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

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HIGHLIGHTS

- The procedure for 4π(PC)β-γ 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.

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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 4π (PC) β-γ 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 ± 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 ^{99m}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 4πβ-γ 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}Cu (n,γ) ^{64}Cu in a thermal neutron flux of 2×10^{13} cm^{-2} s^{-1} obtained near the core of the IEA-R1 research reactor, operated at 4.5 MW. The samples,

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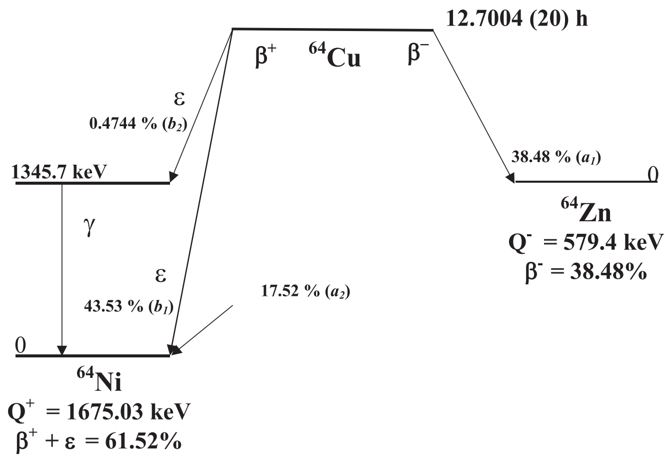

Fig. 1. Decay scheme of ^{64}Cu (Bé and Helmer, 2001–2011).

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 HNO_3 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:

$$N_{4\pi} = N_0 \begin{bmatrix} a_1\ \varepsilon_{\beta^-} + a_2\ \varepsilon_{\beta^+} + b_1\ \varepsilon_{EC1} + b_2\ \varepsilon_{EC2} + \\ + a_2\ (1-\varepsilon_{\beta^+})\ (2\ \varepsilon_{\beta\gamma511} - (\varepsilon_{\beta\gamma511})^2) \\ + b_2\ (1-\varepsilon_{EC2}) \left(\frac{\alpha_T\varepsilon_{IC}+\varepsilon_{\beta\gamma}}{1+\alpha_T}\right)_{1345} \end{bmatrix} \quad (1)$$

where: $\boldsymbol{N_0}$ is the disintegration rate;

$\boldsymbol{N_{4\pi}}$ 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;

$\boldsymbol{a_1}$ and $\boldsymbol{a_2}$ are the branching ratios for beta minus and beta plus decay, respectively;

$\boldsymbol{b_1}$ and $\boldsymbol{b_2}$ 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;

$\varepsilon_{\beta\gamma511}$ and $\varepsilon_{\beta\gamma1345}$ 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: $(1-\varepsilon_{\beta^+}) \cong \mathit{0.01}$; $\varepsilon_{\beta\gamma511} \cong \mathit{0.0013}$ (calculated by Monte Carlo simulation); $\boldsymbol{a_2} = 0.1752$ (Bé and Helmer, 2001–2011); $\boldsymbol{\alpha_{T1345}} = 1.24 \times 10^{-5}$ (Bé and Helmer, 2001–2011); $\boldsymbol{\varepsilon_{\beta\gamma1345}} \cong \mathit{0.003}$ (calculated by Monte Carlo simulation) and the $\boldsymbol{b_2} = 0.004744$ (Bé and Helmer, 2001–2011); the inefficiencies terms $a_2\ (1-\varepsilon_{\beta^+})\ (2\ \varepsilon_{\beta\gamma511} - (\varepsilon_{\beta\gamma511})^2) \cong \mathit{4.6} \times \mathit{10^{-5}}$ and $b_2\ (1-\varepsilon_{EC2}) \left(\frac{\alpha_T\varepsilon_{IC}+\varepsilon_{\beta\gamma}}{1+\alpha_T}\right)_{1345} \cong \mathit{1.2} \times \mathit{10^{-5}}$ 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:

$$N_{\gamma511} = N_0\ (a_2\ (2\ \varepsilon_{\gamma511} - (\varepsilon_{\gamma511})^2)) \quad (3)$$

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

$$N_{\gamma1345} = N_0\ (b_2\ \ \varepsilon_{\gamma1345}) \quad (4)$$

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

$$N_{C511} = N_0\ [a_2\ \varepsilon_{\beta^+}\ (2\ \varepsilon_{\gamma511} - (\varepsilon_{\gamma511})^2)] \quad (5)$$

and

$$N_{C1345} = N_0\ [b_2\ \varepsilon_{EC2}\ \ \varepsilon_{\gamma1345}] \quad (6)$$

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 $\boldsymbol{A} = \boldsymbol{(a_1 + a_2)}$ and $\boldsymbol{B} = \boldsymbol{(b_1 + b_2)}$ and considering that $\boldsymbol{(A + B) = 1}$

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

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 $\boldsymbol{N_{4\pi}}$ 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 $\boldsymbol{F_K}$ and considering $\boldsymbol{A} = \boldsymbol{(1\text{-}B)}$ we will have: