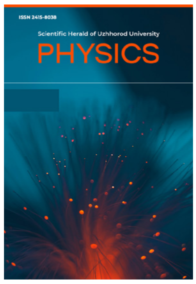
Scientific Herald of Uzhhorod University
Series “Physics”

Journal homepage: https://physics.uz.ua/en
Issue 52, 16-26
Received: 09/15/2022. Revised: 10/28/2022. Accepted: 11/17/2022



UDC 539.1.09, 658.562: 621
PACS 29.17.+w, 29.27._a
DOI: 10.54919/2415-8038.2022.52.16-26

Optimal scheme for stimulating photofission of shielded nuclear materials on the Microtron M-30: a combination of theoretical and experimental studies

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Abstract

Relevance. The photofission of shielded nuclear materials is widely used to develop methods for non-destructive analysis of their isotopic composition. To stimulate the photofission reaction, bremsstrahlung beams obtained on electronic accelerators are used. Their parameters significantly depend on the design features of accelerators and sample activation schemes.

Purpose. Theoretical modelling of parameters of an optimal scheme for stimulating the photofission of shielded nuclear materials on an electronic accelerator – Microtron M-30 for the analysis of their isotopic composition, considering its technical characteristics, and experimental verification of its parameters.

Methodology. Theoretical calculations of the parameters of bremsstrahlung beams for the Microtron M-30 were carried out using the GEANT4 toolkit. For experimental studies of the influence of structural elements of the optimal stimulation scheme on the integral characteristics of inhibitory photons, secondary photoneutrons, the method of activation of detectors made of gold was used; for residual electrons – the transmission method based on a passage chamber and a Faraday cylinder.

Results. As a result of the combination of theoretical and experimental studies, optimal parameters of the activation scheme of nuclear materials on the Microtron M-30 have been established. The scheme provides experimental conditions under which the losses of bremsstrahlung photon beams interacting with the test samples (with energies ≥6 MeV) do not exceed 35% of their initial values, with the practical absence of residual electrons (98% of electrons are absorbed) and secondary photoneutrons (no more than 1E-9 n/e).

Conclusions. The parameters of the developed stimulation scheme for the Microtron M-30 can be applied to various types of accelerators, considering their design features, characteristics of the samples, and implemented activation schemes

Keywords: bremsstrahlung, residual electrons, secondary photoneutrons, GEANT4, activation detector, Faraday cylinder

Suggested Citation:
Pylypchynets I. Optimal scheme for stimulating photofission of shielded nuclear materials on the Microtron M-30: a combination of theoretical and experimental studies. *Scientific Herald of Uzhhorod University. Series “Physics”.* 2022;(52):16-26.

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Introduction

Control of the circulation and non-proliferation of nuclear materials (e.g. fertile – ^{232}Th, ^{238}U; divisible – ^{235}U, ^{239}Pu) is one of the most urgent tasks of nuclear science and technology [1]. Reliable information about their isotopic composition is required to successfully solve this problem. One of the methods for analysing the isotopic composition of nuclear materials, which is widely used and improved (developed) at the present time, is based on the use of information about gamma and neutron radiation from their decay products formed as a result of the photofission reaction [2-4]. As a rule, sealed containers made of stainless steel are used for the storage and transportation of nuclear materials [5; 6]. To stimulate the photofission of shielded nuclear materials, bremsstrahlung beams obtained at electronic accelerators (linear, microtrons) are widely used [1; 3; 5]. For efficient use of bremsstrahlung beams in the analysis, it is necessary that their energy is higher than the thresholds (γ,f) of reactions for nuclear materials, and less than the thresholds of possible photonuclear reactions that can occur (be generated) in the container material [7].

The outputs of bremsstrahlung photons and their energy spectra obtained by converting electron beams into bremsstrahlung (via the (e,γ) reaction channel) depend primarily on the initial electron energy, characteristics (material, geometric dimensions) of converters [8-10]. The electron spans in the materials from which the converters are made will be greater than the thickness at which the photon outputs have maximum values, i.e., residual electrons will be present in the beams [11].

Residual electrons, when interacting with the studied samples, can generate secondary photons [12] and photoneutrons in them (via (e,n) reaction channels) [13; 14]. The generation of secondary photons and photoneutrons is also possible when they interact with stainless steel screens (the material from which containers for nuclear materials are made) [15]. In this regard, there is a need to “clean” the bremsstrahlung beams from residual electrons and photoneutrons, since their presence can significantly affect the final results of the analysis. To reduce the number of residual electrons interacting with the studied samples, without significantly weakening the intensity of bremsstrahlung photons, filters are used – materials with a small charge number. Boron carbide (B_4C) as a material for the manufacture of filters meets these criteria (absorbs residual electrons, secondary photoneutrons without significant deformation of the photon spectrum) [16; 17].

Notably, the quantitative (integral) content of photons, residual electrons and secondary photoneutrons and their ratios in the bremsstrahlung beams interacting with the studied samples of shielded nuclear materials are significantly influenced by geometric factors (the relative position of the converter, the studied sample relative to the axis of the initial electron beam), and the presence of additional structural elements (filters, screens, collimators) used in their generation [19]. In addition, the final parameters of the bremsstrahlung beams will be affected by the design features of devices (nodes) for the output of electron beams into the air, which differ significantly for different types of electronic accelerators [20]. Therefore, these factors should be considered when developing sample activation schemes.

To obtain information about the characteristics of bremsstrahlung beams that interact with the studied samples obtained on electronic accelerators, considering their design features and irradiation schemes, computer modelling is widely used using Monte Carlo programme codes (MCNP6, FLUKA, GEANT4) [21; 22], since experimental studies (for example: activation, track detectors, TLD dosimeters, proportional counters) require expensive and long-term measurements [23; 24].

Purpose of the study: development of an optimal stimulation scheme (generation of bremsstrahlung beams with the maximum content of photons with energies above the thresholds of the photofission reaction with the minimum content of residual electrons and secondary photoneutrons) of the photofission of shielded nuclear materials on the electronic accelerator of the Institute of electronic physics of the National Academy of Sciences (IEP NAS) of Ukraine – Microtron M-30, considering its technical characteristics, for the needs of analysing their isotopic composition, and experimental verification of its parameters.

Materials and Methods

Modelling of parameters of bremsstrahlung beams for the Microtron M-30 electronic accelerator using GEANT4 toolkit

To simulate the spectra of photons, residual electrons, and secondary photoneutrons that fall into the plane of location (installation) of test samples (or on the samples themselves) perpendicular to the beam axis of the primary electrons, and their angular distribution (which allowed visualising their profile images in the plane along the x and y axes perpendicular to the z axis), a C++ programme was developed using the GEANT4 class library [25]. The choice of this toolkit was conditioned by its suitability for modelling the processes of generation of various types of particles and their interaction with the studied samples at electronic accelerators [1; 10; 21], and its availability in terms of free use. The programme provided for the possibility of considering the primary parameters: electron beams (initial geometric dimensions (point source, parallel beam) and beam distribution (Gauss, equally probable)); the presence of converters (converters of electrons to photons) and filters for its generation; samples (particle detectors), and considering their location in three-dimensional space, which allowed reproducing the real irradiation schemes of the samples implemented on electronic accelerators (in this case, on the electronic accelerator of the IEP of the National Academy of Sciences of Ukraine – Microtron M-30 [26]).

The programme was implemented under the Windows platform using multi-threading mode. A computer with a 6-core Intel(R) Core(TM) i7-9750H (2.60 GHz) processor and 36 GB of RAM was used for calculations.

Calculations were performed at the initial electron energies of 12.5 MeV [7] for the geometric dimensions of the output beam, which had the shape of an ellipse with axes of 22×6 mm [27].

Figure 1 shows a scheme for stimulating the photofission reaction of shielded nuclear materials on the Microtron M-30, where 1 – Microtron M-30 output node; 2 – Ti window; 3 – photon converter ((Ta, 52×45×(0.1÷4 mm); 4 – filter B_4C (diameter – 30 mm, thickness – 19 mm); 5 – stainless steel screen (diameter = 23 mm, thickness = 0.1÷9.5 mm); 6 – sample of nuclear material.

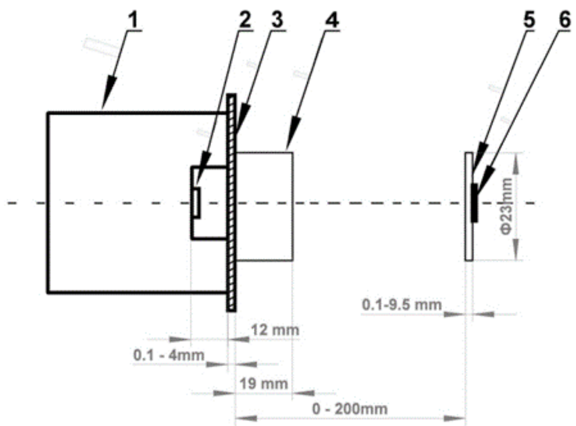

Figure 1. Scheme for stimulating the photofission reaction of shielded nuclear materials on the Microtron M-30

In all calculations, an equally probable distribution of initial electrons along the plane was set (considering the design features of the electron output unit – a titanium window with a thickness of 0.05 mm) [27]. Distance from the output node to the tantalum (Ta) converter (dimensions: 52×45×T mm [T=0.1; 0.5; 1.0; 1.5; 2.0; 3.0; 4.0 mm]) was 12 mm. The test sample (nuclear material; radius – 5.6 mm) was fixed perpendicular to the beam axis at a distance of 0; 25; 50; 75; 100; 125; 150; 175; 200 mm from Ta of the converter. Additionally, the influence of the presence of structural materials (B4C filter (diameter – 30 mm, length – 19 mm) [16], stainless steel screen (0÷9.5 mm)) in schemes for stimulating the photofission of nuclear materials to the final characteristics of bremsstrahlung (the ratio of photons, residual electrons, photoneutrons) was studied. The relative absorption coefficients of photons and electrons (A) were determined by the equation

$$A = (1- I/I_0)\cdot 100\ \% \qquad (1)$$

where I and I_0 – integral values of particle fluxes (photons or electrons) that interact with the sample after passing through the layer with the absorbing material and in its absence.

