Cross-section measurement for the ^{93}Nb(n, 2n)^{92g}Nb reaction at the neutron energy of 14.6 MeV by the AMS method *

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Abstract: The cross-section for the ^{93}Nb(n, 2n)^{92g}Nb reaction has been measured at the neutron energy of 14.6 MeV using neutron activation and accelerator mass spectrometry (AMS) determination of the long-lived product nuclide ^{92g}Nb. The neutron energy was generated from the D+T neutron source at the China Institute of Atomic Energy (CIAE). The neutron flux was monitored by measuring the activity of ^{92m}Nb produced in the competing reaction channel of ^{93}Nb(n, 2n)^{92m}Nb. At the neutron energy of 14.6 MeV, the ^{93}Nb(n, 2n)^{92g}Nb reaction cross-section of (736±220) mb was obtained for the first time.

Keywords: ^{93}Nb(n, 2n)^{92g}Nb reaction, cross-section, D+T neutron source, accelerator mass spectrometry

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1 Introduction

Niobium has been used in the zirconium alloy for reactor fuel rod cladding to improve the mechanical and radiation properties, and ^{92}Nb produced via the ^{93}Nb(n, 2n)^{92}Nb reaction, induced by the high-energy fission neutrons, opens up the possibility for long-term integral neutron dosimetry [1]. The cross-section of this reaction would be useful for nuclear engineering and theoretical research. Due to the long half-life of ^{92g}Nb ($T_{1/2}$=3.47±0.24)×10^7 a), there has been no experimental cross section value reported so far. An AMS method for the measurement of the isotope ratio ^{92}Nb/^{93}Nb was previously established by our group [2]. In this work the cross-section of ^{93}Nb(n, 2n)^{92g}Nb reaction is determined for the incident neutron energy of 14.6 MeV by neutron activation and AMS measurement of the product nuclide ^{92g}Nb.

2 Experiments

2.1 Irradiation

In this work, neutrons were produced by the ^{3}H(d, n)^{4}He reaction with an effective d$^+$ beam energy of 135 keV and a beam current of 300 μA. The tritium-titanium (T-Ti) target was 1.3 mg/cm^2 thick and was placed at a 0^0 angle relative to the d$^+$ beam direction.

High purity metal Nb produced by the Northwest Institute for Non-Ferrous Metals Research of China was used. Two Nb samples were prepared. One sample was used for neutron irradiation and the other as the blank sample. Both samples were machined in a shape adapted to the standard cathode of the cesium beam sputtering source (Fig. 1) for the AMS measurement. The niobium sample was irradiated for about 200 h at 0°with respect to the deuteron (d$^+$) beam at a distance about 6 mm away from the T-Ti target of the Neutron Generator at China Institute of Atomic Energy (CIAE). The neutron yield was about 2.5×10^{10} n/s. The neutron flux variation was monitored by the real-time measurement of the associated alpha particles.

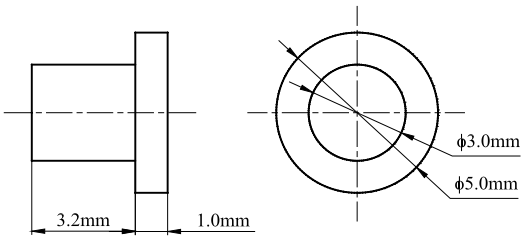

Fig. 1. Sketch of a niobium sample.

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2.2 Activity measurements

The average neutron flux was calculated using the cross-section of the ^{93}Nb(n, 2n)^{92m}Nb reaction and the activity of the product nuclide ^{92m}Nb (with a correction for neutron flux variation by the real-time α-monitoring). The ^{93}Nb(n, 2n)^{92m}Nb reaction cross-section (459±5) mb, was taken from Ref. [3]. The latter was obtained by γ ray spectrometry. The emission rate of the 934.5 keV γ-rays from ^{92m}Nb was measured by a coaxial HPGe detector (model GEM-40190-PLUS, ORTEC Corporation, USA) with a relative efficiency of 40% and an energy resolution of 1.9 keV at 1.33 MeV. The efficiency curve of the detector at the source to detector distance of 25 cm was calibrated using a set of standard γ-ray sources (^{57}Co, ^{137}Cs, ^{60}Co, ^{152}Eu, ^{182}Ta) with relative standard uncertainties in activities of less than 1%, certified by the National Institute of Metrology of China. This counting position is far enough to neglect the coincidence losses of cascade γ-rays. The relative uncertainty in the absolute efficiency curve is below 1.5% in the energy range of 150-1408 keV.

