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^{18}F RADIOISOTOPE YIELD OF POLYTETRAFLUOROETHYLENE ACTIVATION ON THE M-30 MICROTRON

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The experimental and modeled values of the yield of the ^{18}F radioisotope formed during the irradiation of polytetrafluoroethylene (Teflon) with bremsstrahlung photons at the maximum energy of 17.5 MeV are presented. The output was determined using the induced activity method. The activation process was carried out on the M-30 microtron of the Institute of Electronic Physics of the National Academy of Sciences of Ukraine. The yields of the ^{18}F radioisotope for the optimized scheme of stimulation of (γ,n)-reactions were modeled by the GEANT4 toolkit, taking into account the technical characteristics of the M-30 microtron electron output unit. The effectiveness of using the formed beam of bremsstrahlung photons for forming the ^{18}F radioisotope was evaluated, taking into account the reaction cross-section for the optimized irradiation scheme. The possibilities of increasing the output of the ^{18}F radioisotope by optimizing the activation time of the samples and their geometric dimensions were analyzed. The proposed approach can be used to optimize photonuclear reaction stimulation schemes at the M-30 microtron for a wide range of nuclei and predict the yields of the radioisotopes formed.

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INTRODUCTION

Due to its physical characteristics and general chemical properties, the radioisotope ^{18}F is widely used in nuclear medicine for highly sensitive diagnostic studies (positron emission tomography). ^{18}F-labeled fluorodeoxyglucose is the most commonly used pharmaceutical drug in clinical oncology research (PET scanning). This drug accounts for over 95% of all PET scans [1, 2]. Therefore, studies of various methods of ^{18}F production are of great interest [3].

This especially applies to photonuclear methods for producing medical radioisotopes (based on the use of bremsstrahlung due to its high integral intensity), which are implemented on electron accelerators [4–6]. The efficiency of the production of medical radioisotopes is characterized by yield and purity. The yield can be affected by photon energy, cross sections of photonuclear reactions, integral values of photon fluxes, target thickness, irradiation time, etc. Purity can be affected by factors such as the nuclear reaction mechanism and isotopic purity of the target, etc [4–6]. When using photonuclear reactions (primarily the (γ,n)-reaction) to obtain ^{18}F, its yield is, as a rule, lower compared to the traditional method of producing ^{18}F on cyclotrons [3] since the cross section of the reaction is 1–2 orders of magnitude smaller [7]. However, in photonuclear reactions, it is possible to use natural targets ^{19}F (for example HF [8, 10, 13], LiF [9, 10, 11], C_2F_4 [10, 11, 14], CF_4 [9], SF_6 [9], Ca_2F [12], Na_2PFO_3 [13], K_2SiF_6 [14]), which allows for lower costs compared to proton reactions [3]. Available data on the possibility of obtaining ^{18}F due to photonuclear reactions induced in fluoride-containing targets are extremely contradictory [3].

On the one hand, the data of photoneutron catalogs compiled as a result of fundamental research (for example: [15]) are based on a single work in which the total cross section of reactions [(γ,n) + (γ,pn)] for ^{19}F was measured (range of photon energies – 10.90…27.84 MeV), but the (γ,n)-reaction cross section was not determined separately [16]. The cross section of the ^{19}F(γ,n)-reaction was determined only for one energy value – 20.3 MeV (a Teflon sample was used as a target) [17].

In the case of light elements, this can lead to serious inaccuracies in the (γ,n) reaction cross section at energies above the (γ,pn) reaction threshold (the reaction threshold for ^{19}F is 16.039 MeV [7]).

On the other hand, the data obtained during applied work on the production of ^{18}F correspond to fairly high values of the upper energy of the bremsstrahlung photon spectrum, for example, for Teflon: 12.25 [10], 20.3 [17], 25.0 [10, 14], 36.7 [11] MeV. Therefore, it is difficult to find a connection with the general properties of the studied reactions — cross sections and their moments as a function of the energy of bremsstrahlung photons — using the quantities describing the production of ^{18}F presented in works [8–14].

It should be noted that although many technical and economic justifications for producing radionuclide ^{18}F using photonuclear reactions have been published [8–14], the actual production still needs further study [3].

