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Application of Monte Carlo Calculations to Gamma-Spectrometric Measurements of Large Volume Samples

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Abstract: In this paper a calibration method for gamma spectrometric measurements based on computer simulations with GEANT is presented and validated. Proposed calibration method, using self-absorption factors, is especially suitable for measurement of large-volume samples. As demonstrated in the paper, accuracy of proposed method is comparable to other frequently used methods, but it is more effective and less expensive then other available methods.

The current conception of radiation control of discharges and environment of Nuclear power plant (NPP) is based on quantitative measurements of individual radionuclides activity of radioactive discharges from NPP and in samples from the surrounding environment. The semiconductor gamma-spectrometric methods are the basic ones in the determination of radionuclide activities.

The basic condition of the measuring radioactive materials activities with gamma-spectrometric methods is the known peak detection efficiency of used spectrometer. Its value depends on photons energy, spectrometric device, detector-source distance (measurement geometry), form of source (sample geometry) and on absorption of photons by insensitive materials between the source and sensitive detector volume.

Methods used to determine peak detection efficiency can be divided into three basic classes:

a) experimental methods
b) semi-empirical methods
c) Monte Carlo simulation methods.

Quickly developing Monte Carlo methods are based on computer simulation of photons interactions with surrounding materials. Exact knowledge of the composition and experimental setup geometric parameters, beginning with internal parts of detector (sensitive volume dimensions, dead layer thickness etc.), is very important for reliability of calculations and results precision. In the case of good knowledge of necessary parameters the precision of simulated values is abreast of 10 % [1 3].

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1. Experimental measurements

It is possible to resolve experimental measurements into two individual classes:

1. Some measurements of radioactive etalons with cascade emitter contents. Purpose of these measurements was to evaluate precision of Monte Carlo methods for a determination of coincidence-summing correction factors.

2. Some measurements of prepared etalons with various density of material and with contents mostly mono-energy emitters. Results of these measurements led to specification of a calibration method based on application of self-absorption factors. This method eliminates effect of inaccuracies of detector internal parts dimensions to efficiency values precision.

1. Experimental measurements from the first class were accomplished with two liquid nitrogen cooled HPGe detectors with relative efficiency 65 % - detector 1 and 36 % - detector 4. Description of the detectors and their other parameters are listed in Table 1. The parameters of detectors internal parts were determined from cryostat manufacturing drawings.

Detectors were placed in the individual lead shielding with the wall thickness 10 cm and a copper layer from the inside. Spectrometric setup consisted also of computer controlled electronic modules Canberra. The data were transmitted through the Ethernet network to PC. Two etalons made from silicon rubber in plastic cylinder bin with diameter 9 cm and volume 500 ml were used. The first etalon included isotope ^{152}Eu with activity 3.9 kBq. Spectrum was measured by detector 1. The total time of measurement was 1000 seconds. The second etalon included isotope ^{226}Ra with activity 3.3 kBq. The spectrum was measured by detector 4. The time of measurement was 2500 seconds. Etalons were placed on the top of the cryostats in axis of detectors. Having finished the measurements, the background was subtracted from both spectra. The measured spectra were divided into 4000 channels with energy calibration about 0.5 keV/channel.

2. Experimental measurements from the second group were accomplished on five HPGe detectors, whose description and parameters are listed in Table 1. Producers did not provide manufacturing drawings of detectors 2, 3, 5, so experimental setups of these detectors were defined from data sheets. A liquid mixture etalon was used for preparation of measuring standards. Etalon contained 11 mostly mono-energy radionuclides (^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{203}Hg, ^{54}Mn, ^{113}Sn, ^{85}Sr, ^{88}Y). The etalon was diluted and quartered to fractions with the same activities. One fraction was used for preparation of liquid standard with density 1 g.cm ^{3} (HCl acidified water), other three fractions were used for preparation of standards from materials with various composition and density – silicon sand with density 1.6 g.cm ^{3}, wooden sawdust with density 0.13 g.cm ^{3}, iron sawdust of density 1.35 g.cm ^{3}. Standards preparation consisted of uniform instillation of diluted liquid etalon to cleared material, drying up followed by multiple homogenizing. Standards were prepared with volume 3 liters. Besides these standards other one was prepared. Leaden balls with diameter 5 mm were mixed with liquid etalon containing isotopes ^{60}Co and ^{137}Cs. Its volume was 1000 ml and the final density was 7.0 g.cm ^{3}.

