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Measurements of the Cross-sections of Produced Short-Lived Nuclei Induced by Neutrons around 14 MeV on Isotopes of Tungsten *

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New experimental cross-section data for the $^{180}W(n,2n)^{179m}W$, $^{186}W(n,2n)^{185m}W$ and $^{186}W(n,p)^{186}Ta$ reactions at the neutron energies of 13.5 and 14.4 MeV are obtained by the activation technique. The neutron beams are produced by means of the $^{3}H(d,n)^{4}He$ reaction. The gamma activities of the product nuclei are measured by a high-resolution gamma-ray spectrometer with a coaxial high-purity germanium detector. The neutron fluence is determined using the monitor reaction $^{93}Nb(n,2n)^{92m}Nb$. The results in the current work are discussed and compared with the measurement results found in the literature. It is shown that these higher accuracy experimental cross-section data around the neutron energy of 14 MeV agree with some previous experimental values from the literature within experimental uncertainties.

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Tungsten was regarded as one of the most promising candidates for plasma facing materials in future fusion reactors because it has excellent mechanical performance, good thermal conductivity and outstanding irradiation resistance.[1−3] Therefore, a lot of experimental data on its cross-sections for producing relatively long-lived nuclei have been reported around the neutron energy of 14 MeV, which can be found from the International Atomic Energy Agency (IAEA) database.[4] However only a few laboratories[5−16] obtained the experimental data on its isotope cross-sections to produce short-lived nuclei induced by neutrons around 14 MeV. There are discrepancies in those data due to differences in the experimental methods, equipment, data processing methods, samples, nuclear parameters, and so on, which make it necessary to take further measurements.

In the current work, the cross-sections of produced short-lived nuclei at the neutron energies of 13.5 and 14.4 MeV for the ^{180}W(n,2n)^{179m}W, ^{186}W(n,2n)^{185m}W and ^{186}W(n,p)^{186}Ta reactions are studied and a gamma-ray counting technique is applied via using high-resolution gamma-ray spectrometer and data acquisition system. The results obtained are discussed and compared with the previous works.

Natural tungsten foils, 99.99% in purity and 2 mm in thickness, were made into circular samples with diameter of 20 mm. Each was sandwiched between two 99.99% pure circular niobium foils with a diameter of 20 mm and 1 mm thickness, and then wrapped in 99.95% pure cadmium foil with 1 mm thickness to avoid the effect of ^{184}W(n,γ)^{185m}W reaction induced by thermal neutron to the ^{186}W(n,2n)^{185m}W reaction.

The irradiations of the samples used in this work were carried out at the K-400 Neutron Generator at Institute of Nuclear Physics and Chemistry, China Academy of Engineering Physics, and lasted from 2 to 3 hours. The neutrons around 14 MeV were produced by means of the ^{3}H(d,n)^{4}He reaction with a deuteron beam energy of 255 keV and a beam current of 320 μA. Neutrons in the 14 MeV region with a yield of about 5×10^{10} n/s. The tritium-titanium (T-Ti) target used in the neutron generator was about 2.19 mg/cm^2 thick. The neutron flux was monitored by the accompanied α-particles so that the corrections could be made for small variations in the yield during irradiations. Groups of samples were placed at 45° and 135° with respect to the deuteron beam direction. The distances of samples from the T-Ti target were about 3–5 cm. The neutron energies in the measurements were determined beforehand by the cross-section ratios method for the ^{90}Zr(n,2n)$^{89m+g}$Zr and ^{93}Nb(n,2n)^{92m}Nb reactions.[17]

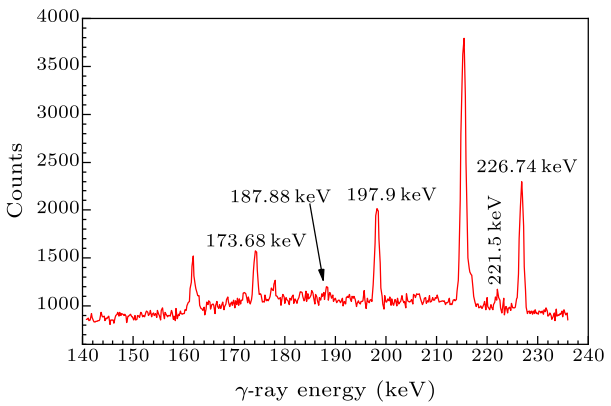

Fig. 1. Part of γ-ray spectrum of tungsten about 5 min after the end of irradiation.

