STANDARDIZATION OF ^{68}(Ge+Ga) BY SUM-PEAK METHOD

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Abstract:
A ^{68}(Ge+Ga) commercial solution had been standardized in LNMRI in Brazil, by Sum-peak Method, in which a 3"x3" NaI(Tl) gamma-ray detector is positioned at the top of a well-type 5"x5" NaI(Tl) gamma-ray detector so that to set up near to a 4π counting geometry.
The ^{68}(Ge+Ga) standardization generally is done using liquid scintillation methods to avoid losses of ^{68}Ge by volatility, if dried sources are done. In this work this behavior was tested using three dried sources and three liquid sources in the sum-peak method measurements and the results showed a standard deviation of 0. 41%.
The activity results were compared with another primary method: 4πβ–γ lived-time anticoincidence counting. The two methods gave activity per unit mass values with differences from the reference value of +0.8 % (anti-coincidence method) and -3.8% (sum-peak-method).

Key words: sum-peak method, standardization, absolute activity measurements, ^{68}Ge; ^{68}Ga.

1. INTRODUCTION

^{68}Ge in equilibrium with ^{68}Ga (daughter) is a potential substitute of ^{18}F in the calibration of the radionuclide calibrators, because his half-life (270.95 ± 0.16) d [1], is longer than the ^{18}F half-life (1.8288±0.0003) h.

The ^{68}Ge disintegrates 100 % by electron capture to ^{68}Ga, producing X-rays and Auger electrons with energies smaller than 10 keV. ^{68}Ga is a positron emitter and his half-life is 67.7 min [1]. Despite the many gamma rays presents in the decay scheme of ^{68}Ga, there is just one that follows up the β^+ decay and it is low intensity, causing low interference in the measurements.

In the Figure 1 it is showed a simplified decay scheme of ^{68}Ge and its daughter ^{68}Ga [8], where it is possible to verify one 511 keV gamma ray originating from annihilation radiation and one 1077 keV gamma ray.

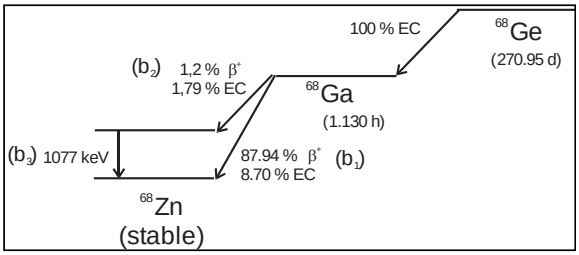

Figure 1. Decay scheme for ^{68}Ge.

2. METHODOLOGY

2.1. *Sum-peak Method*

This method was proposed by Brinkman [3]. The Equation 1 expressed the fundamental equation of the sum-peak method.

$$\frac{A_1.A_2}{A_{12}} + T = N \qquad (1)$$

where, N is the source activity, T is the number of total rate interactions on the spectrum, A_1 and A_2 are photopeak counts rates and A_{12} is the sum-peak counts rates, results from the simultaneous interaction of the two radiations on detector.

For this type of emitters (pure β^+ emitters or β^+-γ emitters), the method is possible only if the source is positioned inside the well, because the geometry must be near to 4π [4]. This restriction is due to the angular correlation between the annihilation rays to be extremely strong. Then, the Equation used in this type of decay is:

$$(\frac{A_1.A_2}{A_{12}} + T) = \frac{N}{W} \qquad (2)$$

where, *W* is a coincidence factor, which is equal 1 if the geometry is very close to 4π.

The photon spectrum of the ^{68}(Ge+Ga) source placed inside the well presents two photopeaks that are used in the sum-peak method: the 511 keV (A_1 e A_2) and the sum-peak originated from the sum of two 511 keV photons. Then, the fundamental sum-peak method equation is:

$$N = T + R \qquad (3)$$

with

$$R = \frac{A_{511keV} \cdot A_{511keV}}{A_{1022keV}} \quad (4)$$

where

A_{511keV} count rate on the photopeak annihilation radiation (511keV)

$A_{1022keV}$ count rate on the two annihilation rays in coincidence (sum-peak = 511+511 keV)

2.2. 4πβ–γ Lived-time anticoincidence counting

The Anti-coincidence measurements may be expressed as:

$$N_0 = \frac{C_\beta C_\gamma}{C_\gamma - C_\gamma^C} \quad (5)$$

where N_0 is the activity of the source and C_β, C_γ and $C^C{}_\gamma$ are β, γ and uncorrelated count rates, respectively. As in the coincidence measurements, an extrapolation curve is obtained and N_0 is determined. This method was introduced in LNMRI in 2006 [5 and 6].

