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A MEASUREMENT OF THE $^{93}Nb(n,n')^{93m}Nb$ CROSS-SECTION IN THE ENERGY RANGE 1 TO 6 MeV

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Abstract: The 30 keV isomer production cross-section in ^{93}Nb excited by neutron inelastic scattering has been measured at ten neutron energies. Small foils of niobium were exposed to high fluences of "monoenergetic" neutrons and the cross-section was determined from the induced K x-ray activities. The detector used in the activation measurements was calibrated with a standard solution of ^{93m}Nb which is used as an international reference material for reactor neutron dosimetry. The neutron fluence of the irradiation was measured with a low efficiency ^{235}U fission chamber in which the fissile deposit is located immediately behind the niobium foil. The ^{93m}Nb production cross-section is thus determined relative to the $^{235}U(n,f)$ standard reference cross-section.

(^{93}Nb, 30 keV isomer, differential cross-section, activation, fission monitor)

Introduction

The reaction for the production of the 30 keV isomeric state in ^{93}Nb is important as a monitor of damage fluence in power reactors because it has a differential cross-section which is similar to the damage function in steel and the metastable state decays with a half-life of about 16 y thus time-integrating the neutron flux. At the time the present measurements were started, there were no reported experimental data on the cross-section for neutron energies between 2.7 MeV and 14 MeV. Below 2.7 MeV, while data on the $^{93}Nb(n,n'\gamma)^{93}Nb$ were available, there are difficulties in deriving the isomer production cross-section from these measurements.

In 1982 a collaborative experimental programme was initiated to measure the neutron production cross-section of the isomer by direct activation, the target accuracy being 5% (1σ). The participants are: the Reactor Physics Division at AEE Winfrith, the Nuclear Physics Division at the Harwell Laboratory, and the Radiation Centre at the University of Birmingham. Niobium samples were irradiated with intense "monoenergetic" neutron beams at various energies between 1 and 6 MeV using the 3 MV Dynamitron accelerator at the Radiation Centre, and at 2.7 MeV on the 5 MV Van de Graaff at Harwell. High neutron fluxes were required, not only because of the small cross-sections and long half-life, but also because the only emitted radiations are conversion electrons and low energy x-rays, allowing only thin samples to be used. The irradiated material was analysed at Winfrith.

A detailed account of the techniques used in this work is given in ref. 1 together with an evaluated cross-section based on the measurements and theoretical calculations. A preliminary account of the experiment was presented at the 1985 Santa Fé Conference[2]. In the present paper the measured cross-section has been updated by using the $^{235}U(n,f)$ reference cross-section from version VI of the ENDF/B evaluated file. In addition, the analysis of the induced niobium activities has been improved.

Experimental

Irradiation procedure and fluence monitoring

Niobium metal samples were irradiated in the low scatter cell of the Birmingham University Radiation Centre using the 3 MV Dynamitron to provide neutrons at nine energies in the interval 1 to 6 MeV. In the energy range from 1 MeV to 2.1 MeV the reaction $^{3}H(p,n)^{3}He$ was used. For energies above 2.8 MeV the reaction was $^{2}H(d,n)^{3}He$. The energy range from about 2.2 to 2.8 MeV could not be covered in these experiments since only the $^{3}H(p,n)^{3}He$ source gives adequate yield and requires proton energies above those available. An additional measurement was made on the Harwell 5 MV Van de Graaff at 2.7 MeV because theoretical calculations[3] had predicted a broad peak in the isomer cross-section in this region.

The niobium foils are placed about 20 mm from the neutron source as a suitable compromise between maximising the neutron fluence and minimising the angular spread of neutrons intercepting the foils. Irradiations at such a short distance do not allow an independent measurement of the neutron fluence to be made with a detector of known efficiency, because the effective solid angle subtended by the foil at the source cannot be determined with sufficient accuracy. It is therefore necessary to determine the fluence by detecting a neutron-induced reaction with a cross-section which is a standard. The fission cross-section of ^{235}U was chosen for this purpose using an ionisation chamber to detect the fission events.

The fission chamber has a 95.1±0.5 μg deposit of 99.94% ^{235}U painted on a 0.127 mm thick platinum plate located on the inside surface of the chamber wall. The deposit has the same diameter as the niobium foil to be irradiated which is positioned in a circular recess on the outer wall of the chamber immediately in front of the ^{235}U deposit.

