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MODELLING THE YIELDS OF RARE EARTH ELEMENT ACTIVATION PRODUCTS FORMED VIA THE (γ,n)-REACTION CHANNEL ON THE M-30 MICROTRON

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To implement the process of non-destructive photon-activation analysis of the content of rare earth elements in the samples under study using electron accelerators and to predict the results, reliable information on the yields of the products (radioisotopes) formed during their activation by generated bremsstrahlung photons is necessary. The results of modelling the yields of radioisotopes ^{44}Sc, ^{88}Y, ^{138}La, ^{141}Ce, ^{140}Pr, ^{147,149}Nd, ^{153}Sm, ^{152}Eu, ^{159}Gd, ^{158}Tb, ^{157}Dy, ^{164}Ho, ^{169}Er, ^{168}Tm, ^{169,175}Yb, ^{174}Lu, formed via the (γ,n)-reaction channel, are presented. Calculations were performed for the case of irradiating rare earth element samples at the electron accelerator of the Institute of Electron Physics of the National Academy of Sciences of Ukraine – the M-30 microtron, operating with bremsstrahlung photons at 12 MeV, taking into account its technical characteristics.

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INTRODUCTION

One of the current challenges is the need to assess the potential natural resources of rare earth elements [1, 2] in Ukraine [3]. To solve this problem, it is necessary to develop methods for determining the content of rare earth elements in the samples under study, which can be implemented using the technical equipment (physical installations) available in Ukraine.

One method for determining the content of rare earth elements in the samples under study is photonic activation analysis, which uses electron accelerators (linear and microtron accelerators) [4–7]. The experience of their use for the implementation of the activation analysis process indicates their high competitiveness with other types of physical installations (for example: reactors, neutron generators, charged particle accelerators [8, 9]), since it allows providing, in addition to photon activation, also neutron activation analysis and their combinations, which is especially important for the determination of rare earth elements in the samples under study due to their nuclear-physical characteristics [10–13].

The effectiveness of using photon activation analysis (analysis accuracy and lower detection limit of the studied elements) to determine the content of rare earth elements in the studied samples is characterized by the numerical values of the yields of their activation products, formed through photonuclear reaction channels (for example: (γ,n)-, (γ,p)-, (γ,α)-, (γ,γ`)-, etc.), when irradiating experimental samples with photons generated in electron accelerators [4,14]. In this context, the key information is about the yields of radionuclides formed through the (γ,n)-reaction channel (which is basic, since its cross sections have larger numerical values compared to the cross sections of other possible photonuclear reactions (for example, (γ,p)-, (γ,α)-, (γ,γ`)-, etc.) at the same photon energies [15, 16]), to ensure the process of photon activation analysis [4, 10].

The yields of activation products of the studied elements depend on the characteristics of the bremsstrahlung photon beams (energy spectra, their integral values and transverse distributions over the potential plane of placement of the analysed samples and intensity (numerical values of the number of particles interacting with the sample)), which are used to stimulate the (γ,n)-reaction, the numerical values of their cross sections [17, 18] and the irradiation time which depends on the nuclear-physical properties of the formed radioisotopes [19], convenient for analysis.

Information on the intensity of photon beams, their energy spectra, and integral values (summed from the thresholds of the (γ,n) reaction to the maximum photon energy) is necessary to predict the yields of formed radionuclides when irradiating experimental samples at electron accelerators and to develop possible activation schemes [20–22].

Information on the transverse distributions of bremsstrahlung photons in the plane of sample placement [23] during irradiation is necessary to optimise the geometric dimensions to increase the yields of the formed radionuclides [24–26].

It should be noted that the characteristics of the beams of bremsstrahlung photons that interact with the studied samples of rare earth elements during their activation (initiating (γ,n)-reactions), in addition to the irradiation schemes used, will be influenced by the characteristics of the initial electron beams at the output of electron accelerators [27], which significantly depend on their technical parameters and may differ among themselves for accelerators of the same type [28, 29]. Therefore, these factors must be taken into account when developing schemes for stimulating (γ,n)-reactions of rare earth elements on electron accelerators to ensure the process of photon activation analysis.

