Measurements of the thermal neutron cross-sections and resonance integrals for ${}^{186}W(n,\gamma) {}^{187}W$ and ${}^{98}Mo(n,\gamma) {}^{99}Mo$ reactions

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Abstract The thermal neutron cross-sections and resonance integrals of the ¹⁸⁶W (n,γ) ¹⁸⁷W and ⁹⁸Mo (n,γ) ⁹⁹Mo reactions in the thermal and 1/E regions, respectively, of a thermal reactor neutron spectrum have been experimentally determined by the activation method using ¹⁹⁷Au (n,γ) ¹⁹⁸Au reaction as a single comparator. The high purity natural W, Mo, and Zr foils; and Au wire diluted in aluminum, were irradiated without Cd shield in two neutron irradiation sites, characterized with different values for the thermal-to-epithermal flux ratios, f at the Second Egyptian Research Reactor (ETRR-2). The induced activities in the samples were measured by high-resolution y-ray spectrometry with a calibrated germanium detector. Thermal neutron cross-sections for 2200 m/s neutrons and resonance integrals for the ¹⁸⁶W (n,γ) ¹⁸⁷W and ⁹⁸Mo (n,γ) ⁹⁹Mo reactions have been obtained relative to the reference values, $\sigma_0 = 98.65 \pm 0.09$ b and $I_0 = 1500 \pm 28$ b for the ¹⁹⁷Au (n,γ) ¹⁹⁸Au reaction. The necessary correction factors for thermal neutron and resonance neutron selfshielding effects, and the epithermal flux index (α) were taken into account in the determinations. The results obtained were: $\sigma_0 = 38.43 \pm 0.4$ b and $I_0 = 502 \pm 65$ b for ${}^{186}W$ (n,γ) ${}^{187}W$, and $\sigma_0 = 0.137 \pm 0.014$ band $I_0 = 6.47 \pm 0.8$ for ⁹⁸Mo (n,γ) ⁹⁹Mo. These results are discussed and compared with previous measurements and evaluated data in literature. The traditional method of determining thermal cross-sections and resonance integrals via neutron irradiation with and without Cd shield in one

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irradiation position was avoided in this work by neutron irradiation without Cd shield in at least two different neutron irradiation positions. This method provides alternative way for determining thermal cross-sections and resonance integrals simultaneously.

Keywords Cross-section · Resonance integral · Self shielding · Flux ratio

Introduction

Tungsten (W) and Molybdenum (Mo) are important structural materials for fusion reactors, accelerator-driven systems and isotope productions [1]. W is used as the target material of the electron accelerator for producing bremsstrahlung and neutrons [1]. Rhenium-188, (¹⁸⁸Re, $t_{1/2} = 16.8$ h) is available from the ¹⁸⁸W/¹⁸⁸Re generator system, made via double neutron capture on ¹⁸⁶W [2, 3]. Mo is very useful as a refractory and corrosion resistant material in accelerator applications [4, 5]. ⁹⁹Mo ($t_{1/2} = 65.94$ h) is produced by the ⁹⁸Mo (n,γ) ⁹⁹Mo reaction [5, 6]. It decays by beta emission to ^{99m}Tc ($t_{1/2} = 6.015$ h). ^{99m}Tc is the most important nuclide used for diagnostic purposes.

The knowledge of the thermal neutron cross-sections and the resonance integrals for ¹⁸⁶W (n,γ) ¹⁸⁷W and ⁹⁸Mo (n,γ) ⁹⁹Mo reactions are important, because these data is used in the production of ⁹⁹Tc and ¹⁸⁸Re and may also used in other studies related to the interaction of neutrons with matter. The thermal neutron cross-section and resonance integral of the ¹⁸⁶W (n,γ) ¹⁸⁷W reaction are important in the calculations of decay heat data and evaluating the radiation damage of the material [7–9]. There are some discrepancies among the experimental

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data for thermal neutron cross-sections and the resonance integrals for the ¹⁸⁶W (n,γ) ¹⁸⁷W and ⁹⁸Mo (n,γ) ⁹⁹Mo reactions [10–17], especially among the resonance integral values.

