

¹³⁴Cs emission probabilities determination by gamma spectrometry

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Abstract: The National Laboratory for Ionizing Radiation Metrology (LNMRI/IRD/CNEN) of Rio de Janeiro performed primary and secondary standardization of different radionuclides reaching satisfactory uncertainties. A solution of ¹³⁴Cs radionuclide was purchased from commercial supplier to emission probabilities determination of some of its energies. ¹³⁴Cs is a beta gamma emitter with 754 days of half-life. This radionuclide is used as standard in environmental, water and food control. It is also important to germanium detector calibration. The gamma emission probabilities (P_γ) were determined mainly for some energies of the ¹³⁴Cs by efficiency curve method and the P_γ absolute uncertainties obtained were below 1% ($k=1$).

Keywords: gamma emission probability, ¹³⁴Cs, HPGe, efficiency curve

1. INTRODUCTION

¹³⁴Cs is a radioisotope of extreme importance for the calibration of HPGe spectrometers, among others. The determination of nuclear parameters beyond the absolute supply activity should be considered as a factor of relevance.

The determination of the gamma emission probabilities, for example, allows improvements in the characterization of this radioisotope

depending on the accuracy of the measured values and serves as quality indicator of the spectrometry system and the methodology used in the determination of this parameter. Another factor that highlights the use of ¹³⁴Cs is that through the determination of efficiency one can use this radioisotope as a plotter to measure ¹³⁷Cs.

2. METHODOLOGY

2.1 Relative Efficiency Curve Method

The efficiency curve was obtained using various radionuclidic standardized sources such as: ^{166m}Ho , ^{152}Eu , ^{137}Cs , ^{241}Am , ^{65}Zn totaling 59 energy points and the graphic can be observed in Figure 1. The HPGe spectrometric system has been calibrated in efficiency through the use of standard point sources [3]. The range of energy was established between 48 keV and 1427 keV originally. Then a cut was made considering only the energies above 300 keV since the low energy region was not necessary for the purpose of the calibration. The efficiency curve fitting was performed by a 5th degree polynomial.

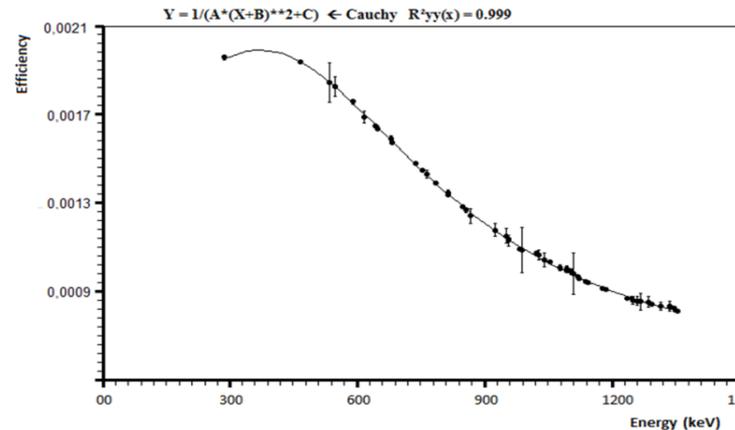
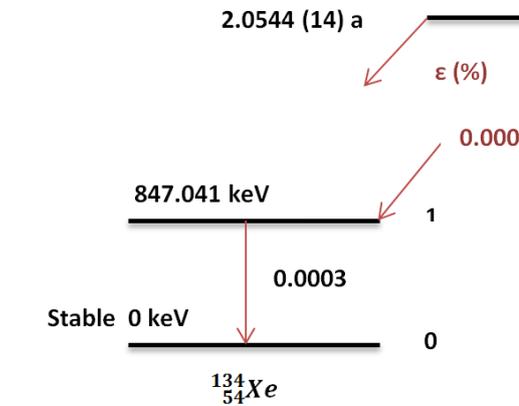


Figure 1. Efficiency curve with ^{166m}Ho , ^{152}Eu , ^{133}Ba , ^{137}Cs , ^{241}Am , ^{54}Mn and ^{65}Zn radionuclidic standards



^{134}Cs has a radioactive half-life of 2.06 years and it has two mechanisms by which it can disintegrate. One mode is by β^- emission which is the most likely occurrence option being responsible for 99.9997% of the disintegrations. Only 0.0003% of the disintegrations occur by electronic capture and positron emission (β^+).

In the event of a nuclear accident, such as the leakage or explosion of a reactor as happened in 1986 in Chernobyl, Ukraine, some radioisotopes produced in the ^{235}U nuclear fission reaction are released into the atmosphere, such as ^{131}I , ^{137}Cs , ^{134}Cs and ^{90}Sr [1]. After this accident, Brazil worried about the importation of foods that could have the presence of radioisotopes ^{137}Cs , ^{134}Cs and ^{90}Sr . These radioisotopes are absorbed by the plants that are consumed by the animals resulting in the possible contamination of imported beef and milk. [2]

2.2 Gamma-ray emission probability measurements

In order to associate the main peaks of the spectrum to the radionuclide, the energy-channel relation of a spectrometer needs to be obtained. Afterwards, the total absorption efficiency curve is determined, as a function of energy, and the radionuclide activity may be calculated from region of interest. The expression that represents an activity area is:

$$\frac{CPS_{corrected}}{P_{\gamma}\epsilon_{\gamma}} \quad (1)$$

where: $CPS_{corrected}$ is the count rate of full energy peak; ϵ_{γ} is the full energy peak efficiency for specific gamma energy; and P_{γ} is the emission probability for specific gamma energy.

