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Evaluation of HPGe Detector Long Term Performance for Environmental Radioactivity Measurements

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Abstract:

A gamma-emitting radionuclide is commonly measured qualitatively and quantitatively using a high-purity germanium (HPGe) detector. The precision of the measurement is determined by the HPGe detectors' performance. In this work, the long-term performance of the HPGe detector in the Central Laboratory for Radioactivity Measurements Inter-comparison and Training (CLERMET) is evaluated using statistical analysis. The minimal detectable activity (MDA) and the detection limit (LD) were computed. The Full Energy Peak Efficiency (FEPE) was calculated using a standard mixed gamma solution source and confirmed using samples from the International Atomic Energy Agency (IAEA) (PTs). The acquired data allowed for the determination of the resolution, peak shape, and peak to Compton ratio of the HPGe detector as a function of gamma-ray energy due to the long-term operation. FWHM, FWTM, FWFM and both ratios of FWTM/FWHM and FWFM/FWHM were determined at 1332 keV (60Co). The findings are used to discuss the detector's long-term performance stability. Even after a lengthy time of use, the results reveal that detector performance has no effect on the full energy peak efficiency (FEPE). The laboratory results were in good agreement with the target value based on the IAEA PTs, confirming the laboratory's dependability and traceability. For method validations, Quality Assurance and Quality Control are required to improve the reliability and accuracy of results as well as to demonstrate the laboratory's performance and trustworthiness.

Different analytical methods are developed for the determination of uranium isotopes in environmental samples using non-destructive and destructive analysis. Choice of an accurate and precise technique to get better performance and quality is essential for the results assessment and improvement. In this work different methods (Non-destructive method based on gamma spectrometer and destructive methods based on alpha spectrometer) were described to implement routine method for uranium isotopes determination in environmental samples. A set of IAEA reference certified samples were used for method verification.

Keywords: Detector stability, Efficiency, Resolution, HPGe Long-term performance, Minimum Detectable Activity (MDA)

1. Introduction

A nondestructive technology for determining the particular radioactivity concentration of natural and manufactured radionuclides is the high purity germanium detector (HPGe). The precision of the measurement is determined by the HPGe detector's performance parameters [1]. Laboratory circumstances, equipment, standards, and sample matrixes all influence performance [2]. Resolution, peak shape, peak-to-Compton ratio, and relative efficiency are all parameters that can affect the performance of an HPGe detector (Full Energy Peak Efficiency). During the detector's lifespan, these factors should be evaluated and validated on a regular basis to ensure precise and accurate measurements. [3].

The breadth of the peak at half its height is denoted as the peak resolution [4]. The photo-peak of 60Co is commonly given at 1332.5 kev. The detector's resolution may be harmed, resulting in the production of a low-energy tail [5]. The peak to Compton ratio is determined at numerous places below the peak to characterize the tail [6]. The Peak-to-Compton ratio [7] is a measurement of the detector's capacity to discriminate peaks from background radiation from the environment or surroundings, as well as electronic noise.

Another factor is the minimum detectable activity (MDA), which assesses the detector's capacity to quantify radiation levels slightly beyond any unavoidable background radiation [8]. The purpose of this study was to assess the performance of an HPGe detector over a lengthy period of time. The detector resolution, peak to Compton ratio, minimum detectable activity, and full energy peak efficiency of the detector were all calculated. The obtained data's statistical analysis was evaluated and discussed.

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2. Material and Method

Characterization of the detector

According to the manufacturer, the HPGe detector is an ORTEC N-type coaxial with a relative efficiency of 30% at 1332 keV (60Co). The detector's major performance specifications are as follows: Both at 1.33 MeV Co-60, the FWHM resolution is 1.9 keV, the peak-to-Compton ratio is 52:1, and the dead layer is 5 mm. -3500 V is the recommended bias voltage. A pre-amplifier (Model A257N) and the Genie-2000 software are used in this study's data gathering system. The detector is insulated with a thin Cu cylinder of 5 mm thickness to decrease background. An empty cylindrical container was used to determine the ambient background around the detector [9, 10, 11, 12]. The sample's counting time is 80,000s. The efficiency calibration was done with a standard mixed solution of 133Ba and 152Eu. The calibration spectra were collected over a period of 80,000s.

