RADIOISOTOPES PRODUCTION AT THE CYCLOTRON IN ALMATY

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ABSTRACT

Kazakhstan variable energy isochronous cyclotron (K = 50 MeV) is a universal facility used both for basic nuclear physics research and applied research in different adjoining fields including material science, elemental analysis, ecology etc. The details of the cyclotron were previously published so only the basic parameters are mentioned [1]. It is 150 cm compact-pole 3 sector, positive ion machine designed to operate in first and third harmonic mode. In first harmonic protons can be accelerated in the range from 6 MeV to 30 MeV, deuterons – from 12,5 MeV to 25 MeV, alpha particles – from 25 MeV to 50 MeV, helium-3 ions – from 18 MeV to 62 MeV. Central magnetic field is changed from 0,6 T to 1,6 T. The accelerating RF system consists of two 180° dees, and operates at voltage up to 80 kV in the frequency range 8,5 – 18,5 MHz. Beam of accelerated ions is extracted with electrostatic deflector with non-uniform electric field. Now one of the main applications of the cyclotron is radioisotope production to meet the needs of the Republic of Kazakhstan. At the time being the main users of radioisotope products are Institutions of Healthcare Ministry and also enterprises of oil-chemistry, metallurgy, mining, scientific institutes etc. The isochronous cyclotron allows to produce high yield of radioisotopes in the reactions of (p,n), (p,2n), (p,3n) type using both external and internal target irradiation. Protons energy of up to 30 MeV allows to produce up to 80% of all radioisotopes, produced at cyclotrons. In this submission a survey on radionuclides production including Tl-201, Co-57, Cd-109, Ga-67, Pu-237, Y-88 is presented.

INTRODUCTION.

Basis of the experiment with using of marked atoms is opportunity to discover, identify

and measure concentration of any radioisotope by difference of types and energy of particles emitted by nuclei of isotopes during decay.

At the time being the main suppliers of radioisotopes are atomic reactors and cyclotrons. Giving up to reactors in volume of radioisotopes production, the cyclotrons essentially enlarge their numbers. Unlike the neutron abundance reactor isotopes, cyclotron isotopes are neutron deficient, emitting easily detected characteristic or positron radiation.

Cyclotron isotopes are distinguished by high specific activity and radioisotope purity, as almost all radioactive atoms of the isotope can be extracted from the target «without bearer».

For radioisotopes production both external for medical isotopes and internal targets, when high beam current is needed, are used. Irradiation of external target is typically being used during some years. Work on internal target was began recently.

Cyclotron and internal target.

One of the most important systems of the cyclotron is the magnet system, in decisive manner defining final parameters of the beam. Three sector magnetic field structure was chosen as leading to maximum axial focusing of the field. Maximum value of the third harmonic of the magnetic field was equal to 0,30.

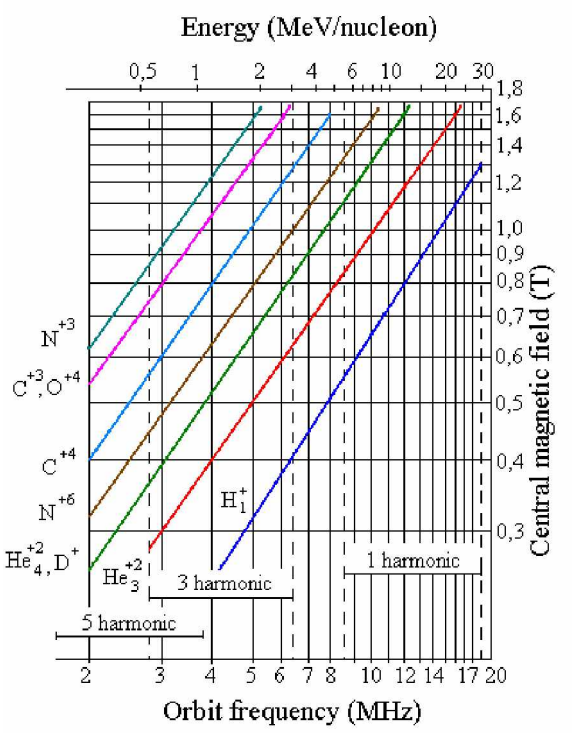


Fig.1. Energy of ions (per nucleon) as a function of magnetic field and acceleration frequency.

