Half-life of $^{92}Nb^{g}$

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We have produced the long-lived ground state of ^{92}Nb by irradiation of niobium foils with 14.8-MeV neutrons, and we have established its half-life from measurements of the disintegration rate and the neutron fluence together with the known $^{93}Nb(n,2n)^{92}Nb^{g}$ cross section. On the basis of two separate experiments we obtain $t_{1/2}(^{92}Nb^{g}) = (3.6 \pm 0.3) \times 10^{7}$ yr.

[RADIOACTIVITY $^{92}Nb^{g}$; measured $T_{1/2}$.]

INTRODUCTION

The first observation of the decay of $^{92}Nb^{g}$ was reported in 1972 by Knight,[1] who produced it by long irradiation of molybdenum in a reactor. On the basis of measurements of the disintegration rates of the $^{92}Nb^{g}$ and $^{94}Nb^{g}$ in a purified sample, of the known half-life of $^{94}Nb^{g}$, and by estimates of the relative production of the two nuclides by the (n,p) reaction, the half-life was estimated to be 1.7×10^{8} yr with an uncertainty of at least a factor of 2.[1,2] Discovery of the ^{92}Nb ground state helped resolve the long-standing puzzle of the apparent anomalously low $^{93}Nb(n,2n)$ cross section at 14 MeV; the 10.15-day ^{92}Nb, previously the only known radioactive species at this mass number, is produced in only about 40% of the $(n,2n)$ events.

More recently, Makino and Honda[3] have reported a value for the $^{92}Nb^{g}$ half-life of $(3.3 \pm 0.5) \times 10^{7}$ yr. Their method was similar to that used by Knight, except that they made a mass-spectrometric measurement of the $^{92}Nb/^{94}Nb$ ratio.

The possible importance of long-lived ^{92}Nb in geochemical and cosmochemical studies has been discussed (e.g., Refs. 1–3). However, the presently accepted half-life of approximately 3×10^{7} yr is sufficiently short to rule out the presence of appreciable primordial ^{92}Nb. Apt *et al.*[2] have reported the measurement of ^{92}Nb in natural niobium, but the amount found is undoubtedly the result of a relatively recent formation process.

We have produced two sources of $^{92}Nb^{g}$ by irradiating niobium with 14.8-MeV neutrons. Our determination of the half-life is based on the measurement of the $^{92}Nb^{g}$ disintegration rates in the two samples, and on the amounts of $^{92}Nb^{g}$ formed as calculated from the $(n,2n)$ cross section and measured neutron fluences.

For a neutron irradiation that is short in time compared to the half-life of the product nuclide, the total number of atoms formed is given by $N = N_0 \sigma \phi$, where N_0 is the number of target atoms, σ is the cross section, and ϕ is the total neutron fluence (n/cm^2). This expression can be rearranged to solve for the half-life of the product, $T_{1/2} = N_A \sigma \phi (\ln 2)/(W_M)(A/M)$, where N_A is Avogadro's constant, W_M is the molecular weight of the target material, and A/M is the disintegration rate per gram. For the irradiation of niobium metal the half-life is given by $T_{1/2}$ (yr) $= 8.54 \times 10^{15} \sigma \phi/(\text{dpm/g})$. [(dpm/g) is disintegrations per minute per gram.]

EXPERIMENTAL DETAILS

Production of the $^{92}Nb^{g}$ sources

Two sources of $^{92}Nb^{g}$ were prepared in separate irradiations at the Insulated Core Transformer-Rotating Target Neutron Source (ICT-RTNS) accelerator at LLL. Niobium foils were placed at 0° and very close to the source of neutrons produced by the reaction of a 400-keV deuteron beam on a rotating titanium tritide target.[4] The first $^{92}Nb^{g}$ sample was produced by irradiating an 0.140-mm-thick foil (79.6 mg) for a total of 37 h over a 3-day period. The neutron fluence in the foil was based on the production of the 10.15-day $^{92}Nb^{m}$. The second $^{92}Nb^{g}$ sample was produced by irradiating an 0.030-mm-thick foil (44.5 mg), which was held tightly between two 0.050-mm-thick stainless steel foils. The foil packet was left in place at the ICT-RTNS for 330 days, during which time it received intermittent irradiation as other researchers used the facility. The stainless steel contained 8.6% nickel, and the $^{60}Ni(n,p)^{60}Co$ reaction was used to measure the cumulative neutron fluence.

Neutron fluence measurement

For the short irradiation, the production of the 10.15-day $^{92}Nb^m$ in the niobium target served as a convenient internal monitor for the neutron fluence. The total production of $^{92}Nb^m$ was measured to be $(4.92 \pm 0.15) \times 10^{14}$ atoms/g Nb, using a Ge(Li) detector and the GAMANAL program.[5] The calculated fluence of 14.8-MeV neutrons in the target was $(1.64 \pm 0.07) \times 10^{17}$ n/cm^2, based on a cross section of 463 ± 14 mb to form $^{92}Nb^m$ (see Ref. 6).

