

⁶⁵Zn and ¹³³Ba standardizing by photon-photon coincidence counting

Jamir S. Loureiro¹, Paulo A. L da Cruz¹, Akira Iwahara¹, José U. Delgado¹, Ricardo T. Lopes²

¹Laboratório Nacional de Metrologia das Radiações Ionizantes/Instituto de Radioproteção e Dosimetria/Comissão Nacional de Energia Nuclear (LNMRI/IRD/CNEN), Av. Salvador Allende s/n, Barra da Tijuca, CEP 22783-127, Rio de Janeiro/Brasil

²Programa de Engenharia Nuclear, Universidade Federal do Rio de Janeiro (PEN/COPPE, UFRJ), Ilha do Fundão, Caixa Postal 68590, CEP 21945-970, Rio de Janeiro, Brasil

E-mail: palcruz@ird.gov.br

Abstract: The LNMRI/Brazil has deployed a system using X-gamma coincidence technique for the standardizing radionuclide, which present simple and complex decay scheme with X-rays of energy below 100 keV. The work was carried on radionuclide metrology laboratory using a sodium iodide detector, for gamma photons, in combination with a high purity germanium detector for X-rays. Samples of ⁶⁵Zn and ¹³³Ba were standardized and the results for both radionuclides showed good precision and accuracy when compared with reference values. The standardization differences were 0.72 % for ⁶⁵Zn and 0.48 % for ¹³³Ba samples.

Keywords: ⁶⁵Zn; ¹³³Ba; standardization; photon-photon coincidence counting.

1. INTRODUCTION

The importance of the development and implementation of an absolute measurement technique for the radionuclide standardizing is highlighted. It is, in short, the state of the art, due to the complexity and specificity of the radionuclide decay-schemes.

The existence of several techniques enriches and makes the metrological chain robust, in order to provide results with traceability and reliability related to the determination of atomic and nuclear data and the activity quantity.

Techniques for absolute radionuclide standardizing are still few and rare, because their recognition and adoption by the international metrology system requires de confirmation of experimentation and theoretical formulation for years, finally enshrined through key-comparisons

promoted by Bureau International des Poids et Mesures (BIPM).

The coincidence $4\pi\beta(\text{PC})-\gamma(\text{NaI})$ counting based on gas flow proportional counter have been developed [1,2,3,4], rising to $4\pi\beta(\text{LS})-\gamma(\text{NaI})$ based on liquid scintillation [5,6]. The anticoincidence $4\pi\beta(\text{PC})-\gamma(\text{NaI})$ also based on gas flow proportional counter have been developed [7,8,9] rising to $4\pi\beta(\text{LS})-\gamma(\text{NaI})$ based on liquid scintillation [10,11], in the actually.

With respect to the beta-emitting radionuclides only from the 80's the theoretical and experimental formulation was developed using the liquid scintillation technique by CNET (CIEMAT/NIST Efficiency Tracing) [12,13,[14] and TDCR (Triple-to Double Coincidence Ratio) [15,16] methods.

The development of solid Germanium and Lithium detectors has given new possibilities to gamma-spectrometry technique such as in radionuclide absolute standardizing by the sum-peak [17,18] and photon-photon [19,20] coincidence counting methods.

This work presents the implementation of absolute radionuclide measurement by photon-photon coincidence counting in LNMRI/Brazil, applied to the standardization of ^{65}Zn and ^{133}Ba , examples of simple and complex decays, respectively. In the specific case, the uses of X-gamma emissions do not involve corrections due to angular correlation and simplifies the experimentation and theoretical formulation.

The ^{65}Zn is used in nuclear medicine and serves to calibrate detectors, as a sodium iodide. The ^{133}Ba is a radionuclide with half-life relatively long and therefore used to calibrate germanium detectors in low energy region, serving as a source for consistency tests in radionuclide calibrator.

2. EXPERIMENTAL PROCEDURE

The photon-photon coincidence system was implemented in the LNMRI to standardize radioactive solution with X-rays below 100 keV.

