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EXPERIMENTAL INVESTIGATION OF THE POSSIBILITY OF ^{99m}Tc ISOTOPE PRODUCTION FOR MEDICAL APPLICATION ON THE BASE OF LINEAR ELECTRON ACCELERATOR LUE50 OF THE YEREVAN PHYSICS INSTITUTE

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Abstract
We describe the process of radioactive ^{99m}Tc isotope production through irradiation of molybdenum by the intensive secondary photon beam of the Linear Electron Accelerator (LUE50) of the Yerevan Physics Institute. An experimental setup for the irradiation has been developed and installed. Experimental investigations of ^{99m}Tc production have been performed with qualitative and quantitative results given here.

Introduction

The radiopharmaceutical industry of practically all industrialized countries uses ^{99}Mo to produce ^{99m}Tc generators used in ~80% of all diagnostic procedures of nuclear medicine.

Currently the world consumption of this radionuclide exceeds $2\cdot10^5$ Ci per year and ^{99m}Tc production is still extremely important [1].

The common method for parent ^{99}Mo production is using a nuclear reactor method where the ^{235}U fission reaction is triggered by thermal and fast neutrons - $^{235}U(n,f) \rightarrow {}^{99}Mo$ [2,3]. But this procedure may have serious ecological consequences, therefore in the last decades alternative methods of $^{99}Mo \rightarrow {}^{99m}Tc$ production are under active discussion in the world community, in particular the ^{99}Mo production method by means of molybdenum irradiation by protons with energies Ep=30 MeV [4] and Ep=70 MeV[5,6].

An alternative method of ^{99}Mo production is the reaction $\gamma + {}^{100}Mo \rightarrow {}^{99}Mo + n$ [7,8] photonuclear reaction using electron accelerators with a specific yield of tens and hundreds of Bk/μA·h·mg of ^{100}Mo. The present paper is devoted to the investigation of the possibility of ^{99m}Tc production on the basis of linear electron accelerator LUE50 of the Yerevan Physics Institute(YerPhI).

1. The linear electron accelerator

The linear electron accelerator LUE50 of the Yerevan Physics Institute with an electron beam energy up to 50 MeV is generally used as an injector for the 4.5 GeV Yerevan Electron Synchrotron but has also other applications. The LUE50 parameters are – electron beam energy up to 50 MeV, average current 5-10 μA, repetition frequency 50 Hz and pulse length up

to 1 μsec. The beam of this accelerator with energy Ee=20 MeV has been used for the first experimental investigation of the ^{99}Mo production.
The results of a calculation of beam dynamics and tracing indicate a possibility of having a beam spot diameter of 10-15 mm at ~5 μA of beam current on the exit window of beam pipe in the ring hall of electron synchrotron. For a qualitative experiment such intensity is sufficient but for industrial production the beam current of an order of magnitude higher is needed. Such an increase of the intensity of this accelerator is generally possible, and this opportunity will be discussed in the Conclusion of this paper.

2. The experimental layout.
The experimental layout has been installed in the ring hall of the Yerevan Electron synchrotron on the beam line of injector. The view of experimental layout is seen on a Figure 1.

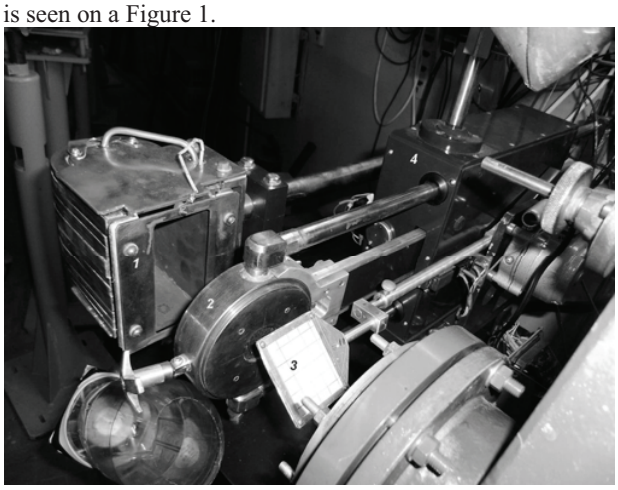

Fig.1 Experimental layout top view. 1 – Faraday cup, 2 – target device, 3 – luminofore frame for the beam spot size and position TV control, 4 – target remote control movement device.

At the exit window of the beam pipe the electron beam hits the luminofore frame 3 which is remotely controlled for moving it in and out of the beam line. A few video cameras monitor this frame and the luminofore frames on the exit window, target module and Faraday cup. The target module 2 was also remotely moved into the beam during exposition and sometimes moved out for direct measurement of beam current. The movement of target module was performed by device 4.
The block-diagram of target module is presented on the Figure 2.

