Applied Radiation and Isotopes 109 (2016) 276–280

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Activity determination of ^{59}Fe by 4π beta–gamma counting using liquid scintillation in the beta channel

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HIGHLIGHTS

- We report on absolute activity measurements of ^{59}Fe made at the NMISA.
- Measurements were made via 4π(LS)β–γ coincidence counting.
- We compare results from extrapolation and non-extrapolation techniques.
- The techniques are based on efficiency analyses.
- The non-extrapolation result is lower than the extrapolation result by 0.33%.

ARTICLE INFO

Article history:
Received 31 March 2015
Accepted 20 November 2015
Available online 22 November 2015

Keywords:
Liquid scintillation coincidence counting
Extrapolation technique
Non-extrapolation method
Iron-59

ABSTRACT

This paper reports on absolute activity measurements of iron-59 made at the National Metrology Institute of South Africa (NMISA) via 4π(LS)β–γ coincidence counting. The exercise formed part of an Asia Pacific Metrology Program (APMP) regional key comparison. Source data were analysed by the extrapolation technique for a number of gamma-ray window settings. In addition, a feasibility study was undertaken on a second technique; a non-extrapolation method based on a detection efficiency analysis. The reported activity concentration of the ^{59}Fe solution was determined with a relative uncertainty of 0.28% ($k=1$), the uncertainty being due mainly to the rate vs. efficiency fitting process. The result from the non-extrapolation method was lower than that given by extrapolation by 0.33%, within two standard deviations. Possible reasons for the small discrepancy are discussed.

1. Introduction

The Technical Committee for Ionizing Radiation (TCRI) of the Asia Pacific Metrology Program (APMP) recently organized a regional key comparison of activity measurements of the radionuclide ^{59}Fe (APMP.RI(II)-K2.Fe-59), in which the National Metrology Institute of South Africa (NMISA) participated. This paper reports on absolute measurements made by the β–γ coincidence technique, with liquid scintillation (LS) counting comprising the 4πβ channel. The literature indicates that LS counting has not previously been reported for ^{59}Fe by this familiar technique, usually incorporating proportional counters (Park et al., 2000). The standardization of ^{59}Fe has however been successfully measured by other LS methods, namely CIEMAT/NIST and TDCR (Günther, 1994; Kossert and Nähle, 2014).

^{59}Fe decays to various excited states of ^{59}Co through five beta branches, two of the branches making up 98.5% of the beta emissions (Fig. 1). Internal conversion is low and the gamma transitions comprise predominantly two high energy gamma rays. A detection efficiency analysis based on the decay scheme was undertaken to confirm that the customary extrapolation analysis does indeed provide the source disintegration rate for ^{59}Fe when applying the 4π(LS)β–γ coincidence counting technique. Experimentally, a locally assembled double phototube LS system in coincidence was used to detect the beta particles, together with a NaI (Tl) crystal for gamma-ray detection. Measurements and analysis by the extrapolation technique provided the results for the APMP inter-comparison (reference date 01 June 2014, 0 h UTC).

Because of the experimental simplicity, where only a single beta, gamma and beta–gamma coincidence counting set are required per source, a study was undertaken to attempt to determine the ^{59}Fe activity by the use of an alternative non-extrapolation analysis technique, which was previously successfully applied at NMISA to the beta-emitter ^{60}Co and the electron-capture radionuclides ^{65}Zn and ^{54}Mn (Simpson and Morris, 2004). This method appears to have been first investigated by Perolat via

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http://dx.doi.org/10.1016/j.apradiso.2015.11.054
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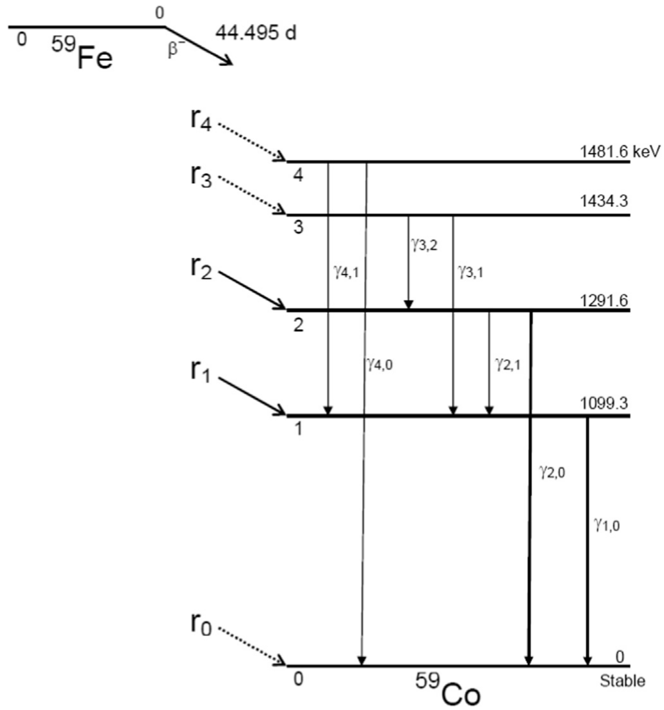

Fig. 1. A simplified decay scheme of ^{59}Fe showing the pertinent features. The two predominant beta branches are shown with solid lines. The intensity of the $\gamma_{1,0}$ gamma ray is 56.59% and that of $\gamma_{2,0}$ is 43.21% (Bé and Chisté, 2004).

