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Uncertainty evaluation in activity measurements with HPGe using two relative methods

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ABSTRACT

Radionuclide metrology laboratories aim to provide radioactive standards for detector calibrations in nuclear safety areas with low uncertainties. Such standards are applied in nuclear industries, according to the requirements of monitoring programmes. The standard radionuclides of ^{133}Ba, ^{152}Eu and ^{166m}Ho are suitable to the determination of efficiency curves in HPGe detectors due to their multi-gamma emissions, which are intense and well separate in the spectrum. With efficiency curves it is possible to do the measurements without needing to use standards. In this work was made a verifying of the uncertainties obtained for the two relative methods: comparative (sample-standard) and efficiency curve. The total uncertainties obtained by sample-standard method varied from 0.4 to 1.2 % (k=1). The results using efficiency curve method are between 0.9 to 2.2 % (k=1).

Keywords: **uncertainty , gamma emitters, germanium detectors , radionuclide metrology.**

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1. INTRODUCTION

Many radionuclides are used as standards in gamma spectrometry technique in order to determine activities and half-lives ($t_{1/2}$) in several sources applied in radioprotection, nuclear safety areas and nuclear technology (medical and industrial). The activity is defined as a number of spontaneous nuclear transformatios in the unity of time, which is a quantity to be measured.

^{57}Co is used as standard in nuclear medicine to intercomparison and calibration programmes. ^{60}Co can be used in radiography, radiotherapy, food-irradiation, and as a standard for checking instruments. ^{133}Ba is used as a reference source to efficiency curve calibration because is important as multi-gamma spectrum emitter at low energy (<400 keV) and is also used as a surrogate for ^{131}I as it has a longer half-life (10.52 years). ^{241}Am is largely applied as reference source because of its monoenergetic low gamma energy (59.54 keV) emission. ^{152}Eu is produced by the fission of U and Pu, and it is normally used as standard source because is more suitable as a multi-gamma spectrum emitter in a wide range of energy besides having several gamma photopeaks well defined and isolated in the spectrum.

The activity determination of a radioactive source needs the detector efficiency response knowledge in function of the energy [1]. The efficiency calibration establishes the relationship between photopeak area and the nuclide activity of the standards as:

$$\varepsilon_f(E) = \frac{S(E)}{[A.I(E).t]} \, .Fc \qquad (1)$$

Where ε_f (E) is the total absorption efficiency at E energy; A is the activity (Bq); S(E) is the photopeak area (counts); I(E) is the absolute probability emission of the energy considered for the specific nuclide; t is the counting time, and Fc is the correction factor that refers to decay correction, detector-source position, dead time, count geometry and weighing. The total absorption efficiency determination can be done from geometric considerations and interaction probabilities or using a semi-empirical relation [2].

There are studies over influence of the geometry parameters on the determination of the efficiency calibration uncertainties for the radiological characterization material and for activated material which is necessary a uncertainty associated of the gamma spectrometry [3-5].

By another path of efficiency determination detector response is using standards with a large energy range, or a multi-gamma standard like ^{166m}Ho (80 to 1000 keV) [6,7], ^{152}Eu (100 to 1400 keV) [6,8] and ^{133}Ba (40 to 400 keV) [6], according to the region of interest.

The efficiency curve depends on radiation energy [9], sample geometry, photon attenuation (sample absorption and absorption between sample-detector), dead time and sample-detector position. This curve is obtained from the acquisition of reference spectra considering the photopeak areas for each energy that corresponding the standard activities.

When the calibration of gamma-ray spectrometry using Ge detector is required for determination of the high activity and/or multi-gamma emitter samples, a large source-detector distance is adapted [10]. This reduces dead-time and true coincidence summing effects [11].

Calin [12] has been demonstrated that the activity values obtained in gamma- ray spectrometers are comparable with low errors respecting with true activity values using standard sources of ^{60}Co, ^{133}Ba, ^{137}Cs and ^{241}Am.

From a high-resolution germanium detector and with well-fitted efficiency curves in a defined geometry using range of energy (50 – 1410 keV) and with appropriate standards, is possible to do a sample calibration in activity. Also the gamma-ray impurities present in the samples can be identified and quantified precisely. Besides, these procedures can be applied as a support to absolute calibration system in the metrological laboratory [7]. If the calibration has been made in accordance between these methods, the activity results could be determined with high accuracy and precision [4]. Even knowing that the accuracy and precision of the relative methods rely mainly on decay scheme data taken from the literature. In this work, it was used the experimental arrangements for measuring precisely the activity of gamma-emitter point and ampoule sources using the efficiency curve and the comparative methods for samples of ^{57}Co, ^{60}Co, ^{133}Ba, ^{152}Eu and ^{241}Am.

