2015 International Nuclear Atlantic Conference - INAC 2015
São Paulo, SP, Brazil, October 4-9, 2015
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN
***ISBN:* 978-85-99141-06-9**

CALIBRATION OF ^{133}BA BY SUM-PEAK METHOD

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ABSTRACT

A calibration laboratory should have several methods of measurement in order to ensure robustness on the values applied. The National Laboratory for Metrology of Ionizing Radiation, (LNMRI IRD), provides gamma sources of radionuclide in various geometries and standardized in activity with reduced uncertainties. Some absolute and relative methods of calibrations could be used routinely. Relative methods require standards to determine the activity of sample to be calibrated, while the absolute methods do not need, simply make the counting and the calculation of the activity is obtained directly. The great advantage of calibrations of radionuclides by absolute method is the accuracy and low uncertainties. ^{133}Ba is a radionuclide enough used in research laboratories and calibration of detectors for environmental analysis and, according to the scheme, it decays 100% by electron capture and emits about 14 energy gamma and x-ray lines, forming several coincidences. However, the classical methods of absolute measurement, as coincidence 4 $\pi\beta$-γ have difficulty to calibrate ^{133}Ba due to its complex decay scheme. The sum-peak method, developed by Brickman, could allow this calibration. It is used for radionuclide calibration that emits at least two photons in coincidence. Therefore, it was developed a methodology that combines gamma spectrometry technique with sum-peak method to standardize ^{133}Ba samples. Activity results obtained proved compatible, with uncertainties of less than 1%, and, when compared with other methods of calibration, sum-peak demonstrated the feasibility of this methodology, particularly, for simplicity and effectiveness.

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1. INTRODUCTION

The National Metrology Laboratory of Ionizing Radiation (LNMRI), subordinate to the National Nuclear Energy Commission (CNEN), is the laboratory designated by the National Institute of Metrology and Quality Technology (INMETRO) to reference standards determination associated with ionizing radiation. In your chart there is a radionuclide service that standardizes the greatness activity. Over the years participates in key international comparisons that aims to ensure the traceability of measurements worldwide network of metrology, coordinated by Bureau International des Poids et Mesures, (BIPM). It is also a participant of the Inter-American Metrology System (SIM). The laboratory has several measures for radioactive sources systems in various geometries, the data generated are analyzed by different methods of calibration, these methods can be absolute or relative.

The gamma spectrometry using high purity germanium detector (HPGe) is an established technique for determining photon emitting radionuclides [1]. The methods of analysis can be: by relative (comparative), when using standards for determining the activity of the samples as they have the same nature and geometry of such samples; by efficiency curve, this curve is generated using multiple standards (^{241}Am, ^{152}Eu, ^{166m}Ho) allowing the determination of impurities generated in the production process or in radionuclide contamination, and, also, the determination of the probabilities emission of gamma and x-rays.

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

The ^{133}Ba has a half-life of 10.54 years and decays by electron capture 100% into two main branches, E1 (86.2%) to the level of 437 keV, and E2 (13.7%) to the level of 383.8 keV, to ^{133}Cs, Figure 1. This radionuclide has x-rays characteristic and various energy ranges totaling 14 lines, forming coincidences emissions to obtain its stability [2]. These energies issued are used for analysis. Because of its nuclear features the ^{133}Ba may be a standard for the sum- peak method.

In 2006 the LNMRI 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 reliable values.

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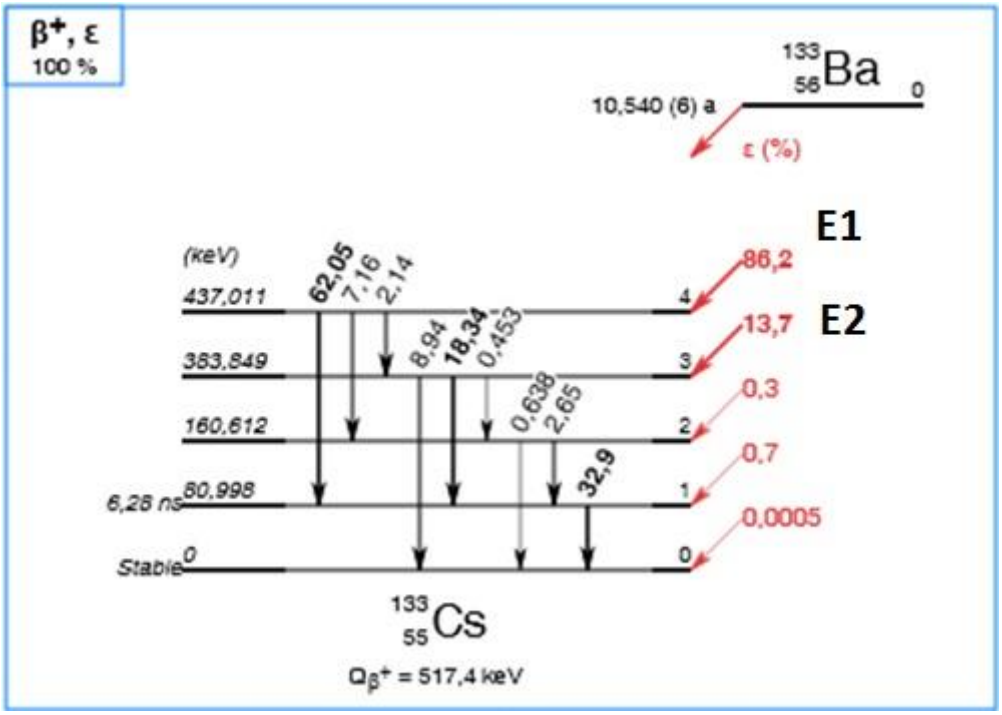
Figure 1. Scheme of ^{133}Ba decay



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 :

$$\frac{A_1 \,.\, A_2}{A_{12}} + T = N \qquad (1)$$

Where: A1 and A2 : area of fotopicos gamma energies observed in coincidence;
A12 : area of fotopicos to energies observed in sum ;
T : full spectrum counts;
N : absolute activity of the source.

