

# MEASUREMENTS OF NEUTRON CAPTURE CROSS-SECTION FOR TANTALUM AT THE NEUTRON FILTERED BEAMS

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The neutron capture cross sections of tantalum have been measured for the neutron energies 2 and 59 keV using the WWR-M Kyiv Research Reactor (KRR) of the Institute for Nuclear Research of the National Academy of Science of Ukraine. The cross sections of  $^{181}\text{Ta}(n, \gamma)^{182}\text{Ta}$  reaction were obtained by the activation method using gamma-spectrometer with Ge (Li)-detector. The obtained neutron capture cross sections were compared with the known experimental data from database EXFOR/CSISRS and the ENDF libraries.

## 1 Introduction

Capture cross-sections in the keV-neutron energy region are important in the design of reactors as well as in the study of nuclear physics. Tantalum (Ta) is the promising construction material, so the study of its interaction with neutrons is very important. Isotope  $^{181}\text{Ta}$  is a heavy deformed nucleus. It is located nearby the  $s$ -strength function maximum and  $p$ -strength function minimum. So, information about capture cross-section for  $^{181}\text{Ta}$  is also very needed for improvement of our nuclear structure knowledge. The evaluated nuclear data libraries BROND-2, CENDL-2, ENDF/B-VI, JENDL-3.3, and JEF-3 contain the data about neutron capture cross-section of tantalum, but the discrepancies between the experimental data, which were used for evaluation in the libraries, were 10-20% in the  $0.5 \div 100$  keV energy range. Moreover, the Request List [1] requires the value of neutron capture cross section for tantalum nuclei with the accuracy at least 10% for energy range 0.025 eV to 1 MeV.

That is why we planned and then performed the series of experiments for determination of the tantalum capture cross-section, using interference filtered neutron beams. In this paper we present the results of the measurements of the capture cross-section of  $^{181}\text{Ta}$  for neutrons with the average energies 1.97 keV and 59 keV. This experiment was done at the 2<sup>nd</sup> experimental horizontal channel of the Kyiv Research Reactor (KRR).

## 2 Experimental details

The activation method was used for definition of the neutron capture cross-section for tantalum nuclei in the reaction  $^{181}\text{Ta}(n, \gamma)^{182}\text{Ta}$ . This method is convenient by the following:

- The decay scheme of a daughter nucleus ( $^{182}\text{W}$ ) is much simpler and more investigated, so that there is no need to gather all the intensities of all

gamma-transitions, because the intensities of each transition are normalized by a single act of decay and are known with good accuracy.

- The capture of neutrons, in the case of filtered neutron beam, is realized in resonance region, this is, so called, average resonance capture (ARC). So, the level population and the gamma transition intensities depend as on energy of neutron line as on its width. At the same time, decay of the daughter nucleus tantalum-182 does not depend on its formation method, that is, neither from energy, nor from width of neutron filter line and this essentially simplifies our task.
- The target activities can be measured after irradiation in so-called “clean” zone, where the background conditions are much better.

That is, this method gives us a possibility to get the larger reliability of results and the better statistical precision.

The experiment scheme is shown in Figure 1. A few remarks are given on methodical details, especially some peculiarities, directed on improvement of measurement accuracy and on reducing of some correction factors:

- Neutron beam collimation was such that the whole sample was in a beam, but the beam did not touch the shield constructions (28 mm sample diameter, 32 mm beam diameter, 51 mm inner shield diameter).
- The irradiated samples were protected from background neutrons of reactor hall. Neither neutrons, scattered on constructive elements, nor neutrons from other experimental installations could get the samples and be captured.
- Permanent monitoring of neutron flux during all irradiation time was realized by neutron spectrometer with  $^3\text{He}$ -counter as a detector. Flux was controled every hour during 3000 s. The  $^3\text{He}$ -peak area was then used as weighting function for averaging of irradiation flux.
- The use of sample pile gave us an opportunity to take into account the neutron resonance self-shielding in the sample.

