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Standardization of a ^{204}Tl radioactive solution

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Abstract

The standardization of ^{204}Tl is described. The efficiency tracing technique was applied using ^{134}Cs as tracer. The 4πβ–γ coincidence system was used for the calibration. The (Laboratorio de Metrologia Nuclear) has participated in this comparison in collaboration with the Laboratório Nacional de Metrologia das Radiações Ionizantes, from Rio de Janeiro. Independent results using different techniques were developed by each of these laboratories and included in the comparison.

Keywords: Activity; Thallium 204; Tracing method; Coincidence system

1. Introduction

The procedure followed by the Laboratório de Metrologia Nuclear (LMN) at the IPEN-CNEN/SP, in São Paulo, for the standardization of ^{204}Tl is described. This radionuclide has been selected by the (Comité Consultative pour les Etalons de Mesures des Rayonnements Ionisants) (CCEMRI) for an international comparison due to its decay scheme characteristics. The LMN have participated in this comparison in collaboration with the Laboratório Nacional de Metrologia das Radiações Ionizantes (LNMRI), from Rio de Janeiro. Independent results using different techniques were developed by each of these laboratories and included in the comparison. A comprehensive report including all results will be published by the BIPM in due time.

^{204}Tl decays 97% by β^- emission and 3% by electron capture, as shown in Fig. 1 (Lagoutine et al., 1983). This feature makes it a suitable nuclide to be standardized by the tracing technique (ICRU, 1994). The radionuclide chosen as tracer was ^{134}Cs, because of its end-point beta energy, which is similar to that of ^{204}Tl. The calibration was performed in a 4πβ−γ coincidence system selecting a γ−ray window in the 796–802 keV energy range. In this window, the expected decay scheme correction for ^{134}Cs is small.

2. Source preparation

The ^{204}Tl solution was taken from an ampoule sent by BIPM. The sources to be measured in the 4πβ−γ system have been prepared by dropping known aliquots of each radioactive solution on a 20 μg cm^{-2} thick Collodion film. This film had been previously coated with a 10 μg cm^{-2} thick gold layer in order to render the film conducting. Seven mixed sources were prepared using a 1:2 ratio (β-pure and β–γ, respectively), in order to obtain similar counting rates for both radionuclides. A seeding agent (CYASTAT SN) was used for improving the deposit uniformity and the sources were dried in a warm (45°C) nitrogen jet (Wyllie et al., 1970). The source masses were accurately determined by the picnometer technique (Campion, 1975). The β–γ tracer has been previously standardized by measuring several sources prepared by the same procedure.

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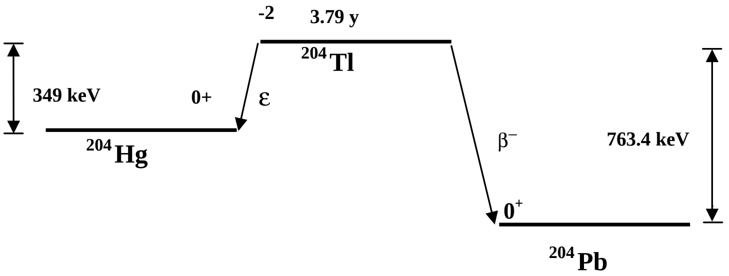

Fig. 1. Decay scheme of 204Tl (Lagoutine et al., 1983).

3. 4πβ−γ coincidence measurements

The system for absolute standardization (Moura, 1969; Fonseca, 1997) consisted of a gas-flow proportional counter with 4π geometry using 90% Ar + 10% CH_4 gas at 0.1 MPa, as the β detector and coupled to a pair of 76 mm × 76 mm NaI(Tl) scintillation counters, as γ detectors. The selected γ-channel window covered both the 796 and 802 keV peaks of ^{134}Cs.

The number of detected events in the proportional counter is given by:

$$N_{(Tl+Cs)} = N_{0Cs}\varepsilon_{\beta Cs} + N_{0\beta Tl}\varepsilon_{\beta Tl}.$$

where $\varepsilon_{\beta Cs}$ is the tracer efficiency in the mixture source, obtained by the ratio N_c/N_γ, $N_{\beta(Tl+Cs)}$ is the counting rate of proportional counter due to the mixture source, N_{0Cs} is the activity of ^{134}Cs tracer, deposited on the mixture source, m_{Cs} is the tracer mass deposited on the mixed source, $N_{0\beta Tl}$ is the ^{204}Tl beta-branch disintegration rate and $\varepsilon_{\beta Tl}$ is the ^{204}Tl beta efficiency.

