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# Experimental Plant for Investigation of the Possibility of Linac-Based Production of Medicine-Intended Isotopes

**R. H. Avagyan[a], A. E. Avetisyan[a], I. A. Kerobyan[a], S. P. Taroyan[a], A. S. Danagulyan[b], R. M. Mirzoyan[a], K. S. Bunyatov[c], R. Ts. Sargsyan[a], S. A. Galumyan[a], V. S. Yeganov[a], A. A. Oganesyan[a], H. S. Vartanyan[a], V. B. Gavalyan[a], V. Ts. Nikogosyan[a], V. S. Ayrapetyan[a†], A. Z. Babayan[a], A. A. Matosyan[a], S. V. Jamkochyan[a], and R. K. Dallakyan[a]**

[a] *Alikhanian National Scientific Laboratory, Yerevan, Armenia*

[b] *Yerevan State University, Yerevan, Armenia*

[c] *ISTC, Moscow, Russian Federation*

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**Abstract**—We describe a technique of obtaining radioactive $^{99m}$Tc isotope by irradiation of molybdenum with high-intensity beam of bremsstrahlung photons from the electron beam of the linear electron accelerator (LUE50) of the Alikhanyan National Science Laboratory (ANSL, former Yerevan Physics Institute). We have elaborated and created an experimental plant for development of $^{99m}$Tc production technology. Upgrading of linac has been performed aimed at raising the beam intensity and density. A system of automated control of the parameters of the plant and accelerator was built up. We carried out preliminary studies of $^{99m}$Tc obtaining technique and give quantitative and qualitative results.

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## 1. INTRODUCTION

Radiopharmaceutical industry of actually all developed countries employs $^{99}$Mo for fabrication of generators of $^{99m}$Tc, which is applied in more than 80% of all diagnostic procedures in nuclear medicine. At present the world consumption of the $^{99m}$Tc radionuclide exceeds $7\times10^6$ GBq yearly [1] and its production remains extremely urgent problem.

For obtaining the parent $^{99}$Mo nuclide the basic is the reactor technique which employs the reaction of fission of $^{235}$U under action of thermal and fast neutrons, $^{235}\mathrm{U}(n, f) \rightarrow {}^{99}\mathrm{Mo}$ [2, 3]. However, production of $^{99}$Mo which is a product of uranium fission is accompanied by considerable ecological problems. Therefore alternative ways to produce $^{99}$Mo and, correspondingly $^{99m}$Tc, are widely considered in world practice. In particular, methods for obtaining $^{99}$Mo by irradiation of molybdenum with protons at energies of $E_p = 30$ MeV [4–6] and $E_p = 70$ MeV [7, 8] are proposed.

It is also possible to obtain $^{99}$Mo in electron accelerators as a result of photonuclear reaction $\gamma + {}^{100}\mathrm{Mo} \rightarrow {}^{99}\mathrm{Mo} + n$ [9–12] with a specific yield of tens and hundreds of Bq/μA mg hour. Our work deals with the study of possibility to produce $^{99}$Mo with use of linac of ANSL.

## 2. LINEAR ELECTRON ACCELERATOR LUE50

The working linear electron accelerator LUE50 of ANSL with the energy of the electron beam of up to 50 MeV serves mainly as injector for ring synchrotron for 4.5 GeV energy, but is also employed for a number of problems including applied ones.

Performed calculations of beam dynamics and tracing have shown the possibility to obtain at the outlet pipe in ring hall of ring electron accelerator EKU a beam of 10–15 mm diameter at the rated intensity of ~5 μA. This intensity is well sufficient for qualitative experiments, but for industrial production at least

twice as high intensity is required. For radical enhancement of the electron beam intensity a new metallic cathode with high emissive ability was mounted in the gun. As a result of this replacement the current of the electron beam rose from 4–5 μA to 9–10 μA. The picture of the new cathode in the gun assemblage is shown in Fig. 1.