4π(PC)β–γ coincidence proportional counting (Perolat, 1973) applied to ^{59}Fe and ^{99}Mo and also provides a case of ^{65}Zn. In our ^{59}Fe study with liquid scintillation counting, the average activity concentration extracted was lower than for the extrapolation method by 0.33%, a potential systematic effect being due to the spurious pulse correction (mainly afterpulsing) if not estimated correctly.

2. Detection efficiency analysis

The beta count rate N_B for ^{59}Fe is given by

$$N_B = N_0 \left[r_0 \varepsilon_{\beta 0} + \sum_1^4 r_i \left\{ \varepsilon_{\beta i} + \left(1 - \varepsilon_{\beta i}\right) g_{i,L} P_{i,L} P_{\gamma i,L} \right\} \right] \tag{1}$$

where N_0 is the source activity; the r_i are the beta branching ratios to levels i ($\sum_0^4 r_i = 1$); the $\varepsilon_{\beta i}$ are the corresponding beta efficiencies; $P_{i,L}$ and $P_{\gamma i,L}$ are the gamma-ray interaction probabilities and detection probabilities in the beta channel, respectively; $g_{i,L}$ is a factor such that the γ-ray intensity is given by $r_i g_{i,L}$ for that branch; and L is a value that identifies the final level of the gamma transition.

With a γ-window set over the photopeak of the highest-energy gamma ray $\gamma_{2,0}$ (assumed to correspond to the r_2 branch only), the gamma-ray count rate given by the NaI detector is:

$$N_G = N_0 r_2 g_{2,0} \left(1 - P_{2,0}\right) \varepsilon_\gamma, \tag{2}$$

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

The β–γ coincidence count rate is thus given by

$$N_C = N_0 r_2 g_{2,0} \left(1 - P_{2,0}\right) \varepsilon_{\beta 2} \varepsilon_\gamma \tag{3}$$

and consequently

$$\frac{N_C}{N_G} = \varepsilon_{\beta 2} \tag{4}$$

when $\varepsilon_{\beta 2}$ reaches 100% efficiency, so do those of the other branches. Thus by extrapolating $\frac{N_C}{N_G}$ to a value of 1, the activity is obtained. The case when a window is set over both the $\gamma_{2,0}$ and $\gamma_{1,0}$ gamma rays is treated similarly to a previous analysis undertaken for ^{131}I (van Wyngaardt and Simpson, 2005).

3. Experimental

3.1. Source preparation

Five liquid scintillation counting sources were prepared in custom made flat-faced cylindrical glass vials by dispensing accurately weighed aliquots of the ^{59}Fe solution received into 12 mL of scintillation cocktail. Source masses (buoyancy corrected) ranged between 39 and 51 mg. The counting vials used were pretreated by soaking each vial with 12–13 mL of carrier solution comprising 5 mg per 100 mL $FeCl_3$ in 1 M HCl for three days, to aid in minimizing adsorption of the active material to the vial surface. The liquid scintillation cocktail used was Quicksafe A from Zinsser Analytic, to which 3 mL L^{-1} of 3 M HCl had been added as an additional precaution against possible adsorption.

3.2. Measurements

Measurements on five sources were undertaken on 22–23 July 2014 with a locally developed double-phototube system viewing each liquid scintillation source in turn, details of which are given in Simpson and Meyer (1988). The gamma rays were counted with a NaI(Tl) detector and beta–gamma coincidences recorded together with the beta and gamma counts (Simpson and van Oordt, 1997). For the extrapolation method, the lowest beta channel threshold was set to just above the third monopeak to ensure no counting of ^{55}Fe if present as an impurity. Variation of the 4πβ counting efficiency was carried out by threshold discrimination, where fifteen bias levels were set (the typical beta rate was 9000 s^{-1} with a background rate of 1.4 s^{-1}). The spurious pulse contribution in this range was small, ranging from 0.15% in the lowest discrimination level down to 0.0%. The system deadtime varied between 1.13 and 1.19 μs and the coincidence resolving time was ~0.48 μs. Fig. 2 shows the four gamma-ray windows investigated, three incorporating one or more of the predominant gamma rays (windows A, B and C) and one was set to count integrally above a threshold set within the Compton spectrum (window D). The maximum efficiency was typically 89%. An adsorption test was performed on one of the sources by decanting

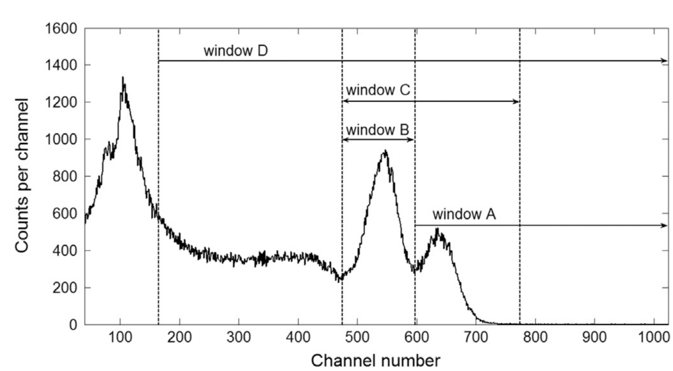

Fig. 2. A plot of the gamma-ray spectrum for ^{59}Fe showing the various windows selected for activity measurement by the 4π(LS)β–γ coincidence counting extrapolation method.