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Absolute activity measurement of the electron-capture-based radionuclides ^{139}Ce, ^{125}I, ^{192}Ir and ^{65}Zn by liquid scintillation coincidence counting

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Abstract

Four radionuclides with electron-capture-based decay schemes have been directly measured by a liquid scintillation coincidence extrapolation technique. ^{125}I, ^{192}Ir and ^{65}Zn were measured as part of international key comparisons held under the auspices of the International Bureau of Weights and Measures (BIPM). The ^{139}Ce measurements formed part of a regional comparison organized by the Asia Pacific Metrology Programme (APMP). Since ^{139}Ce decays purely by electron-capture, the basic method is described for this radionuclide. Results and difficulties encountered are discussed and uncertainty budgets are presented.

Keywords: Liquid scintillation coincidence counting; ^{139}Ce; ^{125}I; ^{192}Ir; ^{65}Zn

1. Introduction

Four radionuclides with electron-capture-based decay schemes have been measured using variations of the 4π(LS)e,x-γ coincidence extrapolation technique. Advantages of using liquid scintillation (LS) counting in the 4π-channel are that self-absorption does not occur, leading to Auger electrons being detected with relatively high efficiency; source preparation is easy; and the source geometry is highly reproducible. The latter leads to good reproducibility of the counting efficiency of the X-rays and Auger electrons, which in turn gives rise to consistent results amongst the counting sources. The application of the technique has been reported on previously for ^{201}Tl (Simpson and Meyer, 1994), but little else has been reported in the open literature.

^{125}I, ^{192}Ir and ^{65}Zn were measured as part of international key comparisons held under the auspices of the International Bureau of Weights and Measures (BIPM) and ^{139}Ce was measured as part of a regional comparison organized by the Asia Pacific Metrology Programme (APMP).

Since ^{139}Ce decays purely by electron-capture, the basic method is reviewed for this radionuclide. ^{192}Ir and ^{65}Zn have more complex decay schemes and resulting differences are discussed. For ^{125}I, a modification of the 4π(e,x)-γ coincidence extrapolation technique was investigated to complement widely used methods that are based entirely on K X-ray counting (Taylor, 1967; Eldridge and Crowther, 1964). Results and difficulties encountered are presented.

2. Liquid scintillation coincidence counting

The basic method is reviewed with respect to ^{139}Ce, which decays purely by electron-capture to the 166 keV excited state of ^{139}La. This state de-excites through the emission of conversion electrons and γ-rays. The detection efficiency analysis is based on a counting source being viewed by two phototubes in coincidence, with a single NaI(Tl) crystal detecting the γ-rays. The 4π coincidence count rate from the LS detector is given by (Steyn et al., 1976)

$$N_{4\pi} = N[f + (1-f)PP_S + (1-f)(1-P)\varepsilon_{AX}], \quad (1)$$

where N is the source activity, f is the conversion electron probability, ε_{AX} is the detection efficiency for the

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electron-capture emissions, P is the probability of the γ-rays interacting with the scintillator solution and P_S is the detection probability of the Compton electrons together with the particles emitted in the electron-capture process (forming a sum pulse). The conversion electrons are detected with 100% efficiency.

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

$$N_\gamma = N(1-f)(1-P)\varepsilon_\gamma, \tag{2}$$

where ε_γ is the efficiency for detecting events in the full-energy peak.

The coincidence count rate is thus

$$N_C = N(1-f)(1-P)\varepsilon_{AX}\varepsilon_\gamma. \tag{3}$$

Thus ε_{AX} can be determined experimentally by measuring N_C/N_γ. Eq. (1) can then be expressed as

$$\frac{N_{4\pi}N_\gamma}{N_C} = N(1-f)(1-P) + N[f + (1-f)PP_S]\frac{N_\gamma}{N_C}. \tag{4}$$

When $(N_\gamma/N_C) \to 1$ so will P_S and the source activity is given by the corresponding extrapolated value of $N_{4\pi}N_\gamma/N_C$. As N_γ/N_C is varied, P_S is expected to change due to the fairly low energy of the Compton electron spectrum. If this variation is linear, it is a simple matter to show that the functional form of Eq. (4) is predicted to be linear.

The approach and complexity of the detection efficiency analyses of the other radionuclides investigated here are dependent on the particular decay scheme.

