PRODUCTION OF SHORT-LIVED RADIONUCLIDES ON THE ELECTRON LINAC FOR PET*

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The comparison of modes of radionuclide production with the use of cyclotron beams ("cyclotron mode") and beams of gamma-quanta (as a result of photonuclear reactions) is carried out. On a beam of the electron linac (E = 25 MeV, $P \cong 20$ kW) positronemitting radionuclides ¹¹C, ¹³N, ¹⁵O, ¹⁸F with the specific activity(in Bq/g· μ A) 1,9·10⁷, 1,67·10⁶, 2,5·10⁶, $1.7 \cdot 10^6$, respectively, are obtained. These data show a possibility of production of short-lived medical radionuclides on a electron linac in amounts sufficient for human diagnostics on PET (tomograph). A large range of gamma-quanta energies in the target in a number of cases ensures total activity of radionuclides compared with a "cyclotron mode". Radionuclide production on high-power electron linacs (10÷20 kW) supplements that on the cyclotron. The use of electron linacs for electron- and gamma-therapy, sterilization and creation of multipurpose neutron fluxes $\approx 10^{13}$ n/s expand possibilities of application of radiation technology on various accelerators.

Most of radionuclide means for medical diagnostics (≈ 80 %) is pharmaceuticals based on ^{99m}Tc. However, a rather essential (and even determining in a number of cases) role in diagnostics of a various kind of diseases is assigned to pharmaceuticals tagged by radionuclides as ¹¹C, ¹³N, ¹⁵O, ¹⁸F. These positron-active radionuclides are widely used all over the world as sources of annihilation gamma-radiation with the energy of 511 keV in positron-emission tomographs (PET). In connection with growing interest to the PET-technique and efforts undertaken for creation of a tomograph in NSC KIPT, it is expedient to study capabilities of production of short-lived radionuclides on the available electron linacs of NSC KIPT.

The present work is undertaken with the purpose of obtaining the experimental data about the values of specific activity of short-lived radionuclides on particular targets in the operating 25 MeV linear accelerator of electrons "EPOS" with the beam power 20 kW.

EXPERIMENTAL RESULTS RESULTS

Experiments were performed to determine the specific activities (Bq/g · μA) of ^{11}C , ^{13}N , ^{15}O , ^{18}F radionuclides of medical purpose for PET (positron-emission topography) which are realized under conditions of the electron linear accelerator LU-20 "EPOS" with power $\cong 20$ kW. For this the following

substances were used as targets: water (in glass capsules of 2 cm² for medical purpose), powders of boron acide H₃BO₃ and boron nitride BN, films of fluoroplastic C₂F₄ and polyethylene (C₂H₂)_n. The powders were packed in containers from aluminium of 0.1mm thick and 20mm diameter. The films of fluoroplastic and polyethylene were in the form of disks of 16mm diameter and manufactured from the tape of 0.2mm thick. The samples were weighted with the analytical balance to the nearest ±5mg; the reference density values for materials being irradiated have been used. The targetsamples prepared were mounted in the assembly with dimensions 25x40 mm² that was placed onto the electron beam directly behind the lead converter of 2mm thick. Especially for these works on the acceleraator the following radiation conditions were created:

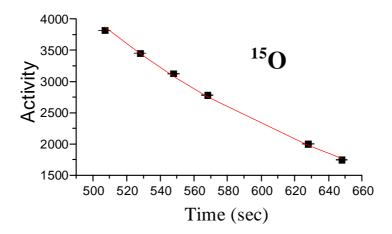
electron energy -25 MeV; frequency of beam scanning -3 Hz; current pulse $-500 \mu A$; area of beam under scanning $-80\text{-}300 \text{ mm}^2$; pulse duration $-4 \mu \text{sec}$ irradiation time -10 min; pulse-repetition frequency -150 Hz.

The specific activity was determined on the measuring spectrometric bench for the counting rate (pulse/sec) of irradiated samples having the known chemical composition and weight and the known density of accelerated electron current.

The spectrometric bench is equipped with a Ge(Li)-detector, electronics in CAMAC standart and computer IBM PC Pentium-133. The measured activity values were corrected for the detector efficiency in the given measurement geometry and for the dead time. After processing of all experimentally obtained spectra the decay curve was plotted over the values of counting sum under the photoabsorption peak (with taking into account the background). By the use of the program REGRESS the experimental data were fitted by the curve of the form $A=A_1 \cdot exp(-\lambda t)$, where λ is the decay constant, t is the time from the irradiation stopping, A_1 is the activity in a moment of irradiation stopping. Besides, the measurement errors were calculated with regard to the main factors.

According to experimental data the curve of ¹⁵O – decay was created (see Fig.).

^{*} The work is supported by STCU contract #285 and project INTAS LA-98-01.



The production yield
$$A_0 = \frac{A_1}{\eta \cdot m \cdot J \cdot \kappa}$$

(Bq/g· μ A), where m is the oxygen mass in the sample, J is the mean current density on the converter (μ A/cm²), η is the detector efficiency with taking into account the solid angle, k is the multiplicity coefficient of γ -quanta, A₁ is the activity of the sample in a moment of

irradiation stopping. The calculated value $A_0=2.5\cdot10^6$ Bq/g· μ A.

A comparison between the experimental data on production of isotopes ¹¹C, ¹³N, ¹⁵O, ¹⁸F (adjusted comparative with the [3]) and calculation results of other authors was done (Table 1).

