THE DEPENDENCE OF Ge DETECTORS EFFICIENCY ON THE DENSITY OF THE SAMPLES IN GAMMA-RAY SPECTROMETRY

by

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The effect of the density of environmental samples on the counting efficiency of Ge detectors used in gamma-spectrometry was studied. The dependence $\varepsilon_{ff}(\rho)$ was determined for two HPGe detectors (relative efficiencies 18% and 20%), using five radioactive standard reference materials (silicone resins, epoxy resin, milk powder, soil) with different matrix densities (0.45-1.22 g/cm³) in Marinelli beakers (V = 500 cm³). The dependence of efficiency *vs*. density was found to be linear and the regression parameters for energies in the range of 60-2000 keV were determined, too. The effect of variation in density on the counting efficiency of Ge detectors is dominant in the range of lower energies (60-600 keV) and decreases with energies in the higher energy range.

Key words: gamma-spectrometry, counting efficiency, environmental samples, Ge detector

INTRODUCTION

One of the crucial problems concerning the validity of the results in gamma-ray spectrometry of voluminous environmental samples by semiconductor Ge detectors is the efficiency calibration of the detector counting system. Ideally, the radioactive reference material used for calibration and the sample under study should be of the same dimensions, measuring geometry, chemical composition and density, and of similar content and past activities of the radionuclides. In that case, detector efficiency could be determined by simply comparing the count rates in the corresponding full energy peaks in the pulls-height spectra. However, as detectors are usually calibrated on efficiency by reference multi-lines or multi-nuclides standard sources with different gamma-energies, different corrections of the full-energy peak efficiency should be performed. Corrections are always necessary when there is a difference between the standard calibration source and the sample with respect to geometry, chemical composition, and density. Also, when either the calibration source or the sample contains radionuclides with gamma-cascade transitions, the corrections for coincidence summing effects are necessary [1-3].

Problems arising from differences in geometry could be solved by adjusting the measuring geometry of the calibration source and the sample. Differences in the chemical composition between the standard calibration source and the sample are usually negligible, since the mass attenuation coefficients for natural materials are very similar. Naturally occurring materials are composed of elements with atomic numbers less than 20 (Z 20) and effective atomic numbers less than 15 ($Z_{eff} < 15$). For environmental samples with characteristic gamma-ray energies in the range of 60-2000 keV, the mass attenuaton coefficients μ mainly depend on photon energy, while the effects on the effective atomic number Z_{eff} can be neglected [4]. But, contrary to the corrections for geometry and chemical composition, if the matrices of the sample and the calibration source differ in density, the differences in self-attenuation effects cannot be neglected and corrections for density are necessary [5]. For environmental samples with characteristic gamma-ray

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energies in the range of 60-2000 keV and densities in the range of 0.2-2.0 g/cm^3, the dependence of the detector's efficiency on the matrix density is often found to be linear [4, 6-9].

Generally, in studies of the effect of the environmental samples' matrix density on the counting efficiency of Ge detectors, secondary reference standards are of the same activity levels and radionuclides content. In this work, the effect of density on detector efficiency was studied on two HPGe detectors, using different radioactive standard reference materials in Marinelli beakers of the same volume (500 cm^3). The standard reference materials were supplied by different suppliers and had different content and activity level of the radionuclides.

MATERIALS AND METHODS

Characteristics of the detectors

The characteristics of detectors D_1 and D_2 (coaxial HPGe, Canberra, relative efficiencies 20% and 18%, respectively) are presented in tab. 1.

Table 1. Characteristics of HPGe detectors D_1 and D_2

Detector		D_1	D_2
Geometry and type		Closed coaxial – p type	Closed reverse coaxial – n type
Relative efficiency (compared to NaI-detector)		20%	18%
Resolution	at 122 keV	0.850 keV	0.759 keV
	at 1332 keV	1.8 keV	1.69 keV
Peak/Compton ratio		51:1	56:1
Cryostat		Vertical (+ preamplifier)	Vertical (+ preamplifier)
Cristal diameter		49.5 mm	48 mm
Cristal length		56.5 mm	48.5 mm
Distance from window		55 mm	5 mm
Entrance window		Al	Be
Voltage		(+) 4500 V	(–) 4000 V

