Emission probability determination of ^{133}Ba by the sum-peak method

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Abstract: The National Laboratory of Metrology Ionizing Radiation (LNMRI/IRD/CNEN) has several measurement methods in order to ensure low uncertainties about the results. Through gamma spectrometry analysis by sum-peak absolute method they were performed the standardization of ^{133}Ba activity and your emission probability determination of different energies with reduced uncertainties. The advantages of radionuclides calibrations by absolute method are accuracy, low uncertainties and is not necessary the use of radionuclides reference standards. ^{133}Ba is used in research laboratories on calibration detectors in different work areas. The uncertainties for the activity and for the emission probability results are lower than 1%.

Keywords: sum-peak method; ^{133}Ba; activity; emission probability; gamma spectrometry

1. INTRODUCTION

The National Laboratory of Ionizing Radiation Metrology (LNMRI), linked to the National Nuclear Energy Commission (CNEN), is the laboratory designated by the National Institute of Metrology and Quality Technology (INMETRO) for reference standards determination to ionizing radiation. In your chart there is a radionuclide service that standardizes the quantity activity.

Over the years participates in key international comparisons that aims to ensure the traceability of measurements in the worldwide network of metrology, coordinated by Bureau International des PoidsetMesures, (BIPM). The LNMRI laboratory is also a participant of the Inter-American Metrology System (SIM). It has several measures for radioactive sources systems in various geometries, the data generated are analyzed by different calibration methods, and these methods can be absolute or relative.

The gamma spectrometry using high purity germanium detector (HPGe) is an established technique for determining photon emitting by radionuclides [1]. The methods of analysis can be: 1-relative (comparative), when using standards for determining the activity of the samples as they have the same nature and geometry of such samples; 2- efficiency curve, this curve is generated using multiple standards (^{152}Eu, ^{166m}Ho) allowing the identification and determination of impurities generated in the

production process or in radionuclide contamination; 3- the determination of the emission probabilities of gamma and x-rays.

The sum-peak method was developed by Brickman [2] and his coworkers in 1963. This method shows that it is possible to dodirect calibration between two coincident radiation x-ray and γ or both. Over the years several articles were published using HPGe [3,4,5] detector.

The ^{133}Ba has 10.54 years of half-life and it decays to^{133}Csby electron capture (100%) into two main branches, E_1 (86.2%) to the level of 437 keV, and E_2 (13.7%) to the level of 383.8 keV, Figure 1. This radionuclide has characteristic x-rays and gamma energy ranges ,about 14 lines, forming coincidences emissions to obtain its stability [6]. These energies issued are used for analysis. Because of its nuclear features the ^{133}Ba can be a standard for the sum- peak method.

In 2006, the LNMRI laboratory participated in an international comparison of ^{133}Ba whose calibration was difficult due to decay. The purpose of this manuscript is the calibration of ^{133}Ba using the sum-peak method, by making a direct calibration, fast and practical, to obtain activity and emission probability values.

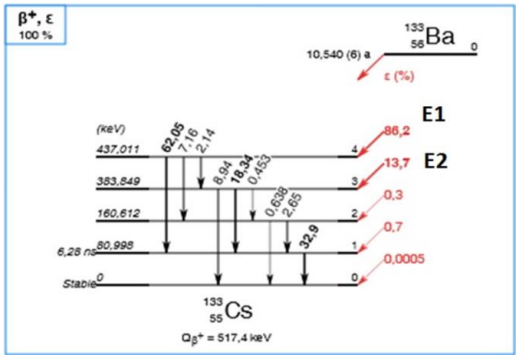

Figure 1. Scheme of ^{133}Ba decay [6]

2. METHODOLOGY

The sum-peak method makes direct measurements and has the same aspects of coincidence count in gamma spectrometry, using only a single gamma radiation detector. It considers that the activity of an isotope that emits two electromagnetic radiation, x-x, x-γ and γ-γ, in coincidence can be extracted from the areas on both photopeaks: the peak corresponding to area of the sum, and an extrapolation for zero energy.

The equation used was described by Brickman in his works is:

$$(A_1 . A_2) / A_{12} + T = N \quad (1)$$

Where:

A_1 and A_2: area of photopeak on the gamma energies in coincidence;

A_{12}: area of photopeak on the energies in sum;

T: full spectrum counts;

N: source absolute activity

The usual absolute calibration method is the 4π(β-γ) coincidence using two detectors to measure β–γ, γ-γ and x-γ radiations are coincident, and the results are obtained through equations of the combinations of the observed channels.

2.1 Activity determination

The activity determination by the sum-peak method has low uncertainty to the same standards of other secondary techniques. The uncertainties in the sum-peak method are basically only type A, this is due to the counts.

The relative combined standard uncertainty associated with any result obtained by gamma-ray spectrometry depends on the uncertainties of the main input parameters, and also of the correction factors. The uncertainties are: type A (counts under the photopeak) and type B (sample mass, decay, half-life and source position). Other type B uncertainties factors such as dead time, sum effect, pile-up, adsorption, self absorption, temperature, pressure and humidity are neglected.

They were fabricated and calibrated five point sources in a thin film, with activities from 1 to 6 kBq. These activity values ensure that the pile-up effect does not occur.

The Canberra GL Model 2020 R germanium planar detector was used, coupled to a voltage divider that allows an increase in work 300 keV to 1200 keV of energy, allowing the full scan of ^{133}Ba spectrum. To compose the spectrometer: the power supply Model 459, Model 572 amplifier, ADCAM MCB 926 and MAESTRO program, Ortec, was used.

The measures of the sources were held on the detector surface (0.1 cm). The time of measurement was in accordance with the counts obtained by providing low uncertainty. The calculations were carried out using spreadsheet Uncertainties follow the recommendations ISOGUM.

