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PRODUCT YIELDS FOR THE PHOTOFISSION OF ^{239}Pu WITH BREMSTRAHLUNG AT 17.5 MeV BOUNDARY ENERGY

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The values of relative cumulative yields of 12 products (^{85m}Kr, ^{91m}Y, ^{92}Sr, ^{97}Zr, ^{99}Mo, ^{105}Ru, ^{133}I, ^{134}I, ^{135}I, ^{138}Cs, ^{139}Ba, ^{142}La, ^{143}Ce) of the ^{239}Pu photofission was measured at a maximum bremsstrahlung energy of 17.5 MeV (average excitation energy ~ 12.03 MeV). ^{239}Pu photofission reaction was stimulated on the electron accelerator of the Institute of Electron Physics NAS of Ukraine – M-30 microtron to simulate the spectra of bremsstrahlung's photons, secondary electrons, and photoneutrons that hit the ^{239}Pu target, the GEANT4 code was used. The input of accompanying nuclear reactions to the yield of ^{239}Pu photofission products for the given experimental parameters was also evaluating. The obtained experimental data of the yields of products ^{239}Pu photofission were compared with the program codes GEF and Talys1.9.5 simulations.

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INTRODUCTION

Information on the yields of ^{239}Pu nuclear fission products plays an important role in the development of methods for transmutation of spent nuclear fuel (which in large quantities contains products of (n,f)- and (γ,f)-reactions) and non-destructive methods of isotope analysis of fissile nuclear materials. The above mentioned directions require reliable experimental information on the yields of ^{239}Pu photofission products, which is currently limited both in the range of studied fragments and in the field of stimulation energies (γ,f)-reactions [1 - 5].

The aim of this work is to experimentally investigate the yields of the fission products of the ^{239}Pu nucleus on an electronic accelerator – M-30 microtron at a maximum bremsstrahlung energy of 17.5 MeV and compare the obtained data with the results of modeling by modern calculation codes (phenomenological fission models) "GEF 2021/1.1" [6] and "Talys 1.95" [7].

To implement this task, the following were performed: modeling of the optimal scheme of the experiment to stimulate the ^{239}Pu photofission reaction taking into account the technical characteristics of the M-30 microtron and the parameters of the fission target using the GEANT4 toolkit [8]; analysis of the contribution of the outputs of related reactions to the outputs of the photofission products of ^{239}Pu; experimental studies of the relative yields of ^{239}Pu photofission products by their gamma radiation; and simulation of product outputs for the fissible nucleus of ^{239}Pu* (E* – 12.03 MeV) using "GEF 2021/1.1" [6] and "Talys 1.95" codes [7].

1. MATERIALS AND METHODS

Determination of relative cumulative yields of ^{239}Pu photofission products was performed by semiconductor gamma-ray spectrometry [9 - 11]. The studied value during measurements is the counting rate in peaks of total absorption (or peak intensity) of gamma quanta from individual fission products, which depends on its activity, absolute measurement efficiency, self-absorption corrections, and gamma line intensity.

1.1. ^{239}Pu FISSION TARGET

A target with plutonium isotopes: ^{239}Pu – 99% and ^{238}Pu – 1% was used for experimental studies. The active layer of plutonium (disc diameter – 11 mm) was applied to a stainless steel substrate disc with a diameter of 24 mm and a thickness of 1 mm. The target was manufactured at the V.G. Khlopin Radium Institute (Saint Petersburg, Russia). The number of ^{239}Pu and ^{238}Pu nuclei was – 1.398xE16 and 1.121x10E7, respectively, at the time of the experiment. The target was not exposed to radiation before the experimental studies.

1.2. SIMULATION OF THE FISSION TARGET ACTIVATION PROCESS

The GEANT410.7 computing code was used to simulate the spectra of bremsstrahlung photons, residual electrons, and photoneutrons (depending on the energy normalised per electron) which hit the target. The input parameters used in the calculations almost completely reproduced the geometric dimensions (design features) of the electron output unit and the activation schemes of fissile nuclei, which was implemented on an electron accelerator – the M-30 microtron [12] (Fig. 1).

