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Disintegration rate measurement of ^{182}Ta

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Abstract

Measurement of the activity of ^{182}Ta sources produced by irradiation at the IPEN research reactor was performed in a $4\pi\beta-\gamma$ coincidence system by using the extrapolation technique. The measurements were undertaken selecting two windows in the γ-channel, in order to check the consistency of the results. A Monte Carlo calculation was performed in order to predict the behavior of the observed activity as a function of the $4\pi\beta$ detector efficiency and the results were compared with experimental values.

Keywords: Activity; Tantalum 182; Coincidence system; Disintegration rate; Monte Carlo simulation

1. Introduction

Spectrometers are useful instruments for the measurement of gamma-ray emitter radionuclides used in several applications, such as environment, medicine, agriculture, and others.

These devices are calibrated with gamma-ray primary standards, some of them with long half-lives such as ^{60}Co, ^{133}Ba, ^{152}Eu, and ^{241}Am, covering an energy range between 59 and 1408 keV. However, some regions lack in standards that can be used to complete the energy range of calibration.

One of these radionuclide is ^{182}Ta which decays with a half-life of 114 days (Firestone et al., 1996) by β^- emission, populating the excited levels of ^{182}W. It emits gamma rays with several energies between 67 and 264 keV and between 1121 and 1289 keV.

In order to use ^{182}Ta as a secondary standard for spectrometer calibrations, the measurement of the activity by means of a primary standard system is required. In the present work, the procedure developed by the Laboratório de Metrologia Nuclear (LMN) of the IPEN-CNEN/SP from São Paulo, for the standardization of this radionuclide by means of a $4\pi\beta-\gamma$ coincidence system is presented.

The measurements were carried out in the $4\pi\beta-\gamma$ coincidence system using two gamma windows. A Monte Carlo simulation program, developed by Takeda et al. (2005), which predicts the behavior of the extrapolation curve, was applied for the two windows. The results were compared with experimental values.

2. Experimental method

2.1. Source preparation

A ^{182}Ta radioactive source was produced from the reaction ^{181}Ta (n,γ) ^{182}Ta. Ta_2O_5 was irradiated in a neutron thermal flux of $1 \times 10^{12}\,\mathrm{cm}^{-2}\,\mathrm{s}^{-1}$ obtained near the core of IPEN research reactor operating at 3.5 MW. After the irradiation, the oxide was mixed with distilled water and aliquots of this mixture were dropped on Collodion film coated with gold. The sources were dried under an infrared lamp and covered with another Collodion film.

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

A $4\pi\beta-\gamma$ coincidence system consisting of a 4π proportional counter coupled to a $76 \times 76\,\mathrm{mm}^2$ NaI(Tl) crystal scintillator was used. The beta counter is a truncated cylinder, made of brass, through which a gas mixture composed of 10% methane and 90% argon is flowing at a pressure of 0.1 MPa. The measurements were undertaken

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using two windows in the γ-channel, in order to check the consistency of the results.

Two windows were selected namely: gamma window 1: from 40 to 300 keV and gamma window 2: from 1000 to 1300 keV.

The activity was obtained by the extrapolation technique. The efficiency was varied using external absorbers of Collodion films previously coated with a $20\,\mu\mathrm{g\,cm^{-2}}$ gold layer, and aluminum foils applied on both sides of the source.

The formulae applied to the coincidence measurement is given by the well-known relationship (Baerg, 1973)

$$\frac{N_\beta N_\gamma}{N_\mathrm{c}} = N_0\left\{1+\left(\frac{1-\varepsilon_\beta}{\varepsilon_\beta}\right)\left(\frac{\alpha\varepsilon_\mathrm{ce}+\varepsilon_{\beta\gamma}}{1+\alpha}\right)\right\} \tag{1}$$

where N_0 is the disintegration rate; N_β the proportional counter counting rate; N_γ the γ-channel counting rate; N_c the coincidence rate; ε_β the proportional counter efficiency for beta particles; $\varepsilon_{\beta\gamma}$ the proportional counter efficiency for γ-rays; α the total conversion coefficient; and ε_ce the proportional counter efficiency for conversion electron.

