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Disintegration rate measurement of a ^{192}Ir solution

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Abstract

The disintegration rate of ^{192}Ir has been measured using the 4πβ–γ coincidence technique. This radionuclide decays by electron capture (EC) and β-emission. Since the EC contribution is low (4.5%), it has been corrected using decay scheme data taken from the literature. This measurement has been performed in collaboration with the Laboratório Nacional de Metrologia das Radiações Ionizantes (IRDDM), in Rio de Janeiro. The results, which were obtained independently and employed different techniques, are compared with the Systéme International Reference (SIR) maintained at the Bureau International des Poids et Mesures. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Activity; Iridium 192; Coincidence system; Disintegration rate

1. Introduction

In recent years, the Laboratório de Metrologia Nuclear of IPEN has been involved in developing procedures for standardization of radionuclides used in nuclear medicine, to improve the standards produced in Brazil for these radionuclides. ^{192}Ir is one of the selected radionuclides because of its applications in medicine and industry.

The decay scheme of ^{192}Ir is shown in Fig. 1 (Lagoutine et al., 1984). This nuclide decays with a half-life of 73.831 days, 95.24% by β-decay and 4.76% by electron capture (EC); both branches are followed by γ-ray emission. The complexity of the decay scheme, in principle, should require the use of two separated coincidence systems, for instance: 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 low γ-ray emission probability from EC-branch, makes the use of a second system very difficult. In addition, the X-rays coming from the electron capture and those from the internal conversion cannot be separated. Nevertheless, since the contribution form the EC-branch is small, this correction has been calculated using data from the literature.

The measurements were carried out in a 4πβ–γ coincidence system (Fonseca, 1997) as described in Section 2. The contribution from electron capture events in the proportional counter, has been estimated by means of 4πβ(PC)–γGe(Li) coincidence measurements.

The present work has been performed in collaboration with the Laboratório Nacional de Metrologia das Radiações Ionizantes (IRDDM), from Rio de Janeiro. Dr. Hino from the ETL in Japan kindly supplied the ^{192}Ir solution.

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2. Source preparation

The counting sources were prepared by dispensing known aliquots of $IrNa_2Cl_6$ in 0.1 N of HCl solution on 20 mg/cm^2 thick Collodion films. This film had been previously coated with a 10 mg/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 warm (45°C) nitrogen jet (Wyllie et al., 1970). The accurate source mass determination was performed using a Mettler 5SA balance by the pycnometric technique (Campion, 1975).

3. Standardization methods

3.1. 4πβ–γ Coincidence method

A conventional 4πβ–γ coincidence system was used (Moura, 1969; Fonseca, 1997) consisting of 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. Two discrimination windows were set in the γ-channel, both related to the β-branch, in order to check the consistency in the results. The γ-ray windows selected were:

$$Window\,1 = 295.96\,\text{keV} + 308.46\,\text{keV} + 316.51\,\text{keV}$$

$$Window\,2 = 588.59\,\text{keV} + 593.40\,\text{keV} + 604.42\,\text{keV} + 612.47\,\text{keV}$$

The discrimination of γ-rays from the EC-branch was very difficult to achieve because of the poor resolution of NaI(Tl) and lower intensity of these γ-rays, as compared to those following the β-decay. Therefore, it was not possible to determine directly the sensitivity of the proportional counter to X-rays and electrons coming from electron capture. This sensitivity had been determined separately, using another system where the NaI(Tl) scintillator had been replaced by a Ge(Li) detector as described below.