2.3. Uncertainties

For the sum-peak method, the statistics components were evaluated using the Equation 3. In this equation there are not nuclear parameters involved, then the principal components that affects the A-type is the calculation of photopeaks net areas, the extrapolation to zero energy (0 keV) and back ground counts. From the derivation of Equation 3, it is possible to evaluate the statistics uncertainties, according to the Equation 6.

$$(\Delta N_0)^2 = \left(\frac{\partial N_0}{\partial N_T}\right)^2 (\Delta N_T)^2 + \left(\frac{\partial N_0}{\partial N_R}\right)^2 (\Delta R)^2 \quad (6)$$

Besides the A-type uncertainties derived from N_0, others uncertainties components also affects the accuracy of the activity of ^{68}(Ge+Ga) solution (sample weight, coefficient dilution and decay correction) considered in this work. According to the masses used to prepare ^{68}(Ge+Ga) sources for sum-peak method, the sample weighting component was 0.10 %, and for the anti-coincidence method it was 0.05 %. The dilution coefficient component may be neglected.

The decay correction component (half-life) is evaluated by the Equation 7.

$$S_{T1/2} = \frac{\ln 2 . \Delta t . \mu T_{1/2}}{T_{1/2}} \quad (7)$$

where,

Δt difference between counting date and reference date.
$\mu T_{1/2}$ uncertainty in the half-life determination [1]
$T_{1/2}$ half-life [1]

3. EXPERIMENTAL PROCEDURES

3.1. *Sum-peak Method*

The ^{68}(Ge+Ga) original solution was in the form of a solution in 0.1 mol/l HCl, carrier free.

Three sources were prepared from a ^{68}Ge/^{68}Ga diluted solution (dilution factor of 20.622298) by dropping deposition of known masses onto a cavity in the center of an acrylic disk fixed in a 0.05 mm thick polystyrene film, as show in the Figure 2. The masses were determined in a micro analytical balance using the picnometer differential weighing technique. Immediately after drying the sources were covered by the same polystyrene film. In the same way more three sources were prepared, but they were not dried. After weighing, the sources were covered by the same film. The six sources were measured with the proposal to investigate if there are losses of ^{68}Ge by volatility (known behavior of ^{68}Ge described in [7]).

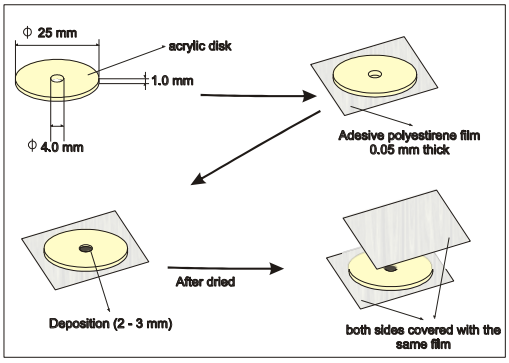

Figure 2. Preparation of the ^{68}Ge/^{68}Ga sources.

The sources were placed inside the well type 5”x5” NaI(Tl) gamma ray detector (bottom). In the top of this well type detector there was a 3”x3” NaI(Tl) gamma-ray detector resulting in a set up near to a 4 π counting geometry. Figure 3 illustrates the experimental arrangement used for the sum-peak method.

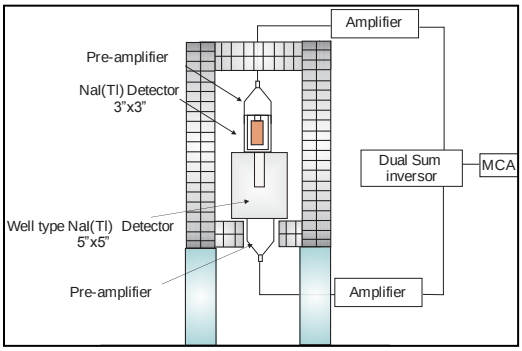

Figure 3. Experimental arrangement used in the sum-peak measurements.

Measurements were done using the coincidence 511 keV, resulting in a 1022 keV (sum-peak).The photon spectrum is shown in the Figure 4. The spectrum exhibits two photopeaks which are used for the activity determination.