As a result of the conducted theoretical and experimental studies, it was established that the output of the photonuclear reaction (or the activity of the formed radionuclide) primarily depends on the energy of electrons, nuclear-physical properties of converters and irradiated samples, and their geometric dimensions [4–6, 11, 18, 19, 20]. It should be noted that the integral output of the reaction is also significantly influenced by the initial parameters of the electron beam (its geometric dimensions (shape), which depend on the design features of the electron output node and differ for different accelerators [6, 19, 21]), geometric factors (mutual location of the bremsstrahlung converter and the irradiated sample relative to the axis of the initial electron beam [18, 19]), and the presence of additional structural elements (filters, screens, collimators [19, 22]) used in their formation.

The listed factors must be taken into account to optimize the process of stimulation of the photonuclear $^{19}F(\gamma,n)$ reaction on electron accelerators, taking into account their technical characteristics (initial parameters of electron beams) since they will significantly affect the activity of the obtained radioisotope (^{19}F). Therefore, studying the possibility of producing medical radioisotopes on certain types of accelerators, taking into account the contribution of possible accompanying nuclear reactions (which can affect the purity of the produced radioisotopes [23]), is an urgent task.

The paper presents the results of experimentally and theoretically (modeled) determined values of the yield of the radioisotope ^{18}F, formed during the irradiation of polytetrafluoroethylene (CF_2, commercial name – Teflon) [24] at the M-30 microtron of the IEP of the National Academy of Sciences of Ukraine and the efficiency of its production, taking into account the technical characteristics of the electron accelerator.

1. MATERIALS AND METHODS

Experimental determination of the 18 F radioisotope yield through the ^{19}F (γ,n)-reaction channel was carried out at the M-30 microtron of the IEP of the National Academy of Sciences of Ukraine [19, 23].

Teflon samples (square plates with edge – 10 mm, thickness – 0.1 mm) [24] in sealed packages made of tracing paper [25] were irradiated at the maximum energy of bremsstrahlung photons – 17.5 MeV. The packages contained from 1 to 3 Teflon plates. At the specified energy, the products of accompanying photonuclear and neutron (initiated photoneutrons) of the reactions did not affect the final results of determining the ^{18}F radioisotope during Teflon activation. Fig. 1 presents the cross-sections of possible photonuclear reactions during the activation of the Teflon sample [26, 27]. The choice of packaging material is also due to the fact that elemental compounds ($_1H$, $_6C$, $_8O$) that are part of the tracing paper have small cross-sections of activation along the channels of photonuclear and neutron reactions [7, 26, 27].

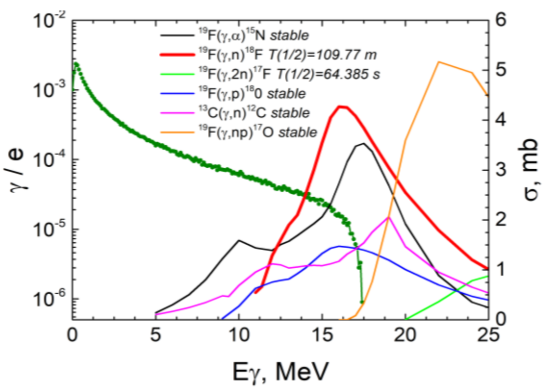


Fig. 1. Spectrum of bremsstrahlung photons and cross-sections of possible photonuclear reactions upon Teflon activation

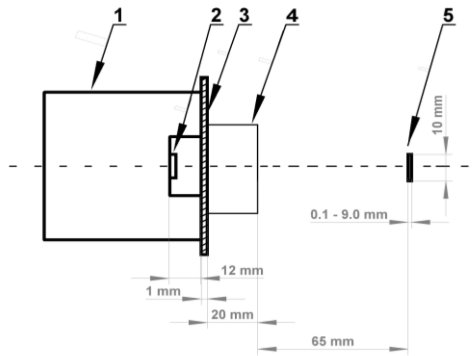
To optimize the Teflon activation process (stimulation of $^{19}F(\gamma,n)$-reactions), previously modeled parameters of the optimal scheme for stimulating photonuclear reactions at the M-30 microtron at an initial electron energy of 17.5 MeV were used. This scheme took into account the technical and design features of the electron extraction unit [19, 23].

