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Disintegration rate measurement of a ^{152}Eu solution

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Abstract

The procedure followed by the Laboratório de Metrologia Nuclear at the IPEN-CNEN/SP, in São Paulo, for the standardization of ^{152}Eu is described. The disintegration rate of ^{152}Eu has been measured using the $4\pi\beta-\gamma$ coincidence technique, using a 4π proportional counter, filled with P-10 gas and operated at 0.1 MPa, coupled to one HPGe detector for the γ-ray emission. Two discrimination windows were set in the γ-channel, one related to the beta branch (344 keV) and the other related to the electron capture events (1408.03 keV), in order to determine the counting efficiencies for β, X-ray and Auger electron events in the proportional counter. The activity of solution was determined by a biparametric extrapolation curve obtained for the two selected γ-windows. © 2002 Published by Elsevier Science Ltd.

1. Introduction

The procedure followed by the Laboratório de Metrologia Nuclear (LMN) at the IPEN-CNEN/SP, in São Paulo, for the standardization of ^{152}Eu is described. This radionuclide was selected by the Comité Consultatif pour les Rayonnements Ionisants (CCRI) for an international comparison due to its decay scheme characteristics. The LMN has 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 obtained by each of these laboratories and included in the comparison. A comprehensive report including all results will be published by the Bureau International des Poids et Mesures (BIPM) in due time.

^{152}Eu decays 27% by β^- emission and 73% by electron capture and β^+ emission and has a half-life of about 4939 days. Both branches are followed by γ-ray emission, as shown in Fig. 1 (Lagoutine et al., 1987). The complexity of the decay scheme, in principle, should require the use of two separate coincidence systems, such as one 4πPC-NaI(Tl) for measuring the β-branch and another NaI(Tl)-NaI(Tl) for the electron capture branch. However, the high internal conversion coefficients and the high number of γ-ray energies from both branches, makes the use of a conventional coincidence system difficult. To solve this problem the gamma detector was changed to a semi conductor HPGe in order to obtain better energy resolution. This made the measurement of gamma rays from different branches without interference possible. The calibration was performed selecting two γ-ray windows: 344 keV following β^- decay and 1408.03 keV following the electron capture and β^+ processes.

Measurements were carried out in a 4πPC-HPGe as described in the following sections. The activity of solution was determined by means of a biparametric extrapolation curve. The efficiencies were changed by external absorbers.

2. Source preparation

The ^{152}Eu solution was taken from an ampoule sent by the BIPM. The sources to be measured in

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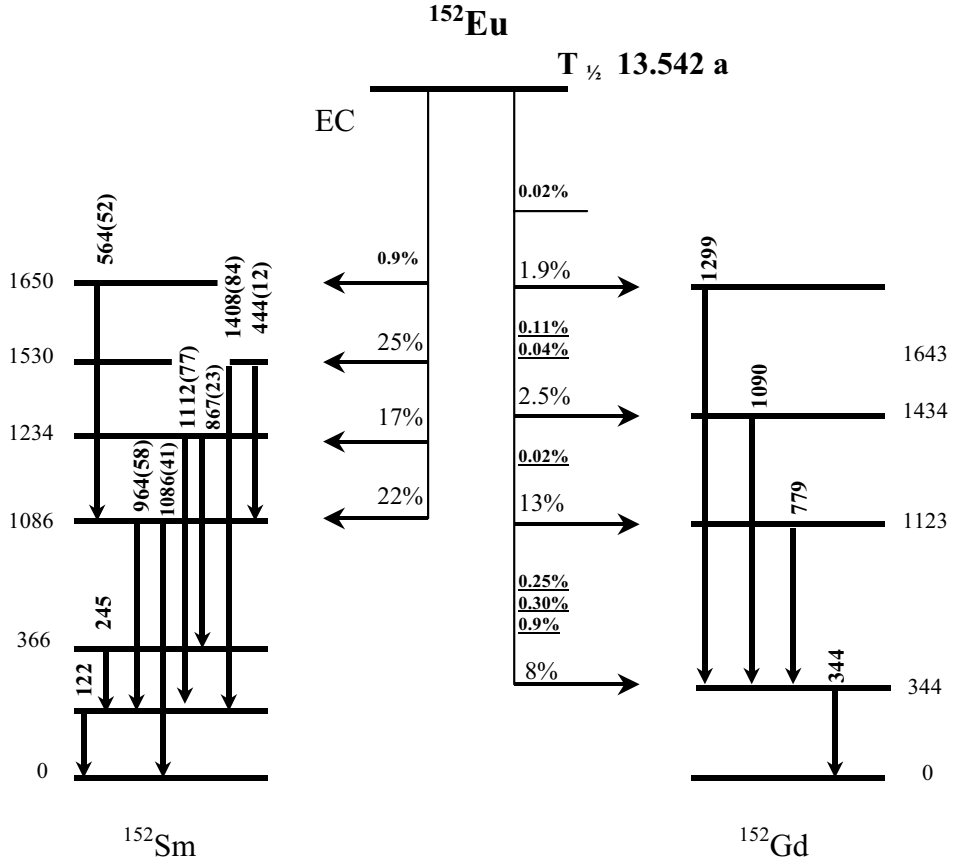

