Radionuclides measurements by the relative method through the germanium detector

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Abstract. The National Laboratory for Ionizing Radiation Metrology (LNMRI/IRD/CNEN) of Rio de Janeiro performed standardization of different radionuclides. Such standards are applied in nuclear industries, nuclear research, radiation protection and nuclear medicine. The comparative method using a Ge detector was applied for obtaining results of some radioactive point and ampoule sources. The activities of ^{22}Na, ^{60}Co, ^{133}Ba and ^{137}Cs radionuclides were determined. The total uncertainties reached values below 1.0 % to liquid samples and below 1.4% to solid samples, in a confidence level of 68 %.

1. Introduction

The National Laboratory of Metrology of Ionizing Radiation (LNMRI) under the National Nuclear Energy Commission (CNEN) is the body designated by the National Institute of Metrology and Quality Technology (INMETRO) as National Metrology Laboratory for quantities associated with ionizing radiation. This laboratory participates in intercomparison in order to ensure the traceability of measurements in the worldwide network of metrology, coordinated by the Bureau International des Poids et Mesures (BIPM) and the Inter-American Metrology System (SIM).

It offers various systems and calibration methods for gamma-emitter radioactive sources in various geometries which can be absolute or relative (comparative or efficiency curve). The high-resolution gamma spectrometry by high purity germanium detector (HPGe) is applied to the analysis of emitting photons radionuclides [1]. This is a relative technique, because uses of standards for determining the activity of the samples is compared to a radionuclide of the same nature.

Gamma spectrometry using the germanium detector is used in nuclear industries, nuclear research, radiation protection and nuclear medicine.

When the calibration of gamma-ray spectrometry using HPGe detector is required for determination of the activity and/or multi-gamma emitter samples, a source-detector distance should be used [2]. This distance should reduce dead-time and true coincidence summing effects [3].

The comparative method is still largely used in direction to obtain the activity results with high accuracy and precision. The equation of the sample (Ax) activity determination is :

$$Ax = Ap \{ CPSx/CPSp \}. Fc \qquad (1)$$

Where :
Ap = standard activity in the same reference date
Ax = sample activity in the same reference date
CPSp = standard counts per second
CPSx = sample counts per second
Fc = correction factors (half-life, pile-up, etc)

^{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 it has a multi-gamma spectrum.

One of the radioisotopes produced in the ^{235}U nuclear fission reaction and can be released into the atmosphere is ^{137}Cs [4]. This radioisotope can be absorbed by the plants and in this way contaminate human food meat, milk) [5].

^{22}Na isotope is used as a radiotracer. It is produced in IPEN/SP cyclotron using collision reaction between ^{24}Mg and ^{2}H (deuterium) [6]. This radioisotope can be used in PET-Scan (positron emission tomography) which is a diagnostic imaging exam.

2. Method

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

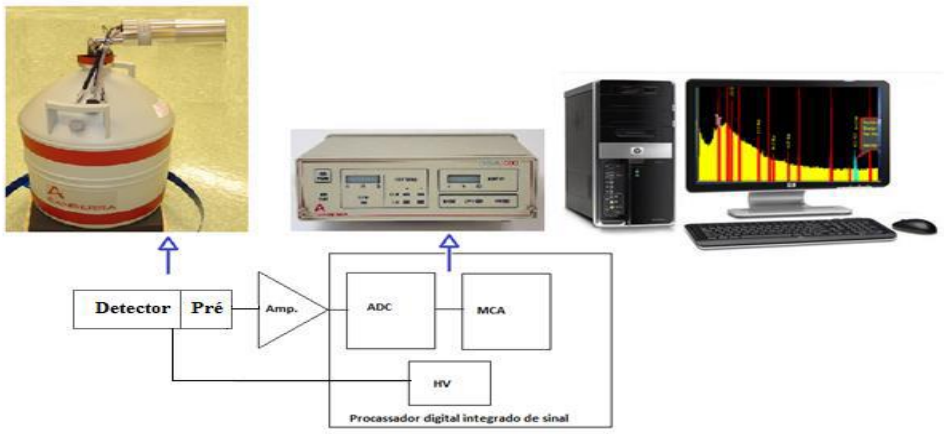

Figure 1. Gamma-ray spectrometric system with germanium detector

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The detector consists of an Ortec HPGe pit detector (GWL120-15). This system is associated with electronic units including an Analog-to-Digital Converter interface module integrated to multichannel analyzer and Maestro II software.

This methodology can require correction factors due to pile-up losses (< 0.1 %), source geometry effects (< 0.2 %), and gamma-gamma summing coincidence effects [3]. In this work it was adopted a 10 cm distance between detector-source in order to reduce these effects.

