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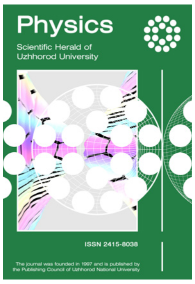

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Simulation the Yields of Actinide Nuclei Photofission Products as Sources of Delayed Gamma Radiation for the Needs of Analyzing their Isotopic Composition

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Abstract

Purpose. One of the most important tasks of the nuclear industry is to control the non-proliferation of fissile nuclear materials (for example ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu) at all stages of their use (movement, storage, etc.). To successfully solve this problem, reliable information about their isotopic composition is required. The present study aims to simulate the yields of pairs of photofission products of ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu nuclei, the delayed gamma radiation of which can be used for nondestructive isotopic analysis of nuclear materials at electron accelerators.

Methods. Calculations of the mass distributions of the photofission products of ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu nuclei were carried out using the GEF code. The bremsstrahlung spectrum was simulated during the interaction of electrons (E=12.5 MeV) with a tantalum converter (1 mm) using a GEANT4 10.7.

Results. Simulations of mass distributions of photofission products of ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu nuclei have been carried out. The yields ratios for product pairs (Y_{88}/Y_{135}, Y_{92}/Y_{135}, Y_{92}/Y_{138}, Y_{87}/Y_{138}, Y_{88}/Y_{138}, Y_{89}/Y_{138}, Y_{87}/Y_{142}, Y_{88}/Y_{142}, Y_{89}/Y_{142}) are calculated for the indicated cores. Estimates of the difference between the numerical values of the ratio of yields of pairs of fragments for pairs of nuclei ^{232}Th and ^{235}U, ^{235}U and ^{238}U, ^{238}U and ^{239}Pu are made on a percentage basis. The values of the difference in the ratios of the yields of these pairs of products are – 5.0÷43.2%, 14.1÷39.3%, and 14.1÷39.3% for nuclear pairs ^{232}Th and ^{235}U, ^{235}U and ^{238}U, ^{238}U and ^{239}Pu, respectively.

Conclusions. The results of the simulation indicate the possibility of using the above pairs of fission products as sources of delayed gamma radiation when performing nondestructive isotopic analysis of nuclear materials. The results obtained can be used to optimize experiments on electron accelerators, which will improve the accuracy and reliability of the results

Keywords: nuclear materials, isotope analysis, photofission, product yields, delayed gamma radiation

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Introduction

One of the most important tasks of the nuclear industry is to control the non-proliferation of fissile nuclear materials (for example ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu) at all stages of their use (movement, storage, etc.). To successfully solve this problem, reliable information about their isotopic composition is required. As a rule, transportation and storage of nuclear materials is carried out in hermetically sealed capsules or containers made of stainless steel [2; 3].

One of the widely used non-destructive methods for the detection and isotopic identification of unshielded and shielded fissile nuclear materials is based on the use of delayed gamma radiation from the products of their separation [4-6]. The essence of this method is to use experimentally obtained information on the ratio of the intensity of stimulated delayed gamma radiation from light and heavy products of their separation. The yields of light products-fragments significantly depend on the mass of fissile nuclei, so at the same excitation energies their mass distributions shift towards larger masses with increasing mass of fissile nucleus. At the same time, the mass distribution of heavy fragments is almost constant and does not depend significantly on the mass of the fissile nucleus.

Fission products that are potential sources of delayed gamma radiation suitable for the analysis of shielded and unshielded nuclear materials must satisfy a number of basic requirements: a) a high probability of their formation in the fission process; b) the presence of a wide set of intense gamma radiation lines with energies> 1000 MeV, free from spectrometric interference (characteristic, background gamma lines, from the products of accompanying reactions) c) with convenient half-lives for analysis. It should be noted that fission product pairs with close half-lives should be used in the analysis.

The following products-fragments meet these requirements: ^{87}Kr, ^{88}Kr, ^{88}Rb, ^{89}BRb, ^{92}Sr, ^{135}I, ^{138}Cs, ^{142}La, which were formed as a result of (n, f)-reactions induced by thermal, fast and high-energy (14 MeV) neutrons [4-6] and bremsstrahlung of electronic accelerators [7-9]. Their nuclear-physical characteristics [10] meet the above criteria. The above works [4-9] demonstrated the possibility of using them for the isotopic identification of unshielded [4-7] and shielded [8; 9] fissile nuclear materials.

