RADIOISOTOPE PRODUCTION AT THE ISOCRONOUS CYCLOTRON IN ALMATY.

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The main results of radioisotope production at the Kazakstan Isochronous Cyclotron (k=50MeV) are presented. Th features of target irradiation and design of internal target are considered. In INP methods for radiopharmaceutical «Tl-201 - chloride» production in small quantities for using in nuclear medicine were developed. At the time being the methods for Co-57, Ga-67, Cd-109 and Pu-237 production are being developed.

1. Introduction.

For the long time cyclotrons have been and continue to be used for radioactive isotope production. The use of cyclotrons for this purpose has significantly increased over past two decades, and nuclear medicine is probably the most important application for radioisotopes because of very important and unique medicine information that can be obtained. Initially in the 1930-s radioisotopes were produced as by-product on cyclotrons designed and built specifically for nuclear science research. Later, beginning from 1960-s special commercial cyclotrons were built. The milestones in this two-three decades process were: «classical» isochronous cyclotrons, positive ion, internal high current ion source, internal targetry, rather high radiation dose to personnel and radiation damage of cyclotron components; then negative ion, internal source, external targetry, moderate vacuum and less radiation damage; and at the latest stage negative ion, external source, external targetry, high vacuum system, the «deep valley» magnet design, high efficiency of the cyclotron, comparatively less radiation damage and radiation dose to personnel [1,2].

At the time being leading radiopharmaceutical companies use one or even more of the latest «third generation», like IBA-Cyclone 30 or EBCO TR 30 cyclotrons which operate at the energies of protons up to 30 MeV, and with target beam intensities at the level of 500-1000 μA. The future tendency is obvious towards using such type of cyclotrons.

2. Cyclotron beam for internal target irradiation.

In spite of all obvious high quality of such cyclotrons, they require rather essential initial investments, and in some cases using of old type-cyclotrons for radioisotope production is justified.

In the Institute of Nuclear Physics (INP) in Almaty [3] the work is undertaken the objective of which is to develop the more or less regular production of radioactive isotopes to meet the needs of the Republic of Kazakstan and their delivery in the Republic and in the nearby regions.

At the same time it is clearly understood that trying to combine high beam intensity isotope production with other applications (Nuclear Physics, Material science research etc.) leads to the difficulties in reliable running cyclotron.

One of the main problem of isotope target irradiation by high current ion beam is to design and manufacture high thermal capacity target capable of sustaining rather high power density at the level of 1 $kW \cdot cm^{-2}$. As a rule internal cyclotron beam has high luminance and compact space structure. Usually surface of the target is inclined to the beam direction at the angle of $6\text{-}12^0$. Sometimes magnetic field of cyclotron is distorted in order to increase beam spot on the target. In our case for Tl-201 production limitation on axial focusing at 30 MeV was used. For this mode of operation axial oscillation amplitude is increased approximately three times. Experimentally obtained ion beam spots for different energies and particles are shown on fig.1.

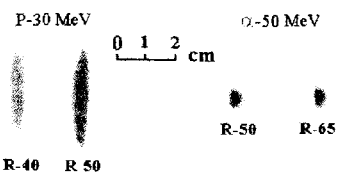

Fig.1 Beam spots for different particles and energies.

3. Beam density distribution on internal target.

The next problem to be solved while using internal target is connected in our case with RF system. It consists of two 180^0 dees and causes high level noise much more higher than beam signal.

the vacuum tank is shown on fig.2 (1-dee, 2-target, 3-target holder).

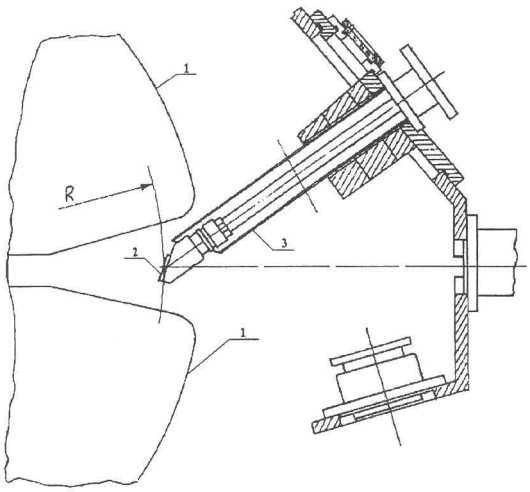

Fig. 2. Schematic view of the target position in the cyclotron tank.

