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Accidental Summing Correction in ^{125}I Activity Determination by the Sum-Peak Method

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The correction for accidental summing (pile-up) in the activity determination of ^{125}I by the Sum-Peak method is described. The results are compared with the X-(X, γ) coincidence method. Theoretical estimates of this correction have been performed by the Monte-Carlo method and predictions were made for other detector efficiencies and electronic shaping times.

Introduction

The Sum-Peak method has been widely used for activity determination of ^{125}I due to its simple experimental procedure and data analysis (Eldridge and Crowther, 1964). However, the results are critically dependent on the counting rate due to accidental summing (pile-up), therefore this effect must be taken into account. Different methods to solve this problem are described in the literature (Taylor, 1967; Martin and Taylor, 1992; Zajic, 1986). In the present paper a simple approach is shown and theoretical predictions by the Monte-Carlo method are given for other experimental conditions.

The decay scheme of ^{125}I is shown in Fig. 1. This radionuclide decays 100% by electron capture, followed by 35 keV γ-ray emission. The K x-rays (27.4 and 31 keV) following electron capture events are coincident with photons emitted in the decay of the 35 keV excited state of ^{125}Te. These photons are either the K x-rays following the internal conversion or the unconverted γ-ray itself. The 35 keV γ-ray transition has a half-life of 1.47 ns and a high internal conversion probability (93.4%). The adopted half-life for ^{125}I was (59.4 ± 0.5) days (Ratel, 1989).

Usually, the Sum-Peak method makes use of a single scintillation counter. In the case of ^{125}I two peaks appear in the pulse height spectrum, as shown in Fig. 2. The first peak with lower pulse height and higher intensity corresponds to pulses arising from K x-rays events overlapping the 35 keV γ-rays, due to the poor resolution of the scintillator. The second peak is produced by the sum of pulses arising from capture K x-rays in coincidence with conversion K x-rays or γ-rays from the 35 keV transition.

The experimental results given in the present paper were obtained during an internal comparison of this radionuclide sponsored by the Bureau International des Poids and Mesures (Ratel, 1989). These results were compared with the $X-(X, \gamma)$ coincidence method, which is not sensitive to changes due to accidental summing. Details of the latter method are not discussed here and can be obtained elsewhere (Taylor, 1967; Dias and Koskinas, 1988).

Correction Method

In the Sum-Peak method, the activity is given by (Eldridge and Crowther, 1964):

$$N_0 = \frac{P_1 P_2}{(P_1 + P_2)^2} \frac{(A_1 + 2A_2)^2}{A_2}, \qquad (1)$$

where A_1 and A_2 are the counting rates corresponding to the first and Sum peaks, respectively, corrected for background and dead time; $P_1 = P_K W_K$ is the x-ray emission probability for electron capture events; and $P_2 = (1 + \alpha_K W_K)/(1 + \alpha_T)$ is the K x-ray emission probability for internal conversion events plus the 35 keV γ-ray emission probability.

It is expected that accidental summing of pulses produced by x-rays or 35 keV γ-rays can change the values of A_1 and A_2 in equation (1). The change in A_1 is usually negligible, but the corresponding change in A_2 may be significant.

The expected change in A_2 is approximately given by:

$$A_2 = A_2^0 + 2\tau_r A_1^2 \quad (2)$$

where τ_r is the effective resolving time of the pulses contributing to the accidental summing; and A_2^0 is the A_2 value extrapolated to zero counting rate. Therefore,

$$\frac{A_2}{A_1} = \frac{A_2^0}{A_1} + 2\tau_r A_1$$

or

$$\frac{A_2}{A_1} = a + bA_1, \quad (3)$$

where a and b can be obtained by linear least square fit.

From equations (1) and (3) the corrected activity is approximately given by:

$$N_0' = N_0\left(1 + \frac{B}{A}A_1\right)^{-1}, \quad (4)$$

where N_0 is the observed activity, given by equation (1).

The parameters A and B are given by:

$$A = \frac{1}{a} + 4a + 4$$

and

$$B = -b\left(\frac{1}{a^2} - 4\right).$$

Experimental

Sample preparation

The radioactive sources were prepared by dropping known aliquots of ^{125}I solution on 30 μg/cm^2 thick COLLODION substrate. The picnometer technique was used (Campion, 1975) and the aliquot masses were determined by means of a METTLER M5SA microbalance. The estimated mass uncertainty was $\pm 15\,\mu$g. To avoid volatility of radioactive material, an aliquot (≈ 10 mg) of aqueous solution containing

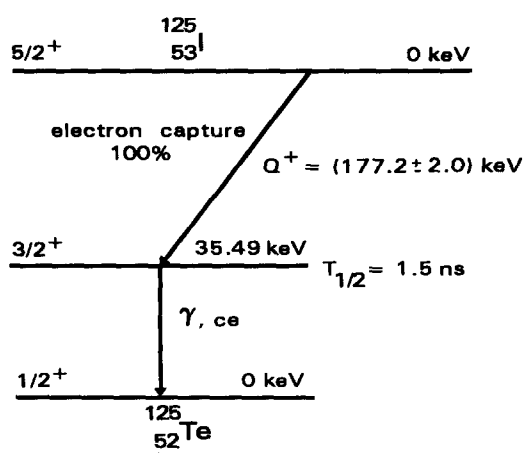

Fig. 1. Decay scheme of ^{125}I (Coursol, 1982).

