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^{99m}Tc PHOTO-PRODUCTION UNDER ELECTRON LINEAR ACCELERATOR BEAM

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Abstract–A method of ^{99m}Tc production by irradiation of molybdenum using high-intensity bremsstrahlung photons from the electron beam of linear electron accelerator LUE50 of the Yerevan Physics Institute is described. The experimental layout for the development of ^{99m}Tc production technology has been designed and constructed. An upgrade of the linear electron accelerator was carried out to increase the electron beam intensity and spatial density. A system of computer-based remote control of the accelerator and experimental layout has been built up. A system for ^{99m}Tc extraction from irradiated MoO_3 has been commissioned and installed. Experimental investigations of ^{99m}Tc production method have been performed and the results presented. The first amount of ^{99m}Tc has been produced.

1. Introduction

^{99m}Tc is the most widely used isotope in nuclear medicine today [1,2] for over 30 million diagnostic medical imaging scans every year around the world [3,4].

^{99m}Tc decays to the technetium-99 ground state (^{99g}Tc) with a half-life of 6 hours by emitting a 140 keV photon that is detected by the imaging devices. Since the ^{99m}Tc half-life is short, it should be produced within a reasonably short distance from where it is used. Fortunately, molybdenum-99 (^{99}Mo) decays predominantly to ^{99m}Tc with a half-life of 66 hours. Medical centers or commercial radiopharmaceutical distributors purchase ^{99}Mo/^{99m}Tc generators from which ^{99m}Tc (and necessarily ^{99g}Tc) can be extracted periodically in a simple chemical process as it accumulates from the parent ^{99}Mo decays. ^{99m}Tc is then bound into the pharmaceutics for the imaging procedure [4-7].

^{99m}Tc is derived as a daughter isotope from ^{99}Mo decay (see Fig.1).

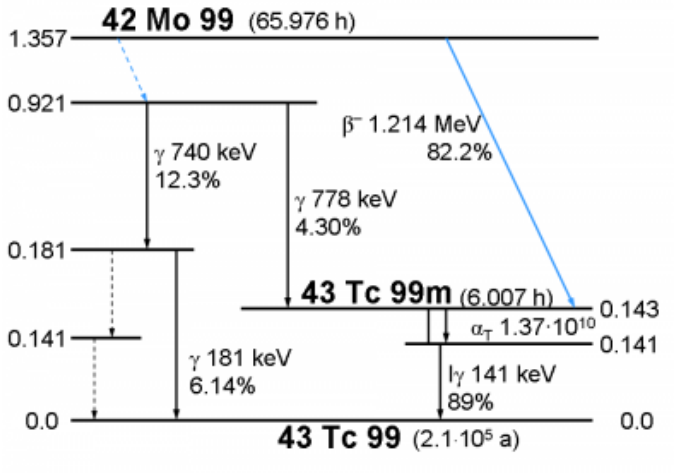

Fig.1. ^{99}Mo decay chain.

One of considered options was a photonuclear reaction [7-12]. Metastable ^{99m}Tc could be obtained in the photonuclear reaction by irradiation of ^{100}Mo under intensive photon beam:

$$\gamma + {}^{100}\mathrm{Mo} \rightarrow {}^{99}\mathrm{Mo} + \mathrm{n} \quad \text{Threshold} = 9.1\ \text{MeV}$$
$$\downarrow$$
$$T_{1/2} \sim 67\,\text{hours} \rightarrow {}^{99m}\mathrm{Tc}\,(T_{1/2} \sim 67\,\text{hours}).$$

For this option the electron beam should be converted to a photon beam via bremsstrahlung. This method does not provide high specific activity and therefore does not allow building Mo/Tc generators but could cover the demand of regional and city clinics.

2. Electron Beam

The linear electron accelerator was designed, built and used many years as an injector for the Yerevan ring synchrotron. To use its electron beam for photon-induced reaction the upgrade of electron gun has been done. A new high intensity metallic cathode was installed with slightly modified gun electrodes so that the maximum intensity grows from 3 up to ~10 μA. From three accelerator sections two were in use providing $E_e = 40$ MeV electron energy. The electron beam was transported to the target magnetic optics in a way that the beam spot diameter on the target was 12 mm, measured by luminophore frame or vibrating wire scanner [13]. The beam pulse length was ~1.1 μs, repetition frequency $f = 50$ Hz.

