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EFFECT OF SAMPLE POSITION ON EFFICIENCY IN GAMMA-RAY SPECTROSCOPY

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GAMA-IŞIN SPEKTROSKOPİSİNDE NUMUNE POZİSYONUNUN VERİM ÜZERİNDEKİ ETKİSİ

ABSTRACT

The full energy peak efficiency for the energies of interest must be known to determine the concentration of activity in gamma-ray spectrometry. This work is focused on examining the effect of sample position on the FEPE. For this purpose, efficiency values were determined by experimental and Monte Carlo simulation method using three different HPGe detectors and standard charcoal cartridges. The cartridges were first counted in the centers of the detectors and then replaced on the X-axis at 5 mm intervals. At maximum distances from the center of 25 mm for detector-1 (called Ge-1), 15 mm for detector-2 (called Ge-2), and 10 mm for detector-3 (called Ge-3), experimental the FEPE values decreased by 11.8-16.4%, 8.2-13.8%, and 4.9-11.0%, respectively. The results show that the sample position significantly affects the full energy peak efficiency. Furthermore, counting must be conducted using sample locator, however, the counting should be carefully completed in the center of the detector unless that is possible.

Key Words: Gamma-Ray Spectroscopy, Full Energy Peak Efficiency, Sample Position, Charcoal Cartridge Filter.

ÖZET

Gama-ışın spektrometrisinde aktivite konsantrasyonunu belirlemek için ilgilenilen enerjilere ilişkin toplam enerji pik verimi bilinmelidir. Bu çalışmada, numune konumunun toplam enerji pik verimi üzerindeki etkisini incelemeye odaklanılmıştır. Bu amaçla üç farklı HPGe dedektör ve standart karbon kartuşlar kullanılarak deneysel ve Monte Carlo simülasyon yöntemiyle verim değerleri belirlenmiştir. Kartuşlar öncelikle dedektörlerin merkezlerinde, daha sonra X- ekseninde 5 mm aralıklarla değiştirilerek sayılmıştır. Merkezden dedektör-1 (Ge-1 olarak adlandırılan) için 25 mm, dedektör-2 (Ge-2 olarak adlandırılan) için 15 mm ve dedektör-3 (Ge-3 olarak adlandırılan) için 10 mm'lik maksimum mesafelerde deneysel toplam enerji pik verim değerlerindeki azalma sırasıyla; %11,8-16,4, %8,2-13,8 ve %4,9-11,0'dir. Sonuçlar, numune pozisyonunun toplam enerji pik verimini önemli ölçüde etkilediğini göstermektedir. Numunelerin mümkün olduğu sürece numune tutucu ile sayılması gerekirken, mümkün olmadığı durumlarda ise sayımın dedektörün merkezinde dikkatli bir şekilde yapılması gerektiğini göstermektedir.

Anahtar Kelimeler: Gama-Işın Spektroskopisi, Toplam Enerji Pik Verimi, Numune Pozisyonu, Karbon Kartuş Filter.

1. Introduction

Especially due to their good energy resolution, HPGe detectors are a useful tool used in many areas such as activity measurements in all kinds of gamma-ray emitting samples, nuclear structure studies, radiation protection, and environmental monitoring (Chuong et al., 2016; Prozorova et al., 2021, Huynh et al., 2023). In determining the activities of radionuclides in the sample by gamma spectrometric method, it is imperative to determine the full energy peak efficiency (FEPE) for each photon energy under the same measurement conditions. The FEPE, the most significant parameter in practical γ-ray spectrometry, is the ratio of the number of counts detected at a peak to the number emitted by the source (Guerra et al., 2017, Gurau et al., 2024). The efficiency of the HPGe detector gamma-counting system depends on many parameters such as the crystal size of the detector, its geometric properties, the detector-source distance, the shape of the source, the materials around the detector, and the absorptions in the source/sample matrix (Khan et al., 2018, Lin et al., 2023, Park et al., 2023). The FEPE is thus a complex function characterized by the detector (its dimensions and composition) and the measurement conditions (composition and geometry of the source) (Montalván Olivares et al., 2017). Therefore, the accuracy of the quantity to be calculated depends not only on the characteristics of the detector and the signal processing system but also

