THE TARGET DEVELOPMENT FOR MEDICAL RADIONUCLIDES ⁶⁷CU AND ⁸²SR PRODUCTION

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Abstract

The RIC-80 (Radioactive Isotopes at cyclotron C-80) radioisotope complex which is constructed at the beam of cyclotron C-80 at the Petersburg Nuclear Physics Institute for the production of a wide spectrum of medical radionuclides for diagnostics and therapy has been discussed. The results of a new method utilization for the target development for the production of generator PET radioisotope ⁸²Sr and radionuclide ⁶⁴Cu are presented.

INTRODUCTION

The production of radionuclides that decay with emission of positrons, allowing their use for PET (Positron Emission Tomography), is very important for diagnostics of different diseases. The nuclear physics experimental methods, combined with very sensitive detectors of nuclear radiation, give a very good possibility for modern medicine in diagnostics and therapies.

In this paper the first results on the development of a new method of a high temperature separation of radioisotopes from different kind of target materials are presented, which has been worked out in the Petersburg Nuclear Physics Institute.

THE RIC-80 FACILITY

The proton beam energy of the C-80 [1] can be varied in the interval 40-80 MeV. The proton beam intensity will be up to 200 µA. This cyclotron is intended mainly for the production of a wide spectrum of medical radionuclides for diagnostics and therapy. A photograph of the C-80 cyclotron with three proton beam lines to the target stations is presented in fig.1. The RIC-80 radioisotope complex [2,3] is being constructed in the cellar of the experimental hall of the PNPI synchrocyclotron. The proton beam line is directed from the ground floor to the cellar where it can be deflected and focused to one of three target stations. The mass-separator with its target station [3] will allow for the production of separated medical radionuclides of a high purity, which will be implanted into corresponding collectors from which they can be easily extracted. The target stations will be equipped with special devices to transfer the highly radioactive targets into protection containers so that they can be transported safely to special storage places, or to hot cells for the after-treatment and corresponding preparations for pharmaceutics. The proton cyclotron C-80 gives a possibility to obtain sources of a high activity practically for the whole list of radionuclides produced at accelerators.



Figure 1: Cyclotron C-80 (ground floor) with three proton beam lines to the target stations (cellar).

These are ^{64,67}Cu, ⁶⁸Ge, ⁸²Sr, ¹¹¹In, ^{123,124}I, ^{223,224}Ra and others, which are at present under discussion in corresponding publications as perspectives for diagnostics and therapy. The mass-separator method [3] will give the possibility of the production of very pure beams of some radioisotopes. As the first steps they can be ⁸¹Rb, ⁸²Sr, ¹¹¹In, ^{223,224}Ra, as radionuclides with respectively low ionization potentials, which can be produced by a mass-separator method with a high efficiency.

Experiment Description and Experimental Results of ⁸²sr Production and Extraction From RbCl Target Material

In the experimental tests for the production of ⁸²Sr the powder of RbCl was used as a target material. Radionuclide ⁸²Sr with a half-live $T_{1/2} = 25.55$ days is a generator for its daughter isotope ⁸²Rb ($T_{1/2} = 1.25$ min) which is widely used in PET diagnostics. For separation of the target material and produced strontium isotopes a new developed, high temperature method was utilized [4]. After irradiation by the 1 GeV proton beam at the PNPI synchrocyclotron RbCl powder was placed into a vessel manufactured from stainless steel which was put into TaW oven heated by the direct current. In a high vacuum the powder was heated slowly to the temperature up to 900 °C to be evaporated to a separated volume specially constructed to minimize the waste of irradiated material in the process of its evaporation. At that temperature the process of complete evaporation of the target material of one gram mass took about one hour. For the evaporation process control the γ -spectrum of the vessel with irradiated RbCl was measured before and after each stage of the heating process. Additionally after each heating the vessel was weighted for the evaporated material mass control. In fig. 2 a), b) a part of gamma-spectra of the irradiated sample of rubidium chlorine is presented. They were measured with a high purity germanium detector. The gamma-line of the energy 552 keV belongs to the decay of ⁸³Rb with half-life 86.2 days and its decreasing indicates the efficiency of the target material evaporation. The gamma-line of the energy 776 keV belongs to the decay of 82Rb with half-life 1.27 min, which is the daughter isotope of ⁸²Sr and its decreasing indicates the strontium radionuclide evaporation. In Fig. 2 by squares the spectrum of the vessel with the irradiated RbCl before the heating is shown. The spectrum after one hour vessel heating at a temperature 500 °C is shown by circles. In Fig. 3 comparison of the spectra after the heating at a temperature 500 °C (circles) and at a temperature 900 °C (triangles) are shown.

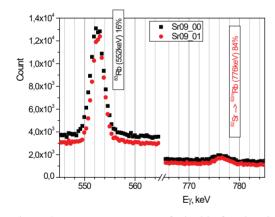


Figure 2: Gamma spectrum of RbCl before heating.

As one can see from Fig. 2, the heating of the irradiated sample at a temperature 500 °C for one hour does not give any effect on the target material evaporation. The same result was obtained by the sample weighting before and after its heating at 500 °C. At the same time Fig. 3 shows, if the vessel with the RbCl is heated up to 900 °C, the target material has been evaporated completely with almost hundred percent conservation of strontium. The fact of complete evaporation of the irradiated target material has been confirmed by the sample weighting before and after its heating at 900 °C. Finally conserved radioactive Sr atoms can be evaporated from the vessel at higher temperature, or washed by a small amount of an acid solution.