3. Experimental Layout for Irradiation

A special experimental layout [14] (see Fig.2) for material irradiation has been designed and mounted that provides remote controlled motion of target module across the beam direction adjusting the center of the target to the beam axes. The body of target module (see Fig.3) was fabricated of copper. A thick tantalum plate has been installed on the entrance window to convert the electron beam to photons. 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 in Fig.4, indicating that the optimum thickness of the tantalum radiator is about 2 mm (0.5 rad).

For direct measurement of beam intensity the Faraday cup (1 on Fig.2) [15] has been used. A Faraday cup is a metal (conductive) cup designed to catch charged particles and consists of lead bottom with 5 cm of thickness and magnet to bring back the secondary electrons. The intensity measurements are performed when the target module was remotely moved out from the beam

position. During the irradiation, only a part of the secondary beam has caught by the Faraday cup, and the calibration needs for the recalculation to real-beam intensity.

Fig.2. The experimental layout. 1 – Faraday cup, 2 – target moveable module, 3 – luminophore frame for the beam spot size and position TV control (on the left photo) or vibrating wire scanner module (on right photo), 4 – movement tool of target module.

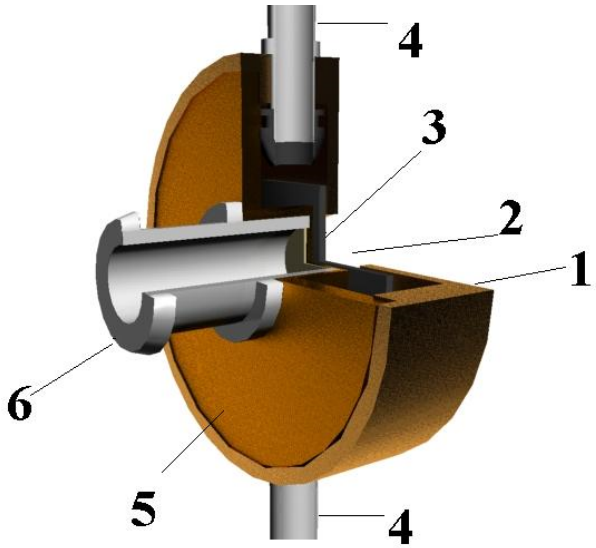

Fig.3. The target module. 1 – framework, 2 – beam entrance window, 3 – tantalum plate, 4 – water cooling pipes, 5 – cup, 6 – target capsule.

At the electron energy $E_e = 40$ MeV, and the beam intensity $I_e \sim 10$ μA total power of the beam is $P = 400$ W. For the transfer of heat, water and air cooling are required for the target module and Faraday cup. To avoid charge leakage from the Faraday cup only pure distilled water (with high specific resistance 0.2 MΩ cm) was used in the cooling system. The values of water temperature and beam current were displayed on the computer monitor and stored in the slow control files. The data acquisition and visualization of these parameters were done by LabVIEW [16] software.

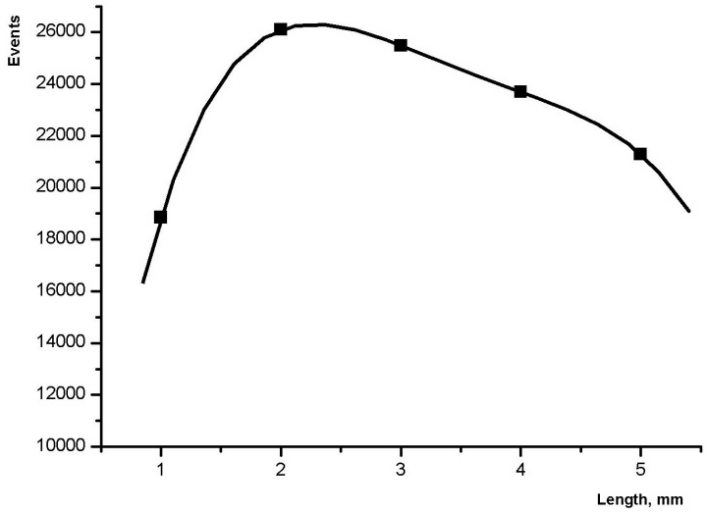

Fig.4. Dependence of the photon yield (in the energy range 9–20 MeV) from the converter thickness.