3. Activity measurements

In all cases, counting sources were prepared in custom made flat-faced cylindrical glass vials, each being viewed in turn by two RCA 8850 phototubes coupled in coincidence. The escaping γ-rays were detected with a 75 mm × 75 mm NaI(Tl) crystal. Conventional analogue pulse processing was used, with slow logic signals fed into a locally designed and built coincidence unit (Simpson and Meyer, 1988) and routed through to a 32-channel scaler, enabling up to 15 datum points to be collected simultaneously (Simpson and van Oordt, 1997). To ensure the detection of the low energy X-rays and Auger electrons, the lowest threshold in the 4π-channel was set below the single electron peak. The counting efficiency was varied by setting additional discrimination levels among the first few monopeaks, counting integrally above these thresholds. In this way all electron-capture events contribute to the counting, thereby eliminating the need of a correction for losses as reported by Funck and Larsen (1983) for a proportional counter. Suitable γ-window settings were set for each radionuclide as described below.

The recorded counts were corrected for background, afterpulsing and rate-dependent effects due to the coincidence resolving time (0.48 ± 0.01 μs) and the inherent non-extending deadtime (Simpson, 1991), which was in the range 1.07–1.39 μs depending on the threshold settings. The afterpulse correction for the lowest threshold setting was measured as 0.81% (^{65}Zn), 0.44% (^{192}Ir), 0.23% (^{125}I) and 0.15% (^{139}Ce). In most cases data in the form $N_{4\pi}N_\gamma/N_C$ vs. N_γ/N_C were generated from the corrected counts.

3.1. ^{139}Ce

The ^{139}Ce solution was received from the coordinating laboratory for measurement within the framework of a regional key comparison. Four accurately weighed sources were prepared from the solution by mixing in 12 ml of liquid scintillator (Quicksafe A from ZINSSER ANALYTIC) to which 12.5 ml/l of a carrier solution comprising 1 g/l $CeCl_3 \cdot 7H_2O$ in 1 M HCl had been added to minimize adsorption to the glass vials.

Three separate windows were set in the γ-channel (166 keV photopeak; spanning a range from the K X-ray peak to the photopeak; and the iodine escape peak and photopeak), providing three different sets of data for each source. Thirteen bias levels were set in the 4π-channel and all levels were counted simultaneously in integral mode, giving 13 data points for each γ-window. Fig. 1 shows typical sets of data from one of the sources. The source activity was obtained from an extrapolation of the data to $N_\gamma/N_C = 1$ using a function that mimics a linear fit at the higher efficiencies coupled to a second-order fit at lower efficiencies (Miyahara et al., 1986). Since all the sets gave the same extrapolated value within counting statistics, an average value was taken. At the present time the results of the comparison are confidential, but the present activity value agrees well with a preliminary analysis of the submissions of the other participants. The uncertainty estimates are given in Table 1.

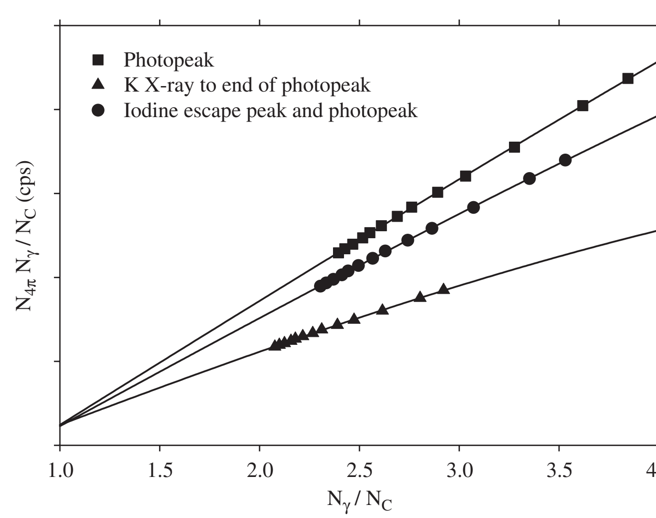

Fig. 1. A series of 4π count rates, $N_{4\pi}$, from a ^{139}Ce source expressed as the inverse of the counting efficiency for various γ-window settings, where N_γ is the γ-channel rate and N_C the coincidence rate. The lines are fits to the data sets using a third-order polynomial with the second-order coefficient set to zero.