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Sum-peak method with two NaI(Tl) crystals: ^{68}(Ge + Ga) standardization

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ABSTRACT

A ^{68}(Ge + Ga) commercial solution has 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, resulting in a set up approximately 4π geometry. In this work the known germanium volatility was tested using three dried sources and three liquid sources in the sum-peak method measurements and the activity results showed a standard deviation of 0.41%. The activities were compared with another primary method: 4πβ–γ live-timed anti-coincidence counting. The two methods gave activity concentration values with differences from the certified value of +0.8 % (anticoincidence method) and -3.4% (sum-peak method).

Keywords: ^{68}Ge, ^{68}Ga, sum-peak method, activity standardization, anticoincidence method.

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1. INTRODUCTION

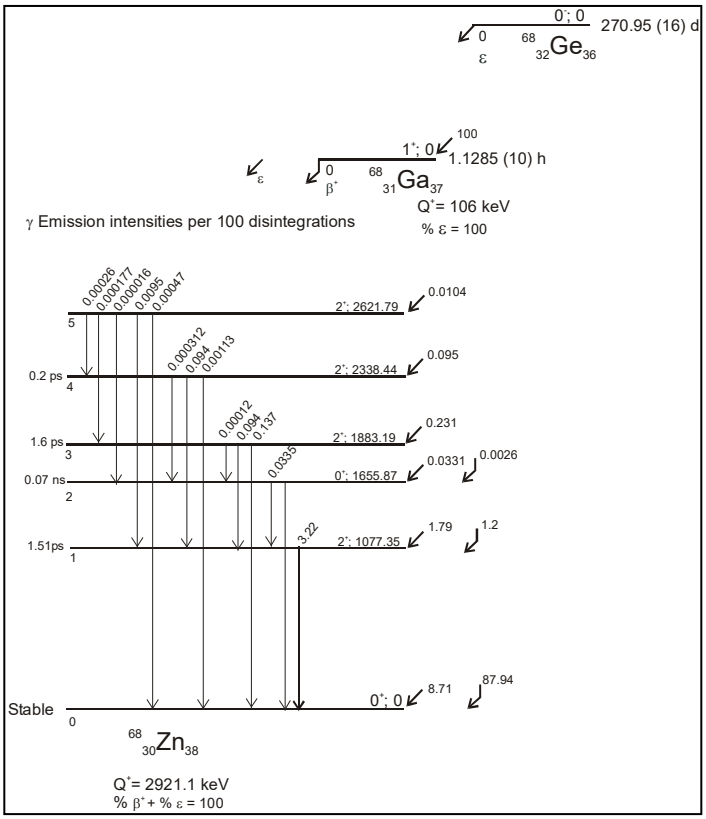
^{68}Ge in equilibrium with its daughter, ^{68}Ga, is a potential surrogate of ^{18}F not only in the checking of the radionuclide calibrators, but also another practices in nuclear medicine [1] because his half-life (270.95 ± 0.16) d [2] is much longer than the ^{18}F half-life (1.8288 ± 0.0003) h [3].

The ^{68}(Ge + Ga) standardization generally is done using liquid scintillation methods [1] to avoid losses of ^{68}Ge by volatility, if dry sources are done [4]. The purpose of this work was to standardize a ^{68}(Ge + Ga) solution by sum-peak method, which uses solid sources. Because of the setup system that requires dry sources, a study for the loss of ^{68}(Ge + Ga) by volatility was done with dry and liquid sources.

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 (1.1285 ± 0.0010) h [2]. Despite the presence many gamma rays in the decay scheme of ^{68}Ga, there is just one that follows up the β^+ decay and it has a low intensity, causing a little interference in measurements.

In the Figure 1 it is showed a decay scheme of ^{68}Ge and ^{68}Ga [2], where it is possible to verify the 511 keV gamma ray originating from annihilation radiation and the 1077 keV gamma ray.

