Half-life of ^{151}Sm remeasured

Ming He,* Hongtao Shen,† Guozhu Shi, Xinyi Yin, Weizhi Tian, and Shan Jiang
Department of Nuclear Physics, China Institute of Atomic Energy, Post Office Box 275-50, Beijing 102413, People's Republic of China
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The half-life of ^{151}Sm was redetermined and compared with literature values. A $^{151}Sm_2O_3$ sample was produced by exposing $^{150}Sm_2O_3$ to the high neutron flux of the heavy water research reactor at the China Institute of Atomic Energy (CIAE). The number of atoms and the activity of ^{151}Sm in the sample were measured by thermal ionization mass spectrometry (TIMS) and liquid scintillation counting (LSC), respectively. The half-life of ^{151}Sm determined in this work is 96.6 yr, with a standard uncertainty of 2.4 yr based on a quadratic summation of the uncertainty components from the measurements of the number of atoms and the activity of ^{151}Sm.

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I. INTRODUCTION

Samarium is a rare earth element. It has been widely used in many fields, for example, as a neutron absorber in nuclear reactors and special magnets. It is a long-lived fission product with a fission yield of ~0.4%. Samarium-{151} can be used as a tracer in the environmental and life sciences. It is also important in astrophysics. Samarium-{151} is one of the key nuclides for characterizing nucleosynthesis in the slow neutron capture process and is an excellent thermometer for the *s* process [1]. The half-life of ^{151}Sm is one of the basic parameters for these applications. However, the half-life of ^{151}Sm has not been well established yet. Table I summarizes the half-life values of ^{151}Sm reported so far. The large uncertainties in most recent values hindered their use in all the aforementioned fields. In this work, the half-life of ^{151}Sm was remeasured in an effort to narrow its uncertainty.

For a long-lived nuclide, the half-life value, T, is usually calculated from the relationship between the radioactivity, A, and the number of atoms of the nuclide, N, that is, $A = N \cdot (\ln 2)/T$. In the present work the half-life of ^{151}Sm was remeasured by determining the number of ^{151}Sm atoms with thermal ionization mass spectrometry (TIMS) and the activity with liquid scintillation counting (LSC).

II. EXPERIMENTAL

A. Production of ^{151}Sm nuclide.

About 50 mg Sm_2O_3, enriched in ^{150}Sm to 87.27%, was exposed to the high neutron flux of the heavy water research reactor at the China Institute of Atomic Energy (CIAE) for about 15 days. The neutron flux was 4.5×10^{13} $n/\mathrm{cm}^2 \cdot \mathrm{s}$ at the sample irradiation site. Samarium-{151} was produced through a (n,γ) reaction. At the same time, a substantial amount of ^{151}Sm was burned up through a $^{151}\mathrm{Sm}(n,\gamma)$ reaction due to the very large cross section of $^{151}\mathrm{Sm}(n,\gamma)^{152}\mathrm{Sm}$ (15 200 b). The ratio of $^{151}\mathrm{Sm}/^{150}\mathrm{Sm}$ was estimated with the Eq. (1) to be about 3.69×10^{-3}.

$$N_{151}/N_{150} = \frac{\phi\,\sigma_1}{\lambda + \phi\,\sigma_2}\{1 - \exp[-(\lambda + \phi\,\sigma_2)\,t_1]\}\exp(-\lambda\,t_2), \quad (1)$$

where φ is the neutron flux at the sample irradiation site, σ_1 and σ_2 are the cross sections for $^{150}\mathrm{Sm}(n,\gamma)^{151}\mathrm{Sm}$ and $^{151}\mathrm{Sm}(n,\gamma)^{152}\mathrm{Sm}$, respectively, $\lambda = 2.4 \times 10^{-10}$ is the decay constant of ^{151}Sm (based on $T_{1/2} = 90$ yr), t_1 is the sample irradiation time, and t_2 is the cooling time. In this equation, λ is negligible compared with $\varphi\sigma_2$. Cross sections of 104 b and 15 200 b [7] were used for σ_1 and σ_2, respectively.

