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Direct activity determination of ^{54}Mn and ^{65}Zn by a non-extrapolation liquid scintillation method

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Abstract

The measurement of ^{54}Mn and ^{65}Zn by liquid scintillation coincidence counting results in low detection efficiencies. The activity obtained from the extrapolation of efficiency data can therefore become problematic if curvature is present. The simple decay scheme exhibited by these radionuclides, with the emission of an energetic gamma ray, allows the absolute activity to be determined from 4πe–γ data by direct calculation without the need for efficiency extrapolation. The method, which relies on determining the probability of the γ-ray interacting with the scintillator solution, is described and validated by measurements made on ^{60}Co.

Keywords: ^{54}Mn; ^{65}Zn; Liquid scintillation coincidence counting; Gamma-ray interaction probability

1. Introduction

The measurement of electron-capture radionuclides by liquid scintillation (LS) counting generally results in low detection efficiencies. For example, the maximum efficiency obtained with a double-phototube coincidence system at the CSIR-NML is about 40% for ^{54}Mn and 60% for ^{65}Zn. The activity obtained from the extrapolation of efficiency data collected by 4π(LS)e–γ coincidence counting can therefore become problematic if curvature is present.

This paper presents an alternative method for those radionuclides exhibiting a simple decay scheme with the emission of an energetic γ-ray. The absolute activity is extracted from the 4πe–γ data by direct calculation based on formulae derived from a detection efficiency analysis. It is only necessary to additionally determine the probability of the γ-rays interacting with the scintillator solution.

The method has been applied to the activity measurement of ^{54}Mn and ^{65}Zn. Besides providing a useful supplement to conventional efficiency extrapolation analysis, the method being reported offers the advantage of experimental simplicity and considerable reduction in data collection since there is no need for efficiency variation.

2. Measurement technique

The method is explained with respect to the detection efficiency analysis for a ^{54}Mn counting source viewed by two phototubes in coincidence, and a single NaI(Tl) crystal detecting the γ-rays. The 4π count rate from the liquid scintillation detector is

$$B = N\varepsilon + N(1-\varepsilon)P_{\mathrm{I}}, \tag{1}$$

where N is the source activity, ε is the double-tube detection efficiency for the electron-capture emissions and P_{I} is the probability that the γ-rays interact with the scintillator solution. The interacting gammas form sum pulses that are detected with essentially 100% efficiency.

The gamma-ray count rate given by the NaI detector is

$$G = N(1-P_{\mathrm{I}})\varepsilon_{\gamma}, \tag{2}$$

where ε_{γ} is the efficiency for detecting events in the full-energy peak.

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The coincidence count rate is thus

$$C = N(1 - P_I)\varepsilon\varepsilon_\gamma. \quad (3)$$

The method takes advantage of the fact that the value of P_I can be determined by Monte Carlo calculation and so the activity can be found directly from the expression given by (1),

$$N = \frac{B}{\varepsilon + (1 - \varepsilon)P_I}, \quad (4)$$

where $\varepsilon = C/G$ is found by measurement.

For ^{65}Zn there is a small positron branch in addition to two electron-capture branches. In this case, the activity is given by

$$N = \frac{B}{(f_1 + f_2)\varepsilon + f_3 + f_1(1 - \varepsilon)P_I}, \quad (5)$$

where f_1 is the electron-capture branching ratio to the $5/2^-$ excited state of ^{65}Cu, f_2 the branching ratio to the ground state and f_3 the intensity of the positron branch. It is assumed that the efficiencies corresponding to the electron-capture branches are equivalent and that the LS counter detects the positrons with 100% efficiency due to the β^+ spectral shape.

