MONTE-CARLO SIMULATION OF RESPONSE OF SEMICONDUCTOR DETECTORS FOR RADIONUCLIDE IDENTIFICATION DEVICES

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Wide band-gap semiconductor detectors have good application prospects in the compact systems intended for detecting low-level radiation and identifying radionuclides. In the present work, the algorithm of identification of radionuclides and determination of their quantitative characteristics was researched. The responses of CdZnTe- and TlBr-detectors to radiation from ^{137}Cs, ^{90}Sr, ^{40}K, ^{131}I and ^{60}Co were simulated by Monte-Carlo method via GEANT4 v. 4.9.6 package for realization of this algorithm. It allowed us to create a databases of spectral signatures for the investigated detectors and to use them as input data for the regression model.

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1. INTRODUCTION

Identification of radionuclides and determination of their quantitative characteristics is a key problem in the environmental remediation. One of the solutions of such problem consists in the preliminary selection and preparation of soil's samples. However, it takes sufficiently long period of time and, in addition to this factor, the given technique is very expensive. The method of in-situ identification of radionuclides and determination of their activity and concentration is cheaper and fast-responding. Over many years, wide band-gap semiconductor detectors have good application prospects in the compact systems intended for detecting low-level radiation and in-situ identifying gamma-radiators. The main advantage of such detectors is the lack of need for their cooling (in contrast to HPGe-detectors). Wide band-gap semiconductor detectors operate at room temperature. Moreover, these detectors can be used for measuring beta radiation spectra [1]. Thus, wide band-gap semiconductor detectors are suitable for working in the mixed radiation fields.

To in-situ identify radionuclides it is necessary to analyze the whole measured spectrum obtained by a detector. The analysis of such spectrum requires knowledge of response of detector to radiation of every radionuclide, which may be contained in the investigated mixture composition. In the present work, the responses of CdZnTe- and TlBr-detectors to mixed gamma-beta radiation and radiation from every radionuclide, which may be contained in the investigated mixture composition, were simulated by the Monte-Carlo method via the GEANT4 v. 4.9.6 package, which is a universal toolkit for the simulating the passage of charged particles, neutrons and gamma quanta through matter. The algorithm of the in-situ identification of radionuclides and the determination of their activity, researched in this work, is based on LASSO (Least Absolute Shrinkage and Selection Operator) regularization [2].

2. DESCRIPTION OF THE MODEL

The measured (or simulated) total spectrum from a mixture of radioactive sources can be presented in the form:

$$\mathbf{Y} = X\boldsymbol{\beta} + \boldsymbol{\varepsilon}. \tag{1}$$

Here, $\mathbf{Y}$ - a vector of the measured (or simulated) spectrum of radiation from the mixture of radioactive sources, X - a detector's response matrix to all individual sources of radioactive radiation, which may be contained in the mixture composition, $\boldsymbol{\beta}$ - a vector of coefficients determining a contribution of each source in the mixed spectrum, $\boldsymbol{\varepsilon}$ - a vector of noise. In detail, the equation (1) can be rewritten in the form:

$$\begin{bmatrix} Y_1 \\ \vdots \\ Y_n \end{bmatrix} = \begin{bmatrix} X_{1,1} & \cdots & X_{1,k} \\ \vdots & \ddots & \vdots \\ X_{n,1} & \cdots & X_{n,k} \end{bmatrix} \times \begin{bmatrix} \beta_1 \\ \vdots \\ \beta_k \end{bmatrix} + \begin{bmatrix} \varepsilon_1 \\ \vdots \\ \varepsilon_n \end{bmatrix}. \tag{2}$$

Here, n – a number of channels in the detector, k – a number of radioactive sources, which may be contained in the mixture composition.

