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Covariance methodology applied to uncertainties in I-126 disintegration rate measurements

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Abstract

The covariance methodology applied to uncertainties in ^{126}I disintegration rate measurements is described. Two different coincidence systems were used due to the complex decay scheme of this radionuclide. The parameters involved in the determination of the disintegration rate in each experimental system present correlated components. In this case, the conventional statistical methods to determine the uncertainties (law of propagation) result in wrong values for the final uncertainty. Therefore, use of the methodology of the covariance matrix is necessary. The data from both systems were combined taking into account all possible correlations between the partial uncertainties.

1. Introduction

The radionuclide ^{126}I is important in fast neutron metrology as a product of the ^{127}I(n, 2n)^{126}I reaction [1], as well as in nuclear medicine where it appears as an impurity in the reactor production of ^{125}I [2]. Therefore, its standardization is of interest. However, due to complex decay characteristics, the absolute measurement of activity requires two detection systems, yielding two set of correlated parameters. The calculation of uncertainties must take these correlations into account.

The decay scheme of ^{126}I [3,4] is shown in Fig. 1. This radionuclide decays 52.7% by electron capture and β^+, and 47.3% by β^- emission; both branches are followed by γ-ray emission. Due to its complex decay scheme, the absolute measurement of activity requires two separate coincidence systems.

The β^- (including a small contribution from β^+) branch measurement has been carried out in a 4π(PC)β–γ coincidence system consisting of a proportional counter, coupled to a pair of 3 in. × 3 in. NaI(Tl) crystals. The electron capture branch was measured in a X–γ coincidence system using two 3 in. × 3 in. NaI(Tl) crystals.

The data from these two systems are correlated and the corresponding uncertainties cannot be combined by any simple propagation law. The present work describes the covariance methodology applied to uncertainties in disintegration rate measurements for this radionuclide.

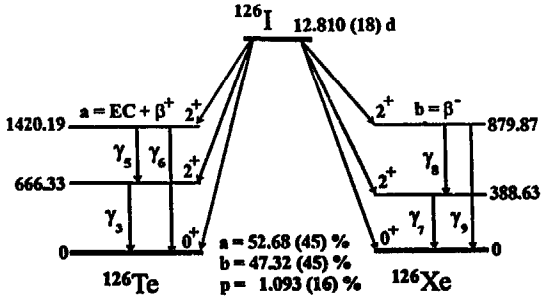

Fig. 1. Decay scheme of ^{126}I [4]. The values of a, b and p and the half-life were taken from Ref. [3].

2. Standardization methods

2.1. X–γ coincidence method

This method makes use of two scintillation counters. The K X-rays following electron capture events are detected by one of the scintillators whereas the other scintillator detects the photons emitted from the decay of the 666 keV excited state of ^{126}Te. Corrections are applied to take into account Compton events under the total absorption peak and possible summing of pulses arising from K X-rays in coincidence with γ-rays.

The number of decay events for this branch is given by:

$$N_0(a-p)=\frac{N_X N_\gamma}{N_c}, \qquad (1)$$

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where $N_0(a-p)$ is the disintegration rate of the electron capture branch, a is the probability of electron capture events plus 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_X and N_γ were corrected for background, dead time and decay in the usual way. A correction factor due to summing the Compton effects [3] was also considered. The coincidence rate N_c was corrected for dead time and accidental coincidences using the Cox–Isham formalism [5].

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

This is the conventional coincidence method using a $4\pi\beta-\gamma$ system [6], consisting of a 4π proportional counter coupled to a pair of NaI(Tl) crystals.

The β^- and β^+ particles were detected in the proportional counter. Two discrimination windows were set for the scintillators: one to measure the photons (γ_7, Fig. 1) emitted in the decay of the 388 keV excited state of ^{126}Xe and another one to measure the photons (γ_3) emitted in the decay of the 666 keV excited state of ^{126}Te. This was done to account for the proportional counter sensitivity to X-rays and electrons.