Foils of materials which provide additional fluence monitoring in the event of fission chamber failure during the long irradiation are included with four niobium foils in a sample pack. Each pack is a multi-layer sandwich of thin discs wrapped in thin aluminium. The reactions of interest are: $^{235}U(n,fp)^{140}Ba$ and $^{58}Ni(n,p)^{58}Co$. In some irradiations a ^{238}U foil was also included to utilize the $^{238}U(n,fp)^{140}Ba$ reaction. The uranium foils were sandwiched between aluminium catcher foils to retain the fission products. The inclusion of both ^{235}U and ^{238}U foils allowed a calculation of the contribution of low energy, room-scattered neutrons to the fission chamber output.

In the Van de Graaff run, the target geometry was different from that used on the Dynamitron, a much thicker layer of cooling water being used. To eliminate the possible background effect of moderated neutrons on the determination of the neutron fluence, the ^{235}U deposit in the fission chamber was replaced by a similar painted deposit of ^{238}U (99.999%). It was not possible to make an accurate determination of the mass of the ^{238}U deposit for the present purpose by α-assay, as was done for the ^{235}U deposit, because of the high purity of the material used. In the absence of a convenient alternative way of determining the ^{238}U mass, the neutron detection efficiency at 2.7 MeV of the fission chamber with the ^{238}U deposit was calibrated against its efficiency with the original ^{235}U deposit[1]. The ^{93m}Nb production cross-section measurement at 2.7 MeV is thus related to the ^{235}U fission cross-section as are the measurements at all the other energies.

Activation Measurements

The potential gamma-rays from the decay of ^{93m}Nb are highly internally converted, so the ^{93m}Nb activity must be measured either by counting the emitted electrons or the resultant x-rays. Electron counting requires the preparation of very thin sources and a method of presenting them to a detector in vacuum. Apart from any problems associated with electron detection, the very low activities obtained from the irradiations preclude the use of the required thin sources. It was therefore decided to count the 16.6 keV K x-rays.

The conflicting requirements involved in the preparation of sources of low specific activity but emitting easily absorbed radiations were resolved by a combination of both relative and absolute counting. For the former the four niobium foils were mounted side-by-side and counted together with an intrinsic Ge X-ray spectrometer (HPGE). This method allowed the relative activities from the various irradiations to be determined reasonably quickly. In order to convert these activities into absolute disintegration rates the following approach was adopted. Selected single Nb foils from the same packs were dissolved and the solution absorbed into filter paper discs. These were mounted on copper substrates and covered with a thin plastic film, and the sources so produced were counted in a reproducible geometry close to the end window of the HPGE.

The x-ray detector was calibrated with a solution of ^{93m}Nb received from CBNM, Geel. This solution has been very carefully calibrated at Geel by Bambynek and Coursey and the errors quoted on its activity are 0.05% random and 0.76% systematic. As this solution is part of a stock that is intended for use as an international reference material for reactor neutron dosimetry using niobium, it seemed appropriate to use it to normalize the cross-section measurements reported here.

The activities of the fluence monitor foils were measured with a germanium detector. The system was again calibrated from sources prepared from calibrated solutions.

Results

Fluence determination

The main purpose of the monitor foils was to provide a measurement of the neutron fluence of an irradiation in the event of an undetected failure in the fission chamber system during the very long exposure. In practice no such failure occurred and the fission chamber provided the most accurate determination of the fluence for all the measurements and this was used in deriving the niobium cross-section. Nevertheless, the monitor foil results provide a useful check of the fission chamber values based on different techniques. The fluences obtained from the monitor foils generally confirm the fission chamber fluences. The lanthanum yields from the ^{235}U foils, however, tend to be slightly lower than the fluences measured with the fission chamber would indicate.

The agreement between the various fluence measurements indicates that the effect of background and secondary source neutrons in the irradiations are small, because the neutron thresholds for the three reactions are different. Fission in ^{238}U has a threshold of ~1 MeV, the effective threshold for the $^{58}Ni(n,p)$ reaction is ~2 MeV while ^{235}U is thermally fissile.

The neutron fluence is obtained from the fission chamber counts after small corrections have been applied for fragment absorption in the fissile deposit and loss of events below the pulse height discrimination level, and account has been taken of any background effect. These effects are considered in detail in ref. 1. Fragment absorption varies from (0.8±0.3)% at 1 MeV to (0.4±0.15)% at 6 MeV. The uncertainty in these values is mainly due to the poorly known value of the fragment range in the painted fissile deposit. The correction for events lost below the discriminator bias amounts to (0.56±0.2)%. Determination of the fluence requires the mass of the ^{235}U deposit to be known. This was determined by α-assay with an uncertainty of 0.5%. Calibration of the ^{238}U chamber for the 2.7 MeV measurement introduces an additional uncertainty[1].