Experimental studies of the influence of structural elements of the photofission stimulation scheme on the parameters of bremsstrahlung beams

Experimental studies of the effect of a combination of structural elements (Ta converter, B_4C filter, stainless steel screen) of schemes for stimulating the photofission of nuclear materials to the final parameters of bremsstrahlung beams (the ratio of photons, residual electrons, secondary photoneutrons) interacting with the samples under study were performed by the transmission method [27; 28]. During the experiment, integral values of photon fluxes and relative absorption coefficients of photons and electrons for a combination of structural elements were determined.

Photons and photoneutrons. For experimental determination of the integral characteristics of photons and photoneutrons in bremsstrahlung beams interacting with the sample, and their relative absorption coefficients for a combination of structural materials (B_4C filter (diameter – 30 mm, length – 19 mm) + stainless steel screen (diameter – 34 mm, thickness – 9.5 mm)) activation detectors containing gold isotope – ^{197}Au were used [24; 28; 29]. The geometric dimensions of the activation detectors (square shape, rib length – 6.6 mm) were close to the size of the samples of nuclear materials in terms of area. Photons (for the energy interval – 8.1÷12.5 MeV) and photoneutrons were determined by activation products ^{197}Au by channels of (γ,n)- reactions: ^{196}Au (gamma energy of the line Eγ = 355.7 KeV, its quantum output Iγ=87%, reaction threshold – 8.01 MeV) and for (n,γ)- reactions: ^{198}Au (Eγ = 411.8 KeV, Iγ = 95.6%) [30], respectively. The relative photon absorption coefficients were determined by the ratios of the gamma radiation intensity from the detector activation products ^{197}Au, irradiated with and without structural materials. The exposure time of the detectors was 15 minutes. Measurements of gamma radiation from generated radionuclides ^{196}Au and ^{198}Au was performed on the ORTEC spectrometric complex [28] for 200 hours. The measurement time of activation detectors was 25 minutes. At least 10 series of measurements were performed for each individual detector. The statistical error of spectrometric measurements did not exceed 3%. The measured intensity values were normalised to the integral values of the primary electron flow, the area of activation detectors, and the number of isotope nuclei of ^{197}Au contained in the detectors.

Electrons. Experimental determination of relative values of electron absorption coefficients separately for structural materials (Ta converter (thickness – 1 mm), B_4C filter (diameter – 30 mm, length – 19 mm), stainless steel screen (diameter – 34 mm, thickness – 9.5 mm)) and their combinations (Ta converter + B_4C + stainless steel), which were used to form photon beams to stimulate the photofission reaction of nuclear materials, were performed by the transmission method described in detail in [27; 28]. Measurements of the integral flow of electrons from the window of the accelerator output unit and the layers of structural materials that passed through and their combinations were carried out using a secondary emission monitor (pass-through chamber) and a Faraday cylinder, respectively. Electron detection was carried out in the plane of placement of the test samples.

Results and Discussion

As a result of the calculations, the dependences of the spectra of photons, residual electrons, and secondary photoneutrons (which were used to determine their integral characteristics and relations between them) interacting with the test sample on the thickness of the inhibitory target – tantalum (Ta) converter (0.1÷4.0 mm) and the distance from Ta converter to the test target (0÷200 mm) were studied. Figures 2 and 3 show the dependences of the spectra of photons and residual electrons normalised to one electron (1e) on the thickness of Ta converter at a fixed distance Ta converter – test sample = 100 mm, respectively.

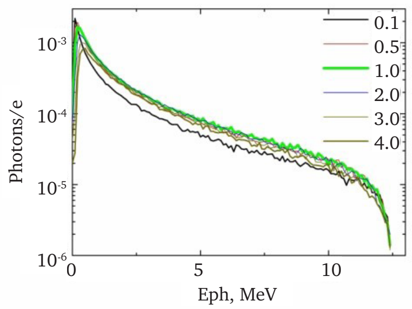

Figure 2. Dependences of bremsstrahlung photon spectra on Ta converter thickness

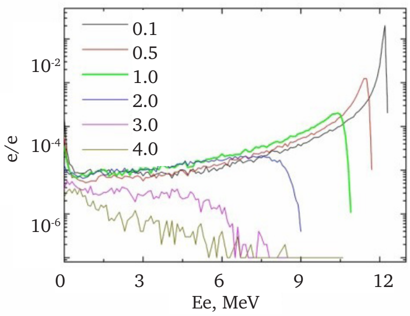

Figure 3. Dependences of residual electron spectra on Ta converter thickness

The relationship between the integral values of photon yields in the low-energy (with energies <6 MeV, which do not affect the sample activation process) and high-energy (with energies from 6 to 12.5 MeV, which stimulate the actinide photofission) parts of the spectra on the thickness of the inhibitory target is established. As the Ta converter thickness increase to ~1.0 mm, the values of the total integral photon outputs increase and reach a maximum value of – 2.11E-2, with a further increase in thickness to 4 mm, the values are constant. For photons with energies ≥6 MeV, the optimal thickness is 1 mm (1.92 E-3). The relationship between the integral values of secondary electron fluxes (which did not participate in the generation of bremsstrahlung photons) in the low-energy (with energies <6 MeV) and high-energy (with energies between 6 and 12.5 MeV, which can generate secondary photons when interacting with the sample) parts of the spectra also depends on the thickness of the Ta tconverter. As the Ta converter thickness increase from 0.1 to 4 mm, the values of the integral fluxes of residual electrons (with energies <6 MeV) decrease from 3.77E-2 to 3.3E-6, respectively. With a thickness of 1 mm, it amounts to 3.36E-3.