The aim of the presented work is to analyze the possibility of using these nuclides-fragments (with A = 87, 88, 89, 92, 135, 138, 142) as potential sources of delayed gamma radiation, to identify 4 main fissile nuclear materials (^{232}Th, ^{235}U, ^{238}U, ^{239}Pu) when they are activated by bremsstrahlung obtained on an electronic accelerator – a microtron. Therefore, it was necessary to calculate the dependence of number values of post-neutron output of pairs of fragments (Y_{88}/Y_{135}, Y_{92}/Y_{135}, Y_{92}/Y_{138}, Y_{87}/Y_{138}, Y_{88}/Y_{138}, Y_{89}/Y_{138}, Y_{87}/Y_{142}, Y_{88}/Y_{142}, Y_{89}/Y_{142}) for the above mentioned fissile nuclei produced at the stimulation of the of photofission reaction.

Modelling Post-Neutron Outputs of Actinide Nuclear Photofission Products

In order to solve this problem, it is necessary to model post-neutron yields of photofission products of actinides ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu at a fixed energy of bremsstrahlung, and to calculate the numerical values of the ratios of the pairs of fragments whose delayed gamma radiation analysis is nuclei. Delayed gamma radiation suitable for isotopic analysis of the specified nuclei.

The GEF code [11], which allows the calculation of characteristics for fissile nuclei in any type of fission (spontaneous, neutron and proton induced, photofission) and which can be used to calculate the post-neutron yields of photofission products of actinides [12; 13], was used for modelling.

The simulation is carried out for brake radiation, which is generated by electricity with energy of 12.5 MeV at a tantalum radiator with thickness of 1 mm. The bremsstrahlung spectrum was simulated during the interaction of electrons (E=12.5 MeV) with a tantalum converter (1 mm) using a GEANT4 10.7 [14; 15], with the following input parameters: the number of events – 10^8, and for the distances: electron source – bremsstrahlung target – irradiated object, which were – 18 and 100 mm, respectively. The choice of the specified energy was due to the possibility of isotopic analysis for both unshielded and shielded fissile nuclear materials [16], i.e. the energy was lower than the thresholds of possible photonuclear reactions for potential elements from which stainless steel containers are made.

The results of the calculation of the spectrum of bremsstrahlung with an energy of 12.5 MeV are presented in Figure 1. The same figure shows the cross sections (γ, f) – reactions for ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu actinide nuclei from the library of estimated nuclear data ENDF [17], which were used to calculate the average excitation energy for these fissile nuclei [12].