The responses of CdZnTe- and TlBr-detectors to mixed gamma-beta radiation, $\mathbf{Y}$, and radiation from every radionuclide, which may be contained in the investigated mixture composition, X, were simulated by the Monte-Carlo method via the GEANT4. The vector of unknown coefficients, $\boldsymbol{\beta}$, was determined by

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a method of LASSO regularization based on the minimization of functional:

$$\|\mathbf{Y} - X\boldsymbol{\beta}\|_2^2 + \mu|\boldsymbol{\beta}|_1 = min, \mu \geq 0, \quad (3)$$

where μ – a regularization parameter. Such kind of the regularization was applied to problems of the radionuclide identification by Bai et al. [3]. They used the LASSO regularization to determine which radionuclides are contained in the mixed radiation source. However, they did not conduct any investigation connected with the determination of the radionuclide activity.

In the present work, the regularization problem was solved numerically using the coordinate descent method. The peculiarities of the application of this algorithm to solving the LASSO regularization problem were researched in detail by Wu and Lange [4].

To determine the regularization parameter, μ, we used a modified cross-validation algorithm. The method essence consists in the minimization of the standard deviation of spectrum, calculated via the investigated linear model, from the spectrum simulated via Monte-Carlo method. Thus, an expression

$$CV = \frac{1}{n}\sum_{i=1}^{n}(y_i - X_{ij}\beta_j)^2 \quad (4)$$

needs be minimal with the optimal regularization parameter.

3. MONTE-CARLO SIMULATION OF CdZnTe- AND TlBr-DETECTORS

The passage of gamma-quanta and electrons through the wide band-gap semiconductor detectors (CdZnTe and TlBr) was simulated by Monte-Carlo method via the user program code, embedded in GEANT4 v. 4.9.6 package. It was described in detail in [5]. This user program code allows the user to specify the detector's characteristics and the initial characteristics of the radiation.

To validate this program we compared the simulated response of the $6{\times}6{\times}3\,mm^3$ planar CdZnTe-detector with the experimental one. The bias voltage, U_b, was 150 V. The detector's dark current was $6\,nA$. The mobility-lifetime products for electrons, $(\mu\tau)_e$, and holes, $(\mu\tau)_h$, were $4{\times}10^{-4}$ cm^2/V and $5{\times}10^{-6}$ cm^2/V, respectively. Even with such characteristics of the CdZnTe-detector, a satisfactory response was obtained. It was shown that the response functions of the CdZnTe-detector simulated via the described user program code [5] agree with experimental radiation spectra of ^{137}Cs (Fig.1) and ^{90}Sr (Fig.2) obtained in [1]. Visible discrepancies can be due to the simplified geometry description.

