ESTIMATION OF RADIATION RISKS UNDER PHOTONUCLEAR PRODUCTION OF ^{67}Cu AND ^{99}Mo ISOTOPES

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The activated bremsstrahlung converter and isotopic target are the main sources of radiation hazard in the isotope production at electron accelerators. In experiments at the NSC KIPT accelerators KUT-30 and LU-40m, intended to produce ^{99}Mo and ^{67}Cu isotopes in the targets based on natural Mo and Zn, the output devices were exposed to radiation. Induced activities of the tantalum converter, of targets and cooling water were measured. The exposure dose rates provided by each element and the activity decrease after the exposure were determined.

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1. INTRODUCTION

The ^{99}Mo and ^{67}Cu isotopes range among the most demandable in nuclear medicine. Thus, the world production of ^{99}Mo, which is the generator of the basic diagnostic radionuclide ^{99m}Tc, currently exceeds 300 000 Ci per annum and is practically all-based on ^{235}U fission in nuclear reactors [1]. This method provides the high-yield production of ^{99}Mo and a high specific activity (up to 10^4 Ci/g), but leads to a great amount of radioactive waste [2]. The technology using the radiative capture reaction ^{98}Mo(n,γ)^{99}Mo appears to be safer [3]. However, in this case the yield and specific activity of the target turn out to be a few orders of magnitude lower, and besides, the method also calls for the use of a nuclear reactor. Taking into account the ecological effects of reactor technologies as well as the problem of fissile material nonproliferation, it becomes currently central to create alternative methods of ^{99}Mo production, one of them being the photonuclear method [4].

In turn, the ^{67}Cu isotope is considered as a most promising beta-emitter for radioimmunotherapy [5]. At present it is mainly produced at high-current proton accelerators through the use of the ^{68}Zn(p,2p)^{67}Cu reaction. The realization of the process also involves the production of a considerable quantity of "hot" impurities [6].

As preliminary studies have shown, the production of the mentioned radionuclides by the ^{100}Mo(γ,n)^{99}Mo and ^{68}Zn(γ,p)^{67}Cu reactions can provide the yield of desired isotopes, which is comparable with that provided by other technologies, but with a substantially less quantity of radioactive waste produced [7, 8]. In this case, the high-current electron accelerator is an essentially more reliable, safe and inexpensive device than the nuclear reactor or the heavy-particle accelerator with the same beam energy and intensity.

Here we analyze the main sources of radiation hazards and estimate their level at different stages of photonuclear production of ^{99}Mo and ^{67}Cu isotopes.

2. RADIATION SOURCES AT PHOTONUCLEAR PRODUCTION OF ISOTOPES

The basis for the technology under consideration is the activation of the isotopic target by means of high-energy bremsstrahlung resulting from accelerated electron beam conversion. The process can be realized directly in the target itself or in a separate device–converter placed between the exit window of the accelerator and the target. The last variant is more preferable, because it provides a high specific activity of the target, and also makes it possible to reduce the radiation power absorbed in the target [9].

The converter generally corresponds to one or several plates, which are made from the material having a high atomic number (e.g., Ta, W or Pb) and are cooled with a flowing water [10]. So, in the general case, the output devices of the electron accelerator operated in the mode of isotope production include the exit window with tandem converter and target units behind it.

The exit window of the high-current electron accelerator usually consists of two thin foils (Al or Ti), between which water is circulated. The contribution of this unit to the radiation hazard can be neglected in the estimation.

On interaction of accelerated electrons with the converter, owing to (γ,n) reactions, apart from bremsstrahlung, a quasi-isotropic photoneutron flux is generated. For example, the main reactions that occur in the W converter are ^{182}W(γ,n)^{181}W and ^{186}W(γ,n)^{185}W, in the Ta converter this is the reaction ^{181}Ta(γ,n)^{180}Ta, etc. In other words, in the process of target activation the converter unit is the source of mixed high-intensity γ,n-radiation [11]. On completion of the activation, the radiation hazard is mainly contributed by the residual activity of the converter and the target. In this case, their dose rates and the activity decrease are determined by the activation mode and the element composition of the devices.