The intensity of the beam current on the external target, located at the distance of 25 m from the cyclotron, is equal to 30 μA for protons and deuterons, 10 μA for ions of He-3 and 20 μA for ions of He-4. Extracted beam intensity is limited by heat dissipation in the deflector septum. Energy spread is 0,6 %. Emittance of the beam for 30 MeV protons and intensity of 15 μA is equal to 16π mm·mrad for radial and axial motion. High intensities, typically several hundred μA are available on internal target. In Fig.1. energy per nucleon is shown as a function of frequency and magnetic field.

Beam currents used are limited more by the target features than by the cyclotron limits. In order to enlarge the beam spot on the target and to reduce beam power density on the target it is

inclined with respect to the beam. In the considered case this angle is equal to 15°. In the cases when 30 MeV protons are used the limitations of axial focusing are manifested in enlarging the axial beam dimensions. The frequency of axial betatron oscillations of accelerated particles is determined as

$$\nu_z^2 \approx -K + \sum \frac{A_n^2}{2}(1 + 2\psi'^2_n) \qquad (1)$$

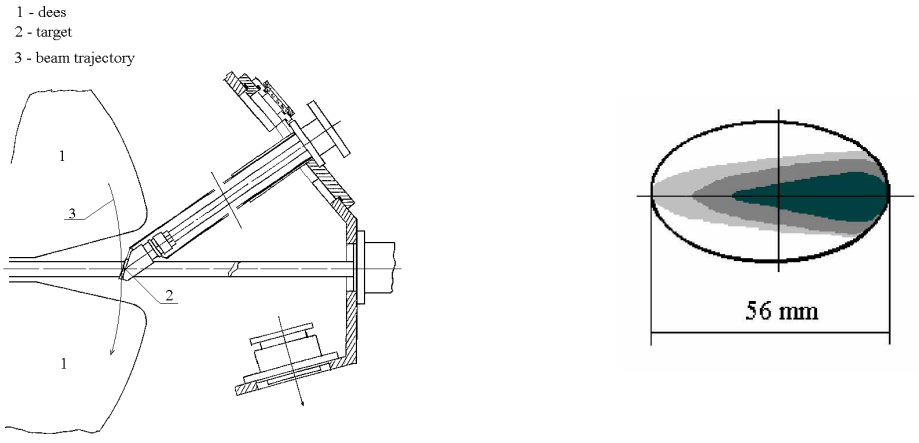
or in the case of isochronous magnetic field

$$\nu_z^2 \approx \frac{-2W}{E_0} + \sum \frac{A_n^2}{2}(1 + 2\psi'^2_n) \qquad (2),$$

here K = magnetic field index, W = energy of particle, E_0 = energy of particle mass rest, ν = frequency of betatron oscillation, A_n = amplitude of magnetic field harmonic, ψ_n = phase of magnetic field harmonic.

The limiting ability of magnetic field axial focusing is about 30 MeV and at this level of protons energy an axial dimension of beam spot increases two-three times. This feature is used to reduce essentially specific beam power density on the target at Tl-201 production. The schematic location of the internal target in the cyclotron tank is shown in Fig.2 a). 30 MeV proton beam spot on the target is shown in Fig.2 b).

Schematic view of internal target device is shown in Figure 3. There also details of target head design are shown and typical temperature distribution evaluation is presented for Ni, plated on Cu backing cooled with water under pressure of 20 bar.



a) Schematic view of the target installation in the cyclotron vacuum tank

b) Beam spot on the internal target, nclined to the beam at the 15°.

Fig.2. Schematic location of the internal target in the cyclotron tank.

