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Measurement of the Gamma-ray Probability Per Decay of I-126

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The emission probabilities per decay of the main γ-ray transitions, as well as the $\beta^-/(\beta^+$, EC) branching ratio and half-life of ^{126}I have been measured. β branch measurements were carried out in a 4π(PC)β–γ coincidence system consisting of a proportional counter coupled to a pair of 3 × 3 inch NaI(Tl) crystals. The electron capture branch was measured in an X–γ coincidence system using two 3 × 3 inch NaI(Tl) crystals. Gamma-ray measurements were performed in a calibrated HPGe spectrometry system. All the uncertainties were treated rigorously by means of covariance analysis. © 1998 Elsevier Science Ltd. All rights reserved

Introduction

^{126}I is important in fast neutron metrology as a product of the ^{127}I(n, 2n)^{126}I reaction (Santry, 1979), as well as in nuclear medicine where this radionuclide appears as an impurity in the reactor production of ^{125}I (Lagoutine *et al.*, 1982). Therefore, the accurate determination of the γ-ray emission probabilities per decay of ^{126}I is of interest.

The decay scheme of ^{126}I is shown in Fig. 1 (Zijp and Baard, 1979). This radionuclide undergoes 52.7% electron capture and β^+ decay and 47.3% β^- decay; both branches are followed by γ-ray emission. The complexity of the decay scheme necessitates the use of two separate coincidence systems. The β^- branch measurement (including a small contribution from β^+) has been carried out in a 4π(PC)β–γ coincidence system consisting of a proportional counter coupled by a pair of 3 × 3 inch NaI(Tl) crystals, while the electron capture branch was measured by an X–γ coincidence system using two 3 × 3 inch NaI(Tl) crystals.

Standardization Methods

4πβ–γ coincidence method

A conventional $4\pi\beta$–γ coincidence technique was used (Moura, 1969; Fonseca, 1997), consisting of a 4π proportional counter coupled to a pair of NaI(Tl) crystals. Both the β^- and β^+ particles were detected by the proportional counter whereas the γ-rays were detected by the scintillators. Two discrimination windows were set for the scintillators: one to measure the photons emitted in the decay of the 388 keV excited state of ^{126}Xe (γ_7, Fig. 1), and another to measure the photons emitted in the decay of the 666 keV excited state of ^{126}Te (γ_3). This study was undertaken to determine the sensitivity of the proportional counter to X-rays and electrons.

The β branch disintegration rate is given by:

$$N_0(b+p) = \left[\frac{N_\beta N_\gamma}{N_c} - N_0(a-p)f_X\right] \times \left[1 + \left(\frac{N_\gamma}{N_c} - 1\right)B\right]^{-1} \quad (1)$$

where $N_0(b + p)$ is the disintegration rate corresponding to β^- and β^+ branches, a is the EC + β^+ emission probability, b is the β^- emission probability, p is the β^+ emission probability, N_β is the proportional counter counting rate, N_γ is the counting rate at 388 keV γ-window, N_c is the coincidence rate and f_X is an experimental correction to account for the sensitivity of the proportional counter to X-rays, as given by (Fonseca, 1997)

$$f_X = \varepsilon_X + k_X - \varepsilon_X k_X,$$

where

$$k_X = \left[\left(\frac{r_3\alpha_3\varepsilon_{ec} + \varepsilon_{\beta\gamma 3}}{1+\alpha_3}\right) + \left(\frac{r_5\alpha_5\varepsilon_{ec} + \varepsilon\beta\gamma_5}{1+\alpha_5}\right)\right]$$

and B is the contribution of non-coincident events to the proportional counter detection efficiency, given by:

$$B = bk_{b7} + 2p\varepsilon_{\beta\gamma a} + (a-p)f_X$$

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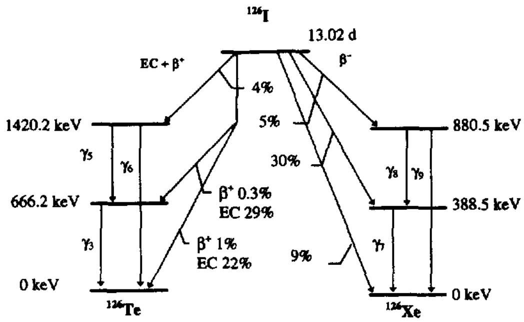

Fig. 1. Decay scheme of ^{126}I (Zijp and Baard, 1979).

where

$$k_{b7} = \left(\frac{r_7\alpha_7\varepsilon_{ec} + \varepsilon_{\beta\gamma7}}{1+\alpha_7}\right)$$

r_3, r_5 and r_7 are the branching ratios of transitions 3, 5 and 7 (see Fig. 1), ε_X is the proportional counter efficiency for X-rays, ε_{ce} is the proportional counter efficiency for conversion electrons and $\varepsilon_{\beta\gamma3}$, $\varepsilon_{\beta\gamma5}$, $\varepsilon_{\beta\gamma7}$ and $\varepsilon_{\beta\gamma a}$ are the proportional counter efficiency for γ-rays of 3, 5, 7 transitions and positron annihilation, respectively.