3. DOSE CHARACTERISTICS OF RESIDUAL ACTIVITY OF OUTPUT ^{67}Cu-PRODUCTION DEVICES

3.1. EXPERIMENTAL CONDITIONS

For experimental estimation of the radiation background at photonuclear production of the ^{67}Cu isotope, a target device shown in Fig.1 has been used. It includes a stainless steel casing with a thin entrance window. Inside the casing, there is a four-plate Ta converter, each plate being 1 mm thick. In the process of irradiation the

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converter is cooled by water (the total thickness of water spacings is 5 mm). Behind the converter, there is a Ti capsule, ~ 52.3 g in weight, which accommodates the isotopic target. The capsule is sealed with a stainless steel cap, ~ 10.5 g in weight. Directly in front of the converter and immediately behind it there were placed two sets of Ø40mm foils (Mo, Ni, Sn) to measure the electron beam profile (Mo^1, Ni^1, Sn^1) and the bremsstrahlung profile (Mo^2, Ni^2, Sn^2) by the photonuclear converter technique [12].

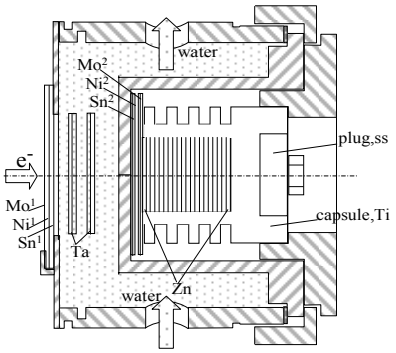

Fig.1. Target device design

To determine the contribution to the radiation background from each target element (Zn, Ti, stainless steel), the following technique was used. Two similar Ti capsules were taken. The first capsule housed 18 Zn discs (Ø=20 mm, h=1 mm), and also 3 molybdenum foils (Ø=20 mm, δ=0.1 mm) at the beginning of the assembly (Mo-1), in its middle (Mo-10) and at the end (Mo-18). This approach enables one to determine the "utilization factor" of bremsstrahlung that is in contact with the Mo foil and Zn disc having the number *i* according to the activity ratio $A_{Mo\text{-}i}/A_{Mo2}$ (see Fig.1).

In the second capsule, an Al cylinder (Ø=20 mm, h=19 mm) was put instead of zinc. Under the same exposure conditions of each capsule, this permitted us to estimate the zinc contribution to the residual activity of both the target and the casing for two variants (Ti or Al) of target casing fabrication. The targets were activated at the accelerator LU-40m operated in the 56 MeV, 5 μA mode. Each capsule was irradiated for 1 hour.

3.2. MEASUREMENTS AND RESULTS

Thirty minutes after the end of irradiation of the first capsule (with zinc) the exposure dose rate (EDR) of the residual activity of the target device was measured. At a distance of 0.5 m the EDR was measured to be 200 mR/h, while the EDR of the capsule with zinc, taken out of the target device, was found to be 189 mR/h at the same distance. Thus, the capsule with zinc appears the main dose contributor of the target device.

In the following 7 days the EDR_L of the capsule with zinc was measured at distances of L cm that provided the correspondence of the measured dose rate to the operative range of the dosimeter-radiometer. The measured values were reduced to the standard distance L=10 cm by the formula:

$$EDR_{10} = EDR_L(L/10)^2 ,$$

and were also normalized to the average beam current value of 1 μA and the exposure time of one hour (Fig.2). After irradiation of the second capsule at the same conditions, it was extracted from the target device and disassembled into components (Ti casing, cap and Al cylinder). Then, with the use of the same technique, dose rate measurements were performed for each component (Fig.3).

The comparison between the data in Figs.2 and 3 shows that immediately after EOB the capsule with zinc mainly contributes to the EDR of the target device.

