Absolute standardization of the $^{133}$Ba by sum-peak method and comparison with other methods

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Abstract. This paper presents the absolute standardization of $^{133}$Ba by sum-peak method. $^{133}$Ba owns a complex decay and was used the sum coincidences between the average energy of characteristic X-rays ($k_\alpha_1$ and $k_\alpha_2$) and gamma-rays. The activity results presented uncertainty values below 0.3 % and were compared with those of other absolute activity measurement techniques. The results obtained are compared with other methods and confirm that this is an alternative, fast and efficient method for $^{133}$Ba standardization.

1. Introduction

The sum-peak coincidence absolute method was introduced in the 1960s by Brinkman and his collaborators for standardization of radionuclides. This simple and challenging method of radiation measurement enables the direct standardization of radionuclides emitting two photons in coincidence using gamma spectrometry. It uses the peak counts of gamma and X-rays and their possible sums, plus the total spectrum count plus the extrapolation for energy equal zero. $^{133}$Ba decays by electron capture emitting X-rays and gamma radiation in coincidence [1-4].

Recently $^{125}$I activity was determined in 2011 using two sodium iodide detectors and compared with that obtained with an HPGe detector [5]. In 2007 de Almeida determined $^{51}$Cr activity using an HPGe detector [2]. All of these authors used the sum-peak method. Many laboratories in recent years have published papers of standardization of simple decay radionuclides by sum-peak method [1, 2, 5].

Here $^{133}$Ba standardization was performed, considering a radionuclide complex to use the sum-peak coincidence absolute method. It was then established the absolute sum-peak method is an efficient, fast and without many corrections for the analysis of this radionuclide when compared with other [6].
2. Experimental

Six sources of $^{133}$Ba with activity varying from 1 kBq to 4 kBq were prepared in geometry 10R glass vial and were used in standardization in the absolute anti-coincidence and Ciemat/Nist methods. A glass ampoule with 2 cm was used for measurement by ionization chamber. An analytical balance model AX205 was used.

However six point sources, also prepared, were measured with a Canberra HPGe gamma-X planar detector, with 20 % relative efficiency and a power range of 3 to 300 keV. Applying a specific voltage divider, inserted between the preamplifier and the amplifier, as Figure 1, it was possible to increase up to 1100 keV the detector operation range [7].

The energies at 53, 276 and 356 keV from the electronic capture (85.4 %) were used for $^{133}$Ba measurements. Other branch at 302 keV from the electronic capture (14.4 %) was also used. These energies are in agreement with the average of the X-rays energies ($\kappa_1 = 30.6$ keV and $\kappa_2 = 30.9$ keV).

The counting time for sources was 24 hours and 48 hours to the background radiation.

The photopeak areas were determined with the aid of deconvolution software that makes the separation of the double or triple photopeaks [8].

![Figure 1. Voltage divider device.](image1)

![Figure 2. The 53 keV energy deconvolution COLEGRAM software.](image2)
3. Methodology

Figure 3 shows the decay of $^{133}$Ba in a different form than conventional [9]. Level 6 represents the ground state of $^{133}$Ba, and from level 5 to level 1 represents the various energy levels of the excited state of the $^{133}$Cs. The decay EC1 represents the branch of electronic capture with 86.2\%. In it are the X-rays kα and kβ with the energies 30.8 and 35 keV, respectively. Similarly, EC2 represents the electron capture branch with 13.7\%, where the energies of the X-rays kα and kβ are respectively 30.8 keV and 35 keV.

![Figure 3. $^{133}$Ba scheme decay modified.](image)

Figure 4 shows the germanium detector spectrum in the energy range from 270 to 450 keV where the gamma energies and sums with the kα and kβ X-rays caused by the coincidence sum effect appear. In this figure some photopeaks are coming from possible escape peaks. The simple sums of the gamma rays with X-rays from the coincidence effect are well evidenced.
Figure 4. Energy spectrum from 270 keV to 450 keV of $^{133}$Ba decay and its sums with the source on detector.

Equation 1 is used by the absolute sum-peak method for the different energies (x) [5].

$$N_0 = T + \frac{N_\alpha N_\gamma}{N_{\alpha\gamma}} - \frac{N_\alpha^2 N_{aa}}{N_{2\alpha\gamma}}$$

(1)

where:

- $N_\alpha, N_\gamma, N_{aa}, N_{\alpha\gamma}$ = keV for X-rays $k\alpha$ for gamma and sum-peak energy peak counts (E);
- $N_0$ = total activity of the source;
- $T$ = the total counts of the entire spectrum.