The β branch disintegration rate is given by:

$$N_0(b+p)=\left[\frac{N_\beta N_\gamma}{N_c}-N_0(a-p)f_X\right](1+k_e)^{-1}, \qquad (2)$$

where $N_0(b+p)$ is the disintegration rate corresponding to β^- and β^+ branches, 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, f_X is an experimental correction to account for the sensitivity of the proportional counter to X-rays [3], $1+k_e$ is a measured decay scheme correction [3].

The observed counting rates N_β, N_γ and N_c were corrected as described in the previous section.

2.3. Activity determination

Since $(a+b)=1$, in principle the activity N_0 could be obtained by summing Eqs. (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 the following equation:

$$R_N-f_X=\left(\frac{N_\beta N_\gamma}{N_c}\Big/\frac{N_X N_\gamma}{N_c}\right)-f_X$$
$$=A\left[1+B\left(\frac{N_\gamma}{N_c}-1\right)\right]. \qquad (3)$$

where

$$A=(b+p)/(a-p)=-[1-(a-p)^{-1}],$$

$$B\left(\frac{N_\gamma}{N_c}-1\right)=k_e \text{ and}$$

B is the contribution of non-coincident events to the proportional counter detection efficiency.

The parameters A and B were obtained by linear extrapolation, changing the efficiency parameter N_c/N_γ in the $4\pi\beta-\gamma$ coincidence system at the 388 keV γ-window.

The parameter given by Eq. (3) is independent of source activity, mass and irradiation time, therefore sources from different batches could be grouped together in a single extrapolation. From the values of A and $(a-p)$, the activity (N_0) has been obtained by means of Eq. (1).

3. Covariance methodology

The covariance matrix is a more complete form of uncertainty representation than the older statistic methods because besides the total uncertainty, it gives information about the existing level of correlation among the partial uncertainties. The method for deriving a covariance matrix (V_i) for experimental data has been discussed in detail by many authors (see for instance Refs. [7,8]).

The elements of this matrix can be given by:

$$V_{ij}=\sum_{l=1}^{L} S_{ijl}e_{il}e_{jl}, \qquad (4)$$

where e_{il} and e_{jl} are the partial uncertainties of the parameters, and S_{ijl} are the corresponding correlation coefficients.

In order to obtain the final uncertainty in the activity, several steps have been followed:

(1) A covariance matrix involving all relative partial uncertainties in the ratio given by Eq. (3) has been obtained making use of Eq. (4), by means of the code CALCOV [7].

(2) A covariance matrix involving the absolute partial uncertainties in f_X was calculated.

(3) Both matrices were combined by means of code CALCOV [7].

(4) A covariance matrix for the linear square coefficients A and B has been obtained by means of code LSSOLVER [9].

The final uncertainty in the activity has been estimated by:

$$\sigma^2_{N_0}=\sigma^2_A+\sigma^2_X+2\rho\sigma_A\sigma_X. \qquad (5)$$

where σ_A is the uncertainty in parameter A (extrapolated value of R_N-f_X). σ_X is the uncertainty in the right side of Eq. (1), and ρ is the correlation coefficient between both parameters.

Uncertainties are quoted at the 67% confidence level

Table 1
Partial uncertainties in the parameter $R_N - f_X$ (%)

Sample	4πβ–γ	X–γ	f_X [a] $\times 10^{-3}$	Resolution time	Summing correction	Compton correction X–γ	Compton correction 4πβ–γ
1	1.38	1.48	1.65	1.10	1.10	0.17	0.61
2	0.84	1.69	1.59	1.10	1.10	0.17	0.61
3	0.54	0.61	0.95	1.10	1.10	0.17	0.61
4	0.49	0.63	0.80	1.10	1.10	0.17	0.61
5	0.75	0.85	0.98	1.10	1.10	0.17	0.61
6	0.40	0.66	0.86	1.10	1.10	0.17	0.61
7	0.91	1.69	0.88	1.10	1.10	0.17	0.61
8	0.63	1.09	0.71	1.10	1.10	0.17	0.61
9	1.58	2.63	2.51	1.10	1.10	0.17	0.61
10	1.31	0.61	0.57	1.10	1.10	0.17	0.61
11	0.45	0.45	1.08	1.10	1.10	0.17	0.61
12	0.34	0.62	0.57	1.10	1.10	0.17	0.61
13	0.56	0.45	0.41	1.10	1.10	0.17	0.61
14	0.60	0.48	0.72	1.10	1.10	0.17	0.61
15	0.61	0.56	0.53	1.10	1.10	0.17	0.61
Correlation factor	0	0	0	1	1	1	1

[a] Absolute.