Figure 1. *Decay scheme for ^{68}(Ge + Ga).*



Source: <http://www.nucleide.org/DDEP_WG/Nuclides/Ga-68_tables.pdf>

2. METHOD

2.1. Sum-peak Method

The sum-peak method was proposed by Brinkman [5, 6] for a well type NaI (Tl) scintillation detector. This method is very simple, using just a single detector, that may be a NaI (Tl) scintillation crystal or an high resolution HP(Ge) detector. To be measured by sum-peak method, the nuclide should be present two or more electromagnetic radiation simultaneously. The radiations may be two gamma rays in coincidence, as ^{60}Co and β+ emitters, or one X-ray followed by one gamma ray, as ^{65}Zn and ^{51}Cr [7].

Equation (1) expresses the fundamental relation of the sum-peak method.

$$\frac{A_1.A_2}{A_{12}} + N_T = N_0 \tag{1}$$

where, N_0 is the source activity, N_T is the number of total rate interactions on the spectrum, $A_1$and A_2 are photopeak counting rates and A_{12} is the sum-peak counting rate, which 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 approximately 4π [5]. This restriction is due to the angular correlation between the annihilation rays to be extremely strong. Because of this, Brinkman [5] proposed the following equation (2) for the source positioned inside de well when the counting geometry is less than 4π:

$$(\frac{A_1.A_2}{A_{12}} + N_T) = \frac{N_0}{W} \tag{2}$$

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

The W value can be experimentally determined by measuring a ^{22}Na source inside the well of a NaI(Tl) detector in the geometry equal or less than 4π. In this case three sum-peaks can be observed in the spectrum with the following equations:

$$(\frac{A_{511}.A_{1274}}{A_{1785}} + N_T) = N_{01} \tag{3}$$

$$(\frac{A_{1022}.A_{1274}}{A_{2276}} + N_T) = N_{02} \tag{4}$$

$$(\frac{A_{511}^2}{A_{1022}} + N_T).W = N_{03} \tag{5}$$

As the activity must be the same in the three equations, the W value may be obtained combining equations (3), (4) and (5), as follow:

$$N_{01} = N_{02} = N_{03} \tag{6}$$

$$W = N_{01}/(\frac{A_{511}^2}{A_{1022}} + N_T) = N_{02}/(\frac{A_{511}^2}{A_{1022}} + N_T) \tag{7}$$

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

$$N_0 = N_T + R \tag{8}$$

with

$$R = \frac{A_{511keV} \cdot A_{511keV}}{A_{1022keV}} \tag{9}$$

where: A_{511keV} is the count rate on the photopeak annihilation radiation (511 keV) and $A_{1022keV}$ is the count rate on the two annihilation rays in coincidence (sum-peak = 511+511 keV).

2.2. 4πβ–γ Live-timed anticoincidence counting

There is no basic difference between coincidence and anti-coincidence counting. The anti-coincidence method is a complementary method of the coincidence method that initially was considered by Bryant [8] in the particular case of nuclides that present meta-stable levels in the decay scheme. Later on, Baerg [9] introduced the use of live time to the anti-coincidence system that eliminates the correction of dead time using an extending dead time device. In the present version LNMRI uses two modules of MTR2 in its anti-coincidence system [10, 11]. These modules have been introduced in the LNMRI laboratory in December of 2005. In this work it was used the minimum dead time of 50 μs in the beta and gamma channels.

The activity of a radionuclide can be determined using equation (10), which is the classic equation of the coincidence method differing only in that N_c, the coincidence count rate that is determined for one given gamma window as the difference between the gamma rate and uncorrelated gamma rate, represented in (10).

$$A = \frac{N_\beta \, N_\gamma^w}{N_\gamma^w - {}^{i}N_\gamma^w} \tag{10}$$

Where, N_β is the count rate in the beta channel; N_γ^w is the count rate in the gamma window and ${}^iN_\gamma^w$ is the uncorrelated gamma count rate.

