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SIMULATION USING GEANT4 OF AXAS-D SYSTEM WITH REGISTRATION K-SERIES OF IODINE IN ACTIVATED CARBON FOR VENTILATION SYSTEMS NPP

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The material shows the results of using the Geant4 program to simulate the operation and capabilities of the SKIF stand when recording radiation K-series with detector the AXAS-D of stable iodine in activated carbon, which is used for air purification at the NPP. The radiation spectra obtained in different situations are shown: at the entrance and exit of the measuring installation; and their processing. The parameters of the registration system were obtained – efficiency and dependence on the thickness of the detector crystal.

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INTRODUCTION

An important impact of air pollution by gas-aerosol emissions, which contain radionuclides, includes the technological waste of nuclear reactor operation, which is removed from the premises of the NPP during ventilation. Taking into account the half-life of radionuclides in these emissions and their radiation effect, the greatest danger is iodine isotopes ^{131}I, ^{133}I and isotopes of inert radioactive gases ^{133}Xe, ^{135}Xe, ^{85}Kr, and ^{41}Ar. Isotope ^{131}I (half-life 8.5 days) is particularly dangerous for humans. This is due to its biochemical identity with stable iodine, which plays a vital role in the body, selectively accumulating in the thyroid gland, being included in the composition of hormones involved in the regulation of metabolic processes, as well as the growth and functioning of various organs and the human body.

Considering the special importance of iodine, the IAEA has developed a standard [1] that regulates the content of iodine in the ventilation air of nuclear power plants. But taking into account the mobility of iodine and methyl iodide molecules, only the methyl iodide molecule was left behind. Similar standards for the certification of activated carbon were approved in the USA [2] and Europe [3]. These documents are based on radioactive methyl iodide with a radiation energy of 364 keV.

For certification, we used samples of activated carbon, which include the stable iodine isotope ^{127}I [4].

The material presents a comparison of the results of modelling using the GEANT4 package [5] with data measured by X-ray radiometric analysis (XRA) on the SKIF stand of a standard sample with concentration stabile iodine of 0.5% in coal.

SKIF STAND AND ITS ELEMENTS

If, as in our case, a stable iodine molecule is used, then the SKIF stand for measuring iodine content by the XRA method in activated carbon looks like this (Fig. 1).

The "IHIA-1m-5" source with the isotope ^{241}Am with an activity of $3.9 \cdot 10^9$ Bq was used as a radiation source. Radiation from a source with an energy of 59.6 keV is used, which excites the radiation of K-lines of the iodine series by the X-ray fluorescence method. Modification of the spectrum of the source and suppression of low-energy lines of 12…20 keV is performed at the expense of a 180 μm thick Pb foil. The "IHIA-1m-5" source is placed in a lead protective case for the safety of personnel.

As a detector of iodine radiation with an energy of 28…34 keV, several detectors [6] were previously used, but the Si detector and spectrometric equipment of the KETEK company [7] was chosen as the final detecting system, for which the simulation was performed.

A sample of activated carbon in the amount of ~ 5 g was placed in a plastic case in the form of a truncated cone with the calculation that this
cone performed two tasks: the entire substance of the cone was irradiated with radiation from a ^{241}Am source; all excitation lines of iodine in the investigated substance were registered by the detector.

EXPERIMENT AND SIMULATION

EXPERIMENTAL MEASUREMENTS

Fig. 2 shows the spectrum obtained by the AXAS-D detector when irradiated with gamma quanta from a ^{241}Am source of a standard sample with a content of 0.5 mass.% of the stable ^{127}I iodine isotope. The range of energies registered by the detector is within 0…40 keV, thus cutting off the spurious signal from Compton gamma quanta. The total area of photopeaks of characteristic iodine quanta in the region of about 28.6 keV (transition KL2.3) (Fig. 3) is 8,670 counts, which at a measurement time of 4,000 s corresponds to the rate of recruitment to the photopeak of ~ 2.17 counts/s.