The formulae applied to the coincidence measurement are the following:

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

$$N\gamma = N_0 I \frac{\varepsilon_\gamma}{1+\alpha_\gamma}$$

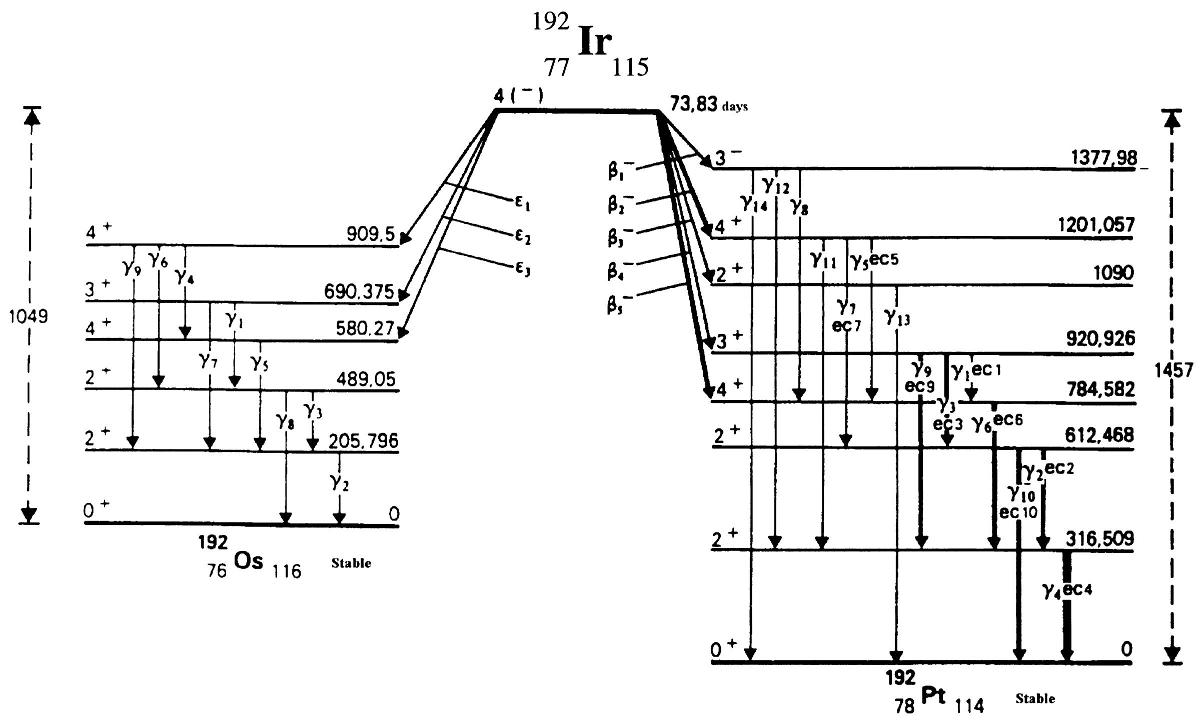


Fig. 1. Decay scheme of ^{192}Ir (Lagoutine et al., 1984). All energies are in keV.

and

$$N_c = N_0 a\left[\varepsilon_\beta \frac{\varepsilon_\gamma}{1+\alpha}\right]$$

where $N_0 a$ is the disintegration rate corresponding to β-branch, a is the β-branching ratio, b is the EC branching ratio, N_β is the proportional counter counting rate, N_γ is the γ-channel counting rate, N_c is the coincidence rate, $\varepsilon_{X,A}$is the proportional counter efficiency for X-rays and Auger electrons, ε_{ce} is the proportional counter efficiency for conversion electrons, $\varepsilon_{\beta\gamma}$ is the proportional counter efficiency for γ-rays, α is the total conversion coefficient, I_{Pt}, I_{Os} are the probabilities of γ-ray transitions of the γ-window selected, ^{192}Pt and ^{192}Os branches, respectively.

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–Isham formalism (Cox and Isham, 1977).