By making use of a well-calibrated GEM-60P coaxial high-purity germanium (HPGe) detector (crystal diameter 70.1 mm, crystal length 72.3 mm) with a relative efficiency of ∼68% and an energy resolution of

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1.69 keV at 1332 keV for ^{60}Co, the activities of these samples were determined from their number of detected γ-ray counts. The cooling time of the sample after the end of irradiation is about 5 min, it takes about 1 hour to measure the sample. Figure 1 shows the typical γ-ray spectrum acquired from the tungsten sample about 5 min after the end of irradiation. The efficiency of the detector was pre-calibrated by using various standard γ sources.

The decay characteristics of the product radionuclides and the natural abundance of the target isotopes under investigation are summarized in Table 1.[18] The abundance of ^{93}Nb is taken from Ref. [19].

Table 1. Reactions and associated decay data of activation products.

Reaction	Abundance of target isotope (%)	Product	$T_{1/2}$	E_γ (keV)	I_γ (%)
^{180}W(n,2n)	0.12	^{179m}W	6.40 min	221.5	8.8
^{186}W(n,2n)	28.43	^{185m}W	1.67 min	187.88	0.81
^{186}W(n,p)	28.43	^{186}Ta	10.5 min	197.9	50
^{93}Nb(n,2n)	100	^{92m}Nb	10.15 d	934.44	99.15

Table 2. Summary of cross-section measurements.

Reaction	This work		Data in literature		
	E_n (MeV)	σ (mb)	E_n (MeV)	σ (mb)	References
^{180}W(n,2n)^{179m}W	13.5±0.2	680±87	13.48	540±86	[5]
	14.4±0.2	518±66	13.64	573±80	[5]
			13.88	490±71	[5]
			14.28	498±95	[5]
			14.47	501±67	[5]
			14.68	459±70	[5]
			14.82	476±65	[5]
			13.5	496±63	[6]
			14.1	567±67	[6]
			14.5	484±52	[7]
			14.7	490±45	[8]
^{186}W(n,2n)^{185m}W	13.5±0.2	575±52	13.47	699±60	[5]
	14.4±0.2	611±50	13.66	676±60	[5]
			13.88	686±60	[5]
			14.04	680±60	[5]
			14.26	683±59	[5]
			14.44	712±66	[5]
			14.63	691±60	[5]
			14.81	707±55	[5]
			14.1	692±68	[6]
			14.7	642±60	[8]
			14.1	689±49	[9]
			14.7	602±74	[10]
			14.8	1152±110	[11]
			14.8	540±80	[12]
			14.8	470	[13]
^{186}W(n,p)^{186}Ta	13.5±0.2	0.82±0.04	13.48	0.80±0.10	[5]
	14.4±0.2	1.40±0.07	13.65	0.93±0.11	[5]
			13.88	1.02±0.12	[5]
			14.04	1.25±0.15	[5]
			14.27	1.55±0.19	[5]
			14.46	1.59±0.18	[5]
			14.67	1.83±0.21	[5]
			14.82	1.97±0.22	[5]
			14.5	1.64±0.12	[7]
			14.7	1.4±0.2	[8]
			14.1	1.33±0.10	[9]
			14.8	11±4	[13]
			13.4	0.59±0.14	[14]
			13.65	1.2±0.35	[14]
			13.88	1.31±0.31	[14]
			14.28	1.78±0.54	[14]
			14.58	1.84±0.41	[14]
			14.87	2.31±0.48	[14]
			14.5	2.6	[15]
			14.5	2.9±0.58	[16]
^{93}Nb(n,2n)^{92m}Nb			13.5±0.3	457.9±6.8	[21]
			14.4±0.3	459.8±6.8	[21]

The measured cross-section values σ_x were calculated by the following formula:[20]

$$\sigma_x = \frac{[S\varepsilon I_\gamma \eta K M D]_0}{[S\varepsilon I_\gamma \eta K M D]_x} \frac{[\lambda A F C]_x}{[\lambda A F C]_0} \sigma_0, \quad (1)$$

where the subscript 0 represents the term corresponding to the monitor reaction and the subscript x corresponds to the measured reaction, $D = e^{-\lambda t_1} - e^{-\lambda t_2}$, $F = f_s \times f_c \times f_g$, $K = [\sum_i^L \Phi_i (1 - e^{-\lambda \Delta t_i}) e^{-\lambda T_i}] / \Phi S$, $S = 1 - e^{-\lambda T}$. The meanings of the letters such as D, F, K, S, ε, I_γ, η, M, t_1, t_2, A, C, λ, f_s, f_c, f_g, L, Δt_i, T_i, Φ_i, Φ and T can be found in Ref. [20].

