

Disintegration rate and gamma-ray emission probability per decay measurement of Cu-64

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HIGHLIGHTS

- The procedure for $4\pi(\text{PC})\beta\text{-}\gamma$ primary standardization of ^{64}Cu is described.
- External absorbers of Collodion and aluminum were applied on both sides of sources.
- An electronic system with a Time to Amplitude Converter (TAC) was used.
- Determined 1345.7 keV gamma-ray emission probability per decay.

ARTICLE INFO

Keywords:

Activity
Coincidence counting
Copper-64
Standardization
Gamma-ray emission probability

ABSTRACT

The procedure followed by the Nuclear Metrology Laboratory (LMN) at the IPEN for the standardization of activity of ^{64}Cu is described. The measurement was carried out in a $4\pi(\text{PC})\beta\text{-}\gamma$ coincidence system. The activity per unit mass of the solution was determined by the extrapolation technique. The events were registered using a Time to Amplitude Converter (TAC) associated with a Multi-channel Analyzer. The gamma-ray emission probability per decay of the 1345.7 keV transition of ^{64}Cu determined with a calibrated REGe gamma-ray spectrometer was $(0.472 \pm 0.010)\%$, in agreement with the literature.

1. Introduction

The Nuclear Metrology Laboratory (LMN) at the IPEN in São Paulo (Brazil) has developed standardization methods for the activity of radionuclides, in particular for some used in nuclear medicine, such as $^{99\text{m}}\text{Tc}$ (Brito et al., 2012), ^{68}Ga (Koskinas et al., 2014), ^{67}Ga (Dias et al., 2014), ^{111}In (Matos et al., 2014). In this paper, ^{64}Cu standardization methodology is presented. This radionuclide is particularly useful for positron emission tomography (PET), and can also be used in diagnostic investigations, as well as in radiotherapy, medical research and clinical practice, due to its beta-plus and beta minus decay.

^{64}Cu decays with a half-life of 12.7004 (20) h, for 38.48 (26) % by β^- to the ground state of ^{64}Zn and 17.52 (15) % by β^+ competing with 43.53 (20) % by EC to the ground state of ^{64}Ni and for 0.4744 (33) % by EC populating the excited state of ^{64}Ni , followed by 1345.7 keV gamma-ray emission (Bé and Helmer, 2001–2011). The decay scheme of ^{64}Cu is shown in Fig. 1. Due to the short half-life of ^{64}Cu and the proximity of a production center, it is of interest that the LMN is able to standardize this radionuclide with good accuracy. Several papers can be found in the literature related to the standardization

of ^{64}Cu , e.g. (Kawada, 1986; Wanke et al., 2010; Sahagia et al., 2012; Havelka and Sochorová, 2014).

The measurement was carried out with a $4\pi\beta\text{-}\gamma$ coincidence system with proportional counter (PC) and NaI(Tl) gamma-ray spectrometer, as described in Section 2. The disintegration rate was determined by means of the efficiency extrapolation technique using external absorbers covering the sources on both sides. The events were registered by a method developed at the LMN, which makes use of a Time to Amplitude Converter (TAC) in combination with a Multi-channel Analyzer (MCA). In addition, a measurement of the 1345.7 keV gamma-ray emission probability per decay of ^{64}Cu was performed by means a REGe gamma-ray spectrometer.

2. Experimental method

2.1. Source preparation

^{64}Cu was produced by means of the reaction $^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$ in a thermal neutron flux of $2 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$ obtained near the core of the IEA-R1 research reactor, operated at 4.5 MW. The samples,

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<http://dx.doi.org/10.1016/j.apradiso.2017.09.007>

Received 10 March 2017; Received in revised form 3 August 2017; Accepted 5 September 2017

