DEVELOPMENT OF CYCLOTRON BASED HIGH BEAM CURRENT TECHNIQUE FOR Ga-67 PRODUCTION.

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Abstract

For the effective production of radioisotopes at the Almaty cyclotron using high beam intensity a new internal target was designed. In particular it is used for production of Ga-67. An additional feature of the developed technique is using of trioctalphosphate oxide (TOPO) as extractant. This feature allows to separate gallium from macro-quantities of zinc from 12 M HCl in one stage. Yield of gallium-67 in a final product reaches 98%, thus the high degree of clearing from copper and zinc radioisotopes is achieved in comparison with traditional extraction method (extraction by diethyl, diisopropyl ethers, sulfo-oxidies). The technology of Ga-67 production is simple in hardware and does not need use of expensive reagents. Obtained results are discussed.

1 INTRODUCTION

Radiopharmaceutical (RPC) gallium-67 citrate is widely used for cancer diagnostics. There are sufficient practical experience and correspondent clinical conditions for use of medical pharmaceuticals radioisotope (RI) Ga-67 in Kazakhstan. Up to now foreign preparations were used for these purposes. A long term program of radioisotopes production for different purposes, including medical RPC, was developed in the Institute of Nuclear Physics (INP). During recent years the investigations for Ga-67 production and development of RPC gallium-67-citrate technology were carried out.

The main basis for medical RI production in the Republic are the variable energy isochronous cyclotron $K=50z^2/A$ MeV and radiochemical laboratory supplied with necessary equipment and having modern technologies for radioisotopes production including their content analysis. The cyclotron gives an opportunity to obtain light ion beams in broad range of energy: protons 6÷30 MeV, deuterons 12÷25 MeV, α-particles 25÷50 MeV and helium-3 ions 18÷62 MeV. Beam's maximal intensity on the external target is 15÷30 μA for different cyclotron operation modes [1]. Irradiated materials' radiochemical processing is carried on in a special building equipped with a chain of "hot cells" and provides the work with radioactivity up to 10 Ci by Co-60. There are all necessary conditions for operation including nuclear waste utilization.

2 INTERNAL CYCLOTRON TARGET

To increase sufficiently the efficiency of radioactive isotopes production, the problem of high intensity internal cyclotron beam mainly was solved. The designing of internal target, sustaining high thermal loading during irradiation, is the basis for this problem solution. The scope of the work for the target manufacturing included: theoretical and experimental determination of beam's parameters on the final radii, target's design development, its manufacturing and adjustment in operation conditions (determination of angle of target inclination to beam trajectory and size of irradiated area). During the experiments on the target model the characteristic form of beam intensity distribution across the target surface was determined.

The obtained data were used for thermal calculations and irradiation conditions determination. On this basis the main parameters of the target design were determined:

- Optimal backing thickness and the form of its cooled surface,
- The cross section of cooling channel and water pressure determining the noted target's surface temperature,
- Maximal integral current value for noted accelerated ions energy.

In the result of the carried out works the target system was manufactured, with remote target mounting into cyclotron tank and automatic target's head disconnection from the rode in sluice chamber. The general target's irradiated area was 8 cm^2. The target reliably sustained cooling water pressure up to 20 bar, providing the use of internal proton beam with energy 30 MeV and beam current up to 300 μA. The target's head and details of its design are shown in Fig.1.

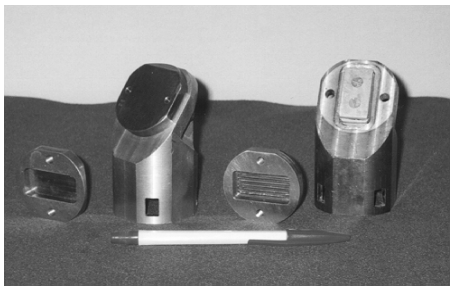

Figure 1: Head of the target.