Characteristics of radioactive standards and standard reference materials

The detectors were calibrated on counting efficiency with the radioactive standard reference materials in Marinelli beakers of the same volume (500 cm^3). The following standard reference materials (SRM 1-5) were used:

SRM 1: *Milk powder* spiked by ^{22}Na, ^{57}Co, ^{60}Co, ^{89}Y, ^{133}Ba, and ^{137}Cs; total activity 1.5 kBq/kg at 01.07.1991; Type MIX-OMH-SZ (SMP), National Office of Measures, Budapest;

SRM 2: *Silicone resin* with homogeneously dispersed radionuclides ^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{113}Sn, ^{85}Sr, ^{88}Y, and ^{203}Hg; total activity 85 Bq/kg at 10. 03. 2001; Type: MBSS 2; Czech Metrological Institute, Inspectorate for Ionizing Radiation;

SRM 3: *Soil* spiked by ^{22}Na, ^{57}Co, ^{60}Co, ^{89}Y, ^{133}Ba, and ^{137}Cs; total activity 1.5 kBq/kg at 01. 07. 1991; Type MIX-OMH-SZ; National Office of Measures, Budapest;

SRM 4: *Epoxy resin* with homogeneously dispersed radionuclide ^{152}Eu; total activity 4.218 kBq at 04. 06. 1987; Type: EGMGE 1; DAMRI, Laboratoire de Metrologie des Rayonnements Ionisants, and

SRM 5: *Silicone resin* with homogeneously dispersed radionuclides ^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{113}Sn, ^{85}Sr, ^{88}Y, and ^{203}Hg; total activity 90 Bq/kg at 10. 03. 2001; Type: MBSS 2; Czech Metrological Institute, Inspectorate for Ionizing Radiation.

Other characteristics of the standard reference materials (densities and reference activities) are presented in tab. 2.

Table 2. Characteristics of the radioactive standard reference materials

Material	Matrix material	Density [g/cm^3]	Reference activity [Bq]
SRM 1	Milk powder	0.45 0.02	390 10
SRM 2	Silicone resin	0.98 0.01	40 1
SRM 3	Soil	1.00 0.01	750 20
SRM 4	Epoxy resin	1.15 0.02	4200 300
SRM 5	Silicone resin	1.22 0.01	52 2

Efficiency calibration

Efficiency as a function of energy ($\log \varepsilon_{ff}$ *vs.* $\log E$) is usually fitted by a polynomial function, but better aproximation for the whole energy range (60-2000 keV) is achieved by two polynomial functions junctioned at about 200 keV [1, 10]. Aproximately linear $\log \varepsilon_{ff}/\log E$ correlation in the energy range of 200-2000 keV for point sources can also be applied for Marinelli beakers of different densities, but better fit is obtained with quadratic regression [7, 11].

In our work, detector D_1 was calibrated for the whole of the energy range (60-2000 keV) by the polynomial function $\log \varepsilon_{ff} = P(\log E)$ of the 4th degree in the lower enery range and a polynomial of the 2nd degree in the higher energy range. Detector D_2 was calibrated in the range of 200-2000 keV, while the dependance $\log \varepsilon_{ff}$ *vs.* $\log E$ was fitted by a linear function.

The efficiency of semiconductor Ge detectors may be expressed as a product of geometric efficiency ε_{geom}, intrinsic efficiency ε_{int}, and sample efficiency ε_{sample} [12]. Geometric efficiency is independent of photon energy E, since it represents the fraction of the emitted photons that are intercepted by the detector. Intrinsic efficiency is defined as a probability that a gamma-ray that enters the detector will interact within the active volume of the detector and generate a pulse which contributes to the full energy peak. Sample efficiency is proportional to the self-absorption factor $f_s(\mu, d, \rho$ *i. e.* the fraction of photons that actually leave the sample. Thus, the efficiency is expressed as

$$\varepsilon_{\mathrm{ff}} \quad \varepsilon_{\mathrm{geom}}\ \varepsilon_{\mathrm{int}}\ f_{\mathrm{s}}(\mu, d, \rho) \tag{1}$$

where μ is the mass attenuation coefficient, d is thickness, and ρ is the density of the sample. When the source-detector geometry is fixed and the values of the mass attenuation coefficients are close enough, as is the case with environmental samples, and assuming the dependence of intrinsic efficiency on the density of the samples is negligible; the efficiency for a certain photon energy E will depend only on the self-absorption factor

$$\varepsilon_{\mathrm{ff}}(E_\gamma, \rho) \quad \mathrm{const} \quad f_{\mathrm{s}}(\rho) \tag{2}$$

where f_s depends only on the density of the sample [4].