Fig. 1. Simplified decay scheme of ^{152}Eu (Lagoutine et al., 1987). All energies are in keV.

the $4\pi\beta$-γ system were prepared by dropping known aliquots of the radioactive solution onto a $20\,\mu g\,cm^{-2}$ thick COLLODION film. This film had been previously coated on both sides with a $10\,\mu g\,cm^{-2}$ thick gold layer in order to render the film conducting. 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, 1970). The source masses were accurately determined by the pycnometer technique (Campion, 1975).

3. $4\pi\beta$-γ Coincidence measurements

The system for absolute standardization (Moura, 1969) consisted of a gas-flow proportional counter with 4π geometry and using 90% Ar + 10% CH_4 gas at 0.1 MPa, as the β detector, and coupled to a $77\,cm^3$ HPGe(RE) detector (22% efficiency relative to a NaI(Tl) scintillation detector at 1.33 MeV).

The formulae applied to the coincidence measurement were:

$$N_\beta = N_0 a\left[\varepsilon_\beta + (1-\varepsilon_\beta) I_{\mathrm{Gd}}\left(\frac{\alpha\varepsilon_{\mathrm{ec}} + \varepsilon_{\beta\gamma}}{1+\alpha}\right)_{\mathrm{Gd}}\right] + N_0 b\left[\varepsilon_{(X,A)} + (1-\varepsilon_{(X,A)}) I_{\mathrm{Sm}}\left(\frac{\alpha\varepsilon_{\mathrm{ec}} + \varepsilon_{\beta\gamma}}{1+\alpha}\right)_{\mathrm{Sm}}\right], \quad (1)$$

$$N_{\gamma 1} = N_0 a I \frac{\varepsilon\gamma}{1+\alpha_\gamma}, \quad (2)$$

$$N_{c_1} = N_0 a I\left[\varepsilon\beta\frac{\varepsilon\gamma}{1+\alpha}\right] \quad (3)$$

for the β^- branch, and

$$N_{\gamma_2} = N_0 b I \frac{\varepsilon\gamma}{1+\alpha_\gamma}, \quad (4)$$

$$N_{c_2} = N_0 b I\left[(\varepsilon_{(X,A)} + \varepsilon_{\mathrm{ec}})\frac{\varepsilon\gamma}{1+\alpha}\right] \quad (5)$$