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on the sensitivity of the measurement conditions in the laboratory (Lépy et al., 2019). While the performance of measurement systems has traditionally been defined in terms of accuracy and precision, it is more accurate to define it in terms of repeatability and reproducibility in terms of the quality of analytical results (Gilmore, 2008). Repeatability is the closeness of the result of consecutive measurements under the same laboratory conditions without changing the experimental condition. Reproducibility is the closeness of measurement results performed under varying conditions (Ağuş, 2019). Therefore, there may be a contribution to the uncertainty budget during the reproducibility of efficiency measurements due to sample location change. That is, many sources of uncertainty such as count statistics, decay, certified activities, random summing correction, and self-absorption correction come with additional systematic uncertainty from sample height, homogeneity, and position in the gamma-ray spectrometer (Yücel et al., 2011). Gilmore (2008) stated that the uncertainty due to sample position can be neglected using the sample locator. To avoid this uncertainty, it is recommended to use a sample locator placed on the detector for routine gamma spectrometric measurements. However, this may not always be possible. Sample locators may not be available for every detector and sample container or the sample locator may not be used in unusually shaped geometries or in samples with geometry that cannot be placed in the sample container.

The effect of sample position, one of the measurement conditions, was investigated using charcoal cartridge filters, associated with the environmental monitoring, towards detector efficiency within the scope of this study. The motivation of this study was to find out what changes in sample position would cause a change in the FEPE. For this purpose, these changes were experimentally determined using the PHITS MC program, together with charcoal cartridges placed at different positions on the end-caps of three different detectors.

2. Materials and Methods

2.1. The HPGe Detectors

Three HPGe detectors, one n-type and two p-type were used in this study. The first p-type detector named Ge-1 (Model: GEM150P4, Cryostat Configuration: PopTop, Preamplifier Model: A257P) has a 1 mm Al window with a relative efficiency of 150%. The n-type detector called Ge-2 (Model: GMX70P4, Cryostat Configuration: PopTop, Preamplifier Model: A257N) has a 0.76 mm Be window with 70% relative efficiency. The other p-type detector called Ge-3 (Model: IGC50195, Cryostat Model: NPR/8, Preamplifier Model: RG11B/C) has a 0.5 mm Al window with 58.49% relative efficiency. Ge-1 and Ge-2 detectors were connected to a digital signal processing analyzer (Ortec DSPECjr.2.0) with a 16K ADC/MCA operating through Gamma Vision spectroscopy software. To reduce fluorescence and scattered X-rays from the lead, the detectors were shielded with 10 cm thick Pb and graded with 1.6 mm Cu and 0.5 mm Sn liners. The amplifiers were calibrated with an appropriate amplifier gain to cover up to approximately 2680 keV on 8192 MCA channels. The Ge-3 detector was also connected to the Canberra DSA 1000 (16K channel) digital signal processing analyzer. The Ge-3 detector was shielded with 10 cm thick Pb, and 5 mm Cu to reduce the fluorescence and scattered X-rays from the lead.

2.2. Simulated Charcoal Gamma Cartridge Standards

Charcoal cartridge standards were used to calculate the experimental efficiency. The charcoal cartridge standards are NIST traceable ^{133}Ba (3.756 kBq), ^{137}Cs (3.811 kBq), and ^{60}Co (3.704 kBq) purchased from Eckert & Ziegler Isotope Products Inc. The cartridges are each 25.4 mm × 57.2 mm in size and the activity is uniformly distributed in the charcoal that

Table 1. Main physical characteristics of detectors.

Description		Ge-1	Ge-2	Ge-3
Crystal polarity		p-type	n-type	p-type
Relative efficiency		150%	70%	58.49%
Crystal diameter		94.8 mm	73.2 mm	65.8 mm
Crystal length		87.2 mm	65 mm	65.9 mm
Core diameter		11.2 mm	10.7 mm	9 mm
Core length		73.4 mm	58 mm	53 mm
Crystal to window distance		5 mm	4 mm	5 mm
End cap window		1.5 mm /Al	0.76 mm /Be	0.5 mm /Al
Dead layer thickness	Outside contact	700 µm Li	0.3 µm B	< 1 mm
	Hole contact	0.3 µm B	900 µm Li	
Full width at half maximum (FWHM)	5,9 keV (^{55}Fe)	-	0,96 keV	-
	122 keV (^{57}Co)	0,89 keV	-	1,22 keV
	1332,5 keV (^{60}Co)	2,11 keV	2,10 keV	2,0 keV
Peak to Compton ratio	1332,5 keV (^{60}Co)	90:1	66:1	67,15:1

fills the cartridge. An experimental set-up of charcoal cartridges is given in Figure 1.

