BEAM APPLICATIONS

THE FUNDAMENTALS OF ^{99m}Tc PRODUCTION CYCLE AT ELECTRON ACCELERATOR

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The report presents the results of investigations into physical and technological fundamentals of ^{99}Mo/^{99m}Tc production with the use of bremsstrahlung of the electron accelerator.

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1. INTRODUCTION

^{99m}Tc (daughter nucleus of ^{99}Mo) is the basic radionuclide used in the present-day medical diagnostics. The main part of ^{99}Mo is produced in nuclear reactors with the use of the ^{235}U fission reaction [1]. The special features of ^{99}Mo/^{99m}Tc generation at the electron accelerator are determined by both a low production cross-section for this isotope in photonuclear reactions (~180 mbn) and a great transport length of high-energy photons in substance. These special features set the requirements on the systems of initial electron beam formation and control, beam conversion into the bremsstrahlung flux, on the target device and beam parameter monitoring in the process of irradiation, and also on the system of radiochemical extraction of ^{99m}Tc from the target. So, while the specific activity of fission-produced ^{99}Mo reaches 10^3 Ci/g [1], in the electron accelerator case, this parameter makes ~0.1 Ci/g for the natural molybdenum target and ~1 Ci/g for the target enriched in the ^{100}Mo isotope. This circumstance restricts the choice of the procedure for ^{99m}Tc discharge by high-selectivity methods (e.g., extraction, electrolysis, etc.), and also calls for optimization of the technology at all its stages.

2. PHYSICAL PRINCIPLES OF THE PROCESS

2.1. Paper [2] has described the results of computer simulation of ^{99}Mo generation processes in liquid targets based on alkaline solutions of natural Mo. The advantages of these targets are the minimum number of procedures involving the solution, and also the ease of organizing a loop-type cycle with target transportation between the accelerator and the setup for ^{99m}Tc extraction. The disadvantages of the above targets lie in a low volume activity of ^{99}Mo (as a result of which it is advisable to realize this variant with the solution of isotopically pure ^{100}Mo as a basis), and also in instability of saturated solution as regards precipitation.

The analysis of the data obtained shows that the highest yield of ^{99}Mo in a solid natural molybdenum target, 14.4 g in mass, makes 5 Ci per day for the 40 MeV beam of 10 kW power at a thickness of W converter of 1 mm and a thickness of converter-cooling water layer of 3 mm (this being in agreement with the results of ref. [3]). However, at these conditions a power of 3 kW is released in the target, and this makes the target cooling problematic. Here, as far as cooling is concerned, the operating conditions with a cylindrical beam of 30 MeV energy and up to 10 mm in diameter, appear real. In this case, the production of ^{99}Mo is reduced down to 2.75 Ci per day, this however remaining acceptable as regards both general and specific activity of the target.

2.2. The main task of the undertaken experimental study of ^{99}Mo generation processes at the electron accelerator was to determine the yield of this isotope per unit charge of the beam as a function of accelerated electron energy for targets of different geometry and phase composition.

The targets were irradiated at the accelerators EPOS and KUT-20 [4].

2.2.1. For experimental studies into the regularities of ^{99}Mo production in water solutions a prototype of the target device has been developed [5]. It consists of a flowing water-cooled converter of bremsstrahlung and four cylindrical vessels axially symmetric to the electron beam. The vessels were filled with a water solution of Na_2MoO_4 with a Mo concentration of 142 mg/ml. The experiments were performed at the EPOS accelerator at dose accumulation conditions for different electron energies E_0.

Table 1 lists the measured volume activity values for the basic isotopes produced in 1 hour at an average beam current of 10 μA for each mode of irradiation.

Table 1. Volume activity (A) of isotopes in target vessels (Na_2MoO_4)

E_0, MeV	№ vess	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

The reactions occurring only in the photonuclear channel can be used to separate the contributions of different channels to the ^{99}Mo generation. So, for the Na_2MoO_4 solution these are ^{23}Na(γ,n)^{22}Na (Q= – 12.4 MeV)

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and ^{92}Mo (γ,2n)^{90}Mo (Q=-13.1 MeV).

The yield via the (n,γ) channel can be investigated with the ^{23}Na(n,γ)^{24}Na reaction. The data obtained with the help of the mentioned reactions on relative yields of ^{99}Mo in (γ,n) and (n,γ) channels - C_γ and C_n ($C_\gamma + C_n$=1) are given in the report [5].

The analysis of the resulting data has shown that the solution of Mo, based on KOH, may appear more promising as a liquid target. This solution provides a higher concentration in Mo, gives less active wastes under irradiation, and also permits later on practically a direct separation of ^{99m}Tc by the extraction method.

2.2.2. Relying on the results of computer analysis, a target device (Fig.1) was designed to investigate the production of ^{99}Mo in the solid phase.

The device is arranged immediately behind the flowing water-cooled exit window (EW) of the accelerator. Inside the casing S there are a converter C (4 tantalum plates, each being 1 mm thick) and a target itself T in the form of a truncated cone which is axially symmetric to the electron beam.

