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MEASUREMENT OF THE ^{93}Nb(n,n')^{93m}Nb CROSS SECTION AT 7.9 MeV

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Abstract: The cross section for the reaction ^{93}Nb(n,n')^{93m}Nb was measured by foil activation to be (265.9 ± 9.8) mbarn at a neutron energy of (7.91 ± 0.02) MeV relative to the ^{238}U(n,f) cross section employing a small ionization chamber as neutron fluence monitor. The mass of the fissile reference deposit was assessed relative to the mass of Al foils by means of a separate irradiation experiment with 14.8 MeV neutrons, thus relying on the ^{238}U(n,f)/^{27}Al(n,α) cross section ratio at this energy. The induced ^{93m}Nb activity was measured detecting the Nb K-shell x rays from the 30.7 keV isomeric transition by means of a Si(Li) x-ray detector. Sample size effects due to self-attenuation of the Nb x rays, extension of the sample, inhomogeneous foil activation and K-shell x-ray fluorescence induced by background radiation were experimentally determined. The result indicates, together with other recently available experimental data, that the cross section information included in the International Reactor Dosimetry File should be revised.

(^{93}Nb(n,n')^{93m}Nb, activation cross section, E_n = 7.9 MeV, reactor dosimetry, fluence monitor.)

Introduction

The reaction ^{93}Nb(n,n')^{93m}Nb is particularly important as a long-term activation monitor in reactor dosimetry due to the long half-life (16.1 ± 0.2 years)[1] of the produced ^{93}Nb isomer and its low reaction threshold (∿ 31 keV). This has been documented by, e.g., presentations at ASTM-EURATOM symposia[2], and by increasing interest of national standardization committees. Tedious recent measurements by Gayther et al.[3] and Wagner et al.[4] have revealed significant discrepancies to the present evaluation of the excitation function of the ^{93}Nb(n,n')^{93m}Nb reaction as included in the IRDF [5]. The new measurement was performed in order to extend the experimental data base for a future reevaluation of the ^{93}Nb(n,n')^{93m}Nb excitation function.

Experimental procedure

Irradiation and neutron fluence monitoring

A Nb foil, ∿ 0.13 mm thick and 20 mm in diameter, with a purity better than 99.9%, was exposed together with monitor foils to ∿ 5 x 10^{12} cm^{-2} D(d,n)^{3}He neutrons for ∿ 60 hours, employing a deuterium gas cell and a deuteron (d^+) beam from the CN-type Van de Graaff accelerator at the Central Bureau for Nuclear Measurements (CBNM), Geel. The air-jet cooled gas cell was 4.0 cm long, operated at 1.5 x 10^5 Pa gas pressure, equipped with a 3.29 mg cm^{-2} Havar entrance window and a 0.5 mm Ta beam stop. The energy of the incident deuterons was (5.091 ± 0.005) MeV, which resulted in a beam energy of (4.753 ± 0.010) MeV at the target center; the deuteron current was about 4 μA. A background run lasting for ∿ 29 hours was performed with the gas cell filled with He instead of deuterium using another set of Nb and monitor foils in order to correct for parasitic neutrons produced in the gas cell structure.

The samples were arranged as shown in Fig. 1, and placed at 0° relative to the incident d^+ beam, together with a small fission chamber in order to measure the neutron fluence. The distance between the Nb sample and the beam stop was 6 mm. The reference fissile material was ^{238}U enriched to 99.98%, in the chemical form UF_4, deposited as a 10-mm-dia layer on a 0.2 mm Al backing. In order to interrelate the average fluence at the position of the Nb foil and the fissile layer 20-mm-dia Ni foils inside and outside the fission chamber served to measure the neutron fluence gradient via the induced ^{58}Co activity. The latter was measured by integral γ-ray counting by means of a 12.7 cm x 12.7 cm NaI(Tl) well-type detector[4], correcting for the contribution of ^{60}Co (0.35%) from the reaction ^{60}Ni(n,p)^{60}Co and for the contribution of background neutrons (2.4% and 3.0% for a 10-mm-dia and a 20-mm-dia Ni foil, respectively). Assuming a fluence dependence like $1/r^{\alpha}$ (r ... distance sample to center of the neutron source) the value of the exponent α was determined to be 1.9 ± 0.1, which includes neutron absorption effects. The conversion factor for the average fluence in the 20-mm-dia foil to the fluence in the 10-mm-dia U layer was determined by measuring the activity of the ring-shaped part and the central part of the inner Ni foil separately thus taking into account the decrease of the neutron fluence with increasing angle. The 0.1 mm thick Ta foil served to estimate the enhancement of the Nb K-shell x-ray emission rate caused by a Ta impurity (∿ 350 ppm according to a spectrochemical analysis) in the Nb sample, by way of measuring the induced ^{182}Ta activity by means of a Ge(Li) γ-ray detector (see section 2.4.2 of Ref. 4).