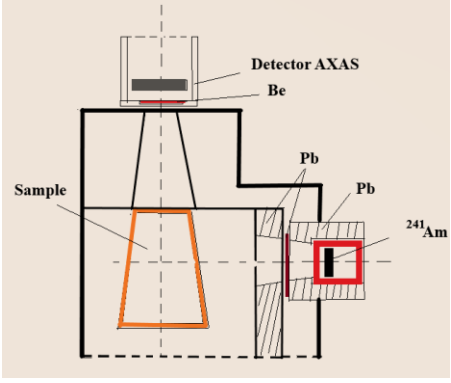

Fig. 1. Measuring part of the SKIF stand

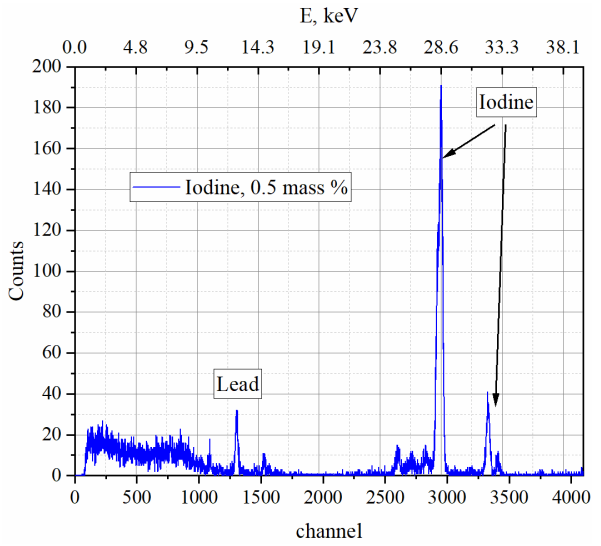

Fig. 2. The spectrum of characteristic radiation of iodine and background radiation obtained by the AXAS-D detector

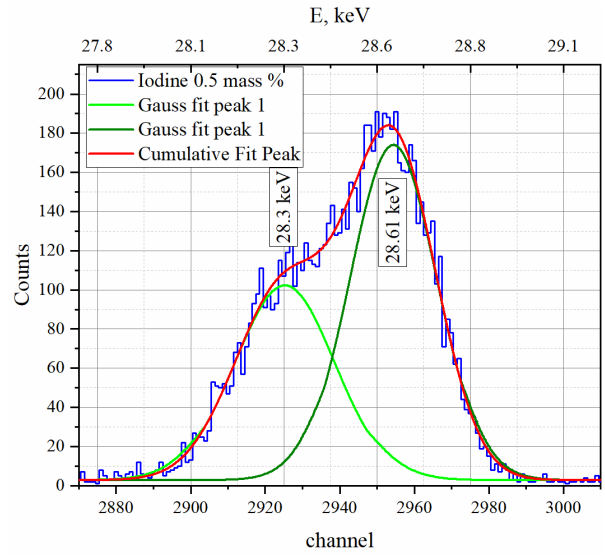

Fig. 3. Spectrum of iodine radiation in the energy range of about 28.6 keV

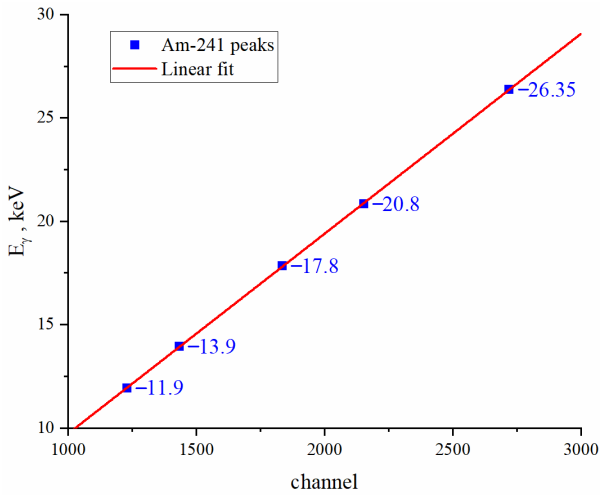

Fig. 4. Dependence of $E_{kx} = f(n)$ for AXAS-D

With the activity of the radiation source ^{241}Am $A_{241Am} = 3.9 \cdot 10^9$ Bq, we obtain that for a test sample of activated carbon with an iodine content of 0.5 mass.%, the coefficient of registration of the characteristic radiation of iodine for the installation XRA of the above configuration (see Fig. 1) $K_{iod\,exp} = 5.56 \cdot 10^{-10}$ quantum/s/Bq (quantum/decay).