To clean the generated bremsstrahlung beam from residual electrons (to exclude the process of destruction of the irradiated sample), a filter made of aluminum was used in the proposed sample activation scheme [18, 19, 23].

Fig. 2 shows the optimized scheme for stimulating Teflon samples on the M-30 microtron.

1.1. MODELING OF THE STIMULATION PROCESS OF THE $^{19}F(\Gamma,N)^{18}F$ REACTION ON THE M-30 MICROTRON

To carry out computer simulation of the process of stimulation of the $^{19}F(\gamma,n)$ reaction on the M-30 microtron, taking into account the parameters of the bremsstrahlung beam and its components (photons, residual electrons) interacting with the Teflon sample, as well as factors (the presence of constructive elements of the activation scheme, their geometric arrangement) affecting the output of the ^{18}F radioisotope, a C++ program was developed using the GEANT4 v11.1 toolkit [28].

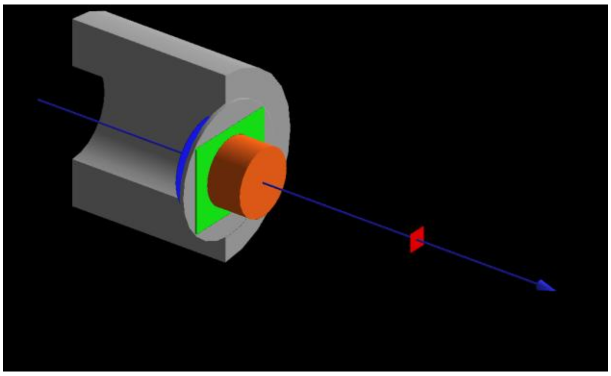


a

b

Fig. 2. Scheme of stimulation of the $^{19}F(\gamma,n)^{18}F$ reaction on the M-30 microtron – a: 1 – block of electron extraction; 2 – window of the output node (ellipse 22x6 mm, thickness – 50 μm); 3 – Ta-converter (52x45x1 mm); 4 – aluminum filter (diameter – 30 mm, thickness – 20 mm); 5 – hermetically packed Teflon samples and b – photo of the output node

The program was implemented under the Windows platform using the multi-threading mode. The calculations were performed for the initial number of 10E8 electrons. During the simulations, all particles generated by the bremsstrahlung target (bremsstrahlung photons, residual electrons, secondary photoneutrons) falling into the plane of potential installation of the irradiated sample (100 x 100 cm) and onto the real sample were recorded. Fig. 3 presents a visualization of the irradiation scheme with the GEANT4 toolkit.



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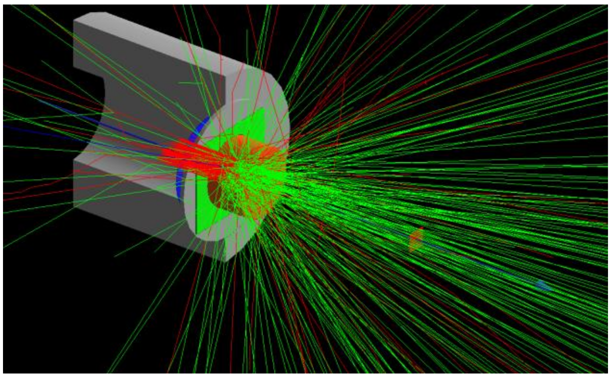

b

Fig. 3. Visualization of the simulation scheme with the GEANT4 toolkit: a – without particles; b – with particles

1.2. ^{18}F RADIOISOTOPE YIELDS DURING ACTIVATION OF POLYTETRAFLUOROETHYLENE SAMPLES

The output of the radioisotope ^{18}F during the irradiation of polytetrafluoroethylene on the M-30 microtron was determined within the framework of the induced activity method [22].

Studies of ^{18}F radioisotope yield dependence (normalized to 1 initial electron) on the activation time (20, 40, and 60 min) of the Teflon sample at a fixed thickness of 0.3 mm were carried out.