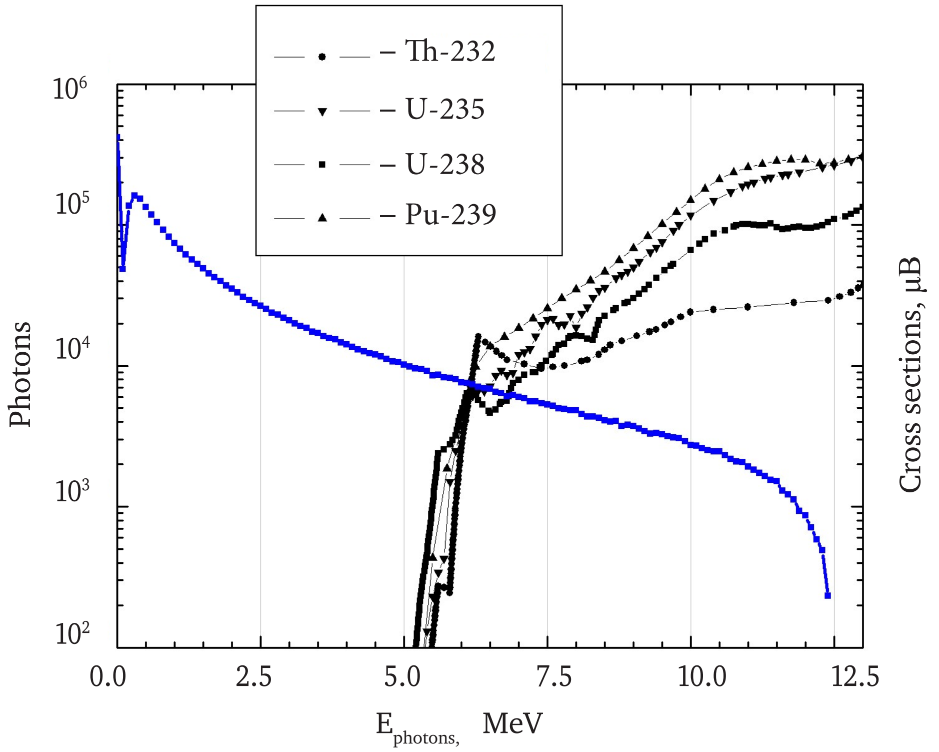

Figure 1. Braking radiation spectrum and photofission cross sections of ^{232}Th, ^{235}U, ^{238}U and ^{239}Pu

The calculated average excitation energy for the photofission of the ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu nuclei at the maximum braking radiation energies of 12.5 MeV were – 8,821; 10,013; 9,744 and 9,922 MeV, respectively. The obtained values were used as input data in the GEF code simulation of post-neutron outputs of products for fissile nuclei ^{232}Th*, ^{235}U*, ^{238}U*, ^{239}Pu*. The latest version of the GEF code – 2020 / 1.1 [18] was used for calculations.

The results of the calculation of post-neutron yields of fission products are presented in the panel of Figure 2. The existing experimental data that were studied at close excitation energies are also presented here. Excitation energy values – for ^{232}Th: 8.8 MeV [19], 8.35 MeV [20]; ^{235}U: 9.7 MeV [21]; ^{238}U: MeV 9.7 [22], 9.09 MeV [23].

The results of modeling the post-neutron yields of ^{239}Pu were compared with the estimated values of the yields of the reaction products ^{238}Pu

(n_{fast}, f) [24] resulting in a similar fissile nuclei $^{239}Pu^*$, as there are no experimental data on the yields of photofission of products at close energies. The average excitation energy $<E^*>$ for the fissile nucleus $^{239}Pu^*$ in neutron fission was calculated from the average neutron energy $<E_n>$ by formula (1):

$$<E^*> = (\Delta^{238}Pu + \Delta n - \Delta^{239}Pu) + <E_n> \quad (1)$$

where $\triangle$ – excess (or defect) of the mass, the values of which were taken from [25]. The value of $<E^*>=6.14$ MeV.

The results of the calculations are consistent with the experimental data within the errors for fissile nuclei ^{235}U, ^{238}U, ^{239}Pu at close excitation energies. For ^{232}Th, there is a discrepancy between the calculated and experimental yields values [19; 20] for both light and heavy products (with maximum yields values). It should be noted that for ^{232}Th there is also a discrepancy between the existing experimental data.