Fig. 4 shows the position of the photopeaks of the ^{241}Am source in the distribution measured by AXAS-D. Linear fitting parameters: $E_\gamma = (9.68 \times k + 11.69)$ eV, where k is the channel number. The coefficient of determination, which characterizes the quality of the fit, is 0.99993. The standard deviation of the angular coefficient of the linear fit is ± 0.088%.

MODEL OF THE MEASURING INSTALLATION

The simulation of the detector response was performed using the Geant4 package [5]. The geometry of the model (Fig. 5) corresponds to the real SKIF installation (see Fig. 1).

When modeling electromagnetic interactions, the G4EmPenelopePhysics model was used, and by analogy with the TestEm5 example, the following options were activated:

/process/em/fluo true;
/process/em/pixe true;
/process/em/auger true;
/process/em/augerCascade true;
/process/em/deexcitationIgnoreCut true.

The G4RadioactiveDecayPhysics model is used to simulate the decay of the ^{241}Am isotope.

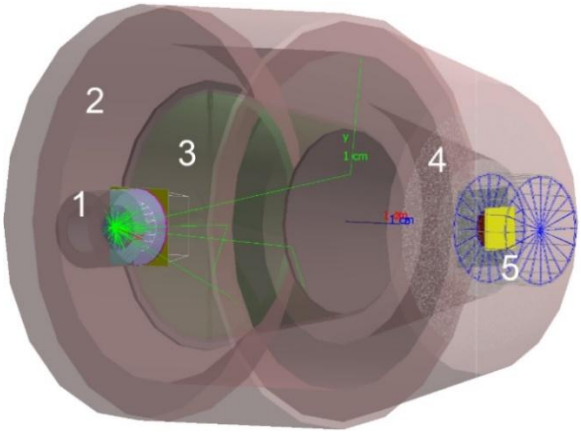

Fig. 5. Geometry model of the measuring installation in Geant4: 1 – the source of gamma radiation ^{241}Am; 2 – body from lead; 3 – container with activated carbon capsules; 4 – output aluminum filter; 5 – Si detector KETEK AXAS-D. The green lines show the trajectories of gamma quanta, the red lines show the trajectories of electrons

The semiconductor detector model used in this work is similar to the model described in detail in [8, 9]. For calculations, the indicated model was adjusted by the specification of the KETEK AXAS-D detector. Table 1 shows the characteristics at which the simulation was performed.

Table 1

Model characteristics of the irradiated iodine filter sample

Parameter	Size
Composition	Carbon – 99.5 mass.% Iodine – 0.5 mass.% The distribution of constituent elements is uniform in volume
Density, g/cm^3	0.5
Mass, g	7.13
Shape	Truncated cone
The density of the cores of the constituent elements, 1/mm^3	Carbon – $2.49 \cdot 10^{19}$ Iodine – $1.19 \cdot 10^{16}$
The ratio of the number of iodine nuclei to the number of carbon nuclei	$k_i \approx 4.8 \cdot 10^{-4}$

EFFICIENCY OF REGISTRATION BY THE KETEK AXAS-D DETECTOR

The calculated full efficiency of registration of characteristic iodine quanta with an energy of 28.6 keV by the AXAS-D detector is ~ 13.47%, the scattering efficiency in the 28.6 keV photopeak is ~ 13.22%. That is, almost all characteristic quanta of iodine (98.21%) interacting with the detector material fall into the photopeak region of the spectrum (Fig. 6).