Dependences of the spectra of bremsstrahlung photons and residual electrons (normalised to 1e) entering the sample on the distance Ta converter – sample (0÷200 mm) with a fixed thickness of the Ta converter = 1 mm are shown in Figures 4 and Figure 5, respectively. The relations between the integral values of photon fluxes and secondary electrons in the low- and high-energy parts of the spectra depend on the distance. As it increases from 0 to 200 mm, the values of Integral photon fluxes and residual electrons (with energies <6 MeV) decrease from 4.21E-2 to 6.69E-4, and from 3.26E-2 to 8.24E-5, respectively.

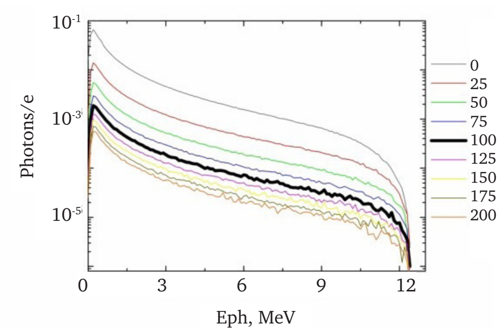

Figure 4. Dependences of bremsstrahlung photon spectra on distance Ta converter – sample

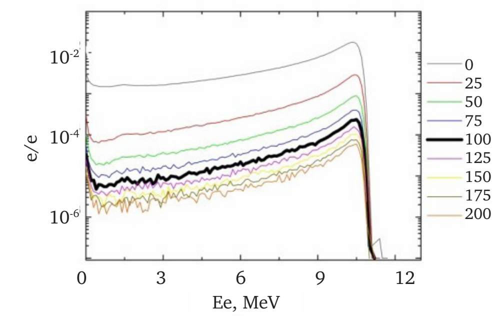

Figure 5. Dependences of residual electron spectra on distance Ta converter – sample

Figure 6 shows the spectrum of secondary photoneutrons generated by the interaction of electrons with Ta converter (normalised to 1e). The numerical value of the integral photoneutron flux is normalised to 1e – 9.3E-6. The integral value of the photoneutron flux interacting with the test sample did not exceed – 1E-8 photoneutrons per 1e, which is consistent with the calculation results in [9; 13].

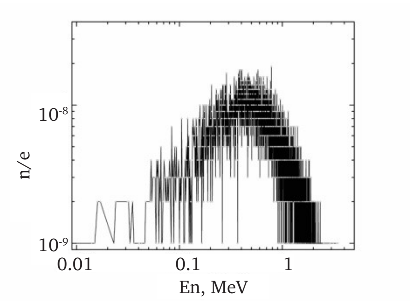

Figure 6. Energy spectrum of photoneutrons

The obtained results reproduce the general patterns that are characteristic of the dependence of photon outputs on the thickness of converters and the influence of geometric factors on the activation process and are consistent with existing data [9; 10; 13], and their difference is associated with the use of different input parameters during simulations [19; 20], which indicates the need to take them into account.

The calculations established the value of the optimal thickness of Ta converter, at which the integral output of the generated photons (in the beam of bremsstrahlung) is the maximum, and the distance Ta converter – test sample, which provides the best ratio between photons, residual electrons, and secondary photoneutrons interacting with the sample (the maximum integral value of the photon flux at the minimum values of electrons, photoneutrons). The optimal thickness of the Ta converter is 1 mm at a fixed distance Ta converter – sample installation plane is 100 mm. Integral values of photon fluxes and residual electrons with energies ≥6 MeV (normalised to 1e) are – 1.92 E-3 and – 3.36E-3, respectively. These values for photons and electrons are 19.9 and 98.5 times less than the zero distance (when the sample is placed close to the converter). A further increase in the distance from 100 mm to 200 mm reduces the value of integral electron fluxes by only 4.0 times.

Notably, when using such a sample activation scheme, additional purification of the bremsstrahlung beam from residual electrons is necessary [16; 28]. Therefore, calculations were made for the absorption of bremsstrahlung photons and residual electrons by a filter made of B_4C [16]. Figures 7 and 8 show the spectra of photons and residual electrons (normalised to 1e) interacting with the test sample in the presence of B_4C filter and its absence for a fixed distance from Ta converter (thickness – 1 mm) – test sample = 100 mm, respectively. It is established for the first time that the total number of electrons and photons absorbed by a B_4C filter – 79.64% and 14.7%, respectively, with energies ≥6 MeV – 99.8% and 9.3%.