The measured cross-section

The measured cross-section is listed in Table 1. The neutron energy and cross-section is a mean value weighted over the calculated neutron energy resolution function for each irradiation. The full width at half maximum of the latter is also listed. The ^{235}U fission cross-sections were derived from the ENDF/B-VI evaluation. The half-life used was 16.13±0.15 y[4], and the detector used in this determination was calibrated against the reference solution provided by the Geel Laboratory, as described earlier. The neutron fluences in these measurements were in the range 5 x 10^{12} to 2.4 x 10^{13} neutrons/cm^2.

Table 1. The $^{93}Nb(n,n')^{93m}Nb$ cross-section

Mean neutron energy (MeV)	Resolution FWHM (MeV)	$^{93}Nb(n,n')^{93m}Nb$ cross-sections (mb)
1.09	0.19	74.8±4.5%
2.10	0.14	201.0±3.9%
2.70	0.20	245.5±3.3%
3.29	0.36	255.5±5.1%
3.73	0.34	274.2±3.7%
4.03	0.14	261.5±7.7%
4.45	0.27	251.3±3.7%
5.05	0.25	280.2±4.3%
5.53	0.18	251.0±4.1%
5.76	0.18	231.5±4.6%

The uncertainties in the niobium cross-section are expressed as standard deviations (1σ). The contributions to the overall uncertainty are:

	1σ (%)
^{235}U mass	0.5
^{93}Nb mass	0.2
$^{235}U(n,f)$ cross-section	1.0
^{93m}Nb activity	2.0-7.0
^{93m}Nb half-life	0.9
Number of fission events	0.2-0.4
Geometry effect	2.0

The uncertainty in the ENDF/B-VI fission cross-section has not been finalized and a conservative value of 1% has been used at all energies. The geometry effect allows for the different fluences at the respective positions of the niobium foil and the ^{235}U deposit.

Discussion

The data from these measurements are shown in Fig. 1 along with existing data below about 2.5 MeV deduced from $^{93}Nb(n,n'\gamma)^{93}Nb$ measurements. The $(n,n'\gamma)$ measurements of Williams[5], Van Heerden et al[6] and Göbel et al[7] were analysed by Strohmaier et al[3] to determine the 30 keV isomer excitation function, but a basic difference in the absolute cross-section scales of these data precluded their use for an evaluation, which was therefore based on a calculation of the cross-section.

The solid curve in Fig. 1 is the isomer excitation function calculated by Strohmaier et al using the statistical model and the level scheme derived by Van Heerden from the $(n,n'\gamma)$ data. The transmission coefficients were obtained using an optical potential which reproduced other neutron cross-section data for niobium including the low energy neutron strength functions. The error bar at 6 MeV is the uncertainty in the theoretical value.

The experimental values of the cross-section at 1.09, 2.10 and 3.29 MeV are significantly lower than the calculated cross-section, and indicate that the pronounced peak in the latter between 2 and 4 MeV may not be genuine. This was confirmed with the measurement carried out at a mean neutron energy of 2.7 MeV.

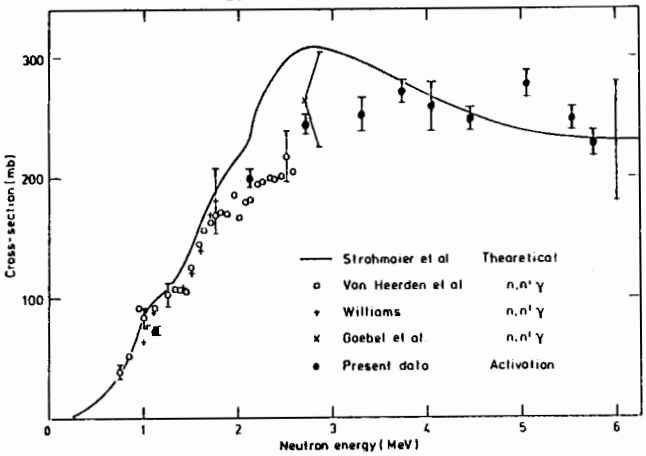

Fig. 1 Measured and calculated $^{93}Nb(n,n')^{93m}Nb$ cross-section

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