The integral values of the initial electron beam during the activation of the Teflon samples were measured by the secondary electron emission monitor at the output window of the electron output. Their values were 2.27E+16, 4.55E+16, and 6.83E+16 for activation times of 20, 40, and 60 min respectively [29]. The instability of electron energy during the irradiation of Teflon samples did not exceed 0.04 MeV.

Additionally, the dependence of the ^{18}F radioisotope yield on the thickness of the Teflon sample (0.1, 0.2, and 0.3 mm) at a fixed activation time of 60 min was investigated.

The output of radioactive nuclei of ^{18}F per one equivalent photon (reduced to one initial electron) was calculated by the formula (1) [22]:

$$Y = \frac{S\cdot(t_R/t_L)\cdot\lambda}{I_\gamma\cdot N_n\cdot N_\gamma\cdot\varepsilon_\gamma\cdot(1-e^{-\lambda t_1})\cdot e^{-\lambda t_2}\cdot(1-e^{-\lambda t_R})}, \quad (1)$$

where S is the peak area of with an energy of 511 keV; ε_γ is the detection efficiency of γ-quanta with an energy of 511 keV; $I\gamma$ is the relative intensity of the γ-line with an energy of 511 keV – 193.46% [27]; λ is the decay constant of the ^{18}F radioisotope (λ = *0.693/T(1/2), T(1/2)* – half-life – 109.77 min [27]); t_1 is the irradiation time; t_2 is the time between the end of the irradiation and the beginning of the measurements (cooling time); t_R and t_L are the real and live time of the gamma spectroscopic measurement; N_n is the number of ^{19}F nuclei per cm^2; N_γ is the integral number of photons with energy > 11 MeV interacting with the Teflon sample.

Measurements of the indicated activity of the ^{18}F radioisotope in Teflon samples were carried out on the ORTEC gamma spectrometric complex with an HPGe detector (volume – 150 cm^3, energy resolution ~ 2.45 keV for the ^{60}So – 1332.5 keV line) [29] within 120 minutes after the end of the irradiation. The statistical uncertainty of measurements did not exceed 5% for the entire time interval.

The WINSPECTRUM [30] was used to process the measured spectra.

Calculations of the correction for the self-absorption of the 511 keV gamma line in Teflon samples depending on their thickness F_s were carried out according to formula (2) [31]:

$$F_s = \frac{1-e^{-\mu(E)x}}{\mu(E)x}, \quad (2)$$

where $\mu(E)$is the energy-dependent mass absorption coefficient (cm^2/g); x is the product of the density of the material by the "effective" thickness of the sample (g/cm^2). The computer program Phy-X/PSD was used for the calculation of μ(511 keV) = 0.084 cm^2·g^{-1} [32]. The numerical value of the density of Teflon was 2.2 g/cm^3 [24].

1.3. EVALUATION OF THE EFFECTIVENESS OF THE ^{19}F(γ,n) REACTION STIMULATION BEAM

Although the results of simulations of the parameters of photonuclear reaction stimulation schemes provide information about the spectral composition and the integral number of photons falling on the irradiated samples, they do not take into account the process of interaction with the samples (energy dependences of photonuclear reaction cross sections).

Evaluation of the efficiency coefficient ε_{eff} of the use of bremsstrahlung photon beam for the used scheme of stimulation of the ^{19}F(γ,n) reaction on the M-30 microtron, taking into account the bremsstrahlung photon spectrum and cross-section of the specified reaction [7] was carried out according to formula (3) [20]:

$$\varepsilon_{eff} = \frac{1}{N_e\sigma_{max}T_0}\int_0^{E_{\gamma max}}\Phi_\gamma(E)\sigma(E)EdE, \quad (3)$$

where N_e is the number of initial electrons; $\Phi_\gamma(E)$ = *dN/dE* is the spectral density of bremsstrahlung photons; *σ(E)* is the ^{19}F(γ,n) reaction cross section [7]; σ_{max} is the maximum value of the reaction cross section; $E_{\gamma max}$ is the maximum energy of bremsstrahlung photons.

2. RESULTS AND DISCUSSION

As a result of the simulations, the integral values of the fluxes of photons and residual electrons (normalized to one electron), which interact with the Teflon sample (energy > 11 MeV), were established – 0.0357 photons/e and 0.49543 electrons/e, respectively. Photoneutrons did not affect the results of experimental studies [7, 19, 23].

