

Radionuclides measurements by the relative method using a germanium detector

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ABSTRACT

The National Laboratory for Ionizing Radiation Metrology (LNMRI/IRD/CNEN) in Rio de Janeiro has conducted the standardization of various radionuclides. These standards are essential for applications in the nuclear industry, scientific research, radiation protection, and nuclear medicine. A comparative method using a high-purity germanium (HPGe) detector was employed to determine the activity of several radioactive point and ampoule sources. The standardized radionuclides included ²²Na, ⁶⁰Co, ¹³³Ba, and ¹³⁷Cs. The methodology ensured high precision in activity measurements, contributing to the reliability of radiometric procedures across multiple sectors. Total measurement uncertainties were maintained below 2.2 % for liquid samples and below 2.8 % for solid samples, with a confidence level of 95 % ($k = 2$). These results demonstrate the laboratory's capability in producing traceable and accurate standards, reinforcing Brazil's metrological infrastructure in ionizing radiation.

Section: RESEARCH PAPER

Keywords: germanium detector; radionuclides; relative method; metrology

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1. INTRODUCTION

The National Laboratory for Ionizing Radiation Metrology (LNMRI), under the National Nuclear Energy Commission (CNEN), is the institution designated by the National Institute of Metrology, Quality and Technology (INMETRO) as the National Metrology Laboratory for quantities associated with ionizing radiation. This designation reflects its role in ensuring the accuracy and traceability of measurements in this specialized field. LNMRI actively participates in international intercomparison exercises to validate its measurement capabilities and maintain traceability within the global metrology network. These activities are coordinated by the Bureau International des Poids et Mesures (BIPM) and the Inter-American Metrology System (SIM), reinforcing the laboratory's alignment with international standards and best practices.

⁶⁰Co is used in radiography, radiotherapy, food irradiation, and as a calibration standard for instrument verification. ¹³³Ba serves as a reference source for efficiency curve calibration due to its multi-gamma emission spectrum.

One of the radioisotopes produced during the nuclear fission of ²³⁵U and potentially released into the atmosphere is ¹³⁷Cs [1].

This radioisotope can be absorbed by plants and, consequently, contaminate human food sources, such as meat and milk [2].

The ²²Na isotope is used as a radiotracer. It is produced at the IPEN/SP cyclotron through a nuclear reaction between ²⁴Mg and ²H (deuterium) [3]. This radioisotope can be used in PET (positron emission tomography) scans, which are diagnostic imaging procedures.

It offers various systems and calibration methods for gamma-emitting radioactive sources in different geometries, which can be either absolute or relative (comparative or efficiency curve). Gamma spectrometry using a high-purity germanium (HPGe) detector is applied to the analysis of photon-emitting radionuclides [4]. This is a relative technique, as it relies on standards to determine the activity of samples by comparing them to a radionuclide of the same nature.

Gamma spectrometry using a germanium detector is employed in the nuclear industry, nuclear research, radiation protection, and nuclear medicine.

Volcanic rocks and granite are used as construction materials, leading to the release of ²²²Rn into indoor air environments. The ²²²Rn gas originates from these natural materials, which contain radioactive ²³⁸U and ²²⁶Ra in equilibrium. The decay of these

radionuclides occurs through α -particle emissions accompanied by gamma-radiation transitions. Through these gamma transitions, the activity of ^{226}Ra and the dose from ^{222}Rn gas can still be estimated using gamma spectrometry [5].

When calibrating gamma-ray spectrometry using an HPGe detector for determining the activity of multi-gamma-emitting samples, a source-to-detector distance should be applied [6]. This distance should minimize dead time and reduce true coincidence summing effects [7].

The comparative method is still widely used to obtain activity results with high accuracy and precision. The equation for determining the activity of the sample is:

$$A_x = A_p \cdot \{CP_{sx}/CP_{sp}\} \cdot F_c \quad (1)$$

where:

A_x = sample activity on the same reference date
 A_p = standard activity on the same reference date
 CP_{sx} = sample counts per second
 CP_{sp} = standard counts per second
 F_c = correction factors (half-life, pile-up, etc.).

