Determination of the number of atoms of the long-lived nuclide ^{126}Sn by γ-ray spectrometry*

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Abstract By using HPGe γ-ray spectrometry, the activity of the long-lived fission product ^{126}Sn in a $SnOB_2$ sample was measured. The number of ^{126}Sn atoms and the ratio of ^{126}Sn to Sn were calculated based on the half-life value of 2.35×10^5a and the chemical stoichiometry. The result of the ratio of ^{126}Sn to Sn, $(1.033\pm0.037)\times10^{-8}$, is consistent with the results measured by the accelerator mass spectrometry (AMS) within uncertainty limits, which confirms our procedures in the measurement of ^{126}Sn by AMS and lays a foundation for the AMS measurement of ^{126}Sn at much lower levels.

Key words ^{126}Sn , activity measurement, γ-ray spectrometry, AMS

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1 Introduction

^{126}Sn is a long-lived beta emitting radionuclide with a half-life of $(2.35\pm0.07)\times10^5$ a [1]. Artificially produced ^{126}Sn has entered our environment through nuclear weapons testing and releases from reprocessing plants and may locally lead to strongly enhanced ^{126}Sn concentrations. So the long-lived ^{126}Sn may have implications on the nuclear pollution in our environment. Furthermore, in supernova explosions ^{126}Sn is predominantly produced by rapid neutron capture (r process). Searches for the presence of live ^{126}Sn at the time of condensation of solar matter by the detection of excess or depletion of its stable daughter nuclide ^{126}Te in meteoritic components have so far not met with success. The primary difficulty in the determination of the ^{126}Sn concentration by common mass spectrometry is the interference of the stable isobar ^{126}Te. AMS is one of the most important methods to detect minute amounts of ^{126}Sn. This work was carried out using the HI-13 tandem accelerator at CIAE National Lab. SnF_3^- ions from the negative ion source were injected into the accelerator whose terminal voltage was set at 8.7 MV. Sn^{10+} ions were selected by an analyzing system and finally counted selectively using a ΔE-E gas ionization detector. The ratio of ^{126}Sn to Sn (1.2×10^{-8}) has been obtained for a $^{126}SnO_2$ sample. In this experiment, the transmission efficiency of ^{126}Sn is derived from ^{124}Sn simulation and is not very accurate. It is necessary to adopt other independent methods to confirm the procedures in the measurement of ^{126}Sn by AMS, so as to lay a foundation for the AMS measurement of ^{126}Sn at much lower levels (10^{-10}–10^{-12}). In this paper, the activity of the ^{126}Sn sample was measured by using HPGe γ-ray spectrometry. The number of ^{126}Sn atoms and the ratio of ^{126}Sn to Sn were calculated based on the half-life value and the chemical stoichiometry to confirm the procedures in the measurement of ^{126}Sn by AMS.

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2 Experiment

2.1 Samples and instrumentation

The three radiochemically pure $^{126}SnO_2$ samples used in this work were provided by the Department of Radiochemistry of CIAE. They were separated from high-level radioactive waste 14 years ago. The mass of SnO_2 carrier for each sample is about 50–100 mg. As shown in Fig. 1, the gamma-ray transitions in the decay of ^{126}Sn to ^{126}Sb are of rather low energy. Therefore, the gamma-ray transitions from the decay of ^{126m}Sb and ^{126}Sb were considered. The branching ratio of 666.3 keV γ ray is 100% in the decay of ^{126}Sb. Therefore, the gamma rays from this transition are best suited for measuring the ^{126}Sb activity. Our ^{126}Sn samples were separated 14 years ago, the activity of ^{126}Sb has been in secular equilibrium with that of ^{126}Sn. So the ^{126}Sn activity is equal to that of ^{126}Sb. The measurements were performed at an ORTEC spectrometer system composed of a HPGe detector of GEM35-PLUS, a DSPEC-PLUS 92X unit with pre-amplifier，low and high-voltage power supply , main amplifier, ADC and the cache built-in, and a computer. The system has an energy resolution of 1.83 keV and a relative efficiency of 47% (both are for 1.33 MeV).