Measuring thermal neutron cross-sections, σ_0 and resonance integrals, I_0 are often performed by the activation method. It is based on irradiation target samples with and without Cd shield in the neutron field from either a reactor or a neutron source. Neutron spectrum parameters characterizing irradiation position should be well-known. There is an alternative scheme to measure simultaneously thermal neutron cross-sections and resonance integrals without irradiation under Cd shield. The target samples can be irradiated in different positions having different values for thermal-to-epithermal flux ratios, *f*. Measuring the induced activities provides coupled equation systems, where σ_0 and I_0 are unknown variables and can be obtained.

Aim of this work is determining thermal neutron crosssections and resonance integrals for ¹⁸⁶W (n,γ) ¹⁸⁷W and ⁹⁸Mo (n,γ) ⁹⁹Mo reactions. The method of neutron irradiations of the target samples without Cd shield in two positions is used as an alternative and instead of the neutron irradiations of the target samples with and without Cd shield in one position. The correction factors of thermal neutron self-shielding ($G_{\rm th}$), and resonance neutron selfshielding ($G_{\rm e}$) effects, and the epithermal neutron spectrum shape factor (α) were taken into account.

Theory

The flux density in a thermal reactor can be characterized by two components, a temperature dependent Maxwellian thermal neutron component and an epithermal neutron slowing down component with an ideal 1/E distribution. A number of formalisms [18–20] have been suggested to describe the reaction rates. A simple convention proposed by Høgdahl [18] for 1/v nuclides is used for the present purpose. According to it, the reaction rate per target nuclei, *R*, of a sample irradiated by reactor neutrons is described by [21, 22]:

$$R = R_{\rm th} + R_{\rm e} = G_{\rm th}\phi_{\rm th}\sigma_{\rm o} + G_{\rm e}\phi_{\rm e}I_{\rm o}(\alpha), \qquad (1)$$

where $R_{\rm th}$ (= $\phi_{\rm th} \sigma_{\rm o}$) is the reaction rate induced by pure thermal, $R_{\rm e}$ (= $\phi_{\rm e} I_0(\alpha)$)) is the reaction rate induced by epithermal neutrons, $G_{\rm th}$ and $G_{\rm e}$ are the thermal and epithermal neutrons self shielding correction factors respectively, and the parameter α , which is independent on the neutron energy [21, 22], corrects for the deviation of the epithermal neutrons from the ideal 1/*E* law.

The reaction rate is measured by Ge detector using the expression:

$$R = (N_{\rm p}M) / (N_{\rm a}t_{\rm m}WSDC\varepsilon_{\rm p}\theta\gamma), \qquad (2)$$

where N_p is the net number of counts in the full-energy peak, W is the weight of the sample, t_m is the measuring time, $S = 1 - \exp(-\lambda t_{irr})$, λ is the decay constant, t_{irr} is the irradiation time, $D = \exp(-\lambda t_D)$, t_D is the decay time, $C = 1 - \exp(-\lambda t_m)/\lambda t_m$, M is the atomic weight, γ is gamma ray intensity, ε_p is the full-energy peak detection efficiency, θ is the isotopic abundance and N_a is Avogadro's number. Similarly the reaction rate for Au as a comparator can be written. Thermal neutron cross-section as a function of the resonance integral for any element x, with respect to Au as a comparator is given by:

$$\sigma_{0,x} = [(G_{\text{th},au}\sigma_{0,au}/G_{\text{th},x}) + ((G_{e,au}\sigma_{0,au}I_{0,au}(\alpha))/(fG_{\text{th},x}))]K - (G_{e,x}I_{0,x}(\alpha)/fG_{\text{th},x}),$$
(3)

where f is the thermal to epithermal neutron flux ratio, and the factor K is given by the relation:

$$K = [(N_{\rm p}/t_{\rm m}WSDC\varepsilon_{\rm p})_x/(N_{\rm p}/t_{\rm m}WSDC\varepsilon_{\rm p})_{\rm au}](M_x\theta_{\rm au}\gamma_{\rm au}) /(M_{\rm au}\theta_x\gamma_x)$$
(4)

To solve Eq. 3, which has two unknown variables, $\sigma_{0,x}$ and $I_{0,x}(\alpha)$, two coupled equations are necessary. This is achieved by neutron irradiation of the target samples in at least two positions characterized with different values for the flux ratio *f*. The solution can be obtained graphically, from the plots of $\sigma_{0,x}$ versus $I_{0,x}(\alpha)$, which intersect in a unique point giving simultaneously the values of $\sigma_{0,x}$ and $I_{0,x}(\alpha)$.