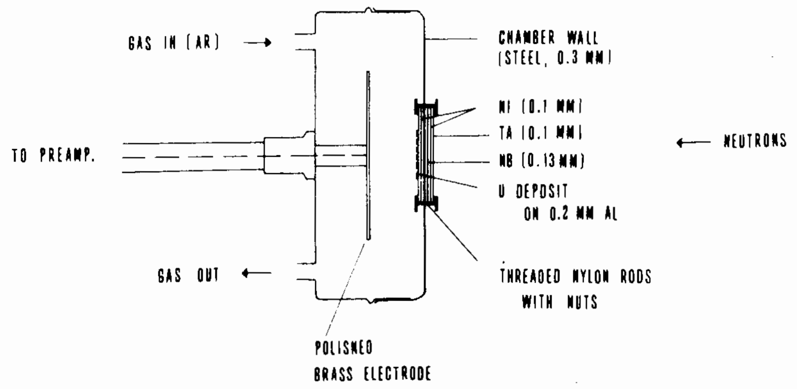

Fig. 1: Sample arrangement inside and outside the fission chamber

The operation of the fission ionization chamber has been described in detail in Ref. 4. The correction applied for the extrapolation of the pulse-height distribution due to the fission fragments to zero pulse height was (0.93 ± 0.31)%; the correction for selfabsorption of the fission fragments considering reaction kinematics and the angular distribution[6] of the fragments was estimated to be (0.6 ± 0.2)% assuming a uniform deposit. The number of fission events from parasitic neutrons amounted to ∿ 4.4% of the total fission fragment counts normalized to the same d^+ charge integral as for the foreground run.

The mass of the ^{238}U deposit was determined relative to the mass of Al foils by means of a separate irradiation experiment with 14.8 MeV neutrons employing the 200-keV Cockcroft-Walton accelerator at the Institut für Radiumforschung und Kernphysik (IRK), Vienna, Austria. The relevant procedures are described in detail in section 2.3 of Ref. 4. The ^{24}Na activity induced in the Al samples was measured absolutely at IRK by integral γ-ray counting [7] with the 12.7 cm x 12.7 cm NaI(Tl) well-type detector mentioned above. Weighting the results from three irradiation runs with different experimental conditions (the reproducibility was within $\lesssim$ 0.5%) resulted in (168.4 ± 2.1)µg for the ^{238}U mass of the fissile deposit. This value thus relies on the ratio of the most recent values for the cross sections for the reactions ^{27}Al(n,α)^{24}Na [8] and ^{238}U(n,f)[9] at a neutron energy of 14.82 MeV. Several arguments are mentioned in Ref. 4 why for the present evaluation of our measurements we have preferred this result to that to be gathered from the measured α-particle emission rate. One among these arguments is: in the course of the fission chamber measurements at 14.8 and 7.9 MeV neutron energy systematic errors in the corrections for the loss of fission fragments due to the microscopic roughness of the deposit and in the extrapolation of the pulse-height distribution will largely cancel.

Measurement of the induced ^{93m}Nb activity

The ^{93m}Nb activities induced in the Nb foils were measured detecting the Nb K-shell x rays from the internal conversion of the 30.7 keV isomeric transition by means of a calibrated Si(Li) detector. The experimental details including necessary corrections will be found in Ref. 4. Due to self-attenuation of the Nb x rays in the irradiated foils ∿ 36% of the emitted x rays were registered as compared to a very thin sample. The self-attenuation correction was determined experimentally with high accuracy in the following way: In a stack of thin inactive Nb foils each of these foils was subsequently replaced by an equivalent Nb foil which had been activated by 14 MeV neutrons in order to simulate the activity sitting in different layers of the sample actually used for the irradiation. The integration of the response of the detector versus the position of the active foil in the stack over the various layers in the sample-equivalent stack provided the self-attenuation factor. From the foreground neutron irradiation the ^{93m}Nb x-ray net count rate in the K_α plus K_β x-ray peaks was ∿ 1.15 counts per minute, obtained from 14 individual measurements lasting for ∿ 10^5 s each, interspaced by background measurements over similar periods. From the background neutron irradiation the ^{93m}Nb net count rate was ∿ 0.13 counts per minute normalized to the same d^+ charge integral as in the foreground run. Due to these very low K-shell x-ray count rates special attention had to be paid to K-shell x-ray fluorescence induced in the