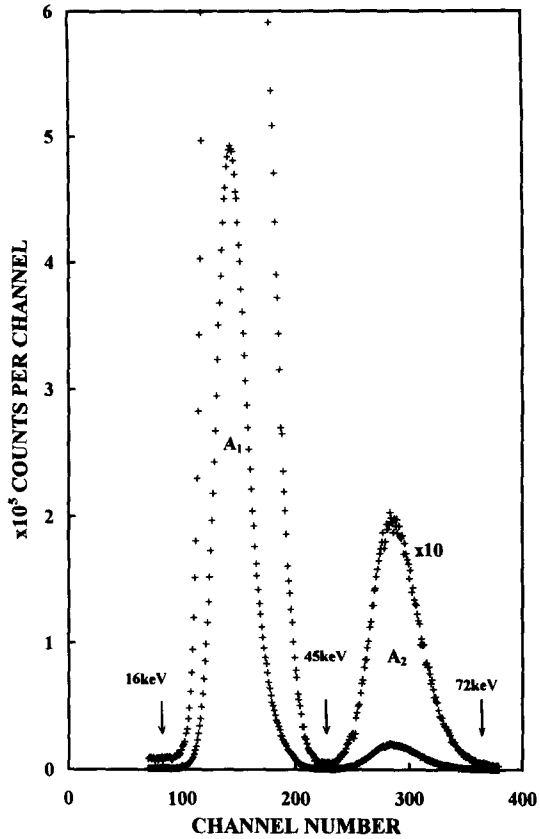

Fig. 2. Typical pulse height spectrum of ^{125}I obtained using a single 3″ × 3″ NaI(Tl) crystal.

160 μg of $AgNO_3$ per gramme of solution was dropped on each ^{125}I source before drying in a desiccator atmosphere. The number of sources was 25 in the mass range from 8 to 50 mg.

Counting equipment

The counting rates obtained in activity measurements by the two methods, namely: Sum-Peak and $X-(X,\gamma)$ coincidence, were measured simultaneously by means of the electronic set-up described in Dias and Koskinas (1988). Two 3″ × 3″ NaI(Tl) scintillation detectors, BICRON Mod. 3M3, were used. The measured resolution FWHM (full width at half maximum) was 22.2% for detector 1 and 24.8% for detector 2, at 28 keV photon energy corresponding to ^{125}I K x-rays (see Fig. 2). The pulse height spectrum windows were 16–45 keV and 45–72 keV for peaks A_1 and A_2, respectively.

The set-up allowed variations in the source-detector distance symmetrically with respect to the source. For most measurements, this distance was fixed around 10 mm.

Theoretical Calculations

A Monte-Carlo code has been developed in order to verify the above assumptions and also for predicting the correction at other experimental conditions.

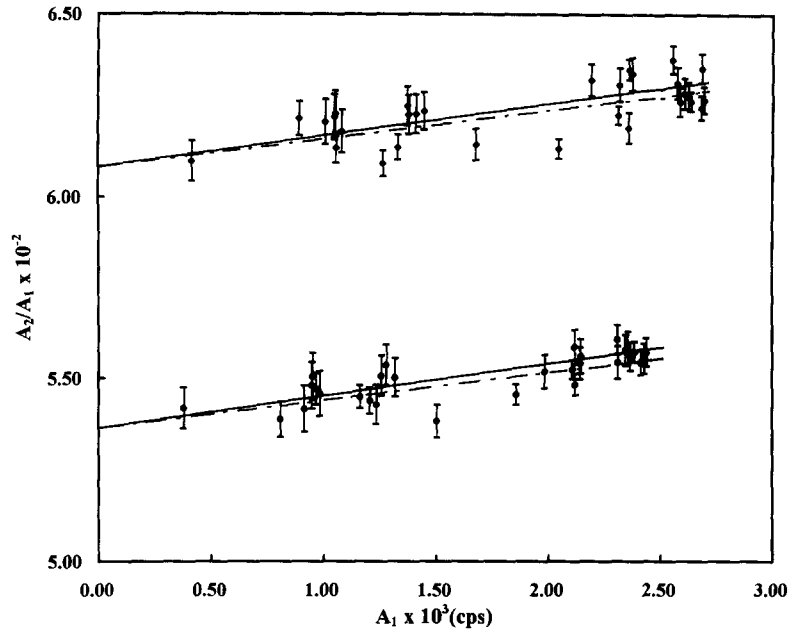

Fig. 3. Values of A_2/A_1 as a function of A_1. The upper set of points were obtained with detector 1 and the lower set with detector 2. The solid lines are least square fits to experimental data. The dashed lines are Monte-Carlo calculations.