The cross-sections of the ^{180}W(n,2n)^{179m}W, ^{186}W(n,2n)^{185m}W and ^{186}W(n,p)^{186}Ta reactions were obtained relative to those of the ^{93}Nb(n,2n)^{92m}Nb reaction. In Table 2 and in Figs. 2–4, the cross-sections measured in this work are shown and plotted together with the values given in the literature for comparison. Here in Fig. 4 in the current work, the result from Ref. [13] 11±4 mb at 14.8 MeV is not adopted because the value is too large to show clearly the relations of the other data near 14 MeV. The cross-sections of the monitor reaction ^{93}Nb(n,2n)^{92m}Nb are obtained from Ref. [21] and also listed in Table 2.

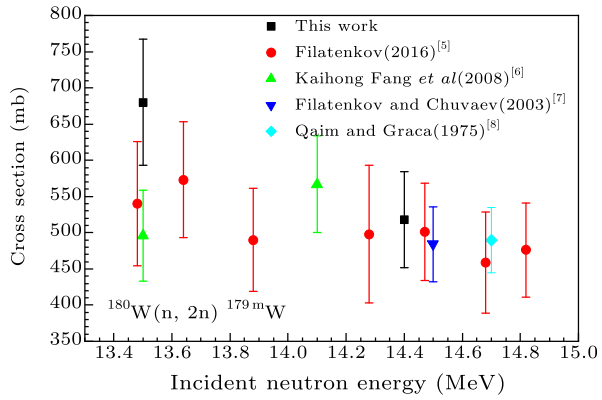

Fig. 2. Cross section of the ^{180}W(n,2n)^{179m}W reaction.

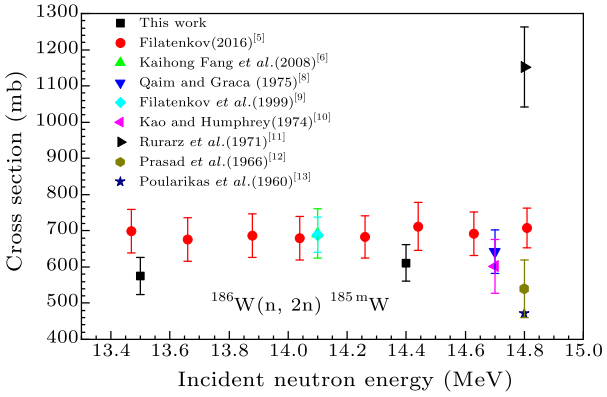

Fig. 3. Cross section of the ^{186}W(n,2n)^{185m}W reaction.

Through the analysis of the possible reactions of tungsten and radioactive gamma-rays produced by their products, it can be seen that the 198.35187 keV gamma-ray (intensity 1.465%) of ^{182}Ta (the half life is 114.74 d) produced by the ^{182}W(n,p)^{182}Ta+^{183}W(n,d)^{182}Ta+^{184}W(n,t)^{182}Ta reaction is close to the 197.9 keV gamma-ray of ^{186}Ta (the half life is 10.5 min) produced by the measured reaction ^{186}W(n,p)^{186}Ta. In the natural sample, the cross-section measurements of the ^{186}W(n,p)^{186}Ta reaction will encounter gamma-ray interference with close energies because of the limit of the energy resolution of the detector, using a cool sample after irradiation was a useless method. The cross-sections of the ^{186}W(n,p)^{186}Ta reaction were calculated using Eq. (1) and the 197.9 keV characteristic gamma ray of ^{186}Ta. In the calculation process, the effect of the 198.35187 keV gamma ray of ^{182}Ta produced by the ^{182}W(n,p)^{182}Ta+^{183}W(n,d)^{182}Ta+^{184}W(n,t)^{182}Ta reaction has been deducted using the formula to subtract the effects of other γ rays with close energies on the measured reaction data.[22]

