

PHOTONUCLEAR PRODUCTION OF ^{111}In ON THE LINEAR ELECTRON ACCELERATOR

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Received 12 November 2012

Abstract–The technique of obtaining of medical intended ^{111}In isotope by irradiation of 91% enriched ^{112}Sn target with intense beam of bremsstrahlung photons from the 40 MeV electron beam of LUE50 linear accelerator of A.I. Alikhanian National Science Laboratory (formerly Yerevan Physics Institute) is described. Experimental methods of irradiation, measurements and the yield calculation are presented. The level of activity of impurities is estimated.

1. Introduction

In recent decades, the radioactive isotopes come into wide use in medicine for diagnosis of various diseases, as well as in therapy. New field was called nuclear medicine. The diagnosis used isotopes emitting gamma rays and positrons. These isotopes can be accumulated in different organs, and their radiation will be registered providing useful information. This is done using SPECT (Single Photon Emission Computer Tomography) and PET (Positron Emission Tomography) studies. For therapeutic purposes, the isotopes with α or β^- activities are more convenient, because they have great ionizing power and can destroy tumor cells in a very small radius, without damaging healthy cells.

^{111}In radioisotope is used in diagnostics, as it emits gamma rays with energies of 171.3 keV and 245.4 keV. But in addition to that this isotope can be used for therapy too entering ^{111}In in the tumor area with some heavy isotope. The photons from ^{111}In give the photoelectric effect on the electrons of heavy isotope. The electrons emitted from the K shell – so-called Auger electrons – have low energy, high ionizing effect on short distances and can destroy cells just in the tumor's area.

Traditionally the radionuclides are produced in reactors and cyclotrons because of the high cross sections of the neutron and proton activation. But for getting of a number of radionuclides the photonuclear reactions cross sections are comparable to corresponding neutron and proton activation cross sections [1-3]. This allows obtaining these isotopes using bremsstrahlung photons from electron accelerators. One of these isotopes is ^{111}In .

This isotope can be produced by the following photonuclear reactions on ^{112}Sn target:

- 1) $^{112}\text{Sn}(\gamma, p)^{111}\text{In}$, the reaction threshold is 7.55 MeV and Coulomb barrier is 10.78 MeV,
- 2) $^{112}\text{Sn}(\gamma, n)^{111}\text{Sn} \rightarrow (35.3 \text{ min } \beta^+) \rightarrow ^{111}\text{In}$, the reaction threshold is 10.79 MeV.

In the Fig.1 the decay scheme of ^{111}In is shown. In the decay process the 100% electronic capture is taking place. The gamma rays are emitting with energies of 171.3 keV (90%) and

245.4 keV (94%), and transition from ^{111}In to the stable ^{111}Cd occurs. The half-life of ^{111}In is 2.8047 d, or about 67.3 h.

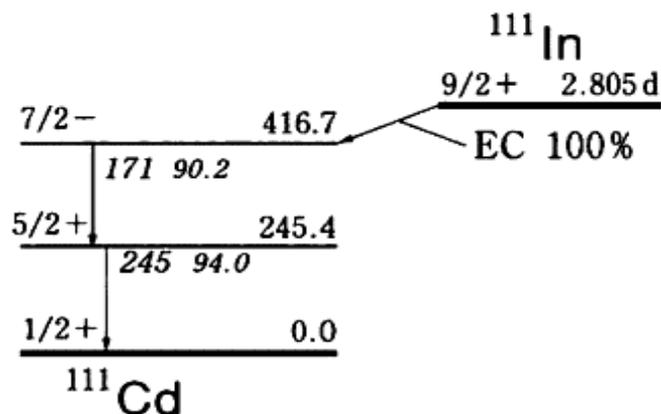


Fig.1. Decay scheme of the ^{111}In isotope.

2. Experimental Layout

Previously, an experimental facility had been developed by YerPhI employees for irradiation of different solid targets by high-energy photons [4]. Facility is mounted in the ring hall of YerPhI electron synchrotron at injector beamline (Fig.2).