In this simulation the pulse shape has been assumed to be Gaussian. The same approximation has been assumed for the pulse height distribution of peak A_1 shown in Fig. 2.

A Gaussian random variable was generated by the following equation (Kalos and Whitlock, 1986):

$$y = \left(\sum_{k=1}^{12} \xi_k - 6 \right), \tag{5}$$

where ξ_k is a uniform random variable.

The distribution of time intervals was assumed to be a random variable given by (Kalos and Whitlock, 1986):

$$t = -\frac{1}{R} \log(1 - \xi) \tag{6}$$

where R is the pulse rate.

The pulse width and pulse rate were input parameters corresponding to different experimental conditions. The pulse FWHM was measured experimentally for different shaping times and used as input data for the code.

Results and Discussion

The experimental results for A_2/A_1 as a function of A_1 are shown in Fig. 3. The main parameters are given in Table 1. The difference between extrapolated A_2^0/A_1 values at the two curves is due to a slight difference in the source–detector distance for each crystal. The two slopes are the same within the experimental uncertainties. This is expected since they are related to effective resolving times which are similar because the electronic conditions were kept the same for both channels. The value of B/A indicates that the accidental summing correction is about 1.2–1.4% for 1000 cps and increases proportionally to the counting rate. The Monte-Carlo calculation is given by the dashed curve and agrees well with

Table 1. Fitting parameters obtained in the extrapolation curve for the Sum-Peak method

Parameters	Detector 1	Detector 2
A_2^0/A_1	$(6.083 \pm 0.026) \times 10^{-2}$	$(5.362 \pm 0.019) \times 10^{-2}$
τ_r	$(0.427 \pm 0.068)\ \mu s$	$(0.444 \pm 0.056)\ \mu s$
B/A	$(-1.10 \pm 0.18) \times 10^{-5}$	$(-1.34 \pm 0.17) \times 10^{-5}$

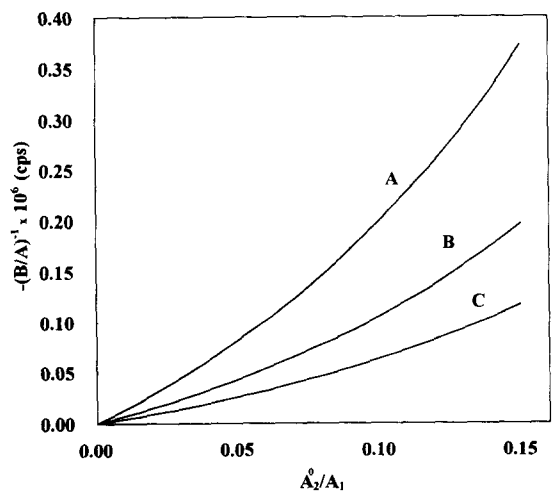

Fig. 4. Behavior of the inverse of B/A as a function of A_2^0/A_1 for different shaping times obtained by Monte-Carlo. Curves A, B and C correspond to 0.5, 1.0 and 2.0 μs shaping times, respectively.

Table 2. Comparison between activity concentrations obtained by different methods (MBq/g)

Method	Master solution	Dilution 1	Dilution 2
Sum-Peak	1.4364 (32)	1.4235 (31)	1.4350 (37)
Coincidence	1.4362 (30)	1.4276 (24)	1.4386 (32)
Ratio	1.0001 (30)	0.9971 (28)	0.9975 (34)

the experimental values. This calculation has been normalized to the extrapolated experimental values, therefore only slopes can be compared.

Figure 4 shows the theoretical behavior of the inverse of B/A as a function of A_2^0/A_1 for different shaping times, as calculated by Monte-Carlo. From this figure one can estimate the accidental summing correction for other crystal sizes, source–detector distances and electronic shaping times. The value of A_2^0/A_1 can be taken from the uncorrected ratio between the two peaks. The value of B/A is then estimated by interpolating at the corresponding shaping time curve.

The value of B/A has little dependance on the peak resolution. Monte-Carlo calculations have been performed varying the peak FWHM from 10 to 40%. The resulting change in B/A was around 10% for 2.0 μs and lower for other shaping times.

Table 2 shows the activity results obtained by the Sum-Peak method together with those obtained by the $X-(X,\gamma)$ coincidence method, for the master ^{125}I solution and two diluted solutions. The number inside the parenthesis is the uncorrelated uncertainty in the last digits. The uncertainty in the ratio has been obtained by propagation. As can be seen, the corrected Sum-Peak value agrees well with the other method, showing that the proposed correction can be considered adequate.

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