TECHNIQUE FOR IRRADIATION OF NB-GA TARGETS AT KAZAKHSTAN ISOCHRONOUS CYCLOTRON

A.A. Arzumanov, V.V. Alexandrenko, A.R.Borisenko, D.N.Ignatenko, V.K.Koptev, S.N.Lyssukhin,
Yu.S.Popov, G.I.Sychikov, B.A.Volkov
Institute of Nuclear Physics, NNC, Almaty, Kazakhstan

Abstract

Institute of Nuclear Physics in Almaty has got considerable experience developing radioisotope production technique that employs 30 MeV protons accelerated by positive ions at INP’s multifunctional cyclotron (K= 50 MeV). Currently, works on irradiation of Niobium-Gallium capsulated targets for production Ge-68 have been initiated. Nb capsule containing Ga for irradiation is water cooled at backside of the target. Proton beam is sweeped azimuthally across target surface at spiral two circles for obtaining more homogeneous irradiation. Heat transfer is evaluated analytically and experimentally by determination of local target temperature with infrared thermometer at the electron beam test stand. Obtained results are discussed. Work is supported by the Department of Energy of the USA.

INTRODUCTION

Nowadays uses of radioisotopes in Nuclear Medicine have become a mainstay in diagnostic imaging, especially in functional imaging which relies essentially on positron emission tomography (PET). When compared with traditional nuclear medicine, PET provides better resolution of images and it also shows the metabolic cell functioning allowing extremely precise diagnosis and reliable disease monitoring by studying the metabolism of pharmaceutical molecules marked with specific radioisotopes.

For getting reliable results of investigations (scan) it is needed to calibrate PET instalment with a positron source. Additionally each clinical scan of a patient must be accompanied with a “transmission” scan. For these purposes an ideal isotope is Gallium-68 (half time is equal to 68.3 min, single gamma-line 1077 keV).

It is applied as a generator Germanium-Gallium. Practically, each PET- facility utilises this isotope for calibration and as a transmission source. Demand in Germanium-68 increased sharply with quick growth of PET applications in medicine.

Considerable half-life of Ge-68 allows its production in accelerators located far from facilities for reprocessing of irradiated gallium targets. This made possible the cooperative work within IPP providing Nb-Ga targets irradiation at cyclotron in Almaty and their processing in Los Alamos.

The cyclotron [1] is a 150 cm compact-pole 3-sector, variable energy, positive ion machine designed to operate in first and third harmonic mode. In first harmonic protons, deuterons, He-3 ions, He-4 ions are accelerated at the cyclotron. Maximal proton beam intensity of internal beam is equal to 500 μA and 30 μA for extracted beam. The cyclotron has been designed for pulse operation with off-duty factor of 2-10 and pulse width 0.5 – 1 ms. Main research activities include studies of nuclear reactions, nuclear fission induced by charged particles, material science investigations, radionuclide production including Tl-201, Co-57, Cd-109, Ga-67, Pu-237.

TARGET

There are several methods known for production of Germanium-68 at cyclotrons. Considering further processing of produced gallium, the usage of Ga in hermetic Nb capsule is the best method for Ga processing. Gallium of natural composition (60.1 % of Ga^{69} and 39.9 % of Ga^{71}) is used as a rule. Cross-section of the reactions Ga^{nat} (p, 2n; p, 4n) Ge^{68}, firstly were measured in the INP, Almaty [2]. Later they were included into database of the IAEA [3]. This method is easy to realise when the proton beam energy overlaps the reaction section curve partly or completely. In this case two-sided cooling and irradiation with beam current of 90 – 100 μA may be used at homogenous current distribution over round target of 20 mm in diameter. At that the energy release density comprises in target about 10 W/mm^2. It seems that this thermal load is close to the limit for capsulated Nb-Ga target. This method is not optimal at INP cyclotron because cooling of the capsule front side will inevitably reduce beam energy down to 25 – 27 MeV and the product yield will be reduced. Output assessment for cases when proton beam with Ep = 30 MeV is absorbed by 300 μm walls of a niobium capsule before it reaches metallic gallium and for a capsule with two-sided cooling and 1-mm aluminium front side and water layer thickness of 0.5 mm gives us values 1.3 MBq/μA.h and 1.0 MBq/μA.h, respectively.

Then it was needed to take into account the possibility of beam manipulation to achieve homogenous thermal load at target. It is important that Ga is a very active metal that dissolves and interacts with other metals quite well. In particular, Nb is dissolved with Ga at temperatures 400 ºC and higher and forms fragile, mechanically weak compound niobium gallate. That is why in the region of contact between Ga and Nb in a target this temperature should not be exceeded at long-term irradiation. So, first of all we investigated possibilities for irradiation at extracted beam; in this case there are possibilities to achieve uniform irradiation.