Experimental measurements of the reaction rates of activation detectors with well-known nuclear data lead to the determination of the neutron flux parameters at the irradiation site and consequently to the measurement of thermal neutron cross-section and resonance integral of the element of interest. It is to be noted that the conversion of $I_0(\alpha)$ to I_0 is necessary to provide a resonance integral independent of the irradiation position, which can then be compared to literature values. This conversion can be made by using the following expression:

$$I = \bar{E}_{\rm r} \alpha \left[I_0(\alpha) - \left(0.316 \,\sigma_0 / \sqrt{E_{\rm cd}} \right) \right. \\ \left. \times \left((1/(2\alpha + 1)E_{\rm cd}^{\alpha}) - \bar{E}_{\rm r}^{\alpha} \right) \right], \tag{5}$$

where $\bar{E}_{\rm r}$ is the effective resonance energy and $E_{\rm cd}$ is the cadmium cut off energy (=0.55 eV).

The necessary corrections factors for thermal and epithermal neutron self shielding are take into account. The thermal neutron self-shielding correction factor for thin slabs was calculated as follows [23]

$$G_{\rm th} = (1 - \operatorname{Exp}(-\zeta))/\zeta, \tag{6}$$

where $\zeta = 2/\sqrt{\pi \sum_0 t}$, \sum_0 is the macroscopic crosssection for thermal neutrons (E = 0.025 eV), and *t* is foil thickness. The epithermal neutron self-shielding factor was calculated as follows [24]:

$$G_{\rm e} = 0.94/(1 + (z/2.7)^{0.82}) + 0.06, \tag{7}$$

where, $z = \sum_{\text{tot}} (E_{\text{res}})\Gamma \ 1.5t \ (\Gamma_{\gamma}/\Gamma)^{1/2}$ is a dimensionless variable, which converts the dependence of G_{e} on the dimension and physical and nuclear parameters into an unique curve and $\sum_{\text{tot}} (E_{\text{res}}) = (\rho N_{\text{a}} \sigma_{\text{res}}/M)$ is the macroscopic cross-section at resonance peak (E_{res}) (where ρ is the density, N_{a} is Avogadro' s number; M is the atomic weight; σ_{res} is the microscopic cross-section at E_{res}), t is the foil thickness, and Γ is the total resonance width $(\Gamma = \Gamma_{\gamma} + \Gamma_n$, where Γ_{γ} and Γ_n are resonance widths for (n,γ) and (n,n') reactions).

Sample irradiations and measurements

Natural W, Mo, and Zr metallic foils of the purity 99.99%; and 0.23 mm, 0.15 mm and 125 µm in thickness altogether with Au samples which are diluted in Al (0.1%) were wrapped with aluminum foils and irradiated without Cd shield in two different positions at the ETRR-2. The irradiation time varied from 2 to 3 h. After proper cooling times, the aluminum foils surrounding the activated samples were removed and the samples were transferred into clean polyethylene vials for gamma ray measurements. The gamma ray spectra were collected using a p-type coaxial EG&G Ortec HPGe detector, with 29.4% relative efficiency and 1.66 keV FWHM at 1332.5 keV of ⁶⁰Co. A Canberra 10 cm thickness ultra low background lead shield with low carbon steel casing is used in shielding the detector. A genie card of 16384 channels ADC is mounted on PC for data acquisition and analysis. The measurements were performed at a distance far from the detector head (10-15 cm) to minimize true coincidence effects. The neutron spectrum parameters characterizing the irradiation positions f and α were determined using the activated sets ¹⁹⁸Au, ⁹⁷Zr/⁹⁷Nb and ⁹⁵Zr/⁹⁵Nb.