Since experimental studies of the characteristics of bremsstrahlung photon beams generated at electron accelerators, taking into account their technical parameters, are associated with significant difficulties (expensive and long-term measurements), Monte Carlo modeling is used to determine them, which provides the possibility of obtaining the necessary information (for example: photon energy spectra, their integral values, and transverse distributions in the plane of potential placement of the samples) in the maximum approximation to the real conditions of bremsstrahlung photon formation [29–31].

Therefore, to ensure the process of determining the content of rare earth elements in the samples (or photon activation analysis) on electron accelerators, reliable information on the yields of their activation products via the (γ,n)-reaction channel is necessary, which will take into account the technical parameters of the electron accelerators.

The paper presents the results of modeling the yields of rare-earth radionuclide products (^{44}Sc, ^{88}Y, ^{138}La, ^{141}Ce, ^{140}Pr, ^{147,149}Nd, ^{153}Sm, ^{152}Eu, ^{159}Gd, ^{158}Tb, ^{157}Dy, ^{164}Ho, ^{169}Er, ^{168}Tm, ^{169,175}Yb, ^{174}Lu), formed via the (γ,n)-reaction channel, at the electron accelerator of the Institute of Electronic Physics of the NAS of Ukraine – the M-30 microtron at the maximum bremsstrahlung photon energy 12 MeV.

1. MATERIALS AND METHODS

To select possible (convenient) radioisotopes formed during irradiation of rare earth element samples via the (γ,n)-reaction channel, suitable for ensuring the process of determining their elemental content in the studied samples, systematised data on nuclear physical constants for photon activation analysis were used [32-34].

The yields of products (radioisotopes) formed via the (γ,n)-reaction channel during the activation of rare earth element nuclei were calculated according to (1) [18, 35]:

$$Y(t) = Nt\int_{E_{th}}^{E_{max}} \phi(E, E_{max})\sigma(E)dE, \qquad (1)$$

where $Y(t)$ – numerical values of the yields of (γ,n)-reactions depending on the irradiation time t; N – number of nuclei of rare earth elements in the studied samples; t – activation time of the studied samples; $\phi(E, E_{max})$ – density of photon fluxes with maximum energy E_{max}; $\sigma(E)$ – cross-section values of (γ,n)- reactions with energy E; E_{th}– threshold energy of reactions; E_{max}= 12.0 MeV.

Modeling of the outputs of activation products of rare earth element samples on the electron accelerator – microtron M-30 was carried out for the following irradiation scheme: bremsstrahlung photons were generated by a tantalum (Ta) converter (rectangular in shape with dimensions – 100x50 mm, thickness – 1 mm), placed perpendicular to the electron beam axis at a distance of 22 mm from the exit window (material – titanium, shape: ellipse 22x6 mm, thickness – 50 µm) of the accelerator [18]; experimental samples (square shape with dimensions – 10x10 mm and 100x100 mm and thickness – 1 mm) were placed perpendicular to the beam axis at a distance of 60 mm from the Ta-converter.

To simulate the characteristics (energy spectra, their integral values and transverse distributions in the plane) of the bremsstrahlung photon beam generated by the Ta converter on the M-30 microtron, which hit the plane of placement of the studied samples (1000x1000 mm) and onto the samples themselves (10x10 and 100x100 mm), a program was developed in C++ [18, 30, 31] using the GEANT4 v11.1 class library [36]. Calculations were performed for the initial number of electrons – 10E9. During the simulations, all particles generated by the Ta converter (bremsstrahlung photons, residual electrons, secondary photoneutrons) that hit the plane of potential placement of the samples and the samples were recorded.

Fig. 1 shows a visualisation of the simulation of the irradiation scheme using the GEANT4 toolkit (Designation: Ta converter – in green, sample (10x10 mm) – in red, electron and photon beams – in red and green, respectively).

