Assessment of calibration parameters for gamma-ray spectrometry systems

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Abstract The Gamma-ray spectrometers are complex instruments and the quality of the data they provide depends strongly on their proper calibration. This paper describes the energy and efficacy calibration of two gamma-ray spectrometers in order to validate the method for measuring the activity of gamma-ray emitting radionuclides. The punctiform standard sources ⁶⁰Co, ¹³³Ba, ¹³⁷Cs and ²⁴¹Am were used in this study. Minimum Detectable Activity, Peak to Compton Ratio and the peak shape parameters were determinate. An inter-comparison of the results obtained with both systems was done. The results demonstrated that the activity values are comparable with low errors respecting with those for convention-ally true activities.

Keywords Energy calibration · Efficiency calibration · Gamma spectrometry · HPGe gamma ray detector

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Introduction

The methods based on gamma-ray measurements are frequently applied for the determination of activity or for nuclide identification. For precise determinations these methods must be validate in conformity to national, regional or international standards or in the absence of the above on the base of validated calibration methods [1, 2].

The validation of the measurement method is based on the determination of the performance parameters of the global method. These parameters are obtained during the development of the method and the inter-laboratory study, or by obeying internal validation protocols [3].

As it is known, the calibration of a gamma-ray spectrometry system evolves three main aspects:

- (1) The energy as a function of the number of the channel;
- (2) FWHM as a function of the number of the channel;
- (3) The efficiency as a function of the energy correlated with the acquisition geometry of the radiation spectrum.

In addition to these aspects, in the case of applied geometries the correlation with the coincidental standard (TCC) can be made. The energy as a function of the number of the channel and the FWHM as a function of the number of the channel do not interfere with the initial calibration in efficiency of the measuring system [4].

This paper presents the energy and efficiency calibration of gamma-ray spectrometry systems with HPGe detector on the base of internal procedures of laboratories.

The measurements and the studies were conducted on two ORTEC, gamma-ray spectrometry systems in two accredited laboratories: SALMROM (*Life Science and Environmental Physics Dept.*) and LAS, (*Radioactive*) Waste Management Dept.) from the "Horia Hulubei"— National Institute for Physics and Nuclear Engineering, Bucharest (IFIN-HH), Romania.

The gamma-ray spectrometry systems

Two gamma-ray spectrometry systems have been used in this study. System 1 (ORTEC, United States) consist in a cooled GEM HPGe detector, 59.1 mm diameter and 54.1 mm length, with a Pop Top vertical cryostat configuration. The detector associated electronics is all digitally implemented. A detector interface module (DIM-ORTEC) includes the high voltage power supply, controlled by a high performance, portable multichannel analyzer, DigiDART and assures the connections for preamplifier. The DigiDART is fully integrated with software Gamma Vision 32 v-6 which can download setup parameters of the high voltage power supply, spectroscopy amplifier and analog digital converter.

The energy range of the HPGe spectrometric system is $50 \div 3000 \text{ keV}$ and the operational parameters of the detector measured for a shaping time of 6 µs and high voltage of +4400 V are: relative efficiency: 30% at 1332 keV (⁶⁰Co); 1.85 keV resolution at 1332 keV (⁶⁰Co) and 0.85 keV resolution at 122 keV (⁵⁷Co) [5].

System 2 (ORTEC, United States) consist in a HPGe germanium detector model GEM40P4 with a diameter of 69.5 mm and a length of 48.7 mm, HV = +4000 V; multichannel analyzer DigiDART, acquisition and data transformation software Vision 32 v-6.01. The energetic interval of the gamma-ray spectrometry system with GeHP detector is 50–2000 keV, and the installation parameters are: HPGe parameters (supplier specifications) obtained with a time constant of 12 µs, relative efficacy 40% at 1332 keV (⁶⁰Co), 1.85 keV resolution at 1332 keV (⁶⁰Co); 0.87 keV la 122 keV (⁵⁷Co) [5].

Results and discussions

System calibration

Energy and resolution calibration

The energy calibration of the System 1 was performed in the energy range of interest, $50 \div 3000$ keV, using certified spectrometric etalon sources, namely, ¹³³Ba, ¹³⁷Cs, ⁶⁰Co, ¹⁵²Eu, ²²Na, ²⁴¹Am. The energetic lines of the natural radionuclides, ⁴⁰K and ²⁰⁸Tl, of 1460 keV, respectively, of 2614 keV, have been also considered for a more accurate energy calibration in the work energy interval.

