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Measurement of the flux-weighted average cross section for the $^{186}\mathrm{W}(\gamma,p)^{185}\mathrm{Ta}$ reaction at the bremsstrahlung end-point energy of 70 MeV

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Abstract. The flux-weighted average cross section for the ${}^{186}W(\gamma, p){}^{185}Ta$ reaction induced with 70 MeV bremsstrahlung end-point energy was measured using the activation method in combination with off-line gamma activity measurement. The ${}^{27}Al(\gamma, 2pn){}^{24}Na$ monitor reaction was used for the determination of the bremsstrahlung flux. The bremsstrahlung photons used to produce the photonuclear reactions were generated from a thin tungsten (W) converter by bombardment with a 70 MeV electron beam. The experimental flux-weighted average cross section is compared with the theoretical prediction, for which the absolute cross section was calculated using the TALYS 1.95 code and the bremsstrahlung spectrum was simulated using the computer program MCNPX. The present experiment was carried out at the 100 MeV electron linac of the Pohang Accelerator Laboratory, POSTECH, Pohang, Korea.

Keywords: ¹⁸⁶W(γ , p)¹⁸⁵Ta reaction; averaged cross-section; bremsstrahlung end-point energy of 70 MeV; activation and off-line γ -ray spectrometry; TALYS1.95; MCNPX. Classification numbers: 25.20.-x; 29.30.Kv.

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1. Introduction

Photonuclear reactions play an important role in various aspects of basic nuclear physics research and in different fields of application [1, 2]. In recent decades, both the well-developed electron linacs capable of generating bremsstrahlung photons with energies of tens or more than tens of MeV and the high-quality γ -ray measuring instruments have offered advantages for research on photonuclear reactions. The high-energy photons are able to produce nuclear reactions with a variety of complexity by emitting not only a single nucleon such as (γ, n) [3–6] or (γ, p) [6–8], but also multi-particle emission reactions such as (γ, xn) [9, 10] or $(\gamma, xnyp)$ with $x, y \ge 1$ [11].

The complexity of the nuclear reactions increases as the energy of incident bremsstrahlung photons increases. At relatively low energies of incident photons, in the range ≤ 30 MeV, the so-called giant dipole resonance (GDR) energy region, photonuclear reactions occur mainly due to a compound mechanism. However, at higher incident energies, nuclear reactions can also be processed via additional mechanisms such as pre-equilibrium and direct. This is also another important reason for studying nuclear reactions with increasing incident energies.

In this work, we chose the ${}^{186}W(\gamma, p){}^{185}Ta$ reaction for study with an incident bremsstrahlung end-point energy of 70 MeV, beyond the GDR energy range. As it is known, W is a potential structural material of a fusion reactor, so the knowledge of the cross section of nuclear reactions on W is necessary. The ${}^{186}W(\gamma, p){}^{185}Ta$ reaction produces a neutron-rich ${}^{185}Ta$ isotope, which was first observed by Butement in 1950 through the ${}^{186}W(\gamma, p)$ photonuclear reaction by 23 MeV X-rays from the synchrotron [12]. However, no literature data for the ${}^{186}W(\gamma, p){}^{185}Ta$ reaction were found. Although we found some reports in the literature about the production of ${}^{185}Ta$ from ${}^{186}W$, but those were not by the (γ, p) reaction. Shizuma et al reported the studies on the production of ${}^{185}Ta$ through the one-proton acceptance reaction ${}^{186}W({}^{18}O, {}^{19}F){}^{185}Ta$ [13], Li et al measured cross section of the ${}^{186}W(n,d){}^{185}Ta$ reaction around 14 MeV neutrons [14], Lovhoiden et al studied the ${}^{186}W(n,x){}^{185}Ta$ reaction with energies between 13.5 and 20.6 MeV [16], and Sakane et al reported the studies on the cross section of the ${}^{186}W(n,n){}^{185}Ta$ reaction by 14 MeV neutron [17].

