

CALIBRATION OF THE HIGH AND LOW RESOLUTION GAMMA-RAY SPECTROMETERS*

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Abstract. Many laboratories accredited according to the requirements of the standard EN ISO/IEC 17025:2005 “General requirements for the competence of testing and calibration laboratories” are implied in the radioactive content measurement of samples collected from the environment, food chain or industrial products. The most used method is the gamma-ray spectrometry measurement, using high resolution HPGe and low resolution NaI(Tl) detectors. In order to improve its sensitivity, many technical and methodological aspects are important: careful choice of detector material and shielding, samples matrix, measurement of high volume samples in close to detector geometries. The results are strongly influenced by the geometry, sample volume and content, and true coincidence summing effects (for cascade gamma-ray emitters), as demonstrated during many proficiency tests. The Radionuclide Metrology Laboratory from IFIN-HH, Primary Activity Standard Laboratory, assures the continuity of the whole metrological traceability chain of activity measurement and is accredited by the national accreditation body RENAR as a Calibration and Testing Laboratory, offering products and services for clients. Within the frame of its QMS, RML elaborated two working procedures for the calibration of the gamma-ray spectrometers and the low level radioactive samples activity measurement. The calibration procedures are described, emphasizing the corrections applicable by the measurement laboratories.

Key words: Gamma-ray spectrometry, calibration, activity measurements.

1. INTRODUCTION

During recent years, many laboratories from Romania were accredited according to the standard EN ISO/IEC 17025:2005 “General requirements for the competence of testing and calibration laboratories”. These laboratories are directly involved in the measurement and certification of the radioactive content of various samples collected from the environment and food chain, or industrial products [1]. For this aim, the most frequently used analysis method is the gamma-ray spectrometry

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method using high resolution HPGe and low resolution NaI(Tl) or equivalent detectors [2]. The method has the advantage of being non-destructive, but for large quantities of materials to be investigated a sampling procedure is necessary.

In order to perform accurate and reliable activity measurements, periodical metrological calibrations of the gamma-ray spectrometry installations of the laboratories are required. The Radionuclide Metrology Laboratory (RML) from IFIN-HH, Primary Activity Standard Laboratory, assures the continuity of the whole metrological traceability chain of activity measurement. Since 2009, the RML is accredited by the national accreditation body RENAR as a Calibration and Testing Laboratory according to the EN ISO/IEC 17025:2005 (www.renar.ro), for products and services offered to the customers. Based on its previous experience in this field, including the production of radioactive standard sources [3, 4] the RML has recently elaborated two working procedures specific for the calibration of the gamma-ray spectrometry systems. These procedures, completed by a framework and other installations calibration working procedures were integrated in the RML Quality Management System (QMS).

This paper gives an overview of the calibration procedures for gamma-ray spectrometry installations, with emphasis on the corrections to be applied by the testing laboratories. Practical examples are presented and discussed.

2. THE GAMMA-RAY SPECTROMETRY METHOD

In accredited laboratories, the gamma-ray spectrometry method is used to perform both qualitative and quantitative radioactivity analysis, for solid, liquid and gaseous samples. A typical gamma-ray spectrometry system is composed of: a detector (usually semiconductor, such as HPGe, or scintillator, for example NaI(Tl) or plastic scintillator) with a shielding – mainly lead, to reduce the background; high voltage power supply; electronics for signal processing (preamplifier, amplifier, multichannel analyzer); computer and dedicated software. The spectrometric system records, stores and processes the gamma-ray spectrum of the analyzed sample, using validated computer software packages [5, 6].

A proper energy calibration is needed to identify the energy of the gamma-ray emissions from the spectrum, *i.e.* the gamma-ray emitter radionuclides contained by the sample (qualitative analysis). The quantitative analysis, *i.e.* the activity (expressed in Becquerel, SI units) and its standard uncertainty determination – for each radionuclide present in the sample, requires a full-energy peak (FEP) efficiency calibration. For both calibrations, various radioactive standard sources with certified activity are necessary. There are many technical and methodological aspects to be taken into account for an optimal radioactivity analysis:

- careful choice of the detector (type, geometry, window) and the radioactive standard sources used for calibration;
- sample geometry, matrix, position relative to the detector;

- passive or active detector shielding;
- various corrections: background, dead time, geometry (sample different from the standard), deconvolution of overlapped spectrum peaks, true coincidence summing (TCS).

The high resolution detectors (HPGe) are suitable for samples containing many radionuclides (*e.g.* from the natural radioactive series), when the gamma-ray spectrum presents a large number of peaks to be resolved. These detectors are expensive and must be kept at very low temperatures (in liquid nitrogen) for a correct functioning. The spectrometry systems based on NaI(Tl) detectors perform faster analysis (due to the high efficiency) and are cheaper, but have the disadvantage of a lower resolution; special attention is needed in defining the spectrum windows for measuring the radionuclide activity.