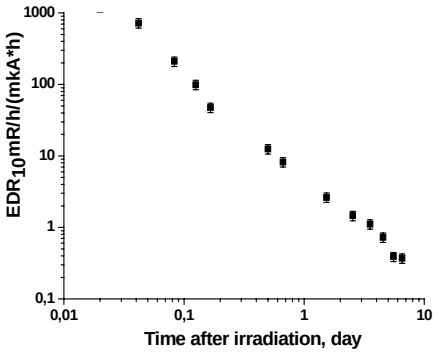

Fig.2. Exposure dose rate (EDR_{10}) of the capsule with zinc versus time after EOB (day)

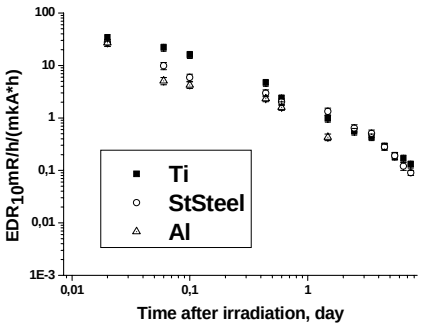

Fig.3. Exposure dose rates (EDR_{10}) of capsule components versus time after EOB (day)

In this case, the radiation background within the first 2-3 hours after the irradiation is determined by comparatively short-lived isotopes Zn-63 and Zn-62 having the half-lives of 38 min and 9.3 hour, respectively. During the first day the main activity of the target is created by Zn-69m and Cu-67 isotopes; then in the following ~ 7 days, it is Cu-67 and Zn-65 that present the main dose-producing factor, and later - only Zn-65 ($T_{1/2}$=243 days).

The EDR is contributed by Sc-46, Sc-47 and Sc-48 isotopes (for the Ti capsule), Ni-56, Ni-57, Co-57 and Co-58 isotopes (for the stainless steel cap), and by isotopes produced on the impurities (Si, Mg, etc.) for the Al target. If within 2 or 3 hours after irradiation the EDR of the Zn-filled capsule is more than order of magnitude higher than the total EDR of other capsule elements, then in a day of "cooling", their EDR values appear comparable.

The present data enable one to choose the optimum regime of irradiated target handling, and also to predict dose characteristics of a similar target device activated at other conditions. D.Ehst has independently calculated the EDR_{10} variations after irradiation of a similar Zn-containing capsule and its separate components for electron energy of 55 MeV and compared the results with the experimental data (see Figs.2 and 3) reduced to a beam current of 31.8 μA (this corresponding to the RPI (USA) accelerator conditions) and 1 hour of exposure. The obtained results are presented in Fig.4.

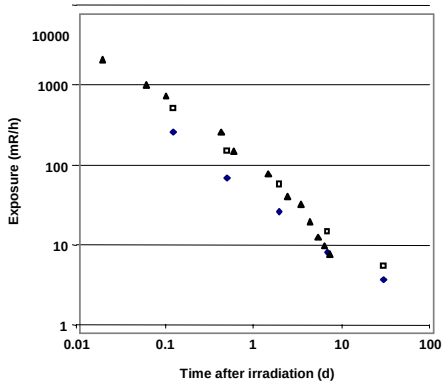

Fig.4. Calculated EDR_{10} versus time after EOB (in days) under RPI conditions: a – Zn-containing capsule, b – capsule components (Ti+cap). Δ – experiment (see Figs.2, 3); □, ◊ – calculation of D. Ehst (□ – upper limit of estimates, ◊ – lower limit of estimates)

As it follows from Fig.4, the measured and calculated EDR_{10} of the activated Zn-containing capsule and its components are in fair agreement.

4. MEASUREMENT OF DOSE CHARACTERISTICS OF ^{99}Mo PRODUCTION DEVICES

4.1. TARGET COMPOSITIONS AND IRRADIATION CONDITIONS

To measure the radiation background of photonuclear ^{99}Mo production devices, two Mo targets of natural composition, each weighing 65.07 g, were used. The target was an assembly of 10 discs, each being ~ 2 mm in thickness and 19 mm in diameter. The target was placed directly behind the converter consisting of four interspaced 1-mm thick tantalum plates. During irradiation the converter and the target were cooled with water at a flow rate of 10 l/min. Molybdenum foils-witnesses of diameter 19 mm were inserted between the Mo discs in the target (Mo-19(1)…Mo-19(10)). In front of the target, there were placed Ø40 mm Mo and Sn foils-witnesses. They were used to determine the bremsstrahlung beam profile (see Fig.2). In addition, measurements of activities $A_{Mo\text{-}19(1)}…A_{Mo\text{-}19(10)}$ enable one to determine the bremsstrahlung intensity variation in depth of the target.