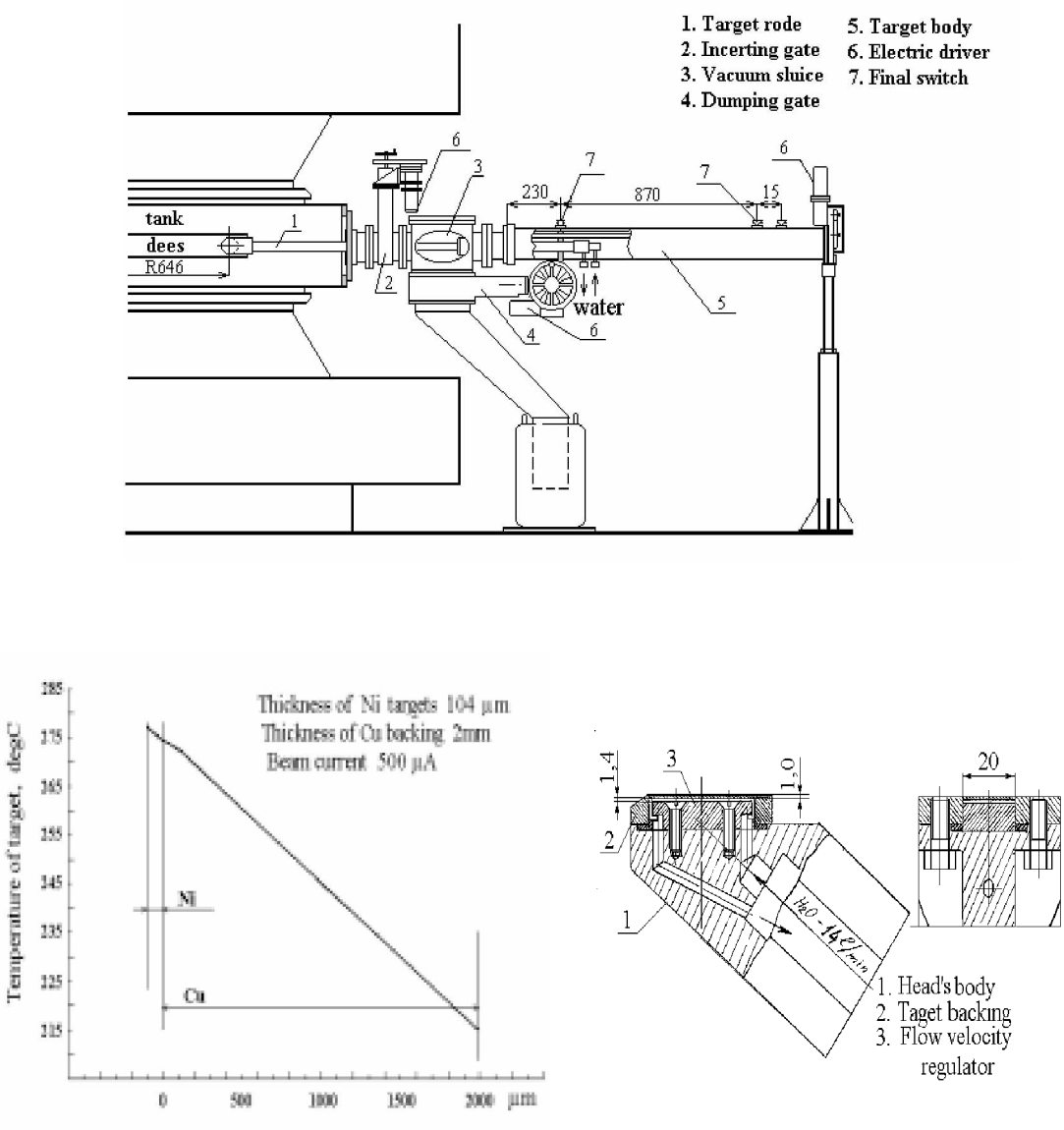


Fig. 3. Schematic view of internal target device.

Thallium-201.

One of the most important commonly used cyclotron produced isotopes is thallium-201 which is used to demonstrate blood flow within the heart muscle and reveal it's disorder. At the same time it is one of the most difficult isotopes to produce.

The production is done on thin metallic (0,5 mm) target enriched (96%) isotope Tl-203, deposited normally by electroplating on a copper backing, strongly cooled by water. The production nuclear reaction is:

Tl-203 (p,3n) Pb-201 → decay Tl-201.

For irradiation 30 MeV proton beam is used [2]. Maximum proton energy is 28,5 MeV and determined by production Pb-200 in the reaction (p,4n). Minimum proton energy is 22 MeV and is determined by production Pb-202 in the reaction (p,2n). The target is irradiated during 16 hours, after which chemical and radiochemical techniques are used for radioisotope extraction from the irradiated target. The primary obtained radioisotope Pb-201 is extracted from the Tl-203 using sulfuric acid as solvent. During the second chemical separation the second product Tl-201 is extracted from the primary product Pb-201 by the co-plating on the buthylacetate technique. Original Tl-203 is recovered from the Pb-201 and different thallium isotopes by cementation technique.

All radiochemistry processes are performed in special radiochemical laboratory equipped for the work with open radioactive samples activity up to 10 Ci for Co-60. Thallium-201 is used for production of pharmaceutical «Thallium-201-chloride» applied in cardiology. Specification of the pharmaceutical is presented in Table 1.

A typical tomographic heart image at rest and under stress using Tl-201 is shown in Fig.4.
These results were obtained in cardiology center in Almaty.