Figure 1. An experimental set-up of charcoal cartridges.

2.3. Calculation of the Full Energy Peak Efficiency

The FEPE for the volumetric charcoal cartridge samples in the energy range of 81-1332.49 keV was calculated using Equation 1.

$$\varepsilon(E) = \frac{C(E)}{A \cdot f_\gamma(E)} \cdot F_c \quad (1)$$

Where C(E) (count s^{-1}) is the photon count rate in the FEPE at a certain energy E (keV), A (Bq) is the activity of the source at the measurement date, fγ (E) is the probability of gamma-ray emission of the energy of interest, and Fc is a correction factor that includes the true coincidence summing and self-absorption correction factors. While determining the FEPE values, self-absorption correction was made in all charcoal cartridges calculations. True coincidence summing correction was performed on ^{133}Ba and ^{60}Co nuclides that emit more than one gamma-ray per decay. Genie 2000™ and Gamma Vision spectroscopy software were used for spectral analysis and peak area calculation. The net areas and percent errors under full energy peaks were calculated using the "Interactive Peak Fit Option" in Genie and the “Interactive In Viewed Area” tools in Gamma Vision. Thus, with these tools, complex peaks around the peaks of a particular nuclide or in a particular energy region can be considered as discrete peaks. For this purpose, user-specified criteria are used as a filter function. Spectrum acquisition times were chosen based on the activity of the cartridges to keep the statistical uncertainty in the peak area below 1%. The mean values of the count rates of the measurements repeated three times were used to increase statistical precision. The uncertainty of the experimental efficiency is calculated according to the JCGM100 (2008) used in the uncertainty calculation.

2.4. Monte Carlo Simulations

PHITS (Particle and Heavy Ion Transport code System) Monte Carlo simulation code (Version 3.28) was used for the simulations in this paper (Sato et al., 2018). PHITS is a general-purpose Monte Carlo particle transport simulation code, developed by collaborations among the JAWA, RIST, KEK and the other institutions. If accessing to an institute in an OECD/NEA Databank participating country is available, it is possible to obtain the latest version of open source PHITS whenever it is required that through the OECD/NEA GitLab. If accessing to an institute in the pre-approved countries is not possible, the latest version of PHITS can be obtained from JAEA. PHITS is a flexible general-purpose code that successfully models HPGe detectors, which are recent in use than other codes in the field of gamma-ray spectrometry. While obtaining the FEPE values in PHITS, the T-deposit tally giving the energy distribution in a specific region in which the HPGe detector operates in pulse height mode, is used. The total number of histories taken into account in each run is 10^7 source particles, from which calculations with a relative error of less than 0.1% are obtained.

3. Results and Discussion

3.1. Experimental Measurements

Before starting the experimental measurements, a homogeneity test was performed with a ^{152}Eu point source. For this purpose, spectra were taken with 5 mm steps from the center in all axes indicated by -x, +x, -y, +y as shown in the Figure 1 on the two-dimensional coordinate plane on the detector surface. It was concluded that the detector surface is homogeneous according to deviations <1% in the net area values of the 121 keV, 778 keV, 1112 keV, and 1408 keV full energy peaks of ^{152}Eu. A similar test was performed examining the change in the 662 keV peak using a ^{137}Cs charcoal cartridge without a true coincidence summing effect, and it was seen that the change was independent of the axis in all detectors. According to these results, it was observed that scanning in the X-axis or Y-axis would be sufficient to determine the position dependence. Then, the experimental measurements were commenced by positioning the cartridges in the center of the top surfaces of the detectors. Since the charcoal cartridge has a diameter of 57.2 mm, the off-axis distances from the center were determined by considering the crystal diameters of the detectors given in Table 1. The off-axis distance was changed in 5 mm steps from the center in all detectors. This change was made on the X-axis in the two-dimensional coordinate plane on the detector surface, in the +X direction from the center (Figure 2). Accordingly, the change of position from the center; 5 mm, 10 mm, 15 mm, 20 mm and 25 mm in Ge-1; 5 mm, 10 mm, and 15 mm in Ge-2; in the Ge-3 detector, measurements were taken as 5 mm and 10 mm.