4. Irradiation Modes

For the irradiation the oxide of natural molybdenum MoO_3 has been chosen. The abundance of the stable isotope ^{100}Mo in natural molybdenum is 9.63%. The irradiated material was packed in a special aluminum capsule (see Fig.5). Two styles of target materials were used – a stack of pressed pellets (left) and full length pressed powder (right) covered by thin copper foil with thickness 0.045 g/cm^2. The first one was used to measure the dependence of activity on the depth of target, and the second one was used for trial production.

Fig.5. Target capsule for different tasks with pellets (left) and full amount (18 g) of pressed powder of MoO_3 (right).

The energy spectra from irradiated materials were measured by 3M3/3-X 905-4 type (producer ORTEC) [17] and HPGe (ORTEC) [18] detectors.

One of the main parameters of production of radioisotopes is the obtained specific activity normalized to the mass of irradiated material, beam current and duration of irradiation Bq/mg μA h. The data on the specific activity published by different experiments has very large variance – from 90 to 3200 Bq/mg μA h [9].

The irradiation has been done under beam intensity $I_e \sim 5.5$ μA during 5 hours. The energy spectrum from irradiated material measured by NaI(Tl) detector is presented in Fig.6. The spectrum was fitted by a Gaussian function; parameters of the fit are presented on the right top corner of the histogram. The mean value of Gaussian function is $E_\gamma \sim 140$ keV. Two peaks are seen in the spectrum with energies $E \sim 140$ keV from ^{99m}Tc and $E \sim 180$ keV from ^{99}Mo (see also Fig.1).

The obtained normalized specific activity calculated from this spectrum is $A \approx 3000$ Bq/mg μA h which is close to the maximum value of published results.

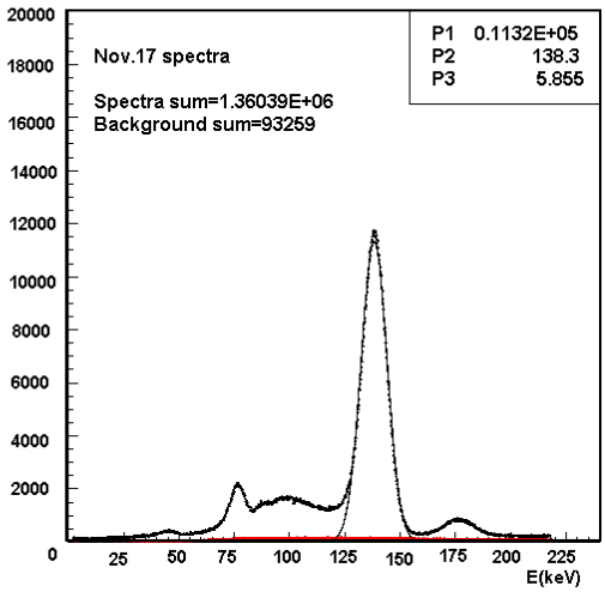

Fig.6. Energy spectrum from irradiated molybdenum.

5. Investigation of the Depth Dependence

To find an optimal thickness of irradiated material inside the target capsule the dependence of activity on the target material depth has been investigated. The Monte-Carlo simulation (see Fig.7) using GEANT 4 code [19,20] has been done analyzing a number of escaped photonuclear neutrons from the MoO_3 target.

To test this simulation results a special experiment has been carried out. The number of identical pellets, 2 g natural MoO_3 each, have been fabricated and then irradiated under beam with

energy $E_e = 40$ MeV and beam intensity $I_e \sim 8$ μA during 2.5 hours. Then activity of each pill was measured by NaI(Tl) detector once per day during 4 days. Results of measurements after 15.7, 37.8, 62.7 and 84.3 hours are presented in Fig.8.

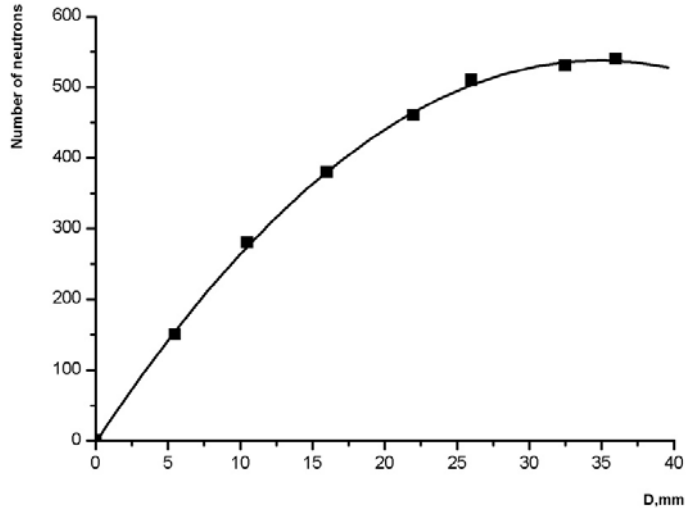

Fig.7. Calculated activity dependence on depth of natural MoO_3 target.