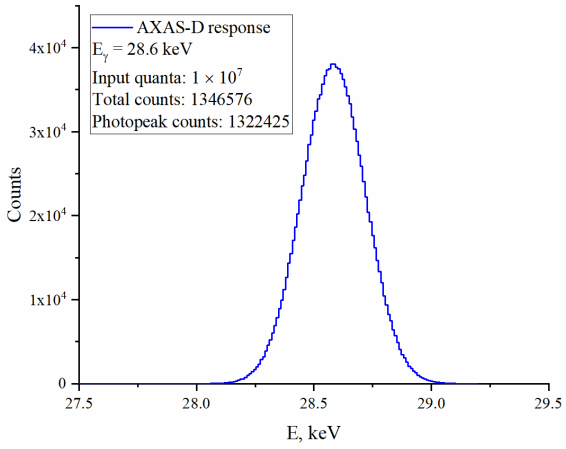

Fig. 6. Response of the AXAS-D detector to gamma quanta with an energy of 28.6 keV

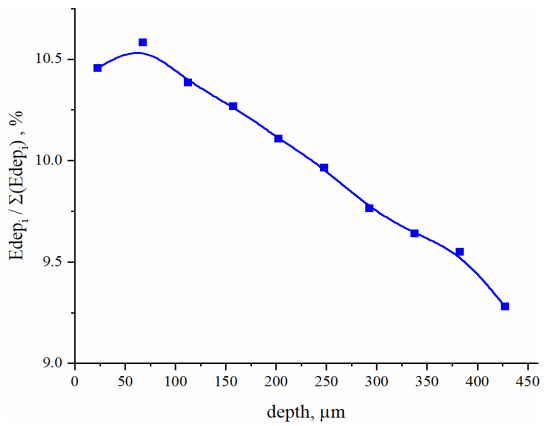

Fig. 7. Distribution of absorbed energy gamma quanta of 28.6 keV across the thickness of the detector

The distribution of the absorbed energy of gamma quanta of 28.6 keV across the thickness of the detector is quite uniform (Fig. 7). In the simulation, the detector was divided into 10 layers with a thickness of 45 μm, E_{depi} is the absorbed energy in the i_{th} layer. The maximum absorbed energy (up to ~ 10.6 % /45 μm) occurs in the region of 0…90 μm with a further gradual decrease to Fig. 7. Distribution of absorbed energy gamma quanta of 28.6 keV across the thickness of the detector ~ 9.3 %/ 45 μm.

INPUT FLUX OF GAMMA RADIATION FROM THE SOURCE

The modeled distribution of gamma quanta at the entrance to the SKIF installation is shown in Fig. 8. n_i is the number of quanta that, during the simulation, got into the i_{th} channel with a width of 1 keV. To demonstrate the fine structure of the input spectrum along the ordinate axis, a logarithmic scale was chosen. The main share of the input spectrum (≥ 97%) is made up of gamma rays with an energy of 59.54 keV. Gamma quanta with lower energies, formed during the decay of the ^{241}Am isotope, do not pass through the output lead filter of the radiation source (see Fig. 1) with a thickness of ≈ 180 μm. The continuous spectrum in the region ~ 30…60 keV is Compton quanta produced by the scattering of gamma quanta with an energy of 59.54 keV in this lead output filter. A minor contribution from scattering in the air is also possible. The 9…16 keV region is the characteristic radiation of lead.

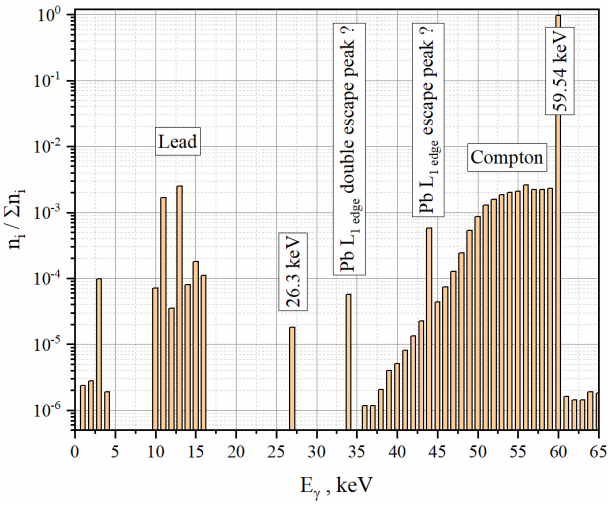

Fig. 8. Distribution of gamma quanta from the source the ^{241}Am entrance to the SKIF installation

In the spectrum of gamma quanta produced by the decay of ^{241}Am nuclei, there is a large number of lines of very low intensity with energies above 60 keV [8]. Their presence in the calculated spectrum can be seen on the right edge of the distribution in Fig. 8. Considering that the 59.54 keV line has an intensity of more than 97%, the probable contribution of other quanta to the spectrum at the output of the installation can be neglected. Also, the calculation showed that with the given geometry of the installation (see Fig. 5) only about 3.3% of the total number of gamma quanta with an energy of 59.54 keV, formed during the decay of ^{241}Am nuclei in the radiation source used. That is,

~ 0.012 quantum/decay enters the model setup. This is a factor that significantly affects the speed of simulation.

