A FAST METHOD FOR PRODUCING SUPER-PURE RADIONUCLIDE ^{57}Co AT THE CLASSICAL 1.5 METER CYCLOTRON OF THE INSTITUTE OF NUCLEAR PHYSICS Ac.Sc.RUZ.

RYLOV A.A., GULAMOV I.R., PORTYANKO A.P., YULDASHEV B.S.

The Cyclotron Center "TEZLATGICH", Institute of Nuclear Physics, Academy of Sciences, Ulugbek, Tashkent, UZ-702132, Uzbekistan.

A very fast method for radiation and chemical purification of an internal water-cooled target consisting of a thin enriched ^{58}Ni layer covering a cooper backing is described. This method allows to obtain the radionuclide ^{57}Co in the chloride form with radionuclide purity less than 0.1% and with total activity of about 1 Ci after 10 days from the beginning of irradiation. The specific thermal power per surface unit during irradiation of the target can be decreased by a first harmonic in the peripheral radial range of acceleration. Remote control of thermal power during irradiation is accomplished by scanning of infrared emission from target surface and by restoring its temperature distribution using computer method. Step-by-step chemical solution of the target and its purification in the earlier stage of holding allow to reduce the ^{56}Co contamination of the total radionuclide purity of ^{57}Co. The intensity of the internal proton beam on the target is 500-600 μa and thermal power is up to 10-12 kW.

1. Introduction

It is shown, that the bombardment of internal target by high intensive proton beam with relatively low energy, 18-20 MeV, and step-by-step chemical solution allow to reduce the time from the beginning of irradiation till delivery of pure ^{57}Co as a final product, in comparison with regular terms, in two times. The scanning of infrared radiation from target surface preventing the production of defective targets is described as well.

2. Internal target irradiation

It is known, that radionuclide ^{57}Co can be obtained from reaction [4] :

$$^{58}Ni\ (p,2p)\ ^{57}Co \tag{2.1}$$

$$^{58}Ni\ (p,pn)\ ^{57}Ni \xrightarrow[37hrs]{ec,\beta^+} {}^{57}Co \tag{2.2}$$

and that its accumulation has a square-law dependence on energy of proton. Simultaneously, the accumulation of such chemically inseparable isotopes as ^{56}Co, ^{58}Co and ^{60}Co increases with the growth of proton energy and reduces radionuclide purity. Therefore, the target irradiation has to be carried out, preferably, with relatively low energy of protons, about 18-20 MeV, and, in that case, the productivity of isotope ^{57}Co is determined by intensity of internal beam entirely.

Cyclotron U-150 -- classic cyclotron with two dees, flat poles that are the lids of vacuum tank and RF resonator ,which has frequency range from 8 MHz to 16.5 MHz allows to accelerate light particles such as proton, deuteron, ^3He, ^4He. E=22 MeV is the maximum of proton energy and can be reached at f=16.2 MHz of RF resonator. The fitting of the magnetic field, providing optimal vertical focusing and right change of particle phases relative to electric potential between dees, allows to reduce beam loss during acceleration and, consequently, give a possibility to increase the intensity of proton beam by increasing burst time of accelerating pulse. The form of magnetic field has been obtained by minimizing a function of beam loss. The phase loss was calculated by numerical integration of the equations of motion in horizontal. In the rectangular frame, when Y-axis is directed between dees and electric field is presumed homogeneous i.e. $E_y = 0$, $E = E_x$, the equations of motion are [1]:

$$\frac{d^2}{dt^2}X = \frac{q}{m}E_m\cos\left(\omega_f \cdot t + \theta_o\right) + \frac{q}{m}B_z\frac{d}{dt}Y \tag{2.3}$$

$$\frac{d^2}{dt^2}Y = -\frac{q}{m}B_z\frac{d}{dt}X \tag{2.4}$$

where: q, m -- charge and mass of proton, B_Z -- magnetic field, E_m --amplitude of electric field, ω_f -- RF generator frequency. First, the form of B_Z is given as a plot and we have to use the fitting polynomial to insert the magnetic field into (2.3). The proton phase is determined by moment of time t_k, when particle crosses Y-axis i.e. as a solution of equation :

$$X(t_k) = 0 \tag{2.5}$$

then the phase: $\theta_k = \omega_f t_k - k\pi$ (2.6)

