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Primary Standardization of ^{152}Eu by $4\pi\beta$(LS) – γ (NaI) coincidence counting and CIEMAT-NIST method

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Abstract: The $4\pi\beta$-γ coincidence counting and CIEMAT/NIST liquid scintillation method were used in the standardization of a solution of ^{152}Eu. In CIEMAT/NIST method, measurements were performed in a Liquid Scintillation Counter model Wallac 1414. In the $4\pi\beta$-γ coincidence counting, the solution was standardized using a coincidence method with ‘‘beta–efficiency extrapolation”. A simple $4\pi\beta$-γ coincidence system was used, with acrylic scintillation cell coupled to two coincident photomultipliers at 180° each other and NaI(Tl) detector. The activity concentrations obtained were 156.934 ± 0.722 and 157.403 ± 0.113 kBq/g, respectively, for CIEMAT/NIST and $4\pi\beta$-γ coincidence counting measurement methods.

Keywords: Standardization; Coincidence counting; CIEMAT/NIST method

1. INTRODUCTION

^{152}Eu is widely used as gamma standard spectrometry sources for the calibration of solid-state detectors. The main method used for the standardization of radionuclide activity by most National Metrology Institutes worldwide is the $4\pi\beta$-γ coincidence counting method, a primary method that has great versatility as may be used for those radionuclides which decay by alpha-gamma and beta-gamma emission, electron capture and pure beta emitters [1].

Another method currently used for determination of activity is the CIEMAT/NIST liquid scintillation method, consisting of three or two photomultipliers based on statistical Free Parameter model of the scintillation photons distribution and their detection probabilities [2].

The decay of ^{152}Eu is divided in branches of β^- decay (27.8%), followed by photon emission, and electron capture (72.2%), with subsequent gamma emission. The gamma-ray spectrum ranges from 121 to 1769 keV, with 85 gamma emissions. The half-life of ^{152}Eu is (13.53 ± 003) years [3, 4]. Due the complex decay of ^{152}Eu, the activity determination is complicated, creating difficulties in the absolute standardization of this nuclide. The main features of the complex decay are shown in Figure 1.

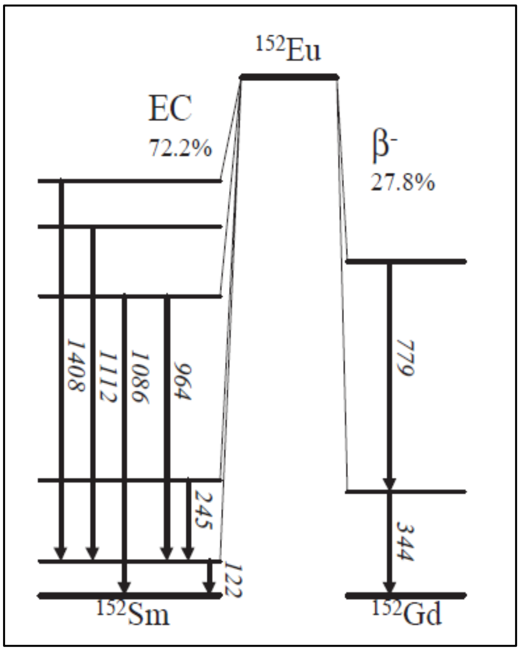

Figure 1. Simplified decay scheme of ^{152}Eu

2. METODOLOGY

2.1. 4πβ-γ coincidence counting method

This well-known counting method for absolute activity measurement is commonly used for decay modes including β-emission, electron capture or a mixture of the two [6, 7].

For the determination of the activity, 14 sources were prepared by adding aliquots of ^{152}Eu solution in a glass vial with 10 mL of commercial cocktail Ultima Gold, manufactured by Perkin Elmer/USA. Sources were measured in 4πβ(LS)-γ (NaI) coincidence system, consisting of an acrylic scintillation cell coupled to two coincident photomultipliers at 180° each other, in which the scintillation vial is placed and the generated signals analyzed and processed in a conventional electronic chain, forming the beta channel.