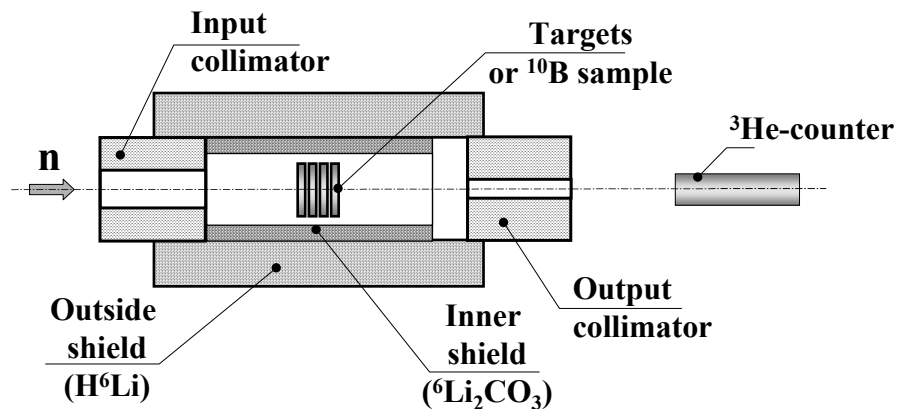


Figure 1. Scheme of tantalum target irradiation.

The irradiation process of the tantalum targets is shown on the scheme (Figure 1). The neutron beam (**n**) was directed with the system of entry collimators (**Input collimator**) to the pile of tantalum targets (**Targets**) in such way that all the targets were inside the beam (the target diameter was 28 mm, and that of the beam – 32 mm); the system of the outgoing collimators (**Output collimator**) focused the neutron beam to the  $^3\text{He}$  counter of the monitoring spectrometer.

The tantalum targets were made of natural tantalum foil and the targets had the 28.0 mm diameter. A pile of four 0.2 mm thickness targets and a pile of eight 0.1 mm thickness targets were used for measurements with 2 keV and 59 keV filter, respectively. Percentage of the  $^{181}\text{Ta}$  in natural tantalum is 99.988%. The diameter of the disks is less than the neutron beam diameter ( $d_{\text{beam}} = 32$  mm) in the position where the disks were placed.

The B-10 target, which is destined to define the neutron flux value, is made of the boric powder, enriched by  $^{10}\text{B}$  isotope. The target is placed inside the container with 28 mm diameter. The container was made of CAB-1T aluminum alloy; the front and back walls were of 0.2 and 0.3 mm thicknesses, respectively. The thickness of a boric sample was 1 mm. Ratio of boron – 97.1%; enrichment with  $^{10}\text{B}$  – 85%.

The spectra of neutrons, for which the cross-sections of the  $^{181}\text{Ta}(n, \gamma)^{182}\text{Ta}$  reaction were defined, were formed by two filters. One of them consists of  $^{10}\text{B}$ , Sc and  $^{60}\text{Ni}$  (2 keV filter), the 59 keV filter consists of  $^{10}\text{B}$ , V, S and  $^{58}\text{Ni}$ . The neutron spectra after the filters were calculated using the JENDL-3.3 and ENDF/B-6 nuclear data and the Filter-5 code.

2 keV filter. The main part of the filtered neutron spectrum is a line in the energy range 0.24-3.0 keV. The energy distribution center of this line is at 1.97 keV; its relative intensity is 95.47% of the entire neutron flux. Besides, in the spectrum there are impurity lines: 6.2+7.5 keV (2.93%), 27 keV (0.39%), 43 keV (0.14%), and a group of lines in the 48-66 keV range (1.08%).

59 keV filter. The main part of the filtered neutron spectrum is a line in the energy range 54.8-60.1 keV. The energy distribution center of this line is at 58.96 keV; its relative intensity is 94.36% of the entire neutron flux. Besides, in the spectrum there are impurity lines: 3 groups of lines in the energy range 1-40 keV (0.01%), 70-230 keV (0.11%) and 235-1570 keV (5.52%).

The neutron flux at the target place was defined using the  $^{10}\text{B}(n, \alpha \gamma)^7\text{Li}^*$  reaction. The neutron flux was defined with area of the full absorption peak of gamma rays with the 478 keV energy (the excited state of  $^7\text{Li}$ ). Gamma ray spectrometer with two Ge(Li) -detectors DGDK-32B was placed at the 2<sup>nd</sup> horizontal channel and used to determine the incident neutron flux. The spectrometer was calibrated on energy and efficiency using the sources  $^{133}\text{Ba}$  and  $^{152}\text{Eu}$  from the SOSGI-M standard set. The value of the absolute efficiency of gamma registration was  $(3.08 \pm 0.17) \cdot 10^{-5}$ .