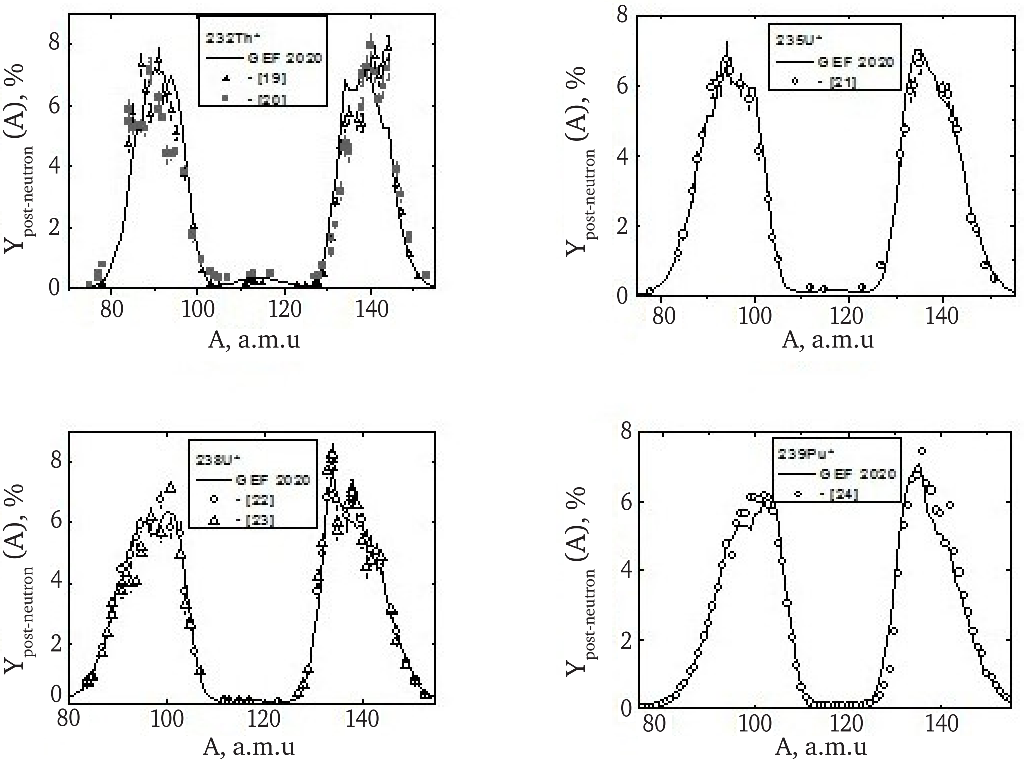

Figure 2. Post-neutron product yields for fissile ^{232}Th, ^{235}U, ^{238}U and ^{239}Pu nuclei

Figure 3 shows the dependences of post-neutron yields of photofission products of ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu on product mass at the same brake radiation energy of 12.5 MeV. As can be seen from the mass distributions shown in the figure, the yields of light products shifts towards larger masses, increasing the mass of the fissile nucleus. At the same time, the mass distribution of heavy fragments remains virtually unchanged and does not depend significantly on the mass of the fissile nucleus. This indicates that the yields of light photofission products of actinide nuclei ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu depend on the mass of fissile nuclei at the same stimulation energies.

The dependence of the ratios (Y_L/Y_H) of the numerical values of the yields of light (Y_L) and heavy (Y_H) products on the mass of fissile nuclei is calculated. The calculations were performed for the following product pairs Y_{88}/Y_{135}, Y_{92}/Y_{135}, Y_{92}/Y_{138}, Y_{87}/Y_{138}, Y_{88}/Y_{138}, Y_{89}/Y_{138}, Y_{87}/Y_{142}, Y_{88}/Y_{142}, Y_{89}/Y_{142}. The fission products selected for analysis are potential sources of delayed gamma radiation suitable for isotopic analysis of shielded and unshielded nuclear materials and meet the above requirements: $Y_{87}=Y_{87Kr}$, $Y_{88}=Y_{88Kr}$, $Y_{89}=Y_{89Rb}$, $Y_{92}=Y_{92Sr}$, $Y_{135}=Y_{135I}$, $Y_{138}=Y_{138Cs}$, $Y_{142}=Y_{142La}$ [10].

The results of the calculation Y_L/Y_H are presented in panel Figure 4. As can be seen from the figure, the output ratio depends on the mass of the fissile nuclei. Here, the Y_L/Y_H yields ratios are presented, for which experimental data were used [19-24]. The obtained data are consistent with each other within the experimental errors.

Table 1 shows the numerical values of the Y_L/Y_H yields ratio for the fissile nuclei ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu and the difference (percentage) in percent (%) between the numerical values of the ratio of pairs of fragments for these nuclei. The percentage difference was calculated between the pairs of nuclei ^{232}Th and ^{235}U (5.0÷43.2%), ^{235}U and ^{238}U (14.1÷39.3%), ^{238}U and ^{239}Pu (14.1÷31.4%).

Thus, the selected product pairs of photofission nuclei ^{232}Th, ^{235}U, ^{238}U, and ^{239}Pu can be used as sources of delayed gamma radiation in non-destructive isotope analysis of these actinides.