The present study focuses on measurements of the flux-weighted average cross section of the ¹⁸⁶W(γ , p)¹⁸⁵Ta reaction induced with a bremsstrahlung end-point energy of 70 MeV. Since the ¹⁸⁵Ta radionuclide has gamma rays and a half-life suitable for the activity measurement, the experiment was performed using the well-known activation method in combination with off-line gamma spectrometry [18–20]. Due to the lack of experimental data, the present result is compared to the theoretical prediction, using the TALYS 1.95 code [21]. For the calculations, the effect of the nuclear level density was taken into account by considering six level density models.

2. Experiment

This experimental procedure contains three main steps, (1) irradiation of the sample to convert the stable target nuclides into radioactive isotopes, (2) measurement of the activity of the radionuclides produced, and (3) data analysis to derive the flux-weighted average cross section of the ¹⁸⁶W(γ , p)¹⁸⁵Ta reaction.

Nguyen Van Do et al.

2.1. Irradiation of the sample

Natural tungsten foil (W) with a purity of 99.95%, a size of 12 mm ×12 mm and a thickness of 0.1 mm was prepared for the irradiation at the electron accelerator of the Pohang Accelerator Laboratory, POSTECH, Pohang, Korea. In the present experiment, the bremsstrahlung flux was monitored by the 27 Al(γ ,2np) 24 Na reaction. For this purpose, the aluminum foil of 99% purity with the same dimension as W sample was also prepared and the stack of Al-W foils was irradiated simultaneously. The bremsstrahlung photon beam with the maximum energy of 70 MeV was generated by bombarding a 70 MeV electron beam into the thin W converter with a size of 100 x 100 mm and a thickness of 0.1 mm. The geometric arrangement for the irradiation was the same as in our previous experiment [11], namely the tungsten converter was placed 15 cm from the electron exit window and the stack of Al-W foils was placed 12 cm from the tungsten converter. During irradiation, the intensity of the electron beam was fixed at 50 mA, the pulse width of the electron beam was 1.2 μ s, the repetition frequency was 15 Hz, and the irradiation time was 60 min. The nuclear reactions of interest in this study are ${}^{186}W(\gamma, p){}^{185}$ Ta and ${}^{27}Al(\gamma,2pn){}^{24}$ Na, and the main decay data of the residual nuclei are listed in Table 1 [22].

Table 1. Investigated nuclear reactions and main decay data of the reaction products 185 Ta and 24 Na 22.

Nuclearreaction	E_{thr} (MeV)	$T_{1/2}$	Decay mode (%)	Spin	E_{γ} (keV)	$I_{\gamma}\left(\% ight)$
186 W $(\gamma, p)^{185}$ Ta	8.40	49.4 m	β^{-} : 100	7/2+	107.80 173.91* 177.59* 243.70	2.75 22.6 25.7 3.8
27 Al(γ ,2pn) 24 Na	31.45	14.997 h	β^- : 100	4+	1368.62* 2754.0	99.99 99.85

* γ -ray used for activity measurement.

2.2. Activity measurement

The γ -spectra of both the irradiated W sample and the Al monitor were measured off-line by the HPGe γ -ray detector (ORTEC, GEM-2018-p) connected to a PC-based 4K channel analyzer. The energy resolution of the HPGe detector is 1.8 keV at the γ -peak of 1332.5 keV from ⁶⁰Co. The detection efficiency of the detector at 1332.5 keV is 20% relative to that of the 76 mm \times 76 mm NaI(Tl) detector. The efficiency of the detector was calibrated at different distances from the detector surface using standard γ -sources as described elsewhere [23].

Considering the advantage of high activity of irradiated samples, the measurement was performed by placing the sample at 10 cm from the surface of the HPGe detector to reduce counting losses due to detector dead time and coincidence summing effect. Typical γ -ray spectra of the irradiated W foil sample and Aluminum monitor are shown in Fig. 1 and Fig. 2, respectively.

As can be seen in Fig. 1, the γ -ray spectrum of the W foil sample shows a number of photo peaks. However, the two γ -rays of 173.91 keV and 177.59 keV are suitable for the activity measurement of the ¹⁸⁵Ta radionuclide. The main reason for this choice was that none of the gamma

rays were interfered or overlapped with the γ -rays of 173.91 keV or 177.59 keV. In addition, both the 173.91 keV and 177.59 keV photopeaks have relatively good statistics, with a statistical uncertainty of about 1.5-2.0%. This makes the analysis of the γ -ray spectra easier and more accurate.