The obtained results reflect the general regularities of the relative absorption coefficients of photons and electrons (for the energy region – 6.5÷12.5 MeV) by a conventionally used filter (reactor graphite) in the generation of bremsstrahlung photon beams to stimulate photonuclear reactions [28]. However, for B_4C filter, the values of the electron absorption coefficients are higher and the photons are less than those of reactor graphite, which confirms the effectiveness of its use for stimulation schemes.

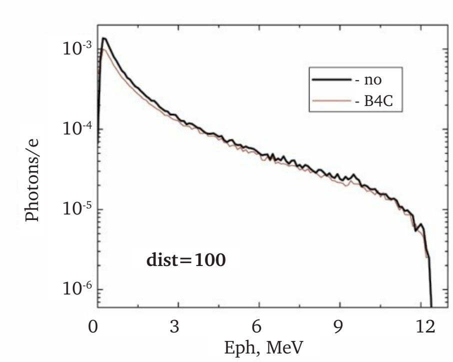

Figure 7. Spectra of photons interacting with the sample without and with B_4C filter.

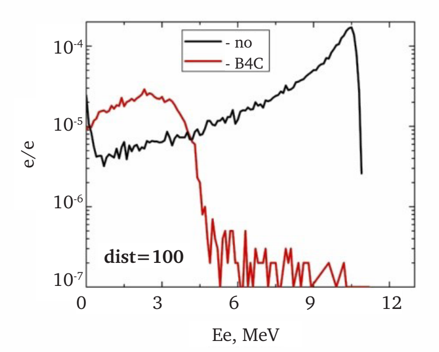

Figure 8. Spectra of electrons interacting with the sample without and with B_4C filter

Additionally, simulations of the influence of the thickness (0.1, 0.5, 1.0, 3.0, 5.0, 7.0, 9.5 mm) of the stainless steel screen on the final parameters of the bremsstrahlung beam (photons, residual electrons) interacting with the sample were carried out. Figures 9 and 10 show the dependences of the spectra of photons and residual electrons interacting with samples at a fixed distance Ta converter (thickness – 1 mm) – test sample = 100 mm, on the thickness of the stainless steel screen.

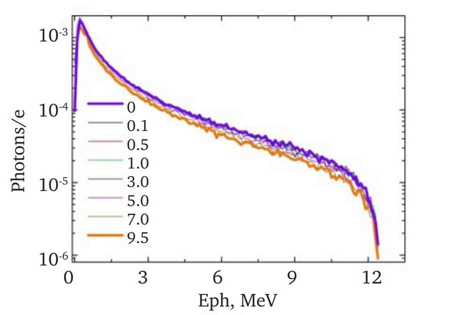

Figure 9. Dependences of the spectra of photons interacting with samples on the thickness of the stainless steel screen

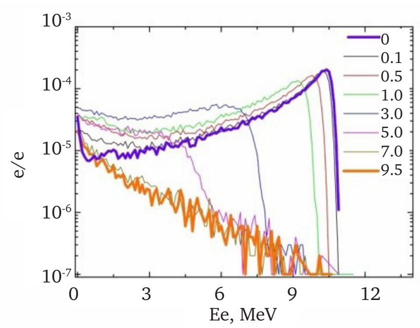

Figure 10. Dependences of the spectra of residual electrons interacting with samples on the thickness of the stainless steel screen

Values of the relative absorption coefficients (according to equation 1) of photons and electrons with energies ≥6 MeV, depending on the screen thickness, are shown in Figure 11. For the first time, it was found that when the screen thickness is more than 5 mm, 99% of electrons and 15% of photons are absorbed. With a further increase in the screen thickness to 9.5 mm, 28% of photons are absorbed.

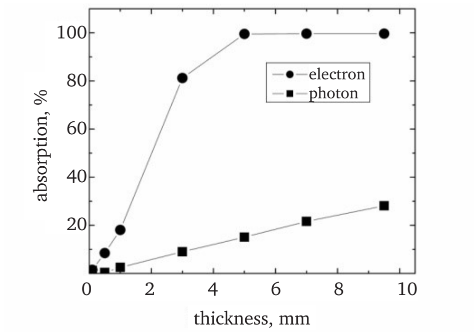

Figure 11. Dependence of photon and electron absorption (with energies ≥6 MeV) on the thickness of the stainless steel screen

The calculations were used as input data for modelling the parameters of the optimal scheme for stimulating the photofission of nuclear materials on the Microtron M-30, considering the technical features of its electron output unit and the characteristics of samples. The activation scheme for shielded (thickness – 9.5 mm – samples of nuclear materials consists of Ta converter (thickness – 1 mm) and B_4C filter. The distance from Ta converter to the test sample – 100 mm. The influence of the structural elements of the activation scheme on the final shape of the spectra of photons and residual electrons that fall on the samples was investigated (Figures 12 and 13, respectively).