The liquid source is a glass ampoule, it has 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 g - 2.7 g that corresponding of 20 mm of solution height.

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 [7]: ^{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.

3. Results and Discussion

Table 1: Values of standards radionuclide sources activities with respective uncertainty

Nuclide	Shape	Code	Activity (Bq/g)	U_A	U_B
^{60}Co	liquid	67	29996	0.17	0.19
^{133}Ba	liquid	05	70014	0.24	0.24
^{137}Cs	liquid	13	47323	0.20	0.36
^{22}Na	solid	129S08	126306	0.06	0.33
^{60}Co	solid	21S02	132756	0.14	0.14
^{133}Ba	solid	129S06	9224	0.32	1.1
^{137}Cs	solid	37S97	44881	0.25	0.42

Table 1 shows the type A and type B uncertainties of the standards that were used in this work . The range of the total uncertainties of the liquid standards activity is between 0.25 % of ^{60}Co to 0.41 % of ^{137}Cs .

The range of the total uncertainties of the solid standards activity is between 0.20 % of ^{60}Co to 1.15 % of ^{133}Ba. The confidence level to be used is 68 %.

The comparative method by gamma spectrometry is advantageous when a laboratory has a lot of sources to be calibrated on a daily basis due to a quick analysis response.

Table 2: Values of liquid radionuclide samples sources activities measured with uncertainty activity (k=1). It was used at 12:00 h to calibrate all samples.

Nuclide	Shape	Measured Activity (Bq/g)	U_A	U_B	Activity uncertainty (%) (k = 1)
^{60}Co	liquid	482.442	0.73	0.31	0.79
^{60}Co	liquid	13.949	0.66	0.31	0.72
^{133}Ba	liquid	109.830	0.43	0.39	0.59
^{133}Ba	liquid	40.736	0.45	0.39	0.60
^{137}Cs	liquid	11.752	0.89	0.46	1.0

In these Table 2 the range of the total uncertainties of the activity determination to liquid samples by comparative method is between 0.59 % to 1.0 %. There were not observed any significant impurities on the samples.

For ^{137}Cs the total uncertainty obtained was higher than others radionuclides due to high type A uncertainty, 0.89 %, probably come from the counting statistic.

Table 3: Values of solid radionuclide samples sources activities measured with uncertainty activity (k=1). It was used at 12:00 h to calibrate all samples.

Nuclide	Shape	Measured Activity (Bq/g)	U_A	U_B	Activity uncertainty (%) (k = 1)
^{22}Na	solid	615.917	0.63	0.41	0.76
^{60}Co	solid	196.310	0.59	0.28	0.66
^{60}Co	solid	103.048	0.37	0.28	0.47
^{60}Co	solid	339.502	0.69	0.28	0.75
^{60}Co	solid	406.825	0.68	0.28	0.74
^{133}Ba	solid	577.298	0.70	1.2	1.4
^{137}Cs	solid	403.638	0.63	0.63	0.90
^{137}Cs	solid	10.707	1.2	0.63	1.3
^{137}Cs	solida	56.590	0.69	0.63	0.94
^{137}Cs	solid	54.916	0.59	0.63	0.87
^{137}Cs	solid	56.361	0.70	0.63	0.94

In Table 3 the range of the total uncertainties of the activity determination solid samples by comparative method is between 0.47 % to 1.40 %. There were not observed any significant impurities on the samples.

For ^{133}Ba the total uncertainty obtained (1.40%) was higher than others radionuclides due to high type B uncertainty, 1.20 %, probably come from the standard (Table 1 line 6). The second radionuclide with higher total uncertainty (1.30 %) is ^{137}Cs (line 8) due to high type A uncertainty, 1.20 %, probably come from the counting statistic.

Here all samples of ^{60}Co and ^{133}Ba measured have activities lower than previously reported [8] for the comparative method . In this reference [8] uncertainties around 0.4 % were obtained for ^{60}Co and 0.7 % for ^{133}Ba.

In this present work, uncertainties around 0.7 % were obtained for ^{60}Co and 1 % for ^{133}Ba. Confidence level used was 68%. The distinct uncertainties measured for these radionuclides were due

to: different standards, the lower concentrations of the samples in this work and different germanium detectors (coaxial and well).

4. Conclusion

The comparative method using a Ge detector was applied for obtaining results of some radioactive point and ampoule sources. The activities of ^{22}Na, ^{60}Co, ^{133}Ba and ^{137}Cs radionuclides were determined. The total uncertainties reached values below 1.0 % to liquid samples and 1.4 % to solid samples, in a confidence level of 68 %. These work results showed the comparative method is fast when the radionuclides of the standards are the same as those of the samples. These measured radionuclides can be used as a reference sources and applied both in medicine and in industry.

5. References

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