Subscripts in k_X and k_{b7} correspond to the X and β^- transitions, respectively, as shown in Fig. 1. 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). An additional correction factor due to Compton events under the 388-keV γ-ray window was applied by measuring a ^{137}Cs source in the same position as the ^{126}I source. The 661.6 keV γ-ray of ^{137}Cs simulates closely the 666-keV γ-ray from ^{126}I, and the correction was given as the ratio of the counts under the 388 and 666-keV γ-ray windows.

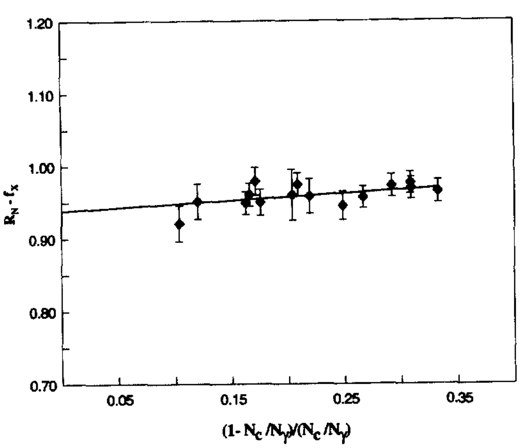

Fig. 2. Extrapolation curve of (R_N–f_X) as a function of the inefficiency factor (N_γ/N_c–1).

Table 2. Fitting parameters obtained from activity measurements of ^{126}I

Parameter	Value	Correlation matrix	
A	0.938 ± 0.017	1	
B	0.093 ± 0.039	−0.602	1

X–γ coincidence method

The X–γ coincidence method makes use of two scintillation counters. K X-rays following electron capture decay are detected by one of the scintillators, while the other scintillator detects the photons emitted from the decay of the 666 keV excited state of ^{126}Te.

The number of decay events for this branch is given by

$$N_0(a-p) = \frac{N_X N_\gamma}{N_c} \tag{2}$$

where $N_0(a\text{–}p)$ is the disintegration rate of the electron capture branch, a is the probability of electron capture events including β^+, p is the probability of β^+ emission, N_X is the K X-ray counting rate in one of the scintillators, N_γ is the counting rate in the 666 keV γ-window and N_c is the coincidence rate.

The observed counting rates N_β, N_γ and N_c were corrected as described in Section 2.1. A correction factor for the Compton events under the X-ray total absorption peak was measured by placing a 0.6 mm thick copper filter between the source and the NaI detector to remove the X-ray counts. The γ-ray attenuation in the filter has been taken into account. Summing of pulses arising from K X-rays in coincidence with γ-rays or between the γ-rays themselves can occur as a consequence of the small distance between the source and the NaI detectors (approximately 3 mm). This effect has been calculated by estimating the total and peak efficiencies for all γ-rays in cascade with the 666 keV γ-ray. The resulting correction was typically (10.4 ± 1.1)%.

Activity determination

Since $(a + b) = 1$, the activity N_0 could be obtained by summing (1) and (2). However, several parameters on the right hand side of these two equations depend on decay parameters. An alternative procedure has been developed by calculating

Table 1. Gamma-ray emission probabilities per decay of ^{126}I

Gamma-ray energy (keV) (Zijp and Baard, 1979)	Emission probability	
	this work	Tamura *et al.* (1982)
388.63	0.3556 ± 0.0048	0.341 ± 0.027
491.24	0.02882 ± 0.00040	0.0285 ± 0.0022
666.33	0.3290 ± 0.0040	0.331 ± 0.025
753.82	0.04148 ± 0.00052	0.0416 ± 0.0031
879.88	0.00743 ± 0.00013	0.00755 ± 0.00059
1420.19	0.003039 ± 0.000075	0.00295 ± 0.00022