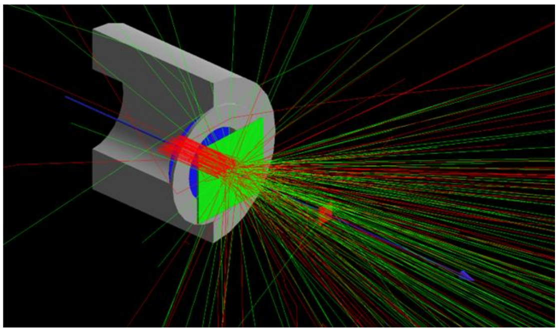

Fig. 1. Visualization of the modeling scheme using the GEANT4 toolkit

From the obtained energy spectra of photons ($\phi(E)$), their integral values (N) were calculated according to formula (2):

$$N = \int_{E_0}^{E_{max}} \phi(E)dE. \qquad (2)$$

When performing calculations, the energy spectra of photons and their integral values were normalised to one initial electron.

To optimise the geometric dimensions of the irradiated samples, probability maps of the transverse distribution of photons in the plane of placement were generated using the inverse Gaussian cumulative distribution [31].

In the process of modeling the yields of radioisotope products formed during the activation of rare-earth elements, according to (1), numerical values of the cross sections of (γ,n)-reactions and their thresholds from the TENDL nuclear data library [37] were used, which is based both on the results of Monte Carlo calculations by the TALYS code [38] by default, and on corrected calculations, as well as on data from other sources [39]. It should be noted that the TALYS code has been successfully used to study the cross sections of (γ,n)-reactions in the interaction of photons with rare earth nuclei [40, 41].

For routine calculations of yields of products formed via the (γ,n)-reaction channel during activation of rare earth element nuclei, the self-created computer program "NPMA Reaction Yield Version 1.1.2304" [42].

2. RESULTS AND DISCUSSION

As a result of the simulations, the energy spectra of bremsstrahlung photons hitting the plane of potential placement of rare-earth element samples (1000x1000 mm) and the samples themselves (with dimensions of 10x10 and 100x100 mm) when they are irradiated by the M-30 microtron were calculated by the program created on the basis of the GEANT4 toolkit [36]. The technical parameters of the electron ejection unit were taken into account when performing the calculations [18, 31].

Fig. 2 presents the energy spectra of photons and the cross sections of (γ,n)-reactions of the interaction of photons with the nuclei of rare earth elements [37].

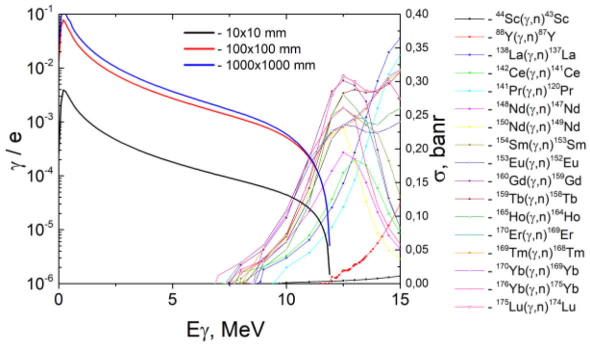

Fig. 2. Energy spectra of bremsstrahlung photons and cross sections of (γ,n)-reactions [37]

Numerical integral values photon energy spectra summed from 0 to 12 MeV and from 8 to 12 MeV are – 0.04816 γ/e and 0.00176 γ/e for a sample with a size of 10x10 mm, respectively. For a sample with a size of 100x100 mm, they are: for the entire energy spectrum – 0.83448 γ/e, for spectra from 8 to 12 MeV – 0.02162 γ/e.

The percentage of photons in the bremsstrahlung beam is 81.71 and 64.21% for the cases of 10x10 mm and 10 0x10 0 mm, respectively.

From the modelling results, the profile of the bremsstrahlung photon beam in a section on a plane measuring 1000x1000 mm at the location of the experimental samples (Fig. 3) and their transverse distribution (Fig. 4) were obtained.

As a result of the simulations, it was found that 4.93E5 photons hit the area of the central cross-section with a size of 10 mm, and 1.74E6 in the area of 100 mm. With a plane of 100x100 mm (namely, an increase in the area by 10 times), the photon concentration increases only by 3.54 times, which indicates that an increase in the area of the sample does not lead to a significant increase in the number of interacting photons, but an increase in the number of accompanying particles, namely residual electrons and secondary photoneutrons, is possible [18, 30, 31].