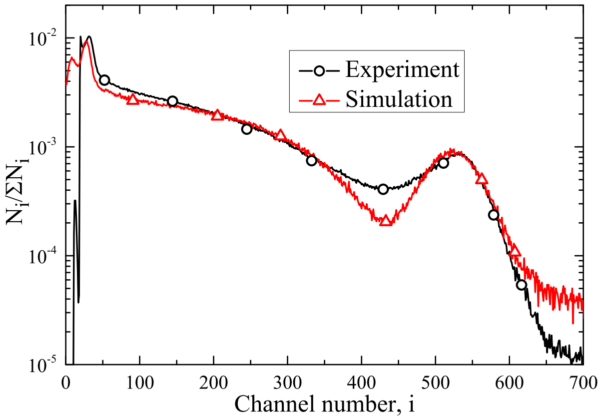

Fig.1. *Spectrum of ^{137}Cs*

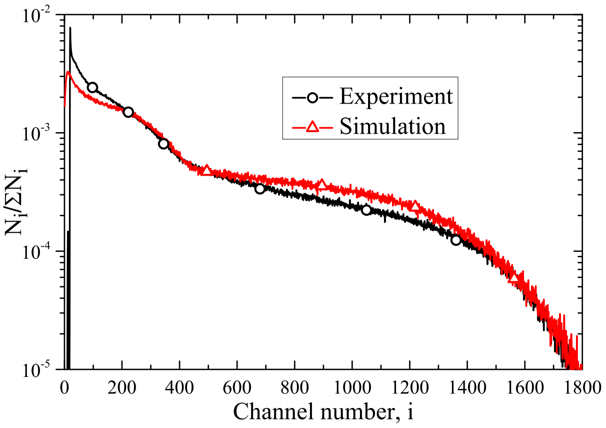

Fig.2. *Spectrum of ^{90}Sr*

In following investigations, the CdZnTe-detector with better characteristics was studied. We carried out a computer experiment, in which the bias voltage, U_b, was 300 V. The total level of noise in the CdZnTe spectrometry systems was specified at about 300 e$^-$. It was assumed that the detector's dark current was $3\,nA$. The mobility-lifetime products for electrons, $(\mu\tau)_e$, and holes, $(\mu\tau)_h$, were selected at $3{\times}10^{-3}$ cm^2/V and $3{\times}10^{-4}$ cm^2/V, respectively. In the present work, we study the in-situ identification of radionuclides, which are contained in the mixed emission source and the in-situ determination of their activity. (^{137}Cs + ^{90}Sr) source is of interest. However, to obtain a universal algorithm apart from ^{137}Cs and ^{90}Sr we researched ^{40}K, ^{131}I and ^{60}Co. Thus, our mixed emission source contains short-lived as well as long-lived radionuclides. Moreover, we simulated pure beta as well as beta- gamma- radiators.

In the most cases of the in-situ identification of radionuclides, a radioactive source is placed under the soil attenuating its radiation. In such situations, for correct operation of the studied algorithm it is not enough to define the response functions of the CdZnTe-detector to radiation from each radionuclide under certain conditions, we must know the dependence of attenuation of each radionuclide radiation from the soil depth. It is necessary to pay attention to that fact that the presence of the radiation source deep under the soil was not included in our investigation. In the present work, we determined the thickness of the surface soil layer, in which the insignificant attenuation of radiation from ^{137}Cs, ^{90}Sr, ^{40}K, ^{131}I and ^{60}Co was observed. Figs.3 and 4 indicate how the distribution of the energy absorbed in the CdZnTe-detector irradiated by ^{137}Cs- and ^{90}Sr-sources, respectively, changes with the increasing depth of soil which covers these sources. In these computer experiments (see Figs.3 and 4), for good statistics it was supposed that all particles move toward the direction of the detector.

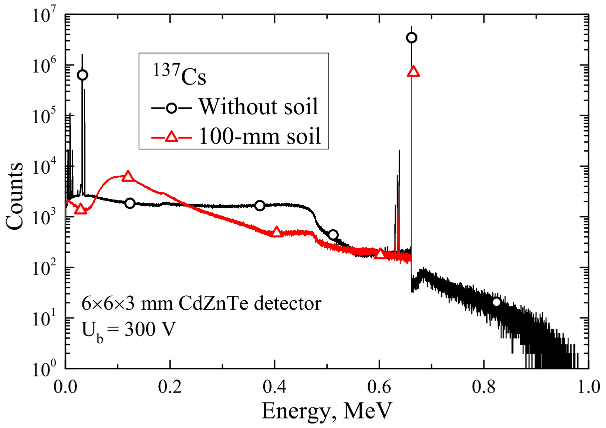

Fig.3. *Change in the distribution of the energy absorbed in the CdZnTe-detector irradiated by ^{137}Cs source with the increasing soil depth, covering the source*

