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Measurement of ¹⁹⁷Au(n, γ)^{198g}Au reaction cross-section at the neutron energies of 1.12, 2.12, 3.12 and 4.12 MeV

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Abstract: The ¹⁹⁷Au(n, γ)¹⁹⁸Au reaction cross-sections at the neutron energies of 1.12, 2.12, 3.12 and 4.12 MeV were determined by using activation and off-line y-ray spectrometric technique. The mono-energetic neutron energies of 1.12-4.12 MeV were generated from the ⁷Li(p, n) reaction by using the proton energies of 3 and 4 MeV from the folded tandem ion beam accelerator (FOTIA) at BARC as well as 5 and 6 MeV from the Pelletron facility at TIFR, Mumbai. The $^{115}In(n, \gamma)^{116m}In$ reaction cross-section was used as the neutron flux monitor. The 197 Au(n, γ) 198 Au reaction cross-section at the neutron energies of 3.12 and 4.12 MeV are reported for the first time. The 197 Au(n, γ) 198 Au reaction cross-sections at 1.12 and 2.12 MeV are close to the literature data of in between neutron energies. The 197 Au $(n, \gamma)^{198}$ Au cross-section was also calculated theoretically by using the computer code TALYS 1.6 and found to be higher than the experimental data of present work and literature data within the neutron energies of 0.8 to 4 MeV.

Keywords: ¹⁹⁷Au(n, γ)¹⁹⁸Au reaction cross-section, off-line γ -ray spectrometric technique, 115 In $(n,\gamma)^{116m}$ In reaction monitor, TALYS calculation.

1 Introduction

For the determination of neutron activation cross-section at low neutron energy, $^{197}Au(n, \gamma)$ reaction cross-section is used as the neutron flux monitor. The $^{197}Au(n,\gamma)$ reaction cross-section at thermal neutron energy is known very accurately based on the evaluation and compilation of Mughabghab et al [1]. However, the reactor neutron has a spectrum within energy of thermal to 15 MeV. If it is desired to use the ${}^{197}Au(n, \gamma)$ reaction as a flux monitor at higher neutron energy then it is necessary to know the 197 Au(n, γ) reaction cross-section accurately at different neutron energies. In earlier days, it was difficult to obtain a mono-energetic neutron source with sufficient flux, except for the neutron energy of 2.45 MeV from the D + D reaction and of 14.8 MeV from the T + D reaction. However, in recent years mono-energetic neutrons from different reactions with good flux have become available. Among them, ${}^{7}Li(p, n)$ reaction is a good mono-energetic neutron source within the proton energy of 6 MeV.

Sufficient ¹⁹⁷Au(n, γ) reaction cross-section data within the neutron energy range of 0.025 eV to 3 MeV and at 14.7 have been compiled in the EXFOR [2] based on the experimental data of various authors [3-16]. The 197 Au(n, γ) reaction cross-section was measured by various authors at the neutron energy of 0.025 eV [3, 4]. Similarly, data are available for the neutron energies of 0.024 to 3 MeV [5-13, 16] and at 14.7 MeV [14, 15]. Besides this, no experimental $^{197}Au(n, \gamma)$ reaction cross-section

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data are available over neutron energy range of 3 to 14.7 MeV. In view of this, in the present work we have determined the reaction cross-section of ¹⁹⁷Au with the neutron energies of 1.12, 2.12, 3.12 and 4.12 MeV by using the off-line gamma ray spectrometric technique. The ¹⁹⁷Au(n, γ)^{198g}Au reaction cross-section was also calculated theoretically by using the computer code TALYS 1.6 [17] and compared with the experimental data from literature and present work.