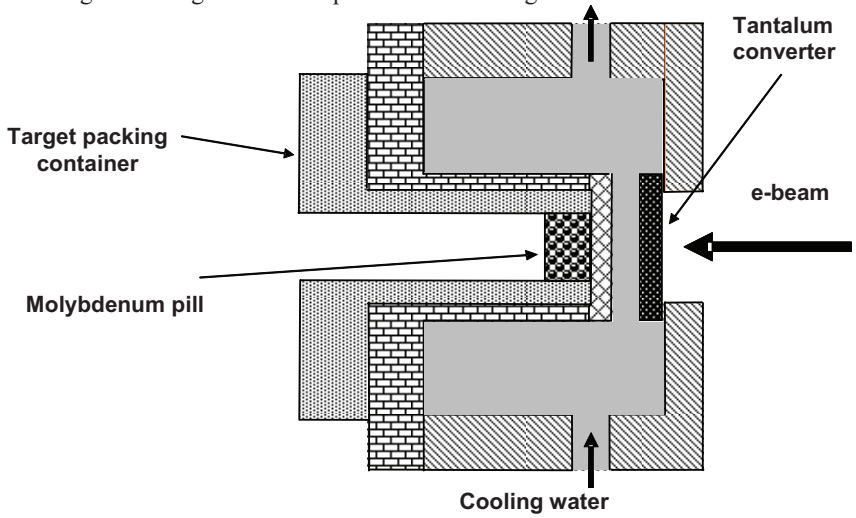

Figure 2. The target module diagram.

The body of target module is fabricated of copper. On the entrance window a 2mm (0.5 radiation length) thick tantalum plate has been installed to convert the electron beam to photons. By increasing the thickness of the converter the number of photons will increased at first but after reaching the critical thickness will fall off due to predominating photon absorption processes in the converter body. A Monte-Carlo simulation of an optimal thickness of the converter has been performed. The resulting dependence of photon yield on the converter thickness is presented on Figure 3, indicating that the optimum thickness of the radiator is about 2 mm.

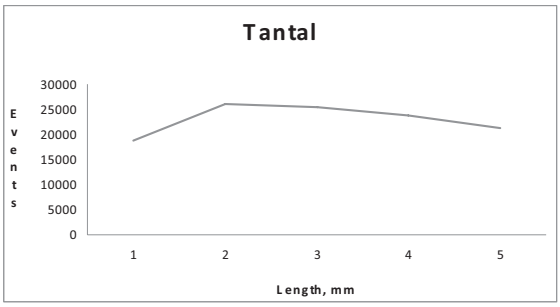

Figure 3. Dependence of photon yield on the converter thickness.

The calculation of electron beam energy absorption shows that about 30% of the energy transforms to thermal form in the target module and remaining part in the Faraday cup. Since the electron energy is Ee=20 MeV and the beam current Ie=5-10 μA – the total beam energy is W=100-200 Wt so that the thermal energy deposit from beam to target module will be on the level of W=30-60 Wt. Since in the nearest future a significant increase of the beam intensity is foreseen the question of target module and also of Faraday cup cooling is very important.
The diagram of water cooling system is presented on Figure 4.

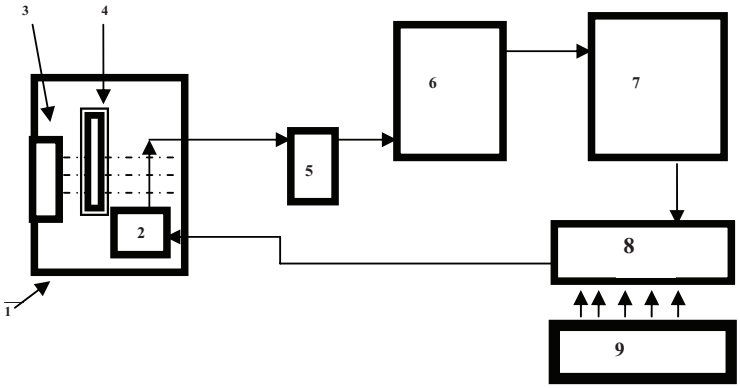

Figure 4. The water cooling system block-diagram. 1 – distilled water tank, 2 – water pump, 3 – water level sensor, 4 – electronic thermometer sensor, 5- water pressure sensor, 6 - Faraday cup, 7 – target module, 8 – cooler radiator, 9 – electric funs.