THE FLOW OF GAMMA QUANTA AT THE OUTPUT OF THE INSTALLATION XRA

The calculated distribution of gamma quanta at the output of the installation XRA with an iodine content of 0.5% in the studied sample is shown in Fig. 9. n_i is the number of quanta that, during the simulation, got into the ith channel with a width of 1 keV. As can be seen, the main part of the flow (~ 80%) consists of Compton quanta, which are formed during the scattering in the installation of gamma quanta with an energy of 59.54 keV. A small amount (< 3.5%) also contains quanta with an energy of 59.54 keV, which underwent elastic scattering.

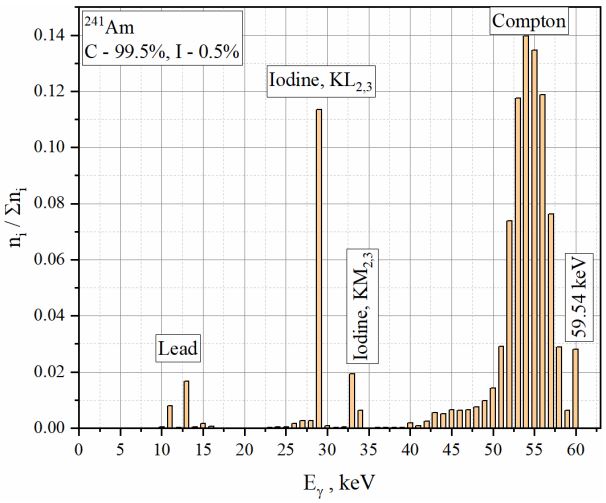

Fig. 9. Distribution of scattered and characteristic gamma quanta at the exit from the installation XRA

The characteristic radiation K of the iodine series is present in much smaller quantities than Compton quanta, but it is sufficient to be registered by the detector. The highest intensity in the output spectrum among the characteristic lines of iodine (≈ 11.3%) is the line with an energy of ~ 28.6 keV (transition KL2.3), which is actually used to calculate the iodine content in the carbon filter. The simulation also shows the presence in the output spectrum of a small amount (~ 2.4%) of the characteristic radiation of lead, from which the protective shell of the installation is constructed.

According to simulation data, characteristic iodine quanta with an energy of ~ 28.6 keV when irradiated from a ^{241}Am source, equal to $\approx 7.52 \cdot 10^{-7}$ quanta/s/Bq/cm^2, were obtained from the XRA installation. When the activity of the ^{241}Am source is $3.9 \cdot 10^9$ Bk, the value of the output flow of characteristic quanta should be out 28.6 keV $\approx 2.9 \cdot 10^3$ quanta/s/cm^2.

ANALYSIS OF REPRODUCTION OF K-SERIES PHOTOPEAKS OF IODINE

The detailed spectrum in the region of the characteristic radiation K of the iodine series is presented in Fig. 10. The shape and amplitude of the peaks with energy ~ 28.6 (transition KL2.3) and ~ 32.2 keV (transition KM2.3) generally agree well. Unfortunately, the spectrum acquisition time in the experiment was insufficient to obtain a stable shape (smoothed curve) of the peaks. However, even under such conditions, the calculated response of the detector shows that the model adequately reproduces the physical processes in the XRA and the features of the AXAS-D detector used.

Quantitative characteristics of the experimental and modeled photopeak ~ 28.6 keV are given in the Table 2. This peak is formed from two characteristic lines (KL2,3), the shape of which can be approximated by a Gaussian distribution (see Fig. 3). According to the passport data, the AXAS-D detector with an active area of 30 mm^2 provides a resolution for the K-line of Mn (≈ 5.89 keV) no worse than 2.26%. As the energy of gamma rays increases, the resolution of the detector improves. For the measurement data, a resolution of 1.0% was obtained on the 28.6 keV line.

