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Half-life measurement of ^{124}Sb

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ABSTRACT

The half-life of ^{124}Sb was determined experimentally by following the decay of a source from a radiopure solution with a Centronic IG12 ionisation chamber. Thousands of measurements were performed over a period of 358 days, i.e. about six half-life periods. However, the data analysis was restricted to the first 221 days, in order to limit the dominant uncertainty component associated with the hypothetical possibility of a systematic error on background subtraction. The resulting value for the ^{124}Sb half-life, 60.212 (11) days, is found to be in very good agreement with published values, but carries a lower uncertainty. Major uncertainty contributions pertain to possible systematic errors in background correction, long-term changes in source-detector geometry and medium- and long-term instability of the instrument. Additional measurements were performed with a high-purity germanium detector to confirm the above value.

1. Introduction

^{124}Sb is a relatively short-lived radionuclide (60.2 d), which disintegrates through β^- transitions to various excited levels in ^{124}Te. The decay scheme is rather complex, involving γ-transitions with energies between 148 and 2807 keV (Bé et al., 2004). Considering that this nuclide emits abundant high-energy gamma rays, it could be valuable as a standard radionuclide for the calibration of gamma-ray detectors in that energy range, provided that the decay data would be known accurately. In the recent Euramet project 907, primary standardisation measurements and x- and γ-ray intensity measurements were performed to establish equivalence among standardisation techniques (Chauvenet et al., this issue) and to derive more accurate emission rates (Bé et al., this issue).

In the frame of this project, a precise measurement of the ^{124}Sb half-life was performed at IRMM. Thousands of activity measurements were performed with an ionisation chamber over a total time span of about six half-lives. High-resolution gamma-ray spectrometry measurements were performed to check for the presence of possible impurities, and to obtain additional data for the half-life determination. In this paper, we present the relevant experimental aspects of the half-life determination, including a detailed uncertainty budget, and compare our result with literature values.

2. Experimental conditions

2.1. Source preparation

The radioactive solution of ^{124}Sb was prepared by LNE-LNHB in the frame of the Euramet project 907 (Chauvenet et al., this issue; Bé et al., this issue). The chemical composition of the solution was 2 M HCl containing $SbCl_3$ carrier (3 μg g^{-1}). Its activity concentration was about 1.56 MBq g^{-1} at the reference date of March 1, 2007, 0 h UTC. An ampoule containing 5 g of this ^{124}Sb solution was sent to IRMM, at the end of February 2007. From this solution, 3.8 g was sealed in a glass ampoule.

The ^{124}Sb sources for use in gamma-ray spectrometry were prepared by gravimetrically depositing an aliquot of the original radioactive solution onto a 10-mg cm^{-2} polyester film. After the liquid had evaporated, the sources were sealed by another polyester film, supported by a 34-mm stainless steel annulus. Two of the prepared sources were used for the measurement of the ^{124}Sb half-life; one carried 24 mg of the original solution and the other 11 mg, or 35 and 15 kBq of ^{124}Sb, respectively, at the reference date.

2.2. Impurity check

The ^{124}Sb was produced by neutron activation of stable antimony in Sb_2O_4. The natural antimony contains 57.21% ^{121}Sb and 42.79% ^{123}Sb. Firstly, the ^{122}Sb formed (cross section σ=5.84 b, Antony, 2002) has a relative short half-life of 2.7 days, and decays to the stable ^{122}Te. A small fraction of the ^{122}Te may have been activated to form ^{123m}Te (σ=1.1 b, $T_{1/2}$=119.7 d) and

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Table 1
Response of the IG12 ionisation chamber to (possible) impurities, relative to the response to ^{124}Sb, taken as 1.

Nuclide	Half-life	Relative response of IG12
^{122}Sb	2.7238 (2) d	0.27
^{125}Sb	2.75855 (25) a	0.26
^{123m}Te	119.3 (1) d	0.13
^{125m}Te	57.40 (15) d	0.00037

^{123}Te (σ=2.3 b, $T_{1/2} > 6 \times 10^{14}$ a). Therefore, ^{123m}Te had to be considered as a possible impurity, while most of the ^{122}Sb should have decayed away by the time the measurements were started. Secondly, from double activation of ^{123}Sb (σ=4.1 and 17.4 b), traces of ^{125}Sb ($T_{1/2}$=2.76 y) may have been formed. The ^{125}Sb is expected to be more apparent when most of the ^{124}Sb has decayed. Another pathway, via activation of ^{124}Te, leads to the possible production of ^{125}Te^{m}.