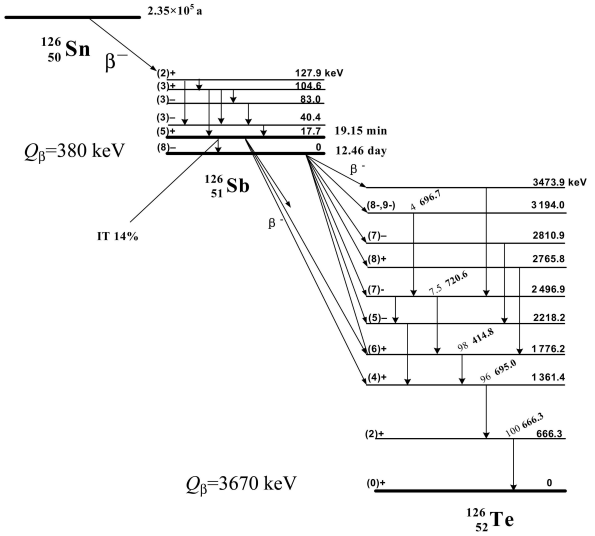

Fig. 1. The simplified decay scheme of ^{126}Sn via ^{126}Sb to ^{126}Te [2].

2.2 Efficiency calibration

As shown in Fig. 1 of the decay scheme of ^{126}Sn [2], there are coincidence summing effects on the measured full energy peak count rates of 666.3 keV γ rays caused by cascade transitions 415 keV, 695 keV, 697 keV, 721 keV γ rays, etc. In order to obtain the emission rate of 666.3 keV γ rays (i.e. the activity of ^{126}Sb and ^{126}Sn), both the intrinsic full-energy peak efficiency of 666.3 keV γ rays at the sample measurement position and the summing effect corrections should be measured with sufficient accuracy.

The energy of 661.6 keV γ rays of ^{137}Cs is close to that of 666.3 keV γ rays of ^{126}Sb , so we can deduce the full energy peak efficiency of 666.3 keV γ rays from that of 661.6 keV γ rays. Since the activities of three available standard ^{137}Cs sources are too strong to be directly counted at the sample measurement position (2.5 mm above the detector surface), a weaker ^{137}Cs source was made and standardized by comparison measurements with the three standard Cs sources at a counting position of 160 mm above the detector surface. The results are shown in Table 1.

Table 1. The results of the weaker source activity calibrated by three Cs standard sources.

source	A_{stan}/ kBq	count rate (stan)/s^{-1}	A_{Weaker}/ kBq	count rate (weaker)/s^{-1}
Cs-1	180.3±1.2	288.9±1.9	24.42±0.17	
Cs-2	81.77±0.82	131.2±1.3	24.40±0.24	39.14±0.31
Cs-3	80.47±0.80	128.9±1.3	24.44±0.24	

The evaluated activity of the weaker Cs source is 24420 Bq with a relative standard uncertainty of 0.49%. The count rate of full-energy at the sample measurement position is 993.5 s^{-1}, and the full-energy peak efficiency of the 661.6 keV γ-ray, $\varepsilon_p(661)$, is 4.781% (as calculated by Eq. (1)) with a relative standard uncertainty of 0.57%.

$$\varepsilon_p(661) = \frac{CPS(661)}{\gamma PS(661)}, \qquad (1)$$

where $CPS(661)$ and $\gamma PS(661)$ stand for peak counts per second and γ rays per second for 661.6 keV γ-rays. The full-energy peak efficiency of 666.3 keV γ-rays at the sample measurement point can be deduced from that of 661.6 keV γ-rays and the efficiency curve. The full-energy peak efficiency curve without coincidence summing effects can be measured by standard multiple γ emitters at counting positions far enough (e.g., 160 mm) from the detector. The peak efficiency curve at the sample counting position (2.5 mm) can be obtained by the transformation from that of a "far" counting position using the "Effective Interaction Depth (EID)" law [3]. The resulting peak efficiency curves at the sample counting position are expressed by Eq. (2) and (3):

$$\ln\varepsilon_p = -12.818489 + 4.54126\ln E - 0.47508\ln^2 E \quad E \leqslant 200 \text{ keV}, \qquad (2)$$

$$\ln\varepsilon_{\rm p} = 1.99273583 - 0.75511\ln E - 0.00306\ln^2 E \quad E > 200\ \text{keV}, \tag{3}$$

The intrinsic full peak efficiency of 666.3 keV, $\varepsilon_{\rm p}(666)$, was calculated using formula (3). The result is 4.754% with a relative standard uncertainty of 1.2% derived from the uncertainties of the peak efficiency curve and the full peak efficiency of 661.6 keV.