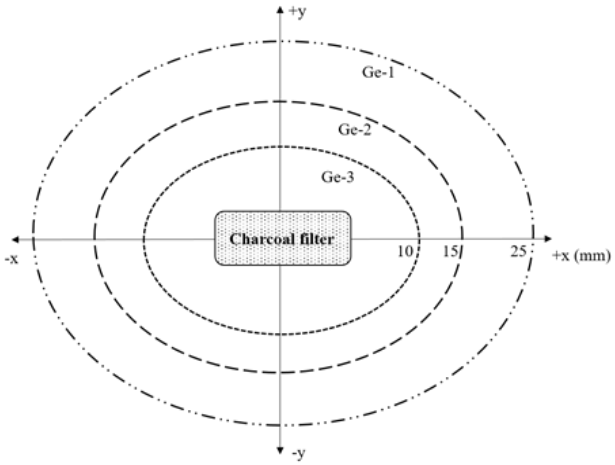

Figure 2. A schematic representation of the measured axes on the detector surface and the charcoal cartridge placed in the center as an example.

The used charcoal cartridges have peaks in the energy range of 81-1332.49 keV. The peaks of 81 keV of ^{133}Ba in the low-energy region, 302.85 keV of ^{133}Ba and 661.66 keV of ^{137}Cs in the medium-energy region, and 1173.23 and 1332.49 keV of ^{60}Co in the high-energy region were studied. Using these energies, 5 mm, 10 mm, 15 mm, 20 mm and 25 mm in Ge-1; 5 mm, 10 mm, and 15 mm in Ge-2; in the Ge-3 detector, the FEPE values obtained at 5 mm and 10 mm are given in Table 2. As it is shown in the Table 2, the efficiency decreases as the sample position moves away from the center. Detector geometry expressed as solid angle; is the detector-to-source distance and the geometric arrangement of the detector and source that depends on the detector/source dimensions. Therefore, the solid angle is the greatest at the position where the source is at the center of the detector. Uncertainties in the experimental efficiency results ranged from 1.3% to 2.1% for Ge-1, 1.5% to 2.0% for Ge-2 and 1.2% to 2.3% for Ge-3.

The percentage differences of all measurements taken off-center compared to the measurement taken from the center are given in Figures 3, 4, and 5. It was observed that the FEPE values decreased between 11.8-16.4% for Ge-1 (Figure 3), 8.2-13.8% for Ge-2 (Figure 4), and 4.9-11.0% for Ge-3 (Figure 5) in changes in sample position from the center. At the smallest displacement 5 mm from the center, the full energy peaks decreased by 0.7-2.0% in Ge-1, 0.5-1.4% in Ge-2 and 1.2-2.4% in Ge-3. In specific-purpose programs such as GESPECOR, DETEFF, and EFFTRAN, the sample position is assumed to be fixed in the center. In the study, true coincidence summing correction factors were obtained from GESPECOR and EFFTRAN softwares. Since the sample position cannot be changed in those, true coincidence summing factors in the center were used.

Table 2. Experimental FEPE values for Ge-1, Ge-2 and Ge-3 detector.