2. MATERIALS AND METHODS

2.1. The efficiency curve method

An efficiency curves were determined for source-detector distance (5 cm, 10 cm and 20 cm) using multi-gamma point source standards of ^{152}Eu, ^{133}Ba and ^{166m}Ho with traceability to the international reference system [7]. This methodology normally requires correction factors due to pile-up losses (< 0.1%), source geometry effects (< 0.2%), and gamma-gamma summing coincidence effects [8].

The Figure 1 shows the efficiency curve obtained by standard sources of ^{133}Ba, ^{152}Eu and ^{166m}Ho, that were positioned at 10 cm distance from the top of detector to reduce pulse stacking, summing effect and dead time (<5%) [13 - 15] in order to determine the activity values .

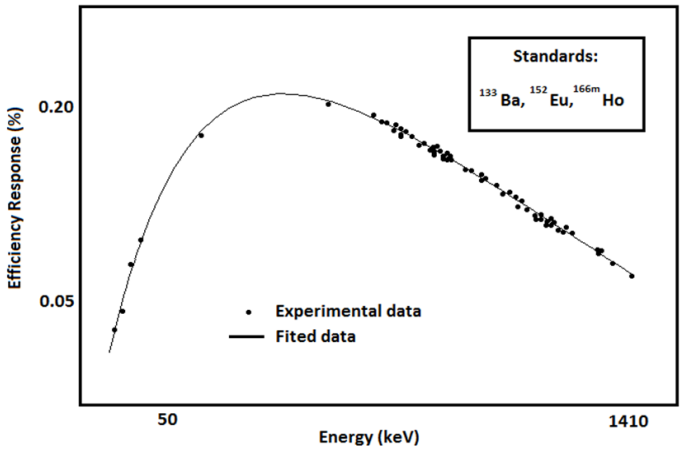
2.2. The comparative method

In this method there is a standard with the parameters previously defined and certified in which it is used as a reference in determination of parameters of a sample with the same radionuclide. Normally it is used to determine the activity.

Both methods, efficiency curve and comparative, were applied to activity determination with associated uncertainties for the following radionuclide samples: ^{57}Co, ^{60}Co, ^{133}Ba, ^{152}Eu and ^{241}Am.

The metrology laboratory used a coaxial HPGe detector, with a volume of 260 cm^3, a resolution (FWHM) of 1.90 keV at 1332.5 keV and a 66:1 Peak-Compton ratio. This system (Ortec) is associated with electronic units including an Analog-to-Digital Converter interface module integrated to multichannel analyzer and Maestro II software.

Figure 1: Efficiency Curve obtained with ^{133}Ba, ^{152}Eu and ^{166m}Ho standard sources [15]



The solid sources to be analyzed were prepared by dropping the original solution on acrylic disc. These acrylic sources have the following dimensions: 25.4 mm of external diameter, 5.0 mm of height, 1.0 mm of thick for deposition area and 1.0 mm of covering. The liquid sources are glass ampoule, they have the following dimensions: 5 mL of capacity, 90 mm of height, 14 mm of external diameter, 0.5 mm of thick, and the sample solution has 2.6-2.7 g that corresponding of 20 mm of solution height. Energies used to activity determination are [6]: ^{57}Co – 122.06 keV; ^{60}Co - 1173.24 keV and 1332.50 keV; ^{133}Ba - 81.00 keV, 276.79 keV, 302.85 keV, 356.01 keV and 383.85 keV ; ^{152}Eu – 121.78 keV, 244.69 keV, 344.27 keV, 778.89 keV, 964.12 keV and 1112.02 keV ; ^{241}Am – 59.54 keV.

3. RESULTS AND DISCUSSION

The original solutions were calibrated by 4πβ-γ absolute method [16] and they were used for comparison two relative gamma-spectrometry methods, which are the comparative and efficiency curve measurements. Typically, absolute methods are also suitable for determination of activity in spite of being costly and time consuming. So to fill this gap, it was used the gamma spectrometry to validate and to furnish the calibration values to users sources after the sample dilutions and drying

treatments. Finally, the samples impurities were investigated and were not identified by gamma spectrometry .

The main typical uncertainty components for a ^{60}Co sample [17], as an example, showed on Table 1

Table 1: ^{60}Co uncertainties to gamma spectrometer method (k=1) [17]

Unc. components	**A (%)**	**B (%)**
live time		0.01
weigh		0.20
half- life		0.05
background		0.02
decay		0.01
counting statistic	0.03	
combined uncertainty	0.23	

The Table 2 shows the reference values [6] of gamma emission probability uncertainties (%) and the uncertainties values obtained in this work from the efficiency curve to range of energies of each radionuclide.