The average neutron flux, I, at the sample position was calculated by using Eq. (1):

$$\frac{1}{I}=\left[K\frac{N_0\sigma_m}{\lambda_m}(1-\mathrm{e}^{-\lambda_m t_1})(\mathrm{e}^{-\lambda_m t_2}-\mathrm{e}^{-\lambda_m t_3})\right]\Big/\left[F\frac{C}{\varepsilon I_\gamma}\right], \tag{1}$$

where N_0 is the number of target nuclides of ^{93}Nb, N_0=$m/A\times R$, where m is the sample mass in grams, A is the gram-atomic weight and R is the Avogadro constant, σ_m represents the cross-section of the monitor reaction ^{93}Nb(n, 2n)^{92m}Nb, ε is the full-energy peak efficiency of 934.5 keV γ-rays, I_γ is the emission probability of 934.5 keV γ-rays, C is the full energy peak counts for 934.5 keV γ-rays, .λ_m is the decay constant of ^{92m}Nb, t_1, t_2 and t_3 are the time interval of the irradiation and time intervals from the end of the irradiation to the start of the counting and to the finish of the counting, respectively, F is total correction factor of the activity, $F=f_s\times f_o\times f_g$, where f_s, f_o and f_g are correction factors for the self-absorption of the sample for 934.5 keV γ-rays, the coincidence sum effect of cascade rays of ^{92m}Nb and the counting geometry, respectively, and K is a neutron flux fluctuation factor expressed by Eq. (2):

$$K=\left[\sum_{i=1}^{L}\varphi_i(1-\mathrm{e}^{-\lambda\Delta t_i})\mathrm{e}^{-\lambda T_i}\right]\Big/\varphi S, \tag{2}$$

where L is the number of intervals into which the irradiation time was divided, Δt_i is the duration of the ith time interval, T_i is the time interval from the end of the ith time interval to the end of irradiation, Φ_i is the neutron flux averaged over the sample in Δt_i, Φ is the neutron fluence averaged over the sample in the total irradiation time T, and S=1−e$^{-\lambda T}$ is the growth factor of the product nuclide.

Relevant nuclear parameters for the activity determination are taken from Firestone et al., 1996 [3], and summarized in Table 1.

Table 1. Relevant nuclear parameters for neutron flux determination.

reaction	abundance of ^{93}Nb	half-life of the products	E_γ/keV	I_γ(%)	σ_m/mb
^{93}Nb(n, 2n)^{92m}Nb	1	10.15d	934.5	99±0.1	459±5

Table 2. Values and uncertainties relative to neutron flux determination.

item	value	unit	relative uncertainty(%)
σ_m	459	mb	1.0
t^*	9656	s	0.1
m	0.4238	g	0.1
C	743895		0.1
ε	0.00027		1.5

* the counting time of the γ-ray spectrum, derived from t_3-t_2.

The correlated values for the neutron flux determination are listed in Table 2.

The average incident neutron flux, I, was obtained to be (9.65±0.02)×10^8 n/s.

2.3 Isotope ratio measurements

Both ^{92m}Nb and ^{92g}Nb decay to ^{92}Zr by positron emitter or orbital electron capture mode. After eighteen months of cooling, almost all of the excited state of ^{92}Nb had decayed to ^{92}Zr, however, the ground state of ^{92}Nb was present in the irradiated sample due to the half-life difference. Profiting from the high particle discrimination ability of the CIAE AMS system, ^{92}Zr can be discriminated from ^{92}Nb, and the two stable isobars, ^{92}Zr and ^{92}Mo, can be clearly separated in the residual energy spectrum. A measurement of the blank sample was conducted as showed in Fig. 2. The distribution of ^{92g}Nb, intermediate between ^{92}Zr and ^{92}Mo, could be estimated according to its atomic number and energy.

The isotopic ratio of ^{92g}Nb to ^{93}Nb in the irradiated sample was measured by the established AMS method. Two-dimensional energy spectra of $\triangle E_1-E_R$ for a blank sample and the ^{92g}Nb sample are shown in Figs. 3 and 4, respectively.