The two targets were irradiated in the accelerator KUT-30 at operating conditions of (36 MeV, 260 μA), one target for 10 min. and the other − for 60 min.

4.2. DOSE CHARACTERISTICS OF RESIDUAL TARGET-DEVICE ACTIVITIES

After 10 minutes after turn-off of the accelerator operated for 10 minutes, the dose rate at the distance L=1m from the target device was measured to be 6800 μSv/h (≈680 mR/h), that on the standard distance L=10 cm basis being 68 R/h. This EDR is formed by the residual activities of the converter, the target and the remaining equipment. After irradiation, the two Mo targets were extracted from the target device for dosimetric measurements. Thus, in 10 min after irradiation, the EDR_{10} of the first target was 32.2 R/h, and in a day it decreased by a factor of 370. In the following period of 1 to 8.8 days the two targets showed a similar fall-off in the EDR due to the decay of Mo-99 isotope. Fig.5 shows the EDR_{10} of the second target as a function of time after irradiation.

High EDR levels of Mo targets of natural composition at the first hours after irradiation are due to ^{97} Nb isotope ($T_{1/2}$=72 min). A further EDR decrease is determined by the decay of the desired isotope ^{99}Mo ($T_{1/2}$=66 hours).

Fig.6 shows the EDR versus time after EOB for the Ta converter. The dose rate is mainly determined by the isotopes ^{180}Ta ($T_{1/2}$=8.12 h) and ^{182}Ta ($T_{1/2}$=115 days).

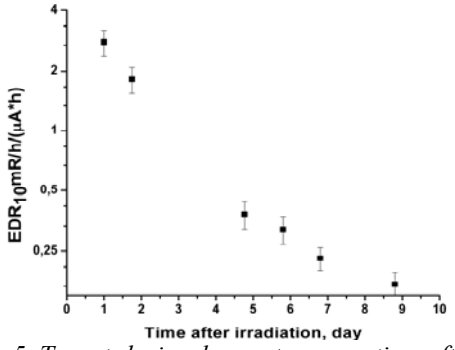

Fig.5. Target-device dose rate versus time after irradiation (day)

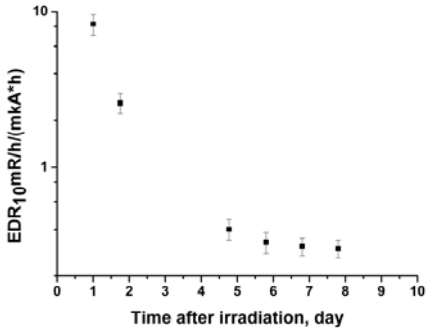

Fig.6. Ta-converter dose rate versus time after irradiation (day)

The converter activity in 4.5 days after irradiation and on was mainly due to ^{182}Ta resulting from the radiative capture reaction ^{181}Ta(n,γ)^{182}Ta. In particular, in 9 days after 10-hour exposure of the Ta converter to the beam having an average current of 240 μA at an electron energy of 36 MeV the EDR at distance of 0.5 m made up 5.4 mR/h. With long exposure of the Ta converter the EDR value will increase practically linearly with time. This fact limits the use of Ta as a converter material in photonuclear production of isotopes.

In realization of the present technology for different periods of target activation it is of interest to make a forecast of the ratio of the desired isotope activity to the EDR provided by the target device. The knowledge of generated partial activities of desired (^{67}Cu or ^{99}Mo) and accompanying (^{62}Zn, ^{63}Zn, ^{69m}Zn or ^{97}Nb) isotopes makes it possible to calculate the target EDR for any activation period. Thus Table 1 lists the measured specific activities A of the ^{67} Cu and ^{99}Mo isotopes (reduced to the irradiation termination), the corresponding EDR values of natural Mo and Zn targets (normalized to 1 g of target weight), and also, the predicted values of the parameters after 24 and 48 hours of irradiation, respectively. It can be seen that the A/EDR ratio substantially increases with increasing time of exposure.

Table 2 gives the predicted activities of ^{67}Cu and ^{99}Mo isotopes, reduced to the termination of irradiation in the (36 MeV, 100 μA) mode, and also, the corresponding EDR values for enriched ^{68}Zn (100%) and ^{100}Mo (100%) targets, 40 and 60 g in weight, respectively, at different time of exposure.