The phase loss is a number of particles, which have phases that lay out of range $(-0.5\pi; +0.5\pi)$. The vertical loss can be found by integration of vertical equation of motion:

$$\frac{d^2Z}{dt^2} + \omega^2(n_1 + n_2 + n_3)Z = 0 \tag{2.7}$$

where: $n_1 = \dfrac{d\ln B_z}{d\ln r}$;

$$n_2 = \left[\frac{qE_m}{W}\right]^{3/2} \frac{\cos^2(\omega_f \bullet t_k + \theta_0)}{\pi};$$

$$n_3 = \frac{qE_m}{\pi W} \sin(\omega_f \bullet t_k + \theta_0)$$

$$W = 0.5m\left[\frac{dr}{dt}\right]^2; \omega = cmq^{-1}\left[\frac{dr}{dt}\right] B_z$$

If given earlier magnetic field does not supply beam with the minimum of its loss, we have to choose another plot and start calculations from (2.1). The distribution of magnetic field along the radius, in the main, is formed by ring shimming nearby the edge of poles and disk shims at the center of accelerator. The fine tuning of magnetic field is determined by additional concentric coils at the periphery of U-150 and at its center. The independent sources of electric current supplying all coils let to have a possibility to tune the median plane at the center as well as on target radius. Obtained optimal magnetic field was approximated by magnetic field of shims and coils. The contribution to main field from them, with accuracy up to $4\pi M$, where M- magnetization of material for shims and $0.4\pi J\omega$, where $J\omega$ ampere-turns of coils were determined by [5]:

$$B_z(r,0) \approx 0.5\Delta h\left[(R+r)^2 + h^2\right]^{-0.5} \left\{F_0(\alpha) + (R^2 - r^2 - h^2)\left[(R-r)^2 + h^2\right]^{-1} E_0(\alpha)\right\} \tag{2.8}$$

where: R_1, R_2 -- radii of shim/coil edges, $R=0.5(R_1+R_2)$ -- its mean radius, $\Delta h=h_1-h_2$ -- the height of shim/coil, $h=0.5(h_1+h_2)$ -- mean height, $F_0(\alpha)$, $E_0(\alpha)$ -- elliptic integrals 1 and 2, α - was given by:

$$\sin\alpha = \left\{4Rr\left[(R+r)^{-1} + h^2\right]\right\}^{0.5} \tag{2.9}$$

All the results of calculations were tested by experiment with beam that was carried out as [2,3] and good coincidence with the theory has been received. Other obstacle that prevented the growth of beam intensity was high-frequency heating appearing in some places of RF resonator with bad contacts and weak water-cooling. This cause made to remove moving plates of resonator, which tuned it on the working frequency and to design new RF-resonator with only one frequency f=16.2 MHz and improved water-cooling system. High intensive beam requires to form special beam distribution on the target surface. The orbits of protons are kept at the center by pairs of harmonic coils installed outside on the edges of poles as well as inside nearby the center of U-150. Internal coils are used to adjust protons trajectories to central optics. The amplitude, phase of first harmonic and position of external coils were received by calculations of last orbits, i.e. by integration of the equations of motion (2.3), (2.4), in assumption that magnetic field had first harmonic. It was found that the bombardment of target could be carried by last three orbits, which touched target surface leaving one-third of their intensity on it. This result was proved by direct measurements of temperature distribution, which had three thermal peaks. The fitted magnetic field and new RF resonator allowed to decrease beam loss to 3-5% and to operate with high intensive proton beam, 500-600 μA. Now, the rate dead time/ burst time for accelerating pulse is equal to 3-5. Internal target is installed inside of the vacuum tank through vacuum lock along the line between dees and the angle from target surface to this line is $81\text{-}86^0$. The working size of target is 100x20 mm^2 . The backside of target is ribbed to improve heat-eliminating ability. The target is cooled by water, water pressure is 6-7 ATM, water consumption is 60 l/min. Before irradiation, proton beam is tuned on the probe installed in target place through another vacuum lock.

