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Application of the Monte Carlo Method for Spectrometer Calibration to Determine the Surface Activity of Radionuclides Deposited on the Ground

F. V. Finkel[a], V. A. Rebyakova[a, b]*, and D. O. Spirin[b]

[a]*Emergency Response Center, State Atomic Energy Corporation Rosatom, St. Petersburg, 194292 Russia*
[b]*Peter the Great St. Petersburg Polytechnic University, Politekhnicheskaya ul. 29, St. Petersburg, 195251 Russia*
**e-mail: victoria.rebyakova@gmail.com*

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Abstract—The results of efficiency calibration and verification of the in situ method with application of a handheld spectrometer with high-purity germanium detector are presented. The conducted studies show that the calibration of spectrometer efficiency with the use of the Monte Carlo method can be applied for measurement of the surface activity of radionuclides deposited on the ground with an uncertainty of not more than 22%.

Keywords: in situ gamma spectrometry, radionuclides deposited on the ground, Monte Carlo method, detector efficiency

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INTRODUCTION

One of the key objectives in responding to radiation emergencies is to determine the radionuclide composition of contaminated soil and the surface activity of deposited radionuclides for comparison with the values of the corresponding intervention levels. This is necessary for timely assessment of the radiological situation and forming recommendations for appropriate protective measures [1].

The sampling method with subsequent sample preparation and spectrometric analysis in analytical laboratories is traditional to examine the areas contaminated by radionuclides.

However, for challenges of rapid response to emergencies with the radiation factor, the use of gamma spectrometry in situ is more rational.

There are many works [2–4] where the use of gamma spectrometry in situ makes it possible to survey contaminated areas.

The aim of this work was to perform the efficiency calibration and testing of the in situ method by the example of a portable semiconductor spectrometer with detector made of high-purity germanium (HPGe).

The geometry is the following: the end cap of the semiconductor detector is oriented down and located at an altitude of 1 m from the surface of soil [1]. In reality, the spectrum obtained at such a geometry contains information on radioactive contamination in an area on the order of a hundred square meters (~100 m^2) and several tens of centimeters in depth (~30 cm). This arrangement of the detector allows one to average the local nonuniformity of the distribution of radionuclides along the surface of the ground.

The spectrometer used for the in situ measurements must be precalibrated; this allows one to measure both the surface activity of recent fallout with preliminarily unknown composition of radionuclides and the activity of old fallout with allowance for their migration into the soil.

EXPERIMENTAL

In recent years, the level of metrological requirements on calibration of equipment detecting radiation of different nature has increased significantly. If for some reason the experimental calibration methods cannot fully meet these requirements, the metrologists have to apply the calculation techniques for calibrating such systems, in particular, to determine the efficiency of gamma-ray scintillation or semiconductor detectors. Currently, in all leading national metrological centers, to calculate the response function and determine the efficiency of registration of detectors, the Monte Carlo method (MCM) based on random modeling of trajectories and subsequent tracing of the fate of each quantum or particle before its full absorption is used. To implement MCM on personal computers, different software was developed (GEANT4 and others) [5]. The initial data for MCM are the experimental geometry, the characteristics of construction materials of detectors, and the nature of radiation. All programs use in the calculations the nuclear databases for cross sections of processes and nuclear decay schemes. All listed software has been repeatedly tested and proven in experiments. The accuracy of detector calibration and detection efficiency of gamma rays using MCM, according to the estimates, is currently competing with calibration accuracy obtained experimentally.

The MCC MT program developed at Peter the Great St. Petersburg Polytechnic University [6] for modeling the processes of transport and registration of ionizing radiation takes into account the cascade summing of gamma quanta [6]. It represents a three-dimensional computer code and allows one to calculate the response function for the most widely used detectors irradiated by various sources of photons or electrons (positrons) in the energy range from 1 keV to 10 MeV.

The MCC MT program makes it possible to considerably simplify the solution of practical problems in the development, optimization, and calibration of detection systems of ionizing radiation.

The main feature of the software is the combination of accuracy of the results with ease of operation and wide availability in solving practical problems. The use of this software makes it possible to optimize and improve the parameters of various radiation detection devices used in the nuclear industry and the systems of radiative environment monitoring. One of the important features of the MCC MT is the developed user interface that allows an operator who is familiar with the work of the standard Windows environment to create complex three-dimensional geometric objects. The operator can set the properties of relevant materials for objects, identify them, specify the properties of detectors, set the desired exposure, and connect various sources of radiation with the possibility of taking into account cascade processes. The calculation data are displayed in the form of an energy spectrum absorbed by the given detector and with the help of special software modules can be easily converted into a detector response function taking into account its energy resolution.