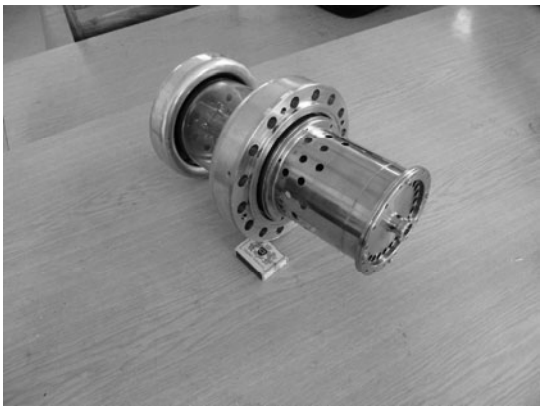

**Fig. 1.** Gun of linac LUE50 with the new metallic cathode.

## 3. EXPERIMENTAL PLANT

The experimental plant is mounted in the ring hall of the electron synchrotron on the beam extractor of injector. The photograph of experimental arrangement is given in Fig. 2.

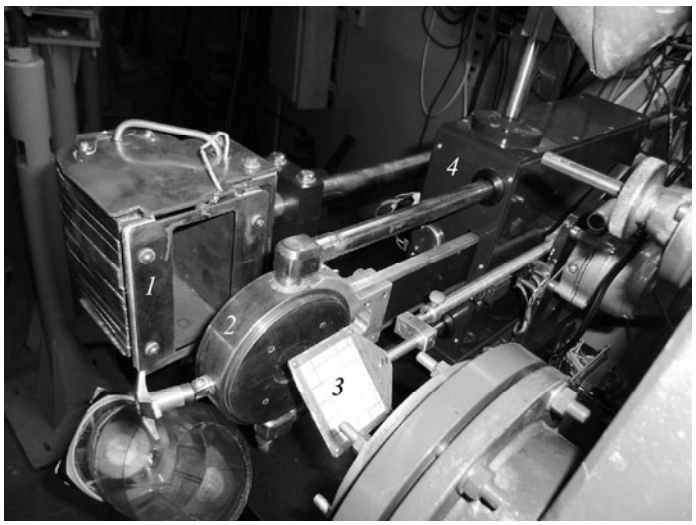


**Fig. 2.** Experimental plant (top view): *1* – Faraday cup, *2* – target facility, *3* – phosphor frame for television checking the beam profile, *4* – device for remote moving the target facility.

Electron beam from the outlet window of extractor got to the phosphor screen *3*, which was remotely put into the beam for observing the beam profile and then removed. Videocameras placed in several points allowed displaying the beam profile on the monitors in panel room in these points: outlet window of beam extractor, frame *3*, inlet to target facility, and the input window of the Faraday cup. The target facility *2* was remotely exposed to beam and removed from the beam when the direct measurement of beam current and position was needed. Movement of the target facility was realized by the device *4*, whose diagram is shown in Fig. 3.

The frame of the target facility is made of copper. For conversion of electrons to γ-quanta a 2 mm thick tantalum plate was placed at the input window. With the increase in converter thickness the number of output photons increases, but starting from a certain thickness it begins to decrease because of

prevalence of photon absorption in the converter material. In order to choose the optimum thickness of converter, we performed Monte-Carlo simulation of electron conversion at different thicknesses of the converter. The curve demonstrated in Fig. 4 shows that the optimum thickness for tantalum is 2 mm.