3. CALIBRATION PROCEDURES CONTENT

The RML elaborated two working procedures for the calibration of the gamma-ray spectrometry equipments.

One working procedure is dedicated for the HPGe detectors and another for NaI(Tl) detectors. The calibrations must be performed in the customer's laboratory, because the delicate electronic components could be damaged during the transport. Another working procedure establishes the framework of the calibration procedures, when the calibration is performed in the customer's laboratory: responsibilities, preliminary conditions, pre- and post-calibration instructions, forms and reports to be completed.

The most important preliminary conditions are:

- the availability of Γ -ray radioactive standard sources containing the necessary radionuclides for the calibration, in similar geometry and density with the samples to be measured;
- the customer's Γ -ray spectrometry installation with shielding and validated software should work normally, in preset conditions, with no radioactive contamination and all the user's manuals available;
- the client must have professionally trained staff to operate the gamma-ray spectrometry equipments.

3.1. ENERGY AND RESOLUTION CALIBRATION

The energy calibration consists in the experimental determination of a function, usually a first or second degree polynomial, describing the energy dependence of the channel number in the spectrum:

$$E_{\gamma} = A + B \cdot Ch + C \cdot Ch^2, \quad (1)$$

where E_γ is the gamma-ray energy, Ch is the spectral channel number for the center of the peak corresponding to E_γ (usually the channel with the maximum number of counts), A , B and C are constants to be determined for calibration.

The energy calibration is performed manually or automatically by the software, by measuring one or several radioactive standard sources emitting gamma-rays of minimum 3–4 different energies covering a wide spectral range between 50 keV and 2000 keV. The energy values adopted are the Bureau International des Poids et Mesures (BIPM) recommended data [7]. Figure 1 presents the energy calibration points of a NaI(Tl) detector from a customer laboratory, obtained using standard sources (^{134}Cs , ^{137}Cs).

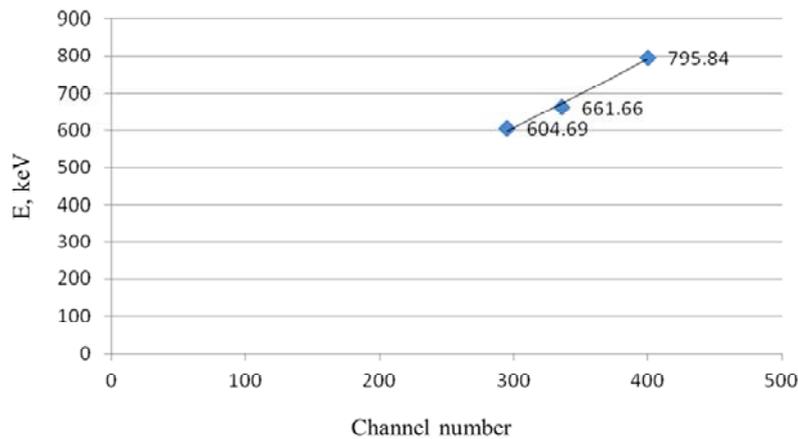


Fig. 1 – The energy calibration of a NaI(Tl) detector (linear function), using standard sources of gamma-rays emitters ^{134}Cs (604.69 keV and 795.84 keV) and ^{137}Cs (661.66 keV).

The resolution calibration, also called FWHM (Full-Width at Half-Maximum) calibration, establishes a function to describe the peak width *versus* the spectral energy. It is an important parameter characterizing the system performance in separating different photon emissions in a narrow energy range. Table 1 compares the FWHM experimental values for two calibrated detectors HPGe and NaI(Tl) type, illustrating the reason why HPGe are considered “high resolution” detectors, while NaI(Tl) are “low resolution” detectors.

Table 1

Comparison of the experimental FWHM values between two detectors of HPGe and NaI(Tl) type

Radionuclide	Energy [keV]	FWHM [keV]	
		HPGe detector	NaI(Tl) detector
^{57}Co	122.06	1.42	16
^{137}Cs	661.66	1.84	47
^{60}Co	1332.49	2.15	69

3.2. EFFICIENCY CALIBRATION

The efficiency calibration consists in determining a function describing the full energy peak (FEP) detection efficiency *versus* the γ -rays energy, E_γ . It is usually a polynomial with the degree higher than 2. Radioactive standard sources with gamma-ray emissions covering a wide energy range (containing several monoenergetic or multienergetic radionuclides such as ^{152}Eu , or mixtures of several radionuclides) are used for this purpose.