For the long irradiation, the stainless steel foils covering the niobium sample were used to monitor the neutron fluence. The nickel content of the stainless steel was analyzed by atomic absorption spectrophotometry and 14-MeV neutron-activation analysis techniques and was found to be 0.086 ± 0.002. We used the ^{60}Co produced by the $^{60}Ni(n,p)$ reaction to measure the neutron fluence, since the long half-life of ^{60}Co (5.26 yr) makes this nuclide suitable for use in a long, intermittent irradiation. The ^{60}Co was measured with a Ge(Li) detector[5] to be $(8.91 \pm 0.27) \times 10^{13}$ and $(8.32 \pm 0.25) \times 10^{13}$ atoms/g steel in the front and back foils, respectively. The ^{60}Co data were extrapolated back to the midpoint of the irradiation, assuming the flux to be approximately constant with time. The cross section for the $^{60}Ni(n,p)$ reaction is 114 ± 6 mb at 14.8 MeV.[7] In addition, the $^{61}Ni(n,np)^{60}Co$ reaction, weighted by the relative isotopic abundances (0.011/0.261), contributes approximately 1.6 mb, producing a total cross section of about 116 mb. The calculated fluence in the niobium foil, averaged from the two steel foils, is $(3.23 \pm 0.20) \times 10^{18}$ n/cm^2. The 6.2% uncertainty (1σ) in the fluence was obtained by combining 5% for the cross section, 2% for the nickel content, and 3% for the ^{60}Co measurement. We also conclude from the measured cobalt content of the steel (0.09%), and from a measurement of the effective $^{59}Co(n,\gamma)^{60}Co$ cross section at the ICT-RTNS, that the contribution from this reaction is negligible.

$^{93}Nb(n,2n)^{92}Nb^g$ cross section

No direct measurements of the cross section to produce the ground state have been made previously. Thus, we must use the difference between the total $(n,2n)$ cross section and the cross section to produce the 10.15-day isomer. The measurement of the total $(n,2n)$ cross section is difficult to make; a measurement of the neutron emission (with a large liquid scintillator) rather than of the decay of a radioactive product is required. Several measurements have been reported for the total $(n,2n)$ cross section in the energy range from 9 to 17 MeV.[8-11] The results of these measurements

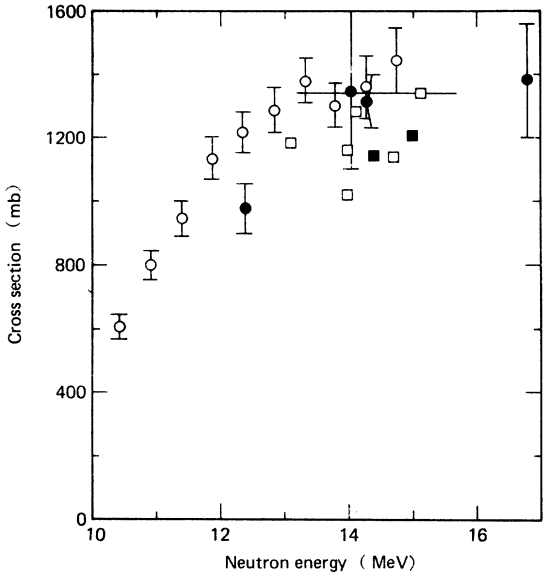

FIG. 1. The total cross section for the $^{93}Nb(n,2n)$ $^{92}Nb^{m+g}$ reaction in the 10- to 17-MeV energy range. Measured values are indicated by open circles (○) (Ref. 11) and by solid circles (●) (Refs. 8–10) with error bars. Estimates based on $(n,2n)$ reaction systematics are indicated by open squares (□) (Refs. 12 and 13), and those based on statistical model calculations by solid squares (■) (Refs. 14–17) without error bars. The average cross section in the region above 13.2 MeV is shown by the straight line at 1340 mb.

are presented in Fig. 1. A number of estimates of the total $(n,2n)$ cross section also have been made, based on $(n,2n)$ reaction systematics[12,13] and statistical model calculations.[14-17] These estimates have somewhat larger uncertainties (±15 to 30%) than do the actual measurements (see Fig. 1). Since the $(n,2n)$ excitation function is almost flat in the energy region above 13 MeV, we have used the weighted average of the measured cross sections between 13 and 17 MeV, which is 1340 mb with an uncertainty of about 4%. The cross section estimates tend to be lower than the measured values, but are in agreement within their limits of accuracy.

The cross section at 14.8 MeV to form the 10.15-day isomer has been measured to be 463 ± 14 mb.[6] The net cross section to form the ^{92}Nb ground state is then, by difference, 877 ± 55 mb.