If a thin cylindrical source with homogeneously distributed activity is placed coaxially with the detector at a far distance, the self-absorption factor is expressed as [1]

$$f_s(\mu, d, \rho) \quad \frac{1 \quad \mathrm{e}^{\mu d\rho}}{\mu d\rho} \tag{3}$$

If eq. (3) is developed (μd < 0.5 cm^3/g and ρ < < 2 g/cm^3):

$$f_{\mathrm{s}}(\mu, d, \rho) \quad 1 \quad \frac{1}{2}\mu d\rho \quad 1 \quad \mathrm{const} \quad \rho \tag{4}$$

one can see that f_s changes linearly with density [4, 7]. Combining eqs. (2) and (4), the counting efficiency as a function of density could be expressed as

$$\varepsilon_{\mathrm{ff}}(E_\gamma, \rho) \quad a\rho \quad b \tag{5}$$

where a and b are linear parametars that depend on the characteristics of the voluminous sample and of the detector itself.

In our work, we determined the efficiency functions for five radioactive standard reference materials (SRM 1-5) in Marinelli beakers (V = 500 cm^3) for gamma ray energies in the range of 60-2000 keV for the detector D_1 and for the energies in the range of 200-2000 keV for the detector D_2. We investigated the dependence of detector efficiency on the sample density $\varepsilon_{\mathrm{ff}}(\rho)$ for gamma ray energies emitted by natural radionuclides from the ^{238}U and ^{232}Th decay series and ^{40}K, and for gamma ray energies from radionuclides often dispersed in standard reference materials.

The efficiency values were calculated for the following energies: 63 keV (^{234}Th); 88 keV (^{109}Cd), 122 keV (^{57}Co), 143 keV (^{235}U); 186 keV (^{226}Ra and ^{235}U); 295 keV (^{214}Pb); 338 keV (^{228}Ac); 352 keV (^{214}Pb); 609 keV (^{214}Bi); 661 keV (^{137}Cs); 911 keV (^{228}Ac); 1120 keV (^{214}Bi); 1460 keV (^{40}K); 1763 keV (^{214}Bi), and 1836 keV (^{88}Y).

Gamma spectrum was analyzed with the standard software package *Genie 2000*.

RESULTS AND DISCUSSION

Efficiency *vs.* energy

The coefficients of the polynomials efficiency functions calculated for detectors D_1 and D_2 (geometry Marinelli) are presented in tabs. 3 and 4. The coefficients were determined by the linear least squares method. Numerical values of the coefficients were established at a confidence level of 95% (2) and efficiency uncertainties were 1-5% (2).

An example presenting a good fit for the efficiency functions of detectors D_1 and D_2 (SRM 3, Marinelli, matrix: soil) is presented in figs.1 and 2.

Efficiency *vs.* density

Calculated efficiencies for detectors D_1 and D_2 (tabs. 3 and 4) *vs.* density for different photon energies are presented in figs. 3, 4, and 5.

As presented in figs. 3-5, the effect of density on detector efficiency is more pronounced in the lower

Table 3. Coefficients of detector D_1 efficiency functions (4th degree polynomial in the energy range 60-180 keV and 2nd degree polynomial in the range 180-2000 keV)

Material	$\ln(E_{\mathrm{ff}}) = p_1(\ln E)^4 + p_2\ \ln E)^3 + p_3(\ln E)^2 + p_4(\ln E) + p_5$ (energy range 60-180 keV)					$\ln(E_{\mathrm{ff}}) = n_1(\ln E)^2 + n_2(\ln E) + n_3$ (energy range 180-2000 keV)		
	p_1	p_2	p_3	p_4	p_5	n_1	n_2	n_3
SRM 1	–0.253	5.846	–50.57	193	–272.7	–0.0257	–0.555	4.984
SRM 2	–0.159	3.747	–33.21	130	–187.5	–0.0363	–0.372	4.172
SRM 3	–0.151	3.625	–32.64	130	–189.7	–0.0356	–0.396	4.386
SRM 4	–0.221	5.102	–44.16	169	–239.2	–0.0524	–0.173	3.608
SRM 5	–0.148	3.488	–30.98	122	–176.3	–0.0286	–0.471	4.449