Therefore, development of a technology for irradiation of Nb-Ga target with one-side cooling is of importance for production of Ge-68 at proton beam of up to 30 MeV. Taking stated above into account there has been designed a target with capsulated gallium. Figure 1 represents one of the possible designs of such target.

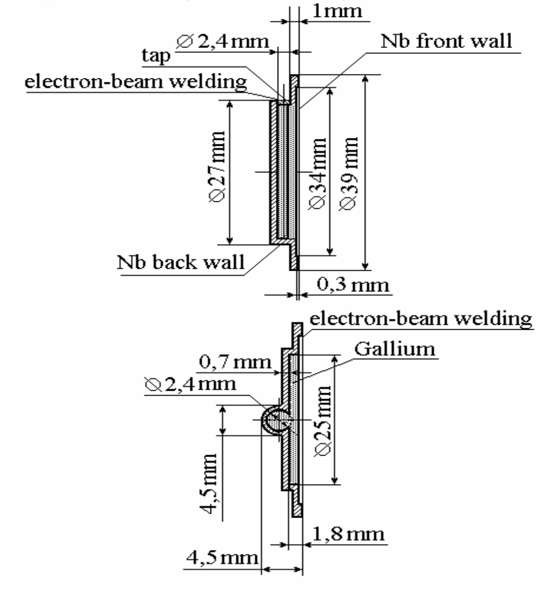

Fig.1: Nb-Ga target design (type 1).

There were investigated two types of capsules:

1. An empty capsule welded with electron-beam welding and then filled in with Ga using a syringe through a special hole at the back lid. The hole was then sealed (welded).

2. Welding was performed along the capsule outline upon filling it with gallium. In this case there is no technological lug in the back lid of the capsule.

For experimental purposes there were manufactured other target dummies:

There was designed a Type 2 capsule (no welding along the outline) for visual assessment of interaction between Nb and Ga by opening it after thermal impact at the electron-beam stand.

Thicknesses of capsule layers (front side, Ga-layer, back side) were chosen taking into account mechanical strength and reliability at influence of cooling water, minimal energy loss in the front side and energy absorption in Ga-layer to the reaction threshold of 11 MeV. So, thermal loads on a target system and Ga-layer were reduced to minimum. In order to irradiate a target is mounted into a unit that assures handling operations, cooling and beam current measurements. Pressure of cooling water is 5 bar, water consumption of about 4 l/min, thickness of cooling flow 0.5 mm, water temperature 14 ºC.

ELECTRON-BEAM STAND

In order to assess thermal load on target there were performed numerical calculations and investigations at the electron-beam stand [4]. Also there was introduced circular electron beam scanning for obtaining homogeneous circle target irradiation.

The gun is equipped with solenoid lens that provides minimal beam size on a target of 1.5 mm. Maximal beam current is 160 mA at electron energy up to 25 keV. Target unit of the stand is identical to the target unit of the cyclotron; their cooling systems are also identical. The stand is equipped with TV-unit enabling targeting and control of beam position on a target and monitoring of irradiation process. Target surface temperature is measured with an IR-thermometer (M-90 ZB Micron) through sodium-chloride window. Spectral respond of the IR-thermometer is equal to 8-14 μm, field of the view for measurement 180:1, temperature resolution 1°C.

Based on available in literature data and additional experiments, radiation coefficient for niobium surface is taken to be 0.13.

Assessed thermal stability of the target depends considerably on electron beam current distribution. Measurements made using wire probes and by the small-hole method showed that the best one in our case is a beam of 4–5 mm in diameter. At that current density distribution is close to the Gaussian one.

One should mention that irradiation at the stand does not completely simulate conditions of a target at the cyclotron. At irradiation with proton beam power is released in the bulk of the target. Range for 20 keV electrons comprises in metals about 10 μm.

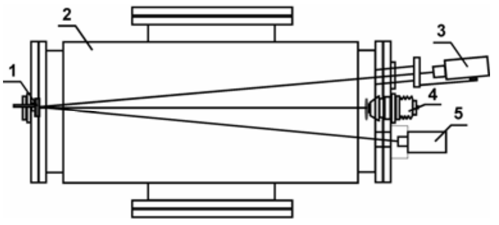

Fig.2 Scheme of target's temperature measurement at the electron-beam stand.

1. Target
2. Vacuum tank
3. Infrared thermometer
4. Electron gun
5. TV camera

Hence, at irradiation with electron beam of materials with low thermal conductivity surface temperature is achieved at a target at lower thermal flows than at a cyclotron.