Cross section of the  $^{10}\text{B}(n, \alpha \gamma)^7\text{Li}^*$  reaction for neutrons that passed the filter, was calculated as an average of the point-wise cross sections, taken from the evaluated nuclear data libraries CENDL-2, ENDF/B-VI, JENDL-3.3 and JEFF-3.1. An average

was done over the shape of neutron filtered beam. The calculation was done using the PREPRO2002 software package. The received values of the cross section for  $^{10}\text{B}(n, \alpha\gamma)^7\text{Li}^*$  reaction were found as 13.61 barn for 2 keV filter and 2.35 barn for 59 keV filter.

Definition of the filtered neutron flux, that pass the interference filter and falls to the target, and its monitoring resolved in two steps:

- Definition of the neutron flux before and after irradiation using the  $^{10}\text{B}(n, \alpha\gamma)^7\text{Li}^*$  reaction; flux monitoring during irradiation of the B-10 target;
- Flux monitoring during irradiation of the tantalum targets.

During all time of irradiation of the tantalum targets, simultaneously the monitoring of the neutron flux has been done by the neutron spectrometer with  $^3\text{He}$ -counter. The measurements of the neutron beam have been done every hour during 3000 s. The  $^3\text{He}$ -peak areas were used as the weighting functions to define the neutron flux, under which the pile of the tantalum targets has been placed. The neutron flux, taken from the measurements due to the boric target before and after irradiation, was averaged by these weighting functions, and the average neutron flux has been defined during total irradiation time.

The irradiation of the tantalum targets was done at the 2<sup>nd</sup> horizontal channel of the KRR. The targets were set up as a single pile from 4 disks for 2 keV and 8 disks for 59 keV, dispose one by one. Exposing the several disks, not just one, allows us to take into account an effect of a resonance self-shielding in tantalum. Activation of tantalum samples was done during 84 hours for 2 keV and 62 + 84 hours for 59 keV, then the targets were removed to the clean zone for the further measurements.

In order to protect the targets from the background neutrons, thermalized in the reactor hall, the pile was exposed inside the system of the protective screens. The external screen was made of paraffin with lithium hydride ( $^6\text{LiH}$ ), and the internal one – of lithium carbonate ( $^6\text{Li}_2\text{CO}_3$ ). Lithium, enriched by  $^6\text{Li}$  to 85% was used in both systems.

As a product of the  $^{181}\text{Ta}(n, \gamma)^{182}\text{Ta}$  reaction the radioactive isotope  $^{182}\text{Ta}$  was received. It decays into the tungsten ( $^{182}\text{W}$ ) with 114.74 days half-life; the gamma-transition intensities of  $^{182}\text{W}$  were measured. For determination of sample activities, nine gamma lines of tungsten-182 were selected: five "soft" lines with energies 152, 179, 222, 229 and 264 keV and four "hard" lines with energies 1121, 1189, 1221 and 1231 keV.

Gamma spectra of the irradiated samples were measured by gamma-spectrometer with Ge(Li)-detector. The spectrometer parameters were as follow:

- Detector type – DGDK-45A;
- Energy resolution – 3.4 keV at 1332 keV gamma-line;
- Lead shield with 15 cm. thickness.

Calibration of the gamma-spectrometer on energy and efficiency was done using etalon sources from the standard set OSGI-3-2. The following sources were used:  $^{133}\text{Ba}$ ,  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ ,  $^{152}\text{Eu}$ ,  $^{228}\text{Th}$ , which covered the energy range from 53 to 2614 keV.

The samples were located 5 cm above the detector cap. The measurements were carried out by series. Each series consists of 8 measurements. In series each of the samples was measured twice: «face» down and «face» up. A «face» is the sample side, turned to incident neutrons during irradiation. Before and after each measurement series the background spectrum was controlled. Acquisition time for each spectrum was 10 000 s for 2 keV and 50 000 s for 59 keV. The spectra were measured in a clean zone (gamma background was less then 0.07 mR/h).

### 3 Data analysis and results

The procedure of the capture cross-section determination for  $^{181}\text{Ta}(n,\gamma)^{182}\text{Ta}$  reaction from measured experimental gamma ray spectra was made according to the following algorithm:

- Finding of full absorption peak (FAP) areas for selected gamma lines.
- Calculation of activities for each sample using every selected line.
- Make correction to the obtained activity for self-absorption of gamma rays in the sample.
- Calculation of the “observed” cross-section values for  $^{181}\text{Ta}(n,\gamma)^{182}\text{Ta}$  reaction for each orientation of each sample using all selected lines.
- Average findings over all selected gamma-lines for each sample and obtain values of “observed” capture cross-section for  $^{181}\text{Ta}(n,\gamma)^{182}\text{Ta}$  reaction for each sample.
- Approximate the “observed” capture cross-section dependence on summary thickness of tantalum before samples and extrapolate this dependence to zero thickness of tantalum.
- Make correction to the obtained capture cross-section for contribution of the impurity lines in the neutron spectrum and for backward scattering of neutrons by the output collimator.