B. Measurement of the number of ^{151}Sm atoms

After a cooling time of more than 5 yr, the irradiated sample was dissolved with 68% HNO_3. The solution was heated on a hot plate and evaporated to near dryness; then 20 ml 0.01 M HNO_3 was added to produce the ^{151}Sm sample solution. Then the sample was measured by a Finnigan MAT 262 TIMS. Nine runs were performed and the results of the abundances of Sm isotopes in the samples before and after irradiation are shown in Table II. The abundance values and uncertainties are based on the averages and the relative standard deviations of the nine runs, respectively. A natural Sm sample and an un-irradiated enriched ^{150}Sm sample were also measured to check the reliability of the measurements. The results of isotopic abundances in the natural Sm sample show that the measured values are in good agreement with literature values [7] (Table III). The ratio of $^{151}\mathrm{Sm}/^{150}\mathrm{Sm}$ in the irradiated sample is $(3.794 \pm 0.002) \times 10^{-3}$, which is quite close to the calculated one (3.69×10^{-3}).

The ratio of $^{151}\mathrm{Eu}/^{150}\mathrm{Sm}$ in the un-irradiated sample is less than 4×10^{-6}, as shown in Table II. To confirm this result from TIMS, a γ-ray spectrum of the irradiated sample for 21.9-hr measurement was obtained by using HPGe γ-ray spectrometry, as shown in Fig. 1. The number of ^{152}Eu atoms was obtained using the peak areas of 244.8-, 344.3-, 1004.9-, and 1112.2-keV γ-ray peaks combined with their branching rations, peak efficiencies, and the half-life of ^{152}Eu. The ^{151}Eu atom number in the un-irradiated sample was calculated by using the equation $N151 = N152/\sigma\varphi t$, where σ is the cross

*minghe@ciae.ac.cn
†shenht@ciae.ac.cn

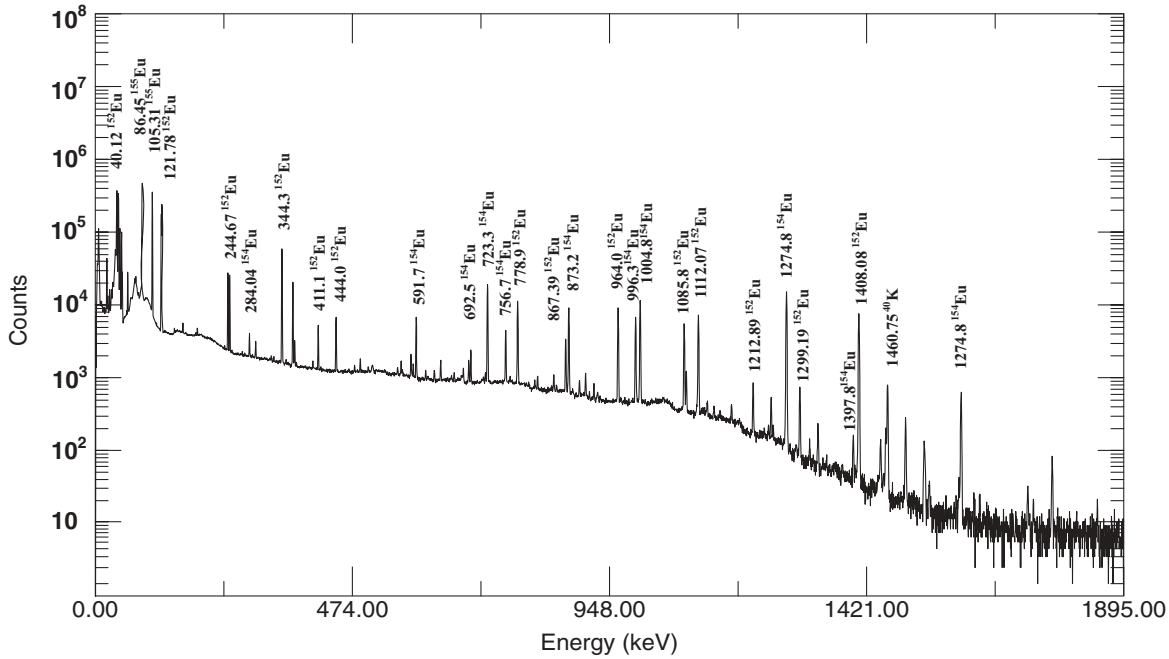

FIG. 1. A γ-ray spectrum of a ^{151}Sm sample.

TABLE I. Summary of experimental values for the half-life of ^{151}Sm.

Investigator	Reported	Half-life (yr)	Relative Uncertainty (%, 1σ)
Inghram *et al.* [2]	1950	122	–
Karraker *et al.* [3]	1952	73	33
Melaika *et al.* [4]	1955	93	–
Flynn *et al.* [5]	1965	87	10
Reynolds *et al.* [6]	1968	93	8.6
This work	2009	96.6	2.5

TABLE II. Abundances of Sm isotopes in Sm samples (enriched in ^{150}Sm) before and after neutron irradiation.