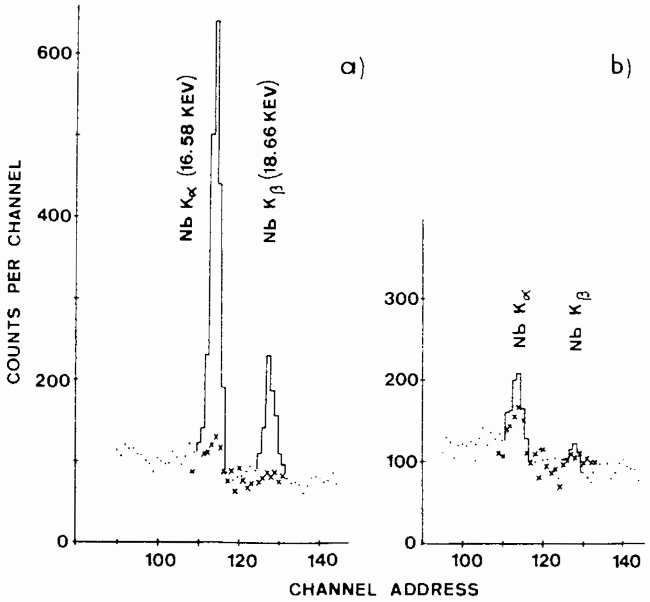

Fig. 2:
(a) X-ray spectrum from the Nb foil irradiated with 7.9 MeV neutrons (-) and background spectrum using a non-irradiated Nb foil (x), both recorded during 10^5 s with a Si(Li)-detector.
(b) The corresponding spectra related to the activation by parasitic neutrons, both recorded during 1.3 x 10^5 s.

sample by background radiation. The background spectrum was measured using a Nb foil of the same dimensions as the irradiated foils, but not activated by neutrons. The background count rate without any sample (0.73 counts per minute) was increased by ∿ 15% - a striking example how the background effect may be changed by the sample itself. Examples of the measured spectra in the relevant energy region stemming from the foreground and the background neutron irradiation are shown together with the non-irradiated sample spectra in Fig. 2.

The Ta impurity in the Nb foils could enhance the count rate in the Nb K-shell x-ray peaks due to the production of a ^{182}Ta activity. This increase via x-ray fluorescence caused by the attenuation of W K-shell x rays and low-energy photon radiation from the decay of ^{182}Ta amounted to ∿ 0.1% only.

The net count rates in the x-ray peaks were corrected for activation enhancement due to elastic neutron scattering in the Nb samples. Inelastic neutron scattering by the Ta beam stop and material close to the Nb sample and the fissile layer increases both the number of fission fragment counts and the activity in the Nb foil. The net effect was estimated to be $(0.25 \pm 0.2)\%$.

Results

The outcome of the present work is summarized in Table 1. The neutron energy distribution profile averaged over the position of the Nb foil and the ^{238}U layer was calculated by means of a Monte Carlo simulation providing the respective energy spread and average neutron energy.

According to the procedure used to determine the number of ^{238}U nuclei in the reference fissile deposit, the result of our activation measurement is proportional to the indicated reference cross sections:

$$\sigma(^{93}\mathrm{Nb}(n,n')^{93m}\mathrm{Nb}) \propto \frac{\sigma(^{238}\mathrm{U}(n,f) \text{ at } 7.97 \text{ MeV}) \, \sigma(^{27}\mathrm{Al}(n,\alpha) \text{ at } 14.82 \text{ MeV})}{\sigma(^{238}\mathrm{U}(n,f) \text{ at } 14.82 \text{ MeV})} \qquad (1)$$

Possibly necessary renormalization can easily be performed.

Table 1: The activation cross section for the reaction ^{93}Nb(n,n')^{93m}Nb as measured in the present work together with the most relevant reference cross section data

Average neutron energy (MeV)	Energy resolution (1/2 FWHM, MeV)	^{93}Nb(n,n')^{93m}Nb cross section (mbarn)	^{238}U(n,f) reference cross section (mbarn) at E_n = 7.97 ± 0.02 MeV	Reference cross sections used for the calibration of the ^{238}U deposit at E_n = 14.82 ± 0.01 MeV (mbarn)	
				^{238}U(n,f)	^{27}Al(n,α)^{24}Na
7.91 ± 0.02	± 0.150	265.9 ± 9.8	992.5 ± 1.2% [a)]	1182.7 ± 0.9% [a)]	112.10 ± 0.6% [b)]

a) Taken from ENDF/B-VI[9]
b) Taken from the evaluation by Tagesen and Vonach[8]

