ECOLOGICAL ASPECTS OF ^{99}Mo/^{99m}Tc PRODUCTION AT AN ELECTRON ACCELERATOR

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The present report deals with experimental investigations into possible processes of accumulation of radioactive by-products (RBPs) during ^{99}Mo/^{99m}Tc production at an electron linac. Foils from natural molybdenum (a target) and a number of typical target setup materials (Al, Ti, Cu, Ta, Ni and stainless steel) were chosen as an object of investigations. The generation of radionuclides in the samples was performed in identical conditions under secondary radiation (bremsstrahlung + photoneutrons) of the linac LU-20 in a typical range of electron energies from 24 to 34 MeV.

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1. INTRODUCTION

In modern practice, more than 8500 medical institutions throughout the world are exploiting up to 20 000 gamma-chambers for diagnostics of diseases by the scintigraphy method. More than 70% of the procedures are carried out with the use of ^{99m}Tc (daughter product of the ^{99}Mo isotope) as a radioactive tracer. The main body of ^{99}Mo (up to 200 000 Ci per year with a steady tendency to increase) is produced in nuclear reactors with the use of ^{235}U fission reactions. In this case, only 6% of the reactions are accompanied by the production of ^{99}Mo. As a result, this method gives up to 50 Ci of radioactive waste (RAW) per 1 Ci of ^{99m}Tc, including long-lived RBPs [2]. In view of the annual growth of ^{99}Mo production and the absence of immediate prospect of replacing ^{99m}Tc by another diagnostic radionuclide, there arises the necessity of creating a new process of ^{99}Mo/^{99m}Tc production with a less ecological load. This technology based on the use of a high-current electron accelerator has been developed (see ref. [3]) and is now being promoted at KIPT.

2. SOURCES OF RBPs

By new technology, the converter, the target substance comprising ^{100}Mo, and also the devices which accommodate them serve as RBPs sources. The composition of RBPs and the rate of their generation are determined by the energy and value of the electron flow, by the isotopic composition of the mentioned structures, and also by their geometry with respect to the secondary radiation field. In this case, only those active products that are not retrapped into the production cycle can be considered as RAW. These are the waste resulting from the process of target treatment, ^{99m}Tc discharges (for the most part, liquid), and also the spent constructives (solid-state RAW).

The technology developed here provides for application of different-type solid targets (metallic Mo or MoO_3 oxide) or liquid targets based on water solutions of molybdenum acid salts, with molybdenum being of both natural composition and enriched in the ^{100}Mo isotope. In view of relatively low total (up to 3 Ci) and specific (≤0.1 Ci/g) activities of the irradiated target, the methyl ethyl ketone extraction of ^{99m}Tc (e.g., see refs. [4,5]) has appeared the most acceptable method for extraction of ^{99m}Tc. In the natural molybdenum case as a target, the spent water phase (at the end of the week-term technological cycle) is the main type of waste.

3. LIQUID TARGET

As it has become obvious from the studies, the liquid target variant is most easily producible [6]. To investigate regularities in the liquid-phase generation of ^{99}Mo and RBPs, a target device prototype was developed (Fig.1).

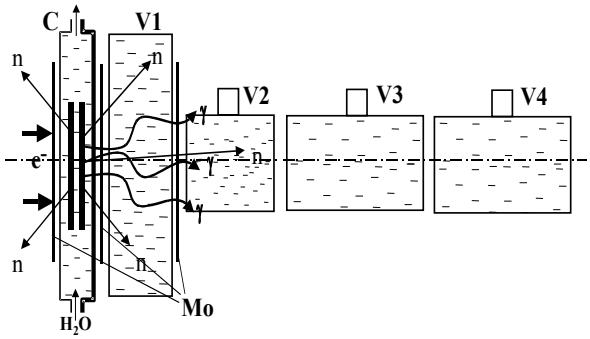

Fig. 1. Schematic of experiment to investigate the production of ^{99}Mo and RBPs in a liquid target

It includes a flowing water-cooled bremsstrahlung converter C. Immediately behind the converter there is a cylindrical vessel (V1), 80 mm in diameter and 40 mm in height, which is axially symmetric to the electron beam. Then sequentially follow a vessel, 32 mm in diameter and 60 mm high (V2), and two vessels (V3 and V4), 100 mm in height.

After irradiation of the target at given conditions, 1 to 2 ml samples of solution were taken from the vessels and were directed for the analysis of induced activity by means of the Ge(Li) detector. Tables 1 and 2 give the results of the analysis of partial activity of targets based on water solutions of Na_2MoO_4 salts (comprising 142 mg/ml of natural Mo) and K_2MoO_4 (208 mg/ml).

4. SOLID TARGET AND CONSTRUCTIVES

To investigate the processes of RBPs generation in structural materials of the target facility, samples of most typical materials (Al, Ti, Cu, Ta, Ni, Mo and stainless steel) were chosen in the form of foils. The foils were assembled as a sandwich and placed directly behind the bremsstrahlung converter.