Fig. 1. Typical γ -ray spectrum of W foil sample irradiated with a bremsstrahlung endpoint energy of 70 MeV. The irradiation, waiting, and measurement times were 30, 50, and 60 min, respectively. A part of the γ -ray spectrum in the upper right corner of this figure shows the main γ -peaks at 173.91 keV (22.6%) and 177.59 kev (25.7%) of the ¹⁸⁵Ta radionuclide, which are used for measuring activity.

The ²⁴Na radionuclide emits two gamma rays at 2754.0 keV (99.85%) and 1368.62 keV (99.99%). In principle, both γ -rays are of high intensity, which can be used for the measurement of ²⁴Na activity. In the present experiment, the 1368.62 keV (99.99%) γ -ray was chosen for the measurement, since its detection efficiency is higher than that of 2754.0 keV. Additionally, as shown in Fig. 2, the 1368.62 keV γ -ray is interference-free and appears in the relatively low background gamma spectrum.

The measured γ -ray spectra were processed using the computer program GammaVision, version 5.10 (EG&G, ORTEC), which was able to accurately determine the energy of the appearing γ -peaks and the number of counts under each photo peak considered in the γ -ray spectra.

2.3. Data analysis

The flux-weighted average cross section for a given photonuclear reaction, over an energy range from the reaction threshold to the bremsstrahlung end-point energy can be expressed as follows [19]:

$$\langle \boldsymbol{\sigma}(E_{\gamma \max}) \rangle = \frac{\int_{E_{thr}}^{E_{\gamma \max}} \boldsymbol{\sigma}(E) \boldsymbol{\phi}(E) dE}{\int_{E_{thr}}^{E_{\gamma \max}} \boldsymbol{\phi}(E) dE},\tag{1}$$



Fig. 2. Typical γ -ray spectrum of Aluminum foil sample irradiated with bremsstrahlung end-point energy of 70 MeV. The irradiation, waiting and measurement times were 30, 180, and 10 min, respectively.

where $\sigma(E)$ is the energy dependent cross section, $\Phi(E)$ is the bremsstrahlung flux at the given energy E, E_{thr} and $E_{\gamma max}$ are the reaction threshold and bremsstrahlung end-point energies, respectively. The integral $\int_{E_{thr}}^{E_{\gamma max}} \phi(E) dE$ gives the integrated bremsstrahlung flux in the energy range from the reaction threshold, E_{thr} to the bremsstrahlung end-point energy, $E_{\gamma max}$, which is incident on W foil sample, resulting in the ¹⁸⁶W(γ , p)¹⁸⁵Ta reaction. The integrated bremsstrahlung flux for the studied reaction (x) is now denoted by $\phi_{int,x}$. So, the flux-weighted average cross section of the nuclear reaction expressed in Eq. (1) can be briefly rewritten as:

$$\langle \boldsymbol{\sigma}(E_{\gamma \max}) \rangle = \frac{Y_{Exp}(E_{\gamma \max})}{\phi_{\inf x}}.$$
 (2)

In order to obtain the flux-weighted average cross section, it is necessary to measure the reaction yield, $Y_{Exp}(E_{\gamma max})$ and the integrated bremsstrahlung flux, $\phi_{int,x}$ incident on the target nuclide to form the nuclear reaction under study. By using the activation method in combination with off-line γ -ray spectrometry, the reaction yield can be obtained as follows:

$$Y_{Exp}(E_{\gamma\max}) = \frac{S_{\gamma}f\lambda(1 - e^{-\lambda T})}{N_0 I_{\gamma} \varepsilon_{\gamma}(1 - e^{-\lambda \tau}) e^{-\lambda (T - \tau)} (1 - e^{-\lambda t_i}) e^{-\lambda t_w} (1 - e^{-\lambda t_m})},$$
(3)

where S_{γ} is the photo-peak area (or total number of counts) of the selected gamma ray of the residual nuclide, λ is the decay constant, f is the correction factor for the counting losses due to the self-absorption, which is calculated by the simple expression: $f = (1 - e^{-\mu d})/\mu d$, where μ is the linear absorption coefficient and d is the sample thickness, I_{γ} is the intensity of γ -ray emitted

from the residual nuclide, ε_{γ} is the detector efficiency, which is determined experimentally as mentioned in subsection 2.2, τ is the pulse width of the electron beam and *T* is the cycle period, t_i , t_w , and t_m are the irradiation, waiting and measurement times.