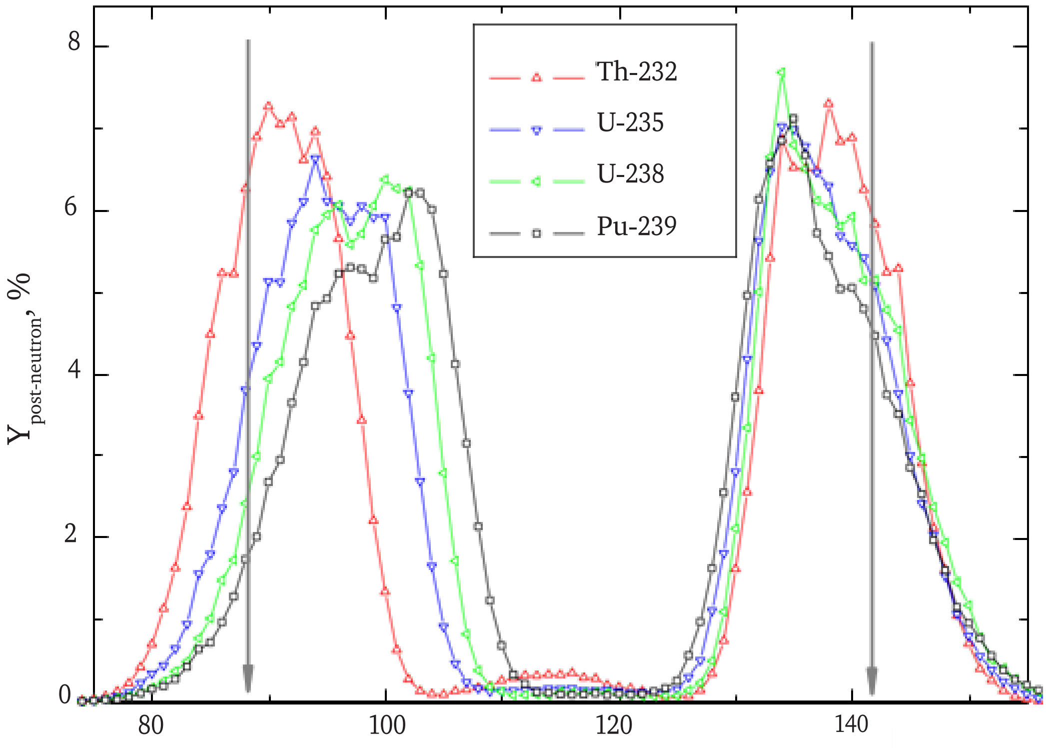

Figure 3. Dependence of post-neutron yields of fission products ^{232}Th*, ^{235}U*, ^{238}U*, and ^{239}Pu* on their mass