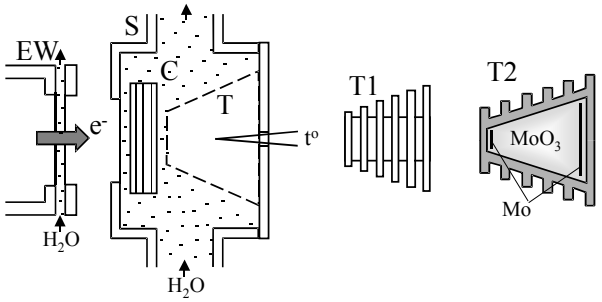

Fig. 1. Device for investigating ^{99}Mo generation in solid targets

Two versions of the target were made: one (T1) - from metallic natural molybdenum, 30 g in mass; the other (T2) - as an aluminum capsule which encloses two foils from Mo and the MoO_3 oxide, 50 g in mass. So, the two targets had nearly the same content of natural Mo. A thermocouple was placed inside each target to measure the temperature during irradiation.

In these versions, the target was irradiated for 30 minutes at the accelerator KUT-20 at an electron energy of 30 MeV, an average current of 7 μA. Then, the total and specific activities of target elements were measured with the help of the Ge(Li) detector (see Table 2). In the process of irradiation, the steady-state temperature value at the center of the MoO_3 target made up 75°C at a cooling water temperature of 36°C.

Table 2. Activity of solid-target elements

Target element	T_1	T_2		
		Mo foil (front)	MoO_3	Mo foil (back)
Activity, MBq	8.249	0.142	5.70	0.065

3. BASIC SYSTEMS AND TREATMENT PROCEDURES

The general scheme of the proposed ^{99}Mo/^{99m}Tc production technology is presented in Fig.2. It is designed to accommodate the equipment within a single radiation-protection zone. The radioactive elements of the production cycle (target, irradiated structures, wastes, etc.) remain within the zone, and only the final product is removed from it in the form of sodium pertechnetate - ^{99m}Tc solution put into compact shipping containers and having the permissible level of radiation.

The undertaken investigations have permitted us to formulate a set of requirements to be met by the basic systems of ^{99m}Tc production at the electron linac.

The accelerator has electron energy up to 40 MeV, beam power no less than 10 kW, minimum transverse dimensions of the beam (<10 mm) and its angular divergence. The exit window of the accelerator at conditions of direct (unscanned) beam must have an operating life more than 1000 hours.

The accelerator monitoring system must provide a continuous unperturbing monitoring of electron energy (average value), beam current (pulsed value and average value), and also of beam position within the exit window.

The target setup is designed in relation to the chosen version of the target (liquid, metallic Mo or MoO_3). In any case, the cooling system of the complex must provide the removal of heat power from the bremsstrahlung converter (no less than 3 kW) and from the target (no less than 0.5 kW). Therefore, it is advisable to make the converter rotating. The radiation durability of the setup structures must be no less than $1 \cdot 10^{10}$ Gy.

The sampling system is also determined by the chosen variant of the target. In all cases, it is reasonable to perform sampling and testing of the target at the initial stage of its irradiation. This is determined by the necessity of measuring the rate of generation of the main isotopic product (^{99}Mo) and by-products (for correcting, if necessary, the conditions of target processing), and by the possibility of safe handling the target.

In the liquid target case, the sampling procedure consists in taking a 1...2 ml sample of irradiated solution with an activity up to 1 μCi.

For the metallic Mo or MoO_3 target, a sample, i.e., Mo foil, is placed at the exit of the target device, coaxially with the target. The accelerator is switched on to be operated in the mode of target treatment, and the sample is activated up to ≤1 μCi. Then, the sample is forwarded to the gamma-spectrometer. To optimize the target irradiation conditions and target adjustment with respect to the flow of braking photons, the method of measuring the surface activity distribution of the foil by means of a collimated CdZnTe radiometer has appeared to be rather efficient.

The irradiated target transport system provides a remote discharge of the target (the equivalent dose rate (EDR) on the surface makes > 10 Sv/h) into a container, transportation of the container to the radiochemical laboratory, a remote-controlled withdrawal of the target and its transfer into the shielding box for further treatment.

The system of target processing is determined by the form of the target. In the realistic version of the target based on natural molybdenum with a comparatively low activity (up to 3 Ci), the efficient separation of ^{99m}Tc is provided by the method of multiple extraction of ^{100}Mo containing water solution by one and the same extrac-

tant (methyl ethyl ketone) volume [6]. The final product is the solution of sodium pertechnetate-^{99m}Tc of standard activity, which is packed into flasks and undergoes sterilization.

If the isotopically enriched $^{100}MoO_3$ target is used, the radiochemical facility can readily be supplemented with a circuit for reduction of trioxide from the solution at the end of the 5-day cycle of ^{99m}Tc extraction for the repeat production of ^{99}Mo at an accelerator.