for the EC-β^+ branch,
where

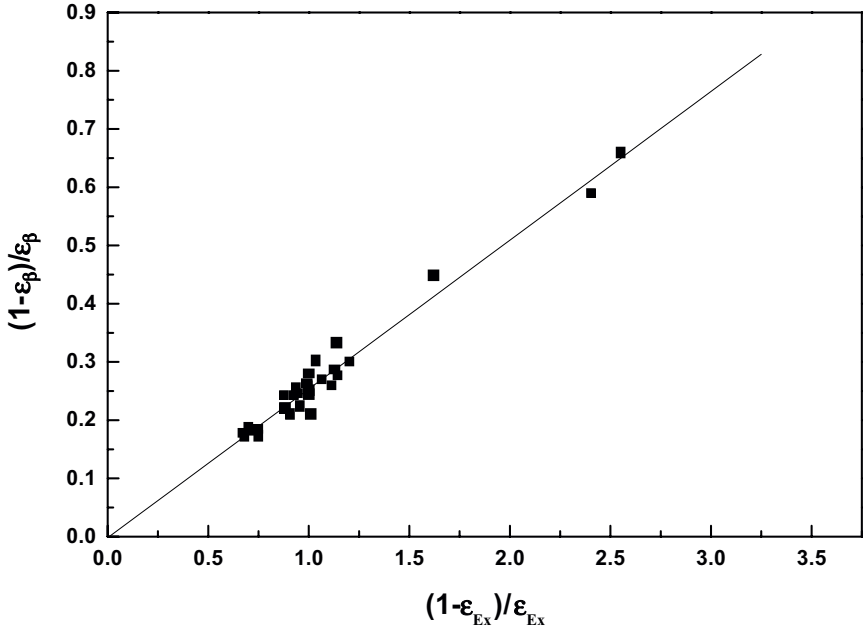

Fig. 2. Regression curve between inefficiency parameters.

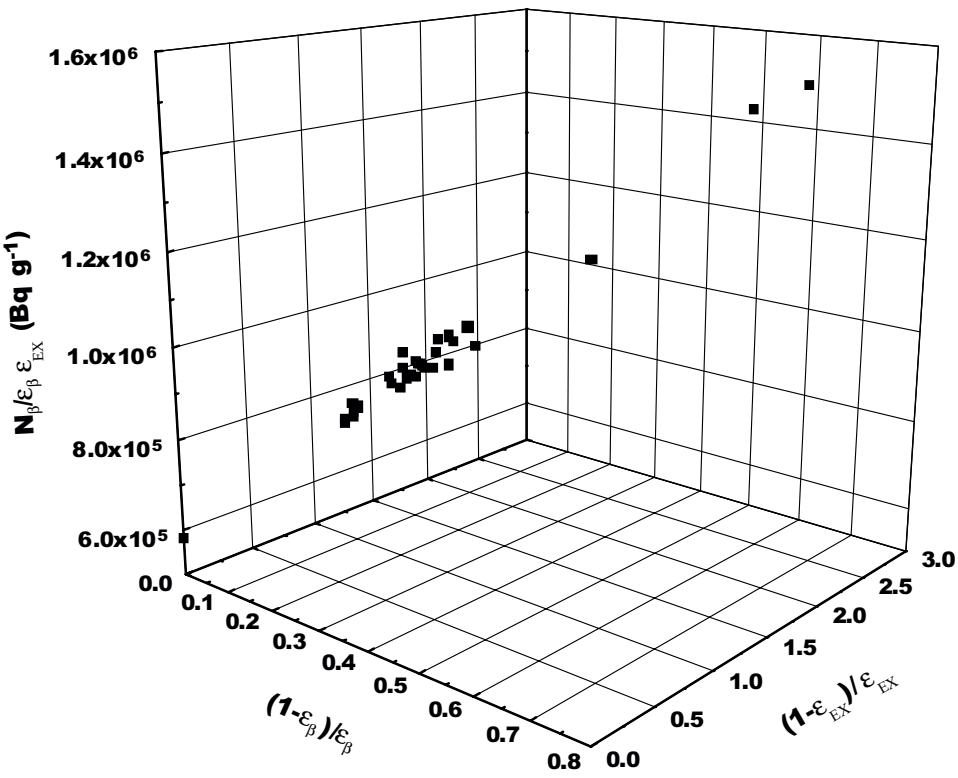

Fig. 3. Biparametric extrapolation curve.