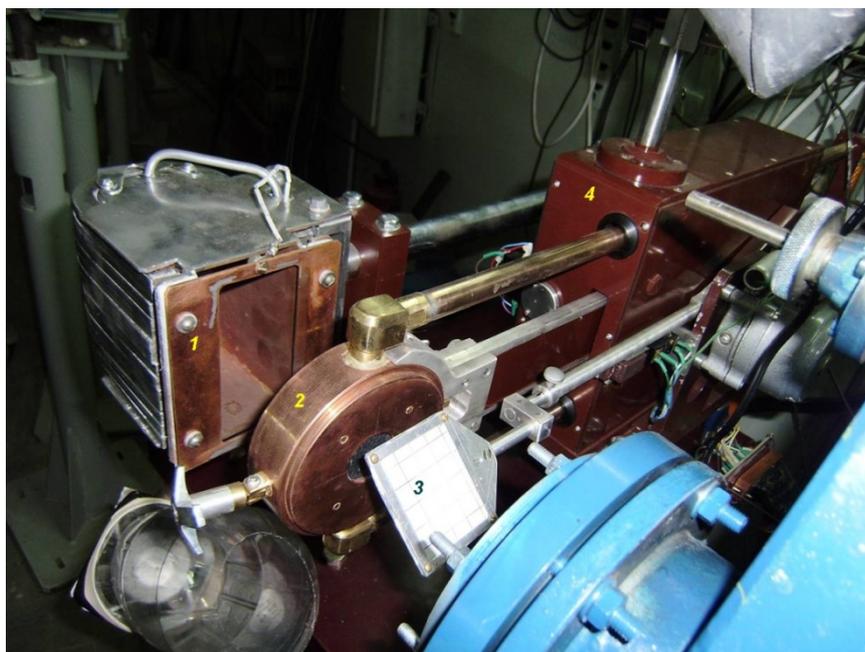


Fig.2. Experimental layout for irradiation under electron beam (top view). 1 – Faraday cup, 2 – target module, 3 – luminophore frame for the TV monitor beam profile, 4 – remote moving target module.

The electron beam passes the beamline window and falls on luminophore screen 3, which was remotely inserted under the beam for monitoring beam profile and then moved out. Installed at several points video cameras can display on video monitors in the control room the beam profile at several points: at the output window of beamline, at the entrance of the target module, and at the

entrance window of the Faraday cup 1. The target module 2 remotely moves to the beam and can be removed from the beam if direct measurement of the beam current is necessary. Movement of the target module is implemented by device 4. The sketch of the target module is shown in Fig.3.

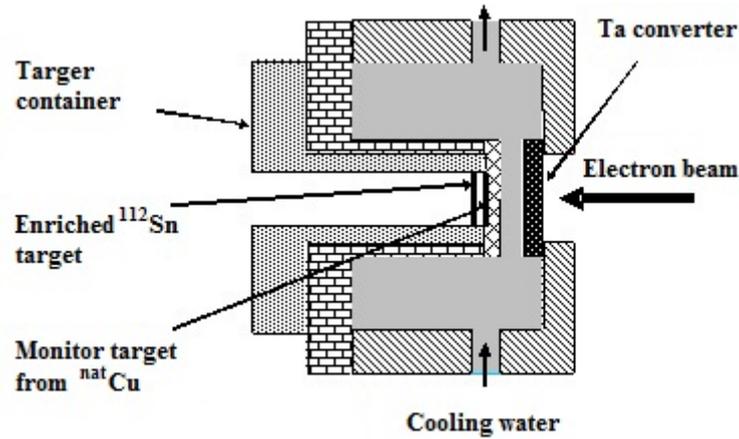


Fig.3. Target module sketch.

The electron beam converts to bremsstrahlung photons in the 2 mm thick tantalum converter. After that, the secondary photon beam hits the copper and the ^{112}Sn targets.

