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Measurements of ^{181}Ta(n,2n)^{180}Ta reaction cross-section at the neutron energy of 14.78 MeV

Bhargav K Soni[a]*, Siddhart Parashari[a], S Mukherjee[a], Rajnikant Makwana[a], M Mehta[b], R Chauhan[a], S V Suryanarayana[c], I Pasha[d], L S Danu[c], H Naik[e], B K Nayak[c], J Varmuza[f] & K Katovsky[f]

[a]Department of Physics, Faculty of Science, The M S University of Baroda, Vadodara 390 002, India

[b]Institute for Plasma Research, Gandhinagar 382 428, India

[c]Nuclear Physics Division, Bhabha Atomic Research Centre, Mumbai 400 085, India

[d]Department of Physics, Bangalore University, Bengaluru 560 056, India

[e]Radio Chemistry Division, Bhabha Atomic Research Center, Mumbai 400 085, India

[f]Department of Electrical Power Engineering, Brno University of Technology, Brno, Czech Republic

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The cross-section of the ^{181}Ta(n,2n)^{180}Ta reaction has been measured with respect to the ^{197}Au(n,2n)^{196}Au monitor reaction at the incident neutron energy of 14.78± 0.20 MeV, using neutron activation analysis and off-line γ-ray spectrometric technique. The present measurement has been done at the energy where discrepant measured results are available in the EXFOR data library. The result has been compared with evaluated data libraries JEFF-3.3 and ENDF/B-VII.1. The present result has also been supported by theoretical predictions of nuclear model code TALYS1.8 and TALYS-1.9. The uncertainty and the correlations among the measured cross-section has been studied using co-variance analysis.

Keywords: Neutron Activation Analysis (NAA), ^{3}H(^{2}H, n)^{4}He reaction neutron, Off-line γ-ray spectrometry, Co-variance analysis, TALYS1.8, TALYS-1.9.

1 Introduction

Nuclear reaction cross-section data are very useful for upcoming nuclear technologies like Accelerator Driven Subcritical System (ADSs) and Advance Heavy Water Reactor (AHWER)[1,2]. Tantalum (Ta) is regarded as a control-rod material for lead-bismuth cooled fast reactors. Moreover, neutron-induced activation cross-sections of tantalum are needed[3] as for the decommissioning of light-water reactors[4]. Also, it is very crucial for both nuclear fission and fusion applications. It has a major constituent of the low activation ferritic-martensitic steel Eurofer which is qualified for future fusion reactors, it means of test irradiations in the International Fusion Irradiation Facility (IFMIF). Tantalum is a candidate material of the spallation target[5] to be used for accelerator driven subcritical systems (ADSs). The cross-section of ^{181}Ta(n,2n)^{180}Ta reaction around 14 MeV has been reported by various authors[6-10], but most of them were obtained before 1992, furthermore, there was disagreement in those data with theoretical nuclear code TALYS-1.8 and TALYS-1.9[11]. The natural tantalum consists of two isotopes, namely, ^{180m}Ta (0.012%) and ^{181}Ta (99.988%). ^{181}Ta is a stable isotope, while ^{180m}Ta is in the meta stable state that decays to ^{180}Ta by isomeric transition. From the compilation of EXchange FORmat[12] (EXFOR) data library it can be seen that, very few cross-section data are available for the ^{181}Ta(n,2n)^{180}Ta reaction from threshold to 20 MeV neutron energies.

In the present work, we have measured the cross-section datum for the ^{181}Ta(n, 2n)^{180}Ta nuclear reaction for the incident particle energy of 14.78 ± 0.20 MeV. The measured cross-sections have also been compared with available different level density models of the theoretical nuclear code TALYS-1.8 TALYS-1.9[11]. The present work is based on experimental measurement of the cross section in which the uncertainties have been calculated by using the covariance analysis[13,14]. This covariance analysis

*Corresponding author (Email: sonibhargav15@gmail.com)

help in identifying the error and the relative correlation among the different quantities used in the cross-section measurement. The present work was carried out to improve the evaluated nuclear data library on ^{181}Ta(n,2n)^{180}Ta reaction by considering the recent knowledge on experimental and theoretical nuclear physics.