It should be noted that, in conducting gamma-spectrometric measurements in situ, the experimental calibration techniques may meet with various difficulties: large volumes of emitting substance, specific or short-lived radiation sources, uneven distribution of radionuclides across the thickness of bedding, etc. Because of this, the modeling with the help of MCM was used to determine the efficiency of a detector, namely, the MCC MT program.

We used a GR3019 HPGe semiconductor spectrometer of coaxial type (Canberra Industries). The detector efficiency with respect to the efficiency of a NaI(Tl)-based crystalline detector with a size of 3″ × 3″ is 30%. The energy resolution of a semiconductor detector at radiation energy of 661.7 keV is 0.2%. The accumulation of the spectrum and measurement of the count rate was performed with the help of the EcoGamma program developed at the Emergency Response Center of Rosatom (ERC), St. Petersburg.

RESULTS

The model of a HPGe detector was created, and the MCM spectra of point sources were calculated, as well as the spectra of surface and bulk distributions of radionuclides. Moreover, the corresponding efficiency of detection depending on the energy of gamma quanta was plotted. The adequacy of the model was tested in the laboratory using reference point sources (RPS), reference volumetric sources (RVS), and samples of soil collected in contaminated areas.

The RPS and RVS spectra of point sources with ^{137}Cs, ^{60}Co, and ^{152}Eu radionuclides were measured at a distance of 10 cm along the axis from the end face of the detector. The activity of the ^{137}Cs radionuclide source was 4830 Bq, $U_{\mathrm{A}} = 1.7\%$ on July 23, 2014. The activity of the ^{60}Co radionuclide source was 5472 Bq, $U_{\mathrm{A}} = 1.8\%$ on the same date.

Figure 1 shows the model of the GR3019 detector created using MCC MT. The spectra of the point sources of the ^{137}Cs and ^{60}Co radionuclides at a distance of 10 cm along the axis from the end cap of the detector unit were calculated using this model.

Table 1 shows the results of experimental and theoretical (calculated) calibration efficiencies of RPS located a distance of 10 cm along the axis from the end cap of the detector. The expression for the assessment of experimental and calculated efficiency of registration has the form [7]

$$\varepsilon_{\mathrm{exp}}(E) = \frac{N(E)}{AI_\gamma}, \tag{1}$$

where $N(E)$ is the count rate of pulses registered in the photopeak of energy E with the deduction of the background signal, count/s; A is the activity of the source on the date of measurement, Bq; and I_γ is the quantum yield of photons with energy E.

Thus, the energy dependence of the detection efficiency at the peaks of total absorption of gamma radiation of the point source with RPS geometry located at a distance of 10 cm along the axis from the end surface of the GR3019 HP Ge detector can be obtained on the basis of the calculated spectrum in the energy range of gamma quanta in which we are interested. In this work, the estimated efficiency values are obtained in the interval from 20 to 1450 keV.

When simulating the spectra of surface contamination, we used the RVS spectra with the ^{152}Eu + ^{154}Eu radionuclides made in the form of laminated plates with dimensions of 10 × 10 cm, onto the surface of which a radionuclide solution was uniformly deposited. Each plate had an activity of 7300 Bq on September 5, 2014, $U_A = 10\%$ ($k = 2$).

For obtaining the modeling spectra using MCC MT, two types of models were used under condition that the radionuclide composition was unknown beforehand: recent surface fallout and old fallout with radionuclide migration into the soil.

Table 2 shows the calculation results for recent surface contamination with the ^{152}Eu radionuclide on an area of 186 × 258 cm. The difference between experimental and calculated efficiency for different energies varies from 1% (the ^{152}Eu line at 444 keV) to 22% (the line 411.1 keV). This is caused by simplification of the model, poor statistics, the errors of RVS calibration, and the presence of impurities of the ^{154}Eu radionuclide in RVS.

In [1], the detection efficiency at the peaks of total absorption of gamma quanta depending on the energy was calculated. It corresponds to gamma quanta with energies of 50–1450 keV and geometry of recent contamination on a surface area of 100 m^2:

$$\ln \varepsilon = \sum_{i=0}^{6} \left(A_i (\ln E)^i \right), \tag{2}$$

where ε is the efficiency of registration; E is the energy of gamma quantum in keV; and A_i are the coefficients of the polynomial: $A_0 = 4.553$, $A_1 = -26.114$, $A_2 = 15.037$, $A_3 = -4.126$, $A_4 = 0.601$, $A_5 = -0.045$, $A_6 = 0.001$.