The data in Fig.8 shows that with increasing the thickness the activity of each pill is reduced. Increasing the thickness of the target may not infinite increase the activity because a part of photons are absorbing and therefore the beam intensity on the beam passing direction is going down.

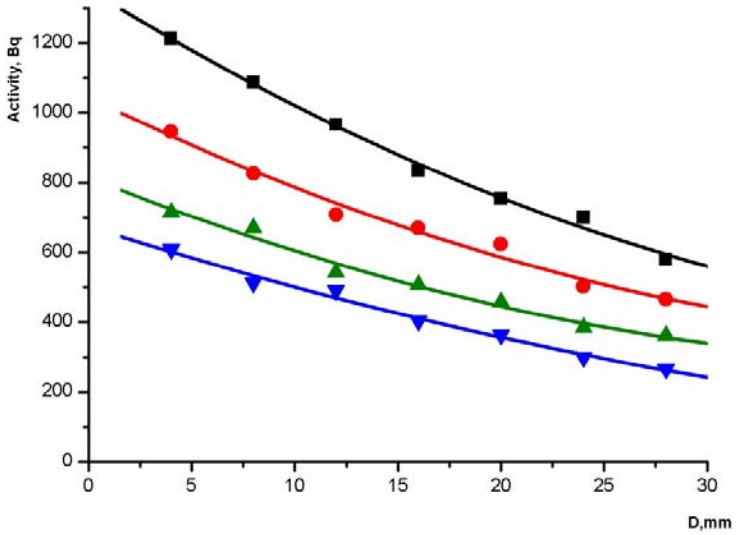

Fig.8. Measured obtained activity dependence on depth of natural MoO_3 target: ■ – 15.7 hours, ● – 37.8 hours, ▲ – 62.7 hours, ▼ – 84.3 hours after EOB.

The behavior of the accumulated in the target activity depending on the target thickness is shown in Fig.9. The red curve shows the increase in accumulated activity with increasing thickness

of the target in the absence of absorption of photons in the target material. The black curve shows the real increase in activity, taking into account the absorption of photons.

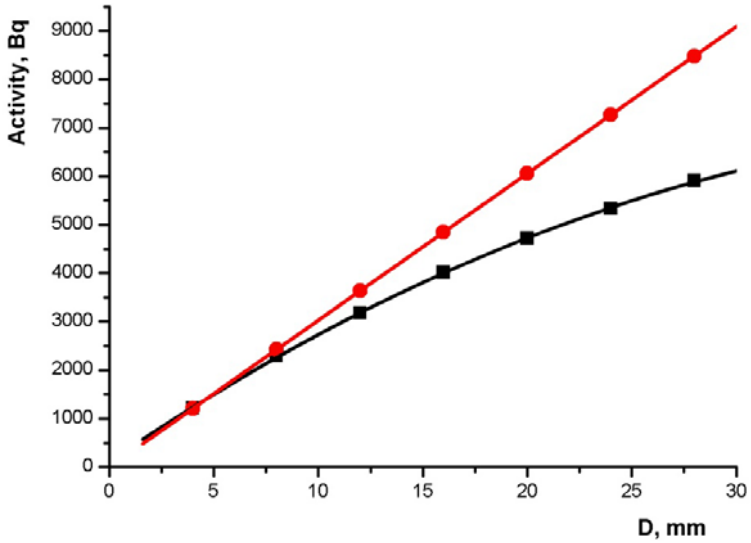

Fig.9. Measured integral activity (squares) and calculated integral activity assuming that the obtained activities of all pellets are the same (circles).

Thus, the determination of the optimum length of the target will provide economic benefits in the production of isotopes ^{99m}Tc. This is particularly important in the case of irradiation of the enriched ^{100}Mo.