The results of modeling the spectrum of bremsstrahlung photons interacting with a Teflon sample, according to the experimental scheme (see Figs. 2 and 3), are presented in Fig. 1.

When using an aluminum filter, the integral value of the flow of bremsstrahlung photons with energies from 11 MeV to the maximum energy value of the spectrum is 0.0334 photons/e. That is, 6.4% of bremsstrahlung photons are absorbed, which practically does not affect the flow of high-energy photons that initiate the ^{19}F(γ,n)-reaction. At the same time, 0.3114 electrons / e hit the sample, of which 1.682E-4 with energy > 11 MeV, that is, 51.46% of all electrons are absorbed, and with energy > 11 MeV – 99.66%.

Based on the simulation results, the profiles of the bremsstrahlung photon beams generated by the Ta-converter and the residual electrons were obtained in a section of a plane measuring 100 x 100 cm at the place of installation of the Teflon sample. Using the Origin package [34], figures were created with a probability map of the photon distribution on a plane based on the inverse cumulative Gaussian distribution.

Profiles of bremsstrahlung photons and residual electrons that hit the plane in place of the Teflon sample installation (100 x 100 cm) without and with the use of an Al filter are presented in Figs. 4 and 5.

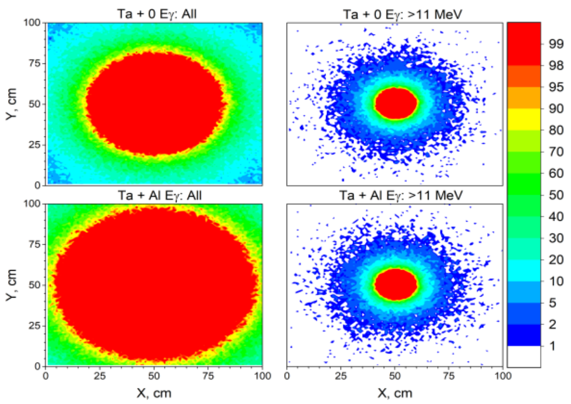

Fig. 4. Profiles of bremsstrahlung photons on the plane in the irradiated Teflon sample installation place, formed by the Ta-converter and the Ta-converter with an Al-filter at an initial electron energy of 17.5 MeV

It can be seen from the figures that high-energy photons have a smaller angular distribution, which leads to their greater concentration in the place of installation of the sample, i.e., in the center of the plane. This is consistent with the results of simulations obtained in [6, 22, 29]. After the aluminum filter, the profile plane of the bremsstrahlung photon beam increases due to the formation of low-energy photons in its material. However, since part of the high-energy spectrum of bremsstrahlung photons (> 11 MeV) remained almost unchanged, the profile picture for bremsstrahlung photons > 11 MeV also did not change.

The resulting profiles of residual electrons falling into the plane at the sample's location confirm the aluminum filter's electron absorption properties [19, 23, 29].

As a result of the experimental studies carried out by the method of induced activity [22], the dependence of the yields of the ^{18}F radioisotope from Teflon samples (normalized by one e) on the activation time of 20, 40, and 60 min with a fixed value of thickness 0.3 mm and a total weight of 0.2514; 0.2613, and 0.2538 g, respectively, as well as dependence on the thickness of the samples 0.1; 0.2, and 0.3 mm (the weight of which is 0.0892; 0.1646, and 0.2538 g, respectively) with a fixed activation time of 60 min were established.

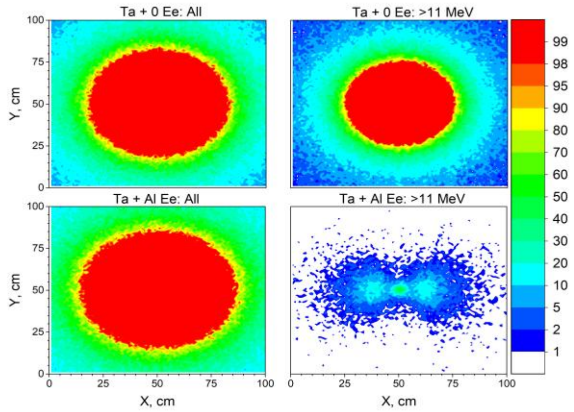

Fig. 5. Profiles of residual electrons on the plane in the irradiated Teflon sample installation place, formed by the Ta-converter and the Ta-converter with an Al-filter at an initial electron energy of 17.5 MeV

