Determination of the cross section for ^{159}Tb(n,2n)^{158}Tb and the half-life of ^{158}Tb

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(Received 18 May 1984)

The half-life of ^{158}Tb was determined to be 180 yr using a combination of isotope dilution mass spectrometry and radioactivity counting techniques. The total uncertainty in this value is 6%. Using the newly measured half-life for ^{158}Tb, the cross section for the ^{159}Tb(n,2n)^{158}Tb reaction at 14.8 MeV was determined to be 1.93 b with an uncertainty of 7%.

INTRODUCTION

To measure the cross section for the reaction ^{159}Tb(n,2n)^{158}Tb, a target consisting of a mixture of the dried hydroxides of ^{159}Tb and ^{169}Tm was irradiated with 14.8-MeV neutrons at the RTNS-1 facility at the Lawrence Livermore National Laboratory. The fraction of target atoms (N_T) converted (FC) to (n,2n) product atoms (N_P) by the irradiation is the product of the neutron fluence and the cross section,

$$\mathrm{FC}=\frac{N_P}{N_T}=(\text{dis rate}/\lambda)_P/N_T=\sigma\phi\ . \tag{1}$$

The fraction converted (FC) is determined by measuring the specific activity (disintegration rate per unit mass) of a purified sample.

Since the (n,2n) cross section for ^{169}Tm is known and the fluence of neutrons is identical for the two nuclides, the cross section can be determined by measuring the fraction converted for each of the rare earth nuclides in the irradiated target and calculating the desired result using Eq. (2):

$$\sigma_{^{159}\mathrm{Tb}}=\frac{\mathrm{FC}_{\mathrm{Tb}}}{\mathrm{FC}_{\mathrm{Tm}}}\sigma_{^{169}\mathrm{Tm}}\ . \tag{2}$$

Obviously, it is critical to have an accurate value of the half-life of the product nuclide. Therefore, the half-life of ^{158}Tb was measured to verify the accuracy of the value currently in the literature.[2,3]

^{158}Tb HALF-LIFE DETERMINATION

Long term irradiations of tantalum targets by high energy protons at the Los Alamos Meson Physics Facility (LAMPF) provide sufficient quantities of neutron deficient rare earth elements (REE) to measure their properties. Terbium was extracted from two tantalum targets, one irradiated in 1977, hereafter referred to as sample 1, and another irradiated in 1980, sample 2. The targets were dissolved in a hot cell, the REE extracted as a group, and terbium isolated from the others by selective high-performance liquid chromatography (HPLC) with α-hydroxyisobutyric acid (α-HIB) on a cation exchange column of 25—43 μm Aminex 50WX12 (Bio-rad). The terbium fraction was taken from the hot cell and again selectively eluted using HPLC to optimize the separation of terbium from gadolinium, the most insidious source of isobaric interference to the mass spectrometric measurements. Master solutions were prepared from the products of these chromatographic separations. Subsequent measurements were made on quantitative aliquots of the master solutions to determine the ^{158}Tb half-life. Isotope dilution mass spectrometry was used to measure the terbium isotope abundances in the solutions. Radioactivity counting techniques were used to measure the decay rate in the same solutions. Once these two quantities are known it is a simple matter to calculate, via the radioactive decay equation, the half-life for the nuclide of interest.

Terbium isotope concentrations

Relative abundances of terbium isotopes were determined by measuring isotope ratios using an NBS type of 30.5 cm radius of curvature solid source mass spectrometer with a Faraday cup detector. No corrections were made for mass fractionation by the thermal ionization source in the instrument. Typically such corrections are on the order of 0.1% per mass unit for rare earth elements.

Isotopes of gadolinium will perturb the determination of terbium isotopic abundances. To monitor the extent of such interferences, the ratio of the signal at mass 156, the first gadolinium isotope with no long-lived terbium isobar, to that at mass 159, the terbium isotope with no isobar of gadolinium, was measured. The ratios 156/159 were all of the order of 5×10^{-6}, values so small that a special mass spectrometer was required to make the measurement. Interferences from gadolinium were of no consequence to the determination of the terbium isotopic composition of the master solutions. Relative isotopic abundances in the two samples are presented in Table I.

TABLE I. Isotopic composition of terbium.

	Isotope abundance (at. %)		
	^{157}Tb	^{158}Tb	^{159}Tb
Sample 1	48.6±0.04	0.091±0.002	51.3±0.04
Sample 2	56.7	0.150	43.2

The abundance of ^{158}Tb is lower than that of ^{157}Tb and ^{159}Tb because of the shielding by stable ^{158}Dy from decay of the higher-Z primary spallation products.

Stated uncertainties are the standard deviations on eight measurements of ^{157}Tb/^{159}Tb and five measurements of ^{158}Tb/^{159}Tb. They are presented to demonstrate the reproducibility of the measurements and do not necessarily reflect their accuracy.