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The usual absolute calibration method is the coincidence $4\pi\beta - \gamma$, uses two detectors to measure the $\beta - \gamma$ radiation , $\gamma - \gamma$ and X- γ that are coincident , the results are obtained through equations of the combinations of the observed channels.
The activity calculation by 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 sources with punctiform geometry 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 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 in two locations : on the detector surface (0 cm) and 10 cm away. The time measurement was in accordance with the counts obtained by providing low uncertainty. The calculations were carried out using EXCEL. Uncertainties follow the recommendations ISOGUM.
Measurements sources made at 10 cm from the detector were compared with a BIPM standard with the same geometry.

3. RESULTS AND DISCUSSION

They used the x-ray characteristic kα averaging 30.8 keV and kβ averaging 35.5 keV decay of ^{133}Ba , and possible sums the energy range for the calculation of activity. The experimental conditions were optimized to provide the best spectra.
Figure 2 shows the energy range spectrum , 270 keV to 450 keV. Observe 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 radiation are displayed, suggesting the application of Equation 1 .

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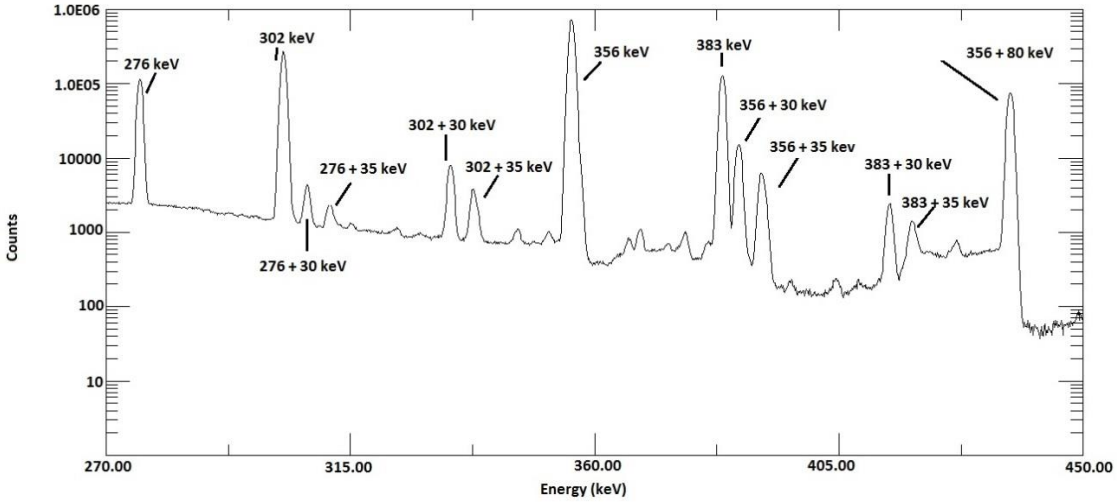

Figure 2: The 270 keV to 450 keV energy spectrum regarding the ^{133}Ba decay and their sums with the source of the detector.

Table 1 shows the results of the various sources at the position 10 cm above the detector by comparison method with the standard BIPM .

Table 1: Results of activity by comparison method using standard BIPM

Source	Activity kBq/g	Ua	Ub
1	**156,465**	**1,2**	**0,48**
2	**156,794**	**0,92**	**0,48**
3	**156,960**	**0,93**	**0,48**
4	**157,366**	**1,2**	**0,48**
5	**157,660**	**0,49**	**0,48**

In Table 2 are shown the values obtained by sum-peak method

Table 2: Results of activity by sum-peak method

Source	Activity kBq/g	Ua	Ub
1	**156,130**	**0,51**	**0,48**
2	**156,210**	**0,51**	**0,48**
3	**156,242**	**0,51**	**0,48**
4	**156,233**	**0,51**	**0,48**
5	**156,250**	**0,51**	**0,48**

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Table 3: Comparative results between activity calibration methods to ^{133}Ba mother source (2015/03/25 at 12:00 h)

Method	**Activity kBq/g**	**Ua**	**Ub**
Sum-peak	**156,213**	**0,51**	**0,48**
Decay	**156,283**	**0,06**	**0,66**
Comparative	**157,049**	**0,95**	**0,48**

Table 3 shows the mother`s source decay (generated the five samples). The reference date is 2015/03/25 at 12:00 h .

Only analyzed the sums of x-rays with gamma energies; some suffer interference from other energy and thus require deconvolution to obtain best results. In table 4 is shown the interference that have been identified in the spectral analysis.

Table 4: Statement of sums and their interference.

Energy keV	**Possible interference**
30,8 + 80,9	**no**
35,5 + 80,9	**no**
30,8 + 276,4	**near the 302,9 keV**
35,5 + 275,4	**no**
30,8 + 302,9	**no**
35,5 + 302,9	**no**
30,8 + 356,0	**near the 383,9 keV**
35,5 + 356,0	**near the 383,9 kev**
30,8 + 383,9	**no**
35,5 + 383,9	**no**
80,9 + 356,0	**with the (53,2 + 383,9) keV sum**

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4. CONCLUSION

The measured values of ^{133}Ba activity are as expected according to the literature. We conclude that 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.
It was observed that the results of activity for some energy sums (x-rays and γ) is influenced by the fact that energy (x-rays) overlap the gamma energy, compromising the reduction of uncertainty and need a deconvolution. All sums must be analyzed and also observe the sums generated by the triple sums that mask the end result.

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