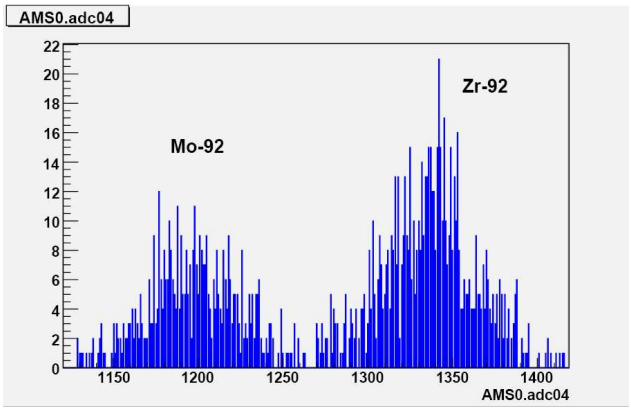

Fig. 2. (color online) Residual energy spectrum of a blank sample.

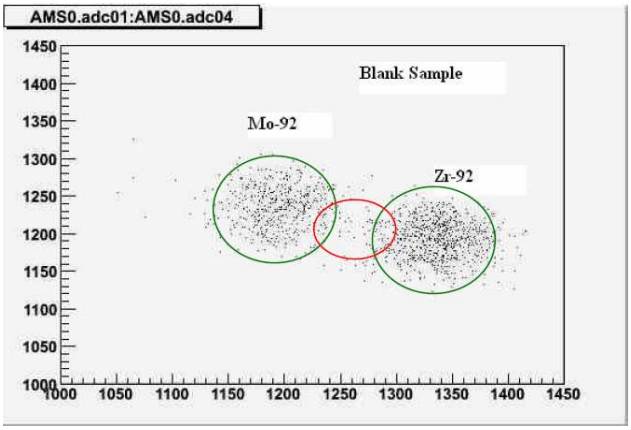

Fig. 3. (color online) E_1-E_R spectra of a blank sample.

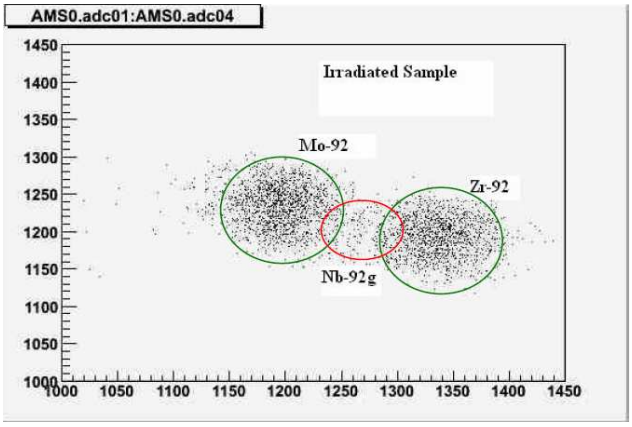

Fig. 4. (color online) E_1-E_R spectra of ^{92g}Nb sample.

Gated on $\triangle E_1$, $\triangle E_2$, $\triangle E_3$, E_R and E_t, net count rates of ^{92g}Nb were obtained by the subtraction of gated count rates of the blank sample from those of the ^{92g}Nb sample after beam current normalization, as described in Ref. [2]. The results of the AMS measurements for the un-irradiated (blank) and irradiated samples are listed in Table 3.

Table 3. Measurements of ^{92g}Nb/^{93}Nb for blank and irradiated samples.

sample	$I_{\rm LEcup}$/ nA	counts of ^{92g}Nb	measure time/min.	^{92g}Nb/^{93}Nb	RSU (%)
irradiated	0.74	47	35	1.05×10^{-9}	15
blank	1.20	14	21	3.20×10^{-10}	26

$I_{\rm Lecup}$ is the beam current before the particle injected into the accelerator.

The net isotope ratio of ^{92g}Nb to ^{93}Nb, $N_{\rm g}/N_{\rm o}$, was obtained by subtracting the ratio for blank sample from that of the irradiated one, giving $(7.30\pm1.76)\times10^{-10}$.

2.4 Data analysis and results

The cross-section of the ^{93}Nb(n, 2n)^{92g}Nb reaction was calculated by Eq. (3):

$$\sigma_{\rm g}[{\rm mb}]=\frac{N_{\rm g}}{N_0}\cdot\frac{1}{I\cdot t\cdot10^{-27}}, \quad (3)$$

where $\sigma_{\rm g}$ represents the cross-section of the measured reaction, $N_{\rm g}/N_0$ is the isotope ratio measured by the AMS method, t is the irradiation time, and I is the average neutron flux calculated by Eq. (1).

Values and corresponding uncertainties relevant to the cross section determination are listed in Table 4.