2.2. Uncertainties

The statistical components were evaluated using the equation (8). In this equation there are not nuclear parameters involved. Then, the main components that affect the A-type are: calculation of photopeak net areas; extrapolation to 0 keV energy; and background counts. From the derivation of Equation (8), it is possible to evaluate the statistical uncertainties, according to:

$$(\Delta N_0)^2 = (\Delta N_T)^2 + (\Delta R)^2 \tag{11}$$

where, N_T is the sum of counts in the spectrum (E=0 to 1350 keV)

The N_T value is withdrawn of the spectrum and it is obtained from the gross counts (N_g=$SUM_{(sample\ \text{spectrum})}$) stripped the background counts.

The equation (8) must be analyzed in parts for clear understanding.

The background spectrum live time ($T_{live(BG)}$) and the sample spectrum live time ($T_{live(sample\ spectrum)}$) are different. The count rate of the background spectrum $T_{(BG)}$ is expressed by:

$$T_{(BG)} = \frac{SUM_{(BG)}}{T_{live(BG)}} \tag{12}$$

if:
$$N_T = N_g - BG \tag{13}$$

and,
$$N_g = SUM_{(sample\ spectrum)} \tag{14}$$

then,
$$N_T = N_g - T_{(BG)}.T_{live\,(sample\ spectrum)} \tag{15}$$

Taking into account the extrapolation to 0 keV energy:

$$N_T = N_g - T_{(BG)}.T_{live\,(sample\ spectrum)} + (extr.p/E = 0keV) \tag{16}$$

Following the rules for uncertainties:

$$(\Delta N_T)^2 = (\Delta N_g)^2 + [\Delta(T_{(BG)}.T_{live(sample\ spectrum)})]^2 + (\Delta ext \to 0)^2 \tag{17}$$

N_γ, $T_{(BG)}.T_{live\ (sample\ spectrum)}$, and $(ext \to 0)$ are counts and from the Poisson distribution the relations below can be used:

$$(\Delta N_g)^2 = N_g \tag{18}$$

$$[\Delta(T_{(BG)}).T_{live(sample\ spectrum)}]^2 = T_{(BG)}.T_{live(sample\ spectrum)} \tag{19}$$

$$(\Delta ext \to 0)^2 = ext \to 0 \tag{20}$$

Then,

$$\boxed{(\Delta N_T)^2 = N_g + T_{(BG)}.T_{live(sample\ spectrum)} + (ext \to 0)} \tag{21}$$

The uncertainty in R is:

$$R = \frac{N_1 N_2}{N_{12}} \tag{22}$$

$$\left(\frac{\Delta R}{R}\right)^2 = \left(\frac{\Delta N_1}{N_1}\right)^2 + \left(\frac{\Delta N_2}{N_2}\right)^2 + \left(\frac{\Delta N_{12}}{N_{12}}\right)^2 \tag{23}$$

R is obtained when N_0 is determined, having $\Delta R/R$ and R:

$$(\Delta R)^2 = \left(\frac{\Delta R}{R}\right)^2 .R^2 \tag{24}$$

The total square uncertainty is the sum of the equations (14) and (17).

$$N_0 = N_T + \frac{N_1 N_2}{N_{12}} = N_T + R \tag{25}$$

$$(\Delta N_0)^2 = \left(\frac{\partial N_0}{\partial N_T}\right)^2 (\Delta N_T)^2 + \left(\frac{\partial N_0}{\partial N_R}\right)^2 (\Delta R)^2 \tag{26}$$

$$\boxed{\Delta N_0{}^2 = 1^2 (\Delta N_T)^2 + 1^2 (\Delta R)^2} \tag{27}$$

Besides the A-type uncertainties derived from N_0, others uncertainties components also affect the accuracy of the activity of ^{68}(Ge + Ga) solution (sample weight, dilution factor 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 uncertainty dilution factor component may be neglected.

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

$$S_{T1/2} = \frac{\ln 2.\Delta t.\mu_{T_{1/2}}}{T_{1/2}} \tag{28}$$

where: Δt is the time interval between counting date and reference date; $\mu_{T1/2}$ is the uncertainty in the half-life value [2]; and $T_{1/2}$ is the half-life [2].