Determination of the Minimum Detectable Activity (MDA)

The detection limit (DL), critical limit (Lc), upper limit (Lu), and minimum detection limit (MDL) are all important statistical factors in Gamma spectrometry. The term "critical limit" refers to whether or not the net count is significant. The statistical significance of the count is determined by the upper limit. The crucial limit is the upper limit's threshold value [13]. The detection limit is the smallest number of counts that can be measured within a given level of confidence. The smallest quantity of activity concentration that is dependent on the counting time, full energy peak efficiency, emission probability, sample mass, and decay constant is defined as the Minimum Detectable Activity (MDA), expressed in Becquerel [14]. Both LD and MDA were calculated using the Curie Formula [15]:

L_D=2.71+3.29 (√(〖B+(N/2n)〗^2 (B_1+B_2))

MDA(Bq /kg)=(2.71+4.66√B)/(T_s 〖ε I〗_"γ" W)

Where B denotes background counts, TS is counting time in seconds, I denotes gamma emission probability, is the detector's absolute efficiency at a given gamma energy, and W denotes sample weight (kg).

Performance Statistical Analysis

The main parameters tested in this work were: energy resolution, peak shape, peak-to-Compton ratio, dead time and relative efficiency.

Energy Resolution

Peak width and peak efficiency are indicators of a detector's capacity to produce photon peaks [16]. Resolution is the width specified in keV as the Full Width at Half Maximum (FWHM). The following equation [17] can be used to manually calculate the resolution:

FWHM=ΔE/((C2- C1+1)) x Nr(keV)

Where:

ΔE is the difference between E2 (1332.5 keV) and E1 (1173.2 keV) known conversion factor,

C1&C2 are the peak position in terms of channel for 1332.5 keV and 1173.2 keV respectively,

Nr be the width of the reference peak in term of channel number, and

(1) is the uncertainty channel count.

At the same live moment, the multichannel analyser (MCA) recorded background count, gross area, count number, and channel number [15]. The Full Width at Half Maximum (FWHM) height, as well as the Full Width at One Tenth (1/10th) of the Peak's Maximum (FWTM) height and the Full Width at One Fiftyth (1/50th) of the Peak's Maximum (FWFM) height, were measured.

Peak Shape

The peak form is determined by calculating the ratio of FWTM over FWHM and the ratio of FWFM over FWHM [16]. For Gaussian peak, the optimal and acceptable resolution ratio is 1.82, and in practise, it should be less than 1.9. While the ideal and acceptable resolution ratio for FWFM/FWHM for Gaussian peak is 2.38, in practise it should be around 2.5 [15, 16].

3-Peak to Compton Ratio

The peak-to-Compton Ratio (P-to-C) is defined as the ratio of counts in the largest channel of the Co-60 energy peak at 1332.50 keV to the average channel count in the Compton continuum between 1040 and 1096 keV in the same spectrum [2]. The ratio was calculated using the information in the spectrum and ten thousand counts (at least 20,000 counts) in the photo-peak.

Peak to Compton ratio =(Highest peak count at 1332.5 keV)/(Average counts per channel between 1040 keV to 1096 keV)

4-Dead Time

The constant separation time of electronic circuits in the gamma-ray spectrometry system is defined as the counting system's dead time. The measuring approaches are based on the fact that the observed count rate differs from the true counting rate nonlinearly [17]. The photons that arrive at the detector during the dead time aren't counted. As a result, the count rate, which is defined as the number of counts per unit of time falls [18].Optimization necessitates careful equipment selection and understanding of the trade-off between resolution and count rate performance in every system [19]. The combination of detector and preamplifier is the most important component of the system electronics.

The resolution of the gamma-ray detector was tested against the dead time. The following equation [20] shows how the Multichannel Analyzer (MCA) system displayed dead time in percentage:

Dead Time $(\tau)=((RT - LT))/RT \times 100\%$

Where, RT is the real time and LT is the live time of the counting system.

