EXPERIMENTAL INVESTIGATIONS OF NEUTRON CROSS SECTIONS FOR TUNGSTEN ISOTOPES ATOMIC NUCLEI: RADIOACTIVE ^{181}W ($T_{1/2}$ = 121.2 DAYS) AND STABLE ^{180}W

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The aim of this paper was the study of the slow neutron interactions with the radioactive isotope ^{181}W ($T_{1/2}$ = = 121.2 days), any data on the neutron cross sections for these interactions are not available. This isotope in macroscopic quantity may be accumulated during the irradiation of the stable isotope ^{180}W in the reactor neutron flux. The data on the neutron cross sections for ^{180}W are needed to evaluate the kinetics of this process. These data are very limited; therefore, the study of all possible cross sections for this stable isotope was the first step of our investigation.

The rare isotope of Tungsten ^{180}W (0.12 % in natural material) was studied on the Kyiv Research Reactor to define the total neutron cross section in the thermal / epithermal region and the parameters of resolved resonances in the region up to 100 eV. Metal powder of Tungsten, enriched with ^{180}W up to 95.1 %, was used to prepare two samples for transmission measurements. The total neutron cross section at the energies 0.025 - 2.5 eV and the resonance parameters in the energy region 2.5 - 87 eV were obtained with rather good accuracy. Then the repacked sample (4.338 g) of this isotope was irradiated in the reactor neutron fluence $4 \cdot 10^{20}$ n/cm^2 to get the radioactive ^{181}W.

The time of flight method with fast chopper and 70 m flight path was used to measure the transmissions of the stable and irradiated samples. All the measurements for the stable and irradiated samples were carried out at the same conditions. The content of the accumulated ^{181}W was evaluated; the results for stable ^{180}W and radioactive ^{181}W and their analysis are presented here in comparison with the previously known data.

1. Introduction

This work has been done at Kyiv Research Reactor (KRR) in the framework of the Program for reception of reactor radioisotopes and technology development to produce the radionuclides for different areas of use: science, medicine, industry and other applications [1].

The values of neutron cross sections for investigated nuclei just as the values of neutron fluxes and their spectra determine the process of radionuclides accumulation. At the same time the expermental investigations have to include as the target nuclei, so as the accompanying ones (present in the irradiated sample and accumulated during the irradiation). The large volume of such investigations has been done at KRR earlier [2, 3].

The aim of this work was the investigation of neutron cross section of tungsten isotopes: radioactive ^{181}W($T_{1/2}$ = = 121.2 days) and stable ^{180}W. Neutron data for these isotopes were very scant [4, 5], as the starting stable isotope ^{180}W is in low abundance (0.12 % in the natural tungsten). In this work the enriched in ^{180}W samples were used for neutron cross section measurements.

Radioisotope ^{181}W for investigations was accumulated in KRR during the irradiation of ^{180}W samples with neutrons under such transmutations:

$$^{180}\mathrm{W}(n, \gamma)\ ^{181}\mathrm{W}(T_{1/2} = 121.2 \text{ days}) \xrightarrow{\beta^-} {}^{181}\mathrm{Ta}. \quad (1)$$

The TOF neutron spectrometer using fast chopper was used in the measurements at the horizontal channel of KRR together with the special installation for high activity samples [6].

2. Samples and investigation proceedings

As the stable ^{180}W is the starting isotope for radioactive ^{181}W accumulation, so the first necessary step was the investigation of starting ^{180}W neutron cross section.

The samples for investigations were prepared with W metal powder, enriched in ^{180}W to 95,1 %.

To remove the system moisture in powder a drying box with ntemperature up to 150 °C was used. Then the powder was packed and pressed in special container from aluminium alloy. The parameters of these samples are presented in Table 1.

Table 1. **Parameters of tungsten-180 samples**

No.	Composition and weight, mg	Dimentions, mm	Content of ^{180}W, n/cm^2 · 10^{-22}	
			Thick direction	Thin direction
1.	^{180}W(95.1 %) - 2547	2.06 × 14.87 × 13.19	2.6227 (L = 13.9 mm)	–
2.	^{180}W(95.1 %) - 4338	2.06 × 15.0 × 23.5	4.401 (L = 23.5 mm)	0.392 (L = 2.06 mm)

Content of other tungsten isotopes in samples was 4,8 % for ^{182}W and 0,1 % for ^{184}W (in accordance with certificate). It is seen that the samples had the parallelepiped form, and this gave the possibility to conduct the measurements with different thicknesses of sample depending of orientation at incident neutron beam. The used directions are shown in Table 1.