The WinSpectrum v.1.5 code was used for processing of the measured gamma ray spectra. This software works in an interactive mode, i.e., it allows to control visually all stages of processing and to choose limits in FWHM, shape of a background distribution under peak, position of a peak center, parameters of both left and right peak “tails”, etc. This code was created by M.V. Strilchuk (KINR, Nuclear Structure Department).

The activity of each sample at the end of irradiation time, calculated for the  $i^{\text{th}}$  gamma-line, was defined with the formula:

$$A_{t_0}^i = \frac{S_{FAP}^i}{I_{\gamma}^i \cdot \epsilon_{\gamma}^i \cdot t_{acq}} \cdot e^{-\lambda \cdot t_D}, \quad (1)$$

where  $A_{t_0}^i$  – activity of the target obtained for the  $i^{\text{th}}$  gamma-line;  
 $S_{FAP}^i$  – area of the full absorption peak of the  $i^{\text{th}}$  gamma-line;  
 $I_{\gamma}^i$  – absolute intensity for the  $i^{\text{th}}$  gamma-line;  
 $\epsilon_{\gamma}^i$  – absolute efficiency for the  $i^{\text{th}}$  gamma-line;

$t_{acq}$  – time of target measurement;  
 $t_D$  – delay time, i.e., time from the finish of irradiation to the beginning of measurement;  
 $\lambda$  – decay constant for  $^{182}\text{Ta}$ .

As the tantalum sample has a non-zero thickness, the obtained activity has to be corrected on self-absorption of gamma-rays. To consider this effect, both the tabulated and experimentally obtained attenuation coefficients of gamma-rays were used. An adjustment for self-absorption of gamma-rays was carried out with the formula:

$$k_{at}^i = \frac{\mu^i x}{1 - e^{-\mu^i x}} \quad (2)$$

where  $k_{at}^i$  – self-absorption coefficient for gamma-rays of the  $i^{\text{th}}$  gamma-line in the sample;  $\mu^i$  – mass self-absorption coefficient of the  $i^{\text{th}}$  -gamma-line;  $x$  – thickness of the target.

The “observed” cross section values for  $^{181}\text{Ta}(n, \gamma)^{182}\text{Ta}$  reaction for each orientation of each sample were calculated by the following formula:

$$\sigma_i^{ob} = \frac{A_i}{\Phi \cdot n_i^n \cdot (1 - e^{-\lambda t_e})} \quad (3)$$

where  $A_i$  – activity of  $i$ -th sample, reduced to the end of irradiation;

$\Phi$  – neutron flux at the 1<sup>st</sup> sample;

$n_i^n$  – number of nuclei in  $i$ -th sample;

$\lambda$  – decay constant;

$t_e$  – time of spectrum acquisition;

$\sigma_i^{ob}$  – “observed” capture cross-section determined for  $i$ -th sample.

As stated above the main neutron lines, created by the 2 keV and 59 keV filters, lie in the energy ranges 0.24-3.0 keV and 54.8-60.1 keV, respectively. In these energy ranges, the total neutron cross-section for  $^{181}\text{Ta}$  has the resonance structure, so the resonance self-shielding of neutrons in the tantalum samples is present, and thus, so-called “observed cross-section” depends on the sample thickness. To take into account this effect, we set up more than one target, but a pile of samples. Depending on the position of the sample in the pile, decreasing of the “observed” cross section for 2 keV was about 2.4, 7.3, 12, and 17%, respectively. And for 59 keV decreasing of the “observed” cross section was about 0.4, 1.8, 2.2, 2.9, 3.4, 5.0, 6.3 and 7.1%, respectively. The real values of cross-sections were received by extrapolation the “observed” cross sections to zero thickness of the samples.

A share of the main neutron line in the total spectrum is 95.47% for 2 keV and 94.36% for 59 keV. So, the obtained cross-section for tantalum responds to the entire filter spectrum. To estimate a share of cross-section from the impure lines, a numerical simulation was performed. The point-wise cross-section data for the  $^{181}\text{Ta}(n, \gamma)^{182}\text{Ta}$  reaction from the evaluated nuclear data libraries BROND-2, CENDL-2, ENDF/B-VI, JENDL-3.3, and JEFF-3.1 were averaged over total neutron spectrum after the filter and over the main filter line, then the values  $f = \langle \sigma \rangle_{\text{line}} / \langle \sigma \rangle_{\text{spectrum}} - 1$  were calculated for each from these 4 libraries, and their average value was considered as additional systematic

inaccuracy in the obtained cross-section. The results of these calculations are presented in Table 1.