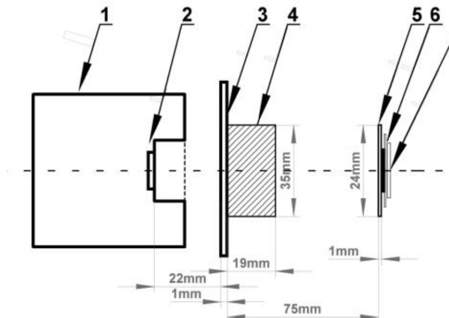

Fig. 1. Scheme of activation of ^{239}Pu target on the microtron M-30:
1 – output node of the microtron M-30; 2 – window of the output node (ellipse, thickness – 50 μm); 3 – photons converter (Ta, 100×50×1 mm); 4 – filter B_4C (D = 30 mm, thickness = 19 mm); 5 – substrate (stainless steel disc, diameter – 24 mm, thickness – 1 mm); 6 – ^{239}Pu layer; 7 – Al collector of fission products

The simulation considered the geometric dimensions of the original electron beam: the shape – an ellipse, the dimensions of the semi-axes – 11 and 3 mm) and characteristic of ^{239}Pu fissioning target. Calculations were performed for 10E9 electrons in the initial beam on two computers with 6-core Intel(R) Core(TM) processors i7-9750HCPU@2.60GHz and 36 GB and 16 GB RAM.

The influence of the elements of the activation circuit (converter, filter [13], stainless steel substrate) on the final shape of the spectra of photons, residual electrons and photoneutrons, which hit on the fissile ^{239}Pu layer, was established (Fig. 2).

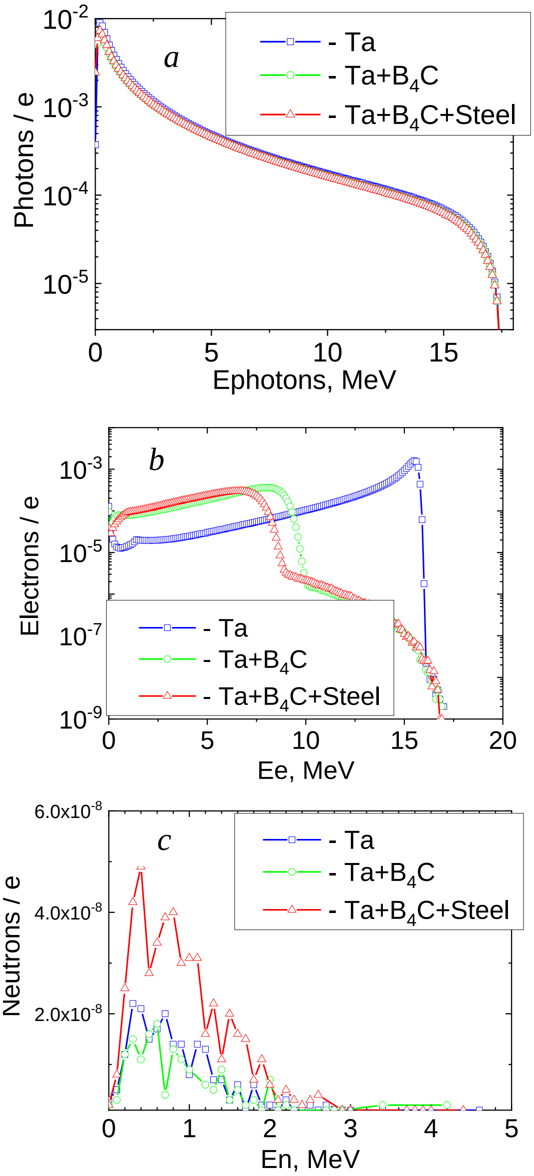

Fig. 2. Spectra of photons (a), residual electrons (b) and photoneutrons (c) on the fissile layer of ^{239}Pu: square – Ta-converter – fissile layer; circle – Ta-converter + B_4C filter – fissile layer; triangle – Ta-converter + B_4C filter + stainless steel substrate – fissile layer

As a result of the simulation, the total number of photons, residual electrons, and photoneutrons normalised per electron hitting a fission target was calculated: 0.12913 photons (with an energy > 6 MeV, involved in stimulating the photofission reaction – 0.01749); – 0.01485 electrons (with an energy > 6 MeV – 0.00571) and ~ 6.1E-7 neutrons.

Residual electrons that fell on the fission assembly did not affect the results of the experiment.

The obtained results were used to estimate the contribution of the outputs of the concomitant (accompanying) nuclear reactions (^{239}Pu(γ,n)^{238}Pu → ^{238}Pu(γ,f); ^{238}Pu(n,f) and ^{239}Pu(n,f) to the outputs of the photofission products. The values of the cross sections of photonuclear ((γ,n)-, (γ,f)-) and neutron ((n,f)- reactions from the ENDF database) [14] (Fig. 3), were used in the calculations (Fig. 4). The calculations of the number of the nuclei ^{238}Pu, which were formed along the reaction channel ^{239}Pu(γ,n)^{238}Pu during the irradiation time (0…4 hours) and the dependences of the number of the photofission and neutron fission acts of the nuclei ^{238}Pu and ^{239}Pu, on the irradiation time (0…4 hours) are represented in Fig. 4.