Sample isotope	Sm enriched in un-irradiated ^{150}Sm, Abundance (%)	Sm enriched in ^{150}Sm, after irradiation Abundance (%)	Rel. std. unc. (%)
^{144}Sm	0.18	0.1853	0.20
^{147}Sm	1.96	1.953	0.11
^{148}Sm	1.30	1.300	0.11
^{149}Sm	3.87	1.316	0.11
^{150}Sm	87.34	89.58	0.15
^{151}Sm(+^{151}Eu)	<0.0004	0.3359	0.15
^{152}Sm	3.83	3.835	0.11
^{154}Sm	1.51	1.503	0.12

section of ^{151}Eu$(n,\gamma)^{152}$Eu, t is the sample irradiation time, and φ is defined as in Eq. (1), N151 and N152 are the numbers of atoms for ^{151}Eu and ^{152}Eu, respectively. The ratio of ^{151}Eu/^{150}Sm in the un-irradiated sample was calculated to be $(1.20 \pm 0.14) \times 10^{-6}$. The consistency of the result from TIMS and γ-ray spectrometry methods confirms that the ^{151}Eu/^{150}Sm ratio in the un-irradiated sample can be neglected compared to the ^{151}Sm/^{150}Sm ($\sim 10^{-3}$) ratio in the irradiated sample.

C. Activity measurement of ^{151}Sm

Samarium-{151} is essentially a pure β-minus decay nuclide with a very weak β branch (0.91%) feeding the first excited state of ^{151}Eu at 21.54 keV. The activity of ^{151}Sm was measured by LSC. The main difficulty in LSC is the discrimination between the nuclide of interest, ^{151}Sm, and the radioactive contaminants present in the sample solution that can induce complex overlapping spectra and quenching effects. As can be seen from the γ-ray spectrum of the ^{151}Sm sample (shown in Fig. 1), ^{152}Eu and ^{154}Eu are the main

TABLE III. Measured and literature values for isotopic abundances of Sm in natural Sm samples.

Isotope	144	147	148	149	150	152	154
Literature abundance (%)	3.07	14.99	11.24	13.82	7.38	26.75	22.75
Measured abundance (%)	3.095	15.03	11.26	13.83	7.379	26.71	22.69

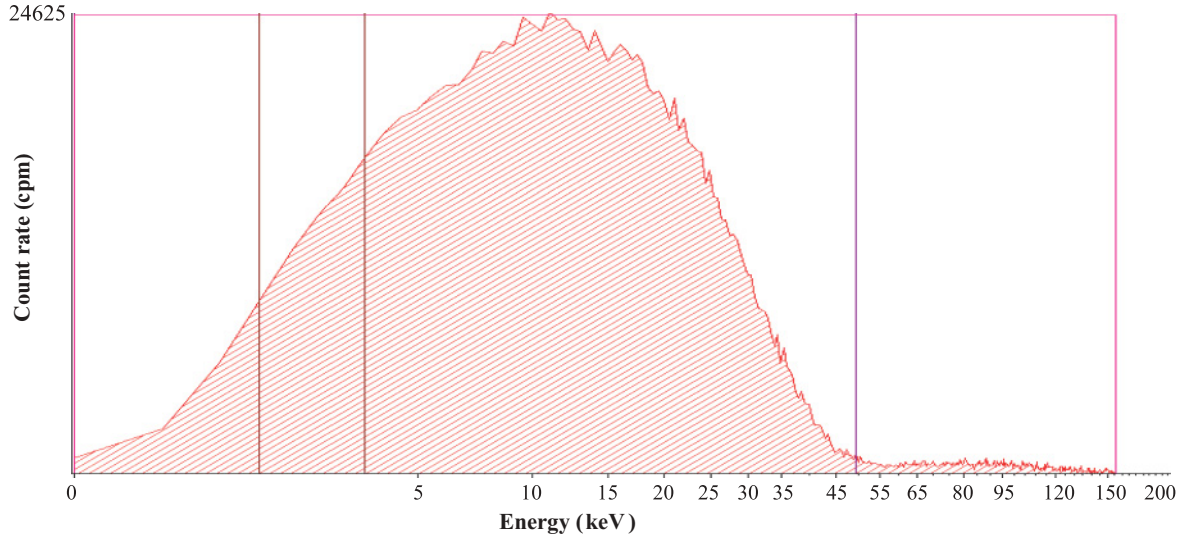

FIG. 2. (Color online) A LSC spectrum of the radiated sample (log-log coordinate.)