Prepared standards were measured in 11 various geometries – cylinder geometry in glass bins with volume 250 ml, 500 ml, 1000 ml, 2000 ml, 3000 ml, ring and Marinelli geometry in plastic Marinelli bins with volume 500 ml and 1000 ml and in iron Marinelli bin with volume 3000 ml. The leaden balls were measured only in geometries with volume 500 ml and 1000 ml. Totally 218 prepared etalons spectra were measured. The count rates

of photo-peaks were determined after the background subtract and then peak efficiencies values were calculated.

Table 1. Detectors parameters.

PARAMETER	Det. 1	Det. 2	Det. 3	Det. 4	Det. 5
Detector type	GC6021	EGPC20	GR1520	GC3520	GC1018
Serial number	b 00148	5430	B 7528	b 00164	P175
Kryostat	7500SL	SHF00	7500SL	7500SL	7500SL
Preamplifier	2002CSL	PSC 821	2002CSL	2002CSL	2002CSL
FWHM/FWTM (1332.5 keV)	1.99/3.84 keV	2.10/3.84 keV	1.90/3.61 keV	1.82/3.39 keV	1.70/3.11 keV
FWHM (122 keV)	0.957 keV	1.2 keV	0.857 keV	0.902 keV	0.803 keV
Peak/Compton (1332.5 keV)	72.5:1	44.0:1	43.0:1	66.1:1	41.1:1
Rel. efficiency (1332.5 keV)	65.4 %	20.0 %	19.2 %	36.0 %	8.6 %
Crystal Volume	255 cm^3	107 cm^3	98 cm^3	147 cm^3	63 cm^3

2. Monte Carlo simulation methods

Computer code GEANT developed in CERN was used for computer simulations of detection efficiency of HPGe detectors. GEANT includes subroutines enabling to make the FORTRAN programs for simulation of various physical processes of interaction and transition of particles through the mass medium [4]. Program GEANT enables:

to construct a model of the experimental setup as a composition of geometrical shapes with the given parameters and a visual control of created model
to generate simulated particles by standard Monte Carlo generator
to control transport of particles through the individual parts of the setup and record all the interactions
to register and interactive to visualize particle trajectories and absorbed energies.

A program compiled by code GEANT contained a particle generator allowing simulation of individual photons or cascade of photons. Photons and electrons had set the same minimal energy threshold 10 keV. All experimental spectra were supplemented with computer-simulated spectra with photons energies emitted by radionuclides included in the standards. Complete photon cascades of given radionuclides for the cascade gamma-emitters were simulated in each step according to literature [5, 6].

Imperfect charge collection in sensitive detector volume and other electronic pulse processing fluctuations of experimental setup result in Gauss normal distribution of impulses in real spectrum. This effect was allowed before including the impulse into the simulated spectrum. In the computer simulations this problem was solved in such a way that the initial energy value E was substituted by the new value E', which was determined on the basis of Gauss distribution probability density (1, 2) assigned from real resolution values FWHM of all the used semiconductor detectors.

$$f(E,E,) \quad \frac{1}{(2 \quad ^2)} \exp \quad \frac{(E \quad E)^2}{2 \quad ^2} \tag{1}$$

$$\frac{FWHM}{2 \quad 1.18} \tag{2}$$

3. Results and discussion

Cascade radionuclides spectra simulations

Two spectra of cascade emitters ^{152}Eu a ^{226}Ra were measured and computer simulated. The number of simulated disintegrations was ten times larger than the number of measured disintegrations. Therefore after the simulation completing, the count rate in each channel was divided by ten. The comparison of these experimental and simulated spectra is shown in Fig. 1 and Fig. 2. Photo-peak count rates of included radionuclides are shown in Table. 2.

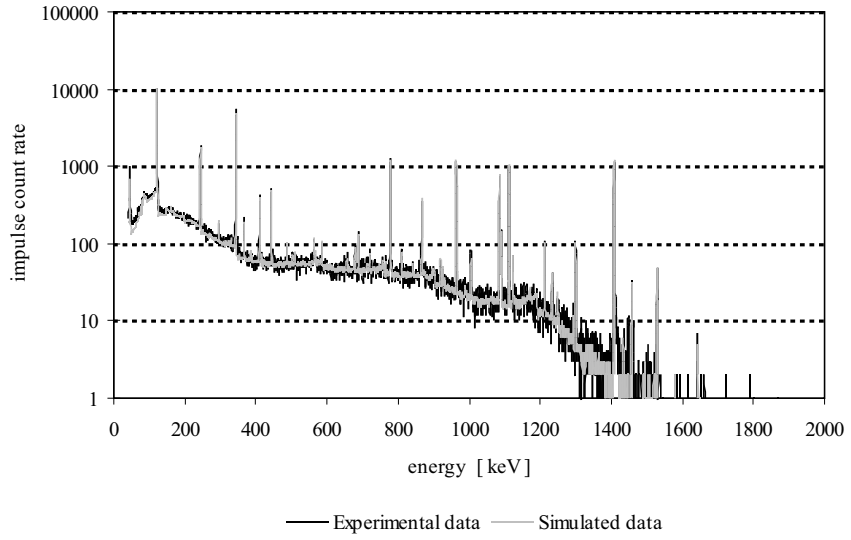

Fig. 1. Nuclide ^{152}Eu, silicone rubber in plastic cylinder bin Vol. = 500 ml, detector 1.