The total efficiencies of 415 keV, 695 keV, 697 keV and 721 keV γ-rays should be obtained in order to correct for the summing effect on 666.3 keV γ-rays. At first the total efficiency of 661.6 keV γ-rays was measured and calculated based on the spectrum of the weaker Cs source using Eq. (4):

$$\varepsilon_{\rm T}(661) = \frac{C_{\rm t}PS(661)}{\gamma PS(661)}, \tag{4}$$

where $C_{\rm t}PS(661)$ is the total count rate of 661.6 keV γ-rays and $\gamma PS(661)$ is defined as in Eq. (1). According to the equation, the value of $\varepsilon_{\rm T}(661)$ is 17.55% with a relative standard uncertainty of 0.55%.

A set of single-energy γ-ray emitters, viz. ^{241}Am, ^{141}Ce, ^{51}Cr, ^{198}Au, ^{137}Cs and ^{54}Mn, were measured, and the P/T (peak-to-total ratio of γ spectrum) values determined for 59.4, 145.4, 320.1, 411.8, 661.7 and 834.8 keV γ-rays, respectively. From these P/T values, the total efficiencies $\varepsilon_{\rm T}$ can be easily obtained by $\varepsilon_{\rm T} = \varepsilon_{\rm p}/(P/T)$.

$$\ln\varepsilon_{\rm T} = -3.54504 + 0.75336\ln E - 0.07102\ln^2 E \quad E \leqslant 200\ \text{keV}, \tag{5}$$

$$\ln\varepsilon_{\rm T} = -1.91326 + 0.34476\ln E - 0.04914\ln^2 E \quad E > 200\ \text{keV}, \tag{6}$$

The total efficiency curves can be expressed by Eqs. (5) and (6). According to the equations, the values of $\varepsilon_{\rm T}(415)$, $\varepsilon_{\rm T}(695)$, $\varepsilon_{\rm T}(697)$, $\varepsilon_{\rm T}(721)$ are 19.78%, 17.18%, 17.17% and 16.99%, respectively. The relative standard uncertainty is 1.5% derived from the uncertainty of the total efficiency curve.

2.3 Determination of the activity and atomic number of ^{126}Sn

Before the measurement of the ^{126}Sn sample, a background γ spectrum was measured as shown in Fig. 2. There is a ^{137}Cs peak (661 keV) in the spectrum. Thanks to the high resolution of the detector (1.83 keV for 1.33 MeV peak), the ^{137}Cs peak does not interfere with the measurement of the 666.3 keV peak.

The sample in a glass container was placed at the position 2.5 mm from the surface of the Ge detector aluminum case, and measured for 68 h. The γ spectrum obtained is shown in Fig. 3.

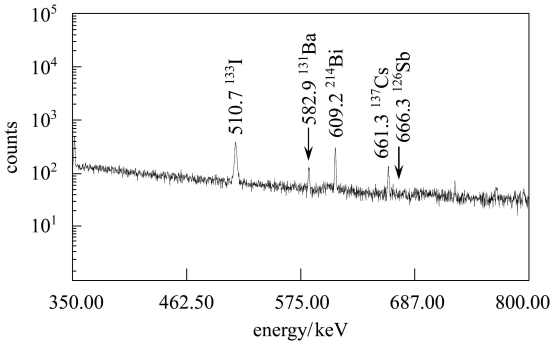

Fig. 2. A background γ spectrum.

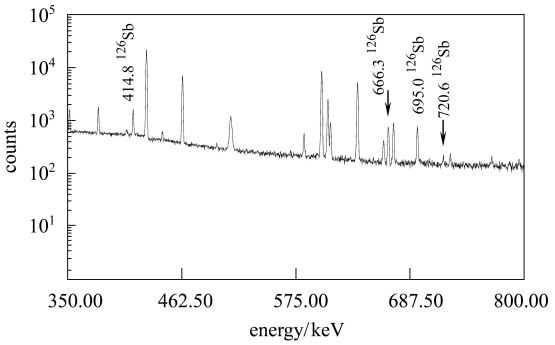

Fig. 3. A γ spectrum of a ^{126}Sn sample.