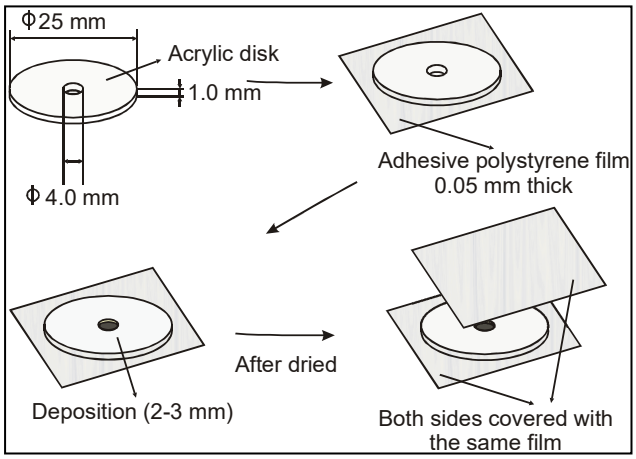
3. EXPERIMENTAL PROCEDURE

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 Mettler Toledo MT5 micro analytical balance (readability: 1 μg; weighing capacity: 5100 mg) using the pycnometer differential weighing technique. The sources were dried in an infrared lamp and 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 purpose to investigate if there are losses of ^{68}Ge by volatility (known behavior of ^{68}Ge described in [8]). In order to achieve the decay chain equilibrium these prepared sources were measured in a time period over 24 hours after drying.

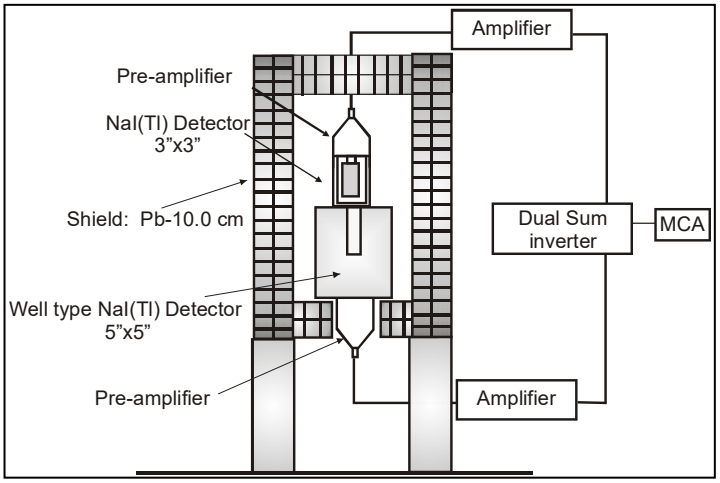
Figure 2. *Scheme for ^{68}(Ge + Ga) source preparation in sum-peak method measurements*.



Source: Oliveira *et al*, 2012 [12].

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 approximately 4π counting geometry. Figure 3 illustrates the experimental arrangement used for the sum-peak method.

Figure 3. *Experimental arrangement used in sum-peak measurements including shield, detectors and electronic modules.*



Source: Oliveira *et al*, 2012 [12].

Measurements were done using the coincidence 511 keV, resulting in a 1022 keV (sum-peak). The measuring time of spectra varied from 2900 to 6000 seconds, which was enough to achieve a statistical uncertainty around 0.03 - 0.04 %. The peak area evaluation and its uncertainties associated were carried out using data acquisition software (Maestro) [13], incorporated within a commercial multichannel analyzer. These peak area evaluation in each analyzed spectrum also included corrections for dead-time and background subtraction. In order to minimize the undesirable but present instrumental pile-up effect that distorts the γ-ray peaks the sources were prepared with low count rate intensity.