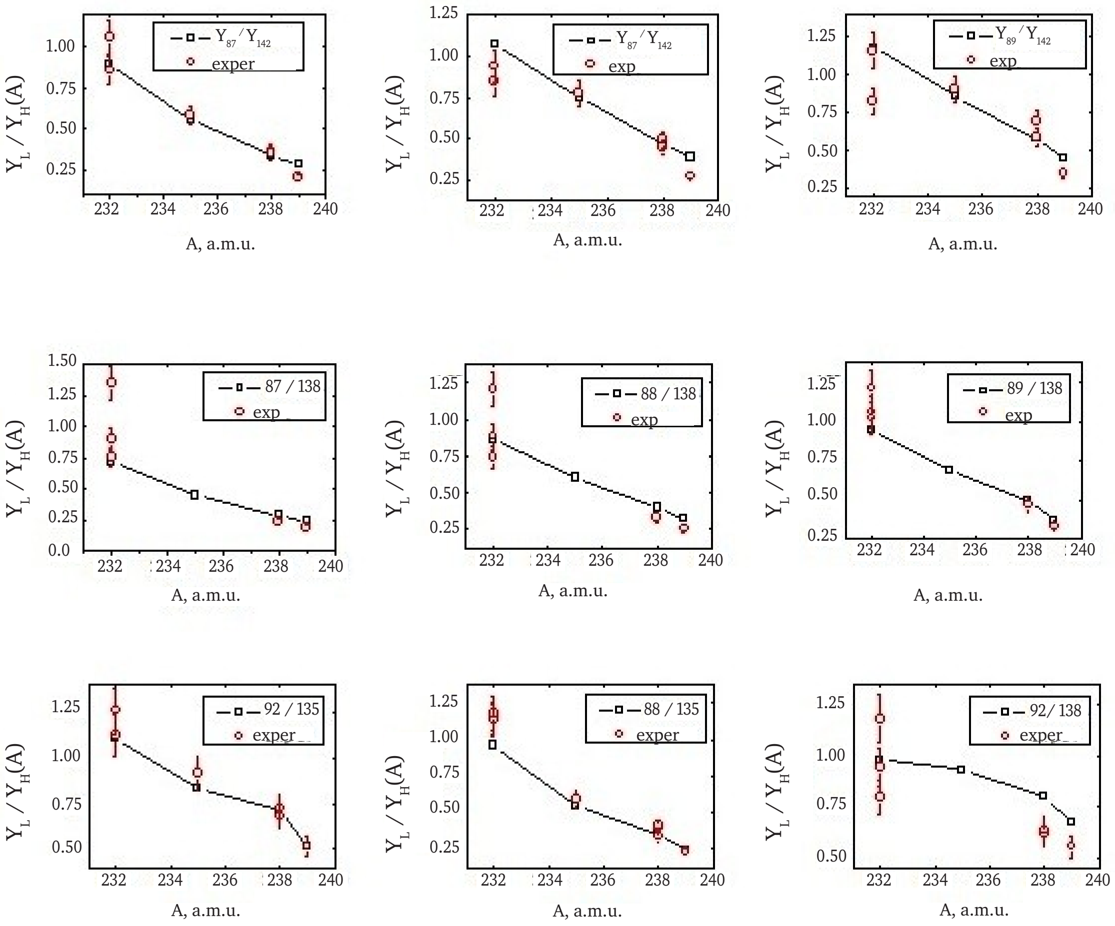

Figure 4. Dependence of the ratios of the numerical values of the yields of light and heavy products on the mass of fissile nuclei ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu

Table 1. Numerical values of the Y_L/Y_H yields ratio for fissile nuclei ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu, and the value of the difference in the yields ratios between adjacent pairs of nuclei

Y_L/Y_H	^{232}Th	%	^{235}U	%	^{238}U	%	^{239}Pu
Y_{88}/Y_{135}	0,95918	43.2	0,54439	34.7	0,35532	31.4	0,24371
Y_{92}/Y_{135}	1,1013	24.0	0,8372	15.2	0,70991	27.8	0,51245
Y_{92}/Y_{138}	0,97806	5.0	0,92957	14.1	0,79844	16.1	0,66998
Y_{87}/Y_{138}	0,71654	37.9	0,44515	36.0	0,28503	17.2	0,23589
Y_{88}/Y_{138}	0,85724	29.5	0,60445	33.9	0,39964	20.3	0,31862
Y_{89}/Y_{138}	0,94446	26.8	0,69173	28.3	0,49569	25.6	0,369
Y_{87}/Y_{142}	0,89638	38.5	0,5129	39.3	0,33437	14.1	0,28735
Y_{88}/Y_{142}	1,0724	30.2	0,74857	37.4	0,46882	17.2	0,38813
Y_{89}/Y_{142}	1,18151	27.5	0,85666	32.1	0,5815	22.7	0,4495

Conclusions

As a result of the calculations, optimal pairs of light and heavy products (with close half-lives) of photofission of actinide nuclei ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu were identified as potential sources of delayed gamma radiation and suitable for analysis of their isotopic composition. It has been demonstrated that combinations of numerical values of the ratio of the fields of light and heavy products (Y_{88} / Y_{135}, Y_{92} / Y_{135}, Y_{92} / Y_{138}, Y_{87} / Y_{138}, Y_{88} / Y_{138}, Y_{89} / Y_{138}, Y_{87} / Y_{142}, Y_{88} / Y_{142}, Y_{89} / Y_{142}) are substantially dependent on the mass of the fissile nuclei.

The calculations made make it possible to determine the ratio of the yields of light and heavy photofission products of actinide kernels for which no experimental data are available, and allow optimization of the procedure of non-destructive analysis of the isotope composition of unshielded and shielded fissile nuclear materials on electronic accelerators – microtron.

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Розрахунок виходів продуктів фотоподілу ядер актинідів – джерел запізнілого гамма-випромінювання для потреб аналізу їх ізотопного складу

І.В. Пилипчинець, Є.В. Олейников, О.О. Парлаг

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Анотація

Мета. Одним з найважливіших завдань атомної промисловості є контроль за нерозповсюдженням ядерних матеріалів, що розщеплюються (наприклад, ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu) на всіх етапах їх використання (переміщення, зберігання тощо). Для успішного вирішення цієї проблеми потрібна достовірна інформація про їх ізотопний склад. Метою дослідження є моделювання масових розподілів продуктів фотоподілу ядер ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu, затримка гамма-випромінювання яких може бути використано для неруйнівного ізотопного аналізу ядерних матеріалів на прискорювачах електронів.