Calculations were made according to formula (1) [22]. The main input parameters were the experimentally obtained values of S (the area of the gamma-line peaks – 511 keV of the ^{18}F radioisotope) and N_γ (simulated integral values of the bremsstrahlung spectra that stimulate the ^{19}F(γ,n) reaction).

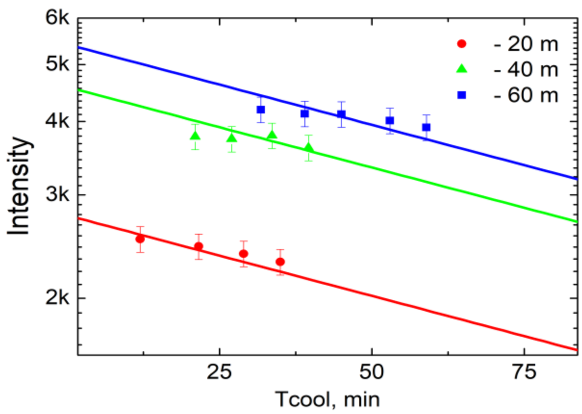

Fig. 6. Dependence of the relative intensity of the gamma line of the ^{18}F radioisotope, formed during the activation of Teflon samples (time of activation: ● – 20 min, ▲ – 40 min, and ■ – 60 min), from their cooling time

As an illustration, Fig. 6 shows the dependence of the intensity (values of the peak area are normalized for the time of measurements) of gamma lines with an energy of 511 keV, emitted by the radioisotope ^{18}F and formed during the activation of Teflon samples with a fixed thickness of 0.3 mm, on the time of their cooling (exposure). The measurement time of all samples was 5 min. For each sample, at least 4–5 measurements were taken over a period of 60 min. Experimental values for different activation times are marked: ● – 20 min, ▲ – 40 min, and ■ – 60 min, respectively.

The statistical uncertainty of spectrometric measurements did not exceed 5%.

The results of theoretical calculations, represented by solid lines, agree with the obtained data within the total uncertainty of the experimental results, the value of which did not exceed 5%.

When calculating the yields of the ^{18}F radioisotope formed during the activation of Teflon, a correction was introduced for the self-absorption of gamma rays with an energy of 511 keV in Teflon samples according to formula (2) [32]. Its numerical value varies from 0.9991 to 0.9908 depending on the thickness of the samples: 0.1…1.0 mm (self-absorption is 0.092…0.918%, respectively).

Fig. 7 presents the dependence of ^{18}F radioisotope yields (normalized to 1e) on the activation time of Teflon samples with fixed geometric dimensions and mass.

The results of theoretical calculations, represented by solid lines, agree with the obtained data within the total uncertainty of the experimental results, the value of which did not exceed – 13%.

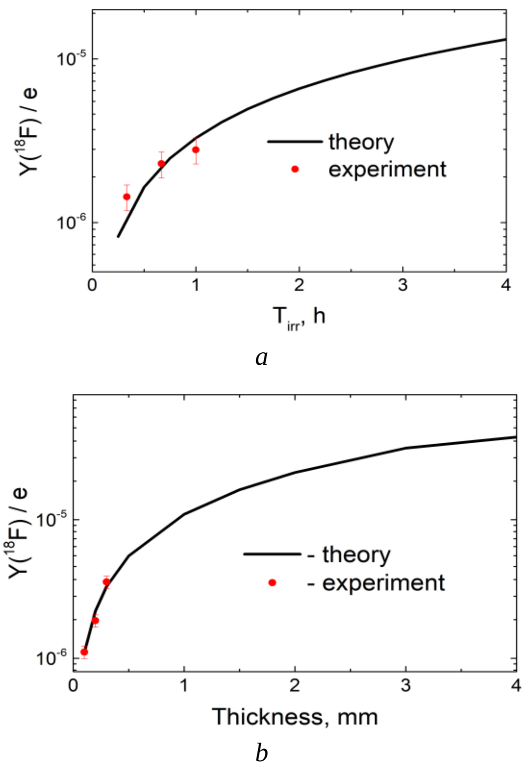

Fig. 7. Dependence of the integral values of the ^{18}F radioisotope yields (a) on the irradiation time of a Teflon sample, 0.3 mm thick, and (b) on the sample thickness at a fixed activation time of 1 h