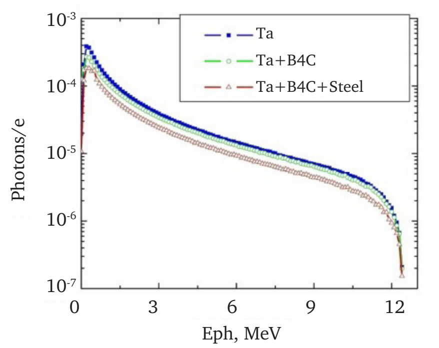

Figure 12. Spectra of photons interacting with samples for optimal stimulation scheme

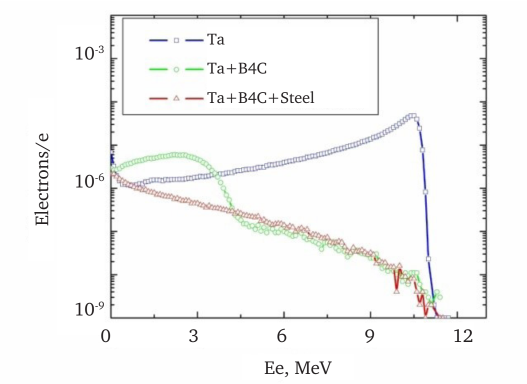

Figure 13. Spectra of residual electrons interacting with samples for optimal stimulation scheme

The integral values of photon fluxes, residual electrons normalised to one electron, falling on samples of shielded nuclear materials are – 2.87E-3 photons (with an energy >6 MeV – 2.86E-4) and 3.89E-5 electrons (with an energy >6 MeV – 2.454E-6). When performing calculations based on a given activation scheme, photoneutrons that fell on samples of shielded nuclear materials were not detected.

For the proposed scheme of stimulating the photofission, 7.87E-1% of photons and 1.48E-2% of electrons from their total number (those that fall into the plane of sample placement) fall on the samples. Similarly, they interact with samples (energies >6 MeV) – 1.09% of photons and 1.08E-3% of electrons, respectively.

Thus, the presented scheme allows almost completely excluding the interaction of residual electrons and secondary photoneutrons with the studied samples of shielded nuclear materials, that is, excluding the stimulation of additional channels of electro-nuclear ((e,γ)-, (e,n)-, (e,f)-) and neutron ((n,γ)-, (n,f)-) reactions [7], which may introduce additional errors in the final results of the analysis of their isotopic composition.

Comparison with the experiment

Photons. Value of integral photon beam fluxes (normalised to 1e), interacting with samples of nuclear materials, without structural elements (straight beam) and when combined with B_4C filter + stainless steel screen was 4.63E-5 and 3.08E-5, respectively. The obtained values are consistent with the results of calculations using the GEANT4 toolkit (4.32E-5 and 2.896E-5) within 6.69% and 5.97%.

Results of measurements of the dependence of the relative intensity values (which were used to determine the photon absorption coefficient) of gamma-rays of the generated radionuclide ^{196}Au from its cooling time are shown in Figure 14. Experimental values of gamma intensity of activation detectors for a direct beam of photons hitting the sample were circles, combinations of filter + steel screen – squares. The results of theoretical calculations [28] for identical experimental conditions (at the same activation, cooling, and measurement times) are represented by solid lines. The experimental values are consistent with the calculation results within 5%. The total measurement error did not exceed 10%. The main contribution to the error was made by the uncertainty of the number of isotope nuclei ^{196}Au, and was ~7%.

The error of the obtained experimental values can be further influenced by the factor associated with the process of irradiation of gold samples, namely, with the instability of the efficiency of the output of the primary electron beam from the microtron output node during their activation. Even with stable operation of the microtron, the output efficiency can vary from 85 to 90%, since it depends on the process of focusing the electron beam in the centre of the Ti window [26-28].

Photon absorption coefficient value for the combination B_4C filter + steel screen was – 33.3%, which was consistent with the calculation result of the GEANT4 toolkit (36.34%) within 9.1%.

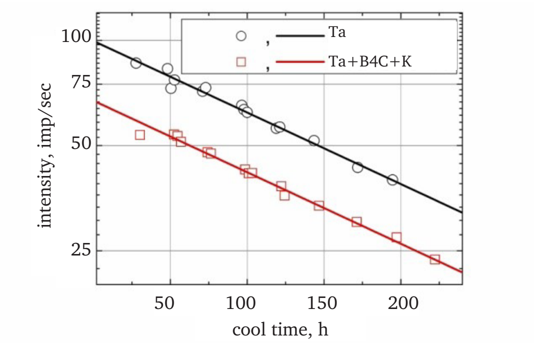

Figure 14. Dependences of the relative intensity of gamma radiation of a radionuclide ^{196}Au on cooling time

Photoneutrons. When performing spectrometric measurements of irradiated gold detectors placed in the plane of installation of test samples, lines from isotopes ^{198}Au, generated along the channel (n,γ)- reactions, the measured gamma-ray spectra are absent (within the background), and their value is less than the detection limit. This indicates the absence of photoneutrons in the bremsstrahlung beam interacting with test samples, which is consistent with the results of the calculations performed.

Electrons. Values of electron absorption coefficients for combinations Ta converter + B_4C filter, Ta converter + B_4C filter + steel – 83.74% and 98%, and are consistent with the results of calculations with GEANT4 toolkit (79.64% and 95.46%) in the range of 4.9% and 2.6%, respectively.

The results of experimental studies of the parameters of bremsstrahlung beams and the effect of structural elements of the activation scheme on their characteristics confirm the results of calculations performed using the GEANT4 toolkit.