The layout of the internal target irradiation is shown on fig.3.

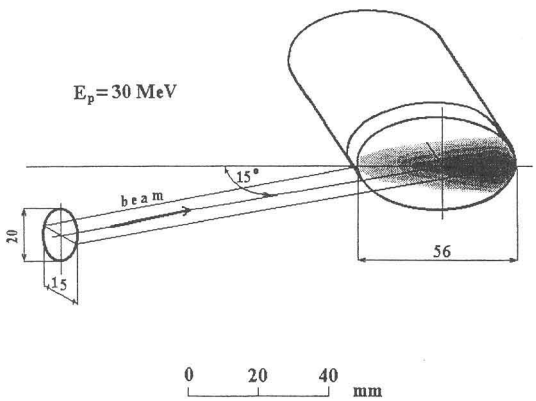

Fig.3. Layout of internal target irradiation

The chosen angle of the target's inclination with respect to beam final orbit trajectory makes possible to use target's area equal to S=10 cm^2. Current density distribution across isotope target surface is shown on fig.4.

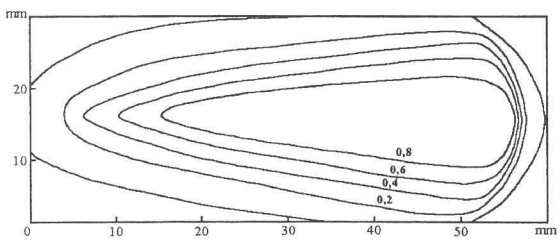

Fig.4. Current density distribution across isotope target surface.

As it is follows from the presented plots there is sufficiently distinct plateau in the current density distribution: 80% of the beam's intensity is account for 50% of irradiated surface of the target.

4. Technology of irradiated target treatment for radioisotopes extraction

Radioactive isotopes (RI) extraction from irradiated targets and available isotopes production is carried on in special radiochemical laboratory equipped for the work with open radioactive samples activity up to 10 Ci for Co-60.

Technology and production of radiopharmaceutical (RPC) thallium-201-chloride applied in clinical cardiology was developed and launched. Table 1 presents the main characteristics of produced RPC TL-201-chloride.

To reduce the manufacturing cost of RPC an efficient technology was developed for recovering of stable thallium concentrated by thallium-203 - substance used for RPC production. Thallium-203 recovering is carried out from liquid radioactive wastes obtained during pharmaceutical production. Initial (for recovering) sulfate solution contains up to 30 mg/ml of thallium-203.

It must be noted that along with RPC Tl-201-chloride production and its technology improvement the work is being carried on for the production of radionuclides applied in radioecology

In particular, the technique for the production of radionuclide Plutonium-236,-237 and -238 was developed. They were produced on the cyclotron of INP NNC RK in reactions on α-particles with energy 50 MeV. As a target samples with UO_2 enriched by isotope U-235 (3,3% and 2,4%) were used. Material for the target with the thickness 0,5 mm was mounted in demountable holder of target device. The target was covered by copper foil (thickness 40 μm). Energy of α-particles, bombarding the target, was 46,5 MeV. Irradiation was carried on by current 10 μA; time of irradiation 168 hours. During the irradiation of the target containing U-235, in reactions (α,n), (α,2n), (α,3n) radionuclides of plutonium with A=238, 237, 236 correspondingly were formed. The activity of produced radionuclides was: Pu-237-150 kBq, Pu-236+Pu-238-10 kBq.

Plutonium fraction was extracted from irradiated targets by the method of extraction chromatography. Trioctylamine (TOA) was used as extragent and teflon crumb as inert support. Trioctylamine is the selective extragent for plutonium, it extracts anion complexes of four-valent plutonium from nitrate solution.. For plutonium stabilization in the form of Pu(IV) it is necessary for the first its reduction down to Pu(III) by ascorbic acid and then oxidization of Pu(III) down to Pu(IV) before sorption with the help of potassium nitrite. Sorbent was prepared by deposition onto the inert support of extragent of the quantity 1÷3% to support material weight.

