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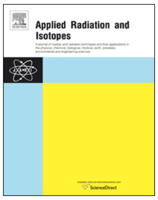

Determination of gamma-ray emission probabilities per decay of Ga-68

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HIGHLIGHTS

- The procedure for $4\pi(PC)\beta-\gamma$ primary standardization of ^{68}Ga is described.
- Multiparametric fitting was performed for the extrapolation curve.
- Gamma-ray emission probabilities per decay were determined for seven transitions.

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ABSTRACT

The measurement of the gamma-ray emission probabilities per decay of 1077 keV of ^{68}Ga is presented. The standardization system consists of a gas-flow proportional counter in 4π geometry coupled to an HPGe detector for the gamma-ray detection. The gamma-ray emission probabilities per decay were measured in an HPGe gamma-ray spectrometer. The weaker gamma-ray intensities of ^{68}Ga were measured in a relative way, making use of an uncalibrated ampoule of ^{68}Ge–^{68}Ga in radioactive equilibrium, and considering the absolute result from the 1077 keV gamma-ray.

1. Introduction

The procedure followed by the Nuclear Metrology Laboratory (LMN) at the IPEN-CNEN/SP, in São Paulo, for the determination of the gamma-ray emission probabilities per decay of ^{68}Ga, a positron emitter used in Positron Emission Tomography (PET), is presented. ^{68}Ga decays with a half-life of 67.8 min by mixed β^+ and electron capture branches mainly to the ground state and to excited states of ^{68}Zn, consequently several gamma-rays are emitted, as shown in Fig. 1.

The activity standardization was carried out applying a Software Coincidence System (SCS) developed at the LMN (Toledo et al., 2007), making easier the standardization of radionuclides which decay with short half-life such as ^{99m}Tc (Brito et al., 2012).

Two gamma-ray windows were set for the coincidence measurements, one including the positron-annihilation quanta and the other located at the total absorption energy peak of the 1077 keV gamma-ray. A multiparametric linear square fit was performed in order to determine the activity and the 4π detector efficiencies were changed by pulse height discrimination.

The ^{68}Ga gamma emission probabilities per decay were measured in a previously calibrated HPGe gamma-ray spectrometer, in the energy range between 244 keV and 2754 keV; the procedure is detailed in Section 2.

In order to determine the 1077 keV gamma emission probability per decay, a ^{68}Ga ampoule calibrated in the $4\pi\beta-\gamma$ system was used. The emission probabilities per decay for the weaker gamma-rays were measured by means of an uncalibrated ampoule of ^{68}Ge–^{68}Ga in radioactive equilibrium. The advantage, in the use of a ^{68}Ge generator, is its longer half-life (270 days) and its decay by 100% pure electron capture process with no gamma-ray emission. Therefore, it does not interfere in the ^{68}Ga calibration and allows longer counting times, achieving better statistics.

2. Experimental method

2.1. Source preparation

The ^{68}GaCl solution was obtained from a ^{68}Ge–^{68}Ga generator, supplied by the IPEN Radiopharmaceutical Center, followed by elution with 0.5 M HCl solution. After this procedure, the radioactive sources were prepared by dropping known aliquots on Collodion substrate, previously coated with 10 μg cm^{-2} gold layer on both sides to make the film conductive. The source masses were determined by the pycnometer technique.

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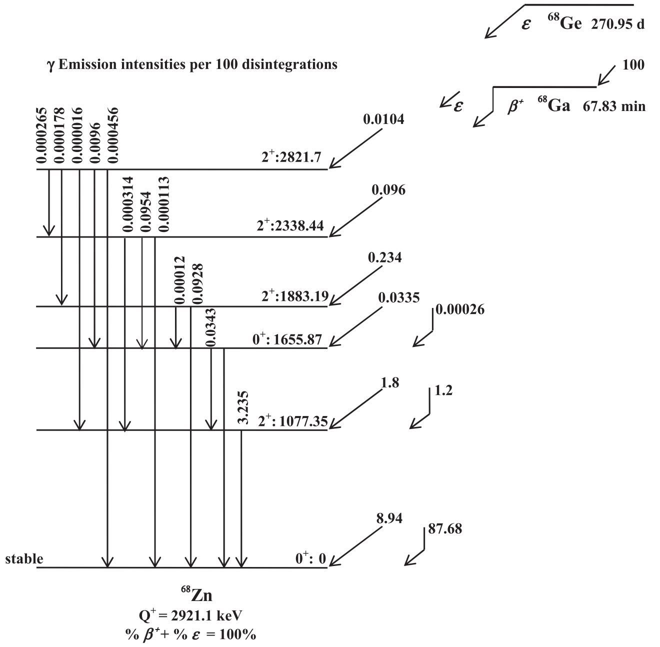

Fig. 1. Decay scheme of ^{68}Ge–^{68}Ga (Bé and Schönfeld, 2013). All energies are in keV.