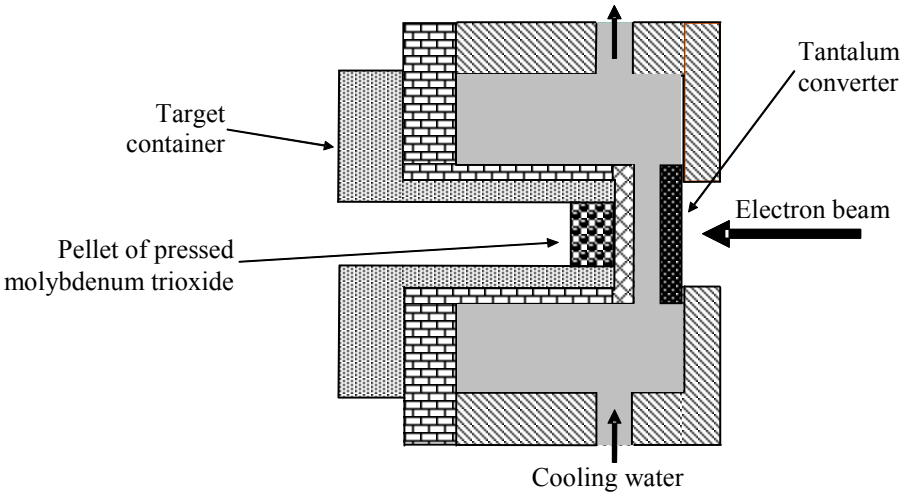


**Fig. 3.** Diagram of the target facility.

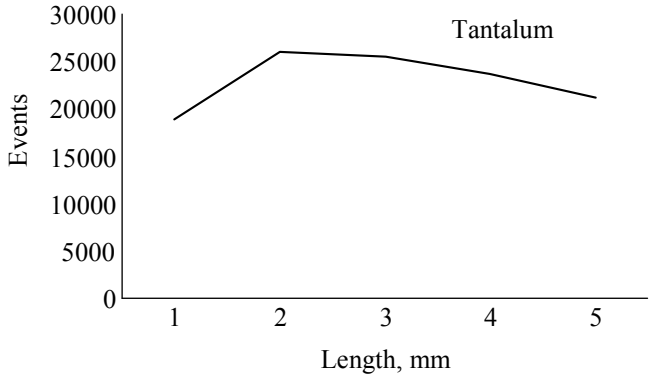


**Fig. 4.** Photon yield versus the converter thickness.

Performed calculations have shown that nearly 30% of the electron beam power is released in the converter material. The first studies were carried out at the electron energy $E_e = 20$ MeV and beam current $I_e = 5$ μA; in this case the beam power is $W = 100$ W and hence the heat power released in the converter amounts to 30 W. At the next stage of studies the electron energy was raised to $E_e = 40$ MeV at the intensity of $I_e = 10$ μA and the problem of water cooling of the target facility became urgent. The scheme of the water cooling system is given in Fig. 5.

For avoiding charge leakage from the Faraday cup only distilled water, i.e., water with high specific resistance, is used in the cooling system. Water, supplied by the pump *2*, passes through pipes of the Faraday cup *6*, then through the body of the target facility *7* (see Fig. 3), removing from them the heat released at absorption of a part of the beam energy, then it travels through the radiator *8* blown by the fans *9* where it transfers the heat to ambient air and returns to the storage tank *1*. Sensors of water level in the tank and of water pressure in the system (*3* and *5*, respectively) prevent emergency of water leakage from the system. Electronic thermometer records the water temperature in the tank.

Similar temperature sensors were placed in accelerating sections of linac, since the beam intensity depends strongly on the temperature of cooling water in the section.

As the whole equipment is placed in the ring and injector halls of the accelerator and operates on the background of strong electromagnetic noises, we have chosen the technique of temperature measurement which transforms proportionally the temperature reading (current in thermoresistor) into the pulse repetition rate, which is transferred via coaxial cable to the panel room and converted by a scaler in the

CAMAC standard. Similarly the current in the Faraday cup was measured. Readings of cooling water temperature and the beam current were displayed on the computer monitor and written in the file of checking the irradiation parameters. Processing and visualization of information was performed by means of LabView software.