The reduction of HPGe detector efficiency was performed moving away the radioactive sample from detector or introducing absorbers between the sample and this detector.

In the figures 1 and 2 are displayed a drawing and a block diagram symbolizing the electronics modules used in the system.

The upper part of the figure 2 displays the modules used for photon spectrometry methods. The bottom part of the figure 2 displays the modules used for coincidence with absolute standardizing methods. The connections with dotted lines indicate the modules used only during previous adjust of the instrumentations.

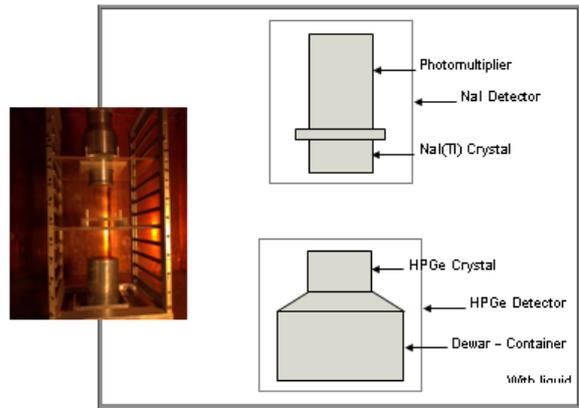


Figure 1. X- γ coincidence counting NaI(Tl) and HPGe detectors.

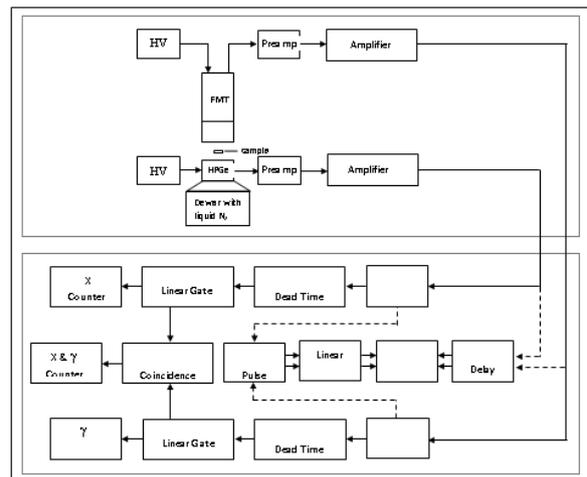
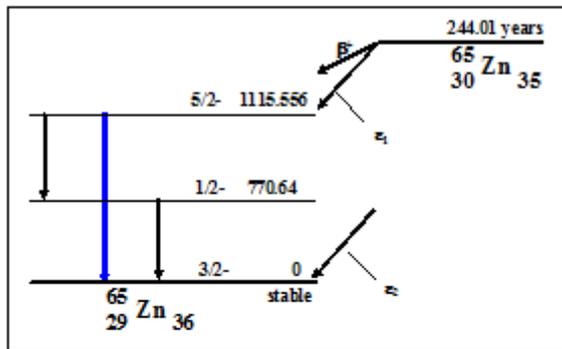


Figure 2. Block diagram of X- γ coincidence system working with the NaI(Tl) and HPGe detectors.

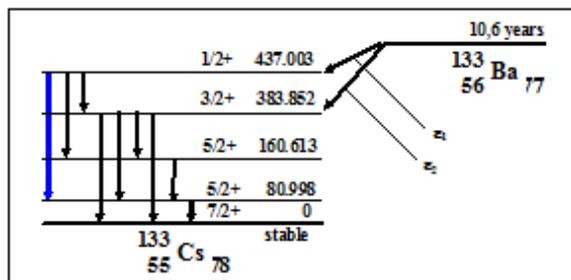
The ^{65}Zn disintegrates by electron capture to the 1115 keV excited level and beta plus emission to the ground state level of ^{65}Cu , as shown in figure 3.

The ^{133}Ba disintegrates by electron capture, mainly to two ^{133}Cs excited levels of 437 keV (85.4 %) and of 383 keV (14.5 %) with three very minor branches to the 160 keV, 81 keV excited levels and the ground state, as shown in figure 4.



