PRODUCTION OF ISOTOPES ^{184}Re, ^{186}Re AND ^{188}Re AT LINEAR ELECTRON ACCELERATORS OF NSC KIPT

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This paper presents the preliminary results on determining the values of specific activity of radionuclides ^{184}Re, ^{186}Re and ^{188}Re. Irradiation of Re targets of a natural isotope composition with gamma-quanta and neutrons was conducted at NSC KIPT linear accelerators. Possible levels of activity of the above-mentioned radionuclides are forecasted.

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It is known that in the world nuclear medicine more than 80 % of diagnostic means are radiopharmaceuticals based on ^{99m}Tc. Developed-to-perfection technologies for commercial production of ^{99}Mo $\rightarrow$^{99m}Tc generators are based on a reactor method of ^{99}Mo production. However, due to a comparatively short half-period of ^{99m}Tc, as well as, quick accumulation of its decay product—^{99}Tc, deteriorating the diagnosis quality this element can not be put in a row of ideal ones [1].

Therefore, in the world practice one observes a tendency to discover new radionuclides having higher characteristics as labels for radiopharmaceuticals [2]. For example, one of promizing radionuclides is an isotope ^{186}Re having, as compared with ^{99m}Tc, a higher complex-forming ability and a high tropy characteristic. An optimum value (137 keV) of the energy of γ-quanta emitted, short half-period and absence of accompanying radiation makes this isotope rather promising. The present work is aimed for obtaining the data on the values of the specific activity of Re isotopes produced at NSC KIPT accelerators under different irradiation conditions.

As targets for isotope production we have used metallic Re in the form of plates and fine dispersed powder. The chemical purity of initial targets was no less than 99.99%. The targets were packed in containers from the aluminum foil 0.1 mm thick.

Accelerators were operating under the following irradiation conditions:

- electron energy – 25 MeV; 11.5 MeV
- mean beam current – 500 μA;
- irradiation time – 20 hours; 105 min.

Prepared and weighed sample-targets were placed at the accelerator EPOS at a distance of ~90 cm from the excit foil of the accelerator. The cassetes with other target after radiation treatment were placed ahead of the samples. Such arrangement of rhenium targets is caused by the necessity to carry out simultaneous seances of irradiation in conformity with different radiation programmes that enables one to use the "spent" electron beam. At the accelerator KUT similar targets from Re were placed ahead other targets immediately under the electron beam. A bremsstrahlung converter was not set in order not to disturb the radiation conditions for simultaneously conducted irradiating programmes.

The specific activity of targets was determined by measuring, on a spectrometric bench, of a number of γ-quanta in the given energy range with a known accelerated electron current density in the site of samples. The similar technique of measurements and data processing was applied in [3]. A measuring bench was completed with a Ge(Li) semiconductor detector DGDK-100A of a 100 cm^3 volume. The energy resolution of the detector is 2.3 keV, detected by ^{60}Co γ–lines.

For mathematical data processing we have used the programmes determining the sum under the peak of full photoabsorption with subtraction of the approximated background. To obtain the necessary accuracy we have used the method of least squares that makes it possible to approach the experimental data by the exponential curve in the form $A = A_0 \exp(-\lambda t)$, where **λ** is the decay constant, **t** is the time after irradiation stopping, $\mathbf{A_0}$ is the activity at an instant of irradiation stopping.

Miscalculations were counted by the following formula:

$$N_0 = \frac{N}{1 - N_s \cdot (\tau / t_1)}, \qquad (1)$$

where **N** is the number of photons in the photopeak detected by the detector, $\mathbf{N_0}$ is the real number of photons, **Ns** is the total number of pulses in the spectrometric channel, **τ** is the dead time of the spectrometer, $\mathbf{t_1}$ is the exposition time of spectrum. The error of N_0 includes the error of the sum value under the photopeak σ_{n0}, the error due to the given exposition time of spectrum σ_t, and the error due to subtraction of the background σ_f. The total error is determined as a mean-square value of all the components.

$$\sigma_{tot} = \sqrt{\sigma_{n0}^2 + \sigma_t^2 + \sigma_f^2}. \qquad (2)$$

The main contribution into the total error is made by σ_{n0} and σ_f [4]:

$$\sigma_{n0} = \sqrt{\frac{1 + N_0 \cdot (\tau / t_1)}{N_0}}, \sigma_f = \sqrt{\frac{1 + N_f \cdot (\tau / t_1)}{N_f}}, \qquad (3)$$

$$\sigma_t = \sqrt{N_0 \cdot \exp(-\lambda \cdot t_1)} / N_0. \qquad (4)$$

When evaluating the sample activity, the detector efficiency η was the essential factor. It was determined as a relationship between the quantity of quanta detected in the photopeak and the quantity of quanta radiated from the γ-source with a known activity. With such determination of the efficiency it was not necessary to evaluate in each case a solid angle being covered by the detector when changing the source position. To determine η we have used a set of standard γ-sources comprising ^{22}Na, ^{137}Cs, ^{54}Mn, ^{241}Am with energies of γ-lines

511, 1275, 661, 834, 59.5 KeV, respectively.

Results on the detector efficiency measured as a function of the γ-quantum energy at different distances from the source are represented in Fig. 1.