A seeding agent (Cyastat SM) was used to improve the deposit uniformity and the sources were dried under a nitrogen jet at 45 °C (Wyllie et al., 1970). A total of three sources were prepared with masses ranging from 7 to 30 mg.

Two flame-sealed ampoules were prepared for the gamma-ray emission probability per decay measurement. The first one was a standard ^{68}Ga ampoule from the same solution used for preparing the sources measured in the $4\pi\beta-\gamma$ system, and the second was prepared from a ^{68}Ge–^{68}Ga uncalibrated generator solution, to be used for determining the weaker gamma-ray intensities. These solutions did not show any impurity that could affect the activity results.

2.2. $4\pi\beta-\gamma$ coincidence measurements

The specific activity of ^{68}Ga was measured by a triple $4\pi\beta-\gamma$ coincidence system consisting of a thin window gas-flow proportional counter (PC) in 4π geometry coupled to a 50.1 mm × 25.4 mm NaI(Tl) scintillator and to a 20% relative efficiency HPGe crystal, for gamma-ray detection. Two gamma windows were set for each gamma detector, one at the 511.0 keV positron-annihilation quanta and the other at 1077 keV gamma-ray full energy peaks. Data acquisition was carried out by means of a Software Coincidence system (SCS).

The SCS was based on a National Instruments PCI-6132 card capable of up to four independent analog inputs, and the signals were processed by means of a LabView version 8.5 acquisition program. Information on pulse height and time of occurrence were registered for both beta and gamma channels.

The activity calculation was performed by means of code SCTAC version 6.0 (Dias, 2010), developed at the LMN, which allows gamma-ray window selection after the measurement has been completed.

The final formula applied to the coincidence measurement is

$$\frac{N_{4\pi}}{\varepsilon_1\varepsilon_2}=N_0\left[1+A\frac{(1-\varepsilon_2)}{\varepsilon_2}+B\frac{(1-\varepsilon_1)}{\varepsilon_1}+C\frac{(1-\varepsilon_1)}{\varepsilon_1}\frac{(1-\varepsilon_2)}{\varepsilon_2}\right] \tag{1}$$

where

$N_{4\pi}$ is the total counting rate of the PC

$$\varepsilon_1=\left(\frac{N_c}{N_\gamma}\right)_{511}=\frac{\left[a_1\varepsilon_{\beta_1}+a_2\varepsilon_{\beta_2}\right]}{(a_1+a_2)} \tag{2}$$

and

$$\varepsilon_2=\left(\frac{N_c}{N_\gamma}\right)_{1077}=\frac{\left[a_1\varepsilon_{\beta_1}+b_1\varepsilon_{ec1}\right]}{(a_1+b_1)} \tag{3}$$

Indexes 511 and 1077 refer to 511 keV annihilation quanta and 1077 keV transitions, respectively; N_c is the coincidence rate between each selected gamma-ray and the PC; $\varepsilon_{\beta i}$ and ε_{eci} are the PC efficiencies for positrons and electron capture events of the i-th branch, respectively. The approximations: $\alpha_{1077}\cong 0$, and, $\varepsilon_{ec1}=\varepsilon_{ec2}=\varepsilon_{ec}$ and $\varepsilon_{\beta_1}\cong\varepsilon_{\beta_2}\equiv\varepsilon_\beta$ were considered; a_i and b_i are the branching ratios for beta plus and electron capture, respectively.

A–C are parameters determined by the least square fitting. In this fitting parameter C was considered as independent. According to Chauvenet (2013) this parameter is linearly dependent on A and B, that is $C=A+B-1$. The final fitting parameters show that this equality is true, within the uncertainties, indicating consistency between these two procedures. The simultaneous extrapolations of $\frac{(1-\varepsilon_1)}{\varepsilon_1}$, $\frac{(1-\varepsilon_2)}{\varepsilon_2}$, and consequently $\frac{(1-\varepsilon_1)(1-\varepsilon_2)}{\varepsilon_1\quad\varepsilon_2}$ to zero yield the activity N_0.

Earlier measurements of ^{68}Ga by multiparametric extrapolation were compared with Monte Carlo simulation by means of code ESQUEMA version 9.0 and showed good agreement, demonstrating the reliability of this method of extrapolation. (Koskinas et al., 2012).

The PC efficiency was changed by moving the PC discrimination lower level by software used as input to SCTAC code. Corrections for background, dead time and decay were applied, and corrections for accidental coincidences were performed by using the Cox-Isham formalism adapted by Smith (1978).