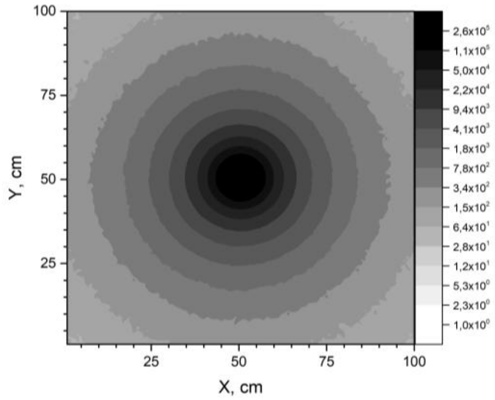

Fig. 3. Profile of bremsstrahlung photons on the plane at the potential location of rare earth element samples

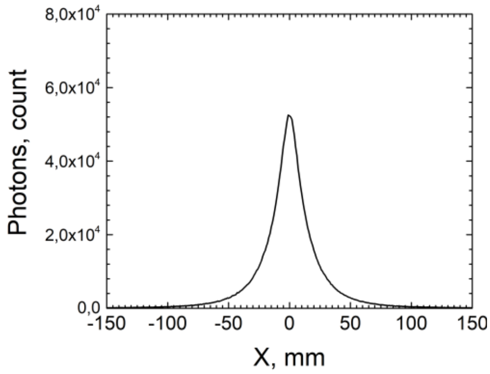

Fig. 4. Transverse distribution of bremsstrahlung photons in the plane of sample placement

Yields of activation products of rare earth elements (normalised to 1 initial electron, 1 second of irradiation and one atom of the element) formed via the (γ,n)-reaction channel at the maximum energy of bremsstrahlung photons – 12 MeV

Element	**Isotope**	**Abundance, % [43]**	**$E_{th\ (\gamma,n)}$, MeV**	**Product**	**T(1/2)***	**Yield 10x10 mm**	**Yield 100x100 mm**	**Increase, times**
$_{21}$Sc	Sc-45	100	11.33	Sc-44	4.042 h	6.795E-36	3.592E-35	5.29
$_{39}$Y	Y-89	100	11.48	Y-88	106.627 d	7.176E-37	3.265E-36	4.55
$_{57}$La	La-139	99.91	8.78	La-138	1.03×10^{11}y	4.436E-33	4.618E-32	10.41
$_{58}$Ce	Ce-142	11.11	7.17	Ce-141	32.504 d	6.388E-33	7.156E-32	11.20
$_{59}$Pr	Pr-141	100	9.40	Pr-140	3.39 m	2.264E-33	2.148E-32	9.49
$_{60}$Nd	Nd-148	5.7	7.33	Nd-147	11.03 d	9.577E-33	1.081E-31	11.29
	Nd-150	5.6	7.38	Nd-149	1.726 h	1.025E-32	1.122E-31	10.95
$_{62}$Sm	Sm-154	22.75	7.97	Sm-153	46.284 h	1.066E-32	1.138E-31	10.68
$_{63}$Eu	Eu-153	52.19	8.55	Eu-152	9.3116 h	8.030E-33	8.293E-32	10.33
$_{64}$Gd	Gd-160	21.86	7.45	Gd-159	18.479 h	1.483E-32	1.641E-31	11.07
$_{65}$Tb	Tb-159	100	8.13	Tb-158	10.70 s	9.603E-33	1.008E-31	10.50
$_{66}$Dy	Dy-158	0.06	9.05	Dy-157	8.14 h	7.336E-33	7.245E-32	9.88
$_{67}$Ho	Ho-165	100	7.99	Ho-164	28.8 m	7.636E-33	7.858E-32	10.29
$_{68}$Er	Er-170	14.91	7.26	Er-169	9.392 d	1.588E-32	1.796E-31	11.31
$_{69}$Tm	Tm-169	100	8.03	Tm-168	93.1 d	1.299E-32	1.410E-31	10.85
$_{70}$Yb	Yb-170	3.04	8.46	Yb-169	32.018 d	1.209E-32	1.285E-31	10.63
	Yb-176	12.76	6.87	Yb-175	4.185 d	1.713E-32	1.966E-31	11.48
$_{71}$Lu	Lu-175	97.41	7.69	Lu-174	142 d	1.106E-32	1.187E-31	10.73

* s – second, m – minute, h – hour, d – day, y – year

The obtained results reproduce the general patterns in the influence of the geometric dimensions of the irradiated samples on the integral values of the interacting photons and are consistent with the results of similar studies [18, 24-26]. Differences in numerical values may be associated with the initial parameters of the calculations (initial electron beam, its energy, sample irradiation schemes [27, 28, 31]).