The results of the comparison of the presented data:
1. Compton count rate in the area from 0 to 50 keV of simulated spectra is less than in experimental ones. This fact is caused by the existence of significant X-ray lines with energy less then 50 keV in the decay scheme of included radionuclides, which were not added to simulations, and also by small inaccuracies in the thickness of individual materials between the measured sample and sensitive detector volume.

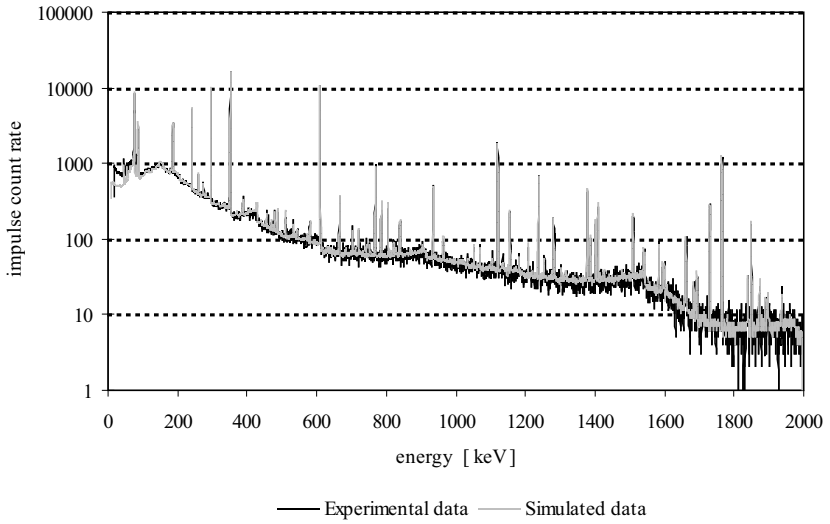

Fig. 2. Nuclide ^{226}Ra, silicone rubber in plastic cylinder bin Vol. = 500 ml, detector 4.

2. From the comparison of total impulse count rate in spectra follows that distinction between experimental and simulated total count rate is less than 5 % for energy interval over 50 keV.
3. Photo-peak count rate of included radionuclides mostly agrees in the interval of 5 %. Distinction of count rate is not over 10 % in any case, which is comparable with the precision of other frequently used methods [7, 8].

A very good agreement of experimental and simulated data indicates that exact knowledge of detector internal parameters enables to eliminate simulation results inaccuracies. The limiting factor of simulations exact germanium dead layer determination and Monte Carlo code precision in the low photon energy area still remains.

Simulations of prepared etalons with different composition (self-absorption factors)

Monte Carlo methods efficiency is subject to good knowledge of experimental setup geometry parameters including cryostat internal parts. Determination of all the needed parameters is uncertain especially at old detectors. In these cases a method based on simple experimental measurements and simulations of self-absorption corrections fs [9] will be effective mainly in the case of measurements of large volume samples with various composition. Self-absorption factor represents photon rate attenuation during sample crossing and it is possible to define it as an attenuation of sample with effective thickness X_{ef} (3).

$$f_s \quad \exp(\quad X_{ef}) \tag{3}$$

It is necessary to determine the effective sample thickness X_{ef} and all variable parameters relations for successful use of self-absorption factors for all required geometries. These parameters are detector type and dimensions, sample material composition and

Table 2. Photo-peaks count rates.

