

EMPIRICAL FORMULA FOR THE DEPENDENCE OF HPGe-DETECTOR EFFICIENCY ON ENERGY AND DISTANCE FOR SHIELDED GAMMA-RAY SOURCES

*I.V. Pylypchynets, O.I. Lengyel, O.O. Parlag, E.V. Oleynikov
Institute of Electronic Physics NAS of Ukraine, Uzhhorod, Ukraine
E-mail: parlag.oleg@gmail.com*

The accuracy of gamma-spectrometric measurements in the isotopic analysis of shielded nuclear materials depends on the accuracy of the detector calibration in terms of energy efficiency, which should take into account corrections related to the measurement geometry and the absorption of γ -radiation by the shield material. The results of experimental studies of the energy efficiency of the HPGe-detector measured at fixed distances between the calibration source and the detector (50 and 100 mm) in the presence and absence of an absorbing shield made of stainless steel 12X18H10T (thickness – 9.6 mm) are presented. On the basis of experimental data, an empirical description of the efficiency dependence for fixed distances between the gamma radiation source and the detector in the presence of a stainless steel absorbing shield was obtained.

PACS: 24.75.+1, 25.85.-w, 25.85.Ec, 25.85. Ca

INTRODUCTION

Knowledge of the qualitative (isotopic) and quantitative characteristics of fertile and fissile nuclear materials is necessary to solve a number of applied problems in nuclear engineering (for example, the issues of their safe use, control and non-proliferation [1]). As a rule, such nuclear materials are stored in sealed containers. One of the main methods for analyzing the isotopic and quantitative composition of shielded nuclear materials is based on gamma-ray spectrometric measurements of their characteristic [2] or stimulated gamma radiation [3]. In addition, during the analysis, there is a need for gamma-spectrometric measurements of high-intensity γ -radiation fluxes from high-level nuclear materials. Therefore, there is a need to attenuate the intensity of γ -radiation to reduce miscalculations associated with detector loading (its dead time) and, accordingly, corrections for cascade summation [4].

To attenuate the intensity of γ -radiation during spectrometric measurements, the distance between the sample and the detector working surface is reduced (this leads to a weakening of the effect of cascade summation of γ -quanta due to a decrease in the geometric efficiency of the detector [5]), or use filters (absorbing screens) that are inserted between the sample and the working surface of the detector (this also reduces the intensity due to the absorption and scattering of gamma-radiation by the material (or materials) from which the filter is made [6]).

The accuracy of spectrometric measurements depends on the accuracy of the detector calibration for energy efficiency, which should take into account corrections related to the measurement geometry and the absorption of γ -radiation by the shield material.

Experimental studies of the efficiency of measurements with absorbing screens are rather limited [7-9]. When performing theoretical calculations of the efficiency-energy dependence for specific types of detectors and measurement schemes, difficulties arise due to the uncertainty of the detector dead layer thickness, which can lead to inaccurate final simulation results [10].

The aim of the present work is to obtain an empirical description of the dependence of the efficiency of the HPGe detector for fixed distances between the gamma radiation source and the detector in the presence of a stainless steel absorbing shield.

To accomplish this task, the following were done:

- experimental studies were conducted to determine the energy efficiency at fixed distances: γ -radiation source – detector in the presence and absence of an absorbing screen;

- refinement of the previously obtained empirical formula for the dependence of the energy efficiency of the detector on the distance [5] by adding parameters that take into account the presence of an absorbing screen between the γ -ray sources and the detector.

1. THEORETICAL BACKGROUND

The absolute efficiency of the detector is determined by measuring γ -radiation from standard sources containing radionuclides with known activity. The total absolute (peak) efficiency is defined as

$$\varepsilon(E_\gamma) = \frac{N_\gamma}{N_S}, \quad (1)$$

where N_γ – is the number of pulses measured in the peak areas from γ -ray sources with used energies E_γ [11].

The number of photons N_S during the measurement time t from a radionuclide with known activity A is given by the relation:

$$N_S = A I_\gamma t, \quad (2)$$

where I_γ – is the intensity of the γ -line with energy E_γ .