The $N_\beta N_\gamma/N_c$ value is given by:

$$\frac{N_\beta N_\gamma}{N_c} = N_0\left[a + \left(\frac{1-\varepsilon_\beta}{\varepsilon_\beta}\right)B + bf_x\right]$$

where f_X is an additional correction, obtained experimentally, to account for the sensitivity of the proportional counter to X-rays, given by: $f_X = \varepsilon_{X,A} + (1-\varepsilon_{X,A})K_{Os}$, $B = aK_{Pt} + bf_x$

$$K_{Os} = I\left(\frac{\alpha\varepsilon_{ec} + \varepsilon_{\beta\gamma}}{1+\alpha}\right) \quad \text{and} \quad K_{Pt} = I_{Pt}\left(\frac{\alpha\varepsilon_{ec} + \varepsilon_{\beta\gamma}}{1+\alpha}\right)_{Pt}$$

.

A 4π PC–Ge(Li) system has been used to determine the f_X factor. This system consisted of a 4π proportional counter coupled to a 10% Ge(Li). Singles and coincidence measurements were made in a multichannel analyser. The selected γ-window of (484 + 489) keV was not affected by Compton events from the 316-keV γ-rays.

The singles and coincidence peak areas of spectra are named A_s and A_c, respectively. Their ratio corresponds to the efficiency of the proportional counter, given by:

$$A_c/A_s = \varepsilon_{X,A} + (1-\varepsilon_{X,A})I_{Os}\left(\frac{\alpha\varepsilon_{ec} + \varepsilon_{\beta\gamma}}{1+\alpha}\right)_{Os}$$

A linear least square fit results:

$$\frac{N_\beta N_\gamma}{N_c} = A + \left(\frac{1-\varepsilon_\beta}{\varepsilon_\beta}\right)B$$

where $A = N_0(a + bf_X)$.

The values of a and b were taken from the literature (Firestone and Shirley, 1996) and the factor f_x has been estimated by the 4πPC–Ge(Li) measurement. Gamma spectrometry measurements by means of the HPGe detector were also performed in order to check for impurities, and to detect possible systematic errors in the adopted methodology.

Table 1
Fitting parameters obtained at the two selected γ-windows, f_X factor and branching ratios

γ-Window	Parameter	Value
γ_1	A	$(256.8 \pm 1.1) \times 10^3$
	B	$(39.5 \pm 2.6) \times 10^3$
γ_2	A	$(258.8 \pm 1.0) \times 10^3$
	B	$(43.2 \pm 3.1) \times 10^3$
γ_1 and γ_2	f_X factor	0.412 ± 0.064
	(a) β-branching ratio	0.9524 ± 0.0004
	(b) EC-branching ratio	0.0476 ± 0.0004

4. Results and discussion

Fig. 2 shows the extrapolation curve obtained for the two selected γ-windows. In this curve the β-efficiency was varied in the range from 65 to 93% using external absorbers.

Table 1 shows the f_X factor and the parameters obtained by linear least square fitting using the code LINFIT (Dias, 1999), which incorporates covariance matrix methodology. Table 2 shows the values of a and b taken from the literature (Firestone and Shirley, 1996) and the resulting activity of the solution. The error in f_X factor is high (around 15%) and is due to poor statistics in the peak areas from Os γ-transitions obtained by the Ge(Li) detector. However, its contribution to the activity is very small because it is multiplied by b.