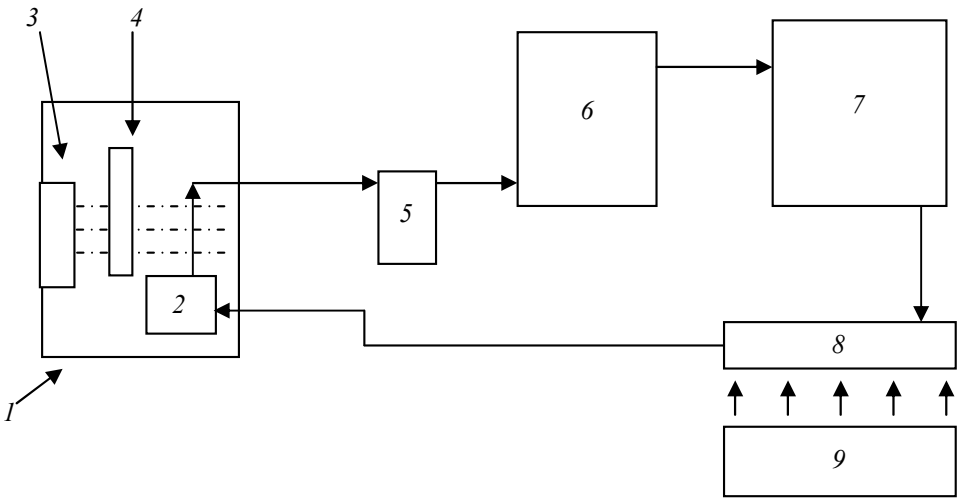


**Fig. 5.** Water cooling diagram: *1* – distilled water tank, *2* – pump, *3* – water-level sensor, *4* – electronic thermometer, *5* – electric water-pressure sensor, *6* – Faraday cup, *7* – target facility, *8* – radiator, *9* – fans.

## 4. CONTROL IRRADIATION

At the first stage we have chosen as irradiated material natural molybdenum trioxide $MoO_3$. The $^{100}$Mo concentration amounts in natural molybdenum to 9.1%. This means that in 3.6 g of irradiated material the content of $^{100}$Mo needed for the photonuclear reaction of obtaining technetium-99 is as small as 0.22 g. The molybdenum trioxide powder pressed into a pellet (see Fig. 3) was placed in the target unit at the distance of 4 mm from the tantalum radiator.

In 2010 a test irradiation session has been performed. The electron energy in the beam was $E_e = 40 \pm 5$ MeV, the beam intensity $I_e \approx 9.5$ μA. The session duration was ~100 hours. After completing irradiation the target material was left in the unit for 12 hours; in this time all short-lived isotopes decayed and the activity of target unit dropped down to safe values.

## 5. RESULTS OF IRRADIATION

Study of $^{99m}$Tc obtained in photonuclear reaction consists in measuring the spectrum of emission from irradiated material. Photon beam irradiation of $^{100}$Mo leads to formation of $^{99}$Mo which becomes $^{99m}$Tc with the decay period of 67 hours. In its turn, $^{99m}$Tc transforms into stable isotope $^{99}$Tc with the half-life of 6 hours. In this two-step process an emission line at $E_\gamma = 180$ keV is observed, which is characteristic for the transition of $^{99}$Mo to $^{99m}$Tc and has the strength 6% of the overall emission strength, and the line at $E_\gamma = 140$ keV, which is the fundamental emission line of $^{99m}$Tc. For detection of emission from irradiated material we used the detector, based on NaI(Tl) crystal, of the firm ORTEC-digiBASE, trademark 3M3/3-X, with the MAESTRO software attached.

Figure 6 shows the emission spectrum of irradiated material measured in 39 hours after completion of irradiation. The demonstrated spectrum displays clearly the peaks at the energies 140 and 180 keV, whose strengths corresponds to theoretical values. The fundamental peak of emission at 140 keV is approximated by the Gauss distribution (solid line) and the energy resolution of the detector at the energy of $E_\gamma = 140$ keV amounts to $\sigma = 5.9$ keV.