2.3. Gamma-ray spectrometry measurements

The gamma-ray spectrometer system consists of an HPGe detector with 500 µm thick Be window, yielding 1.79 keV FWHM at 1332.5 keV. The gamma-ray full energy peak efficiency curve was measured at 17.9 cm of source–detector distance, in the energy range from 244 keV to 2754 keV, by measuring flamed-sealed, 5 mL Schott ampoules with diameter 13.3 mm, of ^{24}Na, ^{60}Co, ^{133}Ba, ^{137}Cs, and ^{152}Eu standardized at the LMN.

These ampoules are traceable to standard sources supplied by the International Atomic Energy Agency (IAEA). The variation in the height was estimated and the geometry and attenuation factors were calculated. The variations in these factors were included in the overall uncertainty.

The area under the peak was evaluated by code Alpino, developed at the LMN (Dias, 2001), which applies the method of numeric peak integration. Dead time and pile-up corrections were applied by measuring a reference pulser peak near the upper edge of the gamma spectrum simultaneously with the sources. The HPGe calibration curve was obtained by a third degree polynomial in log–log scale, applying linear least square fitting applying covariance matrix with all partial uncertainties involved.

The 1077 keV gamma-ray emission probability per decay was determined by measuring the ^{68}Ga ampoule and applying the following relationship:

$$p(E_{1077})=\frac{S(E_{1077})}{\varepsilon(E_{1077})Am} \tag{4}$$

where $S(E_{1077})$ is the counting rate in the 1077 keV peak, $\varepsilon(E_{1077})$ is the peak efficiency for 1077 keV, A is the specific activity determined in the $4\pi\beta-\gamma$ system and m is the ampoule mass.

The emission probabilities for the weaker gamma rays, namely, 578, 805, 1261, 1744, 1882 and 2338 keV were determined by measuring an uncalibrated ^{68}Ge–^{68}Ga ampoule in a relative way, considering the absolute result for the 1077 keV emission probability per decay, obtained at the HPGe spectrometer from the ^{68}Ga calibration. The composition of both ampoules was considered the same once the Ga-68 ampoule was diluted in 0.5 M HCl and the ^{68}Ge–^{68}Ga was diluted with a solution composed by GeO_2 31 μg/mL in 0.5 M HCl.

3. Results and discussion

The 4π(PC) efficiency ranged from 38% to 98% for β^+ (gamma-ray window at 511 keV) and from 6% to 60% for mixed β^+ and electron capture events (gamma-ray window at 1077 keV). The specific activities calculated by a multiparametric least square fitting, applying covariance methodology, for the ^{68}Ga sources are presented in Table 1, together with the average activity and the correlation matrix obtained in the calculation of the average activity by means of the least square method. Typical partial uncertainties for these activity measurements are presented in Table 2. The main uncertainties involved in the measurements were counting statistics, dead time, weight, decay, background, resolution time and efficiency curve extrapolation. The major contribution to the overall uncertainty comes from the multiparametric fitting.

The efficiency curve for the coaxial HPGe detector was fitted by the least square method using LOGFIT code (Dias and Moreira, 2005), the reduced χ^2 value was close to 1, indicating a satisfactory fit. Fig. 2 shows the residuals in percent between the fitted and experimental values.

In the relative measurement the counting time was 200,000 s, and the counting rate was 20 s^{-1} for the 1077 keV transition. For the other gamma transitions, the counting rate varied from 0.75 s^{-1} to 0.005 s^{-1}.

In the 1077 keV gamma-ray emission probability per decay determination, four measurements were performed in sequence, with 2000 s counting time each; the counting rate varied from 36 s^{-1} to 13 s^{-1}. The gamma-ray emission probability per decay was obtained by means of a weighted average.

Table 1
Activity and uncertainty obtained by multiparametric least squares fitting, with the corresponding correlation matrix.

Source	Activity (MBq g^{-1})	Correlation matrix		
1	38.38(25)	1		
2	38.35(35)	0.146	1	
3	37.95(30)	0.366	0.091	1
Average	38.25(20)			

Table 2
Typical partial relative uncertainties in the activity, in percent ($k=1$).

(a) Components	Uncertainty (%)
Counting statistics	0.10
Dead time	0.10
Weight	0.10
Background	0.30
Decay	0.35
Resolving time	0.10
Multiparametric fitting	0.41
Combined uncertainty	0.65

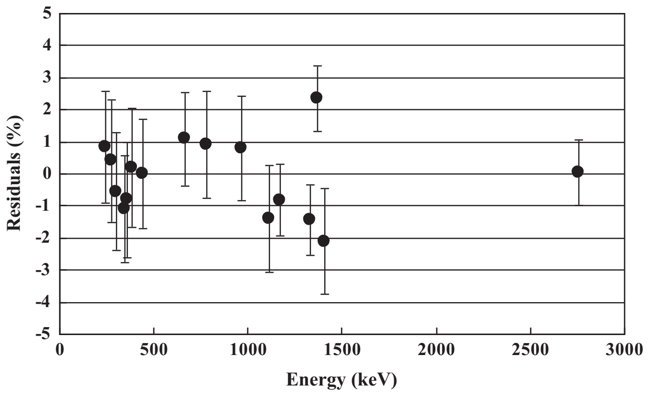

Fig. 2. Residuals between experimental and fitted HPGe efficiency values, in percentage.