N_0 is the disintegration rate, a probability of β^- disintegration, b probability of decay by EC and β^+ processes, N_β proportional counter counting rate, $N_{\gamma 1}$, N_{c1} are β^- branch γ-channel counting rate and coincidence rate, respectively, $N_{\gamma 2}$, N_{c2} are EC-β^+ branch γ-channel and coincidence counting rates, respectively, $\varepsilon_{X,A}$, ε_{ce} are the proportional counter efficiencies for X-rays plus Auger electrons and conversion electrons, respectively. $\varepsilon_{\beta\gamma}$ is the proportional counter efficiency for γ-rays, α is the total internal conversion coefficient, I_{Gd} and I_{ISm} are the probabilities of γ-ray transitions of the γ-window selected for ^{152}Gd and ^{152}Sm branches, respectively.

The observed counting rates were corrected for background, dead time and decay in the usual way (Koskinas, 1988). The coincidence rates were corrected for dead time and accidental coincidences using the Cox—Isham formalism (Cox and Isham, 1977).

The disintegration rate N_0 was determined by changing the efficiency using external absorbers over and below the radioactive source made of conducting collodion films previously coated with a $40\,\mu g\,cm^{-2}$ gold layer. The activity was determined by simultaneous extrapolation to zero of inefficiency parameters $(1 - N_{ci}/N_{\gamma i})/N_{ci}/N_{\gamma i})$.

The biparametric extrapolation function was

$$y = A + Bx_1 + Cx_2, \tag{6}$$

where

$$X_1 = (1 - N_{c1}/N_{\gamma 1})/N_{c1}/N_{\gamma 1}) \tag{7}$$

$$X_2 = (1 - N_{c2}/N_{\gamma 2})/N_{c2}/N_{\gamma 2}), \tag{8}$$

and

$$Y = \frac{N_\beta}{\varepsilon_\beta \varepsilon_{EX}} \tag{9}$$

where $\varepsilon_\beta = N_{c1}/N_{\gamma 1}$ is the efficiency for the beta branch and $\varepsilon_{EX} = N_{c2}/N_{\gamma 2}$ is the efficiency for the electron capture plus β^+ branch.

Table 1
Inefficiency parameters, experimental and fitted values

X_1	X_2	$Y \times 10^5$ (Bq g^{-1})	Uncertainty (%)	Fit $\times 10^5$ (Bq g^{-1})	Residues (%)
0.65755	2.55707	14.93	0.83	15.17	−1.58
0.58848	2.40518	14.66	0.85	14.61	0.34
0.44785	1.62398	11.91	1.04	11.76	1.28
0.29959	1.2058	10.46	1.19	10.22	2.35
0.28597	1.1318	10.15	1.23	9.946	2.05
0.33212	1.1416	10.14	1.23	9.990	1.50
0.27507	1.1468	10.13	1.23	9.998	1.32
0.30066	1.03691	9.676	1.29	9.605	0.74
0.27857	1.00449	9.482	1.31	9.483	−0.01
0.26869	1.0656	9.789	1.27	9.703	0.89
0.26265	0.99299	9.413	1.32	9.439	−0.28
0.25500	0.94262	9.132	1.36	9.255	−1.33
0.25764	1.11842	9.990	1.25	9.892	0.99
0.25026	1.00349	9.390	1.33	9.475	−0.90
0.24783	0.99876	9.363	1.33	9.457	−0.99
0.25660	0.99689	9.447	1.32	9.452	−0.05
0.24517	0.94303	9.160	1.36	9.255	−1.03
0.24138	0.92927	9.083	1.37	9.204	−1.31
0.2414	0.88292	8.864	1.40	9.036	−1.90
0.24258	1.00441	9.437	1.32	9.477	−0.42
0.20958	1.01438	9.433	1.32	9.507	−0.78
0.20932	0.9092	9.028	1.38	9.126	−1.07
0.21911	0.88434	9.044	1.38	9.038	0.07
0.22283	0.9589	9.361	1.33	9.308	0.57
0.18303	0.75138	8.477	1.47	8.550	−0.85
0.18484	0.70605	8.337	1.49	8.386	−0.58
0.18222	0.72236	8.412	1.48	8.444	−0.38
0.17609	0.67434	8.207	1.52	8.269	−0.75
0.17235	0.68481	8.239	1.51	8.307	−0.82
0.17138	0.75217	8.762	1.42	8.550	2.48