The observed counting rates N_β and N_γ were corrected for background, dead time, and decay in the usual way. The coincidence rate N_c was corrected for dead time and accidental coincidences using the Cox and Isham (1977) formalism, adapted by Smith (1978).

The electronic system used has been described elsewhere (Koskinas et al., 2006). The TAC method developed in our laboratory was used (Lavras et al., 2000). In this method a Time Amplitude Converter module coupled to a Multichannel Analyzer was used to collect the N_β, N_γ, and N_c observed rates.

A typical spectrum obtained in the multi-channel analyzer is shown in Fig. 1. In this figure, the first peak is due to the beta counts without coincidences, the second peak corresponds to the coincidence counts from the beta and the gamma window 1, the third peak is due to the gamma window 1 counts without coincidence, and the fourth and fifth peaks are due to the second gamma window set. The total beta counts are given by the integration of the first, second, and fourth peaks, the total gamma window 1 counts are given by the integration of the second and third peaks, the total gamma window 2 counts are given by the integration of the fourth and fifth peaks and the coincidence counts of gamma window 1 and gamma window 2 are given by the integration of the second and fourth peaks, respectively.

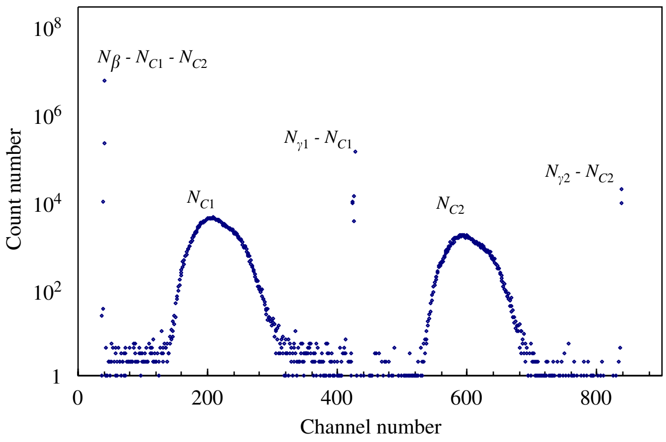

Fig. 1. Typical TAC spectrum with two gamma windows.

The efficiency of gamma window 1 varied from 92% to 58% and the efficiency of gamma window 2 varied from 94% to 63%, respectively.

2.3. Monte Carlo simulation

A Monte Carlo simulation program called ESQUEMA (Takeda et al., 2005) developed at our laboratory was applied for the two gamma windows used. This program uses the decay scheme parameters, the geometry system, and the source characteristics in order to simulate all detection processes in the coincidence system, predicting the behavior of the extrapolation curve.

With the purpose of combining the predicted values obtained with the ESQUEMA code and the experimental data, a least-squares fitting was performed to provide the extrapolated activity N_0. The corresponding χ^2 was given by

$$\chi^2 = (\vec{y}_\mathrm{exp} - N_0\vec{y}_\mathrm{MC})^\mathrm{T} V^{-1}(\vec{y}_\mathrm{exp} - N_0 y_\mathrm{MC}) \tag{2}$$

where $\vec{y}_\mathrm{exp}$ is the experimental vector of $N_\beta\ N_\gamma/N_\mathrm{c}$; $\vec{y}_\mathrm{MC}$ the $N_\beta N_\gamma/N_\mathrm{c}$ vector calculated by Monte Carlo for unitary activity; N_0 the activity of the radioactive source; V the total covariance matrix, including both experimental and calculated uncertainties, and T stands for matrix transposition. The $\vec{y}_\mathrm{MC}$ values used in Eq. (2) correspond to the same efficiency obtained experimentally.

3. Results and discussion

The extrapolation curves of the two gamma windows were obtained by linear least-squares fitting using code LINFIT (Dias, 1999), incorporating covariance matrix methodology. Table 1 shows the extrapolation value obtained for two gamma windows and three sources measured. The ratio of the activity is also indicated.

Table 1
Activity and standard uncertainty obtained by linear least-squares fitting using two gamma windows

Activity[a] (kBq)			
Source	γ Window 1	γ Window 2	Ratio
1	11.145 ± 0.028	11.123 ± 0.030	1.0020 ± 0.0026
2	2.478 ± 0.014	2.477 ± 0.005	1.0004 ± 0.0049
3	2.115 ± 0.010	2.118 ± 0.004	0.9986 ± 0.0041

[a] Standard uncertainties $k = 1$.