Table 1. Result of numerical simulation to take into account the contribution of the impurity lines.

Library	2 keV filter			59 keV filter		
	$\langle \sigma \rangle_{\text{spectrum}}$ , [b]	$\langle \sigma \rangle_{\text{line}}$ , [b]	$\langle \sigma \rangle_{\text{line}} - \langle \sigma \rangle_{\text{spectrum}}$	$\langle \sigma \rangle_{\text{spectrum}}$ , [b]	$\langle \sigma \rangle_{\text{line}}$ , [b]	$\langle \sigma \rangle_{\text{line}} - \langle \sigma \rangle_{\text{spectrum}}$
			$\langle \sigma \rangle_{\text{spectrum}}$ [%]			$\langle \sigma \rangle_{\text{spectrum}}$ [%]
BROND-2	6.060	6.292	3.8	0.4463	0.4624	3.6
CENDL-2	5.935	6.162	3.8	0.4302	0.4424	2.8
ENDF/B-VI	6.597	6.836	3.6	0.4302	0.4424	2.8
JENDL-3.3	5.935	6.162	3.8	0.4722	0.4888	3.5
Average	6.132	6.363	3.78	0.4447	0.4590	3.2

Corrections for backward scattering of neutrons by the output collimator were computed with program package MCNP-4C for real irradiation conditions. The obtained values are less than 0.3% for the 2 keV filter and about 3.7% – for the 59 keV filter.

The experimental value of the  $^{181}\text{Ta}(n, \gamma)^{182}\text{Ta}$  reaction cross-section for the energy range of the 2 keV filter (0.24-3.0 keV) together with the similar experimental data of other authors and the ENDF libraries data, averaged over our 2 keV neutron spectrum, are presented in Table 2.

Table 2. The  $^{181}\text{Ta}(n, \gamma)^{182}\text{Ta}$  reaction cross-section for the energy range of the 2 keV filter.

Author or library	Moxon+ [2]	Konks+ [3]	BROND-2	CENDL-2	ENDF/B-VI	JENDL-3.3	JEFF-3.1	This work
$\langle \sigma \rangle_{\text{line}}$ , b	6.26± ± 0.26	8.52± ± 0.81	6.06	5.94	6.60	5.94	5.94	6.54± ± 0.36

As it is seen from the Table 2, our value  $\sigma_{\text{exp}} = 6.54 \pm 0.36$  b is in good agreement with the result of the measurement [2] within the errors and it lays close to the averaged value taken from ENDF/B-VI.

The experimental value of the  $^{181}\text{Ta}(n, \gamma)^{182}\text{Ta}$  reaction cross-section for the energy range of the 59 keV filter (54.8-60.1 keV) together with the similar experimental data of other authors and the ENDF libraries data, averaged over our 59 keV neutron spectrum, are presented in Table 3.

Table 3. The  $^{181}\text{Ta}(n, \gamma)^{182}\text{Ta}$  reaction cross-section for the energy range of the 59 keV filter.

Author	M.Bokhovko+ [4]	G.Reffo+ [5]	V.Kononov+ [6]	V.Kononov+ [7]	J.Miskel+ [8]	This work
$\langle \sigma \rangle_{\text{line}}$ , b	0.500± ± 0.025	0.440± ± 0.050	0.558± ± 0.047	0.416± ± 0.042	0.431± ± 0.043	0.480± ± 0.041
Library	BROND-2	CENDL-2	ENDF/B-VI	JEFF-3.1	ENDF/B-VII	JENDL-3.3
$\langle \sigma \rangle_{\text{line}}$ , b	0.462	0.442	0.442	0.489	0.442	0.489

As it is seen from the Table 3, our value  $\sigma_{\text{exp}} = 0.480 \pm 0.041$  b is in good agreement with the results of other measurements [4, 5, 8] within the errors. Also within the experimental errors it is in good agreement with the averaged values taken from all ENDF libraries.

The capture cross- sections of  $^{181}\text{Ta}$  for neutrons with the average energies 1.97 keV and 59 keV have been measured by the activation method using gamma-spectrometer with Ge (Li)-detector with errors about 5.4% and 8.5%, less than required by Request List [1].

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