The detection efficiency at the peaks of total absorption of gamma radiation depending on the energy was calculated for the geometry of old surface contamination and gamma quanta with energies of 50–1450 keV under the condition of uniform distribution of radionuclides across the bedding thickness of 20 cm on the surface area of 100 m^2. The soil was modeled as quartz sand with a density of 1.7 g/cm^3.

The energy dependence of the detection efficiency can be expressed by formula (2) with the polynomial coefficients $A_0 = -312.183$, $A_1 = 318.264$, $A_2 = -143.207$, $A_3 = 34.761$, $A_4 = -4.774$, $A_5 = 0.350$, $A_6 = -0.011$.

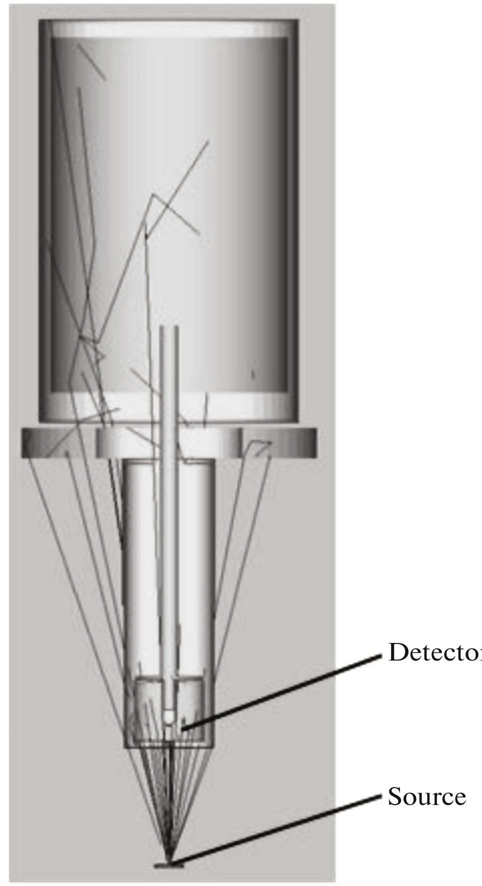

Fig. 1. Depiction of GR3019 high-purity germanium detector and point source.

Samples of soil in radioactively contaminated areas were selected on the islands of the northwestern archipelago of Lake Ladoga within the work carried out at ERC [8].

To assess the uncertainty of the in situ method under conditions of long-time surface contamination related to nonuniformity of radionuclide distribution over the surface, the data on the activity distribution across the depth collected from the soil in contaminated areas were used.

Table 3 shows the activity distribution over depth in the samples of soil extracted. Three representative soil sampling points were located on an area less than 100 m^2.

Table 1. Results of experimental and computed efficiency calibration for RPS

Radionuclide	Energy, keV	Efficiency × 10^{-4} (experiment, RPS), count (s Bq)$^{-1}$	Efficiency × 10^{-4} (calculation), count (s Bq)$^{-1}$	Deviation, %
^{137}Cs	661.7	30.6	31.8	4
^{60}Co	1173.2	19.3	21.1	9
	1332.5	17.1	18.8	10

Table 2. Calculation results for recent surface contamination with the ^{152}Eu radionuclide on an area of 186 × 258 cm

Radionuclide	Energy, keV	Efficiency × 10^{-4} (experiment, RPS), count $(s\ Bq)^{-1}$	Efficiency × 10^{-4} (calculation with consideration of cascade summation), count $(s\ Bq)^{-1}$	Deviation, %
^{152}Eu	121.78	5.805	6.773	17
	244.7	3.573	4.158	16
	344.3	2.723	3.120	15
	367.8	2.189	2.463	13
	411.1	2.376	2.902	22
	444.0	2.439	2.433	1
	778.9	1.341	1.384	3
	867.4	1.291	1.453	13
	964.1	1.140	1.131	1
	1085.9	1.002	1.061	6
	1112.1	1.014	1.126	11
	1408.0	0.826	0.862	4

The specific activities of the ^{137}Cs radionuclide were determined with greater uncertainty but less than 30%.

In carrying out gamma-spectrometric measurements in situ when the detector is located at a height of 1 m above the ground, averaging of the local nonuniformity of distribution of radionuclides on the surface of the ground occurs. In our case, the unevenness of the activity distribution was determined in three samples of soil.