For studies of half-life of irradiated material measurements of spectrum were also performed in 26, 34, 49, 74, 84, 110, and 136 hours of completion of irradiation. Time dependence of the emission intensity is demonstrated in Fig. 7. Triangles stand for experimental values, while the dotted line describes the exponential behavior of the function $I = 16.2 \times 10^6 e^{-t/130.68}$. For this time dependence of the emission intensity the decay period is $T_{1/2} = 64.2$ hours. This result agrees well with the data for the half-life of $^{99}$Mo.

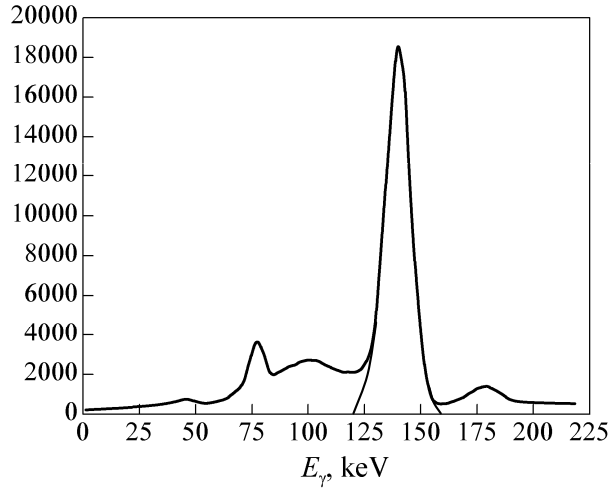


**Fig. 6.** Energy spectrum of irradiated material in 39 hours after irradiation.

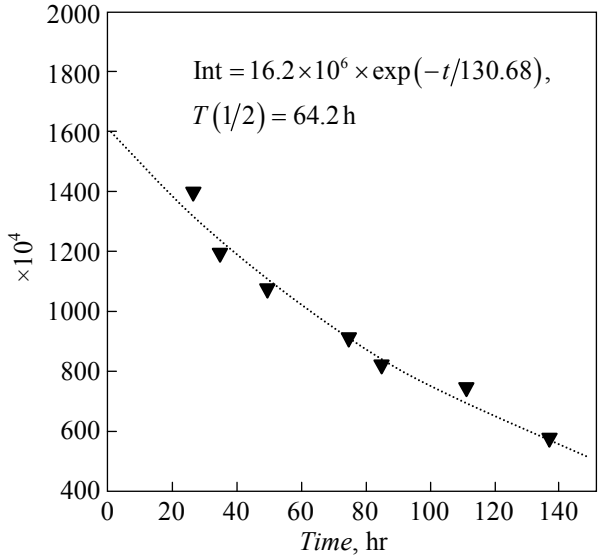


**Fig. 7.** Time dependence of the intensity of emission from the irradiated material.

One of important parameters in production of radioactive isotopes under the beam of electron accelerators is the produced activity reduced to unit mass of target material, unit beam current, and unit time of irradiation, i.e., Bq/mg μA hour. Available in literature data of experiments performed on different accelerators and under different conditions of irradiation are widespread from 90 to 3200 Bq/mg μA hour. The results obtained in our experiment yield the value 3000 Bq/mg μA hour which is near the upper limit of the range of world data.

The irradiated material was processed chemically in order to extract the final product $^{99m}$Tc.

## 6. CONCLUSION

On the electron linac LUE50 of Alikhanyan National Science Laboratory the full-scale experiment demonstrating the principal possibility of producing medicine-intended $^{99m}$Tc isotope has been performed for the first time. The experimental plant was elaborated and fabricated for studies of the possibility to develop technology of production of medicine isotopes on an electron linac.

As a result of experimental production a yield of $^{99m}$Tc isotope was obtained with the activity of 3000 Bq/mg μA hour. Efforts are made on creation of the industrial version of the plant for organization of full-scale production of isotopes on the basis of the developed technology.

## ACKNOWLEDGMENTS

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