Table 4. Coefficients of detector D_2 efficiency functions

Material	$\ln(E_{ff}) = m_1(\ln E)^2 + m_2$ (energy range 200-2000 keV)	
	m_1	m_2
SRM 1	–0.9271	6.095
SRM 2	–0.9049	5.907
SRM 3	–0.9206	6.059
SRM 5	–0.8944	5.791

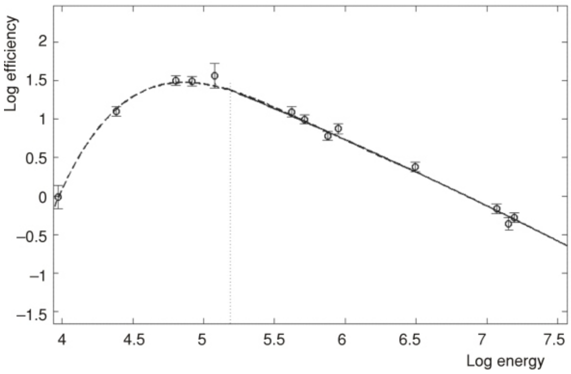

Figure 1. Efficiency function for detector D_1 (SRM 3, matrix: soil)

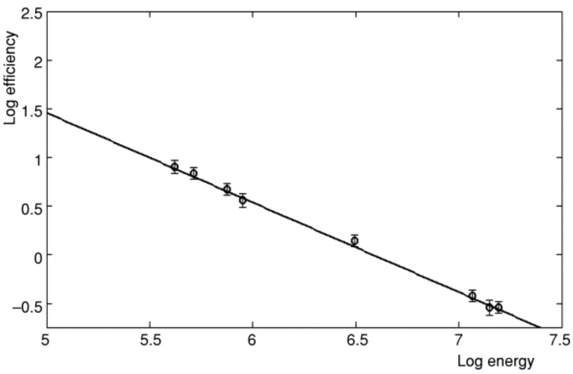

Figure 2. Efficiency function for detector D_2 (SRM 3, matrix: soil)

energy range of 60-600 keV, while in the higher energy range (600-2000 keV) where the self-absorption factors for different matrices are similar, the effect is less important.

According to eq. (5), the efficiency is fitted as a linear function of the density, the parametars of the function depend on the characteristics of the voluminous sample and the detector. In our case, for detectors D_1 and D_2, the values of the linear regresion coefficients a and b, are presented in tab. 5. The coefficients were determined by the least squares method at a confidence level of 95% (2σ).

Correlation coefficients r were calculated, too. For detector $D_{1,}$ in the low energies range from

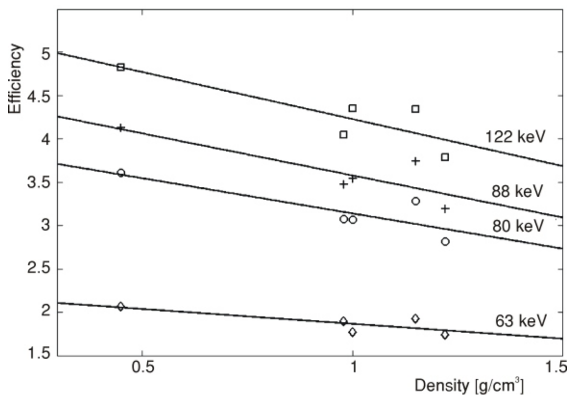

Figure 3. Efficiency *vs.* density for detector D_1 (gamma energies: 63, 80, 88, and 122 keV)

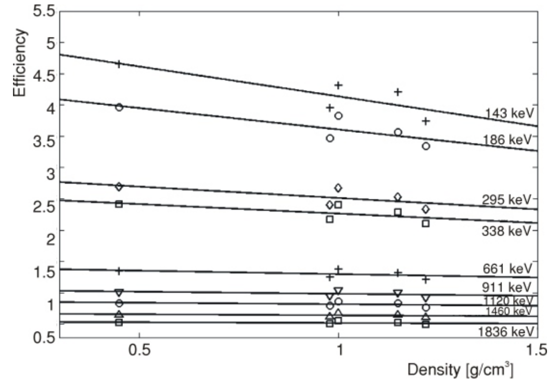

Figure 4. Efficiency *vs.* density for detector D_1 (gamma energies: 143, 186, 295, 338, 661, 911, 1120, 1460, and 1836 keV)