Table 3. Decay parameters of ^{126}I

Parameter	Present work	Zijp and Baard, 1979	Tamura *et al.*, 1982	Anderson *et al.*, 1965	Ok and Kirschner, 1975	Miyano, 1993 (evaluation)
Total EC + β^+ (%)	52.68 ± 0.45		56.3 ± 2.0			
Total β^- (%)	47.32 ± 0.45		43.7 ± 2.0			
Total β^{+-} (%)	1.093 ± 0.016	3.34 ± 0.22				
Half-life (d)	12.82 ± 0.05	13.02 ± 0.07		12.8 ± 0.1	12.93 ± 0.06	13.11 ± 0.05

the following equation:

$$R_N - f_X = \left(\frac{N_\beta N_\gamma}{N_c} \bigg/ \frac{N_X N_\gamma}{N_c} \right) - f_X = A[1 + k_\varepsilon] \quad (3)$$

where $A = (b + p)/(a - p) = -[1 - (a - p)^{-1}]$ and $k_\varepsilon = B(N_\gamma/N_c - 1)$.

Parameters A and B were obtained by linear extrapolation and changing the efficiency parameter N_c/N_γ in the $4\pi\beta$–γ coincidence system at the 388 keV γ-window. The ratio given by (3) is independent of source activity, mass and irradiation time; therefore, sources from different batches could be grouped together in a single extrapolation. Values of A and $(a - p)$ were used to determine the activity (N_0) by means of (2).

Experimental Details

Sample preparation

The ^{126}I source was produced by means of the $^{127}I(n, 2n)^{126}I$ reaction in a $5 \times 10^{12}\ cm^{-2} \cdot s^{-1}$ fast neutron flux at the IEA-R1 research reactor. This sample was made of 100 mg KI powder sealed in a quartz ampoule and surrounded by a cadmium cover in order to avoid thermal neutron activation. The irradiation time was approximately 104 h and the sample was left to decay for about 10 days in order to minimize the activity from impurities (mainly ^{82}Br). The KI powder was dissolved in 6 ml of 0.005 N NaOH solution and the radioactive sources were prepared by depositing 10 to 30 mg of this ^{126}I solution on a 10–20 $\mu g/cm^2$ COLLODION substrate coated with a 10 $\mu g/cm^2$ gold layer. A 10 mg drop of $AgNO_3$ solution (160 mg/ml) was deposited on the source to prevent iodine volatilization and a drop of CYASTAT SN was deposited on the source to avoid crystal growth during the drying process in a desiccator. The uncertainty in the mass of the source was approximately 20 μg.

Coincidence measurements

A total of 15 sources from two irradiations have been prepared and measured by the two coincidence systems. The efficiency parameter N_c/N_γ in (3) has been changed by using external absorbers made of 40 $\mu g/cm^2$ thick COLLODION films and 0.93 mg/cm^2 aluminized MYLAR placed over and under the sources. As shown in Fig. 2, the efficiency parameter N_c/N_γ changed from 0.75 to 0.91. Each source was measured six to ten times for periods of 500 to 2000 s each. The overall statistical error for each series of measurements was in the range of 0.3 to 1.5%.

Gamma-ray spectrometry measurements

A flame-sealed ampoule was prepared from each ^{126}I solution; γ-ray measurements were made with a coaxial HPGe detector of 20% relative efficiency. This detector has been previously calibrated by means of standard sources of ^{60}Co, ^{133}Ba, ^{137}Cs and ^{152}Eu, supplied by the International Atomic Energy Agency. Further details on the calibration procedure can be found elsewhere (Geraldo and Smith, 1990; Fonseca, 1997). The distance from the source to the detector cap was 17.3 cm. Besides the ^{126}I γ-ray measurements, this spectrometer was also used to estimate the activities of the ^{86}Rb, ^{124}Sb and ^{127}Te impurities. These results were taken into account in the ^{126}I activity determination performed with the coincidence systems.

Results and Discussion

The parameters A and B from (3) were obtained by linear least square fitting by means of the code LSSOLVER (Tokai, 1987), which incorporates covariance matrix methodology. Details of the uncertainty analysis have been published in a previous paper (Fonseca *et al.*, 1996). The extrapolation curve for determining the activity is shown in Fig. 2, while the resulting γ-ray probability per decay for the main transitions of ^{126}I are listed in Table 1: these emission probabilities are consistent with the equivalent data of Tamura *et al.* (1982), but with a much lower uncertainty. Parameters A and B are listed in Table 2, and the branching ratios and half-life are given in Table 3. The values obtained for the parameters a (EC + β^+ branching ratio), b (β^- branching ratio) and p (β^+ branching ratio) are not in good agreement with other values in the literature, suggesting that these data need to be revised. A half-life of 12.82 ± 0.05 d was measured and agrees with the values obtained by Anderson *et al.* (1965) and Ok and Kirschner (1975), but is somewhat lower when compared with the other results shown in Table 3.

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