Table 2: Values of gamma emission probability uncertainties and activities range with uncertainty values for radionuclide source samples measured. Both by efficiency curve method (k=1) [6]

Nuclide	Gamma Emission Probability Unc. (%)	Efficiency Unc. Obtained (%)*	Measured Activity Interval (kBq/g)**	Activity Unc. (%)
^{57}Co	0.20	0.79	0.16 – 200.00	0.9
^{60}Co	<0.01	0,81	0.05 – 13.00	1.4
^{133}Ba	1.10 – 2.60	0.52 – 0.86	0.39 – 18.00	1.5
^{152}Eu	0.50 – 2.60	0.52 – 0.79	3.00 – 12.30	1.5
^{241}Am	0.60	1.76	0.30 – 14.00	2.2

* Values obtained from efficiency curves of this work
**There were not observed any significant impurities on the samples

The values from reference [6] and input decay data as intensity, half-life and counting (peak area), contributed to increase the efficiency uncertainty determination by experimental curve. These results were used in total uncertainty determination of activity of the measured radionuclides.

The range of the total relative uncertainties of the activity determination by efficiency curve is between 0.9 % to 2.2 %. In the ^{241}Am case the total uncertainty obtained is higher than the other radionuclides because its low energy (59.54 keV) .

By the comparative method, the activity uncertainties of the standards showed in Table 3 were used to error propagation to determine the samples activity uncertainties that lies in Table 4.

Table 3: Standards from metrology laboratory used by comparative method (k=1)

Nuclide	Standard	Activity Uncertainty (%)
^{57}Co	1	0.26
	2	0.23
^{60}Co	1	0.09
	2	0.31
	3	0.20
^{133}Ba	1	0.31
^{152}Eu	1	0.97
^{241}Am	1	0.37
	2	0.65

Table 4: Activities range and uncertainties values for radionuclide source samples measured by comparative method (k = 1)

Nuclide	Measured Activity Interval (kBq/g)	Activity Uncertainty (%)
^{57}Co	70.00 – 200.00	0.8
^{60}Co	4.00 – 43.00	0.4
^{133}Ba	3.70 – 74.00	0.7
^{152}Eu	8.00 – 170.00	1.2
^{241}Am	0.80 – 50.00	0.8

In these Table 4 the range of the total relative uncertainties of the activity determination by comparative method is between 0.4 % to 1.2 %. There were not observed any significant impurities on the samples. For ^{152}Eu the total uncertainty obtained was higher than others radionuclides because the standard of this radionuclide has a higher activity total uncertainty, 0.97 %, as showed in Table 3.

In a comparison between comparative and efficiency curve methods about activity uncertainty results it was observed that the use of the standards directly (comparative method) to get to lower uncertainties results.

The literature [12] showed that minimum detectable activity for ^{60}Co is 0.22 Bq and in this work the values of the determinate activities were around twice. Respect to ^{241}Am radioisotope the determination of uncertainty by absolute method (4πβ-γ coincidence) is much lower than relative methods by gamma spectrometry using an HPGe coaxial detector, for both efficiency curve and comparative methods, because the uncertainties of standards and of nuclear decay data used in the efficiency curve are higher due to the low energy of this radionuclide, according Tables 2 and 3, respectively.

In Table 5 it can be proven that the absolute methods obviously have better uncertainty values than relative methods (Tables 2 and 4) for all radionuclides considered in this work.

Table 5: Activities with its uncertainty values for radionuclides sources obtained by 4πβ-γ coincidence absolute method from references (k = 1)

Nuclide	**Measured Activity (kBq/g)**	**Unc. (%)**	**References**
^{57}Co	536.5	0.54	[18]
^{60}Co	178.4	0.24	[19]
^{133}Ba	1.2	1.0	[20]
^{152}Eu	94.7	0.33	[21]
^{241}Am	295.6	0.32	[22]

The absolute method is advantageous to calibrate samples routinely due to its low uncertainty and does not require standards, but there are some disadvantages as: the electronic system adjusts, difficulties of the spectrum determination and radionuclide calibration delay (about one month).

In contrast, when it used gamma spectrometry as a relative method to verify the activity of the samples, it is needed a few times to give the results. In this way the gamma spectrometry becomes advantageous when a laboratory has a lot of sources to be calibrated on a daily basis. Thus, both methods are essential in a metrology laboratory, but only the absolute methods provide traceability to users of radioactive standards.

4. CONCLUSION

An efficiency curve method using a Ge detector was applied for obtaining results of some radioactive point and ampoule sources. The total uncertainties reached values below 2.2 % in a confidence level of 68 %. Another method using the comparative method found values below 1.2 % in the same confidence level. These work results showed the comparative method is advantageous to determine the activity to ^{57}Co, ^{60}Co, ^{133}Ba, ^{152}Eu and ^{241}Am radionuclides because the uncertainties of efficiency curve and of the gamma emissions are not included on total uncertainties calculations. So these results showed that the reference sources measured by gamma spectrometry, traceable to the absolute methods and sent to users, could be applied on nuclear programmes to different purposes.

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