Nuclide	Energy [keV]	Experimental data	Simulated data	Difference [ ]
	121.78	24099 316	24547	1.86
	244.70	4596 145	4967	8.07
	344.28	15325 247	15104	1.44
	411.12	1064 73	1104	3.76
	443.98	1445 82	1337	7.47
Eu-152	778.90	4236 131	4106	3.07
	867.39	1232 75	1227	0.41
	964.13	4184 129	4435	6.00
	1085.91	2912 111	2807	3.61
	1112.12	3577 121	3572	0.14
	1408.01	4919 134	5112	3.92
	74.82	7989 221	7885	1.30
	77.11	13697 268	13536	1.18
	87.30	5307 188	5483	3.32
	89.80	2096 142	2022	3.53
	186.21	5968 194	5834	2.25
	241.98	10157 223	10862	6.94
	295.21	22734 313	23827	4.81
	351.92	38364 398	39766	3.65
	609.31	29866 347	30660	2.66
Ra-226	665.45	857 72	853	
	768.36	2606 112	2548	2.23
	934.06	1448 87	1466	1.24
	1120.30	6366 163	6324	0.66
	1155.20	682 62	687	0.73
	1238.10	2375 103	2324	2.15
	1377.70	1628 88	1597	1.90
	1408.00	873 65	936	7.22
	1729.60	1057 67	1108	4.82
	1764.50	5053 144	4994	1.17

density, photons energies. Comparison of X_{ef} values for 5 detectors, 11 geometries, 5 various compositions and densities, and 13 photons energies indicated that X_{ef} values are independent on detector type and dimensions. The next step was a substitution X_{ef} dependence of photon energy and sample composition by the dependence of linear attenuation coefficient. The X_{ef} ( ) curves for all geometries were approximated by secondary logarithmic polynomial fit with least square method (4). The coefficients a_0, a_1, a_2 varied for different geometries. The X_{ef} ( ) curves for some geometries are shown in Fig. 3.

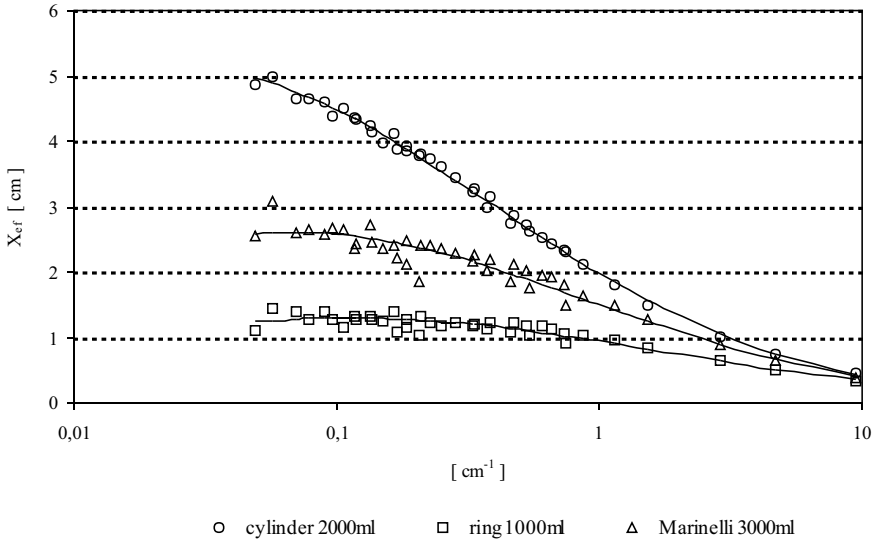

Fig. 3. The X_{ef} ( ) curve for some selected geometries.

$$\ln(X_{ef}) \quad a_0 \quad a_1 \ \ln(\ ) \quad a_2(\ln(\ ))^2 \tag{4}$$

This large-volume geometries calibration method using self-absorption factors was defined on the base of achieved results:

1. experimental measurement of one known sample composition detection efficiency in required geometry (e.g. liquid sample)
2. values determination of required photons energies based on sample density and approximate composition
3. self-absorption factors determination from X_{ef} ( ) curve and detection efficiencies values calculation for analyzed sample.

More than 2000 experimental measured and calculated detection efficiencies were compared for validation of the described calibration method. The compared values were determined for photon energies from 60 keV to 1836 keV and for various composition samples with density from 0.13 g cm ^{3} to 7.0 g cm ^{3} measured on 5 HPGe detectors in 11 various geometries. The liquid etalon represented the known sample from the first step.