Table 2
Typical uncertainties components per data point, in % of the activity

Components	Uncertainty (%)
Counting statistics	0.10
Dead time	0.05
Background	0.30
Decay	0.03
Resolving time	0.10
Extrapolation of efficiency curve	0.30
Combined uncertainty	0.45

Table 3
Comparison between experimental and Monte Carlo simulation values of the activity[a] obtained with gamma window 1

Source	Experimental (kBq)	Monte Carlo (kBq)	Ratio
1	11.145 ± 0.028	11.877 ± 0.020	1.025 ± 0.002
2	2.478 ± 0.014	2.470 ± 0.004	1.003 ± 0.005
3	2.115 ± 0.010	2.113 ± 0.003	1.001 ± 0.004

[a] Standard uncertainties $k = 1$.

Table 4
Comparison between experimental and Monte Carlo simulation values of the activity[a] obtained with gamma window 2

Source	Experimental (kBq)	Monte Carlo (kBq)	Ratio
1	11.123 ± 0.030	11.121 ± 0.017	1.0002 ± 0.0023
2	2.477 ± 0.005	2.476 ± 0.005	1.0004 ± 0.0020
3	2.118 ± 0.004	2.114 ± 0.004	1.0019 ± 0.0019

[a] Standard uncertainties $k = 1$.

The main uncertainties involved in the measurement were: counting statistics, dead time, half-life, and extrapolation curve efficiency. All these uncertainties are included in the fitting. The typical uncertainty components in percentage per data point are presented in Table 2.

The comparison between experimental data and Monte Carlo simulation values are shown in Tables 3 and 4 for gamma windows 1 and 2, respectively. The ratio between these two activity values is also indicated.

The activity values obtained with Monte Carlo simulation applying Eq. (2) agree quite well with the experimental data for gamma window 2. However, the value obtained by Monte Carlo simulation for gamma window 1 does not agree so well with the corresponding fitting value. This may be due to differences in curve shapes between experimental data points and Monte Carlo simulation. The reason for this effect is being investigated.

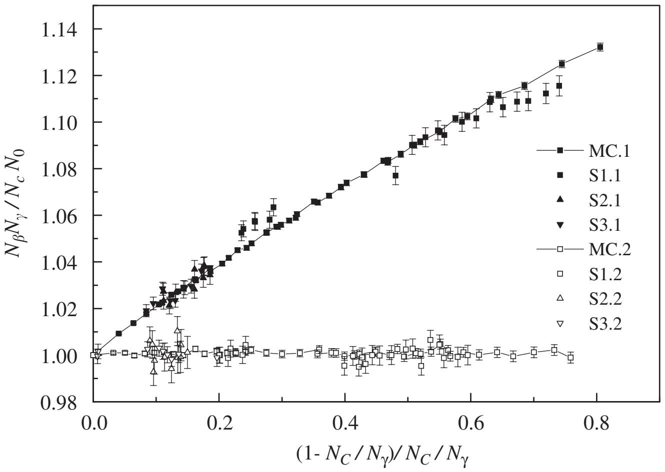

Fig. 2. Behavior of $N_\beta N_\gamma / N_c N_0$ as a function of $(1-N_c/N_\gamma)/(N_c/N_\gamma)$ for two gamma window settings. Closed figures correspond to the experimental data of the three sources measured; open circles correspond to the Monte Carlo simulation.

The behavior of $N_\beta N_\gamma / N_c N_0$ as a function of $(1-N_c/N_\gamma)/(N_c/N_\gamma)$ for the two gamma window settings are shown in Fig. 2. The disintegration rate N_0, of each source, obtained by averaging the ratio $\vec{y}_{\mathrm{exp}}/\vec{y}_{\mathrm{MC}}$ was used in order to normalize the experimental data to be compared with the Monte Carlo unitary activity calculation.

Additional gamma spectrometry measurements are being carried out for determining the emission probability per decay of the most intense gamma rays of ^{182}Ta. These results will be compared with values taken from the literature and published in the future work.

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