4. Results and discussion

Point sources of $^{133}$Ba with low activity were used, aiming at canceling the pile-up effects. This phenomenon occurs within the processing time of the electronic signal generated by the radiation mainly for sources with high activity. It was verified that the pile-up effect was below the detection limit of the system. The dead time in relation to the most active source was less than 5%, this is the difference between the actual time and the counting time of the radiation.

In the standardization of the $^{133}$Ba activity the photopeaks average $k\alpha_{1,2}$, $k\beta_{1,2}$ energies (30.6 and 35 keV) were used in coincidence with the photopeaks of gamma energies (53, 276, 356 and 302 keV). For each source measured, four different activity values were obtained. The other $\gamma$ energies were not analyzed due to interferences caused by other sums or because they are low probability energies.

The peak areas of different energies and their coincidences were determined using the deconvolution software.

Table 1 presents the uncertainty components considered for the determination of the activity.
Table 1. Uncertainty of the $^{133}$Ba activity measured by sum-peak method.

| Uncertainty Components     | A (%) | B (%) |
|----------------------------|-------|-------|
| Dilution factor            | 0.05  |       |
| Live time                  | < 0.01|       |
| Weight                     | 0.20  |       |
| Half-life                   | 0.01  |       |
| Background                 | < 0.01|       |
| Decay                      | < 0.01|       |
| Counting statistic          | 0.11  |       |
| Pile up                    | 0.06  |       |
| Combined uncertainty (k = 1)|       | 0.24  |

The final activity by sum-peak method was determined by the average of the results found by the observed coincidences and compared with the other methods. These results are shown in Table 2.

Table 2. Comparison of $^{133}$Ba activity standardization between sum-peak and other methods.

| Methods           | Activity (Bq/g) | Uncertainties (k=1) |
|-------------------|-----------------|---------------------|
| Sum-peak          | 35.338          | 0.24                |
| Ionization chamber| 35.454          | 0.29                |
| Ciemat/Nist       | 34.959          | 0.48                |
| Anti-coincidence  | 35.254          | 0.66                |

5. Conclusion
The final result of $^{133}$Ba activity obtained by the sum-peak method is suitable for absolute standardization. This result, compared to those obtained by traditional absolute standardization methods, such as anti-coincidence, Ciemat/Nist and BIPM traceable ionization chamber, presented consistent value and coherent uncertainty, below 0.3%. Thus, the sum-peak coincidence method can be used quickly and effectively to standardize in activity $^{133}$Ba a radionuclide with complex decay.
References

[1] Brinkman G A and Aten Jr A H W 1963a Absolute standardization with a NaI (Tl) crystal-III. *Int. J. Appl. Radiat. Isot.* 14, 503-510.

[2] de Almeida M C M, Iwahara A, Poledna R, da Silva C J and Delgado J U 2007 Absolute disintegration rate and 320 keV gamma ray emission probability of $^{51}$Cr. *Nucl. Instrum. Meth. Phys. Res.* A580, 165–168.

[3] Araújo M T F, Poledna R, Delgado J U, Silva R L, Iwahara A, da Silva C J, Tauhata L, Oliveira A E, de Almeida M C M and Lopes R T 2013 Análise de Impurezas Radionuclélicas para o Radiofármaco ($^{123}$I) NaI utilizando a Espectrometria *Gama 7º Congresso Brasileiro de Metrologia* ISSN/ISBN 978-85-86768-07-1.

[4] *Practical Gamma-ray Spectrometry* - Handbooks, manuals, GILMORE GR, Nuclear Training Services Ltd, Warrington,UK, 2008.

[5] *Standardization of I-125 Sum-Peak Coincidence Counting* - Informes Técnicos Ciemat, Carles, A. G., Malonda, A. G., Espana, 2011.

[6] da Silva R L, de Almeida M C M, Delgado J U, Poledna R, Santos A, Veras E V, Rangel J and Trindade O L 2016 Metrological activity determination of $^{133}$Ba by sum-peak absolute method *Journal of Physics: Conference series*, 733 (1).

[7] da Silva R L, Estrada J J S, Delgado J U, Poledna R and de Almeida M C M 2007. Alteração na faixa de detecção e calibração de um sistema de espectrometria gama. *Revista Militar de Ciência e tecnologia*.

[8] Lépy M C Presentation of the COLEGRAM software, Version 4.0, Note technique LNHB/04/26.

[9] Novkovic D, Durasevic M, Kandic A, Vukanac I, Milosevic Z and Nadder L 2007 Coincidence summing of X-and gamma rays of $^{133}$Ba, *Nucl. Instrum. Meth. Phys. Res. A 582*, 592-602.