The used Neutron TOF Spectrometer had the following parameters: fast chopper with diameter 300 mm, flight path – 69.74 m, energy resolution 1.0 mcs/m, 0.22 mcs/m and 0.055 mcs/m.

Transmission measurement data may be received in the energy range 0.025 eV to 30 keV. All the details including the installation for high active samples may be found in our paper [7].

3. Analysis of experimental results

The measured transmission data were used for calculations of total neutron cross sections in thermal energy region and also for identification and definition the neutron resonance parameters of tungsten isotopes. For calculations in thermal and epithermal regionn (0.025 - 2.5 eV) the following expression was used:

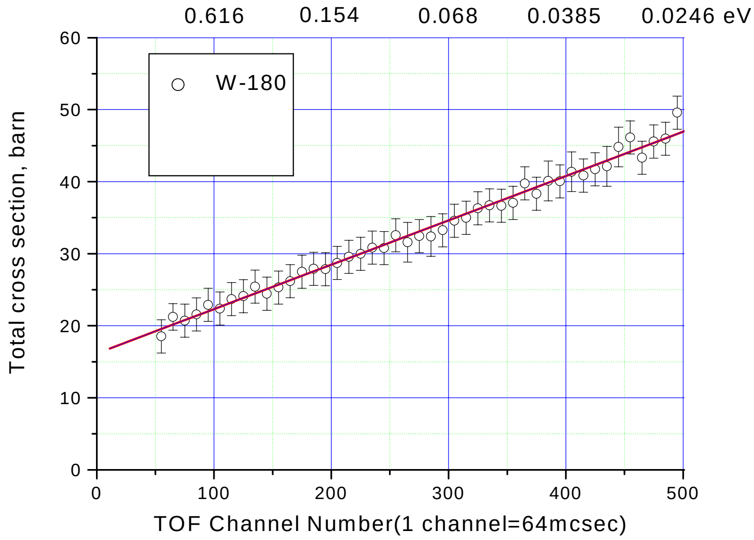
$$\sigma_t^i (E_n) = \ln T_i (E_n) / n_{180}, \tag{2}$$

where σ_t^i – the observed total neutron cross section for sample (related to ^{180}W content); T_i – sample transmission at neutron beam; n_{180} – content of ^{180}W (in neutron beam direction).

Analysis and calculations for neutron resonances were fulfilled using shape method in accordance with single level Breit - Wigner formalism.

3.1. Neutron data for stable ^{180}W isotope

For stable ^{180}W using expression (2) it was calculated the energy dependence of the observed total neutron cross section for investigated tungsten sample. For energy 0.0253 eV it is (48.0 ± 2.3) barn. Correction on admixture of ^{182}W(4,8 %) in sample is 1.4 barn. All other admixture corrections may be neglected. So for ^{180}W at the energy 0.0253 eV we receive the value σ_{tot} = (46.6 ± 2.3) barn. Full energy dependent σ_{tot} values for ^{180}W in the energy range 0.025 - 2.5 eV are presented in the Figure.



Total neutron cross section for ^{180}W (0.025 - 2.5 eV).

Capture cross section value was also calculated for ^{180}W. Potential scattering cross section $\sigma_{pot} = 4\pi R_o^2$ = 9,29 ± ± 0.4 barn, using the value $R_o^{'}$ = 8,6 ± 0,2 fm from [5]. Total scattering cross section may be more or less of this value, but for the first approximation it may be used as $\sigma_s \approx \sigma_{pot}$. After this for ^{180}W at the energy 0.0253 eV the following value is obtained:

$$\sigma_\gamma^o = 46.6 - 9.3 = 37.3 \pm 2.4 \text{ barn.}$$

This value may be used as σ_γ for ^{180}W, as the known value 30 barn [4] has the uncertainty 400 %. Recently, after this investigation has been finished, one more experimental result was published for ^{180}W - σ_γ^r = 21.5 (2.5) barn (capture cross section averaged over Maxwellian spectrum at 0.0253 eV) [9].

Analysis of transmission measurements in resonance region was fulfilled and the results are presented in Table 2.