	Nuclide	Energy (keV)	Experimental FEPE					
			In the center	5 mm off-center	10 mm off-center	15 mm off-center	20 mm off-center	25 mm off-center
Ge-1	^{133}Ba	81.00	0.07338	0.07220	0.07143	0.07004	0.06970	0.06137
	^{133}Ba	302.85	0.07853	0.07318	0.07525	0.07379	0.07262	0.06627
	^{137}Cs	661.66	0.05485	0.05404	0.05311	0.05126	0.04877	0.04659
	^{60}Co	1173.23	0.03765	0.03723	0.03707	0.03622	0.03462	0.03318
	^{60}Co	1332.49	0.03470	0.03446	0.03416	0.03322	0.03295	0.03057
Ge-2	^{133}Ba	81.00	0.14859	0.14661	0.14490	0.12812	-	-
	^{133}Ba	302.85	0.07103	0.07062	0.06970	0.06627	-	-
	^{137}Cs	661.66	0.03142	0.03099	0.03024	0.02884	-	-
	^{60}Co	1173.23	0.01996	0.01968	0.01942	0.01830	-	-
	^{60}Co	1332.49	0.01784	0.01774	0.01759	0.01633	-	-
Ge-3	^{133}Ba	81.00	0.05680	0.05564	0.05052	-	-	-
	^{133}Ba	302.85	0.04393	0.04306	0.04062	-	-	-
	^{137}Cs	661.66	0.02556	0.02495	0.02372	-	-	-
	^{60}Co	1173.23	0.01587	0.01550	0.01508	-	-	-
	^{60}Co	1332.49	0.01442	0.01425	0.01368	-	-	-

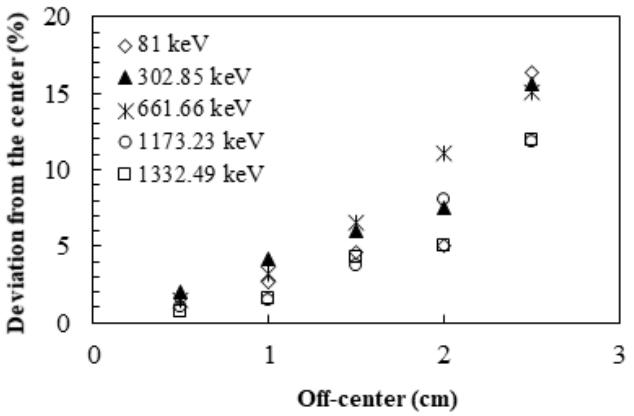

Figure 3. The relative difference between the measurements taken off-center in the Ge-1 detector compared to the measurement taken from the center.

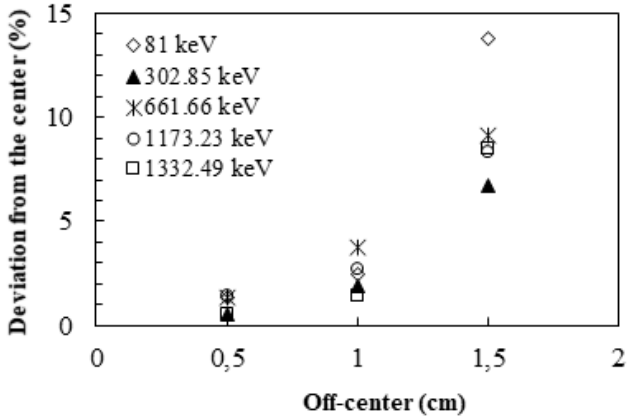

Figure 4. The relative difference between the measurements taken off-center in the Ge-2 detector compared to the measurement taken from the center.

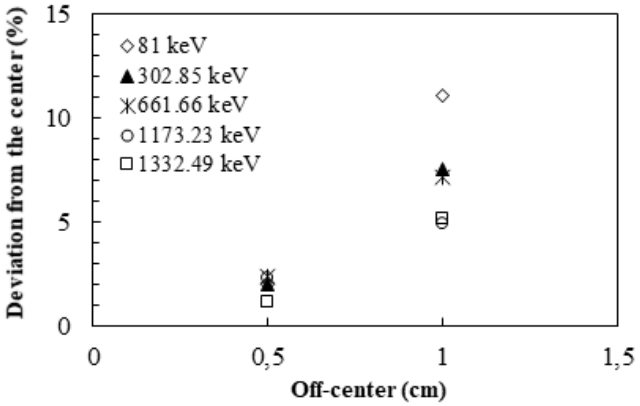

Figure 5. The relative difference of the measurements taken off-center in the Ge-3 detector compared to the measurement taken from the center.

3.2. Monte Carlo Results

The results from both the experimental and PHITS Monte Carlo simulation are given in the Table 3. In the table, the FEPE values at the farthest distances, where the position change is most effective, are compared with the FEPE values in the center. As it is clearly observed, there is a significant decrease in efficiency in all detectors. It can be suggested that the change in the Ge-1 detector, which has the largest crystal diameter, is slightly higher than the other detectors.