Table 3
Results of relative gamma-ray emission probability for ^{68}Ga, in comparison with the literature.

Energy (keV)	This work	Luca et al. (2012)	Schönfeld et al. (1994)	Vo et al. (1994)	Lange et al. (1973)	Carter et al. (1968)	Vaughan et al. (1969)
578.52	1.10(4)	1.35(30)	1.14(15)	1.05(5)	1.00(12)	1.1(2)	0.7(1)
805.83	2.93(4)	2.68(34)	2.90(31)	2.81(14)	2.95(12)	2.8(2)	2.2(2)
1077.34	100	100	100	100	100	100	100
1261.08	2.98(3)	2.60(28)	3.06(31)	2.75(14)	3.00(7)	2.9(2)	3.1(2)
1744.42	0.297(10)			0.295(15)	0.30(4)	0.28(4)	0.5(1)
1883.16	4.29(4)	3.94(42)	3.86(59)	4.6(2)	4.33(12)	4.1(4)	4.8(3)
2338.44	0.049(4)			0.031(3)	0.050(6)	0.04(2)	< 0.1

The behavior of the electronic chain versus counting rate was verified by measuring the reference pulser at the end of each spectrum.

The comparison of the present results of relative emission probability with the literature is shown in Table 3. As can be seen, the present results are in agreement within the experimental uncertainties with the literature for 578, 805, and 1744 keV gamma-rays. For 1261 keV our value is in agreement with Carter et al. (1968), Lange et al. (1973) and Schönfeld et al. (1994), but it is not in agreement with Luca et al. (2012) and Vo et al. (1994); the value for 1883 keV is in agreement with Carter et al. (1968), Schönfeld et al. (1994), and Luca et al. (2012), but it is not in agreement with Lange et al. (1973) and Vo et al. (1994), and the value of 2338 keV does not agree with Vo et al. (1994), and no value is in agreement with Vaughan et al. (1969).

In Table 4 the emission probabilities per decay determined by the 1077 keV are presented. The present work is compared with Bé and Schönfeld (2013) evaluation and more recent values in the literature, namely, Schönfeld et al. (1994) and Luca et al. (2012). As can be seen, our value of 1077 keV is in agreement with these recent values within the estimated uncertainties.

Table 5 shows the uncertainties involved in the final values in order to obtain the combined uncertainty. The final uncertainty was the quadratic sum of the uncertainty considering each relative gamma intensity uncertainties plus the uncertainty of $I_\gamma(1077)$, dead time and in the geometry, which are common for all the weaker gamma rays.

Table 4
Results of the gamma-ray emission probability per decay of ^{68}Ga, in comparison with the more recent values from the literature (in percent).

Energy (keV)	This work	Bé and Schönfeld (2013)	Luca et al. (2012)	Schönfeld et al. (1994)
578.52	0.0354(14)	0.0343(23)	0.044(10)	0.037(5)
805.83	0.0942(17)	0.0928(27)	0.087(11)	0.094(10)
1077.34[a]	3.214(30)	3.235(30)	3.25(11)	3.22(3)
1261.08	0.0958(13)	0.0954(21)	0.084(9)	0.099(10)
1744.42	0.0095(3)	0.0096(5)		
1883.16	0.1379(19)	0.1420(35)	0.128(13)	0.125(19)
2338.44	0.00157(14)	0.00113(16)		

[a] Absolute measurement.

Table 5
Typical relative uncertainty components of the gamma-ray emission probability, in percent ($k=1$).

Component	Uncertainty (%)
Statistics net peak area of weaker gamma-rays	0.37–8.91
Statistics net peak area of 1077 keV	0.06
Dead time	0.29
Efficiency of 1077 keV measurement	0.49
Efficiency of weaker gamma-rays	0.51–0.78
Activity of 1077 keV measurement	0.52
Attenuation factor	0.16–0.32
Ampoule geometry	0.25
Intensity of 1077 keV	0.92
Combined uncertainties	1.35–9.1

4. Conclusions

The emission probability of 1077 keV gamma-ray of ^{68}Ga was measured by an absolute method and the result is in agreement with the literature. The weaker gamma-ray intensities of ^{68}Ga were measured in a relative way, making use of an uncalibrated ampoule of ^{68}Ge–^{68}Ga generator, and considering the absolute result from the 1077 keV gamma-ray. The relative values are in agreement with the literature; however when the absolute values are compared with the last evaluation, some of them are not in agreement.

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