2. METHOD

A schematic view of the gamma-ray spectrometric system with a Ge detector is shown in Figure 1.

The detector consists of an Ortec HPGe p-type detector (GWL120-15). This system is connected to electronic units, including an Analog-to-Digital Converter interface module integrated with a multichannel analyser and Maestro II software.

This methodology may require correction factors due to pile-up losses (< 0.1 %), source geometry effects (< 0.2 %), and gamma-gamma coincidence summing effects [7]. In this work, a 15 cm source-to-detector distance was adopted to minimize these effects. All measurements of standards and samples were decay-corrected to the same reference date and time.

The liquid source is contained in a glass ampoule with the following dimensions: 5 mL capacity, 90 mm height, 14 mm external diameter, 0.5 mm wall thickness, and a sample solution mass of 2.6–2.7 g, corresponding to a solution height of 20 mm.

Solid sources are prepared by dripping the original solution onto an acrylic disc; after drying, it is covered with another acrylic disc and sealed.

Energies used to activity determination are [8]: ^{22}Na – 511.0 keV and 1274.5 keV; ^{60}Co – 1173.2 keV and 1332.5 keV; ^{133}Ba – 81.0 keV, 276.8 keV, 302.8 keV, 356.0 keV, and 383.8 keV; ^{137}Cs – 661.7 keV. Half-lives ($t_{1/2}$) of radionuclides are: ^{22}Na – 2.60 y; ^{60}Co – 5.27 y; ^{133}Ba – 10.54 y; ^{137}Cs – 30.02 y.

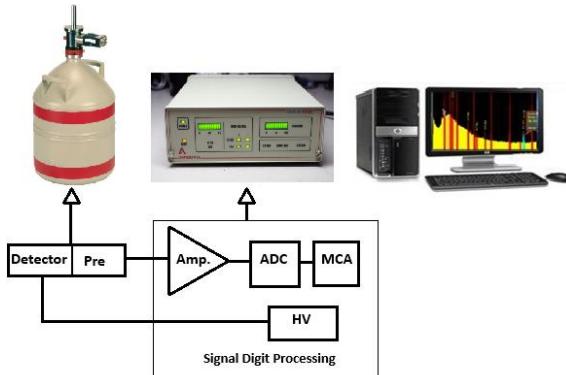


Figure 1. Gamma-ray spectrometric system with a germanium detector.

The statistical uncertainties applied in this work are as follows: A-type uncertainty is calculated from the source counting, while B-type uncertainty depends on factors such as counting geometry, source half-life, detector specifications, and its calibration, among other parameters. Five measurements were performed for each sample. The results were obtained by taking the simple average of three measurement cycles for each standard source, with a counting time of 3600 seconds, ensuring good statistical reliability.

3. RESULTS AND DISCUSSION

Table 1 shows the A-type and B-type uncertainties of the standards used in this work. The total uncertainty range for the activity of liquid standards is between 0.51 % for ^{60}Co and 0.82 % for ^{137}Cs , with $k = 2$.

The total uncertainty range for the activity of solid standards is between 0.40 % for ^{60}Co and 2.3 % for ^{133}Ba . The confidence level used is 95 %. The comparative method using gamma spectrometry is advantageous when a laboratory needs to calibrate many sources daily, due to its fast analytical response.

Table 2 shows the total uncertainty range for activity determination of liquid samples using the comparative method, which is between 1.2 % and 2.0 % ($k = 2$). No significant impurities were observed in the samples. For ^{137}Cs , the total uncertainty obtained was higher than for the other radionuclides due to a high A-type uncertainty of 0.89 %, likely resulting from counting statistics.

Table 3 shows the total uncertainty range for activity determination of solid samples using the comparative method, which is between 0.93 % and 2.7 % ($k = 2$). No significant impurities were observed in the samples.