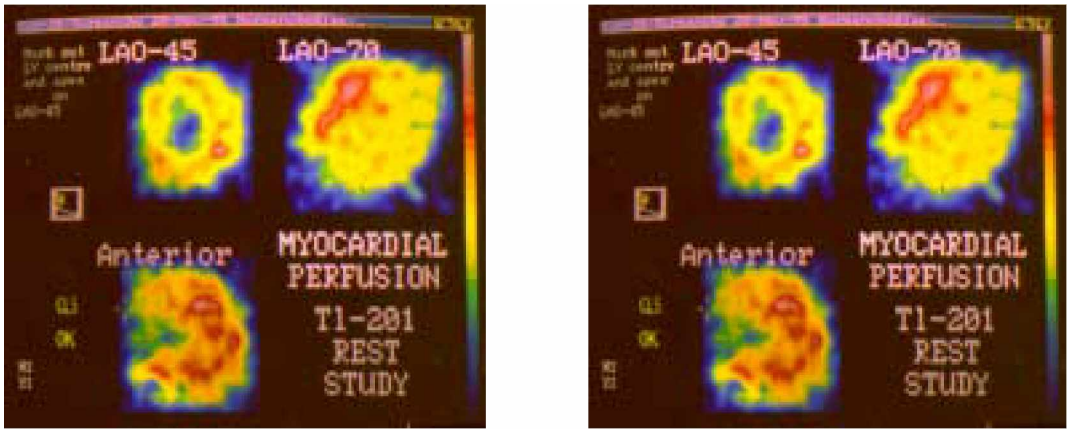


Fig.4. Tomographic heart image using thallium-201.

Table 1. Specification of «Tl-201, chloride», produced in the Institute of Nuclear Physics NNC RK.

Half life	73,1 hours
Energy of gamma-rays	0,072 MeV (Hg K Xrays) 0,135 MeV 0,167 MeV (Tl-201 γ-rays)
Radionuclidic impurity	Tl-202 < 0,2% Tl-200 < 1,5% Pb-203 < 0,05% Pb-201 < 0,05%
Nonactive impurities	Tl ≤ 2,0 μg/ml Fe ≤ 1,0 μg/ml Cu ≤ 0,1 μg/ml
Specific activity	≥ 55 MBq/ml for the date of specification
Chemical form	Sterile, isotopic, aqueous solution of thallium chloride
Expiry day	Average 10 days post preparation
Administration : Normal dosage Whole body absorbed dose Miocard absorbed dose	 50 – 74 MBq or 1,5 – 2,0 mCi 0,057 mZv/MBq 0,35 mZv/MBq

Radioactive tracers.

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 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.

Pu-237 radionuclide yield in reaction $^{237}U(\alpha,2n)\ ^{237}Pu$ for α-particles energy 46,5 MeV was 90 Bq/(μA·h).

The total yield of Pu-236 and Pu-238 was 6 Bq/(μA·h).

Produced plutonium sources were used as radioactive tracers in radiochemical processes for plutonium detection in the soils of Semipalatinsk Nuclear Test Site in Kazakhstan.

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

Other isotopes.

Alongside with Tl-201 some other radioisotopes are produced at the cyclotron [3]. Co-57 is produced using technique developed at the Institute and is used at the isotope laboratories in Medical Centres of Almaty. Necessary quantities of Co-57 for Mössbauer sources production are regularly produced for internal use in Kazakhstan.

In this year research on production of radioactive sources of Cd-109 comes to completion. Cd-109 is produced in reaction $^{109}Ag(p,n)^{109}Cd$. Technology of thermoresistant silver layer deposition on the copper backing production is developed. Extraction-chromatographic technique of cadmium extraction from the irradiated target is worked out. An prototype of sealed radioactive sources was produced and now it is used in ecology research carried out in the Institute.

Work on development of radiopharmaceutical "Ga-67-citrate" production is coming to completion. In the nearest future pharmaceutical will be transferred to hospitals for clinical trials. Ga-67 is produced in nuclear reaction $^{67}Zn(p,n)^{67}Ga$. The irradiated target processing is carried out on the base of chromatography column extraction technique. Optimization of gallium separation from zinc on TOPO was determined on stable gallium, zinc and copper model solutions. Analysis of the final product had shown absence of zinc and copper impurities. "Gallium-67 citrate" preparation was produced in Kazakhstan for the first time and its quality corresponds to similar preparation of international standard.

CONCLUSIONS

Experience acquired at the Institute of Nuclear Physics during research and production of cyclotron based radioactive isotopes assures that many isotopes may be produced in the INP using K=50 MeV cyclotron to meet need of the Republic of Kazakhstan in radioactive isotopes to be used in medicine, industry, science, ecology etc.

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