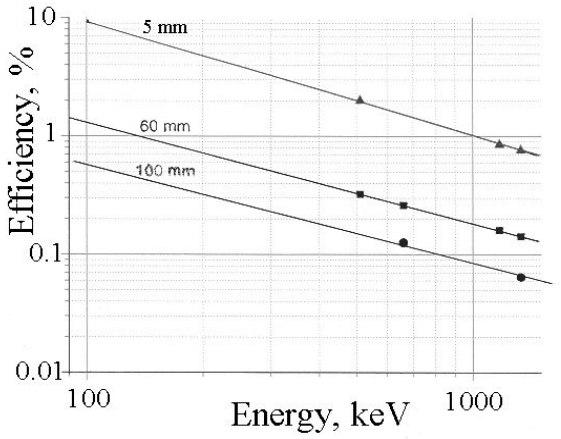

Fig. 1. Detector efficiency as a function of γ-quantum energy at different distances from the source: 5.5 mm, 60 mm, 100 mm.

The radionuclide activity A_0 at the instant of irradiation stopping was calculated by the formula

$$A_0 = \frac{A}{ç \cdot I \cdot m \cdot k \cdot e^{-\lambda \cdot t}} \ (Bq/g \cdot \mu A), \qquad (5)$$

where **η** is the efficiency of γ-quantum detection, **I** is the average current falling onto the sample, $\mathbf{A_0}$ is the specific activity of the sample at the instant of stopping irradiation, **m** is the target mass, **k** is the coeffcient of γ-quantum multiplicity, **A** is the number of γ-quanta detected in the time **t** after irradiation stopping, **λ** is the radionuclide decay constant.

Initially irradiation of the Re sample in the form of a plate was performed at the accelerator EPOS. When measuring the activity of samples in the form of a plate the essential factor is the γ-irradiation absorption with an energy of 137 keV in the target material. For the data be more accurate we have carried out irradiation of Re samples in the form of a powder at the accelerator KUT with an electron beam energy of E_0 =11.5 MeV. Selection of a powdered target significantly decreases the influence of γ-radiation absorption in the target material on evaluation of the specific activity.

Under irradiation of Re samples having a natural isotope composition: ^{185}Re(37%) and ^{187} Re(63%), as a result of γ,n reactions, isotopes ^{184}Re and ^{186} Re are formed (Table 1).

Table 1.

Nuclear reaction	$T_{1/2}$	E_{thres} MeV	Eγ, keV (intens.)
^{185}Re(γ,n) ^{184}Re	38 days	7.8	111(17%) 792(36%) 894(16%) 903(36%)
^{187}Re (γ,n) ^{186}Re	90 hours	7.3	137(12%)
^{185}Re (n,γ) ^{186}Re	90 hours	-	137(12%)
^{187}Re (n,γ) ^{188}Re	17 hours	-	155(21%)

In Fig. 2 shown is a part of the radiation spectrum of the Re sample irradiated at the accelerator KUT during 105 min. For the illustration be clearer the γ-lines of ^{184}Re with energies of 792 and 903 keV are not shown.

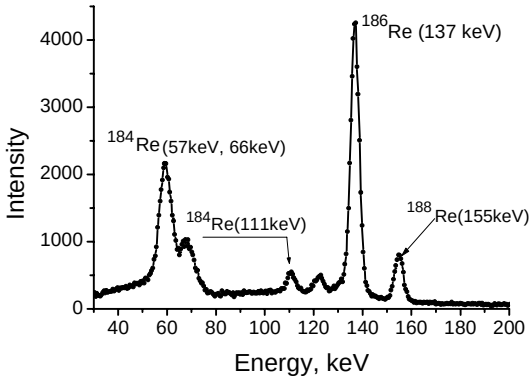

Fig. 2. Spectrum of Re irradiated at the accelerator KUT.

An important result is the presence of the 155 keV peak belonging to ^{188}Re. This isotope formed from ^{187}Re, as a result of neutron capture, has comparatively short half-life and is interesting for radiological investigations. A traditional method of ^{188}Re production is the ^{188}W based generator obtained in nuclear reactors from ^{186}W (28%) by the scheme ^{186}W (n,γ)→ ^{187}W(n,γ) → ^{188}W(69 days) → ^{188}Re. Such a two-step process of ^{188}W production requires availability of high-intensity neutron flows and reactor irradiation of long duration. The results obtained on the ^{188}Re radionuclide shows a possibility to produce this radionuclide at a linear electron accelerator as an alternative of the reactor method that is currently urgent for the nuclear medicine of Ukraine. Irradiation of Re targets with neutrons is more preferable than with γ-quanta because there formed are only isotopes ^{186}Re and ^{188}Re without ^{184}Re.

The levels of induced activity of isotopes at the accelerator KUT are calculated by the following data: irradiation during 105 min, sample mass is 250 mg, beam is spread on the area of 300 m^2, surface of the Re sample is 0.3 cm^2. We have obtained the following data on the specific activity of radionuclides: ^{186}Re – $4.3 \cdot 10^4$ Bq/g·μA, ^{188}Re – $5.3 \cdot 10^3$ Bq/g·μA. At the accelerator EPOS the samples were irradiated during 20 hours. Rest of parameters for calculating the specific activity were similar to parameters of irradiation at the accelerator KUT. The value of activity of radionuclide ^{186}Re was $7.2 \cdot 10^6$ Bq/g·μA. The values of activity allow one to make a conclusion about expedience to continue a research in this direction. Let us note that this experiment was preliminary since the target irradiation was conducted simultaneously with other radiating programs without applying complementary converters and neutron-producing targets. The specific activity of produced isotopes can be increased, firstly, by applying the optimized converter of bremsstrahlung irradiation (Ta (Z=73), thickness ~2 mm); secondly, by increasing the duration of sample irradiation and by focusing the electron beam onto the converter or neutron-producing tar-

get that, according to calculations, should increase the specific activity approximately by a factor of 10^3. This enables one to begin experiments on preparing and using farmaceuticals labelled with isotopes ^{186}Re and ^{188}Re.

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