For a given peak corresponding to the energy E_γ in the gamma-ray spectrum measured with a NaI(Tl) or HPGe detector, the efficiency ε and its expanded standard uncertainty can be computed as follows [1]:

$$\varepsilon = \frac{(A - A_F) \cdot F_D}{t \cdot I \cdot \Lambda \cdot F_C \cdot F_T}, \quad (2)$$

$$U_\varepsilon = k \cdot u_\varepsilon = k \cdot \sqrt{s_{A-A_F}^2 + u_I^2 + u_\Lambda^2 + u_{F_C}^2 + u_{F_D}^2 + u_{F_T}^2}, \quad (3)$$

where A and A_F are the net areas of the considered peak from the gamma-ray spectrum of the standard, respectively the same peak from the installation background spectrum; t is the measurement time, the same for standard and background, expressed in seconds; F_C , F_T and F_D are the multiplicative coefficients for coincidence summing corrections, efficiency transfer corrections and for the decay during the reference time and the time of the sample measurement start (important only for short half-life radionuclides); I is the emission probability of the considered gamma-rays; Λ is the activity of the standard, expressed in Becquerel units [Bq]. In Eq. (3), k is the coverage factor, u_ε is the efficiency combined standard uncertainty, s_{A-A_F} is the result of the type A evaluation of the peak net area measurement uncertainty, while u_I , u_Λ , u_{F_C} , u_{F_T} , and u_{F_D} are the results of the type B evaluations of measurement uncertainties for the emission probability, activity and correction factors for coincidence summing, efficiency transfer, respectively decay during the reference time and the time of the measurement start. The standard uncertainty of the measurement time and the corresponding decay are considered negligible.

As usually the investigated materials present low activity values, the samples are measured directly on top the detector and the true coincidence summing (TCS) corrections are very important for multienergetic radionuclides emitting cascade gamma-rays, as for example: ^{60}Co , ^{134}Cs , ^{152}Eu [2]; the effect can be amplified in volume sources by comparison with point ones. In the case of the NaI(Tl) detectors used in the environmental and food monitoring national networks, the radionuclides to be analyzed in samples are ^{134}Cs and ^{137}Cs , and the TCS corrections are avoided using standards containing the same radionuclides. For high resolution analysis with HPGe detectors, the TCS corrections are very

important. One example of efficiency calibration curve *versus* energy performed in the RML for a HPGe detector (coaxial, p-type, crystal diameter 60.5 mm and length 50.5 mm), with the source placed directly on the top of detector, including the TCS corrective factor, is presented in Table 2.

Table 2

Experimental efficiency calibration with its standard combined uncertainty and the true coincidence summing corrections factor for a HPGe detector and volume radioactive standard source in soil matrix

Radionuclide	Energy [keV]	Efficiency $\times 10^2$		True coincidence summing corrections factor (F_C)
		Value	Standard combined uncertainty	
Am-241	59.54	0.781	0.040	1
Ba-133	80.89	2.32	0.19	0.903
Eu-152	121.78	3.98	0.22	0.915
Eu-152	244.70	3.24	0.18	0.886
Ba-133	276.4	3.05	0.21	0.951
Ba-133	302.85	2.80	0.19	0.976
Eu-152	344.28	2.55	0.14	0.942
Ba-133	356.01	2.48	0.17	0.978
Ba-133	383.84	2.30	0.16	1.038
Eu-152	411.12	2.28	0.14	0.865
Eu-152	443.96	2.10	0.13	0.898
Cs-137	661.66	1.50	0.09	1
Eu-152	778.9	1.31	0.07	0.916
Eu-152	867.38	1.19	0.07	0.871
Eu-152	964.08	1.17	0.06	0.946
Eu-152	1085.84	1.06	0.06	1.029
Eu-152	1089.74	1.06	0.07	0.926
Eu-152	1112.08	1.04	0.06	0.966
Eu-152	1299.14	0.87	0.06	0.918
Eu-152	1408.01	0.871	0.048	0.959
Co-60	1173.23	0.97	0.06	0.936
Co-60	1332.49	0.88	0.05	0.933

From Table 2 data one can conclude: for monoenergetic radionuclides, ^{137}Cs and ^{241}Am , the effect is not present, while for ^{60}Co , ^{133}Ba , ^{152}Eu the effect can influence the calibration up to 13%, in both senses, depending on the recorded „summing-out” or „summing-in” coincidences. The efficiency calibration values of the same HPGe detector, Table 2 data, without and with TCS corrections, are presented respectively in Figs. 2a and 2b.