Then

$$\varepsilon(E_\gamma) = \frac{N_\gamma}{A I_\gamma t}. \quad (3)$$

If an absorbing screen is placed between the detector and the γ -ray source (e.g., to absorb X-rays to reduce the count rate in γ -peaks (or detector dead time) and in some cases to eliminate summing corrections), the absorption process must be taken into account.

For an absorbing screen of thickness x , the basic Beer-Lambert law provides a simplified correction for absorption, since this law is valid only for the “good

geometrical arrangement” (for a narrow beam of γ -quanta) of the absorption coefficients measurements [6]:

$$C_{Att}(E_\gamma) = \exp(-\mu(E_\gamma)x) = \exp\left(-\frac{\mu(E_\gamma)}{\rho}\rho x\right), \quad (4)$$

where $\mu(E_\gamma)$ and $\mu(E_\gamma)/\rho$ – are respectively the linear and mass absorption coefficients of the material from which the screen is made; ρ – is the density of the material. These values can be calculated by the codes XCOM [12], EPICOM [13], etc. only for the “good geometrical arrangement”. However, it should be noted that this is true only for monochromatic photons incident at a normal angle on the absorbing layer or for the “good geometrical arrangement” of absorption coefficient measurements.

During the procedure of calibration of detectors by absolute (peak) efficiency, usually, the measurement mode for the “good geometrical arrangement” is not performed (used), i.e. measurements are performed in the “bad geometrical arrangement” mode (for a broad beam of γ -quanta). In this case, γ -radiation passing through the absorbing screen generates two radiation components inside or outside the screen, namely, colliding and non-colliding γ -quanta. Therefore, it is necessary to introduce a correction for the build up factor, which takes into account this process and significantly depends on the energy of γ -radiation and the absorption properties of the material from which the screen is made [14].

As a rule, empirical formulas for describing the absolute efficiency of detectors for cases of different geometric conditions of measurements with and without absorbing screens, obtained from the experimental data sets on these detectors, take into account geometric corrections and corrections related to the transmission of γ -radiation by absorbing screens [15].

2. EXPERIMENTAL RESEARCH

To study (measure) the energy efficiency of the detector at fixed values of the distance from the γ -radiation source to the detector surface in the presence and absence of an absorbing screen, an installation based on a certified semiconductor HPGe detector from ORTEC was used, the characteristics of which are presented in Table 1 [5, 16].

Table 1
Technical characteristics of the HPGe detector by ORTEC

Model.	Gem 40195
Serial No.	27 P1892A
Manufacturer	Ortec, USA
Crystal volume, cm ³	150
Crystal radius, mm	31.4
Distance cover – surface of the detector crystal, mm	3.0
Detector cover thickness, mm	1.0

The efficiency of the detector was determined using 7 certified standard point sources of γ -radiation: ²²Na, ⁵⁷Co, ⁶⁰Co, ¹³³Ba, ¹³⁷Cs, ¹⁵¹Eu, and ²⁴¹Am, which allowed covering a wide energy range from 59.5 to 1408 keV.

The experimental studies were conducted using γ -radiation sources manufactured by the Institute of

Metrology (St. Petersburg, Russia) and Eckert & Ziegler Isotope Products GmbH (California, USA).

The γ -source of ²⁴¹Am was not used in the efficiency measurements with the absorbing screen due to its relatively low activity at the time of the measurements.

A schematic diagram and a photograph of the spectrometry setup are shown in Figs. 1 and 2, respectively.

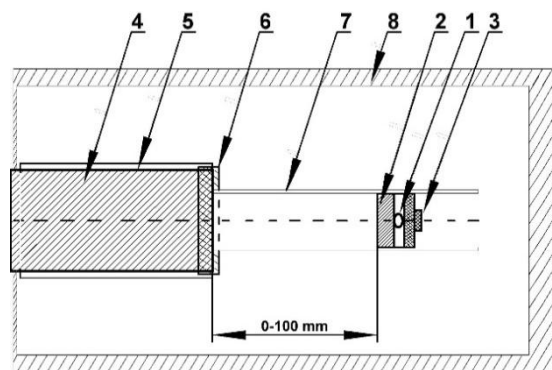


Fig. 1. Calibration scheme of HPGe-detector by absolute efficiency:

- 1 – certified point source of γ -radiation; 2 – absorbing screen made of stainless steel; 3 – fixation of the γ -radiation source; 4 – active volume of the HPGe-detector; 5 – cadmium sheet [0.1 cm]; 6 – tube holder – tube fixation (material - plexiglass); 7 – tube for fixing the position of the point source (material – plexiglass); 8 – passive lead protection (thickness – 100 mm)

A custom holder made of plexiglas was used to measure γ -radiation from sources for two different axial distances in the right direction – 50 and 100 mm from the detector surface in the presence and absence of an absorbing shield. The holder was mounted directly on the detector surface.

The choice of the initial distance between the source and the detector, 50 mm, was due to the need to exclude the effect of summing corrections of registered photons from γ -radiation sources with a complex decay scheme (for example, ⁶⁰Co, ¹⁵²Eu) on the final measurement results [17, 18]. At distances of more than 100 mm, the statistical error of measurements increased due to insufficient activity of the calibration γ -ray sources.



Fig. 2. Photograph of the HPGe-detector with the calibration γ -sources fixation

A disk (diameter – 34 mm, thickness – 9.6 mm) made of stainless steel grade 12X18H10T [19] was used as an absorbing screen. The choice of this material was due to the fact that stainless steel of this brand and its foreign analogues are widely used in nuclear engineering as a structural material in the manufacture of sealed containers for packaging and storage of radioactive substances [2].

The amplitude analysis was performed using a multichannel analyzer from ORTEC (model: N5608X3, serial number 387 1069) connected to a HEWLETT PACKARD computer interface.

The resolution of the spectrometer was – 2.6 keV for the γ -line of ^{60}Co – 1332.5 keV (Fig. 3).

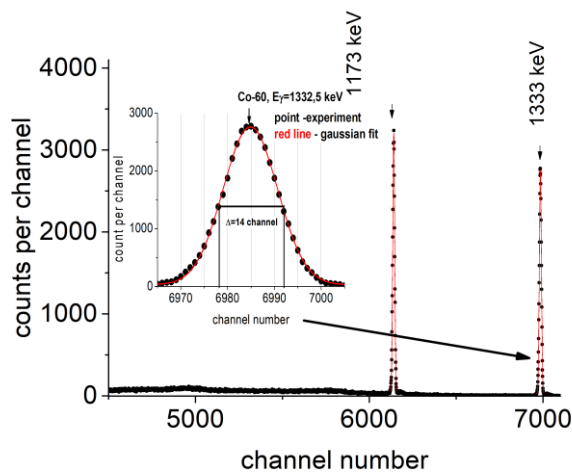


Fig. 3. Fragment of the γ -spectrum of ^{60}Co . Inset – the result of the mathematical description of the experimental points (circles) by the Gaussian function (solid line)

During the measurements, the drift of the energy scale and the spectrometer resolution was constantly monitored using point standard γ -sources ^{57}Co and ^{60}Co . The drift of these parameters did not exceed 1%.

At each fixed position of the γ -sources with and without the absorbing screen, three series of measurements were performed.

The measured γ -ray spectra were processed using the WINSPECTRUM program package [20].

The values of nuclear spectroscopic data from the NNDC library (USA) [21] were used in the efficiency calculations. Their values were: ^{22}Na : 1274.54 (99.94); ^{57}Co : 122.06 (85.6), 136.47 (10.68); ^{60}Co : 1332.49 (99.98); ^{133}Ba : 80.99 (32.9), 276.49 (7.16), 302.85 (18.34), 356.01 (62.05), 383.84 (8.94); ^{137}Cs : 661.66 (85.1); ^{151}Eu : 121.78 (28.53), 778.90 (12.93), 964.06 (14.51), 1112.08 (13.67); ^{241}Am : 59.54 (35.9). The energies of the γ -ray lines are given in keV, with their intensities in percent (%) in parentheses.

Fig. 4 shows the experimental efficiency values averaged over three independent measurements for fixed distances of 50 and 100 mm without an absorbing screen (dark squares and triangles) and with an absorbing screen (light squares and triangles).

