ELECTRON ACCELERATOR BASED SOFT TECHNOLOGY FOR MEDICAL IMAGING ISOTOPES PRODUCTION*

N.P. Dikiy, A.N. Dovbnya, V.L. Uvarov†
NSC "KIPT", Kharkov, 61108, Ukraine

Abstract

Nowadays ^{99m}Tc (the daughter nucleus of ^{99}Mo) is the main isotope used for medical imaging. Its consumption reaches about 150000 Ci yearly and increases continuously [1]. Traditional technologies for ^{99}Mo production are based on research fission reactors and are accompanied by the large amount of radioactive waste. Besides, for a calibration of scintillation cameras the isotope ^{57}Co is commonly used because of the proximity of its gammas energy (122 keV) to ^{99m}Tc one but greater lifetime (270 days). Thus, the elaboration of soft technology for these isotopes seems to be urgent. To obtain ^{99}Mo the authors proposed the irradiation of liquid target (the natural molybdenum solution in the alkali water solution) with braking photons of high-current electron linac. ^{57}Co is produced in its turn by irradiation of the Ni plate cooled with water. The geometry and treatment conditions for each targetry system were previously optimized by means of computer analysis. The results of simulation as well as of experimental isotope production using proposed technology are presented.

1 INTRODUCTION

The authors of papers [2-5] have demonstrated a possibility, in principle, of ^{99}Mo producing in the bremsstrahlung field of the electron accelerator. At the same time, the transition to this technology of ^{99m}Tc production on an industrial scale calls for the solution of a number of physical and technical problems.

The application of natural molybdenum appears more reasonable for the purpose. In this case, considering that in parallel with bremsstrahlung, a flux of photoneutrons also comes from the converter, the ^{99}Mo generation in the field of secondary emission of accelerated electrons can proceed in two channels: with the ^{100}Mo isotope (making up 9.63% of natural molybdenum) in the basic reaction ^{100}Mo(γ,n)^{99}Mo, and with the ^{98}Mo (24.13%) in the reaction ^{98}Mo(n, γ)^{99} Mo. Both the metallic Mo and the concentrated solution of Mo can serve as a target material.

At the initial stage, each version of the target setup was investigated by method of computer simulation using the GEANT code supplemented by the data on excitation functions of the corresponding nuclear reactions. The simulation has revealed that with an increase in the electron energy from 20 to 40 MeV the yield of ^{99}Mo normalized to 1 kW beam power increases nearly twice.

2 EXPERIMENT

For experimental study into the regularities of ^{99}Mo production, a target unit prototype was designed (Fig. 1).

The device includes the flowing water-cooled bremsstrahlung converter. Immediately behind the converter, a cylindrical vessel (V1), 80 mm in diameter and 40 mm in height, was placed, axially symmetric about the electron beam. Next to it, there were three vessels in succession: one (V2), 32 mm in diameter and 60 mm in height, and the other two vessels (V3 and V4), 100 mm in height.

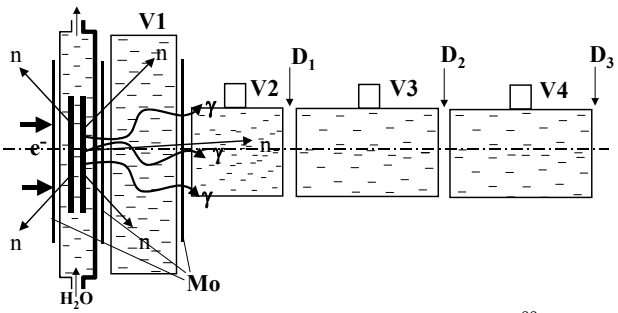

Fig. 1: Scheme of experiment to investigate ^{99}Mo production using the electron accelerator

The vessels were filled with a water solution of NaOH with natural Mo dissolved in it to a concentration of 142 mg/ml. The usage of NaOH as a solvent of molybdenum is of advantage, since Na has a large cross-section of elastic neutron scattering (600 barn for 2.8 keV neutron energy). The presence of water solution also favors the efficient slowing down of neutrons. As a result, there arise the conditions for increasing the yield of the ^{98}Mo(n, γ) reaction with natural molybdenum as a basis.

In the spacings between the target unit vessels there were placed Mo foils and technological dosimeters D_1 ... D_3. This made it possible to determine the divergence of the braking photon beam and its intensity distribution along the axis of the target setup, and also to compare the ^{99}Mo yield for both the solution and the solid target.