Available online 09 September 2017

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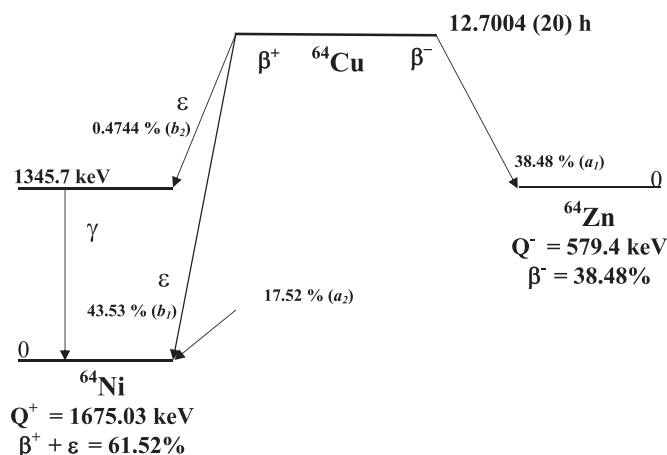


Fig. 1. Decay scheme of ^{64}Cu (Bé and Helmer, 2001–2011).

consisting of 0.3 mg of metallic copper wire, were irradiated for 8 h and left to decay for about 24 h. Next, they were dissolved in 20 μl of HNO₃ 65% and diluted in 20 mL of 0.1 N HCl. From the final solution, several sources to be measured in the 4π β-γ system were prepared by dropping known aliquots on a Collodion film of 20 μg cm⁻² thickness. This film had been previously coated with a 10 μg cm⁻² thick gold layer on each side, in order to render the film conductive. A seeding agent (CYASTAT SN) was used for improving the deposit uniformity and the sources were dried in a desiccator. The accurate source mass determination was performed by the pycnometer technique (Campion, 1975), using a Mettler 56XP balance. A flame-sealed ampoule was prepared for impurity study and for measurement of gamma-ray emission probability per decay. Eight irradiations were carried out, and the solutions obtained did not show any impurity that could affect the activities. From each solution, about six sources were prepared with masses in the range from 10 mg to 50 mg, to be measured in the 4πβ-γ coincidence system, and one ampoule with 1 mL to be measured in the gamma-ray spectrometer.

2.2. 4πβ-γ coincidence measurement

A conventional 4πβ-γ coincidence system was used, consisting of a 4π proportional counter filled with P-10 gas and operated at 0.1 MPa, coupled to a pair of 76 mm × 76 mm NaI(Tl) crystals. The events were registered by a method developed at the LMN, which makes use of a Time-to-Amplitude Converter (TAC) (Baccarelli et al., 2008). Two gamma windows were set: one at the 511 keV positron-annihilation peak and the other at the 1345.7 keV gamma-ray full energy peak, respectively.

The formulae applied to the coincidence measurement, considering the emission probabilities according to the decay scheme shown in Fig. 1 are:

$$N_{4\pi} = N_0 \left[\begin{array}{l} a_1 \varepsilon_{\beta^-} + a_2 \varepsilon_{\beta^+} + b_1 \varepsilon_{EC1} + b_2 \varepsilon_{EC2} + \\ + a_2 (1 - \varepsilon_{\beta^+}) (2 \varepsilon_{\beta\gamma 511} - (\varepsilon_{\beta\gamma 511})^2) \\ + b_2 (1 - \varepsilon_{EC2}) \left(\frac{\alpha_T \varepsilon_{IC} + \varepsilon_{\beta\gamma}}{1 + \alpha_T} \right)_{1345} \end{array} \right] \quad (1)$$

where: N_0 is the disintegration rate;

$N_{4\pi}$ is the total PC counting rate, the subscript 4π was used instead of the usual β, because it includes the beta and the electron capture detection;

a_1 and a_2 are the branching ratios for beta minus and beta plus decay, respectively;

b_1 and b_2 are the branching ratios for electron capture to the excited and ground level, respectively;

ε_{β^-} and ε_{β^+} are the PC detection efficiencies for beta minus and

positrons, respectively;

ε_{EC1} and ε_{EC2} are the PC detection efficiencies for both electron capture events, respectively;

ε_{IC} is the PC detection efficiency for conversion electrons of the 1345.7 keV transition;

$\varepsilon_{\beta\gamma 511}$ and $\varepsilon_{\beta\gamma 1345}$ are the PC detection efficiencies for the 511 keV positron-annihilation quanta and for the 1345.7 keV gamma-ray, respectively;

and α_T is the total internal coefficient of the 1345.7 keV transition.

