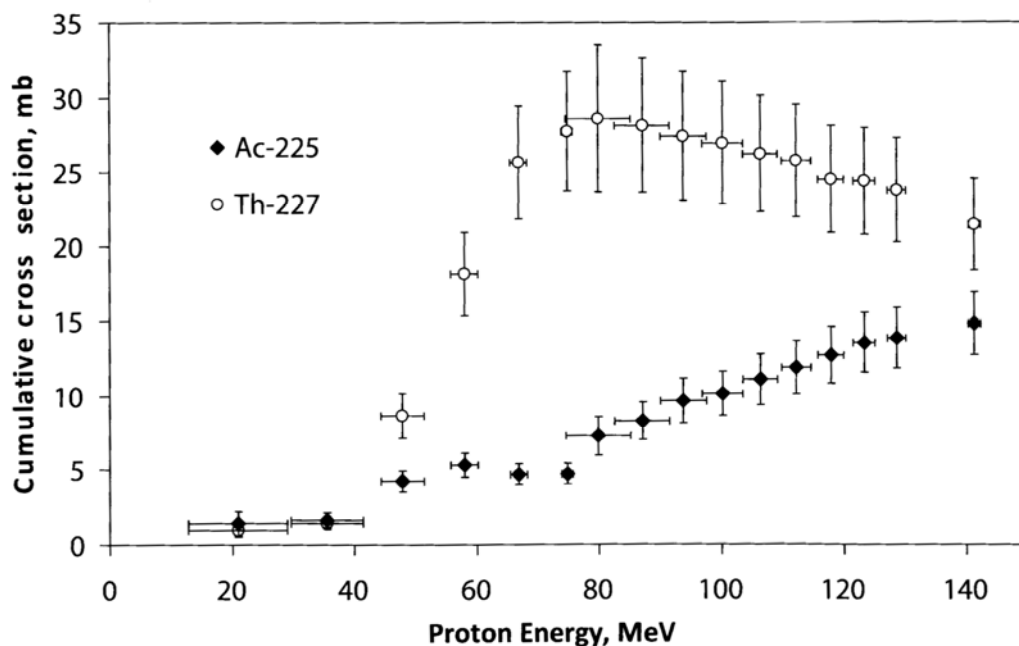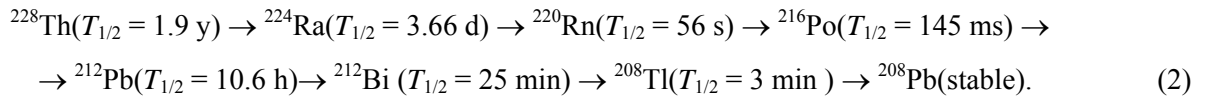
Fig. 10. Cross sections for ^{227}Th production in a thorium target *vs* the energy of protons. The open circles show the cross section for ^{225}Ac production

In our experiments, we used a ^{238}U target (uranium monocarbide of high density [6]) for production and high temperature extraction of radium isotopes. The Uranium Carbide (UC) target with the density of 11 g/cm^3 was irradiated by the proton beam with the intensity of 0.1 μA for 24 hours at the PNPI synchrocyclotron. The target mass was about 1 g. After two months of radiation cooling, the target was placed into the heating tungsten container on the vacuum test bench. The radium isotope extraction was carried out at the temperature of about 2400 °C, which is close to the melting point of the uranium monocarbide target material (2500 °C). The alpha-spectrum of the species evaporated at 2400 °C from the UC target and deposited on a cooled tantalum collector is shown in Fig. 11.
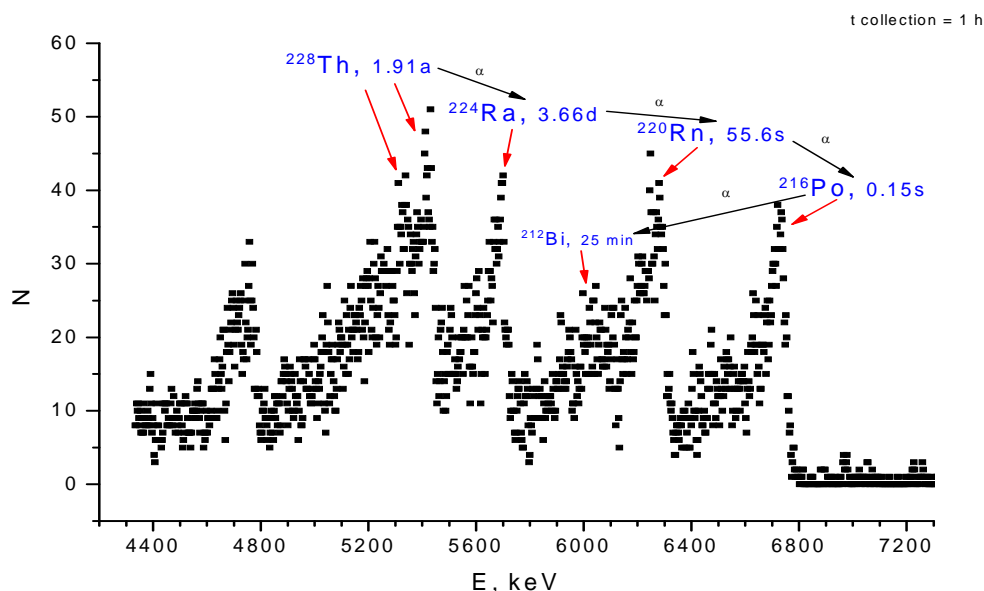
Fig. 11. Alpha-spectrum of species evaporated at 2400 °C from the UC target and deposited on a cooled tantalum collector

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As the irradiated target was cooled down during a rather long period, we detected the alpha particles emitted only by the long-lived ^{228}Th ($T_{1/2}$ = 1.9 y). In Fig. 11, we can see the chain of alpha lines after the decay of the extracted ^{228}Th:

$$^{228}\text{Th}(T_{1/2} = 1.9 \text{ y}) \rightarrow {}^{224}\text{Ra}(T_{1/2} = 3.66 \text{ d}) \rightarrow {}^{220}\text{Rn}(T_{1/2} = 56 \text{ s}) \rightarrow {}^{216}\text{Po}(T_{1/2} = 145 \text{ ms}) \rightarrow$$

$$\rightarrow {}^{212}\text{Pb}(T_{1/2} = 10.6 \text{ h}) \rightarrow {}^{212}\text{Bi }(T_{1/2} = 25 \text{ min}) \rightarrow {}^{208}\text{Tl}(T_{1/2} = 3 \text{ min }) \rightarrow {}^{208}\text{Pb(stable).} \qquad (2)$$

The experiment on ^{224}Ra production demonstrated the principal possibility to obtain alpha emitting radium radionuclides from high density UC targets. There is also a possibility to increase the yields of Ra isotopes by more than an order of magnitude with a ThC target instead of the UC target. The technology of ThC targets has already been developed. Next year, we plan to test this target material for production of radium and other alpha emitting isotopes for medicine.

5. Conclusion

This work can be considered as the first stage of developments of targets for production of several radionuclides from a large set of isotopes for nuclear medicine planned to be produced at the RIC-80 facility. In our tests, several kinds of target materials were explored. The tested target materials will be used for manufacturing of real target prototypes for the PNPI radioisotope complex. All these target materials can be also used to construct the mass-separator targets for production of radioisotopic medical beams of high purity at RIC-80. The following stage will be the construction of a target unit prototype with the amount of the target material of 5–10 g for medical radionuclide production at the RIC-80 complex.

References

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