When the β-emitter and the β–γ tracer are combined in a single source, a functional relationship exists between the detection efficiencies (ICRU, 1994). This relation can be defined by a polynomial function G where

$$(1-\varepsilon_{\beta Tl})/\varepsilon_{\beta Tl} = G((1-\varepsilon_{\beta Cs})/\varepsilon_{\beta Cs}).$$

Applying the extrapolation technique we can write the expression as:

$$\frac{N_{\beta(Tl+Cs)}N_\gamma}{N_c} - N_{0Cs} = N_{0\beta Tl}[1 + G'((1-\varepsilon_{\beta Cs})/\varepsilon_{\beta Cs})].$$

The function G' was fitted by weighted least squares using code LINFIT (Dias, 1998) and the extrapolation to $(1-\varepsilon_{\beta Cs})/\varepsilon_{\beta_{Cs}} = 0$ gave the expected $N_{0\beta Tl}$ value. Suitable corrections for decay were included in the activities.

The final activity is given by

$$N_0 = \frac{N_{0\beta Tl}}{I_\beta + \varepsilon_{EC}I_{EC}}$$

where N_0 is the disintegration rate of ^{204}Tl, I_β and I_{EC} are the beta decay and electron capture branching ratios and ε_{EC} is the efficiency of electron capture events.

In the case of ^{204}Tl the efficiency of electron capture events is dominated by Auger electron detection because the proportional counter, operating at 0.1 MPa, has low efficiency for 8-83 keV X-rays. Neglecting transitions above the L shell, the value of ε_{EC} is given approximately by

$$\begin{aligned}\varepsilon_{EC} &= \varepsilon_{AeK}P_K(1-\omega_k) + \varepsilon_{AeL}P_L(1-\omega_L)\\ &\quad - \varepsilon_{AeK}\varepsilon_{AeL}P_KP_L(1-\omega_k)(1-\omega_L),\end{aligned}$$

where subscripts K and L correspond to K and L shells, respectively, w_K and w_L are the fluorescence yields, ε_{AeK} and ε_{AeL} are the detection efficiency for Auger electrons and P_K and P_L are the relative probabilities of electron capture.

The last part of this equation takes into account simultaneous detection of K and L Auger electrons and it is small for the case of ^{204}Tl.

A comparison was performed between the extrapolation technique and a semi-empirical function developed to establish a relationship between the tracer efficiency and the β-pure emitter efficiency, considering the end-point beta-ray energy of each radionuclide (Dias, 2002).

This function is given by:

$$r = \frac{\varepsilon_{\beta Cs}}{\varepsilon_{\beta Tl}} = \exp\left\{-\ln(\varepsilon_{\beta Cs})\left[1-\left(\frac{E_{Cs}}{E_{Tl}}\right)^{1.14}\right]\right\},$$

where E_{cs} is the end-point beta-ray energy of ^{134}Cs, and E_{Tl} is the end-point beta-ray energy of ^{204}Tl.

The $N_{0\beta Tl}$ value was obtained by the following expression:

$$N_{0\beta Tl} = \left\{\frac{N_{\beta(Tl+Cs)}N_\gamma}{N_c} - N_{0Cs}\right\}\frac{1}{\varepsilon_{\beta Cs}r}.$$

4. Results and discussion

Fig. 2 shows the behavior of $N_{\beta Tl}$ as a function of $(1-N_c/N_\gamma)/(N_c/N_\gamma)$. In this curve the efficiency was