X Rays	Energy (keV)	Relative probability
X _{Kα}		
K _{α1}	8.03	51
K _{α2}	8.05	100
K _{β1}	8.91	21,05
K _{β2}	8.98	
X _L		
L _α	3.80	
L _β	0.81	
L _γ	0.93	
L _δ	5.70	

Figure 3. Simplified ⁶⁵Zn decay scheme with the selected highlighted events for counting.



X Rays	Energy (keV)	Relative probability
X _{Kα}		
K _{α1}	30.63	54
K _{α2}	30.97	100
K _{β1}	34.92	29.5
K _{β2}	34.99	
K _{β3}	35.25	
K _{β4}	35.26	
K _{β5}	35.82	6.7
K _{β6}	35.90	
K _{β7}	35.97	
X _L		
L _α	3.80	
L _β	5.70	

Figure 4. Simplified ¹³³Ba decay scheme with the selected highlighted events for counting.

The activities were determined by extrapolation of apparent activities curves in function of the relative inefficiency factor [3], reducing HPGe detector efficiency. To decrease X-rays near 8 keV of intensity, was increased the distance between of ⁶⁵Zn samples and HPGe

detector by steps of 0.5 cm until the maximum of 5 cm. This movement was done keeping unchanged the distance between the sample and NaI detector, while at the same time the distance from both and HPGe detector was being increased. To decrease X-rays near 30 keV of intensity, were inserted between of ¹³³Ba samples and detector until 9 aluminum discs, each one with 0.5 cm thick.

Three ⁶⁵Zn samples were prepared by deposition of standard radioactive solution on a plastic tape fixed at the bottom of an acrylic ring, covered by another similar plastic tape. A total of four ¹³³Ba samples were prepared on a thin plastic film of polyvinyl chloride (VYNS) and measured by photon-photon system. The validation of this system was carried out with the measuring of the same ¹³³Ba samples by 4πβ-γ(NaI) coincidence counting system.

The measurements were done adjusting each Single Channel Analyzer – SCA with pulse height suitable to detect the selected events. For ⁶⁵Zn samples were considered gamma photons of 1115 keV (SCA windows from 7.40 V to 10 V) and X-rays from 8 keV to 9 keV (SCA window from 0.15 V to 0.30 V). For ¹³³Ba samples were considered gamma photons of 356 keV (SCA windows from 2.60 V to 3.32 V) and X-rays from 30 keV to 36 keV (SCA window from 0.60 V to 0.98 V)

3. RESULTS

The apparent activities were calculated for each detection efficiency variation by software deployed in the LNMRI/IRD, which uses the coincidence method [21,22,23].

As the selected events for counting are mono-energetic gamma photons and the X-rays produced by electron capture, originated in the same decay, and considering too that there's no overlapping in the energy range of the two kinds of selected events, each channel of the instrumentation certainly will register only their

respective events and no other one. In these conditions, the $N\beta$ of the BAERG equation for ideal detectors can be substituted by N_X to determine all apparent activities with good accuracy without need introduction of any correction factor in the value found after the experimental curve extrapolation.

$$N_0 = \frac{N_X N_Y}{N_C} \quad (1)$$

where, N_0 is apparent activity; N_X is X-ray count rate of HPGe detector; and N_Y is gamma count rate of NaI(Tl) detector; and N_C is the X-gamma count rate coincidence. For each experimental curve, the sample activities were determined through linear regression and extrapolation to zero value of the relative inefficiency factor.

The figure 5 displays the apparent activity curve in function of relative inefficiency factor for the ^{65}Zn sources. The mean activity concentration found in these samples was $646.02 \text{ kBq/g} \pm 0.56 \%$, with the uncertainty determination showed in Table 1. The figure 6 displays the apparent activity curve in function of relative inefficiency factor for the ^{133}Ba sources. The mean activity concentration found in these samples was $504.23 \text{ kBq/g} \pm 0.39 \%$, with the uncertainty determination showed in Table 2.