As a result of the simulations, the numerical value of the efficiency evaluation of the bremsstrahlung photon beam usage ε_{eff} for the proposed optimized scheme of stimulation of the ^{19}F(γ,n)-reaction on the M-30 microtron was established according to formula (3) [20], which is 3.125E-5.

From the obtained values *of* ε_{eff}, as well as literature data [6, 20], it was established that increasing the output of the ^{18}F radioisotope (or increasing the efficiency of using the initial electron beam) for the proposed stimulation scheme, first of all, it is necessary to use the maximum initial electron flow (maximum current of the electron output beam), optimize the irradiation time (for example: during the activation time equal to the 3 half-life periods of the ^{18}F radioisotope, 75% of the ^{19}F nuclei undergo the activation process, and when the time is further increased to 5 periods – 98% [6]), and the geometric dimensions of the activated sample, both in the plane (perpendicular to the axis of the initial electron beam) and in thickness [6, 11].

As a result of the conducted simulations, the dependence of the integral values of the bremsstrahlung photon beams with energies > 11 MeV interacting with the activated Teflon samples on their geometric dimensions, namely the radius, was established.

It was found that when the radius of the irradiated sample increases to 10 cm (the limit of the geometric distribution of high-energy photons, see Fig. 4), the effectiveness of the bremsstrahlung beam for this irradiation scheme will increase. A further increase in the radius of the target will lead to a decrease in the efficiency of the beam, which is associated with going beyond the energy angular distribution of high-energy photons and increasing the number of low-energy particles.

CONCLUSIONS

As a result of experimental studies carried out within the framework of the method of induced activity, the output of the radioisotope ^{18}F, formed during the irradiation of polytetrafluoroethylene (Teflon) with the maximum bremsstrahlung photon energy of 17.5 MeV was determined.

The results of computer modeling of the output of the ^{18}F radioisotope using the GEANT4 toolkit agree with the obtained experimental result within the total uncertainty of the experimental value.

^{18}F radioisotope was evaluated, taking into account the cross-section of the ^{19}F(γ,n)-reaction for the optimized stimulation scheme used, and the possibilities of increasing the output of the ^{18}F radioisotope were analyzed.

The proposed approach can be used to optimize photonuclear reaction stimulation schemes at the M-30 microtron for a wide range of nuclei and predict the yields of the radioisotopes formed.

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ВИХІД РАДІОІЗОТОПУ ^{18}F ПРИ АКТИВАЦІЇ ПОЛІТЕТРАФТОРЕТИЛЕНУ НА МІКРОТРОНІ М-30

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Представлено експериментальні та промодельовані значення виходу радіоізотопу ^{18}F, утвореного при опроміненні політетрафторетилену (тефлону) гальмівними фотонами з граничною енергією 17,5 МеВ. Визначення виходу проводилося у рамках методу індукованої активності. Процес активації проводився на мікротроні М-30 Інституту електронної фізики НАН України. Виходи радіоізотопу ^{18}F для оптимізованої схеми стимуляції (γ,n)-реакцій моделювалися інструментарієм GEANT4 з врахуванням технічних характеристик вузла виводу електронів мікротрона М-30. Проведена оцінка ефективності використання сформованого пучка гальмівних фотонів для утворення радіоізотопу ^{18}F з врахуванням перерізу реакції для використаної схеми опромінення. Проаналізовано можливості збільшення виходу радіоізотопу ^{18}F шляхом оптимізації часу активації зразків та їх геометричних розмірів. Запропонований підхід може бути використаний для оптимізації схем стимуляції фотоядерних реакцій на мікротроні М-30 для широкого кола ядер та прогнозування виходів утворених радіоізотопів.