Absolute isotope abundances were determined by isotope dilution mass spectrometry using carefully prepared gravimetric standards of monoisotopic ^{159}Tb. One such standard solution was prepared from high purity terbium metal obtained from the Ames Laboratory at Iowa State University. The other was made from Johnson and Mathey Spec-Pure Tb_4O_7 dried to constant weight at 800 °C. A quantitative aliquot of each of these solutions was mixed with a quantitative aliquot from each of the two master solutions. Isotopic ratios of ^{159}Tb/^{157}Tb were measured on the master solutions (R_x) and on the mixes of standard and master (R_{mix}). The absolute abundance of ^{157}Tb was calculated from the isotope dilution equation shown in Eq. (3):

$$^{157}\text{Tb}_x(\text{moles/g solution}) = \frac{^{159}\text{Tb}_{\text{std}}}{[R_{\text{mix}} - R_x]\text{mass}_x} . \quad (3)$$

x refers to the master solution; $^{159}\text{Tb}_{\text{std}}$ refers to the moles of this isotope added from the standard solution. Abundances of the other isotopes were calculated from their proportions relative to ^{157}Tb as was shown in Table I. The absolute abundances are presented in Table II, the uncertainty is the standard deviation of four measurements on solution 1 and five measurements of solution 2. No bias could be determined between results obtained from the mixes with different standard solutions. It is estimated that the overall uncertainty in the measurement for the abundances of the major isotopes ^{157}Tb and ^{159}Tb is about 0.5% and about 1% for the minor isotope ^{158}Tb.

Disintegration rate

^{158}Tb disintegration rates were determined by gamma ray spectrometry on aliquots of the solutions. Each aliquot was counted several times on each of four different Ge(Li) detectors. Count rates were converted to disintegration rates using 11 γ rays with energies between 80 and 1187 keV. Branching ratios for the γ rays were taken from Ref. 1. For each count the disintegration rate is determined from the average of the result of each peak weighted by its branching ratio. Unfortunately the major isotope ^{157}Tb emits no measurable γ rays and its disintegration rate cannot be determined by this method. The results of the gamma-ray spectrometry measurements are presented in Table III.

TABLE II. Terbium isotope abundance in master solutions.

	Isotope concentration (nanomoles/g of solution)		
	^{157}Tb	^{158}Tb	^{159}Tb
Sample 1	1.82±0.006	0.00341	1.92
Sample 2	1.83±0.003	0.00483	1.39

TABLE III. ^{158}Tb disintegration rate (dis/min g of solution).

Sample 1	$1.50\times10^4\pm0.01$
Sample 2	$2.13\times10^4\pm0.02$

The uncertainties are the standard deviations of the average of 30 measurements of two aliquots of sample 1 on four different spectrometers and ten measurements of a single aliquot of sample 2 on four different spectrometers.

Conclusion

The half-life of ^{158}Tb calculated from measurements on both sample solutions is 180 y. We have arbitrarily assigned an uncertainty of 3% to the measurement of this value. This estimate is considered to be conservative and is based upon a consideration of the imprecision of multiple measurements as expressed in the tables, and systematic uncertainties in the concentration of the primary mass spectrometry standards, in the calibration of the Ge(Li) detectors, and the failure to correct for mass fractionation by thermal ionization in the mass spectrometers. There is an additional 5% uncertainty in the γ-ray branching ratios[1] used to determine the ^{158}Tb disintegration rate. The overall uncertainty in the value is then considered to be 6%. Our measured value is larger than 150 y, the half-life currently used for ^{158}Tb (Refs. 2 and 3), but at the upper limit of the uncertainty associated with that result. The previous result was obtained using a combination of nuclear counting techniques and mass spectrometry;[3] similar, in principal, to the methods used in our work. Unlike our study, the authors of the previous work obtained the ^{158}Tb half-life by comparison with that of ^{160}Tb. They attributed the uncertainty in their result to uncertainties in the decay scheme of ^{160}Tb and analytical uncertainties associated with measuring the isotope ratio. Our result has no dependence upon a knowledge of the decay of ^{160}Tb. In addition, mass spectroscopy has improved such that our isotope abundance measurements are certainly more accurate and precise than the ones made 20 years ago.

CROSS SECTION MEASUREMENT

The target of dried, mixed hydroxides of ^{159}Tb and ^{169}Tm was dissolved in HCl following the irradiation with 14.8±0.3 MeV neutrons at the RTNS-1 facility. The Tb and Tm were separated by elution from a Dowex-50 cation-exchange column. The specific activities of ^{159}Tb

and ^{169}Tm in oxide samples were measured using several Ge(Li) detectors with source-detector distances of 8—10 cm.

Using the newly defined decay constant for ^{158}Tb, the fraction of ^{159}Tb converted was $(8.41\pm0.31)\times10^{-8}$, and for ^{169}Tm it was $(8.61\pm0.19)\times10^{-8}$. The cross section for the ^{169}Tm(n,2n)^{168}Tm reaction using 14.8 MeV neutrons is 1.98±0.04 b (Ref. 4). Using this value and the measured fractions converted, the ^{159}Tb(n,2n)^{158}Tb cross section using 14.8 MeV neutrons is calculated to be 1.93 using Eq. (2). The uncertainty in this calculation is 4%; considering the additional uncertainty of 5% in the ^{158}Tb branching ratio and the 2% uncertainty in the ^{169}Tm cross section, we estimate the overall uncertainty in the measurement to be 7%.

[1]M. A. Lee, Nucl. Data Sheets **31**, 381 (1980).

[2]*Table of Isotopes*, 7th ed., edited by C. M. Lederer and V. S. Shirley (Wiley, New York, 1978), p. 920.

[3]J. M. Prospero, Ph.D. thesis, No. PUC-1963-95, Princeton University, 1963.

[4]B. P. Bayhurst, J. S. Gilmore, R. J. Prestwood, J. B. Wilhelmy, N. Jarmie, B. H. Erkkila, and R. A. Hardekopf, Phys. Rev. C **12**, 451 (1975). The ^{169}Tm(n,2n)^{168}Tm cross section was corrected from 2.01 to 1.98 b after reevaluation of the counting data. Later measurements at both Livermore and Los Alamos confirm a value of 1.98±0.04 b for this cross section at neutron energies between 14 and 15 MeV.