The integrated bremsstrahlung flux incident on the aluminum target, resulting ${}^{27}\text{Al}(\gamma, 2pn)^{24}\text{Na}$ monitor reaction, denoted by $\phi_{\text{int,mon}}^{Exp}$, which represents the integrated bremsstrahlung flux over the threshold energy of 31.45 MeV to the end-point energy of 70 MeV can be obtained from the following expression:

$$\phi_{\text{int,mon}}^{Exp} = \frac{S_{\gamma,mon}f\lambda(1-e^{-\lambda T})}{\langle \sigma_{mon}^{Exp}\rangle N_0 I_{\gamma} \varepsilon_{\gamma}(1-e^{-\lambda \tau})e^{-\lambda (T-\tau)}(1-e^{-\lambda t_i})e^{-\lambda t_w}(1-e^{-\lambda t_m})},$$
(4)

where $S_{\gamma,mon}$ is the number of counts under the 1368.62 keV gamma-peak of ²⁴Na, $\langle \sigma_{mon}^{Exp} \rangle$ is the mean cross section of the monitor reaction ²⁷Al($\gamma, 2pn$)²⁴Na over the energy range from 31.45 MeV to the bremsstrahlung end-point energy of 70 MeV, and its value is 0.158 mb [19, 24], and all other terms have the same meaning as in Eq. (3).

As seen in Table 1, the threshold energy for the monitor reaction ${}^{27}\text{Al}(\gamma,2\text{pn}){}^{24}\text{Na}$ is 31.45 MeV and that for the studied reaction ${}^{186}\text{W}(\gamma,p){}^{185}\text{Ta}$ is 8.40 MeV. Thus, the integrated bremsstrahlung flux for the studied reaction

$$\int_{E_{thr,x}}^{E_{\gamma\max}} \phi(E) dE = \phi_{\text{int,x}}$$

can be derived from that of the monitor reaction as follows:

$$\phi_{\text{int},x} = C_x \times \phi_{\text{int},mon},\tag{5}$$

where $\phi_{\text{int},mon}$ is the integrated bremsstrahlung flux for the monitor reaction ${}^{27}\text{Al}\gamma,2pn$)²⁴Na over the energy range from the threshold energy of 31.45 MeV to the end-point energy of 70 MeV, C_x is the conversion factor. As usual, the conversion factor can be obtained by calculation, taking into account the threshold energies of the two nuclear reactions, ${}^{186}\text{W}(\gamma,p)$ ${}^{185}\text{Ta}$ and ${}^{27}\text{Al}(\gamma,2pn)^{24}\text{Na}$ as follows [19]:

$$C_x = \int_{E_{thr,x}}^{E_{\gamma\max}} \phi(E) dE \bigg/ \int_{E_{thr,mon}}^{E_{\gamma\max}} \phi(E) dE,$$
(6)

where $E_{th,x}$ and $E_{th,mon}$ are the threshold energy of the ¹⁸⁶W(γ, p)¹⁸⁵Ta and ²⁷Al($\gamma, 2pn$)²⁴Na reactions, respectively, $E_{\gamma max}$ is the end-point energy of the bremsstrahlung spectrum and equals 70 MeV. The bremsstrahlung spectrum as a function of photon energy, $\phi(E)$ was obtained by simulation using MCNPX [25] computer program. The simulation took into account the actual arrangement of the experimental geometry, the dimension of the W target and the characteristics of the electron beam as described in subsection 2.1. The simulated bremsstrahlung spectrum is shown in Fig. 3. The conversion factor for the ¹⁸⁶W(γ, p)¹⁸⁵Ta reaction was calculated as $C_x = 3.19$. Finally, it was possible to determine the integrated bremsstrahlung flux for the ¹⁸⁶W(γ, p)¹⁸⁵Ta reaction according to the Eq. (5).



Fig. 3. The bremsstrahlung spectrum generated by impinging a 70 MeV electron beam on the W converter with a size of 100 mm \times 100 mm and a thickness of 0.1 mm. The spectrum was simulated using the computer program MCNPX.