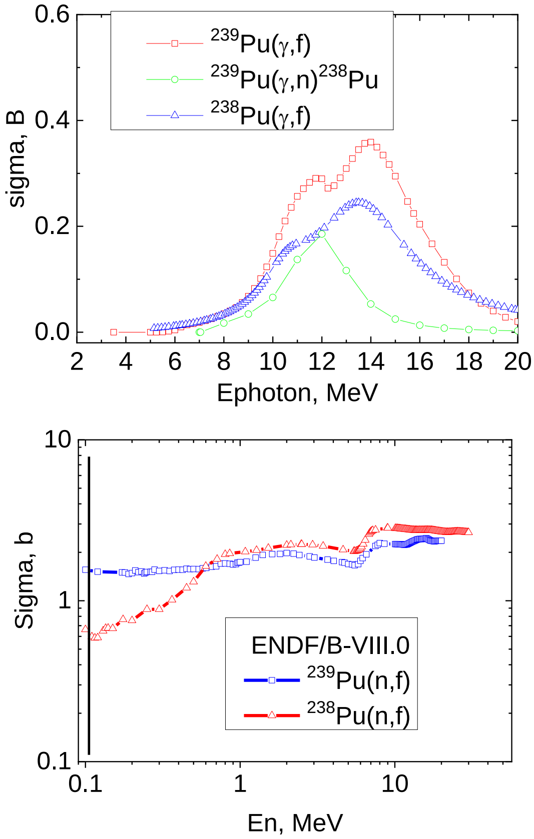

Fig. 3. Cross sections of photonuclear (up) and neutron reactions (down) [14]

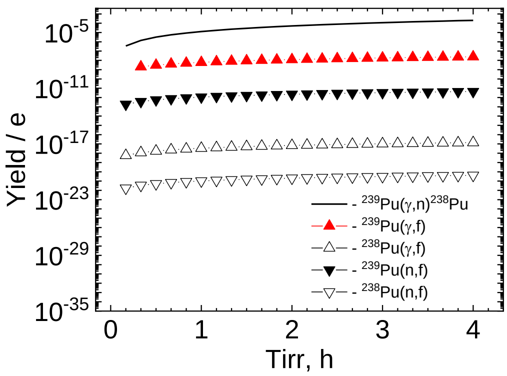

Fig. 4. The dependence of the number of nuclei formed from possible reaction channels, and the reaction rate from the activation time (0…4 h) upon irradiation of ^{239}Pu by bremsstrahlung with an energy of 17.5 MeV

The analysis indicates the lack of influence of the yields of the products of concomitant reactions on the accuracy of the final results.

The results of the calculations allowed to optimize the scheme of stimulation of the $^{239}Pu(\gamma,f)$ reaction on the M-30 microtron.

1.3. STIMULATION OF THE ^{239}Pu PHOTOFISSION REACTION

To accumulate photofission products during activation, 0.2 mm thick aluminum foil collectors were used, which were installed close to the ^{239}Pu layer.

Irradiation of a fissible assembly (which consisted of plutonium fissioning target and collector layer) were performed on an electron accelerator of the Institute of Electron Physics of the National Academy of Sciences of Ukraine – an M-30 microtron (electron energy E=17.5 MeV, average beam current ~ 4 μA) (see Fig. 1). The instability of the electron energy during target irradiation did not exceed 0.04 MeV [12].

To generate bremsstrahlung, a tantalum converter was used (thickness – 1 mm), located at a distance of 22 mm from the output window (Ta, thickness – 50 microns) of the electron output unit. The fission assembly was installed perpendicular to the beam axis at a distance of 75 mm from the Ta converter. The filter of residual electrons and photoneutrons (B_4C) was installed close to the Ta-converter perpendicular to the axis of the beam [13]. The irradiation time of the fission assembly were – 3.5 and 3.75 hours. The choice of time parameters (irradiation, cooling, and measurement times) was made considering the half-lives of the studied photofission products and their precursors (the so-called "parent" products) along the isobaric chain.