The signals come mainly from the Auger electrons and characteristic X-rays generated by the electronic capture events. The counting rate of these events is called N_β. A 10 x 10 cm^2 NaI(Tl) crystal is placed at the bottom of the acrylic scintillation cell for detection of gamma rays whose signals are analyzed and processed in a conventional electronic chain, forming the gamma channel. The counting rate of these events is denominated N_γ. The counting rate of pulses from beta and gamma channels arriving simultaneously in a coincidence circuit with a fixed resolution time of 1.27 µs is denominated N_c. The activity was determined by means of a variation of the mean beta efficiency, ε_β, by electronic discrimination and applying the extrapolation method to the measurement results [8, 9]. The count rate $N_{\beta,}$ can be described by

$$N_\beta = N_0 \sum a_i \left[\varepsilon_{\beta i} + (1-\varepsilon_{\beta i}) \frac{\alpha_i}{1+\alpha_i} \varepsilon_{cei} \right] \qquad (1)$$

where a_i is the branching ratio of *i*th transition, α_i is the internal conversion coefficient for the γ-transition(s) of the *i*th branch and ε_{ce} is the efficiency by which the conversion electrons are detected by the β-detector [10].

2.2. CIEMAT/NIST measurement method

This counting system consists of two photomultipliers placed in coincidence at 180° to each other. The CIEMAT/NIST method uses a set of standard tracer tritium (^{3}H) samples for the commercial scintillator characterizing in terms of the quenching parameter by the introduction of the chemical quenching agent in increasing degree. Figure 2 shows the correlation between Free Parameter, quenching and efficiencies for standard tracer and radionuclide in analysis by CIEMAT/NIST method [2, 5].

The equation 2 gives the detection efficiency for two photomultipliers obtained from Poisson model.

$$\varepsilon = \left(1 - e^{-\frac{\nu m}{2}}\right)^2 \qquad (2)$$

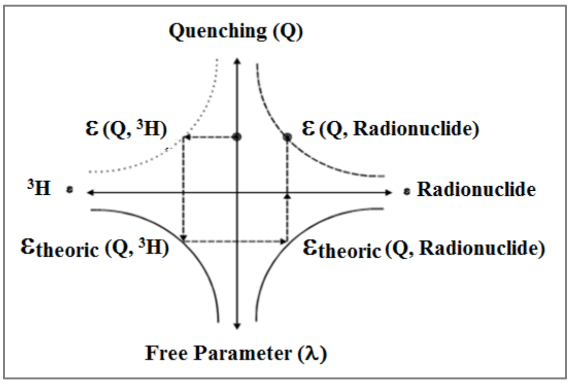

Figure 2. CIEMAT/NIST model for the radionuclide standardization

3. RESULTS AND DISCUSSION

3.1. 4πβ-γ coincidence counting method

The activity was determined through the variation of the beta efficiency by electronic discrimination, and applying an extrapolation technique to the measurement data. Two levels of gamma discrimination were adjusted for the counts in the gamma channel: 100 and 180 keV.

To obtain the extrapolation curve, the expression $(N_\beta N_\gamma) / N_c$ is plotted as a function of $(1 - \varepsilon_\beta) / \varepsilon_\beta$ and extrapolated to $\varepsilon_\beta = 1$. The function is linear in the region of high efficiencies [6]. ε_β efficiencies ranged from 0.78 to 0.55 and 0.81 to 0.72 for the gamma window range of 100 and 180 keV, respectively. The extrapolated value to $(1-N_c/N_\gamma)/(N_c/N_\gamma)$, where $\varepsilon_\beta=1$, provides the activity of the radioactive solution from which the sources were prepared. Figure 3 presents the extrapolation curves on the coincidence measurements of all 14 sources.

The consistency of the results of value of activities from all sources was verified using the statistical test called "Birge ratio" [11]. The calculated value with the experimental results was 1.17. The value adopted for the activity concentration was the weighted average of the 14 results: 157.403 ± 0.113 kBq/g, where the uncertainty refers to the standard deviation of the mean of the results. Table 1 presents the components of the uncertainties in determining the activity of ^{152}E by the coincidence method.