The settings of the System 1 parameters made for the HPGe system calibration were: HV(+) = 4400 V; Gain = 2.2;

Table 1 Peak position of energy lines of calibration sources

Radionuclide/laboratory	Energy (k	eV)	Channel	
	System 1	System 2	System 1	System 2
²⁴¹ Am	59.54	59.54	363.53	236.94
¹³³ Ba	80.99	80.99	495.14	322.38
¹⁵² Eu	121.78	-	744.81	_
¹⁵² Eu	244.7	-	1494.82	-
¹⁵² Eu	276.29	-	1688.46	_
¹³³ Ba	302.71	355.86	1850.33	1420.84
¹⁵² Eu	344.3		2102.92	
¹³³ Ba	355.86	383.70	2174.90	1531.80
¹³⁷ Cs	661.62	661.62	4041.97	2641.38
¹⁵² Eu	778.9	-	4758.66	_
¹⁵² Eu	867.37	-	5299.49	_
⁶⁰ Co	1173.23	1173.23	7168.42	4684.15
²² Na	1274.54	-	7789.12	_
¹⁵² Eu	1299.14	-	7937.78	_
⁶⁰ Co	1332.51	1332.51	8141.7	5320.45
¹⁵² Eu	1408.01	-	8603.12	_
⁴⁰ K	1460	1460	8929.33	5832.73
²⁰⁸ Tl	2614	-	15987.14	-

Rise time: 6 μ s; Flat Top: Width = 0.8, Tilt = -0.0396; ADC Conversion Gain = 16384; LLD = 100; ULD = 16383. From the height amplitude spectra of the etalon sources were obtained the peak positions of their energy lines mentioned in Table 1.

The energy calibration of the HPGe detector of the System 2 has been made in the energetic range of $50 \div 2000$ keV with standard punctiform sources of 60 Co, 133 Ba, 137 Cs, 241 Am taking into consideration the energetic line of 40 K of 1460 keV. The parameters of the system and their settings were: HV(+) = 4000 V; Coarse Gain = ×4, Fine Gain = 0.7645 formation time constant = 12 µs ADC Conversion Gain = 8192; Digital offset = Automatic; LLD = 180; ULD = 8190.

The energy calibration of the System 1 has been made in the mentioned experimental conditions by measuring on 18 channels, and for the System 2 only eight channels were measured (Table 1).

Taking into account these results from Table 1, the energy calibration curve has been obtained, being described by the following 2° polynomial function [6, 7]:

Energy (keV) = $a_0 + a_1 \times \text{channel} + a_2 \times (\text{channel})^2$

where the coefficients, a_i bear the values: $a_0 = -0.291$, $a_1 = 0.163$, $a_2 = 2.155 \times 10^{-9}$, for the System 1 and $a_0 = 0.240$, $a_1 = 0.250$, $a_2 = -1.086 \times 10^{-9}$ for System 2. The energy calibration curves obtained for the two gamma-ray spectrometry systems are presented in Fig. 1.



Fig. 1 The energy calibration curve of the gamma-ray spectrometry systems

The resolution calibration of both systems have been conducted with calibration sources of ⁵⁷Co, ¹³⁷Cs, ⁶⁰Co, ¹³³Ba, ²⁴¹Am, and the values of efficiencies obtained for the two detectors are presented in Table 2.

The resolution (FWHM) calibration curve is also described, by a 2° polynomial function, as follows [6, 7]:

 $FWHM(keV) = a_0 + a_1 \times canal + a_2 \times (canal)^2$

where the coefficients a_i bear the values $a_0 = 4.7361$, $a_1 = 0.000653$, $a_2 = -4.2255 \times 10^{-9}$ for the System 1 and $a_0 = -0.2912$, $a_1 = 0.163542$, $a_2 = 2.15548 \times 10^{-9}$ for the System 2. These are the coefficients of the fitting curve, represented in Fig. 2.