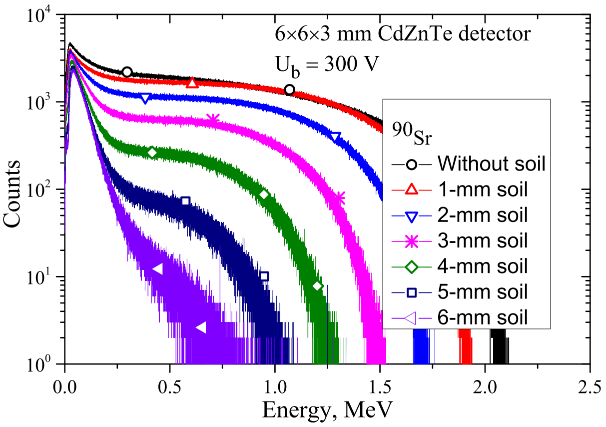

Fig.4. *Change in the distribution of the energy absorbed in the CdZnTe-detector irradiated by ^{90}Sr source with the increasing soil depth, covering the source*

The depth of location of the radioactive point source of ^{137}Cs under the soil changed from 1 mm to 200 mm. In order not to overload the figure, Fig.3 presents a change in the distribution of the energy absorbed in the CdZnTe-detector irradiated by ^{137}Cs source only with the 100 mm soil depth, covering the source. Fig. 3 shows that the soil with 100 mm thickness attenuates gamma-radiation in the 661.7 keV photopeak by 83.7 %. With increasing the soil thickness up to 200 mm this value is attenuated by more than 95 % (it is not shown on the figure). Overall, we can detect a radiation from ^{137}Cs by the CdZnTe-detector if the depth of soil, under which the source presents, is not more than approximately 100 mm.

Fig.4 demonstrates the simulation results obtained with ^{90}Sr source, the location of which under the soil changed from 1 mm to 6 mm with a discretization interval of 1 mm. The soil considerably attenuates beta-radiation from ^{90}Sr. The soil with thickness of 1 mm attenuates beta-radiation from ^{90}Sr by 10.9 %, 2 mm – by 42.8 %, 3 mm – almost by 70 %, 5 mm – by more than 90 %. Thus, it is not possible to detect the ^{90}Sr source by the CdZnTe-detector at the thickness of soil covering the source more than approximately 10 mm.

In the present work, we studied the mixed radioactive volume source placed in the sufficiently thin surface layer of soil, the attenuation by which can be neglected. We simulated the response functions of the CdZnTe-detector to radiation from individual volume ^{137}Cs, ^{90}Sr, ^{40}K, ^{131}I and ^{60}Co sources with radius of 0.5 mm. Some of these curves are presented on Figs.5, 6. The activity of every source was 100 MBq. The measurements were taken in the 4π geometry.