3. Determination of the gamma-ray interaction probability

The value of P_I is dependent on the source geometry and the scintillator composition, necessitating a Monte Carlo calculation for its determination. This information has been determined for the custom made cylindrically-based counting vials used with the counting system (Simpson, 1994), covering the photon energy range of interest. Fig. 1 shows the results for one of the commercial scintillation cocktails used, namely Quicksafe A from ZINSSER ANALYTIC. The values differed slightly for the other scintillators (Insta-Gel from Packard and HP Ready-Solv from Beckman) due to the differences in composition and density. The mass fractions of the hydrogen, carbon, oxygen and nitrogen constituents necessary for the calculations were obtained from elemental analyses by gas chromatography. The photon mass attenuation coefficients for these elements were taken from Hubbell (1982) and combined according to the mass fractions of each cocktail. The specific interaction probability values required for analyzing the activity measurements made are given in Table 1.

4. Experimental

Counting sources were prepared in custom made flat-faced cylindrical counting vials, each being viewed in turn by two RCA 8850 phototubes coupled in coincidence. The escaping gamma-rays were detected with a 75×75 mm NaI(Tl) crystal and a window was set to record only the *upper* portion of the photopeak events so that C/G would better represent the efficiency in the 4π channel by minimizing the γ–γ coincidences from cross scattering. The counting system described in Simpson and Meyer (1988) was used to simultaneously collect 15 datum points of varying efficiency for each set. The recorded counts were corrected for background, after-pulsing and rate dependent effects using a deadtime of 1 μs and coincidence resolving time of 0.47 μs. For the investigation of the non-extrapolation method, the counting data corresponding to the lowest 4π threshold were used to extract the activity.

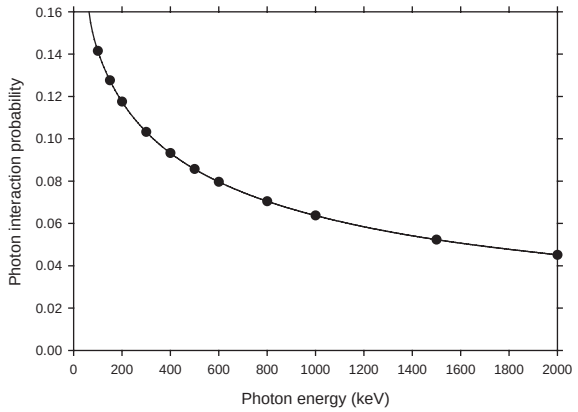

Fig. 1. The simulated average photon interaction probability determined at the indicated energies (closed circles) for the flat-faced cylindrical counting vial containing 12 ml of Quicksafe A liquid scintillator. The solid line is a regression fit to the data (fifth-order polynomial with inverse terms) used for interpolation.

5. Activity measurements

5.1. Validation of the method with ^{60}Co

^{60}Co was selected to demonstrate the direct analysis technique because it not only fits the requirement of comprising a simple decay scheme with an energetic gamma ray, but also has a high detection efficiency. A minor complication is a second coincident gamma ray. In this case

$$B = N\varepsilon_\beta + N(1 - \varepsilon_\beta)(P_1 + P_2 - P_1P_2), \quad (6)$$

where ε_β is the double-tube detection efficiency for the beta particles and P_1 and P_2 are the interaction probabilities for the γ_1 (1173 keV) and γ_2 (1332 keV) gamma rays, respectively.

If the higher energy gamma-ray photopeak is selected in the gamma-channel, the NaI detector count rate is

$$G = N(1 - P_2)\varepsilon_\gamma \quad (7)$$

and the coincidence rate is

$$C = N(1 - P_2)\varepsilon_\gamma\{\varepsilon_\beta + (1 - \varepsilon_\beta)(P_1 - P_1P_2)\}. \quad (8)$$

The extra term is due to γ_1-γ_2 coincidences. Solving gives the activity as

$$N = \frac{B}{C/G + (1 - C/G)P_I}, \quad (9)$$

where $P_I = \left(P_2/(1 - P_1 + P_1P_2)\right)$. The numerical value for P_I was 0.0588, obtained from the interaction probabilities shown in Table 1.