Disintegration rate measurement

The ^{92}Nb decay scheme is shown in Fig. 2.[18] The 561.1-934.5-keV γ-ray cascade provides a unique signature for the decay of the ^{92}Nb ground state. Each γ ray occurs in essentially 100% abundance. The $^{92}Nb^g$ measurements were made first on the niobium foils with calibrated Ge(Li)

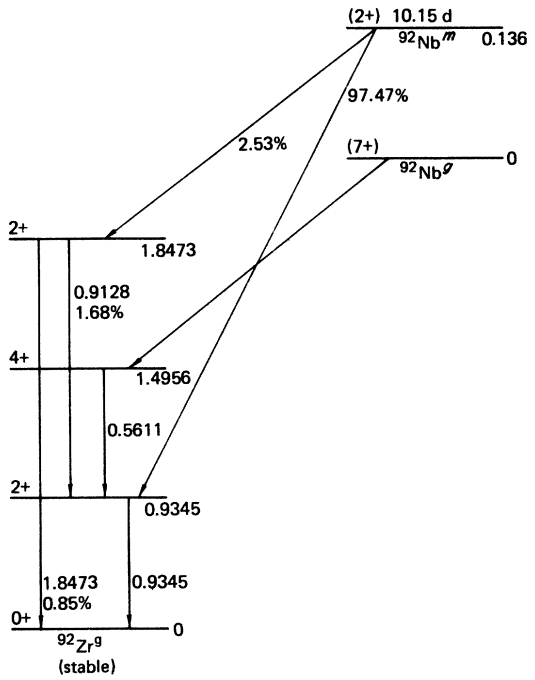

FIG. 2. Schematic drawing of the decay scheme of ^{92}Nb.

detectors[5] about one year after the irradiations had ended, allowing the 10.15-day isomer to decay away sufficiently. Several long-lived radionuclides (e.g., ^{54}Mn, ^{57}Co, ^{60}Co, and ^{182}Ta), formed by activation of impurities in the niobium, interfered with the accurate measurement of the very low levels of $^{92}Nb^{g}$ in the samples. The niobium foils were dissolved and radiochemically purified to remove the contaminants. Samples of Nb_2O_5 were prepared for final counting with Ge(Li) detectors. The γ-ray spectra from these samples contained mainly the photopeaks from the decay of $^{92}Nb^{g}$ and $^{94}Nb^{g}$, plus a few other nuclides present in the detector background. Portions of the spectra showing the photopeaks of interest are shown in Figs. 3 and 4.

Because of the low counting rates, the radioactivity measurements had to be made with the samples close (1.2 to 2.6 cm) to the effective center of the Ge(Li) crystal. This then required a correction for summing losses due to true coincidences occurring with the 561.1- and 934.5-keV γ rays. Fortunately, the $^{94}Nb^{g}$ in the samples provides a fairly accurate means of estimating the extent of the summing corrections. The decay of $^{94}Nb^{g}$ is very similar to that of $^{92}Nb^{g}$, involving a cascade

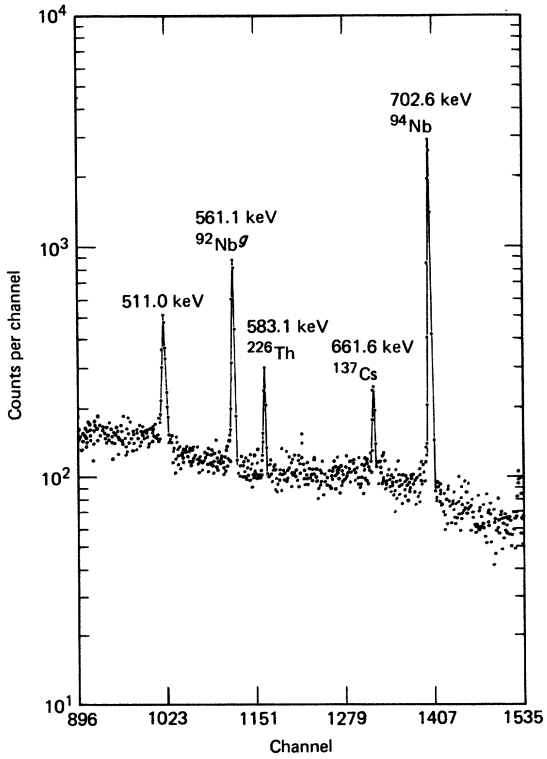

FIG. 3. A portion of one of the Ge(Li) spectra taken of a purified Nb_2O_5 sample showing the energy range from 448 to 768 keV.