To avoid charge leakage from the Faraday cup only pure distilled water (with high specific resistance) is used in the cooling system. Water is pumped through the pipes in the body of the Faraday cup, then through the volume of the target module (see Figure 2) absorbing the heat from them, and then flows through the cooling radiator which is blowing by air fans, and is returned to the water tank. Sensors of the water level in the tank and water pressure in the cooling system prevent emergency situations of water leakage from the cooling system. Electronic thermometer is monitoring the water temperature.
Since all these devices are installed in the ring hall of synchrotron where the level of electromagnetic background is very high the following method of temperature measurement was chosen: at first the current of a thermal resistor was converted to signal (NIM standard) with a corresponding frequency and transmitted to the control room via coaxial cable, where they were counted by a CAMAC scaler. The current in the Faraday cup was measured in a similar way. The values of water temperature and beam current were displayed on the computer monitor and stored in the slow control files. The DAQ and visualization of these parameters was done by LabView software.

3. Irradiation modes

For the irradiation on the first step the oxide of natural molybdenum MoO_3 has been chosen. In the natural molybdenum concentration of stable isotope ^{100}Mo is only 9.1%. For these 2 reasons an amount of ^{100}Mo was really irradiated by photonuclear reaction was only 0.22 gram while the total weight of MoO_3 was 3.6 gram. The molybdenum oxide powder was molded in a pill (see Fig.2) and inserted in the target module on a distance of 4 mm from tantalum converter.
Two shifts of irradiation have been done on end of 2008, in both cases the amount of irradiated material was the same. The energy of electrons was Ee=20±1.2 MeV, the beam intensity was Ie=~5 μA. The duration of the first irradiation was 4.5 hours, and for the second one – two times longer. After irradiation finished the irradiated material was still stored in the target module for ~12 hours – during that time all short-living isotopes were dissociated, and activity level decreased till safe values.

4. Irradiation results

To investigate the produced ^{99m}Tc the radiation spectra of irradiated material has been measured. As a result of photonuclear reaction the ^{100}Mo converted to ^{99}Mo which with $T_{1/2}$~67 hours converted to ^{99m}Tc. The ^{99m}Tc within $T_{1/2}$=6.7 hours converts to stable ^{99}Tc. This two-step process has a radiation characteristic line Eγ=180 KeV from ^{99}Mo->^{99m}Tc transformation, with an intensity ~5% of total radiation, and a radiation characteristic line Eγ=140 KeV which is the main line from ^{99m}Tc. To detect the radiation spectra of irradiated material the NaI(Tl) based detector 3M3/3-X (producer- ORTEC-digiBASE) has been used.
The Figure 5 shows spectra of irradiated material measured after 39 hours after finishing the irradiation. 2 hard peaks with 140 KeV and 180 KeV energies are seen with intensities close to awaited.

The distance from irradiated material to NaI(Tl) detector was 53 cm to avoid an overlaps of registered pulses. The exposition time was chosen 500 sec. For this distance the size of irradiated material pills could be neglect so that the ratio of total solid angle 4 π to the acceptance of detection is $\Omega_{total}/\Omega_{registration}$=774. Taking into account this point the number of registered photons $2.2 \cdot 10^6$ during 500 sec respects to the intensity of radiation $I=3.4 \cdot 10^6$ Bk. The main peak of radiation is fitted by Gaussian function (solid line), fitting parameters are seen in the right top corner, showing that energy resolution of used NaI(Tl) detector is σ = 5.9 KeV for Eγ=140 KeV region.
The same irradiated material energy spectrum has been measured also by Ge solid detector with very high energy resolution (see Figure 6).

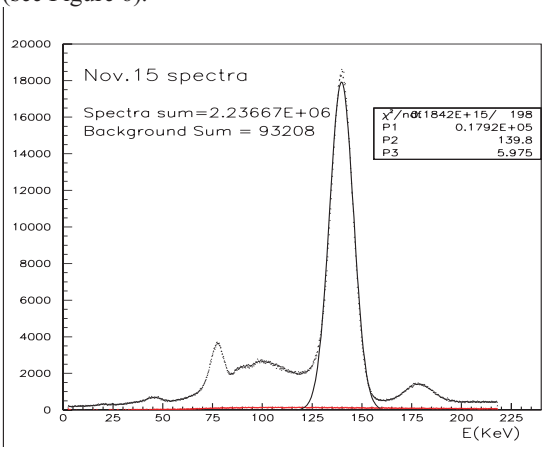

Figure 5. The spectra of irradiated material after 39 hours of irradiation finishing.