The statistical error of the efficiency measurements did not exceed 3...5% for the entire energy range.

3. PARAMETRIC DESCRIPTION OF DETECTOR EFFICIENCY

Experimentally, the efficiency of the detector is determined by measuring standard point γ -sources containing different radionuclides with a certain activity. Given the limited number of experimental points, a relatively small number of parameters should be selected, sufficient for a qualitative description of the efficiency curve.

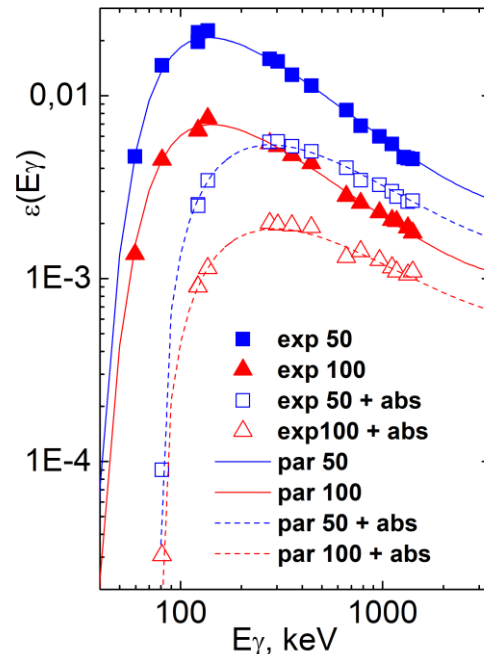


Fig. 4. Detector efficiency at fixed distances point source – detector surface with and without absorbing screen

Therefore, we used an acceptable empirical efficiency formula for fixed distances detector surface - γ -ray source without an absorbing shield (5), obtained in [5], modified by a function with a minimum number of parameters.

The empirical formula for efficiency without an absorbing screen is as follows:

$$\ln \varepsilon(E_\gamma) = -g_0(x) + g_1(x) \ln\left(\frac{E_\gamma}{E_0}\right) - g_2(x) \ln^2\left(\frac{E_\gamma}{E_0}\right) + g_3 \ln^3\left(\frac{E_\gamma}{E_0}\right) - g_4/E_\gamma^\delta. \quad (5)$$

The coefficients reflect the dependence on the distance between the calibration point γ -source and the detector:

$$g_1(x) = a_1 + b_1 \ln x; \quad (6)$$

$$g_0(x) = a_0 + b_0 \sqrt{x}; \quad (7)$$

$$g_2(x) = a_2 + b_2 x. \quad (8)$$

The presented formula well describes the experimental numerical values of efficiency (see Fig. 4, solid curves) measured at fixed distances ($x = 50$ and 100 mm) without an absorbing screen for a given energy calibration range.

The best parameters for formulas (6) through (8) are presented in Table 2.

Table 2
The most fitting efficiency parameters

Parameter	Value
a ₀	10.40
b ₀	1.398
a ₁	5.459
b ₁	0.04589
a ₂	0.962
b ₂	-0.455E-3
g ₃	0.04845
g ₄	3.8E5
δ	3.0

To find the efficiency of the detector in the presence of an absorbing screen, we will process the parameterization (5) with fixed values of the parameters from Table 2.

Given the limited number of experimental points, a relatively small number of parameters should be selected, sufficient for a good description of the curve. Therefore, we chose the efficiency formula as

$$\varepsilon(E_\gamma) = f(E_\gamma) \exp \left[-g_0(x) + g_1(x) \ln \left(\frac{E_\gamma}{E_0} \right) - g_2(x) \ln^2 \left(\frac{E_\gamma}{E_0} \right) + g_3 \ln^3 \left(\frac{E_\gamma}{E_0} \right) - g_4/E_\gamma^\delta \right] \quad (9)$$

with an additional coefficient $f(E_\gamma)$ that takes into account the absorption process

$$f(E_\gamma) = g_5 + g_6 \ln \left(\frac{E_\gamma}{E_0} \right) + g_7 \ln^2 \left(\frac{E_\gamma}{E_0} \right). \quad (10)$$

This approximation with at least three additional parameters (Table 3) allows us to successfully (within the experimental errors) describe the efficiency of the detector with the presence of an absorbing screen, as shown in Fig. 4 (dashed lines).