The experiments were performed using the LU-20 accelerator [6] in the regime of dose accumulation. The conditions of target treatment are given in Table 1.

Table 1. Target irradiation conditions (Na_2MO_4 solution)

	$\bar{E}_{e^-}$, MeV	d_{e^-}*, mm	Dose rate, kGy/h.		
			D_1	D_2	D_3
1	24	13	40.8	12.3	6.8
2	28	12	60.4	22.8	12.7
3	34	10	91.2	33.1	17.4

* at a half-height of current density distribution

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† uvarov@kipt.kharkov.ua

After a recurrent exposure, a sample of irradiated solution, 1 to 1.5 ml in volume, was taken from each vessel for the analysis of isotopic composition by means of the Ge(Li) detector.

3 RESULTS

Fig. 2. shows the characteristic spectrum of the irradiated solution Na_2MoO_4, and Table 2 lists the measured values of volumic activity for the basic isotopes produced at each regime of irradiation for 1 hour at an average beam current of 10 μA.

Simultaneously with irradiation of solutions, 0.1 mm thick natural molybdenum foils, placed ahead of and behind the converter, and also behind the vessel V_1 (see Fig. 1), were exposed to radiation. After irradiation, the surface activity distribution of ^{99m}Tc along the Eγ = 140 keV line was measured by means of a collimated γ-spectrometer with the CdZnTe-based detector.

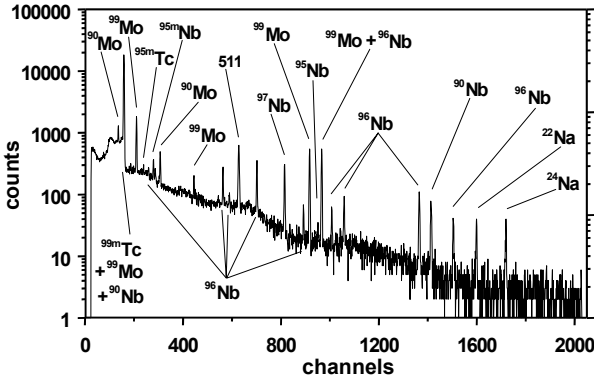

Fig. 2: Gamma-spectrum of the Na_2MoO_4 solution

The activity and isotopic composition of foils were also determined with the Ge(Li) detector.

Table 2: Volumic activity (A) of isotopes in the target vessels

E_e, MeV	№vessel	A, nCi/ml					A, Bq/ml
		^{90}Mo	^{96}Nb	^{99}Mo	^{22}Na	^{24}Na	^{99}Mo
24	1	1.58	6.67	156.12	2.46	44.05	5777
	2	3.52	11.78	183.59	3.32	9.71	6793
	3	1.60	5.23	77.30	2.11	1.57	2860
	4	0.75	2.45	30.42	1.94	0.86	1126
28	1	8.84	30.96	264.43	3.19	88.12	9784
	2	20.47	46.76	371.45	4.87	23.44	13744
	3	9.58	17.66	141.35	1.23	3.07	5230
	4	4.59	6.84	57.49	0.29	1.51	2127
34	1	48.72	52.52	452.56	4.44	106.66	16745
	2	76.16	54.72	404.72	6.74	18.61	14975
	3	38.36	30.49	173.37	3.68	5.54	6415
	4	16.13	13.85	62.61	1.45	3.11	2317

Thus, the ^{99}Mo activity of the foil fragment in the form of a circle, 40 mm in diameter, was found to increase near the distribution maximum from 10.6 μCi (24 MeV) to 37.8 μCi (34 MeV) for the samples placed immediately behind the converter.

4 DISCUSSION

The measurements of the geometry of bremsstrahlung field have shown that it is characterized by a substantial nonuniformity. This nonuniformity is due to both the divergence of the braking photon flux emitted from the converter and its absorption in the solution under irradiation. The divergence angle (measured at the half-height of the induced surface activity distribution of Mo foils) varies in the range from 12.6° for (24 MeV) to 7.9° (34 MeV).

The geometry chosen for the target setup permits the determination of a relative contribution of (γ,n) and (n, γ) channels to the production of ^{99}Mo. Really, the dimensions of vessel V_1 are chosen such that it fully intercepts the photon flux and simultaneously provides the moderation of photoneutrons to resonant capture energies in the ^{98}Mo(n, γ)^{99}Mo reaction. In vessels V_3, V_4 the ^{99}Mo isotope is generated nearly fully at the expense of the (γ,n) channel, only. So, the increase in the volumic activity of the solution, observed in the second vessel, as compared to the first vessel is due to the fact that the isotope production in V_1 via the photonuclear channel occurs within the bremsstrahlung cone, i.e., only in the central part of the vessel.