3. Results and discussion

3.1. Experimental result

The flux-weighted average cross section of the ¹⁸⁶W(γ, p)¹⁸⁵Ta reaction was determined from Eq. (2) based on the measured reaction yield, $Y_{Exp}(E_{\gamma max})$ and the integrated bremsstrahlung flux, $\phi_{int,x}$. It should be noted that when determining the reaction yield, the contribution of the competing reaction ¹⁸⁶W(n,x)¹⁸⁵Ta was removed by slowing down the fast neutrons before entering the studied sample [26]. The flux-weighted average cross section of the ¹⁸⁶W(γ, p)¹⁸⁵Ta reaction induced with a bremsstrahlung end-point energy of 70 MeV obtained in this study is 0.105 ± 0.011 mb. The total uncertainty of the experimental result was determined according to the square root rule and gives values between 9 and 12%. The main sources of uncertainty included the statistical uncertainty (2 – 3%), the nuclear data used such as gamma intensity (4 – 5%) and half-life of the residual nuclide (3.1%), the detector efficiency (3 – 4%), the variation of the electron beam (5 – 6%) and the uncertainty expected from other sources (4 – 7%). In this experiment, by correcting for the gamma-ray self-absorption effect and avoiding the competing reaction ¹⁸⁶W(n,d)¹⁸⁵Ta, the uncertainties caused by both become small and are included among the other sources of uncertainty.

Currently, no experimental data can be found in the literature regarding the flux-weighted average cross section of the ${}^{186}W(\gamma, p){}^{185}Ta$ reaction. We therefore cannot compare the current results with the reference data. Instead, we compare the current result with theoretical predictions.

3.2. Model calculation results

We are unable to compare the current experimental result with others due to the lack of measurement data for the ${}^{186}W(\gamma, p){}^{185}Ta$ nuclear reaction. In order to verify the present result, we calculate the flux-weighted average cross section of the ${}^{186}W(\gamma, p){}^{185}Ta$ reaction for comparison. The flux-weighted average cross section was calculated according to Eq. (1). Accordingly, we need to calculate the reaction yield and the integral bremsstrahlung flux. The energy dependent cross section $\sigma(E)$ for the ¹⁸⁶W(γ, p)¹⁸⁵Ta reaction was calculated using the nuclear model code TALYS-1.95 [21]. The code TALYS was chosen for the calculation because it is user friendly, well documented and widely applied today. In addition, numerous nuclear models are available in the TALYS code that help simulate all possible nuclear reactions that occur with different projectiles such as γ -rays, protons, neutrons, deuterons, tritons, ³He and ⁴He with an incident energy between 1 keV and 200 MeV on nuclides with mass number between 12 and 339. The TALYS code also takes into account different reaction mechanisms, such as compound, pre-equilibrium and direct reactions, depending on the energy of the incident particles. The TALYS input is based on the RIPL-3 database (IAEA Reference Input Parameter Library) [27]. The total bremsstrahlung flux incident on the W foil sample, resulting in the ${}^{186}W(\gamma, p){}^{185}Ta$ reaction was obtained by integrating $\int_{E_{thr}}^{E_{\gamma \max}} \phi(E) dE$ over the energy range from the reaction threshold of $E_{thr} = 8.40$ MeV to the bremsstrahlung end-point energy of $E_{\gamma \max} = 70$ MeV.

It is known that the nuclear level densities and gamma strength functions are the important ingredients to calculate the reaction cross section. In order to verify the level density effect, the cross sections of the ¹⁸⁶W(γ , *n*)¹⁸⁵Ta reaction were calculated taking into account the six level density models, including three phenomenological models and three microscopic models [28–30], namely:

Constant temperature + Fermi gas (Ldmode 1),

Back-shifted Fermi gas (Ldmode 2),

Generalized superfluid (Ldmode 3),

Microscopic level densities (Skyrme force) from Goriely's tables (Ldmode 4),

Microscopic level densities (Skyrme force) from Hilaire's combinatorial tables (SFH) (Ldmode 5), and

Microscopic level densities (temperature dependent HFB, Gogny force) from Hilaire's combinatorial tables (Ldmode 6).