A significant discrepancy between theoretical calculations and experimental results is associated with errors in nuclear data used in GEANT4 toolkit libraries [21; 25], affecting modelling accuracy.

The obtained results complement the existing information on the methods of generating bremsstrahlung beams with specified parameters on electronic accelerators to stimulate the photofission of nuclear materials as unshielded – ^{235}U, ^{238}U, ^{239}Pu [2]; ^{235}U, ^{238}U [3]; ^{235}U, ^{238}U, ^{239}Pu [4], and shielded, packed in stainless steel containers, – ^{232}Th [5]; ^{238}U, ^{239}Pu (additional studies were conducted for the case of a carbon steel container) [6].

These papers present the results of both theoretical calculations performed using Monte Carlo codes: MCNP6 [2] and a combination of PHITS 3/0 and MCNP6 [3] for initial electron energies of 8 and 7, and 13.5 MeV, respectively, and experimental studies conducted on linear accelerators for a wide energy range from 7 to 17.5 MeV (7 and 9 [6]; 10 [5] and 17.5 [4]) at average peak electron currents on bremsstrahlung converters from 30 mA [5] to 100 mA [4; 6]. Metal plates of tantalum (Ta) with a thickness of 0.5 mm [3] and 2 mm [5] and tungsten (W) with a thickness of 5 mm [4] and 1.8 mm [6] were used as converters for generating bremsstrahlung.

Notably, the numerical values of the electron flux currents generated by microtrons are an order of magnitude less than on linear accelerators due to differences in their principle of operation [1]. Therefore, this factor must be considered when developing schemes for stimulating the photofission of nuclear materials for conducting the procedure for their isotope identification.

Cylindrical lead (Pb) collimators were used to focus them on the studied samples of shielded [6] and unshielded [4] nuclear materials during their activation. The need for their use was conditioned by the use of schemes for online detection of gamma and neutron radiation from the decay products of nuclear materials, during the procedure for their isotope identification. Since detectors require protection (the geometric dimensions of which must be cons when placing the studied samples when activating them) from the parasitic background of bremsstrahlung photons and neutrons formed during the operation of linear accelerators [5]. However, their use leads to a weakening of the bremsstrahlung beams and their additional contamination with photoneutrons [19]. In contrast, in [5], during the activation of samples, an uncollimated beam of bremsstrahlung was used, which provided more efficient use of the initial electron flux.

Monte Carlo codes were used to model the characteristics of the bremsstrahlung photon spectra obtained at linear electron accelerators: FLUKA [5] MCNP6 [4; 6]. Only in [4], experimental studies of the characteristics of bremsstrahlung photon fluxes at a maximum energy of 17.5 MeV were conducted using activation detectors made of Au (Gold), Ni (nickel), U (uranium), Zn (zinc), and Zr (zirconium).

When electrons (E_e=7÷10 MeV [3; 5; 6]) interact with Ta [3; 5] and W [6] converters, the fluxes of secondary photoneutrons generated through the channels of electro and photonuclear reactions [9; 13] did not affect the final results of the analysis of the isotopic composition of nuclear materials.

In the case of interaction of electrons (E_e=17.5 MeV) with a W converter with a thickness of 5 mm [4], the resulting secondary photoneutrons can additionally initiate channels of neutron fission reactions (n,f) of nuclear materials, since their cross-sections (for the energy region of fast neutrons, which are the main components of the energy spectra of photoneutrons [9]) are approximately several times larger than the cross-sections of photofission reactions [31], which can affect the accuracy of the analysis of their isotopic composition. Therefore, when activating samples of nuclear materials on a linear accelerator [4], a combined filter was used to purify the bremsstrahlung beam from the photoneutrons present in it, generated by the interaction of electrons (E_e=17.5 MeV) with the W converter and additionally generated by interaction with the Pb collimator. The filter consisted of layers of boron polyethylene (thickness – 200 mm) and cadmium (thickness – 2 mm), which allowed almost completely protecting test samples from interaction with photoneutrons [4].

In the above studies on the use of photofission stimulation schemes for unshielded [2; 3; 4] and shielded [5; 6] nuclear materials, the interaction of residual electrons with the samples under study was not analysed.

The most important indicator of the effective use of activation schemes of nuclear materials in the analysis of their isotopic and quantitative composition is information on the numerical values of the outputs of photofission reactions ((number of acts of photofission)/(1e)·(gram)), which occurred during their stimulation by beams of bremsstrahlung on electronic accelerators [7]. The percentage of occurrence of photofission acts (i.e., the number of photons and neutrons generated from the decay products of nuclear materials used in the analysis process [2; 3; 4]) affects the lower weight limit of their identification.

As a result of the analysis of the yield dependence ((number of photofission acts)/(1e) * (gram)) of the photofission reaction of the ^{238}U nucleus from the energy of electrons (which, when interacting with the inhibitory target, generate high-energy photons), it was found that the numerical values of the yield change by about 4 orders of magnitude for the energy region from ~6 (reaction threshold) to ~20 MeV, and for the energy region ~(6÷13) MeV – by more than 3 orders of magnitude [7]. Therefore, a further increase in the energy of accelerated electrons leads to a change in the yield by less than an order of magnitude.