In operation mode the thermal power of beam for zinc targets irradiation was 3.7÷4.5 kW for proton beam's current value 100÷150 μA. Maximal Ga-67 yield was obtained by optimization of the whole production process, and the choice of energy value and the type of accelerated ions, determining cross section of nuclear reactions, were very important. The information about nuclear reaction cross sections and Ga-67 yield is presented in [2,3,4]. It comes out that for Ga-67 efficient production the target must be made from Zn-68 or zinc natural isotope content. Usually deuterons or protons were used as bombarding particles. In this case 28 MeV proton beam and the target containing zinc of natural isotope content were used. Electrolyte content and optimal mode of zinc electrolytic extraction on the copper plate (cathode) were adjusted experimentally: cathode current density was 22-23 mA/cm^2, for 20-30°C. The thick zinc layer was obtained with plating velocity 38-40 μm/hour. The dissolving zinc anodes were shielded by paper filters to reduce zinc precipitates contamination. For zinc better adhesion on copper, it was preliminary plated from sulfur electrolyte by nickel layer with the thickness 8-10 μm and then zinc of necessary thickness was electroplated on nickel film. The manufactured zinc targets had thickness 150÷250 μm.

3 TECHNOLOGY OF GA-67 PRODUCTION

Adjustment of radiochemical technology of Ga-67 production was carried out on the base of extraction-chromatography method which was advantageous as being compared with other ones.

Chromatography column extraction method was used. The inert support was polychrome with fraction dimensions 0.5÷1 mm. The column preparation was carried out in accordance with usual method of stationary phase plating on the support. Extragent TOPO (0.2 M solution in toluene) was taken to the column (length of the column – 13.5 cm, diameter – 8 mm) as much as 2 ml.

The optimal mode of gallium separation from zinc on TOPO was determined on stable gallium, zinc and copper model solution, as well as with the help of plated Zn-65 and Ga-67 tracers. Gallium elution was carried out by 0.5 M HCl solution. Filtrates, rinsing water and elutes were analyzed on gallium, zinc and copper by ICP method, and for introduced tracers on gamma-spectrometer. The data of spectrometry and ICP analysis had shown that in the concentration range 8 – 12 M of HCl up to 99% of gallium was extracted on TOPO. The dependence of Ga-67 yield during elution by 0.5 M HCl from the volume is presented in Fig.2.

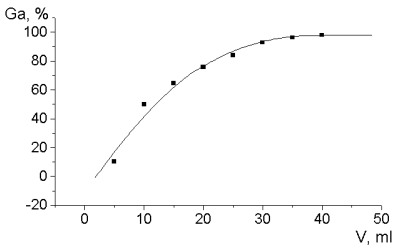

Figure 2: Ga-67 yield versus elute volume.

The data of elutes analysis on zinc and copper had shown that good degree of zinc and copper purification for TOPO application for gallium extraction was obtained when 12 M HCl was used and it was $3 \cdot 10^5$ and $7 \cdot 10^3$ correspondingly. Experiments for Ga-67 production and its extraction from irradiated target were carried out in the condition close to regular production. Zinc target with thickness 200 μm was irradiated by 28 MeV and 50 μA proton beam during 7 hours. After the target dissolution in 55 ml 12 M HCl radionuclides content of induced radioactivity was examined and the data are presented in Table 1. The technological process of gallium extraction from the solution was carried out in accordance with the developed scheme: transmission through the chromatography column, filled with polychrome coated by TOPO (0.2 M solution in toluene), column rinsing by 30 ml 12 M HCl and subsequent gallium elution from the column with 70 ml 0.5 M HCl. Analysis of the final product on radionuclides content had shown the absence of zinc and copper impurities. Gallium yield for the described technology was 97%.

Gallium-67-citrate preparation was produced in the Republic of Kazakhstan for the first time and its quality corresponds to similar preparation of international standard.

Acknowledgements.

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Table 1. Radionuclide content of irradiated target.

Nuclide	Cu-61	Zn-62	Zn-63	Zn-65	Zn-69	Ga-66	Ga-67	Ga-68
Half-life time	3.4 h	9.3 h	38.1 m	244 d	13.6 h	9.5 h	3.26 d	1.13 h
Activity (mCi)	373	34	31 500	5.1	5.4	743	282.5	2 141