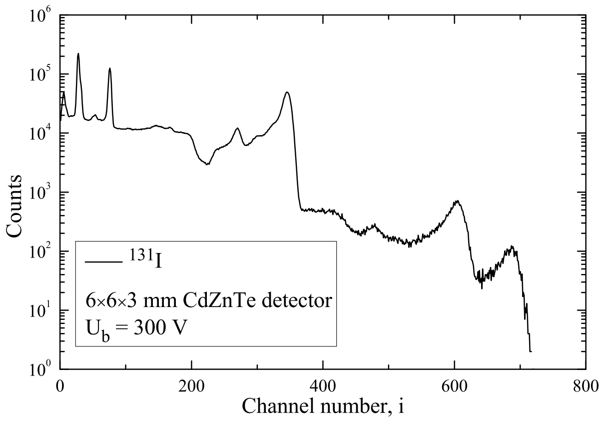

Fig.5. *Spectrum of ^{131}I*

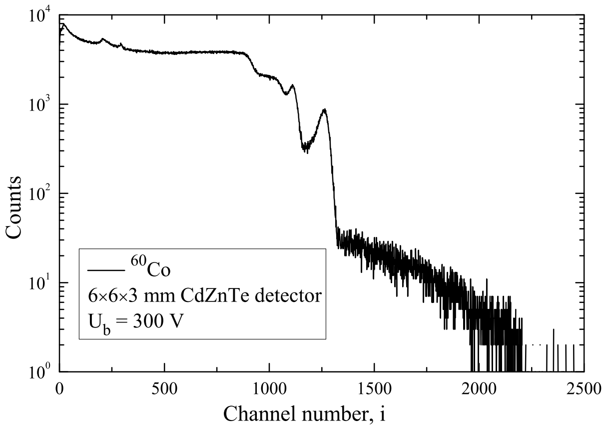

Fig.6. *Spectrum of ^{60}Co*

4. IDENTIFICATION OF RADIONUCLIDES

To check the described algorithm of the radionuclide identification we simulated radiation from the mixed radioactive source with radius of 0.5 mm which consists of ^{137}Cs, ^{90}Sr, ^{40}K and ^{131}I with the ratio of activities of 0.4:0.1:0.1:0.4, respectively (Fig.7).

At the deconvolution of the spectrum of this source we suppose that source can include ^{137}Cs, ^{90}Sr, ^{40}K, ^{131}I and ^{60}Co, which is absent in fact. As the result of the deconvolution of spectrum of the radiation from the given mixed source the β coefficient vector was determined: this value is (0.3986, 0.0979, 0.0916, 0.4023, 0). Thus, it was determined that ^{60}Co is not present in the mixed source. Using this β coefficient vector and our reference individual sources, we determined a mixture composition and calculated the activity of radionuclides.

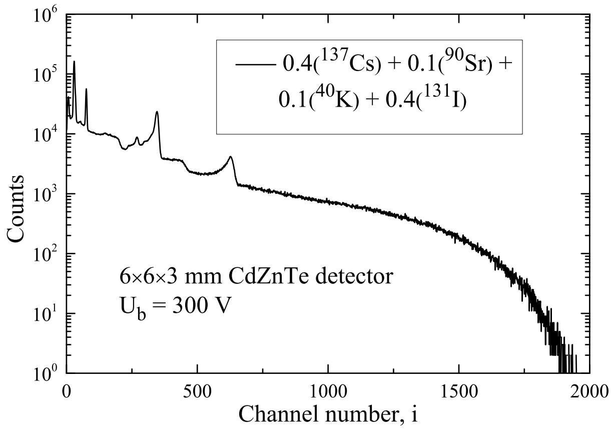

Fig.7. *Spectrum of radiation from a mixed source of (^{137}Cs + ^{90}Sr + ^{40}K + ^{131}I) obtained by CdZnTe-detector*

In the present work, we also simulated the response of 2.7×2.7×2 mm planar TlBr-detector equipped with ohmic contacts (Fig.8). The bias voltage was chosen as 400 V. We specified the total level of noise in the TlBr-spectrometry systems at about 400 e^-. The mobility-lifetime products for electrons, $(\mu\tau)_e$, and holes, $(\mu\tau)_h$, were 5×10^{-4} cm^2/V and 5×10^{-5} cm^2/V, respectively.

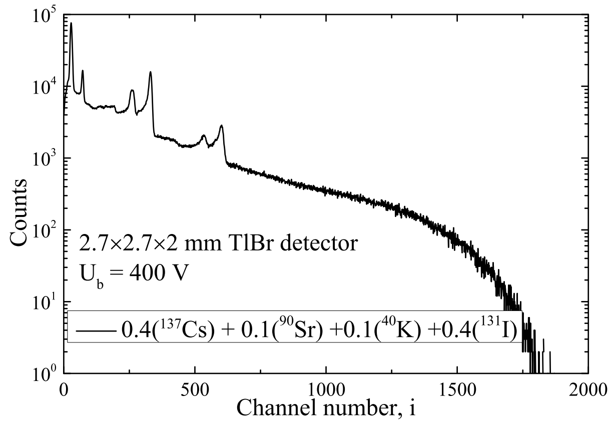

Fig.8. *Spectrum of radiation from a mixed source of (^{137}Cs + ^{90}Sr + ^{40}K + ^{131}I) obtained by TlBr-detector*

The same mixed source was used. Results of the algorithm realization, obtained with using TlBr-detector, agree with results, obtained with CdZnTe-detector. In Table 1, the values of the given activity of radionuclides, which are contained in the mixed source, are compared with the computed values.