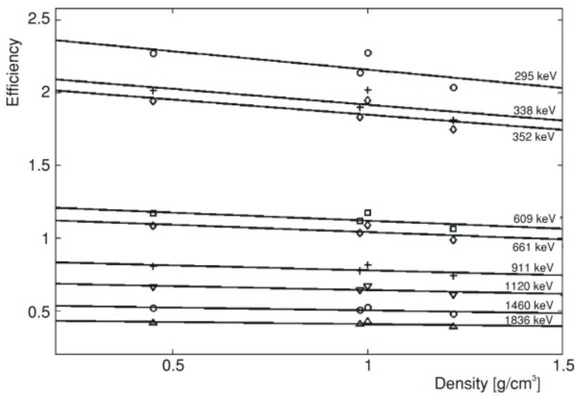

Figure 5. Efficiency *vs.* density for detector D_2 (gamma energies: 295, 338, 352, 609, 661, 911, 1120, 1460, and 1836 keV)

60-200 keV (where there is a clear difference in the photon self-absorption coefficient for each matrix), there is a strong linear correlation between the efficiency and the density (r 0,84). For both detectors D_1 and D_2, in the range of 200-900 keV, the correlation is medium (r 0,6), while in the range of higher energies (900-

Table 5. Coefficients of linear regression *a* and *b* for detectors D_1 and D_2 (geometry: Marinelli, SRM 1-5)

Energy [keV]	$\varepsilon_{ff}(\rho) = a\rho + b$			
	Detector D_1		Detector D_2	
	a_1	b_1	a_2	b_2
63	0.3427	2.209		
88	0.9706	4.546		
122	1.0860	5.311		
143	0.9538	5.090		
186	0.6859	4.293		
295	0.3633	2.876	0.2521	2.408
338	0.3012	2.566	0.2177	2.133
352	0.2841	2.477	0.2080	2.054
609	0.1238	1.513	0.1106	1.228
661	0.1093	1.407	0.1007	1.139
911	0.0660	1.050	0.0691	0.844
1120	0.0474	0.864	0.0537	0.694
1460	0.0312	0.671	0.0386	0.539
1763	0.0239	0.566	0.0309	0.455
1836	0.0222	0.540	0.0290	0.434

-2000 keV), the correlation is low (r 0,35). This could be explained by the fact that the change of the self-absorption factor with density $f_s(\rho)$ and, consequently, the change in efficiency with density $\varepsilon_{ff}(\rho)$, is almost within the experimental uncertainty of the efficiency determination, that is in agreement with the literature data [4, 7, 9].

For gamma-ray energies above 1000 keV, our regression lines are almost horizontal. For that energy range, a significant difference occurs in the values of the linear regression parameters of the $\varepsilon_{ff}(\rho)$ function declared by other authors. For standard reference materials in synthetic matrices (density range: 0-2 g/cm^3), a clear linear fit was found [8], while for standards made of natural materials of different densities (density range: 0-1,5 g/cm^3), the regression lines were almost horizontal [6]. Therefore, it can be concluded that even in the higher energy range (1000-2000 keV) where there are no significant changes in the counting efficiency due to density, more accurate results are obtained if the function "efficiency *vs*. density" is determined using reference materials of different densities but in the same matrix.

CONCLUSION

The effect of sample density on counting efficiency was determined for two HPGe detectors with a series of standard reference materials in Marinelli geometry. A linear fit was found $\varepsilon_{ff}(\rho) = a\rho + b$, following the linear dependence of the sample self-absorption factor with its density. Liner regresion coefficients *a* and *b* were calculated for a series of energies in the range of 60-2000 keV emitted by radionuclides usually found in environmental samples. The effect of density variation on detector counting efficiency was found to be dominant in the range of the lower energies (60-600 keV), while in the higher energy range, there were no significant changes in detector counting efficiency due to density. This should be taken into consideration in gamma spectrometry of the voluminous environmental samples.