The calculated spectra of gamma quanta with energies of 50–1450 keV were obtained for three layers of soil on a surface area of 100 m^2. The soil was modeled

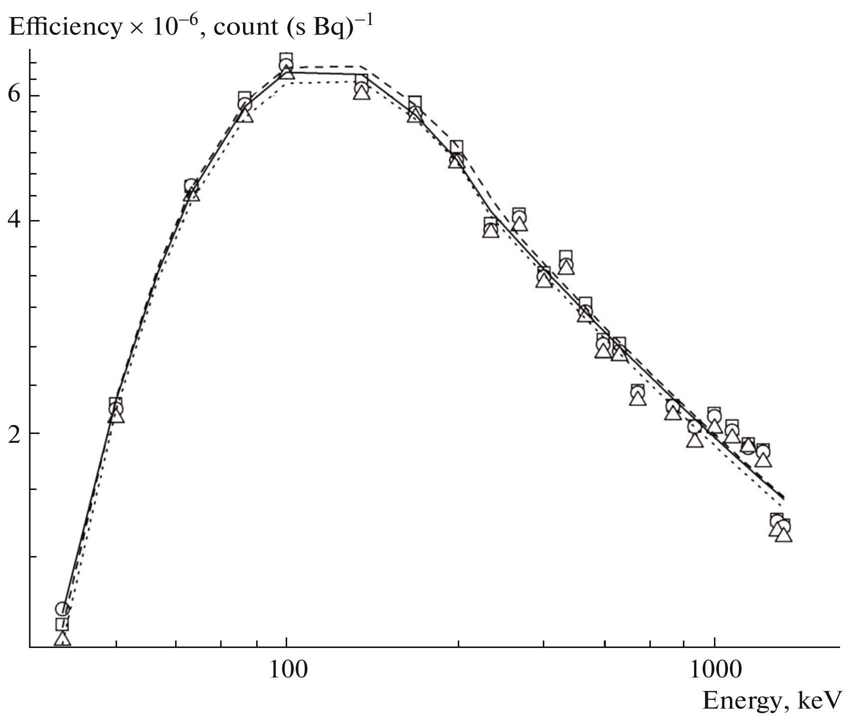

Fig. 2. Efficiency of registration of gamma quanta depending on the energy for the geometry of old surface contamination on an area of 100 m^2 in the case of nonuniform distribution of radionuclides across the thickness of bedding at three points. The circles, triangles, and squares correspond to points 1 to 3.

Table 3. Activity distribution over the depth in extracted samples of soil

Point no.	Location depth, cm	A_{sp} (^{137}Cs), Bq kg^{-1}	Proportion (surface activity of the upper layer is taken as 100)
1	0–5	514.5	100
	5–10	16.2	3.1
	10–15	<0.1	0
2	0–5	444.6	100
	5–10	19.5	4.4
	10–15	4.5	1
3	0–5	181.8	100
	5–10	2.7	1.5
	10–15	10.2	5.6

as quartz sand with a density of 1.7 g/cm^3. The activity of the conditional source (one layer in this case) is 1000 Bq. The probability for the yield of gamma quanta per one decay of the conditional source is 100%.

Figure 2 shows the efficiency of registration of gamma quanta at the peaks of total absorption depending on the energy for the geometry of old surface contamination for gamma quanta with energies of 50–1450 keV under the condition of nonuniform distribution of radionuclides over the depth on the surface area of 100 m^2 (in correspondence to Table 3).

As one can see from Fig. 2, the curves of efficiency differ one from another by no more than 10% in the entire range of energies from 50 to 1500 keV. This fact can be put into the basis of the conclusion that the distinction of a nonuniformity of distribution of radionuclides over the depth from point to point for a single measuring site gives a 10% contribution to the uncertainty of the measurement results.

CONCLUSIONS

We created the model of a detector and calculated the spectra for point sources, as well as for the surface and bulk distributions of radionuclides. The adequacy of the model was tested in the laboratory using RPS and RVS and the samples of soil collected in radioactively contaminated areas.

The conducted studies showed that, using MCC MT, the calibration of spectrometer efficiency upon the registration of gamma quanta with energies from 50 to 1500 keV can be successfully used in the measurements of radionuclide activity under the conditions of natural occurrence with an uncertainty of not more than 22%.

One may also conclude that the use of software based on the Monte Carlo method allows one to reduce the expenses for acquiring the measures of activity. This will make it possible to perform the calibration of various spectrometric facilities of measurements (for example, in no more than an hour in the geometry of a point source), which is important for challenges assigned to ERC.

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