In the Eq. (1), due to the low value of: $(1 - \varepsilon_{\beta^+}) \cong 0.01$; $\varepsilon_{\beta\gamma 511} \cong 0.0013$ (calculated by Monte Carlo simulation); $a_2 = 0.1752$ (Bé and Helmer, 2001–2011); $\alpha_{T1345} = 1.24 \times 10^{-5}$ (Bé and Helmer, 2001–2011); $\varepsilon_{\beta\gamma 1345} \cong 0.003$ (calculated by Monte Carlo simulation) and the $b_2 = 0.004744$ (Bé and Helmer, 2001–2011); the inefficiencies terms $a_2 (1 - \varepsilon_{\beta^+}) (2 \varepsilon_{\beta\gamma 511} - (\varepsilon_{\beta\gamma 511})^2) \cong 4.6 \times 10^{-5}$ and $b_2 (1 - \varepsilon_{EC2}) \left(\frac{\alpha_T \varepsilon_{IC} + \varepsilon_{\beta\gamma}}{1 + \alpha_T} \right)_{1345} \cong 1.2 \times 10^{-5}$ were neglected, so that the Eq. (1) can be written as:

$$N_{4\pi} = N_0 [a_1 \varepsilon_{\beta^-} + a_2 \varepsilon_{\beta^+} + b_1 \varepsilon_{EC1} + b_2 \varepsilon_{EC2}] \quad (2)$$

The gamma-ray counting rate at the 511 keV positron-annihilation quanta is given by:

$$N_{\gamma 511} = N_0 (a_2 (2 \varepsilon_{\gamma 511} - (\varepsilon_{\gamma 511})^2)) \quad (3)$$

The gamma-ray counting rate at the 1345.7 keV gamma-ray full energy peak, subsequent to the electron capture process, is given by:

$$N_{\gamma 1345} = N_0 (b_2 \varepsilon_{\gamma 1345}) \quad (4)$$

The coincidence counting rates from the two gamma gates are given, respectively by:

$$N_{C511} = N_0 [a_2 \varepsilon_{\beta^+} (2 \varepsilon_{\gamma 511} - (\varepsilon_{\gamma 511})^2)] \quad (5)$$

and

$$N_{C1345} = N_0 [b_2 \varepsilon_{EC2} \varepsilon_{\gamma 1345}] \quad (6)$$

The PC efficiencies are given by:

$$\left(\frac{N_C}{N_\gamma} \right)_{511} = \varepsilon_{\beta^+} \quad (7)$$

and

$$\left(\frac{N_C}{N_\gamma} \right)_{1345} = \varepsilon_{EC2} \quad (8)$$

The PC electron capture efficiencies can be considered equal, as suggested by Kawada (1986), so that $\varepsilon_{EC1} \cong \varepsilon_{EC2} \cong \varepsilon_{EC}$.

The PC detection efficiencies for beta minus and beta plus particles were also considered equal based on theoretical assumption (Havelka and Sochorová, 2014), so that $\varepsilon_{\beta^-} \cong \varepsilon_{\beta^+} \cong \varepsilon_{\beta}$.

Therefore, Eq. (2) can be approximated by:

$$N_{4\pi} = N_0 [(a_1 + a_2) \varepsilon_{\beta} + (b_1 + b_2) \varepsilon_{EC}] \quad (9)$$

Defining $A = (a_1 + a_2)$ and $B = (b_1 + b_2)$ and considering that $(A + B) = 1$

$$N_{4\pi} = N_0 [1 - B (1 - \varepsilon_{EC}) - A (1 - \varepsilon_{\beta})] \quad (10)$$

In order to obtain the disintegration rate, the factor suggested by Kawada (1986) to correct the inefficiency for beta plus and beta minus in the $N_{4\pi}$ has been applied.