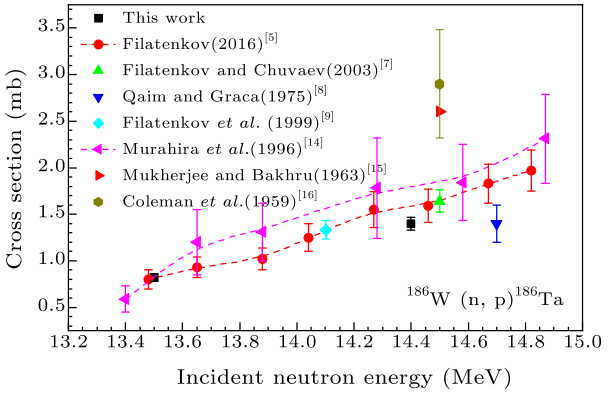

Fig. 4. Cross-section of the ^{186}W(n,p)^{186}Ta reaction.

In the present work, some corrections were made for the fluctuation of the neutron flux during the irradiation, γ-ray self-absorption in the sample, and the sample geometry. The main uncertainties in our work stem from the counting statistics (1–5.9%), the standard cross-section uncertainties (1.1–1.5%), detector efficiency (2%), the weight of samples (0.1%), the sample geometry (1%), γ-ray self-absorption in the sample (1.5%), and the fluctuation of the neutron flux (1%), etc. The total uncertainties are about 4.9–12.8%.

In this work, low energy neutrons may originate the d-d reaction by accumulation of deuterium in the tritium-target with time and from background neutrons, which may originate from the scattering of thermal and epithermal neutrons from room walls etc. We did not measure these low energy neutrons, but the new T-Ti target and natural high-purity tungsten foils were used, the samples were wrapped in thin cadmium foil and placed at the appropriate positions, which were about 3–5 cm away from the center of the T-Ti target and were far away from the walls and the upper or lower floors of the experimental hall during the irradiation. The influence of lower energy neutrons was reduced to a very low level. In addition, the effect of other γ rays with close energies was subtracted in the calculation process of the cross-section values of the

^{186}W(n,p)^{186}Ta reaction, which can make our results more accurate.

For the ^{180}W(n,2n)^{179m}W reaction, it can be seen from Table 2 and Fig. 2 that the data obtained in our work at the neutron energies 13.5 and 14.4 MeV are in excellent agreement with the values of the excitation curve in Ref. [5] at corresponding energies within experimental uncertainties. At the neutron energy of 13.5 MeV, the datum obtained in our work is higher than the result in Ref. [6].

The cross-section values of the ^{186}W(n,2n)^{185m}W reaction are presented in Table 2 and Fig. 3. This shows that the datum obtained in our work is in agreement with the corresponding value of the excitation curve in Ref. [5] within experimental uncertainty at the neutron energy of 14.4 MeV, but the datum obtained in our work is lower than the corresponding value of the excitation curve in Ref. [5] at neutron energy of 13.5 MeV. The datum in Ref. [11] is much larger than the others at the neutron energy of 14.8 MeV.

In the case of the ^{186}W(n,p)^{186}Ta reaction, it can be seen from Table 2 and Fig. 4 that the data obtained in our work are in agreement with the corresponding values of the excitation curve in Refs. [5,14] within experimental uncertainties at the neutron energies of 13.5 and 14.4 MeV. The result in Ref. [13] is much larger than the others at the neutron energy of 14.8 MeV. The value from Refs. [15,16] are larger than the others at the neutron energy of 14.5 MeV.

In the present study, the new experimental cross-section data for the ^{180}W(n,2n)^{179m}W, ^{186}W(n,2n)^{185m}W and ^{186}W(n,p)^{186}Ta reactions producing short-lived nuclei at the neutron energies of 13.5 and 14.4 MeV are obtained. In general, our measured results around the neutron energy of 14 MeV agree with some of the previous experimental values from the literature within experimental uncertainty. These new experimental cross-section data would improve the quality of the neutron cross-section database and are expected to help in new evaluations around the neutron energy of 14 MeV for the cross sections mentioned in this paper.

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