Table 3
Best values for the efficiency description parameters with an absorbing screen

Parameter	Value	Error
g ₅	-2.408	0.0804
g ₆	0.7636	0.0306
g ₇	-0.04871	0.0028
χ^2/dof	0.94	

The parameters presented in Table 3 were obtained by simultaneously fitting the experimental points at fixed distances of 50 and 100 mm with an absorbing screen.

To approximate the experimental values of efficiency, the CERN MINUIT program [22] of the least squares method was used.

CONCLUSIONS

A universal empirical formula for describing the dependence of detector efficiency on γ -radiation energy at fixed distances between point calibration γ -sources and the detector surface in the presence of an absorbing screen made of 12X18H10T stainless steel is proposed, obtained on the basis of experimental data.

The resulting parameterization allows us to obtain numerical values of efficiency for a wide range of energies up to 3000 keV (i.e., for energies that are

higher than those provided by the use of standard calibration sources – up to 1408 keV).

The results of the presented studies are necessary to ensure technical regulations for conducting γ -spectrometric measurements of high-level samples of fertile and fissile nuclear materials stored in sealed stainless steel containers when analyzing their isotopic composition.

ACKNOWLEDGEMENTS

The authors express their gratitude to the microtron group of I.M. Kushtan for the technical support of the experimental studies.

REFERENCES

1. M. Grdeń Non-classical applications of chemical analysis based on nuclear activation // *J. Radioanal. Nucl. Chem.* 2020, v. 323, p. 677-714; <https://doi.org/10.1007/s10967-019-06977-w>
2. C. Agarwal, S. Chaudhury, T.N. Nathaniel, A. Goswami. Nondestructive assay of plutonium in empty stainless steel boxes by apparent mass method // *J. Radioanal. Nucl. Chem.* 2012, v. 294, p. 77-80; <https://doi.org/10.1007/s10967-011-1474-3>
3. I.V. Pylypchynets, O.O. Parlag, V.T. Maslyuk, et al. Isotopic identification of photofissed nuclear materials in stainless steel containers using delayed gamma-rays // *Problems of Atomic Science and Technology.* 2022, №5 (141), p. 103-109.
4. J. Knezevic, D. Mrdja, J. Hansman, et al. Corrections of HPGe detector efficiency curve due to true coincidence summing by program EFFTRAN and by Monte Carlo simulations // *Applied Radiation and Isotopes.* 2022, v. 189, Article number: 110421; <https://doi.org/10.1016/j.apradiso.2022.110421>
5. I. Pylypchynets., A. Lengyel, O. Parlag, et al. Empirical formula for the HPGe- detector efficiency dependence on energy and distance // *J. Radioanal. Nucl. Chem.* 2019, v. 319, p. 1315-1319; <https://doi.org/10.1007/s10967-019-06426-8>
6. M.-C. Lépy, A. Pearce, O. Sima Uncertainties in gamma-ray spectrometry // *Metrologia.* 2015, v. 52, p. S123-S145; <https://doi.org/10.1088/0026-1394/52/3/S123>
7. J. Saegusa, T. Oishi, K. Kawasaki, et al. Determination of gamma-ray efficiency curves for volume samples by the combination of Monte Carlo simulations and point source calibration // *Journal of Nuclear Science and Technology.* 2000, v. 37, p. 1075-1081; <https://doi.org/10.1080/18811248.2000.9714994>
8. P. Reimer P. Fast. *Neutron Induced Reactions Leading to Activation Products. Selected Cases Relevant to Development of Low Activation Materials, Transmutation and Hazard Assessment of Nuclear Wastes:* Thesis, 2002, JRC23077, 115 p.
9. E. Stancu, C. Costache, O. Sima. Monte-Carlo simulation of p-type HPGe detectors – the dead layer problem // *Romanian Reports in Physics (Atomic and nuclear physics).* 2015, v. 67, p. 465-473.
10. S. Kaya, N. Çelik, T. Bayram. Effect of front, lateral and back dead layer thicknesses of a HPGe detector