Методи. Розрахунки масових розподілів продуктів фоторозщеплення ядер ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu було проведено із застосуванням GEF-коду. Спектр гальмівного випромінювання було змодельовано під час взаємодії електронів (E=12,5 MeB) із танталовим перетворювачем (1 мм) за допомогою GEANT4 10.7.

Результати. Проведено моделювання масових розподілів продуктів фотоподілу ядер ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu. Коефіцієнт виходів продуктів для ядерних пар (Y_{88}/Y_{135}, Y_{92}/Y_{135}, Y_{92}/Y_{138}, Y_{87}/Y_{138}, Y_{88}/Y_{138}, Y_{89}/Y_{138}, Y_{87}/Y_{142}, Y_{88}/Y_{142}, Y_{89}/Y_{142}) розраховуються для зазначених ядер. Оцінка різниці між числовими значеннями коефіцієнта виходу фрагментів пар для ядерних пар ^{232}Th та ^{235}U, ^{235}U та ^{238}U, ^{238}U та ^{239}Pu зроблено у відсотках. Значення різниці у співвідношеннях виходу цих пар продуктів становлять -5,0÷43,2 %, 14,1÷39,3 % та 14,1÷39,3 % для ядерних пар ^{232}Th та ^{235}U, ^{235}U та ^{238}U, ^{238}U і ^{239}Pu відповідно.

Висновки. Результати моделювання вказують на можливість використання зазначених вище пар продуктів поділу як джерел затримки гамма-випромінювання при проведенні неруйнівного ізотопного аналізу ядерних матеріалів. Отримані результати можуть бути використані для оптимізації експериментів на прискорювачах електронів, що підвищить точність та надійність результатів

Ключові слова: ядерні матеріали, ізотопний аналіз, фотоподіл, виходи продуктів, запізніле гамма-випромінювання

Расчет выходов продуктов фотоделения ядер актинидов – источников запаздывающего гамма-излучения для нужд анализа их изотопного состава

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Аннотация

Цель. Одной из важнейших задач атомной промышленности является контроль за нераспространением ядерных расщепляющихся материалов (например, ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu) на всех этапах их использования (перемещение, хранение и т.д.). Для успешного решения этой проблемы нужна достоверная информация об их изотопном составе. Целью исследования является моделирование массовых распределений продуктов фотоделения ядер ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu, задержка гамма-излучения, которое может быть использовано для неразрушающего изотопного анализа ядерных материалов на ускорителях электронов.

Методы. Расчеты массовых распределений продуктов фоторасщепления ядер ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu было проведено с применением GEF-кода. Спектр тормозного излучения было смоделировано при взаимодействии электронов (E=12,5 МэВ) с танталовым преобразователем (1 мм) с помощью GEANT4 10.7.

Результаты. Проведено моделирование массовых распределений продуктов фотоделения ядер ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu. Коэффициент выходов продуктов для ядерных пар (Y_{88}/Y_{135}, Y_{92}/Y_{135}, Y_{92}/Y_{138}, Y_{87}/Y_{138}, Y_{88} / Y_{138}, Y_{89}/Y_{138}, Y_{87}/Y_{142}, Y_{88}/Y_{142}, Y_{89}/Y_{142}) рассчитываются для указанных ядер. Оценка разницы между числовыми значениями коэффициента выхода фрагментов пар для ядерных пар ^{232}Th и ^{235}U, ^{235}U и ^{238}U, ^{238}U и ^{239}Pu сделано в процентах. Значение разницы в соотношениях выхода этих пар продуктов составляют 5,0÷43,2 %, 14,1÷39,3 % и 14,1÷39,3 % для ядерных пар ^{232}Th и ^{235}U, ^{235}U и ^{238}U, ^{238}U и ^{239}Pu соответственно.

Выводы. Результаты моделирования указывают на возможность использования указанных выше пар продуктов деления как источников задержки гамма-излучения при проведении неразрушающего изотопного анализа ядерных материалов. Полученные результаты могут быть использованы для оптимизации экспериментов на ускорителях электронов, что повысит точность и надежность результатов

Ключевые слова: ядерные материалы, изотопный анализ, фотоделение, выходы продуктов, запаздывающее гамма-излучение