The isotope product quality control system involves a set of measuring devices and methods for determining the correspondence of the product to the Pharmacopoeia Regulations (PhR).

The radioactive wastes (RAW) handling system provides a stock-piling of wastes in temporary storages, and also a periodic control of RAW activity.

The final-product transportation system includes portable shipping containers, which accommodate flasks with the solution of sodium pertechnetate-^{99m}Tc, and also specially equipped transport means for the delivery of containers to clinics.

The radiation dosimetry system embraces all technological procedures accompanied by the action of radiation on personnel. The requirements on the system and admissible levels of exposure are described in Radiation Safety Standards.

4. CONCLUSION

The undertaken investigations have resulted in the elaboration of the fundamentals of a soft technology of ^{99m}Tc production for nuclear medicine with the use of the electron accelerator. The proposed process is fully compatible with the technologies in service today, based on the ^{98}Mo(n,γ)^{99}Mo reaction [6,7], as regards the procedures of radiochemical extraction of ^{99m}Tc, preparation of the solution of sodium pertechnetate-^{99m}Tc, packing and sterilization, control of product quality and delivery to the customers. These circumstances facilitate the procedure of new technology certification.

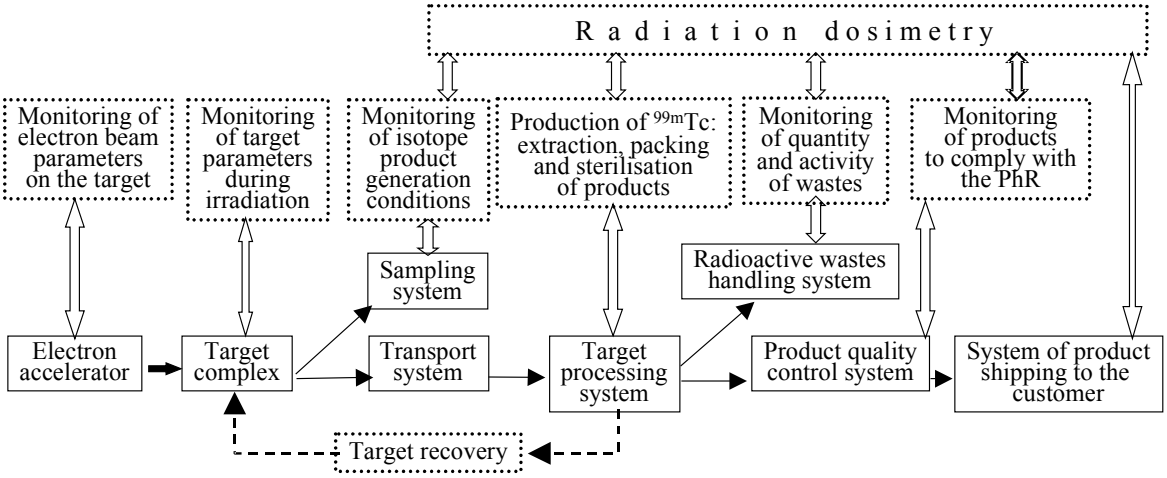

Fig.2. Functional block diagram of ^{99m}Tc production at the electron accelerator

The use of the target enriched in the ^{100}Mo isotope appears most efficient, both economically and ecologically. For example, in the case of >95% enrichment, the yield of ^{99}Mo increases by order of magnitude as compared with the target of natural composition. The activity of liquid RAW produced in one technological cycle is reduced to the same extent. Though ^{100}Mo is expensive (up to $5000 for 1 gram), a single purchase of 30 grams of the isotope provides a long-standing production of ^{99m}Tc. Really, in one cycle of irradiation, no more than 10^{-6} nuclei of ^{100}Mo transmute into ^{99}Mo, and after extraction of ^{99m}Tc, $^{100}MoO_3$ can be easily reduced from the water phase to be used for the repeat irradiation.

The use of reactor Mo accumulated in the wastes from spent fuel processing [8] is less expensive. The concentration of ^{100}Mo in this case is 28% (9.6% in the natural Mo), i.e., the efficiency of the process is 3 times increased.

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ОСНОВЫ ТЕХНОЛОГИЧЕСКОГО ЦИКЛА ПО ПРОИЗВОДСТВУ ^{99m}Tc НА УСКОРИТЕЛЕ ЭЛЕКТРОНОВ

Н.П. Дикий, А.Н. Довбня, В.Л. Уваров

Изложены результаты разработки физических и технологических основ производства ^{99}Mo/^{99m}Tc с использованием тормозного излучения ускорителя электронов.

ОСНОВИ ТЕХНОЛОГІЧНОГО ЦИКЛУ ПО ВИРОБНИЦТВУ ^{99m}Tc НА ПРИСКОРЮВАЧІ ЕЛЕКТРОНІВ

М.П. Дикий, А.М. Довбня, В.Л. Уваров

Викладені результати розробки фізичних і технологічних основ виробництва ^{99}Mo/^{99m}Tc із використанням гальмівного випромінювання прискорювача електронів.