- on full energy peak efficiency // *Nucl. Inst. Meth. A*. 2022, v. 1029, Article number: 166401; <https://doi.org/10.1016/j.nima.2022.166401>
11. M.C. Lépy. *Detection efficiency*: Presented to IAEA-ALMERA Technical Visit at Laboratoire National Henri Becquerel, 2010; http://www.nucleide.org/ICRM_GSWG/Training/Efficiency.pdf
 12. XCOM: Photon Cross Sections Database; <https://www.nist.gov/pml/xcom-photon-cross-sections-database>
 13. Data for: A simple spreadsheet program for calculating mass attenuation coefficients and shielding parameters based on EPICS2017 and EPDL97 photoatomic libraries; <https://data.mendeley.com/datasets/5p3grx7pgg/1>
 14. L. Seenappa, H.C. Manjunatha, N. Sowmya, K.N. Sridhar. A study of energy absorption buildup factors of some steels // *Radiat. Prot. Environ.* 2018, v. 41, p. 123-127; https://doi.org/10.4103/rpe.RPE_52_18
 15. L. Seenappa, H.C. Manjunatha, K.N. Sridhar, Ch. Hanumantharayappa. Semiempirical formula for exposure buildup factors // *Radiation Effects and Defects in Solids*. 2017, v. 172, p. 790-798; <http://dx.doi.org/10.1080/10420150.2017.1393426>
 16. A.I. Lengyel, O.O. Parlag, V.T. Maslyuk. Semiempirical description of Ge(Li)- and HPGe-detectors efficiency for potofission experiments // *Scientific Herald of Uzhhorod University. Series "Physics"*. 2009, v. 25, p. 95-99.
 17. J. Lee, H. Kim, Y.U. Kye, et al. Source and LVis based coincidence summing correction in HPGe gamma-ray spectrometry // *Nuclear Engineering and Technology*. 2022, v. 54, p. 1754-1759; <https://doi.org/10.1016/j.net.2021.11.008>
 18. A.K. Chakraborty, M.S. Uddin, M.A. Shariff, et al. Efficiency calibration of γ -ray detector for extended sources // *Pramana – J. Phys.* 2019, v. 92, Article number: 67; <https://doi.org/10.1007/s12043-019-1735-1>
 19. Steel 12X18H10T. Foreign analogues of steel grade 12X18H10T (old X18H10T) (in rus.). <https://westa.kiev.ua/ru/standarty/marki-stali/stal-12x18h10t>
 20. M.V. Strilchuk. *User manual for Winspectrum. KINR NAS of Ukraine (unpublished)*. Private communication.
 21. Decay Radiation database version of 3/25/2022. https://www.nndc.bnl.gov/nudat2/indx_dec.jsp
 22. F. James, M. Ross. *Function minimization and error analysis*. MINUIT D506. CEREN Computer Centre Program library. 1967, p. 1-47.

Article received 19.04.2023

ЕМПІРИЧНА ФОРМУЛА ЗАЛЕЖНОСТІ ЕФЕКТИВНОСТІ HPGe-ДЕТЕКТОРА ВІД ЕНЕРГІЇ ТА ВІДСТАНІ ДЛЯ ЕКРАНОВАНИХ ДЖЕРЕЛ ГАММА-ВИПРОМІНЮВАННЯ

І.В. Пилипчинець, О.І. Лендел, О.О. Парлаг, Є.В. Олейніков

Точність гамма-спектрометричних вимірів при ізотопному аналізі екранованих ядерних матеріалів залежить від точності калібрування детектора за енергетичною ефективністю, що повинна враховувати поправки, пов'язані з геометрією вимірювань, та поглинанням γ -випромінювання матеріалом екрана. Представлено результати експериментальних досліджень енергетичної ефективності HPGe-детектора, яка виміряна при фіксованих відстанях калібрувальне джерело – детектор (50 і 100 мм) при наявності та відсутності поглинаючого екрана із нержавіючої сталі марки 12X18H10T (товщина – 9,6 мм). На основі експериментальних даних отримано емпіричне описання залежності ефективності для фіксованих відстаней між джерелом гамма-випромінювання і детектором при наявності поглинаючого екрана із нержавіючої сталі.