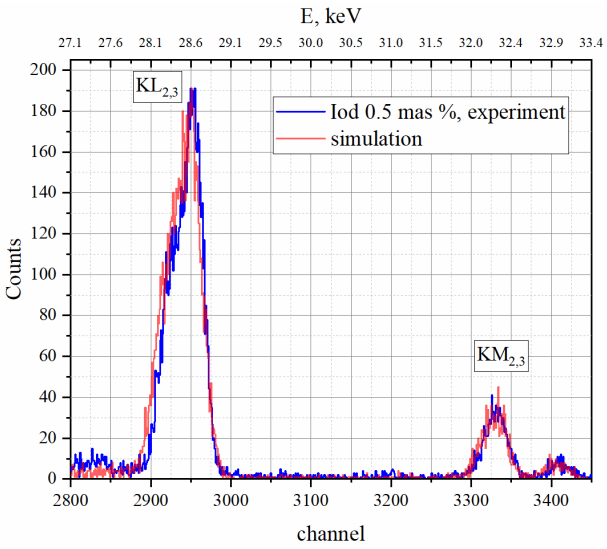

Fig. 10. The range of energies of characteristic radiation K-series of iodine

The data in Table 2 show that the model well reproduced the main characteristics of the component lines of the photopeak. At the same time, the simulated photopeaks turned out to be somewhat wider (by approximately 13%) than the experimentally measured ones.

This led to a proportional underestimation of the amplitude of the main line of 28.61 keV by about 14%. The possible reasons for such an overestimation of the width of the photopeak of 28.61 keV include a slightly higher noise level in the model of the spectrometric path and errors in the dimensions and elemental composition in the model of the measuring installation. However, the total area of the modeled photopeak (cumulative fit peak square) of 28.6 keV exceeds the experimental data by only 4%.

Table 2

Characteristics of the experimental and simulated photopeak ~ 28.6 keV (see Fig. 10)

Experiment						
Line number	Center, canal	Center, keV	FWHM, canal	FWHM, eV	Amplitude, counts	Integral, counts
1	2925.26	28.33	31.39	315.5	99.47	3323.2
2	2954.39	28.61	27.2	275.0	171.17	4956.0
Model						
1	2920.88	28.29	36.22	362.3	99.11	3819.1
2	2951.2	28.58	31.67	318.3	142.71	4810.7

CONCLUSIONS

The use of the KETEK-AXAS-D silicon detector made it possible to significantly reduce the level of background and "parasitic" signals in the PPA spectra of samples with a small concentration of iodine. Also, the good resolution of the detector provides a more accurate measurement of iodine concentration.

The developed PPA model with the AXIS-D detector satisfactorily reproduces the experimental measurements of the characteristic K radiation of the low-concentration iodine series in activated carbon samples.

Although the width of the modeled photopeaks turned out to be somewhat larger than in the experimental spectra, the total photopeak area of 28.6 keV, which actually determines the iodine content in the studied samples, is in good agreement with the experimental data. The obtained results make it possible to use the developed model in designing and changing the design of the PPA installation to determine small concentrations of other elements.

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МОДЕЛЮВАННЯ З ВИКОРИСТАННЯМ GEANT4 СИСТЕМИ AXAS-D З РЕЄСТРАЦІЄЮ К-СЕРІЇ ЙОДУ В АКТИВОВАНОМУ ВУГІЛЛІ ДЛЯ ВЕНТИЛЯЦІЙНИХ СИСТЕМ АЕС

О. Захарченко, В. Левенець, О. Лонін, С. Соколов

У матеріалі наведено результати використання програми Geant4 для моделювання роботи та можливостей стенду СКІФ при реєстрації випромінювання К-серії з детектором AXAS-D стабільного йоду в активованому вугіллі, який використовується для очищення повітря на АЕС. Показано спектри випромінювання, які отримані у різних ситуаціях: на вході і виході з вимірювальної установки, та їх обробка. Отримано параметри системи реєстрації – ефективність і залежність від товщини кристала детектора.