Table 1

Reduced yields of ^{67}Cu ^{99}Mo, target EDRs

Time of exposure, hours	A - desired-isotope specific activity, μCi/g·100μA·h	Target EDR_{10}, mR/h·g	A/EDR_{10} of the target, μCi/ g·100μA·h/mR/h·g
^{67}Cu			
1	68	6,50	10,4
24	1500	83,53	17,9
48	2530	110,79	22,8
^{99}Mo			
1	271	51,37	5,27
24	5810	212,87	27,3
48	10320	295,1	34,9

Table 2

Activity and EDR_{10} of enriched targets

Time of exposure, hours	Desired isotope activity, Ci	Target EDR_{10}, R/h
^{67}Cu		
1	0,0144	0,08
24	0,318	1,77
48	0,538	2,98
^{99}Mo		
1	0,169	3,08
24	3,63	65,88
48	6,45	117,02

5. COOLING WATER ACTIVATION

To estimate the activity of water that cools the target device, its 100 ml aliquot was taken 30 min after the irradiation of Mo was completed.

The gamma-spectrometry analysis has indicated the presence of ^{24}Na and ^{99}Mo isotopes in the water. This can be attributed to their leaching from structural elements, and also to activation of water impurities. The isotope ^{15}O was not detected, because by the measurement time it had fully decayed.

CONCLUSIONS

1. The undertaken experiments have indicated the main background radiation sources of target devices for ^{67}Cu and ^{99}Mo/^{99m}Tc isotope production. The measured EDRs are given in universal units (mR/h/(μA·h) and are reduced to the standard distance from the radiation source. This enables one to predict the radiation environment in the neighborhood of the target device at different modes of its irradiation. The comparison between our experimental data and the calculations by D. Ehst (USA) for the RPI accelerator conditions shows their good agreement.

2. As regards the ratio of the desired isotope (^{67}Cu and ^{99}Mo) activity to the EDR of natural-composition targets, the modes of exposure for much more than 1h are preferred, because in this case the activities of more short-lived impurity isotopes (^{62}Zn, ^{63}Zn and ^{97}Nb) get saturated.

3. The use of enriched ^{68}Zn- and ^{100}Mo-based targets not only increases the yield of the desired isotopes ^{67}Cu and ^{99}Mo/^{99m}Tc, but also substantially reduces the EDR of the targets just after EOB.

4. Of the structural materials considered, the radiation background criterion points to Al (without Fe and Mn impurities) as most preferable. At the same time, the exposure of Al in contact with water may be accompanied by the formation of a great amount of Al_2O_3.

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ОЦЕНКА РАДИАЦИОННЫХ РИСКОВ ПРИ ФОТОЯДЕРНОМ ПРОИЗВОДСТВЕ ИЗОТОПОВ ^{67}Cu И ^{99}Mo

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Основными источниками радиационной опасности при производстве изотопов на ускорителе электронов являются активированные конвертер тормозного излучения и изотопная мишень. На ускорителях КУТ-30 и ЛУ-40М ННЦ ХФТИ проведено облучение экспериментальных выходных устройств для получения изотопов ^{99}Mo и ^{67}Cu в мишенях на основе природных Mo и Zn. Исследована наведенная активность конвертера из тантала, мишеней и охлаждающей воды. Определена мощность экспозиционной дозы, создаваемая каждым элементом, а также скорость ее снижения после облучения.

ОЦІНКА РАДІАЦІЙНИХ РИЗИКІВ ПРИ ФОТОЯДЕРНОМУ ВИРОБНИЦТВІ ІЗОТОПІВ ^{67}Cu І ^{99}Mo

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Основними джерелами радіаційної небезпеки при виробництві ізотопів на прискорювачі електронів є активовані конвертер гальмівного випромінювання та ізотопна мішень. На прискорювачах КУТ-30 і ЛУ-40М ННЦ ХФТІ проведено опромінювання експериментальних вихідних пристроїв для отримання ізотопів ^{99}Mo і ^{67}Cu в мішенях на основі природних Mo і Zn. Досліджена наведена активність конвертера з танталу, мішеней і води, що охолоджує. Визначена потужність експозиційної дози, що створюється кожним елементом, а також швидкість її зниження після опромінення.