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Beta decay of the fission product ¹²⁵Sb and a new complete evaluation of absolute gamma ray transition intensities

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ARTICLE INFO

Article history: Received 12 February 2011 Received in revised form 29 August 2011 Accepted 21 December 2011 Available online 30 December 2011

Keywords: Radionuclide ¹²⁵Sb Beta decay Internal conversion coefficients Relative gamma ray intensities Absolute gamma ray emission probabilities Evaluation

ABSTRACT

The radionuclide ¹²⁵Sb is a long-lived fission product, which decays to ¹²⁵Te by negative beta emission with a half-life of 1008 day. The beta decay is followed by the emission of several gamma radiations, ranging from low to medium energy, that can suitably be used for high-resolution detector calibrations, decay heat calculations and in many other applications. In this work, the beta decay of ¹²⁵Sb has been studied in detail. The complete published experimental data of relative gamma ray intensities in the beta decay of the radionuclide ¹²⁵Sb has been compiled. The consistency analysis was performed and discrepancies found at several gamma ray energies. Evaluation of the discrepant data was carried out using Normalized Residual and RAJEVAL methods. The decay scheme balance was carried out using beta branching ratios, internal conversion coefficients, populating and depopulating gamma transitions to ¹²⁵Te levels. The work has resulted in the consistent conversion factor equal to 29.59(13) %, and determined a new evaluated set of the absolute gamma ray emission probabilities. The work has also shown 22.99% of the delayed intensity fraction as outgoing from the 58 d isomeric 144 keV energy level and 77.01% of the prompt intensity fraction reaching to the ground state from the other excited states. The results are discussed and compared with previous evaluations. The present work includes additional experimental data sets which were not included in the previous evaluations. A new set of recommended relative and absolute gamma ray emission probabilities is presented.

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1. Introduction

Precise decay scheme data of radionuclides are required for a variety of applications in basic research and in applied nuclear fields. Transition intensities in radionuclide decay are particularly important in determining the efficiency response curves of detectors (Rajput et al., 2002, Iqbal et al., 2009) and in nuclear cross section measurements (Rajput et al., 2003, Rajput et al., 2009). The radionuclide ¹²⁵Sb is an important fission product, which decays to ¹²⁵Te by negative beta emission with a half-life of 1008 day and emits several gamma rays in the energy range 20–700 keV. The gamma ray emission probabilities of the isotope are valuable in the investigation of nuclear fuel burn-up, decay heat calculations, fission cross-section studies and activation analysis. The isotope ¹²⁵Sb, due to multiple gamma ray emissions in the low energy region, can also be used effectively as a detector calibration standard.

In this work, all the previously measured experimental data of the relative gamma ray intensities following the beta decay of ¹²⁵Sb have been compiled to perform a consistent and

comprehensive evaluation of the transitions in the decay scheme of the nuclide. The compiled relative intensity data revealed the existence of several discrepancies. In earlier studies, Longoria-Gandara et al. (1990) had pointed out discrepancies in the data of the isotope and adopted an arbitrary inflation of uncertainties method for the evaluation. Later some new sets of experimental measurements of the isotopes were published (Helmer, 1990; Smith et al., 1992; Fawaz and Stewart, 1993; Sainath et al., 1998) and few new evaluation methodologies (described later) were proposed. The availability of these methodologies and later measurements motivated the present evaluation of the relative intensities. In addition, the decay scheme balancing is carried out considering the selected energy levels at 35.5, 144.8 and ground state of ¹²⁵Te to deduce the conversion factor that allows the determination of the absolute gamma ray emission probabilities. Electron and total intensities were also calculated and used to find the prompt and delayed intensity fractions.

2. Evaluation methodologies

Some of the evaluation methodologies existing at present, to deal with discrepant data such as half-life, gamma ray emission probabilities include the Normalized Residual (NR) method

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⁰⁹⁶⁹⁻⁸⁰⁶X/ $\$ - see front matter @ 2011 Elsevier Ltd. All rights reserved. doi:10.1016/j.radphyschem.2011.12.030

(James et al., 1992), RAJEVAL method (Rajput and MacMahon (1992)), Boot Strap method (BS) (Helene and Vanin, 2002) and Limitation of relative statistical weight method (LWM) (Zijp, 1985). The LWM methodology often recommends the unweighted mean as an evaluated value depending upon the fulfillment of certain laid down criterion. It means that the method in such cases ignores the useful information of experimental uncertainties and the efforts made by the experimenter made in error propagation. The BS method does not take at all into account the experimental uncertainties to reach the evaluated value and thus completely ignores the valuable part of the experimental data. A useful comparison study on the convergence of different evaluation techniques was published by MacMahon et al. (2004). In the present work, we selected the following two methodologies as they utilize the full experimental information including the experimental uncertainties.

2.1. RAJEVAL technique

RAJEVAL evaluation methodology was proposed by Rajput and MacMahon (1992) and comprises three stages for the detection of outliers, data inconsistencies and evaluation.