2.2 *Gamma emission probability determination*

The result of activity obtained from ^{133}Ba sources by the sum-peak method was applied to calculate the gamma emission probabilities (Pγ) for the following energy: 276, 302, 356 and 383 keV. The choice of these energies used in calculating Pγ was due to their higher intensity and are the most used range of ^{133}Ba emission probability.

For the calculation of Pγ energies above 276 keV it is necessary to fabricate an efficiency curve of ^{133}Ba using the germanium detector. Through the use of reference standards and sources of ^{166m}Ho and ^{152}Eu, using 27 points gamma energies in the range between 121 keV and 1408 keV. Using spreadsheet and LABFIT curve fitting softwares, the efficiencies of energy were calculated.

The Pγ were calculated using the equation:

$$P\gamma = C / (N_0 * \varepsilon_\gamma) \quad (2)$$

Where:

C is the count rate in photopeak;
No is the source activity measurement (Bq) at the reference date;
ε_γ is the photopeak efficiency of γ rays emitted.

The main components of total uncertainty in the determination of Pγ were derived: 1- uncertainties of the ^{133}Ba sources activities; 2- type A uncertainty (by the areas of photopeaks determination); 3- type B uncertainties (by the decay correction and efficiencies interpolated from efficiency curve).

3. RESULTS AND DISCUSSION

Through the ^{133}Ba decay they used the x-ray characteristic kα averaging 30.8 keV and kβ averaging 35.5 keV and, possible sums the energy range for the activity calculation. The experimental conditions were optimized to provide the best spectra.

Figure 2 shows the energy range spectrum, 270 keV to 450 keV. It can observed the gamma energies and their sum with x-rays. Unidentified peaks in this figure are the background radiation. The simple sum of the x- ray with gamma rayare displayed, suggesting the application of equation 1.

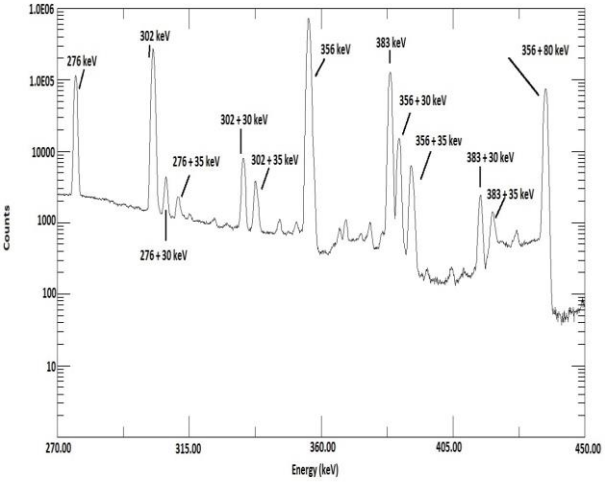

Figure 2. The 270 keV to 450 keV energy spectrum regarding the ^{133}Ba decay and their sums with the source of the detector.

In table 1 are shown the activity values obtained by the sum-peak method from 5 samples generated from the mother`s source.

Table 1: Results of ^{133}Ba mother`s sourceactivity by the sum-peak method (2015/03/25 at 12:00 h)

Source	Activity (kBq/g)	u_a	u_b	U_t (k=1)
1	156.13	0.51	0.48	0.35
2	156.21	0.51	0.48	0.35
3	156.24	0.51	0.48	0.35
4	156.23	0.51	0.48	0.35
5	156.25	0.51	0.48	0.35
Average	156.21	0.51	0.48	0.35

The sums of x-rays with gamma ray energies were analyzed; some energy sums suffer interference from other energy and therefore require deconvolution to obtain better results. In table 2 is shown the interference that have been identified in the spectral analysis.

Table 2: Presenting the summing energies and their interference.

Summing energies (keV)	Possible interference
30.8 + 80.9	no
35.5 + 80.9	no
30.8 + 276.4	near302.9 keV
35.5 + 275.4	no
30.8 + 302.9	no
35.5 + 302.9	no
30.8 + 356.0	near383.9 keV
35.5 + 356.0	near 383.9 keV
30.8 + 383.9	no
35,5 + 383,9	no
80.9 + 356.0	(53.2 + 383.9) keV sum

Table 3. Gamma probability emission to ^{133}Ba energies data by LNHB laboratory (2016) [7]

Energy (keV)	P_γ (%)	U_t (k=1)
276.4	7.13 (6)	0.84
302.9	18.31 (11)	0.60
356.0	62.05 (19)	0.31
383,9	8.94 (6)	0.67

Table 4: Results of ^{133}Ba intensity of gamma emission probability (P_γ) to each energy considered using activity result from sum-peak method (table 1).

Energy (keV)	P_γ (%)	u_a	u_b	U_t (k=1)
276.4	7.13	0.05	0.81	0.81
302.9	18.31	0.01	0.86	0.86
356.0	62.05	0.18	0.94	0.96
383,9	8.94	0.05	0.99	0.99

The uncertainty results of the table 4 are higher than LNHB reference (table 3) because it was also used the efficiency curve results to obtain the gamma emission probability. This efficiency curve was gotten by ^{152}Eu and ^{166m}Ho energies and the uncertainties of the efficiency values were higher for the 302, 356 and 383 keV.

4. CONCLUSIONS

The ^{133}Ba activity measurement values are as expected according to the literature. The sum-peak absolute method can be applied to the calibration of this radionuclide in addition to being practical, fast, simple and with getting low uncertainties. This method can be used and enhanced so that it becomes a useful tool for radionuclide metrology in general.

All summing energies must be analyzed and also can observe that the sums generated by the triple sums mask the end of results.

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