Table 2. **Neutron resonances of ^{180}W and admixture isotopes**

Isotope	This work			Atlas 2006 [ ]		
	E_0, eV	Γ_γ, MeV	Γ_n^o, MeV	E_0, eV	Γ_γ, MeV	Γ_n^o, MeV
^{182}W	4.140 ± 0.001	59.3 ± 4.0	0.63 ± 0.03	4.155 ± 0.005	48 ± 3	0.72 ± 0.02
^{180}W	15.88 ± 0.01	(70)	2.63 ± 0.03	15.9 ± 0.1	(70)	3.1 ± 0.4
^{186}W	18.78 ± 0.01	(43.8)	66.3 ± 1.5	18.83 ± 0.3	43.8 ± 1,4	69.1 ± 2.3
^{182}W	21,03 ± 0,01	(65)	7.59 ± 0.14	21.06 ± 0.03	65 ± 6	8.5 ± 0.2
^{180}W	49.04 ± 0.01	(70)	0.75 ± 0.03	49.3 ± 0.1	(70)	0.88 ± 0.10
^{180}W	62.25 ± 0.03	(70)	0.16 ± 0.01	62.7 ± 0.2	(70)	0.18 ± 0.03
^{180}W	74.70 ± 0.02	(70)	3.55 ± 0.09	75.2 ± 0.2	(70)	4.0 ± 0.2
^{180}W	86.81 ± 0.05	(70)	0.50 ± 0.05	87.4 ± 0.3	(70)	0.78 ± 0.05

Among observed resonances one can see the one belonging to ^{186}W, whose content in the sample is not shown in certificate of tungsten (accuracy low limit - 0.1 %). But this resonance is very strong (Γ_n = 300 MeV [4], or even 315.6 MeV as it follows from recent measurements[8]), so the amount of ^{186}W in sample is about 0.06 %, less than low limit of certificate, it is enough to bee seen in our measurements.

The observed neutron widths of ^{180}W are 15 % less than those from Atlas 2006 [ ], but as our sample had high enrichment 95.1 % in compare with 6.93 % in [4], we recommend our data, as more reliable. Resonance Integral, calculated using these data, was found as I_γ = 194.5 ± 6.0 barn in compare with Atlas 2006, where this value is I_γ = 214 ± 30 barn.

3.2. Neutron data for radioactive isotope ^{181}W($T_{1/2}$ = 121.2 days)

Neutron parameters of radioactive ^{181}W were studied with joint analysis of two transmission curves of sample N2 (^{180}W - 4.401 · 10^{22} n/cm^2), measured accordingly till and after the irradiation in neutron flux. To accumulate ^{181}W, the sample was irradiated in the vertical experimental channel N46/60, where the undisturbed neutron flux density was 6 · 10^{13} n/cm^2 s. Total irradiation time can be considered as 116.15 days, reduced to nominal reactor power 10 MWt, and nominal total fluence as 6 · 10^{20} n/cm^2. But the real fluence was less due to the effects of self-shielding in the sample and it was defined in analysis of well known neutron resonances of stable isotopes observed in our transmission curves.

Using the obtained data for ^{180}W - σ_γ^o and I_γ, the dynamics of isotope transmutations during the neutron irradiation in accordance with expression (1) was calculated and presented in Table 3.

Table 3. **Transmutations in ^{180}W at the irradiation with reactor neutrons**

Neutron fluence, 10^{20} см$^{-2}$	^{180}W, %	^{181}W, %	^{181}Ta, %
0,3	99,88	0,11	-
0,9	99,64	0,33	0,03
1,5	99,40	0,52	0,07
2,1	99,16	0,70	0,13
2,7	98,92	0,86	0,20
3,3	98,68	1,00	0,29
3,9	98,44	1,14	0,39

Table 4. **^{181}W capture cross section calculated with resonance parameters at E_n = 3.18 eV**

E_n , eV	σ_γ , barn
2.46	10.58
0.615	1.66
0.273	1.95
0.154	2.39
0.098	2.88
0.068	3.40
0.050	3.92
0.038	4.43
0.030	4.96
0.0246	5.49

Sample transmissions at ^{180}W after irradiation in reactor flux showed new resonances with energies 3.18, 4.20, 10.26 eV. The last two belong to stable ^{181}Ta, accumulated in the sample. Neutron parameters of these resonances are well known [4], so using these data it is possible to determine the content of ^{181}Ta and the corresponding fluence (see Table 3).