Purity checks of the solution were done by means of x- and γ-ray spectrometry at the various participating laboratories. No impurities were found, and detection limits were determined at LNE-LNHB (in February 2007) for the possible impurities: $A(^{125}Sb)/A(^{124}Sb) = 4 \times 10^{-4}$ Bq Bq^{-1}; $A(^{122}Sb)/A(^{124}Sb) = 2 \times 10^{-4}$ Bq Bq^{-1}; $A(^{125}Te^{m})/A(^{124}Sb) = 10^{-3}$ Bq Bq^{-1} (Chauvenet et al., this issue). At IRMM, γ-ray spectrometry was performed on a low-energy germanium detector, at several phases of the measurement campaign, to quantify the traces of ^{125}Sb and ^{123m}Te, and to confirm the decay of ^{122}Sb. The derived activity concentrations relative to ^{124}Sb at the reference date were 3.2×10^{-6} Bq Bq^{-1} for ^{125}Sb and 4.5×10^{-6} Bq Bq^{-1} for ^{123m}Te, respectively.

To obtain an estimate of the response of the ionisation chamber to the possible impurities ^{122}Sb, ^{125}Sb, ^{123m}Te and ^{125m}Te, the mathematical model of Švec and Schrader (2002) was applied. Table 1 presents the response to the impurities relative to the response to ^{124}Sb. From this table it is clear that for half-life measurements performed with our IG12 ionisation chamber, traces of ^{125m}Te have a negligible effect.

2.3. Ionisation chamber

The ampoule was measured in a Centronic IG12 well-type ionisation chamber, containing argon gas at a pressure of 2 MPa. The steel well has a thickness of 0.8 mm. A 50-mm thick lead shield surrounds the chamber. The ionisation current is integrated over a custom-made air capacitor, which is placed in another lead shield to reduce discharge effects by radiation. The capacitor voltage is sampled using a Keithley 6517A electrometer operating in voltage mode, triggered every 2 s by a stable crystal oscillator. The trigger period is measured with a calibrated frequency meter, traceable to the SI unit second by comparison with a standard frequency generated by a DCF-77 receiver, of which the oscillator is synchronised with the primary atomic clocks of PTB in Braunschweig Germany. During the measurement campaign, measurements of background and of a long-lived ^{226}Ra source were regularly repeated.

The raw data consist of voltage samples of the capacitor, taken every 2 s, in cycles from 0 to 9 V. When a voltage of nine volts is reached, the capacitor is discharged and after a waiting period of 30 s another measurement cycle is started. For every two consecutive voltage samples, the voltage difference is calculated and these differences are checked for outliers, e.g. due to electronic spikes. If there is at least one outlier within a series of samples, the complete charge cycle is rejected from further processing. In this way, 276 out of 20 101 cycles were rejected. The duration of one cycle varied with the activity of the source. The integration time was 26 s at the beginning of the measurement campaign and 334 s at the end.

The total integrated charge over one cycle was calculated from the product of the voltage difference between first and last sample and the capacity of the feedback capacitor. After background subtraction, this integrated charge was corrected for decay during the cycle, using a preliminary half-life value of 60.20 d. Then, each cycle result was also corrected for impurities.

The measurement campaign was started on 19 March 2007. While the total time span of the measurement campaign was 358 days, only data acquired in the first 221 days were taken into account for the half-life calculation. This was done because of uncertainty considerations, where the possible presence of a systematic uncertainty component in the background subtraction method would propagate into a major source of uncertainty on the half-life value after about 221 days.

2.4. High-purity germanium detector

A low-background, 93% relative efficiency, coaxial high-purity germanium (HPGe) detector and a 36% relative efficiency coaxial HPGe detector were used for the measurements. The first detector was housed in a 5-cm thick Pb shield of square intersection and the second in a 10-cm thick Pb shield of circular intersection.

In the acquisition system, a very stable 100-kHz quartz oscillator provided the time base of the live-time clock gates. As time base, the legal time broadcasted through the LF transmitter DCF77 was used. The spectra were acquired for 86 400 s each and the three stronger peaks of 603, 723 and 1691 keV were analysed. In total, 77 spectra recorded over a period of 210 days with the larger detector and 68 spectra taken over a period of 170 days with the second detector were analysed. The analysis consisted of summing the accumulated counts under each peak and subtracting a linear background. Corrections for dead time and decay during measurement were applied.

3. Measurement results

3.1. Ionisation chamber

Because of the large number of ionisation current measurements performed (10 138 cycles in 221 days), the data were grouped into time intervals of one day. For each group, the mean and variance were calculated, again using a preliminary half-life of 60.20 d to correct for the decay within the day. This reduced the number of data points to 46 mean ionisation currents as a function of time, expressed in days since the reference date. An exponential decay curve was fitted to the data, using the inverse of the variances as weights. The background count rate was included in the fitting process. The grouping was only done to facilitate the fitting procedure.

The least-squares fitting procedure yielded a half-life value of 60.212 d with an uncertainty of (only) 0.006 d. Relative residuals from the fit (scaled by a factor of 10^{-4}) are presented in Fig. 1. To check the randomness of the data, the autocorrelation plot of the residuals was constructed. The data appeared to be random, although the correlation coefficient approaches the 5% significance level a few times.

3.2. High-purity germanium detector

The half-life of ^{124}Sb was measured additionally with HPGe detectors. In general, HPGe detectors cannot compete with