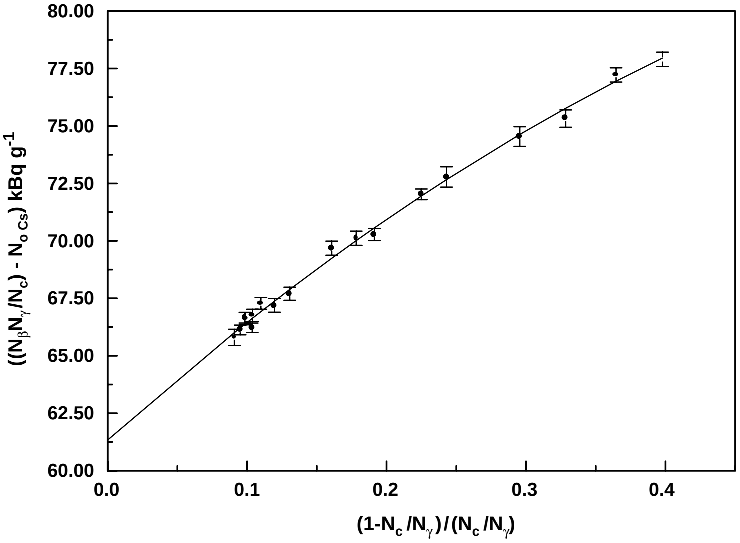

Fig. 2. Extrapolation curve of $(N_{\beta(Tl+Cs)}N_\gamma/N_c - N_{0Cs})$ as a function of $(1 - N_c/N_\gamma)/(N_c/N_\gamma)$.

Table 1
Fitting parameter obtained with the tracing technique

Value (kBq g^{-1})	Correction for β and EC branching ratios	Activity (kBq g^{-1})
Intercept 61.33±0.37	0.987±0.004	62.13±0.47

Table 2
Partial uncertainties in percent (one standard deviation) involved in the activity determination

Components	Uncertainty
Counting statistics	0.14
Background	0.002
Dead time	0.0001
Weighing	0.19
Decay scheme parameter	0.40
Half-life	0.017
Extrapolation of efficiency curve	0.60
Combined uncertainty	0.76

Table 3
Values obtained by applying the semi-empirical factor *r*

$(N_{\beta(Tl+Cs)}N_\gamma/$ $N_c-N_{0Cs})/\varepsilon_\beta$ $Cs \times 10^3$ kBq g^{-1}	Factor *r*	$N_{0\beta Tl}\times 10^3$ kBq g^{-1}
65.79	1.0663	61.70
66.12	1.0693	61.83
66.65	1.0717	62.22
66.61	1.0719	62.14
66.21	1.0753	61.57
66.75	1.0754	62.07
67.28	1.0796	62.32
67.19	1.0863	61.85
67.69	1.0939	61.88
69.68	1.1142	62.54
70.11	1.1259	62.27
70.27	1.1343	61.95
72.02	1.1557	62.32
72.78	1.1669	62.37
74.54	1.1985	62.19
75.31	1.2173	61.87
77.22	1.2372	62.42
77.90	1.2545	62.10
Average	62.09	

varied in the range of 71–91%. The best fit has been obtained with a second degree polynomial and the reduced chi-square was 1.03.

Previous measurements of ^{54}Mn and ^{57}Co, taken in similar conditions, showed that the detection efficiency for 5 keV Auger electrons was around 50%. This value increases to 100% for energies of 10 keV and above. Since the L Auger electrons are emitted by ^{204}Tl in the 5–14 keV energy range, an averaged value of (75±25)%

was assumed for ε_{AeL}. The K electrons are emitted in the 53–83 keV, therefore it was assumed 100% for ε_{AeK}. Considering $I_{EC} = (2.6 \pm 0.1)\%$ (Lagoutine et al., 1983) the value of $\varepsilon_{EC} I_{EC}$ resulted was $(1.3 \pm 0.4)\%$.

The fitting extrapolation values of $N_{0\beta}$ and activity are presented in Table 1. The final activity value was $(62.13 \pm 0.47)\,\mathrm{kBq\,g^{-1}}$. The uncertainty components of the tracing method are shown in Table 2. The main component of the overall uncertainty is due to the extrapolation curve shown in Fig. 2 and amounts to 0.61%. This result needs to be compared with the values obtained by other laboratories which took part in this International Comparison organised by the Bureau International des Poids et Mesures.

The last column in Table 3 shows the values obtained using the semi-empirical function (r). The average value of $N_{0\beta}$ was $62.09\,\mathrm{kBq\,g^{-1}}$ and agrees within 1.2% with the intercept shown in Table 1. This indicates that this procedure can be useful for measuring β-pure radionuclides in a simple and accurate way. Other β-pure emitters are being investigated and a comprehensive report will be published in due time (Dias, 2002).

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