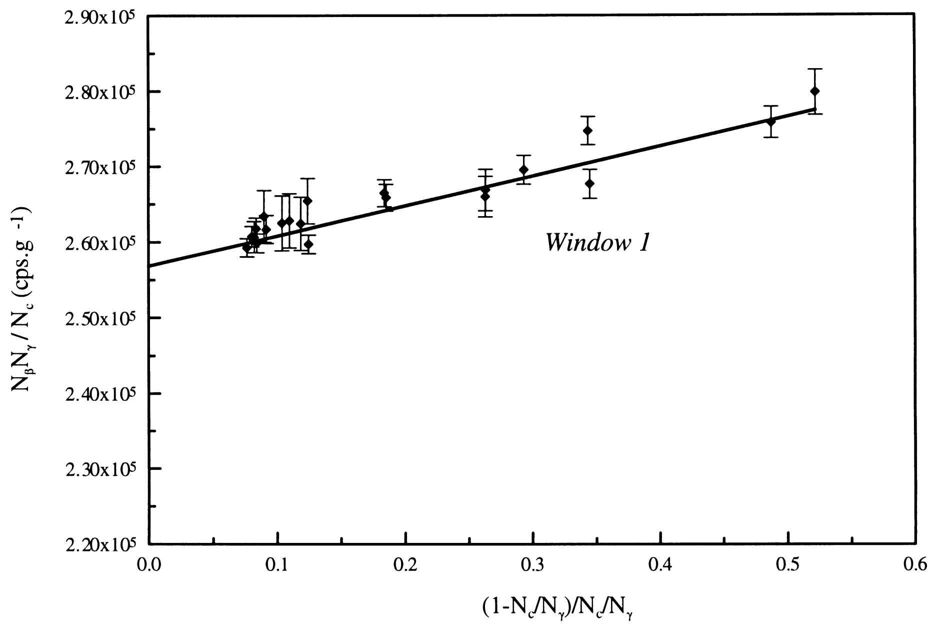
The final value obtained in the present measurement was (265.2 ± 1.1) kBq/g. This value is the average from the activities using the two γ-windows.

As can be seen, this result is in good agreement with

Table 2
Activity of ^{192}Ir solution

Reference	Condition	Activity (kBq/g^{-1})
Present work	γ_1-Window	264.2 ± 1.1
	γ_2-Window	266.2 ± 1.0
	Average	265.2 ± 1.1
IRDDM	4πβ–γ with NaI(Tl)	263.7 ± 1.1
BIPM (SIR)	4πγ (IC)[a]	265.1 ± 2.8

[a] Ionization chamber.



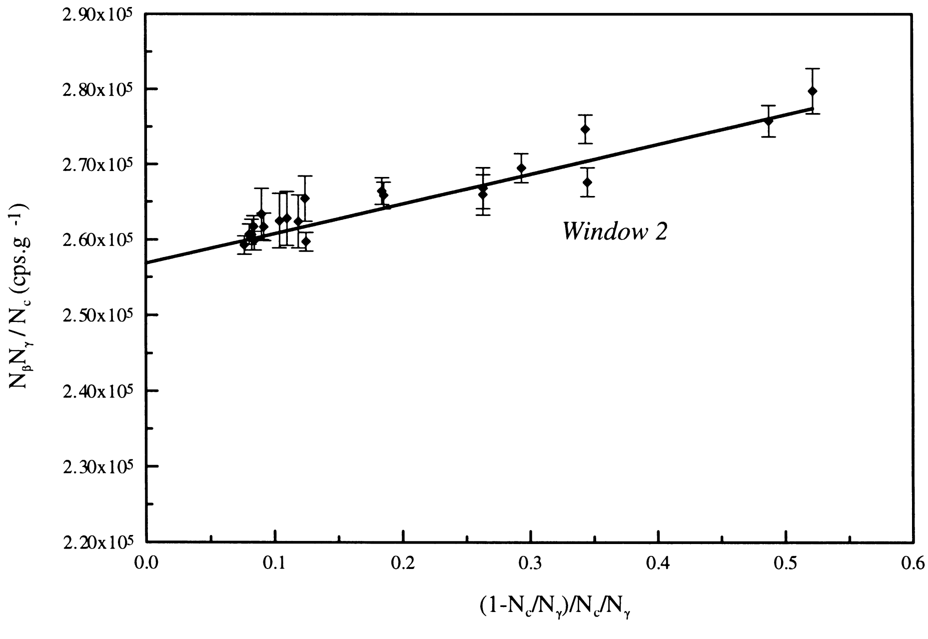


Fig. 2. Extrapolation curves of $N_\beta N_\gamma/N_c$ as a function of $(1 - N_c/N_\gamma)/(N_c/N_\gamma)$.

those obtained by BIPM (SIR) and IRDDM (Iwahara et al., 1998)

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