γ-emitting contaminants. The activity of ^{152}Eu in a 1-mg Sm sample, $A(^{152}\mathrm{Eu})$, was calculated using Eq. (2) to be 3.74×10^3 Bq, with a relative standard uncertainty of 1.3% mainly derived from the uncertainties of the peak efficiency curve.

$$A(^{152}\mathrm{Eu}) = \mathrm{Cps}(344.3)/[\varepsilon(344.3) \cdot B(344.3)], \qquad (2)$$

where Cps(344.3), $\varepsilon(344.3)$, and $B(344.3)$ stand for peak counts per second, peak efficiency, and branching ratio of a 344.3-keV γ ray, respectively. The activity of ^{154}Eu was calculated using the same method to be 4.18×10^3 Bq/mg Sm sample, with a relative standard uncertainty of 1.3%. Several other γ rays of these two nuclides (such as 244.7-, 1085.8-, 1112.1-keV γ rays of ^{152}Eu and 591.7-, 723.3-, and 1004.8-keV γ rays of ^{154}Eu) were also used in calculating the activities of ^{152}Eu and ^{154}Eu, and the results are consistent with the previous results within uncertainty limits, which confirms that the total activity of ^{152}Eu and ^{154}Eu was less than 8×10^3 Bq/mg Sm sample and can be neglected compared to the activity of the ^{151}Sm in the sample ($\sim 3 \times 10^6$ Bq/mg Sm sample). Furthermore, the end-point energy for ^{151}Sm β decay is 76.7 keV, while that for ^{152}Eu and ^{154}Eu β decay is about 500 keV; therefore the contribution from $^{152+154}$Eu to the energy region of ^{151}Sm in LSC spectrum will be even smaller.

A model 3170 TR/SC Tri-Carb low-background LSC spectrometer with double photomultiplier tubes was used to measure the activity of ^{151}Sm. The detection efficiency is a function of the emission spectrum of the radionuclide and the light yield of the scintillator, which depends on the LSC cocktail itself but also on the chemical composition of the source. The LSC was calibrated by an efficiency tracing method using a ^{14}C standard source with the same chemical and physical parameters (i.e., volume, source to cocktail volume ratio, acidity concentration) and quenching agent (nitromethane) as for the ^{151}Sm sample. A blank source containing natural Sm was prepared, using the same LSC cocktail, and measured. A ^{151}Sm source containing 26.1 ml of sample solution was measured. A LSC spectrum is shown in Fig. 2. Taking into account the detection efficiency, the activity of ^{151}Sm in the solution was (626.8 ± 13.9) Bq/μl. The concentration of Sm in the sample solution, measured with ICP-MS, was $(2.044 \pm 0.031) \times 10^{-4}$ g/ml. The specific activity of ^{151}Sm in the sample solution is therefore $(3.067 \pm 0.076) \times 10^6$ Bq/mg Sm. The uncertainty budget is given in Table IV.

III. HALF-LIFE DETERMINATION

The ^{151}Sm half-life was calculated from the number of ^{151}Sm atoms (N) and its activity (A) by Eq. (3):

$$T_{1/2} = (N\ln 2)/A, \quad \text{and} \quad N = (N_0 C_{151})/M, \qquad (3)$$

where $T_{1/2}$ is the half-life of ^{151}Sm, N_0 ($=6.0221 \times 10^{23}$) is Avogadro constant, M ($=150.044$) is the average atomic mass of the irradiated Sm sample measured with TIMS as shown in Table II, C_{151} ($=(3.359 \pm 0.005) \times 10^{-3}$) is the ^{151}Sm abundance in the irradiated Sm sample (Table II), N ($=(1.348 \pm 0.002) \times 10^{-3}$) is the number of ^{151}Sm atoms in 1 mg of irradiated Sm sample, A ($=(3.067 \pm 0.076) \times 10^6$ Bq)

TABLE IV. Uncertainty budget in liquid scintillation counting.

Uncertainty component	Relative standard uncertainty (%)
Impurities	2.3
Detection efficiency for ^{151}Sm	1.0
Counting statistics	0.2
Sample weighing	0.1
Total	2.5

is the activity of ^{151}Sm in 1 mg of irradiated Sm sample. Using these values, the half-life of ^{151}Sm with its associated standard uncertainty is (96.6 ± 2.4) yr.

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