Efficiency calibration

The efficiency calibration of the System 1 has been done using the same certified spectrometric etalon sources, ²⁴¹Am, ¹³³Ba, ¹³⁷Cs, ⁶⁰Co, ¹⁵²Eu, ²²Na. The efficiency values were determined using the amplitude spectra acquired for every radioactive source, namely, from the net area in the region of interest of the energy peak of interest, divided by the activity of the sources, acquisition time and intensity of the energy line. The standard radioactive



Fig. 2 The resolution calibration curve

sources were positioned at 11 cm distance from the end cap of the detector, using a special distance piece (Table 3).

These results emphasize the capability of the System 1 to measure complex spectra and to identify radionuclides with very close energy lines, in the limits of the values calculated for the resolutions. The efficiency calibration of the System 2 has been conducted with punctiform sources at a distance of 30 cm from the detector. The efficiency values are presented in Table 4.

The efficiency fitting curve is described by a 6-term polynomial function to fit the natural logarithm of efficiency to the energy [8]:

$$\operatorname{Ln}(\varepsilon) = \sum_{i=0}^{5} a_i E^{2-i}$$

where ε is the efficiency at energy E (MeV) and a_i is the fitting coefficients.

Using the determinated values of the fitting coefficients from the System 1: $a_0 = -0.3821104$, $a_1 = -6.343216$, $a_2 = 0.5304888$, $a_3 = -0.05512359$, $a_4 = 0.002084932$, $a_5 = -0.3127718 \times 10^{-4}$ and the System 2: $a_0 = -0.412965$, $a_1 = -4.983256$, $a_2 = 0.782049$, $a_3 = -0.0712944$, $a_4 = 0.0120845$,

Table 2 Measured values of the resolution

Radionuclide/laboratory	Energy (ke	V)	FWHM (keV) max. warranted (certificate)		FWHM (keV) measured	
	System 1	System 2	System 1	System 2	System 1	System 2
²⁴¹ Am	_	59.54	Not specified		_	0.7019
¹³³ Ba	-	80.99	Not specified		-	0.7170
⁵⁷ Co	122	_	0.85	-	0.65	-
¹³³ Ba	-	355.86	Not specified		-	1.0320
¹³³ Ba	-	383.70	Not specified		-	1.1527
¹³⁷ Cs	661.62	661.62	Not specified		1.23	1.2548
⁶⁰ Co	1332.86	1332.51	1.85	1.85	1.63	1.5294
⁴⁰ K		1460	Not specified		-	1.7933

Table 3 Efficiency measurements-System 1

Radionuclide	Energy (keV)	Efficiency $(s^{-1} \times \text{GPS}^{-1})$	Efficiency (fitting curve) $(s^{-1} \times \text{GPS}^{-1})$
¹³³ Ba	30.97	2.729×10^{-5}	2.723×10^{-5}
¹³⁷ Cs	32.19	6.166×10^{-5}	6.148×10^{-5}
¹³³ Ba	35	2.152×10^{-4}	2.219×10^{-4}
¹³⁷ Cs	36.4	3.472×10^{-4}	3.444×10^{-4}
²⁴¹ Am	59.54	4.849×10^{-3}	4.845×10^{-3}
¹³³ Ba	79.52	8.563×10^{-3}	8.561×10^{-3}
¹³³ Ba	80.99	7.96×10^{-3}	7.98×10^{-3}
¹⁵² Eu	121.78	8.683×10^{-3}	8.753×10^{-3}
¹⁵² Eu	244.7	6.326×10^{-3}	6.372×10^{-3}
¹⁵² Eu	276.29	5.727×10^{-3}	5.757×10^{-3}
¹³³ Ba	302.71	5.262×10^{-3}	5.319×10^{-3}
¹³³ Ba	355.86	4.659×10^{-3}	4.61×10^{-3}
¹⁵² Eu	383.7	4.445×10^{-3}	4.313×10^{-3}
¹⁵² Eu	443.96	3.878×10^{-3}	3.793×10^{-3}
¹⁵² Eu	488.68	3.567×10^{-3}	3.489×10^{-3}
¹³⁷ Cs	661.62	2.637×10^{-3}	2.704×10^{-3}
¹⁵² Eu	688.67	2.728×10^{-3}	2.616×10^{-3}
¹⁵² Eu	867.37	2.166×10^{-3}	2.169×10^{-3}
⁶⁰ Co	1173.23	1.712×10^{-3}	1.698×10^{-3}
²² Na	1274.54	1.539×10^{-3}	1.585×10^{-3}
¹⁵² Eu	1299.14	1.621×10^{-3}	1.56×10^{-3}
⁶⁰ Co	1332.51	1.540×10^{-3}	1.527×10^{-3}
¹⁵² Eu	1408.01	1.505×10^{-3}	1.456×10^{-3}
¹⁵² Eu	1528.1	1.258×10^{-3}	1.354×10^{-3}