2 Experimental Techniques

The measurements of the ^{181}Ta(n,2n)^{180}Ta and ^{197}Au(n,2n)^{196}Au reactions were carried out at the Cockcroft–Walton voltage multiplier accelerator of Purnima at Bhabha Atomic Research Center (BARC), Mumbai, India by using Neutron Activation Analysis (NAA) technique followed by γ-ray spectroscopy. D^+ ions were used to produce suitable energies of neutrons. The D^+ ions were produced in an RF ion-source and then accelerated up to 300 kV, the accelerated D^+ ions were incident on titanium–tritium (TiT) target to produce 14.1 MeV neutrons through the ^{3}H(^{2}H,n)^{4}He (Q-value=17.59 MeV) reaction[15]. In the present experiment the D^+ ions were accelerated to 99.71 keV which was impinge on titanium–tritium (TiT) target. This collision produces neutrons of energy 14.78 ± 0.20 MeV in the laboratory frame through the ^{3}H(^{2}H,n)^{4}He reaction, in at nearly forward angles. A Schematic diagram of the experimental set up for the neutron irradiation set up is given in Fig. 1. In our irradiation set up, the Ta and Au sample of area about 1 × 1 cm^2 were separately wrapped with Al foil of thickness 0.011 mm to shield the radioactive contamination during irradiation. The weights of ^{nat}Ta and ^{nat}Au metal foils were 239.12 mg and 334.30mg with 99.97 % purity, respectively. The stack of Ta-Au placed at a distance of 1.5 cm from the neutron generating (T-Ti) target at zero degree angle relative to the beam direction. The stack foils of Ta-Au were irradiated for 1.5 h. After the irradiation, samples were allowed to cool for 0.3 h to accumulate the radiation dose. The radioactive samples of Ta and Au along with Al wrapper were mounted on different Perspex plates and then taken for γ-ray spectrometry. The γ-ray counting of the irradiated samples (Ta & Au) were carried out using a lead shielded pre-calibrated 185-cc Baltic HPGe detector coupled with the PC-based 4096 channel analyzer. In the γ-ray counting, dead time of the detector was always kept less than 4% by keeping the mounted samples at a suitable distance from the detector end cap. The data acquisition was done using a CAMAC based LAMPS (Linux Advance Multi Parameter System) software. A standard ^{152}Eu source was used for the energy and efficiency calibration. The resolution of the detector system during counting was measured as 1.8 keV at 1332 keV of ^{60}Co. A typical gamma ray spectrum obtained from the irradiated ^{nat}Ta sample at 14.48 ± 0.20 MeV neutron energy is shown in Fig. 2.

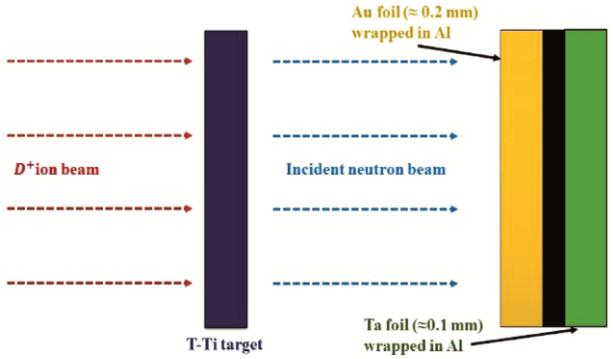

Fig. 1 — Schematic diagram showing the arrangements used for the neutron irradiation.

3 Data Analysis

3.1 Estimation of HPGe detector efficiency

The efficiency (ε) of the HPGe detector system is determined by using the standard ^{152}Eu source with the help of their known characteristic γ-ray energies. The variation in the efficiency with the γ-ray energy is totally independent with the detector geometry, but its absolute value depend on geometry[16]. The geometry dependent efficiency of the HPGe detector is given by the relation:

$$\varepsilon = K_c \frac{C}{A_0 I_\gamma e^{-\lambda T} \Delta t} \quad \ldots (1)$$

where C is the count of the photo peak of the characteristic ^{152}Eu γ-ray spectrum, A_0 is source