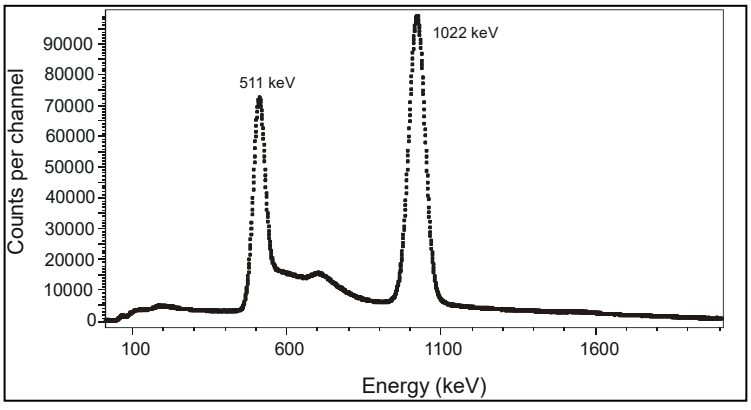
The correction factor for decay during counting related to the initial count time is done obeying the equation (29) below:

$$F_D = \frac{\lambda t_c}{1 - e^{-\lambda t_C}} \tag{29}$$

where, $\lambda$is the decay constant *(λ= ln(2)/$T_{1/2}$) and* t_c is the period of time.

The photon spectrum is shown in the Figure 4. The spectrum exhibits two photopeaks which were used for the activity determination, from the equation (1).

Figure 4. *^{68}(Ge + Ga) gamma energy spectrum measured with a 5"x5" NaI (Tl) well type detector covered by a 3"x3" NaI (Tl) detector resulting in 4π geometry for sum-peak measurements.*



Due to the low resolution of NaI (Tl) detector, counts from the 1077 keV gamma ray from the decay of ^{68}Ga not appear in the spectrum but interferes in the counts of 1022 keV sum peak and must be accounted for as follow:

$$N(sum) = N_0 \varepsilon_1^2 P_{\gamma 1}^2 + N_0 \varepsilon_2 P_{\gamma 2} \tag{30}$$

Where, *N(sum)* includes the contribution from 1077 keV represented by $N_0 \varepsilon_2 P_{\gamma 2}$ [5].

This interference is small, but the following procedure to correct was adopted. First, it was established that the lines for 1077 and 1115 keV from ^{65}Zn decay have the same photopeak efficiency. A ^{65}Zn reference source was used (in the same geometry measurements conditions that ^{68}Ge sources) to obtain the value of photopeak efficiency. After knowing the interference counts, it was obtained the counts values for 1022 keV alone, which corresponds an increase of 0.26 % in the final value of activity concentration.

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 was possible to determine the value of *W* from equation (7). For these measurements, five sources from a ^{22}Na solution were prepared in the same way that ^{68}Ge. The *W* value found is 1.0058 ± 0.0005.

3.2. Live-timed anti-coincidence counting 4πβ–γ

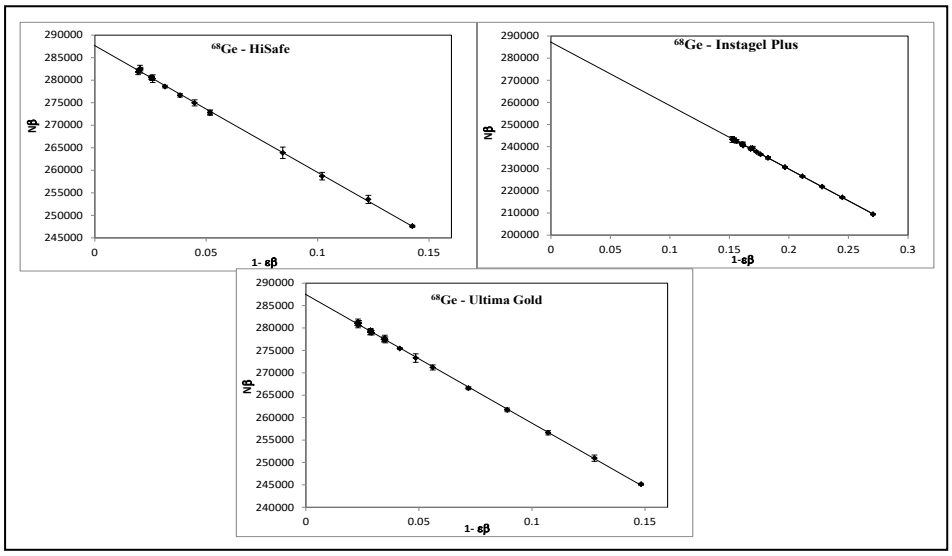
To the anticoincidence measurements nine sources were prepared from the same diluted solution used in the sum-peak method. A set of vials were prepared by adding know masses of the diluted solution to 15 ml commercial scintillator cocktail. It was used three commercial scintillators: Ultima Gold, HI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