Table 4 Efficiency measurements—System 2

Energy (keV)	Efficiency $(s^{-1} \times \text{GPS}^{-1})$	Efficiency (fitting curve) $(s^{-1} \times \text{GPS}^{-1})$
59.54	1.2469×10^{-3}	1.2469×10^{-3}
88.03	2.1043×10^{-3}	2.1063×10^{-3}
122.07	2.2529×10^{-3}	2.2466×10^{-3}
165.85	2.1287×10^{-3}	2.1356×10^{-3}
279.17	1.5752×10^{-3}	1.583×10^{-3}
391.69	1.1779×10^{-3}	1.1854×10^{-3}
661.66	7.6387×10^{-4}	7.5950×10^{-4}
898.02	5.9550×10^{-4}	5.9348×10^{-4}
1173.24	4.8751×10^{-4}	4.8088×10^{-4}
1332.50	4.2480×10^{-4}	4.3493×10^{-4}
1836.01	3.3686×10^{-4}	3.3434×10^{-4}

 $a_5 = -0.5723974 \times 10^{-4}$, we obtained the in efficiency calibration curves presented in Fig. 3.

The relative errors of the measuring interval obtained after the efficiency calibration are presented in Table 5, taking into account the measuring conditions: the dead time correction made by the system, measurements of the calibration defined geometry, the used sources are radioactive punctiform standard sources, with the characteristics described in the calibration certificates.

Method validation

The activity measurement of punctiform standard sources

For the method validation, gamma-ray spectrometric measurements were carried out using standard sources such as ⁶⁰Co, ¹³³Ba, ¹³⁷Cs and ²⁴¹Am. Minimum Detectable Activity (MDA), the stability and repeatability of the energetic resolution, Peak to Compton Ratio and the peak shape parameters were determinate.

The activities measured for the punctiform standard sources are presented in Table 5.

Note that the values of the activity of measured sources with both systems are in close connection with the true conventional values, the relative measurement errors being situated within tenths of percent.

Minimum detectable activity

The MDA for the two systems (Table 6) was determined for 137 Cs and 60 Co using the formula:

MDA (Bq) =
$$(2.71 + 4.65\sqrt{B})/Eff. \times P_{\gamma} \times t_m$$

where B is the background in the energy region of interest; t_m (s) the measuring time; Eff. (cps/Bq) the peak efficiency; and P_{γ} is the radiation intensity.

The results obtained by the two gamma-ray spectrometry systems were very good and in concordance with the standard sources used.

The stability and repeatability of the energetic resolution

The stability of the energetic resolution (energetic repeatability) for the 122 keV peak of the 57 Co source and for the 1.33 MeV peak of the 60 Co source was monitored with System 1.

In addition 10 measurements for each measuring were made with an acquisition time of 1,800 s for each measurement without modifications of the parameters. The average calculated values for the resolutions of the gamma-ray spectrometry systems for the 60 Co and 57 Co are 1.674 keV at the 1.33 MeV line and 0.808 keV at the 122 keV line. These values are lower than the maximum values guaranteed by the producer and mentioned in the technical documentation/specification of the HPGe detector for the same energy lines: 1.85 keV at the 1.33 MeV line of 60 Co and 0.85 keV at the 122 keV line of 57 Co. (Table 7).

The time stability of the peaks position has been determined with System 2 for the 137 Cs peak by repeated



Fig. 3 The efficiency calibration curve of the gamma-ray spectrometry systems

measurements on a period of 6 h and a measuring time of 600 s without ups or downs of the high tension on the detector (Table 8).