3. Calculation of Bremsstrahlung Photons Number Incident on the Target

A 91% enriched ^{112}Sn was used as a target material with 0.163 g of weight, 17 mm of diameter and 100 μm of thickness. Duration of irradiation was 18 min, the beam current was 7.6 μA , electron energy 40 MeV. A target from natural copper with 0.095 g of weight, 17 mm of diameter and 50 μm of thickness was chosen as monitor. These two targets are irradiated simultaneously, and the number of photons incident on the both targets is almost the same as the targets are very thin and the photons absorption is negligible. The photonuclear monitor reactions occur in the copper. With these reactions one can determine the number of photons incident on the targets by measuring the spectrum intensity of the irradiated copper and having (from world data) measured yields for those reactions at the same maximum energy of bremsstrahlung photons – 40 MeV.

The natural copper contains two isotopes – ^{63}Cu (69.15%) and ^{65}Cu (30.85%). The following photonuclear reactions occur on these isotopes – $^{65}\text{Cu}(\gamma, n)^{64}\text{Cu} - \sigma_q = 53 \text{ mb}$, and $^{63}\text{Cu}(\gamma, 2n)^{61}\text{Cu} - \sigma_q = 2.6 \text{ mb}$ [5]. ^{64}Cu has a gamma line with energy 1345.8 keV and its half-life is 12.7 h. ^{61}Cu has gamma line with energy of 283 keV and its half-life of 3.333 h. Inserting the values of activation yields of these reactions in the equation (1) of activation [6], one can obtain the number of photons incident on the targets.

$$N_\gamma = \Delta N \lambda / \left(\sigma_q N_{\text{nuc}} k \varepsilon \eta \left(1 - e^{-\lambda t_1} \right) e^{-\lambda t_2} \left(1 - e^{-\lambda t_3} \right) \right), \quad (1)$$

where ΔN is the number of events under the photopeak measured by the detector during the time t_3 , λ is the decay constant, σ_q is yield of the reaction, N_{nuc} is the number of nuclei involved in the reaction on 1 cm² of target, k is coefficient of absorption of gamma rays in the target, air and detector's cup, ε is detector efficiency, η is the intensity of the gamma line, t_1 is duration of exposure, t_2 is the time between exposure and measurement, t_3 is the duration of the measurement. The average value for N_γ from data of both reactions is 2×10^{16} ph/h.

4. Irradiation Results

The ¹¹¹In isotope can be obtained by two channels (reactions) simultaneously:

- 1) (γ, p) reaction forms the final radioisotope – ¹¹²Sn (γ, p) ¹¹¹In,
- 2) (γ, n) reaction produces ¹¹¹Sn, which quickly decays to ¹¹¹In with 35.3 min of half-lives:
¹¹²Sn (γ, n) ¹¹¹Sn \rightarrow (35,3 min β^+) \rightarrow ¹¹¹In.

In Fig.4 the gamma spectrum of the irradiated material is shown.