3. Results

Determination of the Minimum Detectable Activity

To assess long-term performance, the collected spectra of the detector background were displayed. The only peaks found were at 609 and 1460 keV. Figures 1 and 2 show integral background counts for both the gamma peaks. The data obtained showed that the average limits were lower than $\pm 2\sigma$. Points falling between $\pm 2\sigma$ are considered to be satisfactory, the ones inside $\pm 3\sigma$ (Uper Count Limit) are warning and those exceeding $\pm 3\sigma$ indicate that a problem in measurement of the background has occurred [15].

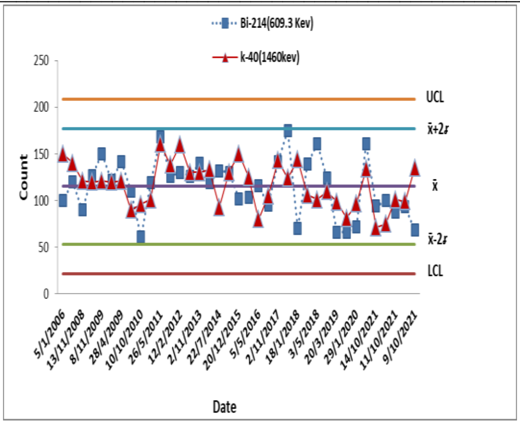

Figure1. Control Chart of Bi-214 (609keV) and K-40 (1460 keV) count rate

At a typical counting duration of 80,000s, the lower limit of detection (LD) in counts and the Minimum Detectable Activity (MDA) in Becquerel were obtained for a detected radionuclide, as given in Table1. MDA were computed with a 95 percent confidence level.

Table 1: Detection limit and Minimum Detectable Activity (MDA) of the HPGe gamma spectrometry system

	E(kev)	DL	MDA	B.R
Eu-152	121.78	2.71	0.002096	0.2856
	244.7	2.71	0.013771	0.0758
	344.28	2.71	0.005171	0.265
	778.9	2.71	0.020362	0.129
	964	2.71	0.021316	0.14605
	1085.7	2.71	0.033556	0.102
	1112	2.71	0.025576	0.1364
	1408.01	2.71	0.020051	0.21
Ba-133	81	2.71	0.001272	0.34
	276	2.71	0.016047	0.0716
	302.85	2.71	0.006873	0.18
	356.02	2.71	0.002268	0.6205
U-238 series	351	2.71	0.0037	0.376
	609	48.15	0.083222	0.461
	1120	2.71	0.023236	0.151
	1764	2.71	0.032722	0.154
	295	2.71	0.006278	0.193
Th-232 series	338	2.71	0.023324	0.0894
	583	2.71	0.00992	0.3
	911	2.71	0.012117	0.258
	969	2.71	0.025494	0.17
k-40	1460	46.9	0.043812	0.11
Am-241	60	2.71	0.010803	0.36
Na-22	1274.5	2.71	0.002146	0.9994
Cs-134	604	2.71	0.002736	0.976
	795	2.71	0.002699	0.855
Cs-137	662	2.71	0.004273	0.852
Co-60	1173	2.71	0.004032	0.999
	1332	2.71	0.004007	0.999

Full Energy Peak Efficiency (FEPE) Calibration of HPGe

The full energy peak (FEPE) efficiency for each photopeak at a given energy E is describing detection efficiency versus the γ-rays energy, Eγ [22]. The efficiency curve was plotted and fitted as shown in Fig. 1.

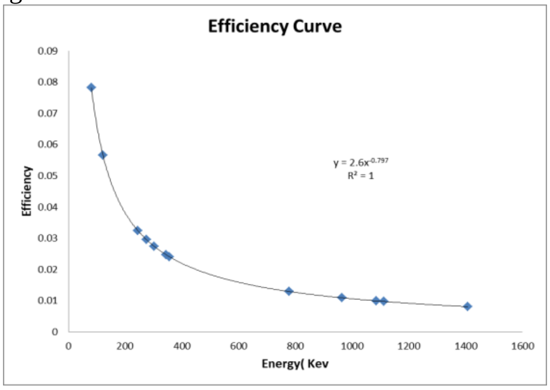

Figure 2: Experimental Full Energy Peak

Efficiencies Using Standard Solution

The genuine coincidence summing corrections for the HPGe detector were not taken into account in an efficiency calibration approach [23]. The determination of numerous gamma emitters in different sample matrices was used to ensure regular participation in proficiency examinations. The laboratory performance and data are considered satisfactory for z-score between -2 and +2. Z score values from 2006 till 2021 for different radionuclide in soil, sediment, water, and plant samples. The validation parameters for IAEA-PT 2018 soil sample are presented in Table 2. The proficiency tests' performance evaluations revealed that the laboratory results were in good agreement with the goal values. The results show that the laboratory measures were accurate and traceable.