Results and discussion

The neutron spectrum parameter f and α characterizing the two irradiation positions were determined using the activated isotopes of Zr and Au standards—details will be reported elsewhere. The results are shown in Table 1. The determined values of f and α for the two irradiation positions were used in Eq. 3 to determine the thermal neutron cross-sections and resonance integrals of the reactions ¹⁸⁶W (n,γ) ¹⁸⁷W and ⁹⁸Mo (n,γ) ⁹⁹Mo simultaneously relative to that ¹⁹⁷Au (n,γ) ¹⁹⁸Au.

The results are shown in Figs. 1 and 2. As one can see, the curves intercept in unique points giving simultaneously σ_0 = 38.43 b and $I_0(\alpha)$ = 425 b for ¹⁸⁶W (n,γ) ¹⁸⁷W; and σ_0 = 0.137 b and $I_0(\alpha)$ = 4.75 b for ⁹⁸Mo (n,γ) ⁹⁹Mo. The values of the resonance integrals determined from the curves were corrected for the value of the parameter α in the two irradiation positions with average values of 502 b and 6.47 b for the reactions ¹⁸⁶W (n,γ) ¹⁸⁷W and ⁹⁸Mo (n,γ) ⁹⁹Mo respectively—see Table 1. The thermal and epithermal neutron self shielding corrections were taken into



Fig. 1 Thermal cross-section and resonance integral of 186 W (n,γ)

Table 1 Determined neutron spectrum parameters, thermal neutron cross-sections and resonance integrals in the two irradiation positions

f	α	186 W (n,γ) 187 W			⁹⁸ Mo (<i>n</i> ,γ) ⁹⁹ Mo			
		σ_0 (b)	$I_0(\alpha)$ (b)	<i>I</i> ₀ (b)	σ_0 (b)	$I_0(\alpha)$ (b)	<i>I</i> ₀ (b)	
13.7	0.053	38.43	425	497	0.137	4.75	6.33	
17.3	0.06			508			6.603	
				Average $= 502$			Average $= 6.47$	



Fig. 2 Thermal cross-section and resonance integral of $^{98}\mathrm{Mo}~(n,\gamma)$ $^{99}\mathrm{Mo}$

account using Eqs. 6 and 7 respectively. These corrections and the nuclear data used in the calculation of the thermal neutron cross-sections and resonance integrals are shown in Table 2.

The results obtained were compared with previous measurements and the evaluated data in literature; and shown in Table 3. The determined thermal neutron cross-section for ¹⁸⁶W (n,γ) ¹⁸⁷W reaction, agrees with most of the reported results with deviations less than 9%, however the present result deviates by -10.2 and 16.45% from the

reported values in [12] and [33] respectively. The obtained resonance integral of ¹⁸⁶W (n,γ) ¹⁸⁷W reaction agrees with most of the literature values with relative deviation less than 9%. Too high deviations (up to 73%) are observed between the current result and the reported values in [16, 33, 37–39].

The determined thermal neutron cross-section for the 98 Mo (n,γ) 99 Mo reaction is in a very good agreement with the literature results with deviations less than 7%, however the higher deviations of 14.1 and -23.8% are observed between the present result and the results found in [14] and [15] respectively. Similarly, the obtained resonance integrals of the 98 Mo (n,γ) 99 Mo reaction agrees with most values, however with some deviations—see Table 3.

Total uncertainties of the obtained cross-sections and resonance integrals are determined. The uncertainty for thermal cross-sections and resonance integrals are found less than 13% of the determined values.

In this work, the two neutron irradiation positions used in determining cross-sections and resonance integrals are in the same reactor, however, these positions can belong to two different reactors. Moreover, three or more neutron irradiation positions can be used for the same purpose. In addition, the method of two neutron irradiation positions can be extended to perform neutron activation analysis using the k_0 method. Namely, it can be used to determine the neutron spectrum parameters as well as elemental concentrations for any number of samples of unknown compositions using only one comparator. The details of this extension and procedures will be reported in a forthcoming paper.