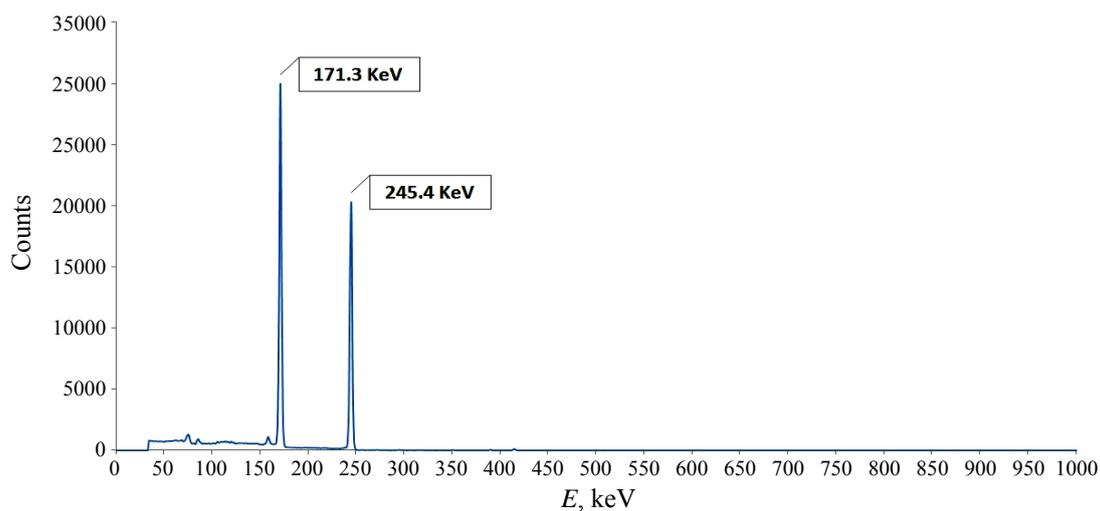


Fig.4. Gamma spectrum of the irradiated material after 332 h from the end of irradiation.

¹¹¹In has the gamma lines with energies 171.3 keV (90%) and 245.4 keV (94%). A measured gamma spectrum of irradiated material shows these lines and their intensities according to experimental data, proving the presence of the desired isotope. Here, however, the peak at 171.3 keV with intensity of 90% with higher amplitude than the 245.4 keV peak of 94%, should not cause a contradiction, because the 10/25 efficiency of the detector is higher in energy 171.3 keV (0.76% at a distance of 10 cm from the source to the detector's cup) than at 245.4 keV (0.6% at a distance of 10 cm from the source to the detector's cup).

Now for yield of ¹¹¹In one can find the value $\sigma_q = 113$ mb from the equation (2) inserting ΔN from spectra and N_γ from calculations:

$$\sigma_q = \Delta N \lambda / \left(N_\gamma N_{\text{nuc}} k \varepsilon \eta \left(1 - e^{-\lambda t_1} \right) e^{-\lambda t_2} \left(1 - e^{-\lambda t_3} \right) \right). \quad (2)$$

The activity of medical radioisotope is important factor in the nuclear medicine. In our case just after the end of irradiation there was still a huge amount of ^{111}Sn , which decays into ^{111}In with half-life of 35.3 min. So we will bring the activity of ^{111}In after a day from the end of irradiation, when almost all ^{111}Sn nuclides have already decayed into ^{111}In . The activity of ^{111}In at that time was 5.8×10^5 Bq, or the specific activity – normalized to the mass of irradiated material, beam current and duration of irradiation - was 1.7×10^3 Bq/mg mA h.

5. Efficiency Measurement of HPGe Detector

The spectrum of the irradiated material was measured with high resolution HPGe (ORTEC) detector [7]. For that the 0/10/25 efficiency calibration of HPGe detector was done using ^{155}Eu , ^{57}Co and ^{22}Na isotopes. These isotopes have the following lines: 86.5 KeV (^{155}Eu), 105.3 keV (^{155}Eu), 122.06 keV (^{57}Co), 136.47 keV (^{57}Co), 511 keV (^{22}Na) and 1274.6 keV (^{22}Na). In Fig.5 measured efficiency at 0 cm, 10 cm and 25 cm distance from the cup of detector is shown.