2 Experimental details and calculations

For the measurement of $^{197}Au(n, \gamma)^{198g}Au$ reaction crosssections, experiments with four different neutron energies were carried out at two different places. The fast neutrons were produced from the $^{7}Li(p, n)^{7}Be$ reaction by using the proton beam of 3, 4, 5 and 6 MeV. The Qvalue for the ${}^{7}Li(p, n){}^{7}Be$ reaction to the ground state is -1.644 MeV, whereas the first excited state is 0.431 MeV above the ground state leading to an average Q-value of -1.868 MeV. The threshold value of the ⁷Li(p,n) reaction to the ground state of ⁷Be is 1.881 MeV. Thus for the proton energy of 3, 4, 5 and 6 MeV the resulting peak energy of the first group of neutrons (n_0) would be 1.12, 2.12, 3.12 and 4.12 MeV, respectively. The corresponding neutron energy of the second group of neutrons (n_1) , for the first excited state of ⁷Be, will be 0.63, 1.63, 2.63 and 3.63 MeV, respectively [18-20]. This is because above the proton energy of 2.37 MeV, the n_1 group of neutrons is also produced. However, the contribution from the n_1 group neutrons is negligible within the proton energies of 6 MeV. For the proton energies of 4–6 MeV, the neutron spectrum consists of the full energy peak due to the ${}^{7}Li(p, n){}^{7}Be$ reaction and a continuum component attributable to the multi-body break up process, i.e. ${}^{7}\text{Li}(p, n^{3}\text{He})\alpha$ (Q = -3.231 MeV). However, the energies of the emitted neutrons under the main peak were estimated to be 1.12 ± 0.11 , 2.12 ± 0.15 , 3.12 ± 0.21 , and 4.12 ± 0.32 MeV, respectively.

The first experiment with the neutron energies of 1.12 and 2.12 MeV was performed using the folded tandem ion beam accelerator (FOTIA) at Van-de-Graff, BARC, Mumbai. A circular LiF pellet of 1 cm diameter and 3 mm thickness was used for neutron production. It was fixed on a stand inside in a zero degree angle of the beam exit window. A beam collimator of 10 mm diameter was used before the target. The current of incident proton beam during the irradiations was 100 nA for both 3 and 4 MeV. The 3 mm thick LiF pellet is sufficient to stop the proton beam of 3 and 4 MeV. About 52.7 to 59.1 mg of Au metal foil with purity ~99.99% was wrapped with 0.025 mm thick Al foil. Similarly, 98.8 to 163.7 mg of In metal foil was also wrapped with 0.025 mm thick Al foil to prevent contamination from one another. The ¹¹⁵In(n, γ)^{116m}In reaction of the In metal foil was used as neutron flux monitor. Two sets of Al wrapped Au and In metal samples stack were made for two different irradiations. The Au-In stack was wrapped with additional Al foil and kept at a distance of 3 mm behind the LiF target. The stacks of samples were irradiated one set at a time with the neutron beam energies of 1.12 and 2.12 MeV, for 8.6 h and 5.6 h, respectively.

The second experiment was performed using the 14 UD BARC-TIFR Pelletron facility at TIFR, Mumbai with neutron energies of 3.12 and 4.12 MeV [21]. They were generated from the ${}^{7}Li(p, n){}^{7}Be$ reaction by using the proton energies of 5 and 6 MeV. The current of incident proton beam during the irradiations was 50 nA at 5 MeV and 60 nA at 6 MeV. These runs were carried out at 6 m height above the analysing magnet of the Pelletron facility to utilize the maximum proton current from the accelerator [21]. At this port, the terminal voltage is regulated by generating voltage mode (GVM) by using a terminal potential stabilizer. Furthermore, we use a beam collimator of 6 mm diameter before the target. The lithium foil was made up of natural lithium with thickness 3.7 mg/cm^2 , sandwiched between two tantalum foils of different thicknesses. The front tantalum foil facing the proton beam was the thinnest one (3.9 mg/cm^2) , in which the degradation of proton energy is 50-85 keV [22]. On the other hand, the back tantalum foil (beam stopper) was thicker (0.025 mm), which is used to stop the proton beam. For each irradiation, separate samples were made by packing 59-59.4 mg of Au metal foil wrapped with 0.025 mm thick aluminium foil. Similarly, 58.1-64.3 mg of In metal foil was also wrapped separately with 0.025 mm thick Al foil. Then two different sets of Al-wrapped Au and In metal stacks were prepared for different irradiations at neutron energies of 3.12 and 4.12 MeV, respectively. The individual stacks of Au-In samples were additionally wrapped with two different Al foils. The Au-In stack was mounted at zero degree angle with respect to the proton beam direction at a distance of 2.1 cm behind the Ta-Li-Ta stack [21]. A schematic diagram of the Ta-Li-Ta stack and of the Au-In stack is given in Figure 1. The samples were irradiated one set at a time with the neutron beam energies of 3.12 and 4.12 MeV. The irradiation time was 11.8 h for the neutron energy of 3.12 MeV and 13.4 h for 4.12 MeV. The neutron background during and before experiment was also



Fig. 1: Schematic diagram showing the arrangement used for neutron irradiation.

tested by keeping an inactive Au foil outside the irradiation chamber and was found to be absent.