For ^{133}Ba , the total uncertainty obtained (2.8 %) was higher than for other radionuclides due to a high B-type uncertainty of 1.2 %, probably coming from the standard (Table 3, line 6). The second radionuclide with the highest total uncertainty (2.7 %) is ^{137}Cs (line 8), due to a high A-type uncertainty of 1.2 %, probably coming from the counting statistics.

Here, all samples of ^{60}Co and ^{133}Ba measured have activities lower than those previously reported [9] for the comparative

Table 1. Values of standards radionuclide sources activities with respective uncertainty ($k = 2$).

Nuclide	Form	Activity (Bq/g)	U_A	U_B	U_T
^{60}Co	Liquid	29996	0.17	0.19	0.51
^{133}Ba	Liquid	70014	0.24	0.24	0.68
^{137}Cs	Liquid	47323	0.20	0.36	0.82
^{22}Na	Solid	126306	0.06	0.33	0.67
^{60}Co	Solid	132756	0.14	0.14	0.40
^{133}Ba	Solid	9224	0.32	1.1	2.3
^{137}Cs	Solid	44881	0.25	0.42	0.98

Table 2. Activity values of liquid radionuclide sample sources measured with associated uncertainty ($k = 2$).

Nuclide	Form	Activity (Bq/g)	U_A	U_B	U_T
^{60}Co	Liquid	482442	0.73	0.31	1.6
^{60}Co	Liquid	13949	0.66	0.31	1.5
^{133}Ba	Liquid	109830	0.43	0.39	1.2
^{133}Ba	Liquid	40736	0.45	0.39	1.2
^{137}Cs	Liquid	11752	0.89	0.46	2.0

Table 3. Values of the solid radionuclide sample sources' activities measured and their uncertainty ($k = 2$).

Nuclide	Form	Activity (Bq/g)	U_A	U_B	U_T
^{22}Na	Solid	615917	0.63	0.41	1.5
^{60}Co	Solid	196310	0.59	0.28	1.3
^{60}Co	Solid	103048	0.37	0.28	0.93
^{60}Co	Solid	339502	0.69	0.28	1.5
^{60}Co	Solid	406825	0.68	0.28	1.5
^{133}Ba	Solid	577298	0.70	1.2	2.8
^{137}Cs	Solid	403638	0.63	0.63	1.8
^{137}Cs	Solid	10707	1.2	0.63	2.7
^{137}Cs	Solid	56590	0.69	0.63	1.9
^{137}Cs	Solid	54916	0.59	0.63	1.7
^{137}Cs	Solid	56361	0.70	0.63	1.5

method. In this reference, uncertainties of around 1.3 % for ^{60}Co and 2.8 % for ^{133}Ba were obtained for $k = 2$.

Analysis using the gamma spectrometry technique is still widely used today, and it was chosen for this work due to its speed and the uncertainties accepted by the Radionuclide Metrology Laboratory for determining activity [10]. Different materials can be analysed using this technique. Research on the application of uncertainties can be conducted using spectral simulation programmes [11].

In this work, uncertainties of around 1.5 % were obtained for ^{60}Co and 2.8 % for ^{133}Ba . A confidence level of 95 % was used. The distinct uncertainties measured for these radionuclides were due to different standards, the lower concentrations of the samples in this work, and a different counting geometry.

4. CONCLUSION

The comparative method using a Ge detector was applied to obtain results for radioactive point and ampoule sources. The activities of ^{22}Na , ^{60}Co , ^{133}Ba , and ^{137}Cs radionuclides were determined. Total uncertainties were below 2.0 % for liquid samples and 2.8 % for solid samples, at a 95 % confidence level. The results of this work demonstrate that the comparative method is quick when the standard and sample radionuclides are the same. The measured radionuclides can be used as reference sources for applications in both medicine and industry. Gamma spectrometry with a Ge detector is suitable for determining activity in various materials. This technique is also applied in spectral simulation programmes and for evaluating external exposure from gamma rays in soils and rocks.

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