However, as the source activity is known, P_{γ} was calculated for ranges of 475 keV, 563 keV, 569 keV, 604 keV, 796 keV, 802 keV, 1039 keV, 1168 keV and 1365keV to ^{134}Cs by means of the following expression, taking into account the corrections as decay, background and positioning:

$$P_{\gamma} = \frac{CPS_{corrected}}{\epsilon_{\gamma} \cdot A} \quad (2)$$

where A is the absolute activity. The P_{γ} determination depends on the precision achieved in efficiency curve

2.3 Source preparation and measurements

^{134}Cs point source was prepared by means of the pycnometer technique, depositing drops of radionuclide solution in a polystyrene film, with a thickness of 0.05 mm, set in one acrylic ring. The ring has an external diameter of 25 mm, inner diameter of 4 mm and a thickness of 1 mm. Once dried, the source was covered with the same polystyrene film.

The spectrometry system used consist of HPGe detector type a planar with beryllium window with 20% of relative efficiency (d3gem). Some appropriate electronics composed basically of the elements: High voltage supply, signal amplifier and a multichannel analyzer. The multichannel analyzer associated with the data acquisition program is responsible for subtracting background beyond managing dead time.

The conditions of ^{134}Cs source measurements are :HPGe detector position-d3gemp4 (20cm); source activity : 2820.71 Bq at 12:00h of date 20170601. This activity was determined in the LNMRI laboratory by $4\pi\beta(\text{LS})-\gamma(\text{Na}(\text{Tl}))$ anticoincidence method with 0.38% of uncertainty (k=1) [4].

3.RESULTS

The components of the uncertainties considered for the determination of gamma emission probability were :uncertainty of the photopic area, uncertainty in the activity, uncertainty in efficiency and uncertainty in ^{134}Cs decay.

TABLE 1. Counts per second to the different energies of ^{134}Cs

Energy (keV)	Area	Uncertainty u_a (area)	CPS _{corrected} (Bq/s)
475	16538	322	0.0476
563	82224	421	0.2366
569	149331	499	0.4297
604	854624	965	2.4591
796	630766	811	1.8149
802	62050	268	0.1785
1039	5796	92	0.0166
1168	9681	105	0.0279
1365	14951	123	0.0430

The gamma spectrometry system was stable throughout the measurement. During ^{134}Cs spectrum acquisition some problems can occur when considering the peaks of 563 keV and 569 keV that overlap over the 605 keV peak when using a NaI (Tl) type detector but in this case the use of the HPGe detector stands out for such use because it has a much greater resolution power and facilitates the separation of these peaks with greater ease.

TABLE 2. Results of gamma emission probabilities to ^{134}Cs energies

Energy (keV)	GammaEfficiency	P_γ (%)
475	0.001278	1.509(7)
563	0.001135	8.446(44)
569	0.001115	15.625(39)
604	0.001034	96.41(85)
796	0.000861	85.48(77)
802	0.000857	8.440(24)
1039	0.000673	1.0039 (19)
1168	0.000618	1.827(18)
1365	0.000579	3.013(21)

The results of gamma emission probabilities (P_γ) to ^{134}Cs energies are near with the LNHB results [5].

The absolute uncertainties results, Table 2, for the P_γ of each energy are within the expected although the values were higher than those of the LNHB, Table 3. This fact is also due mainly to the uncertainties of the efficiency curve and the source activity.

TABLE 3. LNHB [5] data of the gamma emission probabilities (P_γ) to ^{134}Cs energies

Energy (keV)	P_γ
475	1.479 (7)
563	8.342 (15)
569	15.368 (21)
604	97.63 (8)
796	85.47 (9)
802	8.694 (16)
1039	0.9909 (33)
1168	1.791 (5)
1365	3.019 (8)

TABLE 4. Deviation between this work results and LNHB [5] data of the gamma emission probabilities (P_γ) to ^{134}Cs energies

Energy (keV)	ΔP_γ (%)
475	+ 2.03
563	+ 1.25
569	+ 1.67
604	- 1.25
796	+ 0.01
802	- 2.92
1039	+ 1.31
1168	+ 2.01
1365	- 0.20

The calculation of the deviations of P_γ results, Table 4, from each energy was:

$$\Delta P_\gamma (\%) = \frac{[(P_\gamma_{\text{WORK}} - P_\gamma_{\text{LNHB}}) / P_\gamma_{\text{LNHB}}] * 100}{(3)}$$

The experimental results of P_γ showed a tendency of higher values than those of the literature [5], anyway the results were close.

4. CONCLUSION

The efficiency curve method is widely used for radioisotopes whose half-life is not long as in the case of the ^{134}Cs . Satisfactory results were obtained for the gamma emission probabilities in which a mean deviation was obtained compared to the literature of 0.43%. The gamma emission probabilities were determined mainly for some energies of the ^{134}Cs by efficiency curve method and the absolute uncertainties obtained were below 1% ($k=1$).

5. REFERENCES

[1] Gazal S and Prange H2005 *Radioprotection, Suppl.1.* **40S747**

[2] Pawel J and Kalita S J 2010 *Nukleonika* **55** (2)143

[3] Dabb H M 2015 *Arab Journal of Nuclear Science and Applications* **48** (4) 53

[4] da Silva C J et al . “The ^{134}Cs activity determination by $4\pi\beta(\text{LS})-\gamma(\text{Na}(\text{Tl}))$ anticoincidence method” will be published.

[5] LNE-LNHB/CEA 2012 Table de Radionucléides :

www.nucleide.or/Cs-134-tables