The relative bias between the experimental and simulation results according to the FEPE values at the center and off-center were calculated according to Equation 2.

$$RB = \frac{|FEPE_{Experimental} - FEPE_{Simulated}|}{FEPE_{Experimental}} \quad (2)$$

Table 3. The FEPE values from the experimental and simulation for the center and maximum distance from the center.

	Nuclide	Energy (keV)	Experimental FEPE			Simulated FEPE		
			In the center	Off-center*	% Diff.**	In the center	Off-center*	% Diff.**
Ge-1	^{133}Ba	81.00	0.07338	0.06137	16.4	0.07612	0.06313	17.1
	^{133}Ba	302.85	0.07853	0.06627	15.6	0.08093	0.06813	15.8
	^{137}Cs	661.66	0.05485	0.04659	15.0	0.05685	0.04818	15.3
	^{60}Co	1173.23	0.03765	0.03318	11.8	0.03918	0.03396	13.3
	^{60}Co	1332.49	0.03470	0.03057	11.9	0.03621	0.03184	12.1
Ge-2	^{133}Ba	81.00	0.14859	0.12812	13.8	0.15458	0.13256	14.2
	^{133}Ba	302.85	0.07103	0.06627	6.7	0.07405	0.06885	7.0
	^{137}Cs	661.66	0.03142	0.02884	8.2	0.03265	0.03005	8.0
	^{60}Co	1173.23	0.01996	0.01830	8.3	0.02085	0.01901	8.8
	^{60}Co	1332.49	0.01784	0.01633	8.5	0.01845	0.01703	7.7
Ge-3	^{133}Ba	81.00	0.05680	0.05052	11.0	0.05885	0.05259	10.6
	^{133}Ba	302.85	0.04393	0.04062	7.5	0.04586	0.04258	7.2
	^{137}Cs	661.66	0.02556	0.02372	7.2	0.02674	0.02452	8.3
	^{60}Co	1173.23	0.01587	0.01508	4.9	0.01664	0.01575	5.3
	^{60}Co	1332.49	0.01442	0.01368	5.1	0.01508	0.01415	6.2

* It is the FEPE value at 25 mm for Ge-1, 15 mm for Ge-2, and 10 mm for Ge-3.
** It is the percent difference between the measurement taken off-center and the measurement taken from the center

The mean relative bias between the two methods was similarly found to be around 3-5% for all detectors (Figure 6).

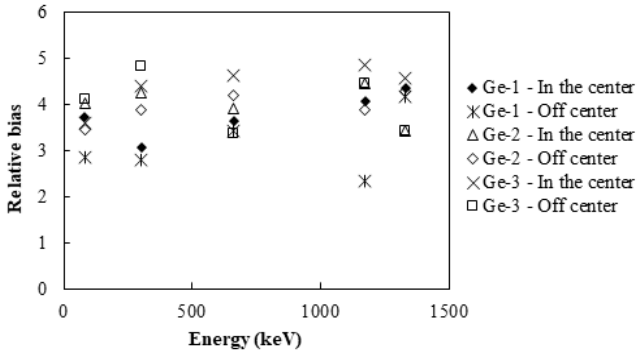

Figure 6. The relative bias between experimental and simulation results in the center and off center.

4. Conclusions

In this study, the effect of the change in the sample position on the efficiency of the samples with volumetric geometry placed on the detector end-cap was investigated. Compared to the values obtained from three different detectors and three different charcoal cartridge standards, it was observed that the sample position affected the efficiency at all energies between 81-1332.49 keV, being slightly more predominant at 81 keV. In gamma-ray spectrometric measurements, if possible, a sample holder that allows the sample to be counted in the center should be used. Since it will not be possible to provide a holder for every geometry, the samples must be carefully placed at the center of the detector and then measured. It is quite important that standard source used in the experimental efficiency calibration and the sample to be determined for its activity concentration are counted in the same position. The results show a reasonable association between the simulated values and the experimental value, with a maximum difference of 4.9 %. Therefore, sample position also has an effect on the combined uncertainty from many parameters (source-to-detector distance, sample height, source activity, efficiency, full energy peak count rate, etc.) in gamma-ray spectroscopy and must be added.

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