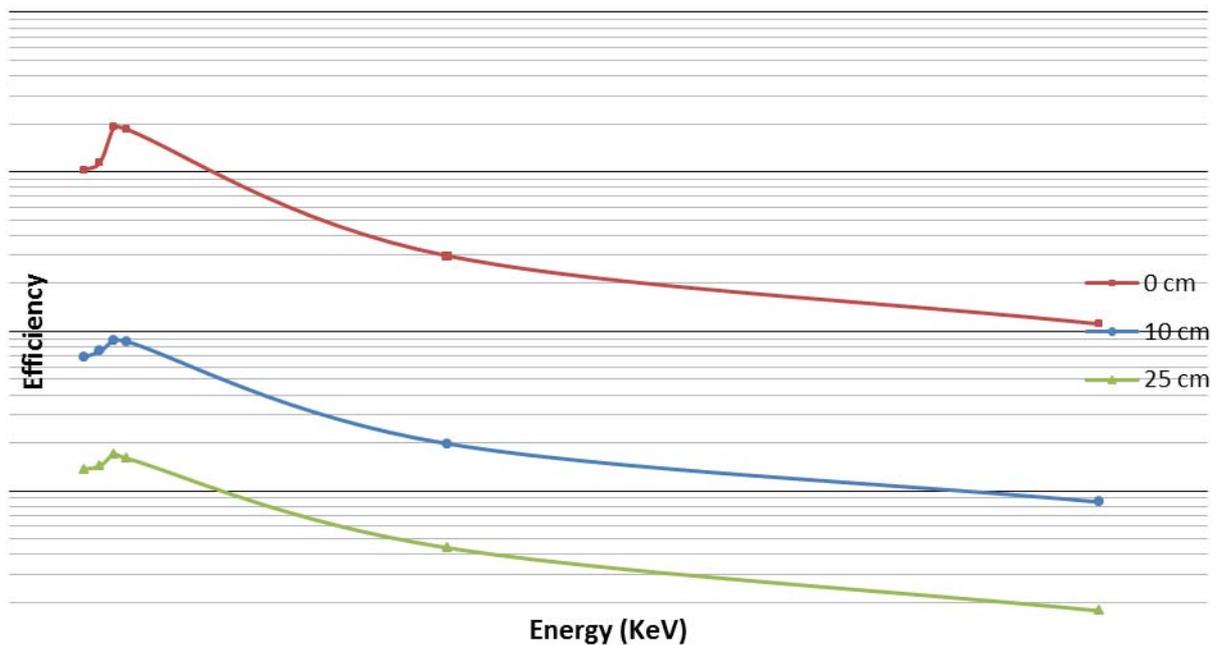


Fig.5. Efficiency of HPGe (ORTEC) detector at 0 cm, 10 cm and 25 cm distance from the detector's cup.

6. Impurities

For obtaining the radiopharmaceutical of ^{111}In , one must separate ^{111}In from other isotopes of the target. It can be done chemically. But this method does not separate ^{111}In from other isotopes of tin. In consequence of photonuclear reactions other isotopes of In can produced along ^{111}In . The activity of other isotopes should be very low compared to activity of ^{111}In . The ^{110}In can be produced by $^{112}\text{Sn}(\gamma, np)^{110}\text{In}$ reaction, but with a very low probability. In addition, the reaction

threshold is 17.61 MeV (the Coulomb barrier is 10.78 MeV) and flux of bremsstrahlung photons decreases sharply when increases their energy. And the half-life of ^{110}In is 4.51 h, hence its activity will decrease by 40 times after a day. Since we have a target of 91% enriched ^{112}Sn , then the activity of other produced isotopes of In from the remaining Sn isotopes, which together constitute only 9%, will be rather lower. The half-lives of these isotopes are shorter than that of ^{110}In , except for $^{114\text{m}}\text{In}$ (half-life is 49.51 d). But this isotope can mainly produced by the reaction $^{115}\text{Sn}(\gamma, p)^{114\text{m}}\text{In}$ with very low probability too.

So, nearly after a day from the end of the irradiation activity of impurities will decrease many times and will be very low compared to the activity of ^{111}In . It allows getting radiopharmaceuticals by this method.

7. Conclusion

The experimental investigations show the opportunity of obtaining ^{111}In on the 40 MeV electron accelerator using 91% enriched ^{112}Sn target. The value $\sigma_q = 113$ mb for photonuclear yield of ^{111}In was carried out from calculations. After a day from the end of irradiation the activity of ^{111}In was 5.8×10^5 Bq, or the specific activity – normalized to the mass of irradiated material, beam current and duration of irradiation was 1.7×10^3 Bq/mg $\mu\text{A h}$. The estimation of activity of impurities shows that after a day from the end of irradiation all impurities activity will be rather low, which allows to obtain ^{111}In for medical purposes.

Acknowledgements

The author gratefully acknowledges Prof. Robert Avakian, Dr. Albert Avetisyan, Dr. Ivetta Kerobyan and Prof. Alita Danagulyan for their kind help and support.

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