Table 2: Validation Parameters for HPGe gamma spectrometer

IAEA-TEL2018-S04-SOIL SAMPLE							
Parent	E(Kev)	accuracy	BIAS	z-score	A1	A2	Precision
U-238 series	351	0.04	3.99	0.40	1.00	6.99	0.65
	609	0.02	2.27	0.23	0.57	6.93	0.65
	1120	0.16	15.65	1.56	3.91	7.41	0.65
	1764	0.14	13.58	1.36	3.39	7.33	0.65
	295	0.12	11.75	1.18	2.94	7.27	0.65
k-40	1460	0.10	9.52	0.95	35.60	90.16	0.49
Cs-134	604	-0.01	-0.61	-0.06	0.23	7.04	0.31
	795	-0.13	-13.01	-1.30	4.92	6.54	0.31
Cs-137	662	0.10	9.81	0.98	5.91	10.03	0.25
Co-60	1173	0.02	2.42	0.24	2.24	15.55	0.26
	1332	-0.01	-1.24	-0.12	1.14	15.14	0.26
Ba-133	276	0.04	4.07	0.41	1.87	7.27	0.25
	302.8	0.06	5.87	0.59	2.69	7.38	0.25
	356	0.06	6.24	0.62	2.86	7.40	0.25

Performance Statistical Analysis

Energy Resolution

FWHM, FWTM, FWFM and both ratios of FWTM/FWHM and FWFM/FWHM were determined at 1332 keV (60Co). FWTM/FWHM of the full-energy peak is a common parameter to evaluate the degree of asymmetry of the peak (tailing). The ratio of the FWTM and the FWHM were varied from 1.92 to 2.81. For a Gaussian of both coaxial n- and p-type HPGe-detectors at a photo peak of 1332.5 keV, the standard ratio values quoted for FWTM/FWHM is 1.82 and the FWFM/FWHM is 2.38 [21,22]. It was observed that the experimental data for the HPGe-detector was very close to the Gaussian shape for the FWTM/FWHM and FWFM/FWHM peak shape at the period from year 2006 to year 2013 as shown in table 2. These results agreed well with the energy resolution values indicated from the manufacture. While the Gaussian shape for the FWTM/FWHM and FWFM/FWHM peak shape at the year 2014 showed non-conformity. It was found that the reason was attributed to preamplifier troubleshooting. The corrective actions taken were the decrease of high voltage (from 3500 V to 2000 V). After that the system was run again, energy calibration was carried out, performance test was repeated, and acceptable data were showed ratios are bigger than that indicated from the manufacture. However, peaks of poor shape, having, for example, either a low or high energy tail can still give reasonable ratios. Peak shape with ratio should be regarded as a necessary but not a sufficient criterion [21]. The control chart for the FWHM for the two periods were presented in Figure 3 and Figure 4.

Peak to Compton Ratio %

The Peak-to-Compton ratio was determined, as shown in table 3, and its result was lower than the manufacturer's value (31.15: 1). Low Peak-to-Compton ratios might lead to peaks that aren't well defined (less counts can be accumulated over the same counting time). The capacity to measure low-energy peaks in the presence of Compton continuum from higher-energy gamma is referred to as big Peak-to-Compton [24].

Dead Time

Dead durations should be kept below 10% for system stability; in this investigation, the calculated dead time given in table 4 was within an average of 1.7 percent throughout long-term operation. The dependence of the dead time with the count rate for the 1173 keV peak, as supplied by the acquisition system, was employed [9].