3. Thermal scanning

The scanner can measure the temperature of heated spot on the target surface in the range of 100-500 detecting infrared radiation. The horizontal canal is cut through left dee and body of main magnet to observe target projection. The angle from cannel axis to the line between dees is 27^0. UR-radiation is focused by spherical mirror with focus distance R=440 mm on pyroelectrical crystal with sensivity 0.5 V/W. The horizontal motion of crystal produces the same motion of the spot on the target surface, because the mirror remains motionless. For the vertical scanning, a heavy- walled glass is installed on the way of UR-radiation so, when glass rotates and its horizontal width changes the spot on the target has to move vertically. The lead container is driven by a stepping motor providing the measurement of horizontal temperature distribution and another stepping motor rotates thick glass for vertical measurements. Scanner resolution determined by direct measurement of thermal standards is 5x5mm^2. Mechanical modulator interrupts UR-radiation with frequency 30Hz and prevents receiver from saturation. The head amplifier with differential input and filter for all frequencies above 1 kHz allows to overcome RF noise. After additional amplifying, the signal is transformed into digital form by ADC and stored in IBM PC. For fast ADC, the program software has the possibility for fast

Fourie-analysis filtering the signal additionally. Program software builds full thermal image of the target. The vertical and horizontal motion of the spot on the target can be governed by coordinates of special marker moving on the screen of display. Thus, the operator has opportunities to get not only full target image automatically, but to choose an interesting area or point on the target.

4. Chemical purification

The main obstacle in receiving pure ^{57}Co with total content of ^{56}Co, ^{58}Co, ^{60}Co about 0.1% is their accumulation during proton bombardment. 80% of contamination is supplied by ^{56}Co, which is obtained from reaction [4] :

$$^{58}Ni\left(p,2pn\right)^{56}Co \qquad (4.1)$$

during irradiation and:

$$^{58}Ni(p,p2n)^{56}Ni\xrightarrow[6.1days]{ec}{}^{56}Co \qquad (4.2)$$

from ^{58}Ni decay[4] during target holding, because of decay of short-lived isotopes. The accumulation of ^{56}Co can be decreased (up to 30% from its total content) by reducing the holding time to 2-3 days, when the accumulation of ^{57}Co from decay (2.2) is over. After 2-3 days the target remains hot and it has made us to design a new radiochemical box with extra shield that let to dissolve hot targets with activity above 5 Ci from all isotopes. It is known, energy limit for ^{56}Co-formation~19,9 MeV, according to (4.1), is higher that energy limit for ^{57}Co (8,33 MeV), see (2.1),. therefore, ^{56}Co has tendency to accumulate itself in the upper layers of the target rather than in lower one[6]. The elimination of upper layer does not reduce total activity of target substantially (about 5% from total activity) but allows to decrease ^{56} Co content in final product up to 20%. Using both opportunities we succeed to reduce content of ^{56}Co on 50%.

Acknowledgment

We would like to express our gratitude to Prof. Krasnov N.N. and Dr. Ognev A.A. for their long-standing and fruitful cooperation that was very helpful in preparation of the article.

References

1. Livingood J., Principles of cyclic particle accelerators, Princeton, 1961
2. Krasnov N.N., Ognev A.A. et al, PTE, 2, Moscow, 1968 (in Russian)
3. Krasnov N.N., Ognev A.A. et al, PTE, 3, Moscow, 1971 (in Russian)
4. ICRP Publication, 38, Radionuclide Transformation, Pergamon Press, 1987
5. Danilov V.I., Zaplatin N.L. et al, JINR, P-702, Dubna, 1961(in Russian)
6. Dmitriev P.P. , The yields of radoinuclides in he reaction with proton; deutronos, ^{4}He, ^{3}He. Moscow. 1986 (in Russian)