As can be seen from Fig. 3, the peak of 666.3 keV is a single peak with no interference. The peak count rate is (0.0144±0.0003) s^{-1}. According to the decay scheme of ^{126}Sn via ^{126}Sb to ^{126}Te, 666.3 keV γ-ray is in cascade with 415 keV, 695 keV, 697 keV and 721 keV γ-rays. The actual full peak efficiency (after summing effect corrections) of 666.3 keV, $\varepsilon'_{\rm p}(666.3)$, can be obtained by Eq. (7) [3]:

$$\begin{aligned}\varepsilon'_{\rm p}(666.3) = {} & \varepsilon_{\rm p}(666.3)\cdot\left[1-\frac{P(695)}{P(666)}\varepsilon_{\rm T}(695)\right]\cdot\left[1-\frac{P(415)}{P(666)}\varepsilon_{\rm T}(415)\right]\cdot\left[1-\frac{P(721)}{P(666)}\varepsilon_{\rm T}(721)\right]\times \\ & \left[1-\frac{P(697)}{P(666)}\varepsilon_{\rm T}(697)\right]\cdot\left[1+\frac{P(695)}{P(666)}\varepsilon_{\rm T}(695)\frac{P(415)}{P(666)}\varepsilon_{\rm T}(415)\right]\cdot\left[1+\frac{P(695)}{P(666)}\varepsilon_{\rm T}(695)\frac{P(721)}{P(666)}(721)\right]\times \\ & \left[1+\frac{P(695)}{P(666)}\varepsilon_{\rm T}(695)\frac{P(697)}{P(666)}\varepsilon_{\rm T}(697)\right]\cdot\left[1+\frac{P(415)}{P(666)}\varepsilon_{\rm T}(415)\frac{P(721)}{P(666)}\varepsilon_{\rm T}(721)\right]\times \\ & \left[1+\frac{P(415)}{P(666)}\varepsilon_{\rm T}(415)\frac{P(697)}{P(666)}\varepsilon_{\rm T}(697)\right]\cdot\left[1+\frac{P(721)}{P(666)}\varepsilon_{\rm T}(721)\frac{P(697)}{P(666)}\varepsilon_{\rm T}(697)\right],\end{aligned} \tag{7}$$

where P stands for branching ratio and the number in parentheses is the energy of the corresponding γ ray. The value of $\varepsilon'_{\rm p}(666.3)$ is 3.261% with a relative standard uncertainty of 1.3% derived from the uncertainties in the intrinsic peak efficiency of 666.3 keV γ-rays and the total efficiencies of the 415 keV, 695 keV, 697 keV and 721 keV γ-rays. The apparent emission rate of 666.3 keV rays from sample 1, $rps(666.3)$, is calculated by Eq. (8), to be 0.441 s^{-1}.

$$rps(666.3) = Cps(666.3)/\varepsilon'_{\rm p}(666.3). \quad (8)$$

After corrections for the self-absorption correction factor $C_1 = 0.975$ and the glass absorption correction factor $C_2 = 0.993$, the real emission rate of 666.3 keV rays, i.e., the activity of ^{126}Sn in sample 1, is calculated to be (0.456±0.013) Bq, as given in Eq. (9).

$$A_{126\rm Sn} = rps(666.3) \times C_1 \times C_2\,. \quad (9)$$

Four literature values of the ^{126}Sn half-life measured in the past 10 years are listed in Table 2. The evaluated value $T_{1/2}$=(2.34±0.05)×10^5 was obtained by Eq. (10). In this paper, the number of ^{126}Sn atoms was calculated using this value of the half-life.

$$\overline{T}_{1/2} = \frac{\sum_i^N T_{1/2i}/\sigma_i^2}{\sum_i^N 1/\sigma_i^2}, \quad \overline{\sigma} = \frac{1}{\sqrt{\sum_i^N 1/\sigma_i^2}}. \quad (10)$$

Table 2. Half-life data of ^{126}Sn.

investigator	reported	$T_{1/2}$/a	error(%, 1σ)
Zhang [4]	1996	2.47×10^5	8.0
Haas [5]	1996	2.07×10^5	10.1
Oberli [1]	1999	2.35×10^5	3.0
Catlow [6]	2003	2.33×10^5	4.1

The number of ^{126}Sn atoms was obtained by the evaluated half-life and the activity of ^{126}Sn using the equation $N_{126} = AT_{1/2}/\ln 2$; the value is $N = (4.85 \pm 0.17) \times 10^{12}$.

3 Results

In this paper, three ^{126}Sn samples were measured by using HPGe γ-ray spectrometry. Details of the measurements and data reductions are given in previous sections with sample 1 as an example (the same procedures were used for the other two samples). The activity of ^{126}Sn was determined by measuring the count rate of the 666.3 keV peak (in combination with the actual peak efficiency $\varepsilon'_{\rm p}$). The number of ^{126}Sn atoms was obtained by the evaluated half-life and the activity of ^{126}Sn. The P ratio of ^{126}Sn to Sn was calculated based on chemical stoichiometry. The activity, the number of atoms and the ratio of ^{126}Sn to Sn in the three $^{126}SnO_2$ samples are listed in Table 3 and uncertainties are summarized in Table 4.