In all those above experiments, the irradiated samples of Au and In were cooled for 2.3–19 h and 0.7–1.25 h, respectively. Then the γ -ray counting of the samples was done by using pre-calibrated 80 cm³ HPGe detector couple to a PC based 4094 channel analyser. A ¹⁵²Eu standard source was used for the energy and efficiency calibration. The resolution of the detector system during counting was 2.0 keV at 1332 keV of ⁶⁰Co. Typical gamma ray spectra of irradiated Au and In metal foils are given in Figures 2 and 3, respectively.

From the photo-peak activity ($A_{\rm obs}$) of the 138.33, 416.86, 818.72, 1097.33 and 1293.56 keV γ -lines of ^{116m}In, the neutron flux (Φ) was obtained by using Equation (1) from ref. [21]

$$A_{\rm obs}({\rm CL/LT}) = N\sigma_R \Phi I_{\gamma} \varepsilon (1 - \exp(-\lambda t)) \\ \times \exp(-\lambda T) \times (1 - \exp(\lambda {\rm CL}))/\lambda$$
(1)



Fig. 2: Gamma ray spectrum of irradiated ^{nat}In foil with the neutron energy of 1.12 MeV; irradiation time = 8.583 h, cooling time = 42 min, and counting time = 10 min.



Fig. 3: Gamma ray spectrum of irradiated 197 Au foil with the neutron energy of 2.12 MeV; irradiation time = 5.55 h, cooling time = 44.47 h, and counting time = 3.216 h.

where, *N* is the number of target atoms and $\sigma_{\rm R}$ is the crosssection of the ¹¹⁵In(n, γ)^{116m}In reaction. λ is the decay constant ($\lambda = \ln 2/T_{1/2}$) of the reaction product of interest with half-life = $T_{1/2}$. I_{γ} is the branching intensity of the 138.33, 416.86, 818.72, 1097.33 and 1293.56 keV γ -line of ^{116m}In [23] and ε is its detection efficiency. *t*, *T*, CL and LT are the irradiation time, cooling time, real and live-time of counting, respectively.

The 115 In(n, γ) 116m In reaction cross-section is available for a wide range of neutron energies in literature from the work of different authors [24-34]. However, the 115 In(n, γ)^{116m}In reaction cross-sections within the neutron energy range of 1.96 to 7.66 MeV are available in the refs. [29, 31, 32, 34]. Among them, the cross-sections obtained by Husain et al. [31] are within the neutron energy range of 2.44 to 4.5 MeV and are higher than the data of others [29, 32, 34]. Thus the ${}^{115}In(n, \gamma){}^{116m}In$ reaction cross-sections from the refs. [29, 32, 34] were used in the present work for the neutron flux calculation. The evaluated $^{115}In(n, \gamma)^{116m}In$ reaction crosssections as a function of neutron energy are available in ref. [35]. Thus the neutron flux based on the evaluated 115 In(n, γ)^{116m}In reaction cross-sections were also used to determine the neutron flux. The neutron fluxes obtained from the method described above were used in rearranged Eq. (1) to calculate the ${}^{197}Au(n, \gamma){}^{198g}Au$ reaction cross-section by using the photo-peak activity of the 412.8 keV γ -ray of ^{198g}Au. The nuclear spectroscopic data used in the above calculations were taken from the ref. [23] and are presented in Table 1. Since the neutron spectrum has some tail, it has effect on the ${}^{197}Au(n, \gamma){}^{198}Au$ reaction cross-section. At the same time, it also affects the 115 In(n, γ)^{116m}In monitor reaction cross-section. This is

Nuclide	Spin-parity	Half-life	Decay mode	γ-ray energy	γ-ray abundance (%)
^{116m} In	5+	54.29 ± 0.17 min	β^- (100%)	138.29	3.7
				416.9	27.2
				818.68	12.13
				1097.28	58.5
				1293.56	84.8
¹⁹⁸ <i>g</i> Au	2 ⁻	$\textbf{2.6947} \pm \textbf{0.0003} \; \textbf{d}$	$eta^{}$ (100%)	411.8	95.62

Table 1: Nuclear spectroscopic data of the radionuclides from the 197 Au(n, γ) 198g Au and 115 In(n, γ) 116g In reactions used in the calculation from Ref. [23]. The bold numbers are the γ -ray energies, whose activities were

used in the calculation.

because both the ¹¹⁵In(n, γ)^{116m}In and ¹⁹⁷Au(n, γ)¹⁹⁸Au reaction cross-sections decrease with neutron energy. The contribution in the reaction cross-section due to small tail part is discussed in the next section given below.