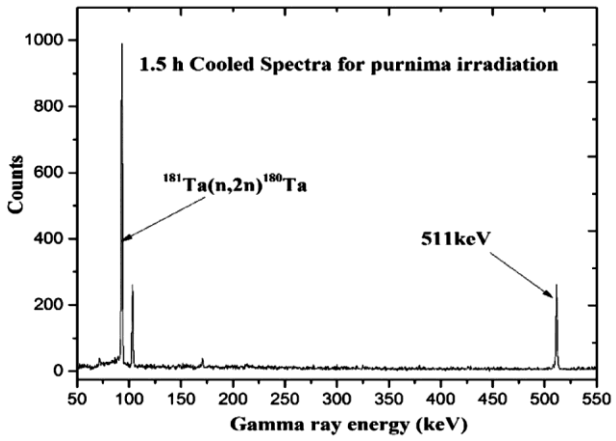

Fig. 2 — Gamma ray spectrum from irradiated natural Ta isotope at 14.78 ± 0.20 MeV neutron energy.

activity (6659.21 ± 81.60 Bq as on 1 October 1999), I_γ is the branching intensity of γ-rays were retrieved from NuDat 2.7[17] database, λ is the decay constant of product nucleus, T is the time interval between the source manufacturing data and observation. The correction factor (K_c) in the measured efficiency due to coincidence summing were estimated using Monte Carlo simulation code EFFTRAN[18]. The measured values of efficiency along with the polynomial fitting are shown in Fig. 3.

3.2 Neutron flux calculation

In relative cross-section measurement, the accurate neutron flux calculation is essential. For the resent work, ^{197}Au(n, 2n)^{196}Au monitor reaction was taken for the measurement of the neutron flux. The daughter product of reaction^{196}Au has a half-life of 6.169 ± 0.006 d[19]. The neutron flux of the reaction has been calculated by using the following relation:

$$\Phi = \frac{C_{obs}\,\lambda\left(\frac{t_c}{t_r}\right)}{N_0 I_\gamma \varepsilon\sigma\,(1-e^{-\lambda t_i})(1-e^{-\lambda t_c})(e^{-\lambda w})} \quad \ldots (2)$$

where C_{obs} is the counts observed from the measured ^{nat}Au γ-ray spectrum, λ is decay constant of ^{197}Au nucleus (s^{-1}),N_0number of target atom, I_γ branching intensity (87 ± 3 %) of γ-ray of ^{196}Au nucleus, ε efficiency of the detector related (^{196}Au = 355.7 keV) to chosen γ-ray, σ cross-section of the ^{197}Au(n, 2n)^{196}Au reaction at neutron energy of 14.78 MeV from EXFOR[12] data library, t_r clock time (s), t_c counting time (s), t_w cooling time (s), t_i =irradiation time (s), Φ is bombarded neutron flux(n cm^{-2}). The spectroscopic data related to the above reaction is given in Table 1. This measured value of the neutron flux from the monitor reaction have been used for estimation of the cross-section.

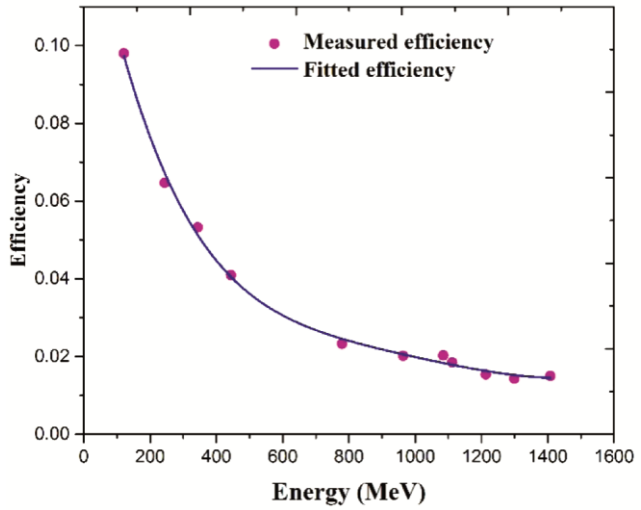

Fig. 3 — The measured efficiency with the 152Eu source keeping at 2cm from the detector head.