Table 4. Values and uncertainties relevant to cross-section determination.

items	value	relative uncertainty(%)
t	1.03×10^{6}	<0.1
I	9.65×10^{8}	1.8
$N_{\rm g}/N_0$	7.30×10^{-10}	28.3
$\sigma_{\rm g}$	736 mb	28.6

The cross-section for the ^{93}Nb(n, 2n)^{92g}Nb reaction, $\sigma_{\rm g}$, has been obtained to be 736±220 (mb) at the neutron energy of 14.6±0.11 MeV.

3 Discussion

To our knowledge, the value obtained in this work is the first ever direct experimental data of the ^{93}Nb(n, 2n)^{92g}Nb reaction cross-section.

Previously reported total cross-sections of ^{93}Nb(n, 2n)$^{92m+g}$Nb reaction at neutron energy of 14 MeV, measured by the activation method or online method, are listed in Table 5 [4-11]. The difficulty of the activity measurement of the irradiated sample, due to the very long half-life of ^{92g}Nb, always results in large uncertainty in the ^{93}Nb(n, 2n)$^{92m+g}$Nb reaction cross-sections. The online method can barely obtain the cross-sections data accurately, due to interference from the intensive sample scattered neutrons and relatively large system errors. There are three cross-section values of the ^{93}Nb(n, 2n)$^{92m+g}$Nb reaction at the neutron energy point of 14.6 MeV.

Table 5. Reported cross-section values of ^{93}Nb(n, 2n)$^{92m+g}$Nb reaction.

neutron energy/MeV	cross-section/ mbarn	author	time	Refs.
14.6	1300±200	Yu.E.Kozur	1977	[4]
14.3	1360±96	A.A.Lychagin	1984	[5]
14.6	1562±212	E.Holub	1976	[6]
14.6	1150±300	D.Hermsdorf	1973	[7]
14.1	1350±250	M.Haering	1971	[8]
14.3	1312±83	D.S.Mather	1972	[9]
14.76	1313±99	J.Frehaut	1980	[10]
14.7	1279±88	L.R.Veeser	1977	[11]

Subtracting (459.0±5.6) mb, the ^{93}Nb(n, 2n)^{92m}Nb reaction cross section of Ref. [12], from the total cross-section values reported by Yu. E. Kozur [4], E. Holub [6] and D. Hermsdorf [7], the cross-sections of ^{93}Nb(n, 2n)^{92g}Nb reaction are indirectly inferred to be 841±200, 1103±212 and 691±300, respectively.

The values of the ^{93}Nb(n, 2n)^{92g}Nb reaction cross section of our work and those of Ref. [4] and Ref. [7] agree with each other within errors. The difference between Holub's values [6] and ours is larger. Holub obtained the cross-section of the ^{93}Nb(n, 2n)$^{92m+g}$Nb reaction theoretically, based on statistical theory combined with the cross-section of the ^{93}Nb(n, 2n)^{92m}Nb reaction. The difference is thought to be dominated by the theoretical defect. The measured ^{93}Nb(n, 2n)^{92g}Nb reaction cross-section value allows more accurate analysis of the outgoing neutron spectrum, induced by the ^{93}Nb(n, 2n)^{92g}Nb and ^{93}Nb(n, 2n)^{92m}Nb reactions.

The uncertainties of our value are mainly from the AMS method, one is the low statistics in the isotopic ratio measurement and the other is from the AMS absolute method.

4 Summary

It is very difficult to measure the ^{93}Nb(n, 2n)^{92g}Nb reaction cross section by the activation method because of the very low radioactivity of the long lived product nuclei, ^{92g}Nb. It is also impossible by ordinary mass spectrometry such as secondary ion mass spectroscopy, because the measuring limit for the isotopic ratio measurement of ^{92}A/^{93}A is always larger than the actual value of ^{92g}Nb/^{93}Nb of the irradiated sample. By the online method, direct detection of the two outgoing neutrons from the reactions of ^{93}Nb(n, 2n)^{92m}Nb and ^{93}Nb(n, 2n)^{92g}Nb, the influence of the half-life can be avoided, but one can only obtain the total cross-section restricted by the capability of distinguishing between the two reactions. In our AMS method the shortcoming is that the deficient of the standard sample and the transmission efficiency had to be determined by using the beam current of ^{93}Nb^{+}. Different transmission between the beam and the particle always induce some efficiency divergence. If there is an intensive D-T neutron source with neutron yield more than 10^{13} n/s, a more accurate result could be expected.

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