To develop the regime for target irradiation with 30 MeV proton beam the following has been completed:

- Considered two variants for irradiation with 17 keV electron beam
- Assessed temperature gradients along target depth
- Modeled 3-D field of temperatures
- Temperatures on a target surface were experimentally measured.

Based on obtained results there were assessed maximal allowable heat loads on targets at irradiations with proton beams considering circular scanning over the target surface. Therefore, stand tests enable us, from one side, assess maximal temperature differences allowable for a target and, from the other side, assure thermal stability of a target at irradiation with proton beams of known energy.

Table 1: Assessments and experimental measurements of temperature at vacuum surface of a target.

I, mA	**Q, W**	**q_s, W/cm2**	**T_{max} caculated, degC**	**T_{max} measured, degC**
20.4	346.8	70.78	85	91
30.2	513.4	104.78	119	117
40.4	686.8	140.16	155	141
50.1	851.7	173.82	188	175
60.4	1026.8	209.55	224	212
70.2	1193.4	243.55	258	238
80.4	1366.8	278.94	293	264
90	1530	312.24	311	280

Described above numerical simulation of 3-D distribution for temperature field at irradiation currents 50 and 90 mA showed maximal temperatures to be 190 and 329 ºC, respectively. In the Fig. 3 temperature distribution from cooled surface to vacuum surface of the Nb-Ga-Nb target is shown.

Similar method was used for assessment of irradiation with proton beam.

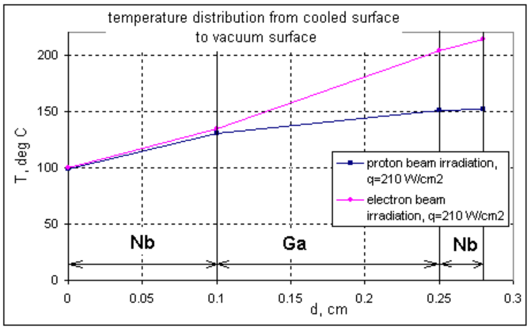

Fig. 3. Temperature distribution of Nb-Ga-Nb target at the irradiation with protons and electrons at the same heat flow (210 W/cm^2).

According to this consideration and irradiation of a target with 30 µA proton beam, temperatures at vacuum-facing surface reached 151 ºC and maximal allowable current the scanned proton beam comprises about 80 µA. In this case temperature of vacuum facing surface reached 400 ºC.

FORMATION OF IRRADIATION FIELD BY EXTRACTED BEAM.

Uniformity of temperature distribution depends on time structure of the beam (pulse duration and duty factor) and, even more, on uniformity of irradiation field over target surface. In order to reveal irradiation conditions series of target dummies were irradiated with varied macro-pulse width, duty factor, beam scanning frequency and scanning method (one- or two-beam), diameter of collimated initial beam (at no scanning), different cooling water flow rates. So, there was chosen optimal irradiation regime.

- Considering irradiation of Ga target with extracted beam there were performed the following works in order to increase beam current:
- optimized ion source performance,
- electrical reliability of the deflector was increased,
- there was increased aperture of the initial part of beam transportation system what resulted in lower beam loss while in the channel,
- there was introduced the system of double circle azimuthally beam scanning on the target surface.
- As a result of cyclotron systems' upgrade it becomes possible to irradiate targets in conditions of continuous and long-term irradiation with extracted beam of 40 µA and 30 MeV.
- For further processing in LANL were produced two gallium targets – an experimental one manufactured in INP (Type 2) and a target provided by LANL (Type 1). Total accumulated Ge-68 activity of these targets was about 100 mCi.

CONCLUSTION

Obtained results made it possible to start irradiation of capsulated niobium-gallium targets for Ge-68 production at the cyclotron in INP, Almaty, with their following processing in LANL. In order to increase production effectiveness it is planned to irradiate capsulated targets in the cyclotron chamber.

ACKNOWLEDGEMENT

The authors are grateful for support of this work by the USA Department of Energy Initiatives for Proliferation Prevention Program.

REFERENCES

[1] A.A.Arzumanov "Kazakhstan Isochronous Cyclotron – Features and Applications". Proc. Of the 3-rd Conference and Workshop on Cyclotrons and Applications. Feb. 1999, Cairo, Egypt, 90-100
[2] Levkovsky, V.N., Activation cross-sections for the nuclides of medium mass region (A=40-100) with protons and a-particles at medium (E=10-50 MeV) energies. (Experiment and systematics), Inter-Vesi, Moscow (1991).
[3] IAEA TECDOC – 1211,2001, pp 273 – 277.
[4] A.Arzumanov, A.Borissenko et. al. "The Electron Beam Test Facility for Cyclotron Isotope Targets Examination". Proc. of the 9-th IWTTC, Turku, Finland, May, 2002, pp 34-37.