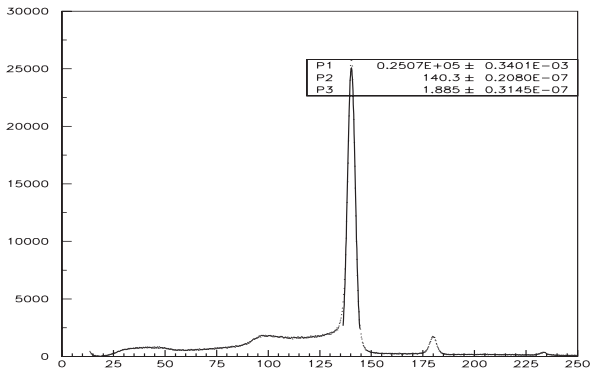

Figure 6. Energy spectra of irradiated material measured by Ge semiconductor detector.

Comparing spectrum from Figures 5 and 6 one can see that peaks position and spectra structure is the same and the only lines width is different. For second case Gaussian fitting results σ = 1.9 KeV. The resume of this comparison is – for describing task the accuracy and energy resolution of NaI(Tl) detector is quite enough.
To investigate the half-life time $T_{1/2}$ of irradiated material the energy spectra have been measured after 39, 66 and 86 hours of irradiation finishing. The dependence of radiation intensity on time is seen on the Figure 7. Triangles are experimental values, dotted line – exponential analytic function describing their behavior.
$I=3.22*10^6*e^{-t/9.61}$

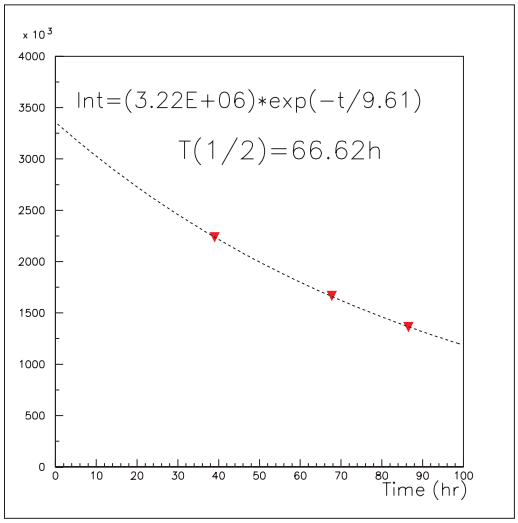

Figure 7. Dependence of radiation intensity from irradiated material on time.

The simple calculation shows that for this dependence of intensity on time the half-life time $\boldsymbol{T_{1/2}}$ =66.2 h which is in a good coincidence with that parameter for ^{99}Mo.
One of the main parameter by production of radioisotopes under electron beam is the specified activity normalized to mass of irradiated material, beam current and time – Bk/mg·μA·h.The data published from different experiments have very big dispersion – from 90 to 3200 Bk/mg·μ·h. Results from presenting experiment is 695 Bk/mg·μA·h which is in the middle of wide spectra of world data.
Chemical extraction of ^{99m}Tc from irradiated material has been carried out.

Conclusion

First time a full-scale experiment on the linear electron accelerator LUE50 of the Yerevan Physics Institute has been done showing the fundamental possibility of medical-isotope ^{99m}Tc production on that beam. Calculation shows that in case of serious increasing of electron beam intensity (which is possible in general and R&D of that option are in progress) the real test-production of medical-isotopes is possible which could cover the need of a part of Armenian hospitals.

Acknowledgment.

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NEUTRON ACTIVATION ANALYSIS OF TRACE ELEMENT CONTENTS IN THE CORTICAL BONE SAMPLES OF HUMAN FEMORAL NECK

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Introduction

Osteoporosis represents a significant social and medical problem in terms of treatment and rehabilitation.[1,2] The disease causes a decrease in the bone mineral part and subsequent bone strength. The bone can be considered a mineralized protein complex consisting of calcium hydroxyapatite [$Ca_{10}(PO_4)_6 \cdot (OH)_2$], collagen, marrow, fat, non collagen proteins, water, minor and trace elements also. Since bone is a biomaterial that is structurally adapted to different functions and loading situations in adults, the exact composition may very depending on sex, age, type and site, but also with alterations known to occur in bone metabolic diseases.[3]

Bone strength depends mainly on the level of mineralization and contents of Ca and P consequently. The importance of minor and trace elements in provision of the bone function and strength has been yet insufficiently studied.[4,5] There are several reviews and texts regarding chemical element analysis of different human bones, using chemical techniques and instrumental methods.[6-9] However, the majority of these data are based upon non intact bones. In most cases, bone samples are treated with solvents in order to remove collagen, fat, marrow and then are ashed and acid digested. There is evidence that by these methods some amount of chemical elements is lost upon treatment.[10-13] In addition, by this way the measured chemical element mass fractions are referred to the mineral part of bone and not to the whole intact bone. Thus, to understand the role of major, minor, and trace elements in the etiology and pathogenesis of bone diseases including osteoporosis it is necessary first of all to determine the normal levels and age- and gender-related differences of chemical element contents in intact bone in a large scale study.