Table 3: Performance statistical analysis of the HPGe gamma spectrometry system

Date	FWHM	FWTM	FWFM	FWTM/FWHM	FWFM/FWHM
1/5/2006	1.89	3.58	5.4	1.89	2.86
2/10/2007	1.89	3.6	5.5	1.89	2.89
2/6/2008	1.89	3.6	5.4	1.89	2.82
2/10/2009	1.89	3.6	5.55	1.90	2.94
2/10/2010	1.89	3.6	5.61	1.89	2.95
30/5/2011	1.89	3.6	5.8	1.91	3.06
15/10/2012	2.05	3.6	6.5	1.77	3.2
7/5/2013	2.04	3.6	6.6	1.76	3.22
24/6/2014	3.5	10.1	14.2	2.8	3.95
4/7/2016	3.57	10.5	14.5	2.9	4.1
10/8/2017	3.57	10.7	15.4	3.00	4.3
29/1/2018	3.8	10.76	15.4	2.8	4.04
17/3/2019	3.8	9.36	15.7	2.5	4.14
21/10/2020	3.8	10.5	16.2	2.76	4.24
26/7/2021	3.8	10.9	15.8	2.88	4.2

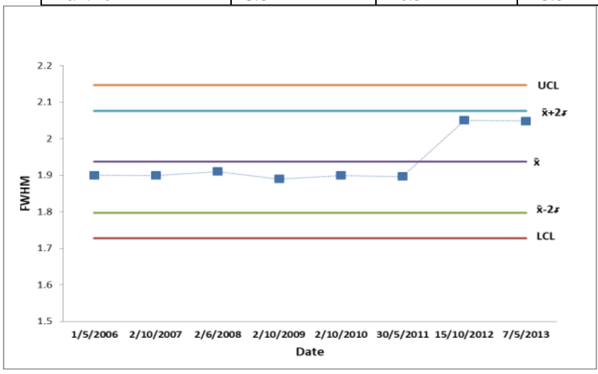

Figure 3: Control chart FWHM

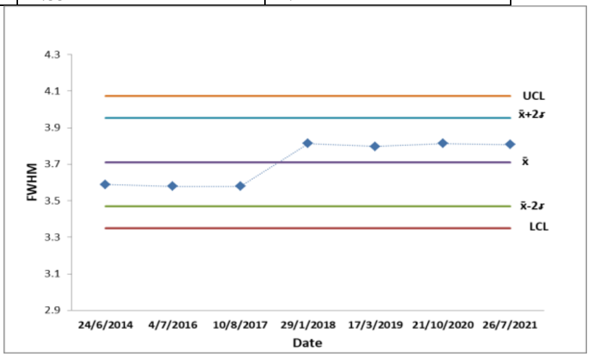

Figure 4: Control chart FWHM

Table 3: Peak to Compton ratio

date	max count at 1332.5	total count from 1040-1096 Kev =integrated	Total channel between 1040-1096 Kev	Average Count per channel	peak to Compton ratio
26/7/2021	**3041**	**42075**	**431**	**97.62181**	**31.15083**

Table 4: Dead time

Year	2011	2012	2013	2014	2016	2017	2018	2019	2020	2021
Dead Time	1.76	2.08	2.09	2.08	1.67	1.98	1.67	0.65	1.4	1.7

4. Conclusion:

A n-type HPGe detector's long-term behaviour was investigated. With a typical 60Co source, whose peaks are intense and cleanly differentiated, resolution, peak shape, and peak-to-Compton ratio were measured weekly. After seven years of operation, the detector's peak shape value for FWFM/FWHM was found to be 3.5. This implies that the detector's peak shape is larger than the typical values of 2.38 for both co-axial n- and p-type detectors, resulting in lower detector performance. Peak to Compton ratio is a measure of detector resolution as well as full-energy peak efficiency. The detector was also observed to have low peak-to-Compton ratio and this characteristic decrease the ability of the detector to distinguish peaks from the background influence because of radiation from environment or surrounding and electronic noise. Care should be taken when measure low-energy peaks in the presence of Compton continuum from higher-energy gamma

A set of IAEA worldwide proficiency test reference samples were used to validate the results and the calibration process. The acquired results revealed that all of the performance parameters were met, as well as the laboratory measurements' accuracy.

It can be determined that the detector is in good functioning order based on the findings of performance specifications. This suggests that the HPGe detector's counting efficiency has remained stable over the last fifteen years.

To increase the quality of radioactivity readings, all necessary factors should be investigated.

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