REFERENCES

[1] Debertin, K., Helmer, R. G., *Gamma- and X-ray Spectrometry with Semiconductor Detectors*, Elsevier, New York, USA, 1988

[2] Debertin, K., Jianping, R., Measurement of the Activity of Radioactive Samples in Marinelli Beakers., *Nucl. Instr. and Meth. A, 278* (1989), pp. 541-549

[3] Djurić, G., Popović, D., Adžić., P., Efficiency of Ge-Li Detector for Different Geometries and Radionuclide Carriers, *Proceedings*, XII Yugoslav Symposium on Measurement and Instrumentation, Belgrade, 1986, pp. 535-542

[4] Boshkova, T., Minev, L., Corrections for Self-Attenuation in Gamma-Ray Spectrometry of Bulk Samples, *Apll. Radiat. Isot., 54* (2001), 5, pp. 777-783

[5] Park, T. S., Jeon, W. J., Measurement of Radioactive Samples in Marinelli Beakers by Gamma-Ray Spectrometry, *J.Radioanal. Nucl. Chem., 193* (1995), 1, pp. 133-144

[6] Taskaeva, M., Taskaev, E., Penev, I., On the Preparation of Efficiency Calibration Standards for Gamma-Ray Spectrometers., *Apll. Radiat. Isot., 47* (1996), 9-10, pp. 981-990

[7] Alfassi, Z. B., Lavi, N., The Dependence of the Counting Efficiency of Marinelli Beakers for Environmental Samples on the Density of the Samples, *Apll. Radiat. Isot., 63* (2005), 1, pp. 87-92

[8] Khan, A., Grey, C., Ramirez, A., Germanium Detector Efficiency Variations due to Chances in Matrix Density for Standard Marinelli Beakers, *Proceedings*, 41st Bioassay Analytical and Environmental Radiochemistry Conference, Boston, USA, November 12-16, 1995

[9] Melquiades, F. L, Appoloni, C. R., Self-Absorption Correction for Gamma Spectrometry of Powdered Milk Samples Using Marinelli Beaker, *Apll. Radiat. Isot., 55* (2001), 5, pp. 697-700

[10] Helmer, R. G., Cline, J. E., Greenwood, R. C., The Electromagnetic Interaction in Nuclear Spectroscopy (Ed. W. D. Hamilton), North-Holland Publishing Company, Oxford, UK, 1975

[11] Alfassi, Z. B., Lavi, N., Development and Application of Marinelli Beaker Standards for Monitoring Radioactivity in Dairy-Products by Gamma-Ray Spectrometry, *Apll. Radiat. Isot., 61* (2004), 6, pp. 1437-1441

[12] Reilly, D., Ensslin, N., Smith, Jr. H., Kreiner, S., Passive Nondestructive Assay of Nuclear Materials, (Eds. D. Reilly, N. Ensslin, H. Smith Jr.), U. S. Nuclear Regulatory Comission, Washington, DC, 1991

Ивана ВУКАШИНОВИЋ, Драгана ТОДОРОВИЋ, Драгана ПОПОВИЋ

ЗАВИСНОСТ ЕФИКАСНОСТИ ГЕРМАНИЈУМСКИХ ДЕТЕКТОРА ОД ГУСТИНЕ УЗОРАКА У СПЕКТРОМЕТРИЈИ ГАМА ЗРАЧЕЊА

Испитиван је утицај густине узорака из животне средине на ефикасност бројања германијумских детектора у спектрометрији гама зрачења. Зависност $\varepsilon_{\mathrm{ff}}(\rho)$ одређена је за два HPGe детектора (релативних ефикасности 18% и 20%) за различите радиоактивне стандардне референтне материјале (силиконске смоле, епокси смолу, млеко у праху, земљу) различитих густина (0,45-1,22 g/cm^3) у Маринели посудама запремине V = 500 cm^3. Утврђена је линеарна зависност ефикасности бројања од густине узорака при чему су одређени параметри линеарне регресије за низ енергија из интервала од 60-2000 keV. Ефект промене густина на ефикасност бројања германијумских детектора доминантан је у интервалу нижих енергија (60-600 keV) и смањује се са порастом енергије у интервалу виших енергија.

Кључне речи: гама спектрометрија, ефикасност бројања, узорци околине, германијумски детектор