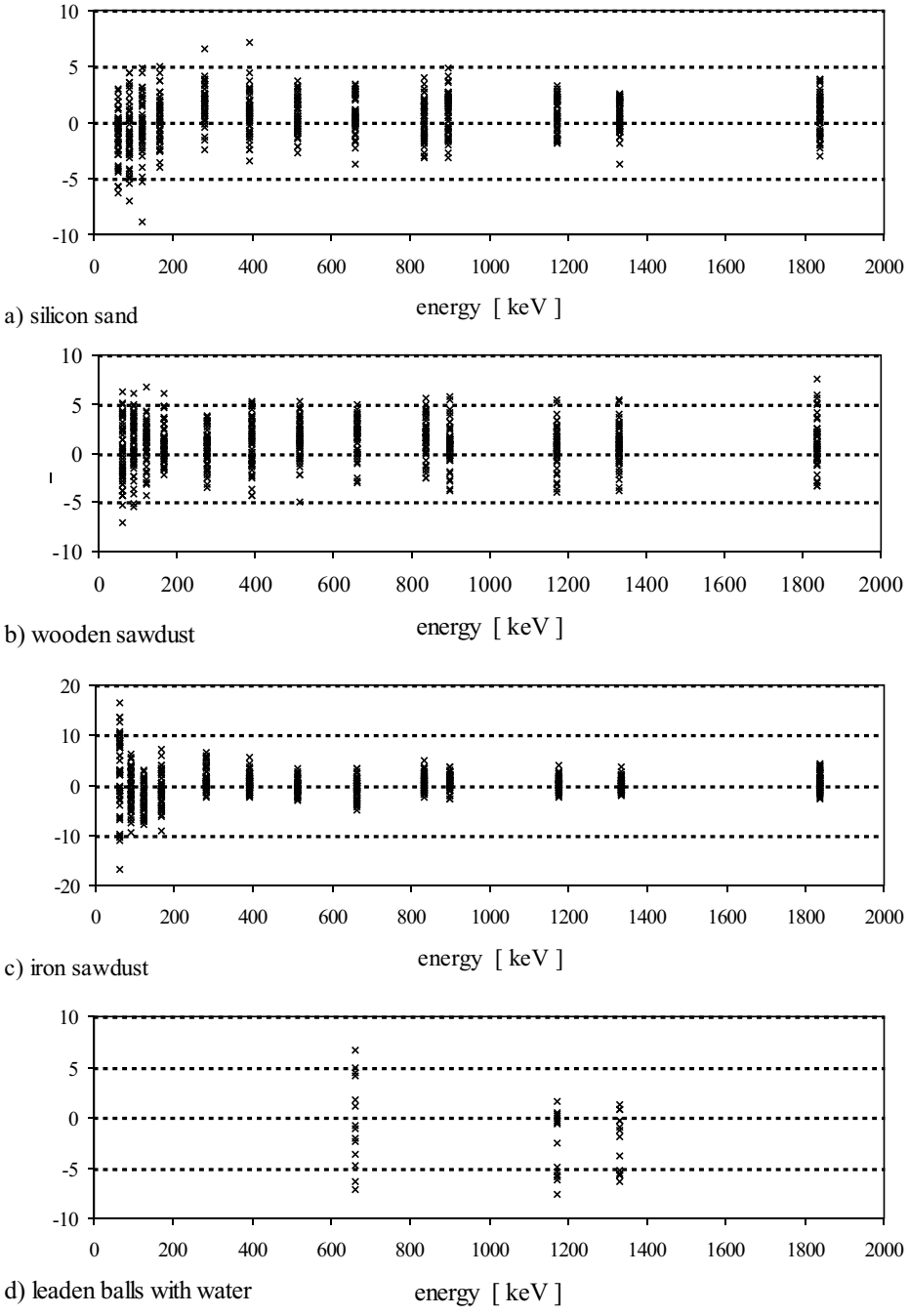

Fig. 4. The comparison of the measured and calculated detection efficiencies.

Efficiencies for other etalons – silicon sand, wooden and iron sawdust and leaden balls with water – were calculated by self-absorption factors.

The comparison of experimental measured (e_{exp}) and calculated (e_{cal}) efficiencies is shown in Fig. 4. The difference was calculated according to formula (5).

$$\frac{(\quad cal \quad \exp)}{\exp} \quad 100 \tag{5}$$

The calculated results are in a very good agreement with the experimental data to within about 5 %, in some cases to within about 10 % for some geometries in the energy region under 100 keV. The greater differences in the low energy range were probably caused by use of one X_{ef}( ) curve for all photons energies and sample compositions. But X_{ef} values can slightly vary for different energies and samples. These differences occur mostly in the case of large volume geometry and great sample material value (low photon energy range and high density samples). The combination of these two conditions occurred in the case of iron sawdust and photon energy 60 keV, where the maximum differences between $_{exp}$ and $_{cal}$ were registered.

In spite of the all above mentioned limitations, precision of the described method highly exceeds NPP radiation safety terms. Advantages of this method are the universal application and cheapness in comparison with other used methods.

4. Conclusions

The comparison of the achieved results indicates a very good agreement of experimental and simulated data. Advantage of Monte Carlo method is the possibility of fast and accurate determination of self-absorption and coincidence summation correction factors. At present computer simulation is the most effective method of semiconductor detectors calibration for gamma-spectrometric measurements of large volume samples with various geometry and composition.

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