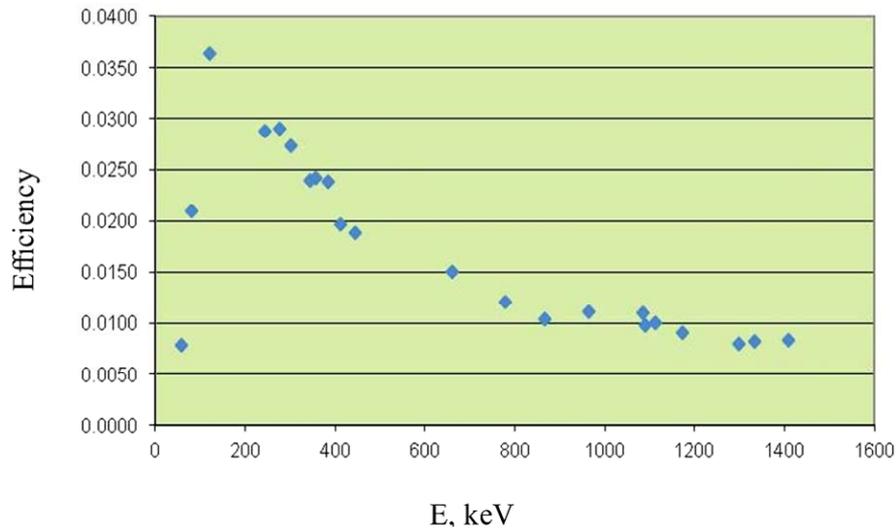


Fig. 2a – Experimental efficiency calibration (HPGe detector, volume standard sources with soil matrix, placed on top the detector), without true coincidence summing (TCS) corrections.

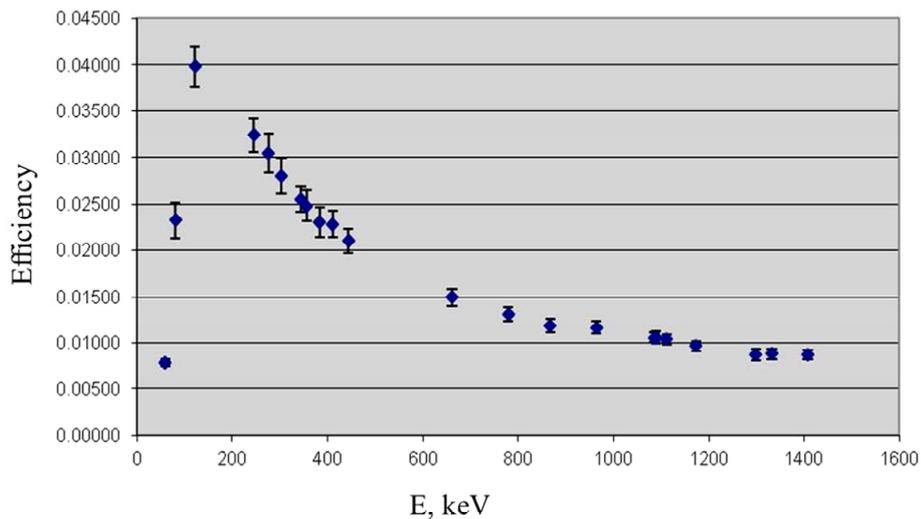


Fig. 2b – Experimental efficiency calibration (HPGe detector, volume standard sources with soil matrix, placed on top the detector), with true coincidence summing (TCS) corrections.

From Figs. 2a and 2b one can conclude that the efficiency curve discontinuity from case (a) are corrected when TCS correction is applied, case (b). For the first time in Romania, an efficiency calibration procedure taking into account the true coincidence summing corrections for HPGe detector was elaborated and implemented, based on the GESPECOR software [8]. The necessary input data are

provided by the detector producer, and completed by Monte Carlo simulations and other experimental data. The correct calibrations of the system for various measurement geometries were validated by the participation of RML in national and international comparisons and proficiency tests [9, 10].

3.3. ADDITIONAL TESTS

The calibrations are completed by:

- the determination of the Minimum Detectable Activity for the radionuclides of interest (based on the background measurement);
- the time stability check for a reference peak position, net counting rate and FWHM.
- the measurement of a standard source of the same type (radionuclide, matrix, volume), using the calibration data, and comparison of the obtained value with the conventionally true value, certified by a metrology laboratory. The two values should agree within the limit of their expanded uncertainty (coverage factor, $k = 2$).

4. ISSUE OF THE CALIBRATION CERTIFICATE

The calibration certificate content is described in a dedicated work procedure. The important points refer to: use of the logo – RENAR; short description of the equipment; calibration conditions (climatic conditions, electric parameters, etc.); activity unit traceability route; calibration method; calibration results, expressed as equations, tables, etc; recommendations, when necessary.

5. CONCLUSIONS

Two working procedures were elaborated and implemented in the calibration of high and low resolution γ -ray spectrometry installations, belonging to clients throughout the country.

All the necessary calibration corrections and the customers feedback were analyzed and applied to improve the quality of the radioactivity measurements performed by the laboratories from the public networks for environmental monitoring, food chain and health, research.

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