This factor is given by:

$$F_K = \left[\frac{1 - B (1 - \varepsilon_{EC})}{1 - B (1 - \varepsilon_{EC}) - (1 - \varepsilon_{\beta}) (1 - B)} \right] \quad (11)$$

Therefore, multiplying both sides of the Eq. (10) by F_K and considering $A = (1 - B)$ we will have:

$$N_{4\pi} F_K = N_0 [1 - B(1 - \varepsilon_{EC}) - (1 - B)(1 - \varepsilon_\beta)] \left[\frac{1 - B(1 - \varepsilon_{EC})}{1 - B(1 - \varepsilon_{EC}) - (1 - \varepsilon_\beta)(1 - B)} \right] \quad (12)$$

Thus, we obtain

$$N'_{4\pi} = N_{4\pi} F_K = N_0 [1 + B(1 - \varepsilon_{EC})] \quad (13)$$

As $B = (I - A)$, dividing the Eq. (13) by Eq. (8), and summing and subtracting A , the conventional equation for coincidence can be written as:

$$\left(\frac{N'_{4\pi} N_\gamma}{N_c} \right)_{1345} = N_0 \left[1 + A \left(\frac{1 - N_c/N_\gamma}{N_c/N_\gamma} \right)_{1345} \right] \quad (14)$$

In this equation, corrections for background, dead time and decay were applied as usual. The coincidence rate N_c was corrected for dead time and accidental coincidences using the Cox-Isham (1977) formalism adapted by Smith (1978).

2.3. Gamma emission probability per decay

The determination of the 1345.7 keV gamma emission probability was performed with a REGe gamma-ray spectrometer with a 500- μ m-thick Be window, having a 1.79 keV FWHM energy resolution at 1332.5 keV. The full peak efficiency curve was measured at 18 cm away from the REGe crystal, in the energy range from 244 keV to 1400 keV, using flamed-sealed 5 mL Schott ampoules with 13.3 mm diameter. Standard solutions of ^{133}Ba , ^{60}Co , ^{152}Eu and ^{137}Cs , previously standardized in the $4\pi\beta-\gamma$ coincidence system in the LMN and prepared in sealed ampoules with 1 mL, were used for calibrating the spectrometer. The geometry of the ^{64}Cu source measured in the gamma spectrometer was the same as for the standard sources.

The area under the peak was evaluated by code ALPINO (Dias, 2001b), which applies the method of simple integration of counts under the total energy absorption peak. Dead time and pile-up corrections were applied by measuring a reference pulser near the upper edge of the gamma-ray spectrum simultaneously with the sources (Debertin and Helmer, 1988). The cascade-summing effects were determined by a Monte Carlo simulation (Dias et al., 2002).

3. Results and discussion

The extrapolation curve from all irradiations normalized by means of the germanium measurements is presented in Fig. 2. The disintegration rate was obtained by a linear least squares fit using code

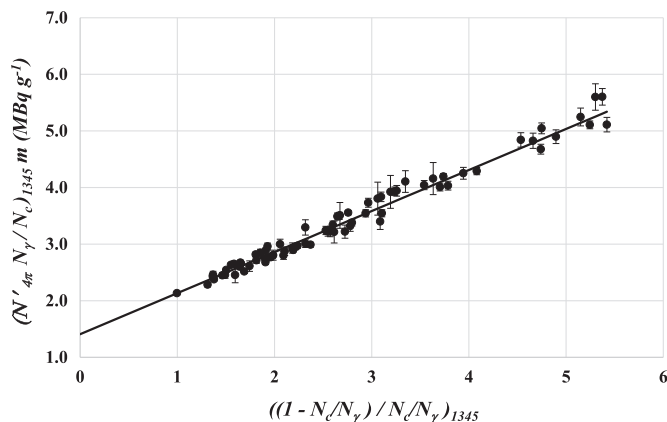


Fig. 2. Extrapolation curves of $(N'_{4\pi} N_\gamma / N_c)$ as a function of $((I - N_c / N_\gamma) / (N_c / N_\gamma))_{1345}$. The dots with uncertainty bars are the experimental points and the continuous line the fitted extrapolation line.