The results obtained allow us to estimate the minimum sample size required to achieve satisfactory representativeness for the specified research task – optimisation of the photon activation analysis process on the M-30 microtron.

The results of calculations of the characteristics of bremsstrahlung photon beams generated on the M-30 microtron, interacting with the studied samples of rare earth elements (sizes 10x10 and 100x100 mm) during their irradiation, were used to model the yields of radioisotopes formed via the (γ,n)-reaction channel, according to formula (1).

When performing calculations, the yields of activation products of rare earth elements formed via the (γ,n) reaction channel were normalised to 1 initial electron, 1 s of irradiation, and one atom of the element.

Results of simulations of the yields of activation products of rare earth elements – radioisotopes ^{44}Sc, ^{88}Y, ^{138}La, ^{141}Ce, ^{140}Pr, ^{147,149}Nd, ^{153}Sm, ^{152}Eu, ^{159}Gd, ^{158}Tb, ^{157}Dy, ^{164}Ho, ^{169}Er, ^{168}Tm, ^{169,175}Yb, ^{174}Lu by bremsstrahlung photons with an energy of 12 MeV, formed via the (γ,n)-reaction channel, are presented in Table.

The obtained values of the yields of rare earth element activation products for different geometric sizes (10x10 and 100x100 mm) of the studied samples can be used to optimize the stimulation scheme of the (γ,n)-reaction on the electron accelerator – the M-30 microtron to ensure the process of determining their isotopic content.

CONCLUSIONS

The simulated values of the yields of activation products of rare earth elements – radioisotopes ^{44}Sc, ^{88}Y, ^{138}La, ^{141}Ce, ^{140}Pr, ^{147,149}Nd, ^{153}Sm, ^{152}Eu, ^{159}Gd, ^{158}Tb, ^{157}Dy, ^{164}Ho, ^{169}Er, ^{168}Tm, ^{169,175}Yb, ^{174}Lu by bremsstrahlung photons with an energy of – 12 MeV, formed via the (γ,n)-reaction channel, supplement the existing literature data necessary to ensure the process of photonuclear activation analysis on electron accelerators.

The proposed modelling scheme (using nuclear Monte Carlo codes: GEANT 4, TALYS, and the self-created computer program “NPMA Reaction Yield Version 1.1.2304”) of activation product yields can be used to optimise photonuclear reaction stimulation schemes at the electron accelerator – the M-30 microtron for a wide range of nuclei and predict the yields of the formed radioisotopes.

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МОДЕЛЮВАННЯ ВИХОДІВ ПРОДУКТІВ АКТИВАЦІЇ РІДКІСНО-ЗЕМЕЛЬНИХ ЕЛЕМЕНТІВ УТВОРЕНИХ ПО КАНАЛУ (γ,n)-РЕАКЦІЇ НА МІКРОТРОНІ М-30

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Для реалізації процесу недеструктивного фотонного активаційного аналізу вмісту рідкісно-земельних елементів у досліджуваних зразках на електронних прискорювачах та прогнозування отриманих результатів необхідна достовірна інформація про виходи продуктів (радіоізотопів), утворених при їх активації згенерованими гальмівними фотонами. Представлено результати моделювання виходів радіоізотопів ^{44}Sc, ^{88}Y, ^{138}La, ^{141}Ce, ^{140}Pr, ^{147,149}Nd, ^{153}Sm, ^{152}Eu, ^{159}Gd, ^{158}Tb, ^{157}Dy, ^{164}Ho, ^{169}Er, ^{168}Tm, ^{169,175}Yb, ^{174}Lu, утворених по каналу (γ,n)-реакції. Розрахунки проводилися для випадку опромінення зразків рідкісно-земельних елементів на електронному прискорювачі ІЕФ НАН України – мікротроні М-30, гальмівними фотонами з енергією 12 МеВ із врахуванням його технічних характеристик.