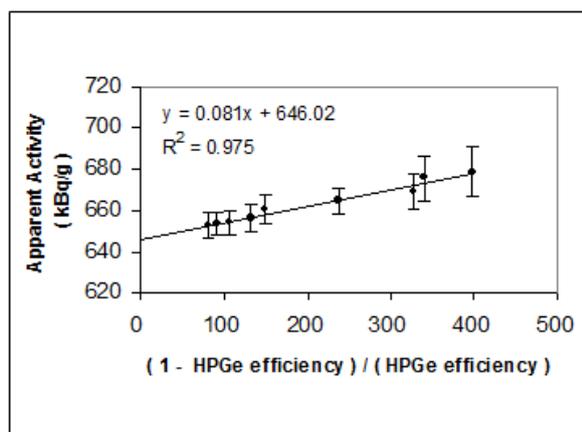


Figure 5. Apparent activity produced by relative inefficiency factor for the ^{65}Zn samples.

Table 1. Uncertainty budget ($k = 1$) of the ^{65}Zn solution standardization.

Components	Type	Uncertainty (%)
Statistic counts	A	0.45
Half-life	B	0.04
Background	A	0.03
Mass	B	0.05
Extrapolation	A	0.033
Combined uncertainty ($k=1$)		0.56

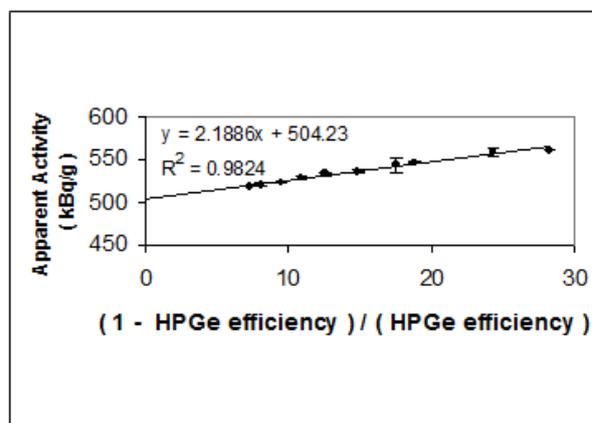


Figure 6. Apparent activity produced by relative inefficiency factor for the ^{133}Ba samples.

Table 2. Uncertainty budget ($k = 1$) of the ^{133}Ba solution standardization.

Components	Type	Uncertainty (%)
Statistic counts	A	0.38
Half-life	B	0.04
Background	A	0.02
Mass	B	0.05
Extrapolation	A	0.004
Combined uncertainty ($k = 1$)		0.39

The Tables 3 and 4 display the comparisons between standardization result for ^{65}Zn and ^{133}Ba samples using the X-gamma coincidence system and their registered reference values for the same solutions.

Table 3. Comparison for ^{65}Zn standardization ($k = 1$).

Measurement system	Activity (kBq/g)	Uncertainty (%)	Δ (%)
Reference (Ionization chamber)	639.51	0.80	-
Photon-photon coincidence	646.02	0.56	0.72

Table 4. Comparison for ^{133}Ba standardization ($k = 1$).

Measurement system	Activity (kBq/g)	Uncertainty (%)	Δ (%)
Reference (Ionization chamber)	501,84	0.25	-
Photon-photon coincidence	504.23	0.39	0.40

4. CONCLUSION

The deployment work of this new X-gamma coincidence system was well succeeded, as shown in table 4 by the comparisons of the ^{133}Ba samples, which produced difference 0.48 % between the results obtained by X-gamma and $4\pi\beta(\text{PC})-\gamma(\text{NaI})$ coincidence counting, showing that the new system has introduced an important improvement in the LNMRI routines for standardization of samples with radionuclides of complex decay scheme.

By the other side, with the difference of 0.72 % obtained with the ^{65}Zn samples was obtained another important improvement introducing the

possibility of standardize samples with X-rays in order of 8 keV.

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