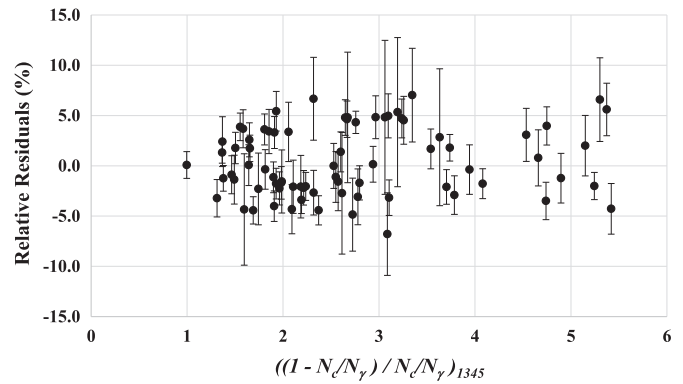


Fig. 3. Relative residuals (in percent) between experimental points and the extrapolation line.

LINFIT (Dias, 2001a). A total of 76 data points were included in the fit. Fig. 3 shows the relative residuals between the experimental points and the fitting, showing no noticeable bias. The electron capture efficiency was changed from 50% to 15% using external absorbers prepared with Collodion films and aluminum foils.

The main uncertainties involved in the measurement were the counting statistics in the electron capture efficiency and the uncertainty in the Kawada factor, included in the fitting. The counting statistical uncertainty in the electron capture efficiency determination was severely enhanced by the lower intensity of the 1345.7 keV gamma-ray. Table 1 shows the typical partial uncertainties in the activity, measured in $4\pi\beta-\gamma$ system, by means of the extrapolation technique and in the determination of the gamma-ray emission probability per decay.

The emission probability per decay of the 1345.7 keV gamma-ray and the corresponding activity obtained were $(0.472 \pm 0.010) \%$ and $(1.408 \pm 0.027) \text{ MBq g}^{-1}$, respectively. The value of the emission probability per decay is compared with the literature in Table 2. It is in agreement with several values presented in the literature, and also with the recommended value by Bé and Helmer (2001–2011).

4. Conclusion

A ^{64}Cu solution was successfully standardized within 1.9% uncertainty by means of $4\pi\beta-\gamma$ coincidence counting. The disintegration rate was determined by the extrapolation technique, applying the Kawada factor, which corrects for the β^+ and β^- inefficiencies. The activity had a relatively high uncertainty due to the short half-life and low gamma-ray intensity. The 1345.7 keV gamma-ray emission probability per decay was determined combining the absolute activity with measurements in a calibrated REGe gamma-ray spectrometer. The result was in good agreement with the literature data.

Acknowledgements

The authors are grateful to the IPEN reactor staff for irradiations and to the National Council for Scientific and Technological

Table 1

Typical partial uncertainties in the activity, measured in $4\pi\beta-\gamma$ system, by means of the extrapolation technique and in the determination of the gamma-ray emission probability, in percent ($u = 1$).

Component	Uncertainty (%)
Counting statistics in electron capture efficiency	0.87–7.4
Kawada factor	0.07–5.7
Extrapolation curve	1.94
HPGe efficiency (1345.7 keV)	0.62
Counting statistic in peak area (1345.7 keV)	0.19
Ampoule geometry	0.83

Table 2

Comparison of the emission probability of the 1345.7 keV gamma-ray per ^{64}Cu decay obtained in the present work, with values from the literature.

Reference	Value ^a
Christmas et al. (1983)	0.471(11)
Kawada (1986)	0.487 (20)
Qaim et al. (2007)	0.543(3)
Wanke et al. (2010)	0.474 (5)
Luca et al. (2012)	0.481 (17)
Amiot et al. (2012)	0.472 (12)
CMI – Bé et al. (2012)	0.476 (6)
This work	0.472 (10)
Evaluation Bé and Helmer (2011-2001)	0.4748 (33)

^a Absolute uncertainty is given in brackets.

Development (CNPq) for partial support to the present research project.

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