Results and discussion

Fig. 3 shows the biparametric extrapolation curve obtained for the two selected γ-windows. In this curve, the β efficiency was varied in the range from 60% to 85% and the X-ray, Auger electron efficiency was varied in the range from 28% to 60%, using external absorbers.

Inefficiency parameters, experimental values (y) with uncertainties and fitting values obtained by linear least square fitting using the code LINFIT, developed at our laboratory, are presented in Table 1. An additional fit was performed, indicating a linear correlation between the two inefficiency parameters. The resulting curve is shown in Fig. 2, and the fitting parameters are presented in Table 2. The extrapolated value was nearly zero within the experimental errors, indicating that the EC-β^+ efficiency goes to one, as the β^- efficiency goes to one.

The extrapolated disintegration value was $579.1\,\mathrm{kBq\,g^{-1}}$. The impurity ^{154}Eu present in the solution was measured using an HPGe spectrometer previously calibrated with a ^{152}Eu standard source from International Atomic Energy Agency (IAEA). The impurity contribution (0.7%) was subtracted from the extrapolated value. Table 3 shows the partial errors involved in the measurement. The main contribution comes from the extrapolation procedure. The final value obtained in the present measurement was $575.2\,\mathrm{kBq\,g^{-1}}$ with 1.1% total uncertainty.

Table 2
Fitting values of linear regression between inefficiency parameters

$X_1 = A + BX_2$	Fitting parameters
A	-0.0019 ± 0.010
B	0.2554 ± 0.0087

Table 3
Uncertainties components involved in the measurements

Uncertainty components	%
Counting statistics	[a]
Weighing	0.19
Dead time	<0.01
Impurities	0.02
Half life	0.07
Extrapolation of efficiency curve	1.08
Combined uncertainty	1.1

[a] The uncertainty in the counting statistics is included in the uncertainty in the extrapolation of efficiency curve.

References

Campion, P.J., 1975. Procedures for accurately diluting and dispensing radioactive solutions. Bureau International des Poids et Mesures, Monographie BIPM - 1.

Cox, D.R., Isham, V., 1977. A bivariate point process connected with electronic counters. Proc. Roy. Soc. A 356, 149.

Koskinas, M.F., 1988. Desenvolvimento de um sistema de coincidência para a medida absoluta da atividade de radionuclídeos empregando detectores de barreira de superfície. Ph.D. Thesis, University of São Paulo.

Lagoutine, F., Coursol, N., Legrand, J., 1987. Table of radionucléides. Laboratoire de Métrologie des Rayonnements Ionisants. Bureau National de Métrologie.

Moura, L.P., 1969. Generalized coincidence method for absolute activity measurements of radionuclides. Application for determining the internal conversion coefficient of Tl-203 279-keV transition. Ph.D. Thesis, University of Campinas.

Wyllie, H.A., Johnson, E.P., Lowenthal, G.C., 1970. A procedure for stirring aliquots of radioactive solutions when making thin 4π counting sources. Int. J. Appl. Radiat. Isot. 21, 497.