Measurements were made with a previously standardized ^{60}Co solution, a sample of which had been sent for comparison in 2002 to the International Reference System (SIR), which is maintained by the International Bureau of Weights and Measures (BIPM), (Rytz, 1983). The standardization was based on efficiency extrapolation of the LS coincidence counting data, giving the solution activity concentration as $121.27 \pm 0.12\,\mathrm{kBq\,g^{-1}}$ on the chosen reference date. This translates to an SIR equivalent activity of 7075 ± 9 kBq, which is 0.14% higher than the key comparison reference value of 7065 ± 4 kBq. Measurements for the non-extrapolation method were made on the same coincidence apparatus using the same counting sources (weighed aliquots mixed in 12 ml Quicksafe A). The lowest beta-particle threshold was again set above the single electron peak to minimize intruding afterpulses. The only change to the electronic settings was that of the γ-ray window/gate, which was set to select only those events falling in the upper portion of the 1.332 MeV photopeak. Table 2 shows the results obtained from some of the sources, measured with the gamma-rays counted integrally above the gate setting. The solution activity concentration obtained from eight sources was $121.12\,\mathrm{kBq\,g^{-1}}$ with a statistical uncertainty of $0.05\,\mathrm{kBq\,g^{-1}}$. The result with a tight γ-ray window was $121.19 \pm 0.08\,\mathrm{kBq\,g^{-1}}$. The selection of the beta-particle threshold was not particularly critical, results being about 0.1% lower for $C/G = 0.80$. The uncertainty component in the activity as propagated through the uncertainty in P_I is of the order of 0.012% (1σ).

5.2. ^{54}Mn

Sources of ^{54}Mn were prepared for various purposes over the years. Those data sets fulfilling the counting requirements were utilized to demonstrate the non-extrapolation method. In all cases the 4π threshold was set to just below the single electron peak to maximize the counting efficiency. The results of the data analysis of source measurements covering a broad range of count rates are shown in Table 3. Whereas the uncertainty from the fit in the extrapolation method is estimated to be 0.30%, the uncertainty component in the direct analysis method due to P_I is between 0.15% and 0.20% depending on the efficiency obtained.

Table 1
The relevant γ-ray interaction probabilities interpolated from fits to the Monte Carlo calculations

Radionuclide	γ-ray energy (keV)	Scintillator	Volume (ml)	Interaction probability
^{54}Mn	834.84	Insta-Gel	12.0	0.0668 ± 0.001
^{54}Mn	834.84	Insta-Gel	20.5	0.0808 ± 0.001
^{54}Mn	834.84	HP Ready-Solv	12.0	0.0648 ± 0.001
^{65}Zn	1115.54	Quicksafe A	12.0	0.0605 ± 0.001
^{60}Co	1173.23	Quicksafe A	12.0	0.0591 ± 0.001
^{60}Co	1332.49	Quicksafe A	12.0	0.0555 ± 0.001

Table 2
Activity concentration of a ^{60}Co solution as determined by the non-extrapolation method

Source mass (mg)	Beta rate (cps)	C/G	Source activity (Bq)	Solution activity concentration (kBq/g)
42.175	4639	0.900129	5120 ± 6	121.40 ± 0.15
81.224	8954	0.906471	9818 ± 9	120.88 ± 0.11
99.081	10884	0.900534	12008 ± 11	121.20 ± 0.11
146.590	16011	0.896067	17747 ± 12	121.07 ± 0.08

Shown are data for a few selected sources. The measured rates, after correction for rate dependent effects, were decay corrected to the reference date, 28 March 2002. The uncertainties shown are a consequence of counting statistics only.