Table 1. Volumic activity (A) of isotopes in liquid target vessels (Na_2MoO_4, 1 hour, 10 μA)

E_0, MeV	№ v.	A, nCi/ml				
		^{90}Mo	^{96}Nb	^{99}Mo	^{22}Na	^{24}Na
24	1	1.58	6.67	156.12	2.46	44.05
	2	3.52	11.78	183.59	3.32	9.71
	3	1.60	5.23	77.30	2.11	1.57
	4	0.75	2.45	30.42	1.94	0.86
28	1	8.84	30.96	264.43	3.19	88.12
	2	20.47	46.76	371.45	4.87	23.44
	3	9.58	17.66	141.35	1.23	3.07
	4	4.59	6.84	57.49	0.29	1.51
34	1	48.72	52.52	452.56	4.44	106.66
	2	76.16	54.72	404.72	6.74	18.61
	3	38.36	30.49	173.37	3.68	5.54
	4	16.13	13.85	62.61	1.45	3.11

Table 2. Volumic activity of isotopes in target vessels (K_2MoO_4)

E_0, MeV	№ v.	A, nCi/ml		
		^{90}Mo	^{96}Nb	^{99}Mo
24	1	-	13,18	293,93
	2	-	23,66	386,40
	3	-	9,80	139,43
	4	-	4,25	61,22
28	1	-	40,89	458,00
	2	-	79,30	659,74
	3	-	26,32	246,68
	4	-	10,68	91,64
34	1	86,19	93,18	623,81
	2	211,33	119,15	947,52
	3	98,33	62,40	346,71
	4	92,67	24,70	134,55

The accelerator LU-20 was operated in the mode of direct (unscanned) beam, the parameters of which are similar to the parameters described above. After irradiation the foils were forwarded for the measurement of their partial activity by means of the Ge(Li) spectrometer. Table 3 lists the measured values of RBPs generation in the chosen set of structural materials and in natural Mo.

The columns of the table contain the following information: 1 - modes of target irradiation at accelerated electron energies of 24 MeV (mode I), 28 MeV (mode II) and 34 (mode III), the average accelerator current being 10 μA, 2 - basic reaction, 3 - half-life $T_{1/2}$ of the radionuclide produced (days), 4 - specific partial activity of radionuclide in the sample, 5 - $^{i}A_{max}$ is the estimated maximum activity of the i- radionuclide in the technological cycle (300 μA, 24 hours), 6 - $^{i}A_{max}/\ ^{Mo\text{-}99}A_{max}$ is the ratio of $^{i}A_{max}$ to the maximum activity of generated Mo-99.

The maximum activity of RBPs in the technological cycle, $^{i}A_{max}$, was estimated in the assumption that the given material is concentrated in the infinite layer of thickness $(\mu\rho)^{-1}$, where μ is the attenuation coefficient of braking photons in the material, the photon energy being in the region of photonuclear reaction, ρ is the density of material.

For mode III (its conditions in the electron energy are the closest to the ones realized in the technological process), Table 4 gives both the calculated values of RBPs activity accounted for by 5 Ci of the generated ^{99}Mo immediately after completion of target irradiation, and the data obtained from RBPs residual activity (total A_i and gamma A_i^{γ}) at the end of the 5-day technological cycle.

Table 3. Generation of basic radionuclides in structural elements of the target facility

1	2	3	4(kBq /g)	5(Ci)	6(Ci/ /Ci)
I	^{100}Mo(γ,n) $\rightarrow^{99}$Mo	2.75	54,4	3,24	1
	^{48}Ti(γ,p) ^{47}Sc	3.35	19,5	1,27	0,39
	^{58}Ni(γ,n)^{57}Ni ^{58}Ni(γ,p) $\rightarrow^{57}$Co	270.9	102,3	7,48	2,3
	^{181}Ta(n,γ) $\rightarrow^{182}$Ta	115.0	0,260	0,00586	0,0018
	^{65}Cu(γ,n) $\rightarrow^{64}$Cu	0.53	644	38,7	11,9
II	^{100}Mo(γ,n) $\rightarrow^{99}$Mo	2.75	80,8	4,53	1
	^{48}Ti(γ,p) $\rightarrow^{47}$Sc	3.35	54,8	3,68	0,81
	^{58}Ni(γ,n)^{57}Ni ^{58}Ni(γ,p) $\rightarrow^{57}$Co	270.9	204,7	14,5	3,28
	^{181}Ta(n,γ) $\rightarrow^{182}$Ta	115.0	0,357	0,00838	0,00184
	^{65}Cu(γ,n) $\rightarrow^{64}$Cu	0.53	1195	75,5	16,65
III	^{100}Mo(γ,n) $\rightarrow^{99}$Mo	2.75	103,0	5,87	1
	^{48}Ti(γ,p) $\rightarrow^{47}$Sc	3.35	103,6	7,04	1,2
	^{58}Ni(γ,n)^{57}Ni ^{58}Ni(γ,p) $\rightarrow^{57}$Co	270.9	298,3	21,4	3,65
	^{181}Ta(n,γ) $\rightarrow^{182}$Ta	115.0	0,457	0,01089	0,00185
	^{65}Cu(γ,n) $\rightarrow^{64}$Cu	0.53	1545	94,5	16,1

5. DISCUSSION

The analysis of the present data shows that the equivalent dose rate (EDR) at the maximum of its distribution on the surface of the samples under study is approximately proportional to the activity of radionuclides produced in them as a result of irradiation, and increases with electron energy in all cases. The data make it possible to make the proper choice for the target facility components. The last ones should give the minimum contribution to the total EDR of the radiated target setup as compared with the radiation of the Mo target itself, and also to produce mainly short-lived radionuclides.