 Table 2
 Nuclear data used for the determination of thermal neutron cross-sections and resonance integrals and correction factors for neutron self-shielding^a

Nuclear reaction	Half-life	Gamma-rays		Isotopic abundance (%)	$\bar{E}_{ m r}({ m eV})$	$G_{ m th}$	G_{e}	
		Energy (KeV)	Intensity (%)					
186 W (<i>n</i> , γ) 187 W	23.72 h	134.25	9.4	28.6	20.5	0.983	0.3578	
		479.55	21.8					
		551.52	5.08					
		618.26	6.28					
		625.54	1.17					
		685.73	27.3					
		772.91	4.41					
⁹⁸ Mo (<i>n</i> ,γ) ⁹⁹ Mo	2.748 d	140.51	4.52	24.13	241	0.9815	0.999	
		181.06	5.99					
		366.44	1.21					
		739.50	12.13					
		777.92	4.26					
187 Au (<i>n</i> , γ) 198 Au	2.695 d	411.80	95.58	100	5.65	1	1	

^a Data were taken from reference [25]

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Table 3 Comparison of σ_0 and I_0 for ¹⁸⁶W (n,γ) ¹⁸⁷W and ⁹⁸Mo (n,γ) ⁹⁹Mo reactions with literature results

Ref.	186 W (n,γ) 187 W				Ref.	⁹⁸ Mo (<i>n</i> , γ) ⁹⁹ Mo			
_	σ_0 (b)	D (%)	<i>I</i> ₀ (b)	D (%)		σ_0 (b)	D (%)	<i>I</i> ₀ (b)	D (%)
This work	38.43 ± 4	-	502 ± 65	-	This work	0.137 ± 0.014	-	6.47 ± 0.8	_
[1]	37.2 ± 2.1	3.3	461 ± 39	8.9	[44]	0.136 ± 0.007	0.7	7.02 ± 0.62	-7.8
[10]	39.5 ± 2.3	-2.7	493 ± 40	1.8	[14]	0.12 ± 0.005	14.1	-	
[26]	41.8 ± 2.9	-8.06	-	-	[15]	0.18 ± 0.02	-23.8	-	
[27]	37 ± 2	3.86	510 ± 50	-1.6	[16]	-	-	4.72	37.07
[12]	42.8 ± 0.8	-10.2	486 ± 5	3.3	[17]	0.14	-2.14	8.2	-21.1
[28]	38.7 ± 1.9	-0.7	530 ± 28	-5.3	[29]	0.13 ± 0.006	5.4	7.3 ± 1.8	-11.4
[29]	37 ± 1.5	3.86	490 ± 15	-2.4	[31]	0.13 ± 0.006	5.4	6.9 ± 0.3	-6.23
[30]	35.4 ± 0.8	-8.55	534 ± 50	-6	[34]	0.1307	4.8	_	-
[31]	37.9 ± 0.6	1.4	485 ± 15	3.5	[35]	0.13	5.4	6.553	-1.3
[32]	37 ± 1.8	3.86	507 ± 27	-0.1	[36]	0.129	6.2	6.954	-7
[33]	33	16.45	318	57.8	[40]	0.144 ± 0.0053	-4.9	7.42 ± 0.3	-12.8
[34]	37.5	2.5	522	-3.8	[41]	0.145 ± 0.015	-5.51	5.2 ± 0.2	24.4
[35]	39.45	-2.6	529	-5.1	[42]	0.131 ± 0.002	4.6	_	_
[36]	38.27	0.4	528.6	-4.9	[43]	0.14 ± 0.01	-2.14	_	_
[37]	36.5 ± 4.2	5.28	290.3	73					
[16]	_	-	290	73					
[38]	_	-	345	45.5					
[39]	-	-	320	56.8					

Conclusions

The thermal neutron cross-sections and resonance integrals of the ¹⁸⁶W (n,γ) ¹⁸⁷W and ⁹⁸Mo (n,γ) ⁹⁹Mo reactions were determined via neutron irradiation without Cd shield in two different neutron irradiation positions instead of the traditional method of irradiation with and without Cd shield in one neutron irradiation position. Neutron self shielding and the deviation from the 1/*E* law corrections were taken into account. The determined cross-sections were compared with the previous measurements in literature. Good agreements are obtained for all values determined, however with some deviations for the resonance integrals of the ⁹⁸Mo (n,γ) ⁹⁹Mo and ¹⁸⁶W (n,γ) ¹⁸⁷W reactions.

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