The given values of the radionuclide activity agree with the values of the radionuclide activity obtained via the algorithm investigated in this article for CdZnTe- and TlBr-detectors.

Table 1. *Activity of radionuclides from the mixed radiation source of (^{137}Cs + ^{90}Sr + ^{40}K + ^{131}I)*

Radiation source	Given activity, MBq	Activity calculated via the researched algorithm, MBq	
		CdZnTe	TlBr
^{137}Cs	4	3.99	3.88
^{90}Sr	1	0.98	0.93
^{40}K	1	0.92	0.95
^{131}I	4	4.02	4.05
^{60}Co	0	0	0

If we increase the distance between the investigated mixed radioactive source and the CdZnTe-detector, the intensity of radiation from source and β coefficients will decrease, respectively (Table 2). However, one can observe which radionuclides are present or absent in the researched source and how values of their activity correlate to each other.

Table 2. *The values of coefficients, β, at different distances between the mixed radiation source and CdZnTe-detector*

Source	Distance, mm				
	0.5	1.0	1.5	2.5	4.5
^{137}Cs	0.40	0.34	0.29	0.21	0.12
^{90}Sr	0.10	0.09	0.08	0.07	0.04
^{40}K	0.09	0.06	0.05	0.04	0.04
^{131}I	0.40	0.33	0.27	0.19	0.10
^{60}Co	0	0	0	0	0

Similar results will be obtained, if we increase the distance between the investigated mixed radioactive source and the TlBr-detector (Table 3).

Table 3. *The values of coefficients, β, at different distances between the mixed radiation source and TlBr-detector*

Source	Distance, mm				
	0.5	1.0	1.5	2.5	4.5
^{137}Cs	0.39	0.28	0.20	0.11	0.05
^{90}Sr	0.09	0.07	0.05	0.03	0.01
^{40}K	0.10	0.08	0.07	0.05	0.03
^{131}I	0.40	0.28	0.20	0.12	0.05
^{60}Co	0	0	0	0	0

If we increase a square of source, remaining the same distance between this source and CdZnTe- (or

TlBr-) detector, correct values of β coefficients and correct values of radionuclide activities will be obtained.

Overall, as we can see from testing the investigated algorithm the obtained method allows identifying the radionuclides and determining the ratio of their activities. This ratio does not change with the increasing or decreasing distance between the CdZnTe- (or TlBr-) detector and soil. The measurement of spectra of an unknown mixed source at the same distance as the spectra of individual sources were measured gives a possibility to determine not only qualitative characteristics of radionuclides but also their quantitative characteristics, for example, activity. We would remind that all investigations and calculations were conducted for contaminated very thin surface layer of soil. However, in reality, in the most cases, contaminated elements are depth distributed in the soil that is a drawback for in-situ measurements. Usually, it is supposed that natural radionuclides are uniformly distributed, and artificial ones are exponentially distributed [6]. The existent methods of the in-situ activity measurements of radionuclides depth distributed in the soil are based on three algorithms: the multiple photopeak method, the collimation or lead plate method and the peak-to-valley ratio method [7]. In following research we plan to study a use of the algorithm considered by us together with some existent algorithm of in-situ activity measurement or together with its modification.