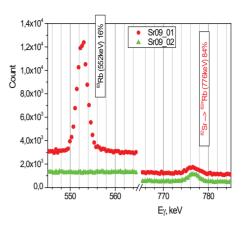


Figure 3: Gamma spectrum after 1 hour heating.

Therefore, as one can see from Fig. 2, 3 for the separation of strontium isotopes, should be some stages of the evaporation of the target material and produced species in the process of the target heating in a high vacuum at different temperatures. To separate strontium from the rubidium chloride target, the target heating was started at a low temperature (500 - 900) °C to evaporate the target material RbCl which has considerably lower boiling point, than strontium. After that, strontium was selectively extracted by washing of internal vessel volume by the HCl solution. Another way of strontium extraction was the niobium or tantalum vessel use with the heating it up to 1700 °C after the target material evaporation at 900 °C. The evaporated strontium atoms were directed to the collector cooled by floating water. The experiments carried out gave the efficiency of the target material separation better than 99,9%. The efficiency of the strontium radionuclide extraction was about 95%.

Experiment Description and Experimental Results of ⁶⁷Cu Production and Extraction from Zn Target Material

In the experimental tests for the production of ⁶⁷Cu natural metallic Zn was used as a target material. Radionuclide ⁶⁷Cu with a half-live 2.57 days is considered as a very perspective radioisotope for therapy of some kinds of malignant tumours. For separation of the target material and produced ⁶⁷Cu radionuclide a new so called "dry", high temperature method, similar to the described method of strontium isotope extraction [4] was utilized. After irradiation by the 1 GeV proton beam at the PNPI synchrocyclotron metallic zinc was placed into a vessel manufactured from tantalum, which was put into Ta-W oven heated by the direct current. In a high vacuum irradiated zinc was heated slowly up to the temperature 700 °C to be evaporated to a separated volume specially constructed to minimize the waste of irradiated material in the process of its evaporation. At that temperature the process of complete evaporation of the target material of one gram mass took about one hour. For the evaporation

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process control the γ -spectrum of the vessel with irradiated zinc was measured before and after the heating process. Additionally after the heating the vessel was weighted for the evaporated material mass control. In Fig. 4, 5 a part of gamma-spectra of the irradiated sample of zinc is presented. The gamma-line of the energy 1115 keV belongs to the decay of 65Zn with half-life 244.3 days and its disappearance indicates the efficiency of the target material evaporation. The fact of complete evaporation of the irradiated zinc material has been confirmed by the sample weighting before and after its heating at 700 °C. The gamma-line of the energy185 keV belongs to the decay of ⁶⁷Cu (T_{1/2}=2.57 days), which is the produced required radioisotope. In Fig. 4 by squares the spectrum of the vessel with the irradiated Zn before the heating is shown. The spectrum after one hour vessel heating at a temperature 700 °C is shown by circles.

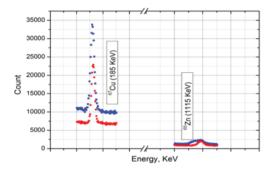


Figure 4: Gamma spectrum of the vessel before and after heating at 700 °C.

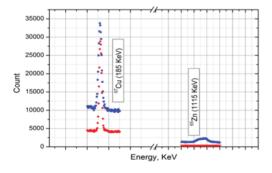


Figure 5: Gamma spectrum before and after heating at 1460 °C.

As one can see, after the heating the target material has been completely evaporated, that was confirmed by the weighting the vessel before and after heating. At the same time radioactive atoms of cooper, having considerably higher boiling point (2562 °C), than the target material zinc (907 °C), remained in the vessel. Also the presence of the gamma-line of the energy1121 keV of ⁴⁶Sc (T_{1/2}=83.8 days) at the spectrum measured after the target material evaporation demonstrates, that atoms of scandium, which is a rather hard volatile element (boiling point 2830 °C) does not evaporate from the target vessel at a temperature of 700 °C. In Fig. 5 in lower part the spectrum of evaporated cooper atoms collected at the cold finger cooled by floating water after the vessel heating at a temperature 1460 °C in two hours (circles) is presented. For comparison by squares the spectrum of the vessel with the irradiated Zn before the heating is shown. Therefore, as one can see from Fig. 4, 5 for the separation of cooper radionuclides and zinc target material should be two stages: the first one is a slow evaporation of the target material at a temperature about 700 °C; the second one is the evaporation of produced cooper species in the target heating process at a temperature 1460 °C.

The first experiments carried out gave the efficiency of the target material separation better than 99%. The efficiency of the cooper radionuclide extraction and collection was about $(90 \pm 15)\%$.

SUMMARY

At PNPI a high current cyclotron C-80 with the energy of extracted proton beam of 40-80 MeV and the current up to 200 µA will be put into operation at the end of 2016 - beginning of 2017. One of the main goals of C-80 is production of a large number of medical radio nuclides for diagnostics and therapy. At present time the construction of radioisotope complex RIC-80 at the beam of C-80 is carried out. The peculiarity of the proposed radioisotope facility is the use of the mass-separator with the target-ion source device as one of the target stations for on-line, or semi on-line production of a high purity separated radio isotopes. The important part of the work was devoted to the target and ion source developments for the new project RIC-80. The tested target materials and developed ion sources will be used for manufacture real target prototypes for PNPI radioisotope complex. R@D of new high temperature methods of separation of produced radionuclides ⁸²Sr and ⁶⁷Cu from rubidium and zinc irradiated targets has been carried out. The following stage of the work will be the target unit prototype construction with the amount of the target material 40-60 grams, which is required for the effective medical radionuclide production at the RIC-80.

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