Table 1. The main characteristics of RPC Tl-201-chloride, produced by Institute of Nuclear Physics NNC RK.

Properties	Quantity estimation
Volume activity for the date of certification	not less than 55MBq/ml
Content of natrium chloride	8.0÷10.0 mg/ml
pH value	5.0÷9.0
Radionuclide purity for the date of certification:	
Tl-201	≥98.2
Tl-202	≤0.2
Tl-200	≤1.5
Pb-201	≤0.05
Pb-203	≤0.05
Nonactive impurities, μg/ml:	
Tl	≤2.0
Fe	≤1.0
Cu	≤0.1
Term of validity from the date of production, in days	10

Irradiated target was dissolved under heating in 30 ml of concentrated nitric acid. Solution was dried up and the dry residual was dissolved in 3M of nitric acid. The following technique of plutonium extraction from the resulting solution was as follows: to the solution heated up to 60^{0}C ascorbic acid was added - 10 mg per 1 ml and exposed for 15 min, and then gradually potassium nitrite was added-30 mg per 1 ml of solution and was blown by air during 10÷15 min. The resulting solution passed through prepared chromatography column with velocity 1,5÷2 ml/min (1,5 g of sorbent placed in stuffing part of the column having diameter 6÷7 mm and height 80 mm). Then the column is rinsed by three portions of 3 M HNO_3 5÷6 ml each. Plutonium was eluted by 7 ml 3% $(NH_4)_2C_2O_4$ in 1,5 M solution of nitric acid.

Concentration of α-isotopes in elution was determined by α-spectrometry.

Carried on investigations had shown that Pu-237 radionuclide yield in reaction ^{237}U(α,2n) ^{237}Pu for α-particles energy 46,5 MeV was 90 Bq/(μA·h), which coincides with the calculated value.

The total yield of Pu-236 and Pu-238 in our experiments was 6 Bq/(μA·h). Keeping in mind the fact that these radionuclides are formed on U-235 (^{235}U(α,n) ^{238}Pu) as well as on U-238 (^{238}U(α,p3n) ^{238}Np→^{238}Pu), experimental date are in agreement with calculated ones.

The developed technique of plutonium radionuclides extraction from UO_2 target irradiated by α-particles provides 95% yield of necessary radionuclides. Produced plutonium sources were applied as radioactive tracers in radiochemical processes for plutonium detection analysis in the soils of Semipalatinsk Nuclear Test Site.

The certain amount of ^{88}Y which is used as tracer during radiochemical procedure for ^{90}Y separation from the environmental objects was produced.

In some cases while using external beam irradiation the combined targets were used, what is not possible in the case of large-scale commercial isotope production. This technique was used for simultaneous production of Tl-201 and Ga-67. Combined targets were consisted of Tl-203 and natural Zn separated by thin copper foil. In spite of inefficient energy range for high quality Ga-67 production it is possible to minimize Ga-66 content to acceptable level by time exposition (half life for gallium-66 is 8 times lower than that for gallium-67)

Main stages of gallium radiochemical extraction from zinc target irradiated by protons: target's dissolvation, gallium extraction by butyleacetate, reextraction and evaporation were developed and implemented.

Usually in one batch for 150 mCi of Tl-201 the activity of Ga-67 in final product was 30 mCi. The quality of obtained Tl-201 and Ga-67 are in correspondence with required parameters.

Necessary quantities of Co-57 for Mössbauer sources production and Cd-109 for X-ray spectroscopy are regularly produced as well for internal use in Kazakstan.

Work for Ag, Ni, Zn and Tl targets manufacturing and testing in the conditions of internal irradiation with current on the target 100÷150 μA was began. First experimental results create the base for positive conclusion about INP NNC RK isochronous cyclotron fitness for semicommercial production of such isotopes as Co-57, Cd-109, RPC Tl-201-chloride and RPC Ga-67-citrate. Thus the isochronous cyclotron in Almaty is used not only for commercial medicine radioisotopes production but also for production of different radionuclides used in the Republic of Kazakstan in various fields of technology, science, environment research etc.

Acknowledgments

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