5. CONCLUSIONS

The algorithm of the reconstruction of spectra of radiation from mixed beta- gamma-sources placed in the sufficiently thin surface layer of soil, the attenuation by which can be neglected, is presented. The investigated method based on the linear regression gives a possibility to in-situ identify the radionuclides, which are contained in these mixed sources, and to in-situ calculate the ratio of their activities. The response functions of the CdZnTe- and TlBr-detectors to the radiation from individual sources of ^{137}Cs, ^{90}Sr, ^{40}K, ^{131}I and ^{60}Co were simulated via the Monte-Carlo method using the GEANT4 v.4.9.6 package. The obtained response functions allowed creating the databases of spectral signatures for two researched CdZnTe- and TlBr-detectors and using them as the input data for the linear regression model. We identified the radionuclides included in the mixed source of (^{137}Cs + ^{90}Sr + ^{40}K + ^{131}I) with radius of 0.5 mm, simulated at the same distance between source and detector, and calculated their activity with a good accuracy at radiation registration by CdZnTe- and TlBr-detectors. It was shown how values of coefficients determined via the researched model for each radionuclide from the mixed radiation source of (^{137}Cs + ^{90}Sr + ^{40}K + ^{131}I) with radius of 0.5 mm change with the increasing distance between this source and the CdZnTe- (or TlBr-) detector.

The maximum thicknesses of soil, under which ^{137}Cs and ^{90}Sr isotopes can be placed for detecting their radiation by the CdZnTe-detector with the specified characteristics placed above the soil, were calculated. We concluded that one can detect the radiation from ^{137}Cs and ^{90}Sr sources by the researched CdZnTe-detector if the thicknesses of soil, under which the sources are placed, are not more than approximately 100 mm and 10 mm, respectively.

Overall, it was shown that the investigated algorithm can be used for identifying radionuclides and determining the ratio of their activities. The values of the radionuclide activities can be obtained under certain conditions. To do a universal algorithm we plan to modify it for determining the values of radionuclide activities at different distance between the CdZnTe- (or TlBr-) detector and the source, which will be depth distributed in the soil.

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МОДЕЛИРОВАНИЕ МЕТОДОМ МОНТЕ-КАРЛО ОТКЛИКА ПОЛУПРОВОДНИКОВЫХ ДЕТЕКТОРОВ ДЛЯ ПРИБОРОВ, ПРЕДНАЗНАЧЕННЫХ ДЛЯ ИДЕНТИФИКАЦИИ РАДИОНУКЛИДОВ

А. И. Скрыпник, М. А. Хажмурадов

Широкозонные полупроводниковые детекторы перспективны для использования в компактных системах, предназначенных для регистрации низкоэнергетического излучения и идентификации радионуклидов. В данной работе исследовался алгоритм идентификации радионуклидов и определения их количественных характеристик. Для реализации данного алгоритма были промоделированы с помощью GEANT4 отклики полупроводниковых CdZnTe- и TlBr-детекторов на излучение ^{137}Cs, ^{90}Sr, ^{40}K, ^{131}I и ^{60}Co. Это позволило создать базы спектральных сигнатур для исследуемых детекторов и использовать их в качестве входных данных для регрессионной модели.

МОДЕЛЮВАННЯ МЕТОДОМ МОНТЕ-КАРЛО ВІДГУКУ НАПІВПРОВІДНИКОВИХ ДЕТЕКТОРІВ ДЛЯ ПРИЛАДІВ, ЯКІ ПРИЗНАЧЕНІ ДЛЯ ІДЕНТИФІКАЦІЇ РАДІОНУКЛІДІВ

А. І. Скрипник, М. А. Хажмурадов

Широкозонні напівпровідникові детектори перспективні для використання в компактних системах, призначених для реєстрації низькоенергетичного випромінювання та ідентифікації радіонуклідів. У даній роботі досліджувався алгоритм ідентифікації радіонуклідів та визначення їх кількісних характеристик. Для реалізації даного алгоритму були промодельовані за допомогою GEANT4 відгуки напівпровідникових CdZnTe- і TlBr-детекторів на випромінювання ^{137}Cs, ^{90}Sr, ^{40}K, ^{131}I та ^{60}Co. Це дозволило створити бази спектральних сигнатур для досліджуваних детекторів і використати їх в якості вхідних даних для регресійної моделі.