The gamma strength function used is the Brink-Axel Lorentzian model, which is often called the default model. The cross sections for the ¹⁸⁶W(γ , p) ¹⁸⁵Ta reaction, calculated using the TALYS-1.95 code [21], taking into account the six level density models (Ldmode1-Ldmode6) are shown in Fig. 4. In addition, in Fig. 4, we also plot the values of TENDL-2021 library [31] for comparison.

As can be seen in Fig. 4, the reaction cross sections calculated using the TALYS-1.95 code [21] considering six different level density models exhibit considerable dispersion. The cross section values peak at a photon energy of around 25 MeV, and the difference between the values calculated using different level density models can reach a factor of ~ 1.4 . However, the difference between the cross section values at photon energies above 50 MeV is not significant, except for the cross section curve calculated using Ldmode 2. The comparison also indicates that the cross section curve from the TENDL-2021 library [31] overestimates all cross section curves

350



Fig. 4. (Color online) Energy dependent cross section of the ${}^{186}W(\gamma, p){}^{185}$ Ta reaction, calculated using the TALYS-1.95 code, considering six level density models (Ldmode1-Ldmode6) and the theoretical data from the TENDL-2021 library [31].

calculated using the TALYS-1.95 code [21] with six level density models. Meanwhile, the cross section values of TENDL-2021 library [31] is lower than the evaluated values of ENDF/B-VII library [32]. The reason for this discrepancy may be due to the inconsistency of the level densities used.

Based on the energy-dependent cross sections calculated with six level density models using the nuclear model code TALYS-1.95 [21], we calculated the flux-weighted average cross sections for the ¹⁸⁶W(γ , p)¹⁸⁵Ta reaction. The measured and calculated flux-weighted average cross sections are given in Table 2, namely the experimental result is given in the third column and the theoretical results calculated using the six level density models (Ldmode 1-Ldmode 6) are given in the fourth to ninth columns. For better visualization, the measured and calculated results are also presented in Fig. 5.

As shown in Fig. 5, the results calculated using the six level density models differ significantly. Further comparison shows that only the result calculated with Ldmode 2 agrees with the experimental result, namely at the bremsstrahlung endpoint energy of 70 MeV the difference between the measured and calculated results is only about 4%. Meanwhile, all other calculation results differ significantly from the experimental result, in particular the calculated result with Ldmode 3 differs about 60% from the experimental result. This fact suggests that more experimental data on the ¹⁸⁶W(γ , p)¹⁸⁶Ta reaction are needed for comparison and verification of the level density model of ¹⁸⁵Ta.

Nuclear reaction	Bremss. end-point energy (MeV)	Flux-weighted average cross section (mb)									
		Experimental	TALYS-1.95 calculation								
$^{186}\mathrm{W}(\gamma,p)^{185}\mathrm{Ta}$	70	result	Ldmode1	Ldmode2	Ldmod3	Ldmod4	Ldmod5	Ldmode6			
		0.105 ± 0.011	0.078	0.101	0.065	0.081	0.075	0.076			

Table 2. Measured and calculated flux-weighted average cross section of the ${}^{186}W(\gamma, p){}^{185}$ Ta reaction at the bremsstrahlung end-point energy of 70 MeV.



Fig. 5. (Color online) The measured and calculated flux-weighted average cross section of the ${}^{186}W(\gamma, p){}^{185}$ Ta reaction. The calculations were carried out with the six different level density models (Ldmode 1- Ldmode 6).

4. Conclusion

The flux-weighted average cross section of the ${}^{186}W(\gamma, p){}^{185}Ta$ reaction was measured at the bremsstrahlung end-point energy of 70 MeV. The measurement was carried out by the activation method in combination with off-line gamma spectrometry. Theoretical calculations were also carried out by incorporating the cross section values obtained from the TALYS 1.95 code and the bremsstrahlung flux simulated using the computer program MCNPX. When calculating the

352

reaction cross section, six level density models were considered and the results show that the fluxweighted average cross section of the ¹⁸⁶W(γ , p)¹⁸⁵Ta reaction calculated using Ldmode2 with the back-shifted Fermi gas model gives the best fit to the experimental result. The present result is published for the first time.

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