The measurements in the anticoincidence system have 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 [14, 10]. The MTR2 modules allow operating with dead time values from 25 to 200 μs. In the measurements of ^{68}Ge it was used a minimum dead time of 50 μs. The LS positron efficiency, ε_β was varied between 0.97 and 0.85 using the electronic

discrimination and extrapolated for 1.0. In order to avoid the contribution of electron capture events from ^{68}Ga and ^{68}Ge, we cut the extrapolation curve in the region of low energy (adjusted to 20 keV), in our experimental condition its mean 1.5 V. The NaI (Tl) was gated on 511 keV region using a single channel analyzer in this condition the mainly γ-ray counts events were due positon-annihilation decay. But a contribution from Compton scattering of 1077 keVγ-rays correspond to electron capture events and not due positron emission therefore a correction to the intercept was necessary. In order to determine this correction, it was used a ^{60}Co source for this factor in a similar proceeding proposed by Zimmerman [1].

Figure 5 presents one extrapolation curve obtained for each cocktail.

Figure 5. *Extrapolation curve counting for each cocktail (HiSafe, Ultima Gold and Instagel Plus) used in anticoincidence method.*



4. RESULTS AND DISCUSSION

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

The results of each method and the reference value are showed in the Table 1(Reference time: January 1st, 2011-12 h – Official time of Brasília).

Table 1. Result of the standardization of ^{68}(Ge + Ga) solution with the sum-peak method and comparison with the value obtained by anti-coincidence method. (Reference time: January, 1st, 2011-12 h – official time of Brasília).

MEASUREMENT SYSTEM	ACTIVITY/MASS (kBq/g)	U (%) k=1
Live- timed anticoincidence counting 4πβ–γ	6.624	0.18
Certified value (traceable to NIST-Gamma spectrometry)	6.572	1.4
SUM-PEAK METHOD	6.347	0.14

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

Table 2.Uncertainty components evaluated in the determination of the activity concentration of ^{68}(Ge + Ga) solution using sum-peak method.

Uncertainty Component	Type	u (%)
Mass determination	B	0.10
Live time	B	0.01
Decay corrections	B	0.01
Statistic counts*	A	0.10
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.Uncertainty components evaluated in the ^{68}(Ge + Ga) activity determination by anti-coincidence method.

Uncertainty Component	Type	u (%)
Statistics counts	A	0.02
Fitting procedures	A	0.05
Mass weighing	B	0.05
Live time	B	0.01
Background	B	0.04
Decay corrections	B	< 0.01
Decay branch	B	0.13
Correction due to detection of 1077 keV photons	B	0.10
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 due to the volatility. 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 measured in this experimental arrangement.

The difference between results obtained by sum-peak method and $4\pi\beta-\gamma$ live-timed anticoincidence counting shows that an improvement should be done in the sum-peak method arrangement. Due to the better resolution of the Ge detector compared to the NaI (Tl) detector, an option to improve the measurements of the sum peak method will be to exchange the 5"x5" NaI (Tl) well type detector and the 3"x3" NaI (Tl) detector by well type Ge detector and a planar Ge detector. Thus, the peak 1077 keV of the ^{68}Ge can be separated from the peak of 1022 keV (sum), eliminating the interference that is probably causing the difference in the results between the sum peak method and the anticoincidence method.

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