The identical yield ratios of the ^{99}Mo, ^{57}Ni and ^{47}Sc isotopes in the samples from natural Mo, Ni and Ti for different modes of irradiation have appeared the same as in the stainless steel samples comprising similar elements. ^{57}Ni is found to be the main source of induced activity in stainless steel.

Table 4. Activity of basic RBPs in the technological cycle

Isotop	$A_i/5Ci_{Mo-99}$, mCi	A_i after 5 days,mCi	A^{γ}_i after5 days,mCi
^{95m}Nb	2600	651	635
^{95}Nb	3.1	26.9	26.9
^{88}Zr	3.15	2.97	2.88
^{92m}Nb	8.0	4.9	4.9
^{96}Nb	730	4.2	~4.2
^{91m}Nb	160	148	4.5

The data on the radionuclide yields in different materials ($^iA_{max}$) at a target thickness equal to the mean path of photons having the energy in the excitation region of the corresponding photonuclear reaction should be considered as certain ultimate estimates. In particular, at a material thickness $d << (\mu\rho)^{-1}$ the produced activity is $Ai \sim d_i \cdot \mu \cdot \rho \cdot {}^iA_{max} << {}^iA_{max.}$ Besides, if the given material is located in the target facility behind the Mo target of thickness $(\mu\rho)^{-1}$, then the maximum yield of side radionuclides is reduced by a factor of about ***e***.

The measurement of surface activity distribution of foils by the collimated radiometry method makes it possible to determine geometrical characteristics of the region of generation of different radionuclides and thus to characterize the beam quality for the isotope production process, and also to optimize the geometry and the elemental composition of the target facility.

6. CONCLUSION

Methods have been developed and studies have been made into the generation of radionuclides in targets and structural materials at production of ^{99}Mo/^{99m}Tc on the electron accelerator. The photonuclear channel was the main channel of radionuclide generation. The effect of photoneutrons may manifest itself only in the reactions ^{98}Mo(n,γ)^{99}Mo, ^{92}Mo(n,p)^{92m}Nb, ^{181}Ta(n,γ)^{182}Ta and ^{23}Na(n,γ)^{24}Na.

Among the structural elements under study, ^{54}Mn has the longest half-life (312.3 days). As regards the activity produced, the use of such materials as Ni, Cu and stainless steel is found to be the least desirable.

The data obtained on the yields of side radionuclides with respect to the yield of ^{99}Mo in the case of natural Mo-based target are independent of the phase composition of the target (metal, solution or trioxide). These data are required for the calculation of biological shield of the "hot" chamber, which accommodates the extraction generator of ^{99m}Tc, and also, the vessel-collector for liquid radioactive waste.

The results of determining the radionuclide composition of irradiated materials and their activity with due regard for possible optimization of target facility materials and geometry give evidence for considerable advantages of the proposed technology of ^{99}Mo/^{99m}Tc production in relation to the ecology load.

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ЭКОЛОГИЧЕСКИЕ АСПЕКТЫ ПРОИЗВОДСТВА ^{99}Mo/^{99m}Tc НА УСКОРИТЕЛЕ ЭЛЕКТРОНОВ

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Изложены результаты экспериментального исследования возможных процессов наработки побочных радионуклидов при производстве ^{99}Mo/^{99m}Tc на ускорителе электронов. В качестве объекта исследования отобраны фольги из природного молибдена (мишень) и набора характерных конструкционных материалов (Al, Ti, Cu, Ta, Ni, а также нержавеющая сталь). Генерация радионуклидов в образцах производилась в идентичных условиях в поле вторичного излучения (тормозное+фотонейтроны) ускорителя ЛУ-20 в характерном диапазоне значений энергии электронов 24...34 МэВ.

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ЕКОЛОГІЧНІ АСПЕКТИ ВИРОБНИЦТВА ^{99}Mo/^{99m}Tc НА ПРИСКОРЮВАЧІ ЕЛЕКТРОНІВ

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Викладені результати експериментального дослідження можливих процесів наробітку побічних радіонуклідів при виробництві ^{99}Mo/^{99m}Tc на прискорювачі електронів. Як об'єкт дослідження відібрані фольги з природного молібдену (мішень) і набору характерних конструкційних матеріалів (Al, Ti, Cu, Ta, Ni, а також нержавіюча сталь). Генерація радіонуклідів у зразках виконувалося в ідентичних умовах у полі вторинного випромінювання (гальмівне+фотонейтрони) прискорювача ЛП-20 у характерному діапазоні значень енергії електронів 24...34 МеВ.

Робота виконана за підтримкою НТЦУ, контракт № 2185