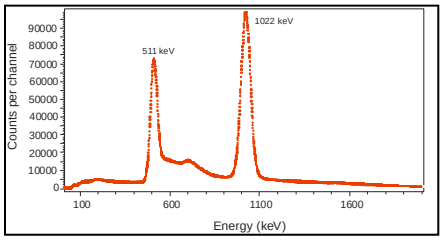

Figure 4. Spectrum obtained in the ^{68}(Ge+Ga) sum-peak measurements.

To prove that the coincidence factor (*W*) is 1 in this set-up, it was used a ^{22}Na reference solution, due to the fact that this nuclide emits both annihilation radiation and one γ-rays (1274 keV), so, it is possible to determine the value of *W*. For these measurements, five sources were prepared in the same way that the ^{68}Ge. The value was 1.0058.

3.2. Lived-time anticoincidence counting 4πβ–γ

To the anticoincidence measurements nine sources were prepared from the same diluted solution used in the sum-peak method. A set of vials were prepared by adding know masses of the diluted solution to 15 ml commercial scintillator cocktail. It was used three commercial scintillators: Ultima Gold, HiSafe III and Instagel Plus (three vials of each cocktail).

The measurements in the anti-coincidence system has been made using liquid-scintillation counting in the beta channel and a NaI(Tl) scintillation detector in the gamma channel. In both beta and gamma channels were used the MTR2 modules [2 and 6]. The MTR2 modules allow operating with dead time values from 25 to 200 μs.

The β channel discrimination level was adjusted to 20 keV to avoid counts Auger electrons originated from electron capture events.

Figure 5 presents one extrapolation curve obtained for each cocktail.

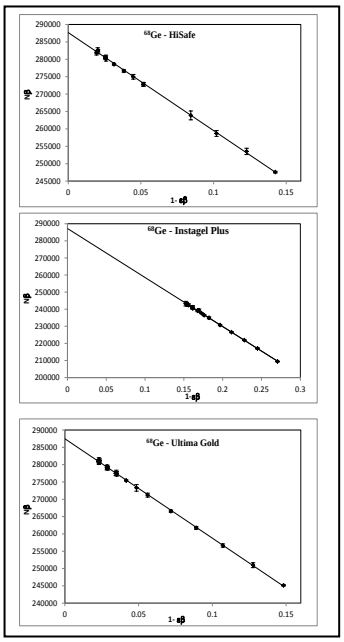

Figure 5. Extrapolation curve for each cocktail used in Anti-coincidence method.

4. RESULTS

The six sources (drieds and liquids) prepared for sum-peak method showed activity values with standard deviation of 0.41 %.

The results of each method and the reference value are showed in the Table 1(reference Date: 01/01/2011-12 h).

Table 1. Results for ^{68}Ge standardization.

Measurement system	Activity/mass (kBq/g)	U (%) k=1
lived- time anticoincidence counting 4πβ–γ	6.624	0.18
Reference value (certificate)	6.572	1.4
Sum-peak method	6.330	0.14

Table 2 shows the main uncertainties components considered in the activity determination by the sum peak method.

Table 2. Main uncertainties components in ^{68}(Ge+Ga) activity determination by sum-peak method.

Uncertainty Component	Type	U (%) k=1
Mass determination	B	0.1
Live time	B	0.01
Decay corrections	B	0.0125
Statistics counts*	A	0.0988
Combined uncertainty (k=1)		0.14

***including background and extrapolation to zero keV uncertainties.**

Table 3 presents the uncertainties components considered in the activity determination by the Anti-coincidence method.

Table 3. Uncertainties components in ^{68}(Ge+Ga) activity determination by anti-coincidence method.

Uncertainty Component	Type	U (%) k=1
Statistics counts	A	0.022738
Fitting procedures	A	0.052134
Mass determination	B	0.05
Live time	B	0.01
Background	B	0.037022
Decay corrections	B	0.003021
Decay branch	B	0.13
Correction due to detection of 1077 keV photons	B	0.1
Combined uncertainty (k=1)		0.18

5. CONCLUSION

The small value for the standard deviation (0.41%) for activity values in the sources dried and liquids shows that if the drying is fast and the source is immediately covered, there aren't significant losses of ^{68}Ge.

The cocktails HiSafe III and Ultima Gold were more suitable for preparing samples for liquid scintillation counting resulting in higher detection efficiencies than those prepared with Instagel plus.

The value of W proves that β^+ and β^+-γ emitters can be measure in this experimental arrangement.

The differences between results obtained by sum-peak method and $4\pi\beta-\gamma$ lived-time anticoincidence counting shows that an effort should be done in the two methods to study others corrections that are necessary in these methods.

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