(1σ). The value of ρ has been calculated by the following equation:

$$\rho = \frac{\mathrm{cov}(R_N, X)}{\sigma_{R_X}\sigma_X}, \qquad (6)$$

where

$$\mathrm{cov}(R_N, X) = E\left(\frac{N_\beta N_\gamma}{N_C}\right) - E\left(\frac{N_X N_\gamma}{N_C}\right)E(R_N),$$

E corresponds to expected values (averaged) for each irradiation, and σ_{R_N}, σ_X are the standard deviations in $N_\beta N_\gamma / N_C$ and $N_X N_\gamma / N_C$, respectively.

4. Results and discussion

Two irradiations have been performed in order to obtain ^{126}I by means of the ^{127}I(n, 2n)^{126}I reaction. A total of 15

Table 2
Total uncertainties and correlation matrix for parameter $R_N - f_X$

Sample	Total absolute uncertainty ($\times 10^{-2}$)	Correlation matrix														
1	1.94	100														
2	1.48	59	100													
3	1.50	58	78	100												
4	1.50	57	77	76	100											
5	1.53	56	76	75	74	100										
6	1.56	55	75	73	73	71	100									
7	2.47	35	47	46	46	45	44	100								
8	2.41	37	50	48	48	47	46	29	100							
9	1.59	55	75	73	73	72	70	45	47	100						
10	1.57	56	75	74	73	72	71	45	47	71	100					
11	1.78	49	67	65	65	64	62	40	41	62	63	100				
12	1.57	56	76	74	74	73	71	45	47	71	72	63	100			
13	2.38	36	49	48	48	47	46	29	30	46	46	41	47	100		
14	1.91	47	63	62	61	60	59	38	39	59	60	53	60	39	100	
15	3.39	25	34	33	33	33	32	20	21	32	32	28	32	21	27	100

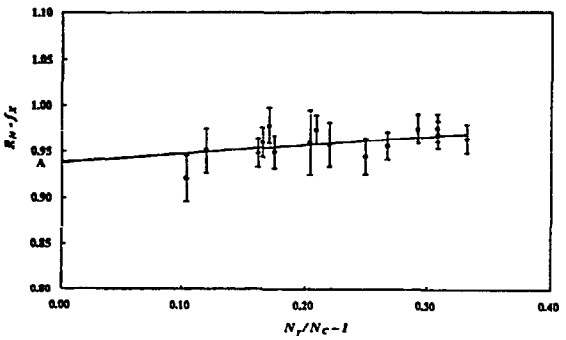

Fig. 2. Extrapolation curve between $(R_N - f_X)$ as a function of the inefficiency factor $(N_T/N_c - 1)$.

sources have been standardized. A list of all partial uncertainties involved in Eqs. (2) and (3) are listed in Table 1. The correlation coefficients are shown in the bottom of the table. The first two columns give the total uncorrelated uncertainties calculated from the measurements using the two coincidence systems, namely: $4\pi\beta-\gamma$ and X–γ. These uncertainties were obtained by simple propagation of all partial uncertainties involved. The third column gives the uncertainty in f_X. The main contribution to this uncertainty was in the 4π detection efficiency for X-rays. All other uncertainties shown in Table 1 are systematic and were assumed to be totally correlated. Table 2 shows the total uncertainties and the corresponding correlation matrix calculated by code CALCOV [7]. These values were used as input data for the linear fit presented in Fig. 2. This fit has been performed by code LSSOLVER [9] taking into account the input correlation matrix.

Fig. 2 shows the extrapolation curve applying Eq. (3). The final uncertainty in the activity for each sample was obtained from the data given in Tables 1 and 2, by means of Eq. (5).

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