Peak to Compton Ratio and the peak shape parameters

Other parameters of the HPGe system, determined as part of the validation method, were the Peak to Compton Ratio and the peak shape parameters FWTM/FWHM and FWFM/FWHM, [9]. These parameters have acquired, using a certified standard point source of ⁶⁰Co.

The result of 67:1, for the Peak to Compton Ratio calculated using the values from Table 8, confirm the value of 67:1, calculated and declared in the HPGe detector technical documentation (Table 9).

 Table 7 Repeatability measurements—System 1

Measurement	Resolution (keV)	
	⁵⁷ Co	⁶⁰ Co
1	0.82	1.66
2	0.81	1.66
3	0.81	1.68
4	0.81	1.67
5	0.81	1.71
6	0.80	1.65
7	0.81	1.70
8	0.80	1.67
9	0.80	1.67
10	0.81	1.67
Average value	0.808	1.674

 Table 8 Repeatability measurements—System 2

Measurement	Background (imp)	Centroid peak (keV)
1	92465	661.61
2	93069	661.62
3	91534	661.62
4	91227	661.63
5	91450	661.63
6	90985	661.62
7	91883	661.63
Average value	91802 ± 735	661.62 ± 0.01

This result emphasizes another important characteristic of the HPGe spectrometric system, namely, that, to distinguish low energy lines in the presence of high energy sources.

Table 5 Measurements of radioactive standard sources

Radionuclide Source type Conventionally tr	Source type	Conventionally true activity (Bq)	Measured activity (Bq)		Relative error (%)	
		System 1	System 2	System 1	System 2	
¹³⁷ Cs	SEG 7-737	13027	13287	13308	+2.10	+2.16
⁶⁰ Co	SEG 7-834	10892	10946	10949	+0.50	+0.50
¹³³ Ba	SEG 7-864	16099	16204	16199	+065	+0.60
²⁴¹ Am	SEG 7-882	9872	9898	9751	-0.20	+1.20

Table 6 The Minimum Detectable Activity (MDA) of the systems

Radionuclide Energy (keV	Energy (keV)	rgy (keV) Background (s ⁻¹)		Efficiency (cps/GPS)		AMD (Bq)	
		System 1	System 2	System 1	System 2	System 1	System 2
¹³⁷ Cs	662	0.217	0.229	2.704×10^{-3}	7.6387×10^{-4}	13.20	12.09
⁶⁰ Co	1332.86	0.220	0.113	1.698×10^{-3}	4.2480×10^{-4}	11.80	13.00

 Table 9
 Peak to Compton Ratio

Source type	Radionuclide	Peak value ⁶⁰ Co (1.33 MeV) (counts)	Average area in the range 1040–1096 keV (counts)	Peak to Compton Ratio
SEG 7-834	⁶⁰ Co	2727	40.08	67:1

Table 10 Peak shape parameters

Source type	Radionuclide	FWHM (channels)	FWFM (channels)	FWTM (channels)	FWFM/ FWHM ratio	FWTM/ FWHM ratio
SEG 7-834	⁶⁰ Co	10.03	15.83	19.25	1.6	1.9

Table 11 Inter-comparison results

Source type	e Radionuclide Conventionally true activity (Bq) Meas	Measured act	Measured activity (Bq) Relative		ive error (%)		
		System 2	System 1	System 2	System 1	System 2	System 1
SEG 7-737	¹³⁷ Cs	13024	13027	13232	13301	1.7	2.1
SEG 7-834	⁶⁰ Co	10884	10892	11162	10946	2.5	0.5

The values calculated for the peak shape parameters, FWFM/FWHM and FWTM/FWHM, of 1.6 and 1.9, respectively, are less than those warranted in the HPGe detector technical documentation, namely, 1.9 and 2.43, respectively (Table 10) [5].

Inter-laboratory comparisons

An important part of the measurement validation was the inter-comparison of the results obtained with both systems. The results obtained are presented in Table 11, indicating comparable activity values, with low errors respecting with those for conventionally true activities.

Conclusions

The validation of the results and of the calibration method was made in a series of measurements to check the values of the parameters declared in the HPGe detector technical documentation, using certified standard point sources and respecting the initial measuring conditions. The results obtained validate the measuring method, emphasizing the fact that the new gamma spectrometric system with HPGe detector is correctly calibrated in energy and efficiency, respecting the measuring geometry.

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