3.3 Neutron activation analysis

The experimental data were evaluated by using the delayed gamma-ray neutron activation analysis (DGNAA) technique. This technique is based on the measurement of the cross-section by irradiating the samples with neutrons in which, the rate of creation of the daughter isotope depends on the number of nuclei available in the target and the incident neutron flux[20]. This proportionality is given by the cross-section of the reaction. The activated sample during neutron irradiation emits characteristic γ-rays having an adequately long half-life and γ-ray branching abundances. By using the following equation, the cross-section of the reaction can be calculated:

$$\sigma = \frac{C_{obs}\,\lambda\left(\frac{t_c}{t_r}\right)}{N_0 I_\gamma \varepsilon\Phi(1-e^{-\lambda t_i})(1-e^{-\lambda t_c})(e^{-\lambda w})} \quad \ldots (3)$$

where all the symbols have their meanings similar to Eq. (2). In the Eq. (3), the peak area of the ^{197}Au(n,2n)^{196}Au reaction from the γ-ray spectrum was measured using LAMPS software. The number of target nuclei (N_0) was calculated using the mass of Ta target. ε efficiency of the detector at 93.32 keV γ-ray energy was calculated by extrapolation, Φ is calculated neutron flux from ^{197}Au(n, 2n)^{196}Au reaction were used in the Eq. (3). Other standard parameters of the reactions were taken from theNuDat[17] data library.

3.4 Covariance analysis

In the present experiment, the cross-sections were measured relative to the monitor reaction, and a

Table 1 — Nuclear Spectroscopic data used in the present measurements

Reaction	Threshold energy (MeV)	Spin state J^π	Half-life	Decay mode (%)	E_γ (KeV)	I_γ (%)
^{197}Au(n,2n)^{196}Au	8.113 ± 0.296	$(3/2)^+$	6.1669 ± 0.0006 d	ε(93) + β^-(7)	355.71 ± 0.6	87 ± 3
^{181}Ta(n,2n)180Ta	7.618 ± 0.135	1^+	8.154 ± 0.006 h	ε(85) + β^-(15)	93.32 ± 0.2	4.5 ± 0.4

common detector setup was used for recording of the γ-ray spectra from Au and Ta irradiated sample foil. Therefore, the measured cross-section is in correlation with the monitor reaction and with the efficiency of the detector used in the present measurement. The covariance analysis[13,14] was performed using a ratio method[14]. In this analysis, first we calculate the relative correlations and covariance among the efficiencies of the standard (^{152}Eu) and the sample (^{196}Au and ^{180}Ta) gamma lines. By using these correlation factors, which came in the first step, we deduce the correlation factors and the related covariance among the measured cross-section. The partial uncertainty contributing in the efficiency measurement given in Table 2, by using it the correlation and co-variance matrix for the efficiencies are calculated and it shown in Tables 3 and 4. The uncertainty in the measured cross-section of the estimated by multiplying the fractional uncertainties in various parameters to obtain cross-section of ^{181}Ta(n,2n) ^{180}Ta reaction and was found to be 15%, which is the least value for the present cross-section measurement by considering the error from each of the quantity used in the calculations.

4 Results and Discussion

The cross sections of the ^{181}Ta(n,2n)^{180}Ta reaction have been measured with respect to^{197}Au(n,2n)^{196}Au monitor reaction at the neutron energy of 14.78 ± 0.20 MeV, which is obtained as 1.83±0.15 (barns) and plotted in Fig. 4. The efficiency of HPGe detector was carried out using a standard ^{152}Eu source and the cross section datum of the reaction were determined using NAA and off-line γ-ray spectrometry technique. We have estimated the uncertainties in the measurement by considering various attributes in the data using the covariance analysis and correlations between them. The theoretical nuclear codes TALYS-1.8 and TALYS-1.9[11] were used for the analysis and prediction of nuclear reaction cross-section values based on the different level density model available in it.