Table 3. Activity, number of atoms and the ratio of ^{126}Sn to Sn in three $^{126}SnO_2$ samples.

samples	activity of ^{126}Sn/Bq	number of ^{126}Sn atoms	^{126}Sn/Sn
1# ^{126}Sn	0.456±0.013	(4.85±0.17)×10^{12}	(1.033±0.037)×10^{-8}
2# ^{126}Sn	0.279±0.013	(2.97±0.15)×10^{12}	(8.31±0.43)×10^{-9}
3# ^{126}Sn	0.133±0.007	(1.42±0.09)×10^{12}	(4.67±0.29)×10^{-9}

Note: All numbers after "±" stand for standard uncertainty.

Table 4. The main sources of uncertainty in the measurement of the ^{126}Sn activity concentration.

source of uncertainty	uncertainty (%, 1σ)
activity of ^{137}Cs	0.57
activity of ^{126}Sn–^{126}Sb	2.5
the full energy peak efficiency	1.2
correction for γ-ray attenuation	0.3
the value of half-life	2.2

4 Discussion

The three available standard ^{137}Cs sources were originally used for detector performance tests, and were usually counted at a position 250 mm from the surface of the Ge detector. The activities of these sources were so high that a considerable (and hardly quantifiable) dead-time effect could occur when they were counted at the position of 2.5 mm above the detector surface (the sample measurement position). So a weaker source of ^{137}Cs was made and calibrated by the three standard ^{137}Cs sources at a distance of 160 mm from the surface of the detector, and used for the determination of the full energy peak efficiency and the total efficiency of the 661.6 keV γ-rays at the sample measurement position (rack height of 2.5 mm). The additional uncertainty induced by this transition is very small and quantifiable compared with the direct use of the strong standard sources in

the calibration. The internal conversion coefficients of the 666.3, 695, 415 keV γ-rays related to the coincidence summing are 0.004, 0.003, 0.01 and those of the 721 and 697 keV γ-rays are negligible. Since the maximum correction introduced from internal conversion is less than 0.1%, this correction item is not applied in our calculations.

Table 5. The correction factors for the emission rate of the 666.3 keV γ-rays in the SnO_2 sample and a glass container.

material	μ/mm^{-1} [8]	D/mm	C
SnO_2	2.52×10^{-2}	1	0.975
glass (SiO_2)	1.32×10^{-2}	0.5	0.993

The attenuation of γ-rays at the container bottom was corrected by the equation: $I/I_0 = \mathrm{e}^{-\mu D}$, where μ is the attenuation coefficient, D is the material thickness, I and I_0 represent the intensity of γ-rays before and after the material, respectively. Assuming that the distribution of ^{126}Sn is homogeneous in the sample and the change of solid angle caused by sample thickness is ignored, the correction factor for self-absorption in the wafer-shaped sample can be calculated by the following equation:

$$\frac{I}{I_0} = \frac{1-\mathrm{e}^{-\mu D}}{\mu D}\,. \tag{11}$$

The equation is suitable for γ-rays with energies higher than 150 keV [7]. The value of total correction for 666.3 keV γ-rays attenuation is 2.5% and the uncertainty is less than 0.3%. The results for γ ray absorption corrections (C) are shown in Table 5. A total-attenuation correction factor of 0.968 was used in calculating the final results.

5 Conclusions

The ^{126}Sn/Sn values of three samples vary from 10^{-8} to 10^{-9}. One of the three samples (sample 1) was measured by AMS and the result ($(1.11\pm0.35)\times10^{-8}$) is consistent with the value obtained by γ-ray spectrometry ($(1.033\pm0.037)\times10^{-8}$) within uncertainty limits, which confirms our procedures in the measurement of ^{126}Sn by AMS. One of the major problems in our AMS measurement of ^{126}Sn is the lack of a calibration standard, as the content of the ^{126}Sn in the samples available is too low to be accurately determined by TIMS or ICPMS. The gamma ray spectrometric method established in this work provides an alternative for absolute calibration of the ratio of ^{126}Sn to Sn. The consistency of the result from this work and the value obtained by our absolute AMS measurement of ^{126}Sn (to be published) confirms the reliability of both methods, and lays a foundation for the AMS measurement of ^{126}Sn at much lower levels.

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