1.4. GAMMA-RAY SPECTROMETRIC STUDIES OF ^{239}Pu PHOTOFISSION PRODUCTS

At the end of the accumulation of fragments, aluminum collectors measured their gamma activity for from 0.25 to 70 hours after the end of irradiation. The duration of individual measurements varied from 0.5 to 3 hours. For the research, spectrometric complexes based on semiconductor detectors were used: HPGe (Ortec) and Ge (Li), the volumes of which were 150 and 100 cm^3 with an energy resolution of ~ 2.45 and ~ 3.5 keV for the line ^{60}Co (1.332.5 keV) [15, 16]. When studying the relative cumulative yields of fission products, the final error of the obtained results is primarily affected by the error value of the measured energy efficiency of the detector.

The energy dependence of the peak efficiency of gamma-ray quantum registration was determined using a set of standard certified point sources ^{22}Na, ^{57}Co, ^{60}Co, ^{109}Cd, ^{133}Ba, ^{137}Cs, ^{151}Eu, ^{241}Am (produced by D.I. Mendeleev Institute for Metrology, Saint Petersburg, Russia). The value of the statistical measurement error during the calibration procedure did not exceed 4%.

Gamma-ray spectra from photofission products were measured in real time. The dead time of the spectrometer did not exceed 5…8% during all measurements. During the measurements, the drift of the energy scale, resolution and recording efficiency of the spectrometric complex were constantly monitored using point standard gamma-active source ^{60}Co. The drift of these parameters did not exceed 1%. Spectroscopic information was processed using the Winspectrum software package [17]. Fission fragments were identified by the energies of their characteristic gamma lines, considering their half-lives and measurement, accumulation, and cooling times. Additionally, the half-lives of their predecessors along isobaric chains were considered. The values of nuclear spectroscopic data of the identified fission products (energies and intensities of gamma lines, half-lives of the formed products and their precursors along the isobaric chain) were taken for calculations from the "Decay Radiation database" [18].

RESULTS AND DISCUSSION

During the experiment, we measured the peak intensity of gamma lines belonging to the following products of the ^{239}Pu nuclear photofission: ^{85m}Kr (151.2), ^{91m}Y (555.6), ^{92}Sr (1383.9), ^{97}Zr (743.4), ^{99}Mo (739.5), ^{105}Ru (724.3), ^{133}I (529.9), ^{134}I (847.0; 884.1), ^{135}I (1260.4), ^{138}Cs (1435.8), ^{139}Ba (165.9), ^{142}La (641.3), ^{143}Ce (293.3). The energies of gamma lines are presented in parentheses (in keV). The statistical error of measurements of the peak intensity of gamma product lines used in the analysis did not exceed 5% for the entire time interval of measurements. Cumulative yields were determined relative to the yields of the ^{97}Zr reference product (4.63% [1]). The total error of relative cumulative yields was estimated considering statistical errors of peak intensity of gamma product lines, analysis of time dependencies, spread of values averaged over individual measurements, as well as errors of interpolated efficiency values and nuclear physical constants (energy and intensity of gamma lines, half-lives of products). The total error in determining the relative cumulative yields of fission fragments did not exceed 10%.

Due to the small size of the plutonium sample, a limited number of fission products were measured.

Experimental values of yields of ^{239}Pu photofission products at a maximum bremsstrahlung photon energy of 17.5 MeV are shown in Fig. 5 by black circle. Squares and up triangles represent yields of fission products at a maximum bremsstrahlung photon energy of 28 and 30 MeV [1, 2]. Rombes and down triangles – at 11 and 13 MeV with monoenergetic photons [4, 5]. At close excitation energies, our data agree with the existing experimental data.

Fig. 5 also show the yields of fission products for a ^{239}Pu* fission nucleus at an excitation energy of ~ 12.03 MeV, calculated by the GEF code [6] (solid curve) and Talys 1.95 code [7] (dashed line). The obtained theoretical output values calculated using the GEF 2020/1.1 code adequately describe the mass distribution of ^{239}Pu photofission products. Talys 1.95 codes describe in general terms the mass distribution of ^{239}Pu photofission products only.