Table 1

Radionuclide	Specific activity, Bq/g·μA at 25 MeV		
	Mac Gregor[1]	Lutz[2]	Our experiment (up to saturation)
¹¹ C	$9.25 \cdot 10^6$	5.3·10 ⁶	$1.9 \cdot 10^7$
^{13}N	$3.76 \cdot 10^6$	$1.04 \cdot 10^6$	$1.67 \cdot 10^6$
¹⁵ O	$3.2 \cdot 10^6$	$8.4 \cdot 10^6$	$2.5 \cdot 10^6$
¹⁸ F	2.04·10 ⁶	2.12·10 ⁶	1.7·10 ⁶ (extrapolation for 1 hour irradiation)

The Table 1 shows that the experimental values of specific activities of ¹¹C, ¹³N, ¹⁵O are in satisfactory agreement with calculation data of Lutz and Mac Gregor.

However, the distinctions between calculated and measured activity values can be explained by differences in the irradiation geometry for different authors as well as by uncertainties in the bremsstrahlung spectrum.

The observed exceeding (approximately by a factor of 5) of the experimental value of radionuclide ¹⁸F production as compared to the calculated one is related to the contribution of the process of production from neutrons according to the reaction ¹⁹F(n, 2n)¹⁸F that can take place under conditions of the "EPOS " accelerator.

In Table 2 comparative data on production ¹⁵O in various accelerators of electrons are given [3].

Table 2.

Source of information	Linac "Fakel" Kurchatov IAE, Moscow[4]	Linac-30MeV Center of Nuclear Medicine, USA, Cincinnati[5]	Linac "EPOS" NSC KIPT, Kharkov
Conditions of irradiation	$ \begin{array}{l} E_0{=}30 \text{ MeV} \\ I_{av}{=}100 \mu\text{A} \\ T_{ir}{=}7 \text{ min} \\ V{=}200 \text{ cm}^3 \end{array} $	$E_0=26 \text{ MeV}$ $I_{av}=100 \mu\text{A}$ $T_{irr}=4 \text{ min}$ $V=500 \text{ cm}^3$ $6\times18 \text{ cm}$	$\begin{array}{l} E_0{=}25 \; MeV \\ I_{av}{=}300 \; \mu A \\ T_{irr}{=}10 \; min \\ V{=}200 \; cm^3 \end{array}$
Integral activity in a moment of the end of irradiation	700 mCi	184 mCi	130 mCi (recalculation to 1 cm ³)

From Table 2 one can see, that the production of ¹⁵O on the accelerator "EPOS" in 200 cm³ of a water for 10 minutes of irradiation is 26 times more, than is required (5mCi) for examination of one patient.

DISCUSSION

The given data show capabilities of medical isotopes production on the accelerator "EPOS" to be sufficient for examination on a PET. The comparison of "cyclotron mode" with "electron linac mode" shows, that higher (10-100 times) values of cross-sections (p, n), (d, n), (d, 2n) and (p, α) - reactions in a comparison with cross-sections of photonuclear reactions - (γ , n), (γ , p) etc. give a possibility to receive higher values of specific activity of radionuclides on cyclotrons.

This circumstance results in the necessity to increase the weight of the irradiated target when using of gamma-quanta beams. The weight of the processed target ($\approx 20\text{--}30$ g) is large enough for irradiation by neutrons, high-energy protons and, especially, for extraction of isotopes from spent fuel rods.

On the other hand, the conditions of irradiation on an electron linac are more favorable, as the basic thermal load is born in the converter, which transforms the flow of electrons in a gamma-quanta. The high penetrating ability of gamma-quanta allows one to simplify the design of the converter and containers for irradiated substances. It allows one to irradiate targets in various states of aggregation (solid, liquid, gaseous) and in various chemical compounds. The preparation of the targets for irradiation and radiochemical detachment is much easier, than in "cyclotron mode".

The isotope structure of targets is important too. For example, when obtaining the radionuclide ¹⁸F on cyclotrons the reactions ¹⁸O (p, n) and ²⁰Ne (d, ⁴He), which require expensive gas targets enriched with the isotopes ¹⁸O and ²⁰Ne, are most convenient.

On an electron beam the production of the radionuclide ^{18}F is carried out on targets of natural composition (100 % by content) of the isotope ^{19}F on the channel of reaction ^{19}F (γ , n) ^{18}F . Our analysis shows the advantage in production of radionuclides with high nuclear number by the "electron linac method" because of the higher cross-section of photonuclear reactions.

Besides, on electron accelerators the radionuclides can be obtained which cannot be obtained on cyclotrons, for example 67 Cu, 101 Ph and, in particular, 99m Tc

CONCLUSION

The data are shown that 25 MeV electron linac with power of the beam about 20 kW able to meet competition with cyclotron in short-lived isotopes for PET [3,6] and ^{99m}Tc [7] production. Additional advantages of the electron linac application for medical radionuclides production is its comparability with other traditional accelerators programs such as sterilization, activation analysis, radiation modification of polymers and semiconductors etc. [7] A rather low cost of the electron accelerator with the beam power ≈ 20 kW creates some additional perspectives. It allows one to use an "electron linac" not only as a tool for production of short-lived radionuclides for diagnostics in a combination with PET, but also for sterilization, gamma-therapy, and for creation of a multi-purpose source of neutrons with a flow of $\approx 10^{13}$ n/s, thus expanding both scientific and applied capabilities of radiation technology.

Authors greatly appreciate the participation of G.L. Bochek, V.I. Kulibaba, A.V. Torgovkikn, I.M.Prokhorets, D.G.Pugachev, A.A.Kapliy, and V.D. Ovchinnik.

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