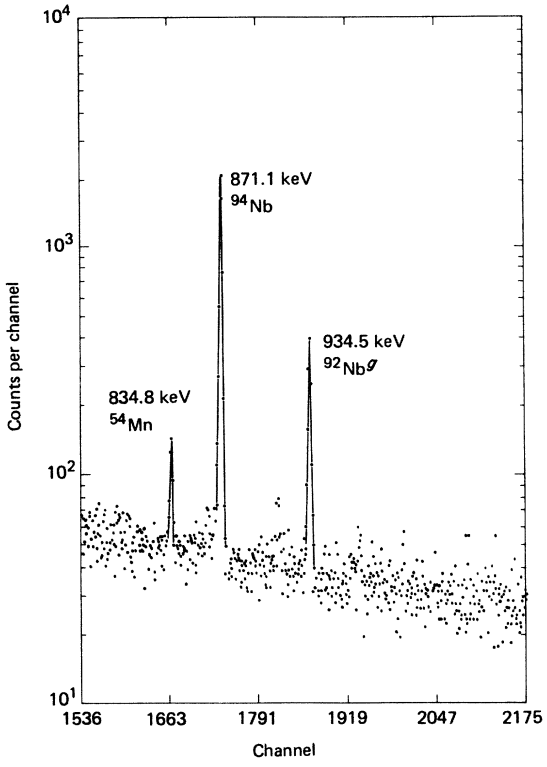

FIG. 4. A portion of one of the Ge(Li) spectra taken of a purified Nb_2O_5 sample showing the energy range from 768 to 1152 keV.

TABLE I. Results for the half-life of $^{92}Nb^{\varepsilon}$.

Irradiation	Measured $^{92}Nb^{\varepsilon}$ (dpm/g)	Error (%)	14.8-MeV fluence	Error (%)	Calculated half-life (yr)	Error (%)
Short	36.8	10	1.64×10^{17}	4.2	3.30×10^{7}	10.9[a]
Long	634	5	3.23×10^{18}	6.2	3.82×10^{7}	7.9[a]
Average					3.60×10^{7}	9.0

[a]The percent error does not include the 6.3% error in the $(n,2n)$ cross section common to both measurements; this error was added in separately after averaging the two results.

of two γ rays with energies of 702.6 and 871.1 keV (see Figs. 3 and 4).[19] The $^{94}Nb^{\varepsilon}$ in the Nb_2O_5 sample from the long irradiation was sufficiently radioactive so that measurements could be made at several sample-to-detector distances ranging from 1.3 to 16.1 cm. A plot was made of the coincidence loss as a function of sample-to-detector distance, assuming that the loss was negligible at the largest distance. We then made the further assumption that, because of the similar decay schemes and γ-ray energies, this plot was also appropriate to use for $^{92}Nb^{\varepsilon}$.

The disintegration rate for the Nb_2O_5 sample from the short irradiation finally was determined to be 2.51 dpm $^{92}Nb^{\varepsilon}$ in the sample, or 36.8 dpm/g Nb, with an uncertainty (1σ) of ±6.5%. A correction of 9.4% was made for the coincidence loss. We estimate that the total uncertainty in this measurement is about ±10%. The final result for the Nb_2O_5 sample from the long irradiation is 20.7 dpm $^{92}Nb^{\varepsilon}$, or 634 dpm/g Nb with an uncertainty (1σ) of ±2.6%. Corrections of 2.3 and 8.6% were made for the coincidence losses in measurements taken at two different sample-to-detector distances (2.55 and 1.34 cm). We estimate that the total uncertainty in the measurement for the long irradiation is about ±5%.

RESULTS AND DISCUSSION

A summary of the data used for the calculation of the $^{92}Nb^{\varepsilon}$ half-life is given in Table I. Using the equation derived in the Introduction, $T_{1/2}(\mathrm{yr}) = 8.54 \times 10^{15}\, \sigma\phi/(\mathrm{dpm/g})$, with $\sigma = 877$ mb ± 4.8%, the half-lives are (3.30 ± 0.36) and $(3.82 \pm 0.30) \times 10^7$ yr for the short and long irradiations, respectively. The average of the two measurements is $(3.6 \pm 0.3) \times 10^7$ yr, with the uncertainty in the $(n,2n)$ cross section included. This result is in agreement with the value of $(3.3 \pm 0.5) \times 10^7$ yr reported by Makino and Honda.[3]

The logft value is 14.5 for the electron capture decay, based on $\log_0^{\varepsilon}$ values[20] and a decay energy of 0.510 MeV.[18] This logft value is consistent with the $\Delta I = 3$ decay ($7+ \rightarrow 4+$) suggested for $^{92}Nb^{\varepsilon}$. In addition, if a mass-spectrometric measurement can reduce the error in the half-life determination to only a few percent,[2,3] then our measurement can be used in turn to infer a more accurate value of the $(n,2n)$ cross section, based on a knowledge of the $^{92}Nb^{\varepsilon}$ half-life.

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