Table 3
Results of activity measurements on ^{54}Mn liquid scintillation sources, analysed by the non-extrapolation method as well as by conventional efficiency extrapolation. The uncertainties shown result from counting statistics

Source	Scintillator	Volume (ml)	Extrapolation method (BG/C vs. G/C)			Non-extrapolation method			
			Efficiency range (%)	Order of polynomial fit	Activity (Bq)	Rate B (cps)	C/G	Activity (Bq)	Difference from extrapolation method (%)
1	Insta-Gel	12.0	10–31	First	60630±100	21706	0.312528	60557±80	−0.12
2	Insta-Gel	20.5	10–30	First	60147±125	20986	0.294562	59697±85	−0.75
3	HP Ready-Solv	12.0	9–30	First	25218±56	8699	0.297416	25366±45	+0.59
4	Insta-Gel	12.0	1–30	Second	1383±10	487.65	0.304136	1391±7	+0.54
5	Insta-Gel	12.0	1–32	Second	1398±9	514.59	0.321040	1405±7	+0.49
6	Insta-Gel	12.0	3–39	Second	10784±23	4690	0.391279	10859±18	+0.69
7	Insta-Gel	12.0	4–40	Second	10515±21	4682	0.402755	10578±16	+0.60
8	Insta-Gel	12.0	4–39	Second	11091±22	4848	0.393498	11169±17	+0.70
9	Insta-Gel	12.0	2−32	Second	23331±59	8484	0.316596	23420±40	+0.38

Table 4
Results of activity measurements on ^{65}Zn liquid scintillation sources, analyzed by the non-extrapolation method as well as by conventional efficiency extrapolation. The uncertainties shown result from counting statistics

Source	Extrapolation method (BG/C vs. G/C)			Non-extrapolation method			
	Efficiency range (%)	Order of polynomial fit	Activity (Bq)	Rate B (cps)	C/G	Activity (Bq)	Difference from extrapolation method (%)
1	23–59	First	4096±13	2500	0.586868	4130±15	+0.83
2	22–55	Second	4617±38	2650	0.553916	4618±16	+0.02
3	21–57	Second	4691±25	2782	0.565085	4758±11	+1.43
4	20–56	First	6595±11	3856	0.562049	6629±14	+0.52
5	19–56	First	5665±11	3301	0.560582	5689±13	+0.42
6	18–55	Second	9234±30	5326	0.550304	9337±15	+1.12

5.3. ^{65}Zn

The ^{65}Zn solution used was received from the BIPM for measurement within the framework of an international key comparison. The sources were prepared in 12 ml of Quicksafe A liquid scintillator. The ^{65}Zn decay parameter values required for the calculations were taken from the "Nucleide" data base (Bé et al., 1998). Thus, $f_1 = 50.61(22)\%$, $f_2 = 47.97(22)\%$ and $f_3 = 1.42(2)\%$. As for ^{54}Mn, the 4π threshold was set just below the single electron peak to maximize the counting efficiency. The analysis of source data collected with a narrow γ-ray window set over the upper portion of the 1115 keV photopeak is given in Table 4. The uncertainty component due to P_I is 0.04%. The mean activity given by the non-extrapolation method is about 0.7% higher than for the efficiency extrapolation analysis. This corresponds to a solution activity concentration that is 1.0% higher than the result specified for the comparison (uncertainty 0.31% at 1σ). It is also noted that for ^{65}Zn the requirement of a narrow γ-ray window is not crucial for the non-extrapolation method. Setting a window over the full photopeak gave results that were on average just 0.56% higher than a second-order polynomial efficiency analysis that in turn was 0.28% higher than that specified for the comparison.

6. Conclusions

The investigation into gauging the viability of applying direct analysis to extract the activity from data collected by liquid scintillation coincidence counting has proved successful. The non-extrapolation technique has been demonstrated by measurements made on ^{60}Co, where the activity obtained agrees well with that given by the extrapolation method. The result is also in excellent agreement with the mean reference value for ^{60}Co in the BIPM's International Reference System.

The method has been applied to the more difficult cases of ^{54}Mn and ^{65}Zn. Agreement with the activity given by the extrapolation method was within experimental uncertainty, being ~0.5% higher for ^{54}Mn and ~1.0% higher for ^{65}Zn. Besides providing a useful supplement to conventional efficiency analysis via extrapolation, the method reported offers the advantage of experimental simplicity and considerable reduction in data collection since there is no need for efficiency variation.

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