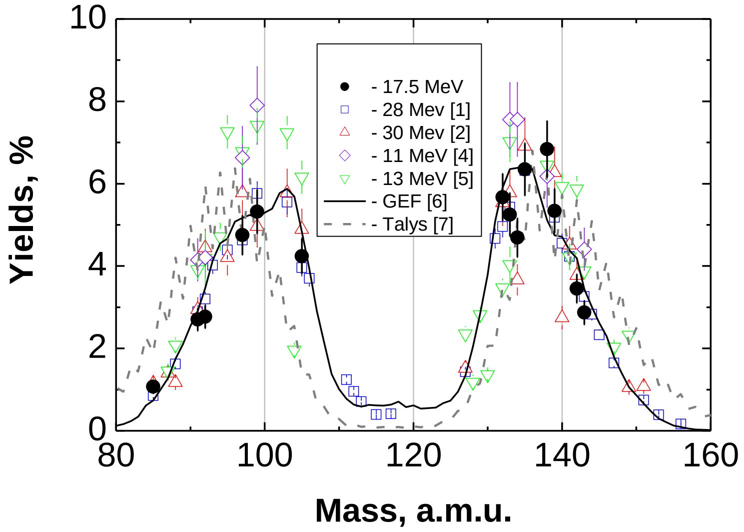

Fig. 5. Yields of ^{239}Pu photofission products

CONCLUSIONS

Results of simulation the characteristics of bremsstrahlung radiation beam and its components (relation of photons, residual electrons, and photoneutrons which hit the target for M-30 microtron (taking into account the constructive features of its electrons output node and the characteristics of the fissile target), with help the Geant 4 toolkit, allowed to develop an optimal scheme for stimulating the reaction of the ^{239}Pu photofission, which ensures the lack of exposure to the yields of related reactions to the precision of the final results.

The cumulative yields of 14 ^{239}Pu photofission products were determined by semiconductor gamma spectroscopy at the maximum bremsstrahlung photon energy of 17.5 MeV (E* ~ 12.03 MeV). The simulations performed using the GEF and Talys1.95 codes describe and predict of mass distributions for a fissile ^{239}Pu* nucleus at an average excitation energy of ~ 12.03 MeV.

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ВЫХОДЫ ПРОДУКТОВ ФОТОДЕЛЕНИЯ ^{239}Pu ПРИ МАКСИМАЛЬНОЙ ЭНЕРГИИ ТОРМОЗНОГО ИЗЛУЧЕНИЯ 17,5 МэВ

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Значения 12-ти относительных кумулятивных выходов продуктов (^{85m}Kr, ^{91m}Y, ^{92}Sr, ^{97}Zr, ^{99}Mo, ^{105}Ru, ^{133}I, ^{134}I, ^{135}I, ^{138}Cs, ^{139}Ba, ^{142}La, ^{143}Ce) фотоделения ^{239}Pu были измерены при максимальной энергии тормозного излучения 17,5 МэВ (средняя энергия возбуждения ~ 12,03 МэВ). Стимуляция реакции фотоделения ^{239}Pu проводилась на ускорителе электронов Института электронной физики НАН Украины – микротроне М-30. Для моделирования спектров тормозных фотонов, вторичных электронов и фотонейтронов, попадающих на мишень ^{239}Pu, использовался код GEANT4. Проведена оценка вклада сопутствующих ядерных реакций в выходы продуктов фотоделения ^{239}Pu при заданных параметрах эксперимента. Полученные экспериментальные данные по выходам продуктов фотоделения ^{239}Pu сравнивались с результатами симуляции кодами GEF и Talys1.9.5.

ВИХОДИ ПРОДУКТІВ ФОТОПОДІЛУ ^{239}Pu ПРИ МАКСИМАЛЬНІЙ ЕНЕРГІЇ ГАЛЬМІВНОГО ВИПРОМІНЮВАННЯ 17,5 МеВ

О.О. Парлаг, В.Т. Маслюк, Є.В. Олейніков, І.В. Пилипчинець, О.І. Лендел

Значення 12-ти відносних кумулятивних виходів продуктів (^{85m}Kr, ^{91m}Y, ^{92}Sr, ^{97}Zr, ^{99}Mo, ^{105}Ru, ^{133}I, ^{134}I, ^{135}I, ^{138}Cs, ^{139}Ba, ^{142}La, ^{143}Ce) фотоподілу ^{239}Pu були виміряні при максимальній енергії гальмівного випромінювання 17,5 МеВ (середня енергія збудження ~ 12,03 МеВ). Стимуляція реакції фотоподілу ^{239}Pu проводилася на прискорювачі електронів Інституту електронної фізики НАН України – мікротроні М-30. Для моделювання спектрів гальмівних фотонів, вторинних електронів і фотонейтронів, що попадали на мішень ^{239}Pu, використовувався код GEANT4. Було проведено оцінку внеску супутніх ядерних реакцій у виходи продуктів фотоподілу ^{239}Pu при заданих параметрах експерименту. Отримані експериментальні дані по виходах продуктів фотоподілу ^{239}Pu порівнювалися з результатами симуляції кодами GEF і Talys1.9.5.