To separate the contributions of different channels to ^{99}Mo generation, one can use the reactions in the vessels taking place only through the photonuclear channel. So, for the Na_2MoO_4 solution, these are ^{23}Na(γ,n)^{22}Na (Q = - 12.4 MeV) and ^{92}Mo(γ, 2n)^{90}Mo (Q = - 13.1 MeV). The yield via the (n, γ) channel can be investigated using the ^{23}Na(n, γ)^{24}Na reaction. The data obtained with the use of these reactions on the relative yield of ^{99}Mo in (γ,n) and (n, γ) channels - C_γ and C_n , $C_\gamma + C_n = 1$, are listed in Table 3.

Table 3: Relative yield of ^{99}Mo in (γ,n) and (n, γ) channels

E_{e-}	№$_{vess.}$	c_γ	c_n
24 MeV	1	0.73	0.27
	2	0.94	0.06
28 MeV	1	0.58	0.42
	2	0.89	0.11
34 MeV	1	0.61	0.39
	2	0.93	0.07

The ratio of ^{99}Mo concentrations in V_3 and V_4 correlates with the photon intensity (measured by the dosimeters D_2 and D_3), and this confirms the manifestation of mainly the (γ,n) channel of ^{99}Mo production in these vessels.

The foil activity data are in agreement with the results of computer simulation and the data on the volumic activity of solution samples (normalized to Mo content in the solution). This points to a small contribution of the solvent to the attenuation of the flux of braking photons, and makes it possible to estimate the ratio of the volumic activity of the solution $(dA/dV)_{liq}$ and the specific activity of the solid target $(dA/dm)_{sol}$ by the formula

$$\left(\frac{dA}{dV}\right)_{liq} = C_{Mo} \cdot \rho_{liq} \cdot \left(\frac{\partial A}{\partial m}\right)_{sol},$$

where C_{Mo} is the weight concentration of molybdenum in the solution, ρ is its density.

For formula it follows, in particular, that the maximum volumic activity of natural molybdenum solution (close to the converter) reaches ~100 mCi/ml and falls off with distance R away from it ~kR^{-2}, where k is the coefficient dependent on the transverse size of the electron flux, the electron energy, converter and solution compositions.

5 CONCLUSIONS

The undertaken investigations have demonstrated that each of the target versions (solid and liquid) to produce ^{99}Mo at the electron accelerator has its advantages and disadvantages. Thus, the liquid target based on Mo-saturated alkaline solution makes, in principle, it possible, owing to moderation of photoneutrons generated in the converter, to involve in addition the (n, γ) channel for ^{99}Mo generation with the use of natural molybdenum. Besides, with a further extraction of ^{99m}Tc from the irradiated solution there is no need for an additional procedure of metallic molybdenum dissolving. If the isotopically pure ^{100}Mo is used, the liquid target version readily permits the realization of the loop-type technology providing a high ^{99}Mo yield through the minimum number of radiochemical procedures and a cyclic circulation of the solution between the accelerator and the technetium extractor [7].

At the same time, the difficulties in the realization of this version are specified by a limited volume of the existing ^{99m}Tc extractors (generally, no more than 200 ml), whence the requirement of a high volumic activity of the solution under treatment follows (as a rule, no less than 10 mCi/ml. in ^{99}Mo) [8]. With the usage of natural molybdenum, this requirement is difficult to meet. The irradiation of liquid target in the field of a high-current accelerator also involves the problem of removing a power of about a few kW from a small-volume solution with keeping the chemical stability of the latter.

The solid target version is associated, in its turn, with the necessity of concentrating the electron beam of ≥ 10 kW power within the region of several centimeters, including the bremsstrahlung converter and the target itself with the provision of their continuous efficient cooling. This version appears more preferential in operation with natural molybdenum. In any case, one must solve the problem ejecting a ≥ 10 kW direct (unscanned) electron beam from the accelerator, the beam cross-section being no more than 1 cm.

From the results of computer analysis and the experimental data it follows also that up to 15 standard ^{57}Co sources, each having an activity of 2 mCi, can be produced for 24 hours operation with the use of a powerful electron accelerator (30 MeV, 500 μA) when optimized geometry of the target setup [9]. With this technology, the problems of beam ejection from the accelerator and target cooling are not so pressing.

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