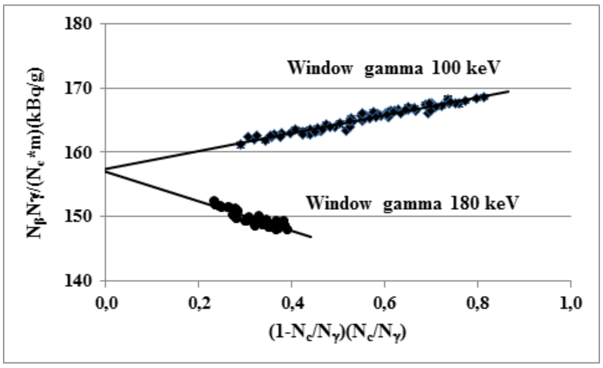

Figure 3. Extrapolation applied to the measurements by 4πβ-γ coincidence counting on all sources, measurements at gamma channel of 100 and 180 keV.

Table 1. ^{152}Eu uncertainty components by coincidence method.

Components	Type	Relative standard uncertainty (%)
Counting statistics	A	0.50
Weighing	B	0.050
Background	A	0.030
Dead-time	B	0.033
Resolving time	B	0.20
Gandy effect	B	0.093
Fitting procedure	A	0.072
Decay correction	B	0.00033
Combined		0.60
Expanded (k=2)		1.2

3.2. CIEMAT / NIST measurement method

8 sources were prepared by adding aliquots of ^{152}Eu solution in a glass vial with 10 mL of commercial cocktail Ultima Gold, manufactured by Perkin Elmer/USA. During the measurement period the samples remained stable, showing no tendency. Measurements were performed in a Liquid Scintillation Counter model Wallac 1414.

The activity was calculated using the CN2003 computational code [12]. The value adopted for the activity concentration was the weighted average of the 8 results: 156,934 ± 0,722 kBq/g, where the uncertainty refers to the standard deviation of the mean of the results. Table 2 presents the components of the uncertainties in determining the activity of ^{152}E by the CIEMAT / NIST method.

Table 2. ^{152}Eu uncertainty components by CIEMAT/NIST method.

Components	Type	Relative standard uncertainty (%)
^{3}H activity	B	0.20
Weighing	B	0.05
^{3}H Quenching (SQPE)	A	0.05
Nuclear and Atomic data	B	0.13
Photomultiplier asymmetry	B	0.23
kB value	A	0.10
^{152}Eu quenching (SQPE)	A	0.09
Statistic counts	A	0.28
Combined		0.46
Expanded (k=2)		0.92

Figure 4 summarizes the results of two absolute measurements methods applied in the determination of the activity concentration of the ^{152}Eu solution. The uncertainties bars refer to combined uncertainty (coverage factor k = 1).

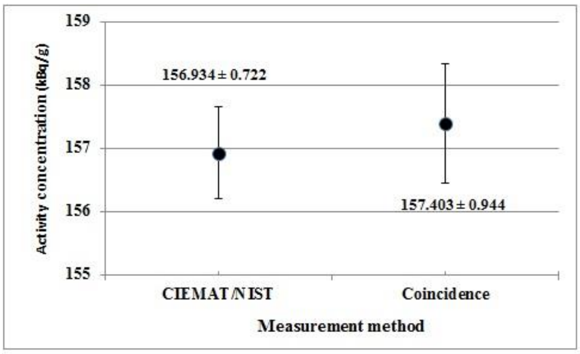

Figure 4. Activity concentration of the ^{152}Eu solution determined by two absolute methods.

4. CONCLUSION

The results of the standardization of ^{152}Eu solution by the absolute CIEMAT / NIST and $4\pi\beta$(LS)-γ(NaI) methods were consistent within the uncertainties evaluated at a confidence level of ≈ 68% (k = 1). The largest contributions to assessing the combined uncertainty came from the counting statistics for both methods indicating the need to increase the counting time or the mass of the sources to minimize these uncertainties. As a reference activity for calibration of other secondary calibration systems in operation at LNMRI, the value of the CIEMAT / NIST method activity was adopted because it presented a lower combined uncertainty (0.92%) than that of coincidence (1.2%).

5. REFERENCES

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