One can see that for 30 mm of thickness the real activity is 1.5 times less than without considering the absorption.

6. Trial Production of ^{99m}Tc

For the low specific activity option only direct extraction of ^{99m}Tc from irradiated material is reasonable. For that a centrifuge extractor with Methyl Ethyl Ketone (MEK) solvent technology was chosen which successfully was used many years in Russia [21]. The irradiated MoO_3 is dissolved in KOH alkali and then MEK liquid is added to that solution. The irradiated MoO_3 dissolves in KOH while ^{99m}Tc dissolves in MEK so that we have mixture of two solutions with very different densities. The centrifuge extractor designed in Moscow A.N. Frumkin Institute of Physical Chemistry and Electrochemistry [22] separates them with high purity and then ^{99m}Tc is separated from MEK by evaporation. The complete automatic equipment commissioned by Moscow "Federal center of nuclear medicine projects design and development" of FMBA of Russia factory was installed in a "hot" cell (see Fig.10).

Fig.10. Main part of the centrifuge extractor complex.

The pressed natural MoO_3 target of 20 g mass and material density 4.96 g/cm^3 (a real density 0.8 g/cm^2) has been irradiated under electron beam with energy $E_e = 40$ MeV and average intensity $I_e \sim 9.5$ μA during 100 hours. Irradiated material was processed by centrifuge extractor and the first trial amount of ^{99m}Tc has been produced. The activity was ~80 mCi via reverse calculated to the EOB (end of bombarding). On subsequent days a new part of ^{99m}Tc is produced due to ^{99}Mo decay and it could be extracted daily during 5–6 days with a coefficient of extraction $k \sim 0.7$ related to the previous day.

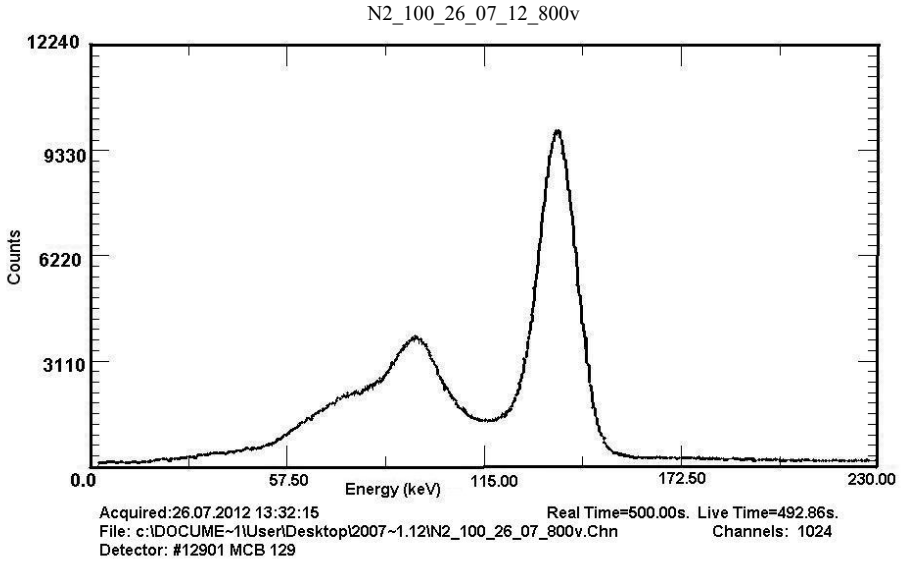

Fig.11. Energy spectrum from extracted ^{99m}Tc.

The energy spectrum from extracted ^{99m}Tc is shown in Fig.11. The huge peak at energy $E \sim 140$ keV from ^{99m}Tc is apparent. The peak around 180 keV from ^{99}Mo is absent. The left part of the spectrum is the edge of Compton scattering in the detector.

7. Conclusion

The theoretical and experimental investigation of ^{99m}Tc has shown that on the photon beam of the electron accelerator due to the reaction on the ^{100}Mo it is possible to carry out effective generation of ^{99}Mo by irradiation of the target of natural molybdenum. The used technologies of extraction ^{99m}Tc from irradiated molybdenum allow obtaining the final product with the purity suitable for delivering the clinics.

The next step should be the increasing of the beam intensity by repetition of the frequency and increasing of the pulse length to enhance the intensity 5–10 times. We aim to increase production intensity in a way that it will cover a part or full ^{99m}Tc demand of Armenian clinics.

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