The comparison of the ^{181}Ta(n,2n)^{180}Ta reaction cross-section with the evaluated data from JEFF-3.3[21] and ENDF/B-VII.1[22] libraries, literature data[6-10]from

Table 4 — Measured efficiency with correlation matrix

E_γ (KeV)	Efficiency	Correlation Matrix	
355.71	0.059811 ± 0.0008	1	
93.32	0.104731 ± 0.0012	0.91	1

Table 2 — Uncertainity contributing in the efficiency measurement

Energy (KeV)	Partial uncertainty (× 10^3)	Total uncertainty (× 10^3)			
	C	I_γ	N_0	$T_{1/2}$	(σ_{eii})
121.8	0.501621	2.786854	5.657249	0.017541	6.230154
244	0.692513	1.564532	3.351265	0.010125	3.651169
344	0.265107	1.653215	2.452846	0.007545	3.016710
411	0.993210	0.874145	1.718635	0.005123	2.187452
778.9	0.340125	0.603151	1.094105	0.003214	1.321628
867	0.923510	0.635410	1.011235	0.003151	1.532147
964	0.253974	0.398471	0.879845	0.002591	1.012105
1112	0.213503	0.403814	0.782146	0.002255	0.901768
1212	0.942130	0.384639	0.652149	0.001898	1.132546
1299	0.738596	0.334648	0.662456	0.001947	1.087452
1408	0.121142	0.251646	0.251486	0.001911	0.689526

Table 3 — Co-variance matrix (× 100) for the detector efficiency

0.004132										
0.001852	0.001467									
0.001546	0.000824	0.000987								
0.001014	0.000522	0.004317	0.000474							
0.000654	0.000321	0.000287	0.000197	0.000170						
0.000584	0.000125	0.000256	0.000174	0.000214	0.000227					
0.000519	0.000312	0.000122	0.000156	0.000864	0.000412	0.000365				
0.000352	0.000170	0.000156	0.000287	0.000197	0.000150	0.000101	0.000094			
0.000325	0.000215	0.000321	0.000256	0.000132	0.000120	0.000156	0.000099	0.00035		
0.000369	0.000174	0.000214	0.000198	0.000095	0.000321	0.000200	0.000410	0.000325	0.000156	
0.000312	0.000235	0.000864	0.000789	0.000523	0.000400	0.000345	0.000215	0.000113	0.000049	0.000035

EXFOR[12], as well as the theoretically prediction nuclear code TALYS-1.9[11] within 8-24 MeV are shown in Fig. 4. It is observed from Fig. 4, that the ^{181}Ta(n,2n)^{180}Ta reaction cross-section of present measurement at the neutron energy of 14.78 ± 0.20 MeV is in excellent agreement with the ldmodel-3 which is generalized superfluid model of the TALYS-1.9[11] and in close agreement with the previous experimental data of Frehaut *et al*.[9]. However, the prediction of the L. R. Veeser *et al*.[8], as well as evaluated from JEFF-3.3[21], and ENDF/B-VII.1[22] libraries are slightly higher. It suggests that more experimental data are needed to be obtained and compared with the different nuclear models. A detailed covariance analysis has also been carried out in order to find out the exact values of the uncertainties in the measured data. Covariance analysis is one of the methods which can help us to calculate the uncertainty in the measured datum by propagating the error in each quantity used.

5 Conclusions

The experimental cross-section for the ^{181}Ta(n, 2n)^{180}Ta reaction have been measured at the neutron energy of 14.78 ± 0.20 relative to the ^{197}Au(n,2n)^{196}Au monitor reaction by using the NAA technique and off-line γ-ray spectrometry technique. The efficiency of HPGe detector system was calculated by using ^{152}Eu standard source along with coincidence summing effect. The polynomial fitting is chosen to estimate the efficiency of unknown γ-ray energies. The uncertainty in the present measurement calculated with the help of covariance analysis was found to be 15%. The measurement has been compared with the literature data[6-10], JEFF-3.3[21], ENDF/B-VII.1[22]and the theoretical modular code TALYS-1.8 and TALYS-1.9[11]. A comparison of the present result shows a good agreement with literature data Frehaut *et al*.[9] as well as with ldmodel-3 of the TALYS-1.9[11]code. The present work highlights that the nuclear reaction datum can be measured within the uncertainty of ≈15% by using the ^{3}H(^{2}H, n)^{4}He reaction neutron generator. The cross-section data presented in this work are essential for the data libraries, and for the future ADSs and IFMIF reactor technology.

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