The number of photofission acts of nuclear materials depends on the energy and quantitative parameters (fluxes per second) of bremsstrahlung beams obtained at electronic accelerators, the nuclear and physical characteristics of the samples under study, since it directly depends on the cross-sections of photofission [1; 31], and on their weight.

The weights of the test samples used in the process of stimulating the photofission of nuclear materials for unshielded samples were: 1.5 g – ^{235}U, 2.1 g – ^{238}U, 0.47 g – ^{239}Pu [2]; 1.9 g – ^{235}U + ^{238}U [3]; 1.0÷1e+2 g – ^{235}U, 1e+2÷1e + 3 g – ^{238}U, 1.0 g – ^{239}Pu [4]; for shielded samples: 15.6 g – ^{232}Th [5]; 241 G – ^{238}U [6]. Their use allowed obtaining statistically accurate and reliable results when analysing the isotopic composition.

When performing the procedure for identifying nuclear materials directly in stainless steel containers, it is necessary to consider the attenuation of photons and neutrons generated during photofission when passing through a steel container. The solution to this problem is achieved by increasing the weight of the studied samples and the current strength of electronic flows generated by accelerators.

The proposed stimulation scheme allows more efficient use of the initial electron beam (the number of photons with energies ≥6 MeV interacting with the sample formed at 1e) to stimulate the photofission of shielded nuclear materials, i.e., reduces the cost of the analysis procedure.

Conclusions

The use of a combination of theoretical and experimental studies establishes optimal parameters of the scheme for stimulating the photofission of shielded nuclear materials on an electronic accelerator – Microtron M-30 for analysing their isotopic composition. Namely, to obtain the information necessary for the generation of bremsstrahlung beams with the maximum photon content with the minimum (or practically absent) content of residual electrons, secondary photoneutrons interacting with test samples, with mandatory consideration of their characteristics, design features of the Microtron M-30, and the activation scheme. This approach can be used to develop schemes for photonuclear activation analysis of the isotopic composition of special materials for various types of electronic accelerators.

The conducted studies allow predicting the sensitivity of the method for identifying nuclear materials depending on the strength of electron beam currents during sample irradiation. And, for the first time, the established values of the relative absorption coefficients of photons and electrons with energies ≥6 MeV, depending on the thickness of the stainless steel screen, allow adjusting the parameters of the photofission schemes of nuclear materials in containers of arbitrary geometric shape.

For the first time, the values of the relative absorption coefficients of photons and electrons with energies ≥6 MeV were established by filters made of B_4C, allow developing more efficient schemes for generating bremsstrahlung photon beams to stimulate photonuclear reactions compared to conventionally used filters.

This approach can be used to develop schemes for photonuclear activation analysis of the isotopic composition of special materials for various types of electronic accelerators.

Funding

The work was partially carried out within the framework of the project of research works of young scientists of the National Academy of Sciences of Ukraine in 2021-2022. State registration number – 0121U111832).

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Оптимальна схема стимуляції фотоподілу екранованих ядерних матеріалів на мікротроні М-30: комбінація теоретичних та експериментальних досліджень

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Анотація

Актуальність. Реакція фотоподілу екранованих ядерних матеріалів широко використовується для розробки методів неруйнівного аналізу їх ізотопного складу. Для стимуляції реакції фотоподілу використовуються пучки гальмівного випромінювання, отримані на електронних прискорювачах. Їх параметри суттєво залежать від конструктивних особливостей прискорювачів та схем активації зразків.

Мета. Теоретичне моделювання параметрів оптимальної схеми стимуляції реакції фотоподілу екранованих ядерних матеріалів на електронному прискорювачі – мікротроні М-30 для потреб аналізу їх ізотопного складу, з врахуванням його технічних характеристик, та експериментальна перевірка її параметрів.

Методологія. Теоретичні розрахунки параметрів пучків гальмівного випромінювання для мікротрону М-30 проводилися з застосуванням інструментарію "GEANT4". Для експериментальних досліджень впливу конструктивних елементів оптимальної схеми стимуляції на інтегральні характеристик гальмівних фотонів, вторинних фотонейтронів використано метод активації детекторів із золота; для залишкових електронів – метод пропускання на базі прохідної камери та циліндру Фарадея.

Результати. У результаті поєднання теоретичних і експериментальних досліджень встановлено оптимальні параметри схеми активації ядерних матеріалів на мікротроні М-30.Схема забезпечує експериментальні умови, за яких втрати пучків гальмівних фотонів, що взаємодіють із досліджуваними зразками (з енергіями $\geq$ 6 МеВ), не перевищують 35 % від їх початкових значень, при практичній відсутності залишкових електронів (98 % електронів поглинаються) і вторинних фотонейтронів (не більше 1Е-9 n/e).

Висновки. Параметри, розробленої схеми стимуляції для мікротрону М-30, можуть бути застосовані для різних типів прискорювачів при врахуванні їх конструктивних особливостей, характеристик досліджуваних зразків і реалізованих схем активації

Ключові слова: гальмівне випромінювання, залишкові електрони, вторинні фотонейтрони, GEANT4, активаційний детектор, циліндр Фарадея