The content of ^{181}Ta thus calculated appeared to be 0.4 % relative to ^{180}W. This corresponded to neutron fluence 3.9 × × 10^{20} n/cm^2 and accumulation of ^{181}W − 1.14 % (to ^{180}W) and nuclear concentration 5.1 · 10^{20}nucl/cm^2. This value was used for normalization of neutron parameters for resonance at 3.18 eV, which was identified, as ^{181}W neutron resonance. As a result, the following resonance parameters were obtained: E_o = 3.18 ± ± 0.01 eV; $g\Gamma_n^o$ = 0.148 ± 0.006 MeV; Γ_γ = 89 ± 18 MeV.

Other possible resonances of ^{181}W were not identified. This may be explained with the small concentration of ^{181}W in sample, then with the screening of energy range 3 - 100 eV by ^{180}W, ^{182}W, ^{186}W and ^{181}Ta neutron resonances. Nevertheless, as a rule, the basic contribution to capture cross section in thermal region follows from the lowest resonances, so it was calculated this cross section in the energy range 0,025 - 2,5 (Table 4) using the parameters of this single resonance.

4. Comparative analysis

The comparative analysis of obtained results and averaged nuclear parameters of other tungsten isotopes (Table 5) gave the possibility to do some evaluations for radioactive ^{181}W.

Table 5. **Average neutron-nuclear parameters of tungsten isotopes**

W isotope	σ_γ^o, barn	σ_s, barn	g_γ	D, eV	Γ_γ, MeV	S_n, keV	U_n, keV	R_o', fm
180	37.3 ± 24 *	9.29 ± 0.4	(1,0) *	22.5 ± 6.5	(70)	6681	5690	8.6 ± 0.2
181	5.4 *	(8.95) *	-	≈ 10 [10]	89 ± 19 *	8064	5503	(8.4) *
182	20.7 ± 0.5	8.84 ± 0.3	1.0031	66 ± 4	54 ± 5	6190	5282	8.3 ± 0.2
183	10.1 ± 0,3	2.4 ± 0.6	0.9989	12 ± 1	70 ± 10	7411	5136	8.1 ± 0.2
184	1.7 ± 0.1	7.35 ± 0.17	0.9989	81 ± 5	57 ± 4	5753	4462	8.0 ± 0.2
186	37.9 ± 0.6	0.147 ± 0.012	1.0014	87 ± 7	50 ± 5	5466	4746	7.6 ± 0.2

In Table 5 the data in columns 2 - 6 are taken from Atlas 2006, excluding our results marked by *; the data in the columns 7 - 8 came from nuclear mass tables and in column 9 – from experimental work [5].

The nearest-neighbor nuclei with close parameters to ^{181}W is the nucleus of ^{183}W, also even-odd. Both nuclei have rather small thermal capture cross sections (≈ 10 barn).

Radiation widths are also close, though ^{181}W has somewhat higher excitation energy – see column 8 (radiation energy is also higher). Level density systematic [10] give for ^{181}W the average level distance 10 eV, and this is close to ^{183}W, where this value equals to 12 eV.

It is worth to note, that the linear dependence of potential scattering radius R_o' from mass number A takes place. The interpolation for ^{181}W gave the value 8.4 fm, then the potential scattering cross section has to be $\sigma_{pot} = 4\pi\ (R_o')^2 =$ = 8.95 barn.

The total neutron cross section for thermal neutrons may be σ_{tot} = 895 + 5.50 = 14.45 barn. This is very rough approximation until new data appear.

For stable ^{180}W the value σ_{pot}= 9.29 barn, while the interpolation to zero energy our $\sigma_{tot}(E)$ (see Figure) gives the value of scattering cross section equal to 16.1 barn. The difference 16.1 - 9.3 = 6.8 barn may be the contribution of low lying levels and also bound level, showed in [4].

5. Summary

^{180}W (stable):

- The first $\sigma_{tot}(E)$ dependence was received at thermal and epithermal energies and with good accuracy (5 %).
- Resonance parameters were received with better accuracy up to 90 eV.

^{181}W(radioactive):

- First investigation of neutron parameters at macroscopic sample of ^{181}W (about 50 mg).
- Resonance parameters of the lowest neutron level are measured and possible capture cross section in the energy range 0.025 - 3.2 eV is evaluated.

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