A population test is performed in stage-1. The quantity y_i is calculated for each value to detect outliers in the data set:

$$y_i = \frac{x_i - \mu_i}{\sqrt{\sigma_i^2 + \sigma_{\mu i}^2}}$$

where, μ_i and $\sigma_{\mu i}$ are respectively the un-weighted mean and associated standard deviation of all the experimental data excluding the ith-measurement (x_i). At the 5% significance level, the critical value of $|y_i|$ for the two tailed test is 1.96. The measurements in the data with $|y_i| > 3 \times 1.96$ are considered as outliers and may be excluded from further stages of evaluation.

In the second stage, internal inconsistencies of the remaining data, after the expulsion of any outlier as a result of the above test, are determined. It requires the calculation of the standardized deviates Z_i :

$$Z_i = \frac{x_i - \overline{x}}{\sqrt{\sigma_i^2 - \sigma_w^2}}$$

where σ_w represents the weighted uncertainty in the weighted mean $\overline{x}.$

The probability integral P(z) for each Z_i is determined by

$$P(z) = \int_{-\infty}^{z} \frac{1}{\sqrt{2\pi}} \exp\left(-\frac{t^2}{2}\right) dt.$$

The values of P(z) may be obtained from normal distribution tables.

The absolute difference between the probability integral values and 0.5 determines the central deviations for which the critical value is decided by the expression:

$$CV = 0.5^{N/(N-1)}$$
 for $N \ge 2$

If the central deviation of any value is greater than the critical value, that value is regarded as inconsistent with the remainder of the data set.

In the third stage of the evaluation, the uncertainties of the internally inconsistent measurements are increased to σ_i , where

$$\sigma_i = \sqrt{\sigma_i^2 + \sigma_w^2}$$

This requires an iteration procedure; each time σ_w is recalculated and added in quadrature to the uncertainties of those values with central deviations greater than the critical value. The iteration terminates when all the central deviations are less than the critical value.

2.2. Normalized Residual Method

James et al. (1992) introduced the Normalized Residual Method. This method defines the normalized residual R_i and a quantity R_o as

$$R_i = \left(\frac{w_i.W}{W - w_i}\right)^{1/2} . (x_i - \overline{X})$$

$$R_0 = (1.8 \ln N + 2.6)^{1/2}; \ 2 \le N \le 100$$

where, $W = \sum w_i = (\text{sum of the weights of all individual measurements})$, \overline{X} is the weighted mean and N is the total number of measurements

For any measurement with condition $|R_i| > R_o$, the weight w_i is adjusted such that the normalized residual is changed to R_o . If more than one measurement has a normalized residual larger in magnitude than R_o , an iterative procedure is used, reducing at each stage the largest $|R_i|$ to R_o .

These two methods adjust only a few experimental uncertainties and the evaluated value is obtained using the weighted mean of the experimental data computed utilizing the adjusted uncertainties. In this work the adopted value is taken as the average of the two evaluated values obtained from these methods as suggested by MacMahon (2006) and this is the approach adopted for the first time in the evaluation of ¹²⁵Sb.

3. Results and Discussion

3.1. Relative intensity data compilation and evaluation

The experimentally measured relative gamma ray intensity data in the beta decay of ¹²⁵Sb have been compiled from 1970 and shown in Table 1. The compilation is complete in the sense that it also includes the additional data sets that were not included in previous work of Helmer et al. (2004). The tabulated experimental measurements show that nearly all the experimenters have identified the main intense gamma rays whereas weaker ones were detected on case-to-case basis. The latest measurements of Sainath et al., (1998) have not only provided verifying signatures to the measurements of all the other experimenters data presented in Table 1 but also found new gamma ray energies at 61.85[0.0068(27)], 81.02[0.017(1)], 132.8[0.0029(19)], 209.3[0.152(9)], 331.8[0.0085(8)], 366.6[0.027(2)], 401.9[0.021(2)], 489.7[0.0046(23)], 497.4[0.009(1)], 503.1[0.013(6)], 538.6[0.0047(25), 617.4[0.018(2)], 652.8[0.009(3)] keV, thus describing the total of 38 gammas in the beta decay of ¹²⁵Sb. However, in the Table 1 we include only those gamma rays that have been detected by at least two experimenters thereby giving the data of 25 gamma rays. The subjective assessment of the Sainath data (Sainath et al., 1998) reveals an interesting fact: the gamma ray energy at 314 keV has been measured with an intensity of 0.014(15) and is confirmed by two other experimenters, but 366.6 keV is only measured by Sainath et al. (1998) with almost twice the intensity value [0.027(2)] in comparison with the intensity value of 314 keV. As the magnitude of 366 keV intensity is considerably higher we expected that some other experimenters should have detected it but this was not the case. Hence such a kind of irregularity requires a fresh and careful measurement to avoid the doubts and verify the beta decay scheme profile of ¹²⁵Sb.

The consistency analysis of the compiled data (Table 1), was carried out by calculating the weighted mean and computing the chi squared value. If chi square values fall within the limits of 5% significance level, the data is considered consistent. However, the generally accepted criterion for chi-square test: that for a good data set the chi-square per degree of freedom (χ^2/ν) be around 1,

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