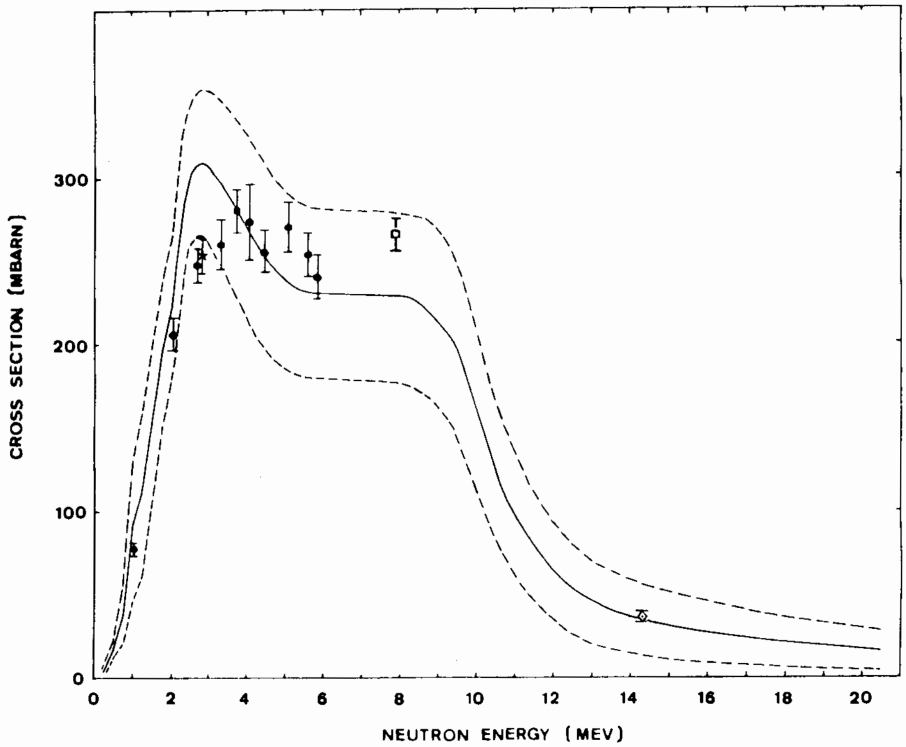

Fig. 3:
The ^{93}Nb(n,n')^{93m}Nb cross section measured in the course of this work and compared with data from the literature:

— IRDF (1982)[5], from a model calculation by Strohmaier et al. (1980)[10],
-- uncertainty limits thereof;
◇ Ryves and Kolkowski (1981)[11];
● Gayther et al. (1987)[3];
× Wagner et al (1988)[4];
□ present work.

Table 2 summarizes the various sources of uncertainties taken into account and provides a survey of the relevant procedures and corrections during the reduction of the experimental data. The partial uncertainty values stated represent standard deviations or equivalent standard deviations, which were added in quadrature to give the total uncertainty. Estimated correlations to a previous measurement of the ^{93}Nb(n,n')^{93m}Nb cross section at 2.8 MeV are included in Table 2. The result for the ^{93}Nb(n,n')^{93m}Nb cross section from this work is shown in Fig. 3 in context with activation data which became recently available. The experimental data indicate that the cross section information contained in the IRDF [5] should be revised.

Table 2: Sources of uncertainty and resulting errors in the measured ^{93}Nb(n,n')^{93m}Nb cross section.

Source of uncertainty	Resulting uncertainty (± %)	Correlation to Ref. 4 (%)
Measurement of the ^{93m}Nb activity:		
Counting statistics including the contribution by interfering neutrons and K-shell x-ray fluorescence due to background radiation	2.26	---
Correction for K-shell x-ray fluorescence caused by Ta impurity in the Nb sample	negligible	---
Efficiency of the Si(Li) detector for Nb K-shell x radiation, averaged over the sample area	0.98	100
Reproducibility of the geometry	0.40	---
Self-attenuation in the Nb sample	0.65	100
Half-life of ^{93m}Nb, survival factor	1.24	100
Mass of the Nb sample	negligible	---
Correction for activation enhancement due to elastic neutron scattering	negligible	---
Fluence related measurements:		
Fission-fragment-counting statistics (gas-in-run)	0.06	---
Correction for fission fragments produced by background neutrons	0.03	---
Extrapolation correction for fissions	0.31	100
Correction for finite thickness of the deposit	0.20	50
Dead time correction	0.05	---
Fluence extrapolation from sample to reference deposit including fluence gradient and irradiation geometry uncertainty	1.51	---
^{238}U(n,f) reference cross section at 7.97 MeV according to ENDF/B-VI	1.2	a)
Neutron energy uncertainty contribution therein	negligible	---
Total uncertainty of the ^{238}U-deposit mass including interference from other isotopes (see Ref. 4)	1.26	100
Net correction for inelastic neutron scattering	0.20	---
d^{+}-charge integral (for normalization of the data obtained from the background neutron irradiation)	negligible	---
Energy uncertainty due to the slope of the ^{93}Nb(n,n')^{93m}Nb cross section	negligible	---
Total uncertainty	3.70	

a) To be taken from final version of ENDF/B-VI

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