3 Results and discussion

The 197 Au(n, γ) 198g Au reaction cross-sections determined in the present work at the neutron energies of 1.12, 2.12, 3.12 and 4.12 MeV are given in Table 2. The uncertainties associated with the measured cross-section values of present work are from the replicate measurements. The overall uncertainty is the quadratic sum of both statistical and systematic errors. The random error in the observed activity is primarily due to counting statistics, which is estimated to be 3.1%-5.5% for ^{198g}Au. This can be determined by accumulating the data for an optimum time period that depends on the half-life of the nuclide of interest. The systematic errors are due to uncertainties in the irradiation time ($\sim 0.1\%$), the half-life of the reaction products and the γ -ray abundances (~2%) and the detection efficiency $(\sim 3\%)$, which arises from the fitting error. Thus the total systematic error is about ~3.6%. The combined uncertainties from both statistical and systematic errors lie within 4.8%–6.6% for the 197 Au(n, γ) 198g Au reaction cross-section. The energy degradation of the proton beam within the lithium metal foil as well as the 115 In(n, γ) 116m In monitor reaction cross-section can cause additional uncertainty. However, this uncertainty is less prominent than the neutron energy uncertainty due to the different extent of population of n₀ and n₁ groups of neutrons.

In order to examine the above aspect, the neutron spectra from the 7 Li(p, n) reaction at the proton energies of 4, 5 and 6 MeV were calculated as done in our earlier work [21] based on refs. [18-20] and the results are plotted in Figure 4. As mentioned before, the neutron spectrum consists of the full energy peak due to the ${}^{7}Li(p, n_{0,1}){}^{7}Be$ reaction and a continuum component attributable to the multi-body break up process, i.e. ⁷Li(p, n³He) α (Q = -3.231 MeV). This is primarily for the proton energy of 6 MeV but not for 3-5 MeV. However, from the Figure 4, the different extent of population of n_0 and n_1 groups of neutrons, which causes the broadening of the neutron spectrum, can be very well seen. Since the neutron spectrum for the proton energy of 6 MeV has low energy tail, it contributes to the crosssection of the ¹¹⁵In(n, γ)^{116m}In reaction monitor as well as to the ${}^{197}Au(n, \gamma){}^{198}gAu$ reaction. For this purpose, the 115 In(n, γ) 116m In and 197 Au(n, γ) 198g Au reaction crosssections were theoretically calculated by using TALYS 1,6 [17]. This computer code can be used to calculate the reaction cross-section based on physics models and pa-

	¹¹⁵ In (n,γ) ^{116m} In	¹⁹⁷ Au(n, γ) ^{198g} Au		
Neutron energy (MeV)	Cross section (mb)	Flux n cm ^{-2} s ^{-1}	Cross section (mb) Experimental [ref.]	TALYS-1.6
$ \begin{array}{r} 1.12 \pm 0.12 \\ 1.12 \pm 0.12 \\ 1.232 \end{array} $	174.4 ± 6.7 [35] 247.1 [34]	$(9.421 \pm 0.122) \times 10^{6}$ $(6.649 \pm 0.086) \times 10^{6}$	$60.441 \pm 3.403 \\ 85.639 \pm 4.822 \\ 82.5 \pm 3.5 [12]$	82.872
2.12 ± 0.15 2.12 ± 0.15 2.0	104.1 ± 4.5 [35] 129.0 [29]	$(2.257 \pm 0.155) \times 10^{7}$ $(1.821 \pm 0.125) \times 10^{7}$	40.588 ± 3.063 50.306 ± 3.795 56.3 ± 2.4 [12]	79.866
3.12 ± 0.21 3.12 ± 0.21 3.0	37.5 ± 1.5 [35] 44.6 [32]	$(2.243 \pm 0.272) \times 10^{7}$ $(1.886 \pm 0.229) \times 10^{7}$	19.771 ± 2.673 23.514 ± 3.182 25.0 ± 1.1 [12]	41.255
4.12 ± 0.32 4.12 ± 0.32	15.6 ± 0.7 [35] 16.7 [32]	$(3.062 \pm 0.319) \times 10^{7}$ $(2.860 \pm 0.298) \times 10^{7}$	13.944 ± 1.611 14.929 ± 1.725	19.180

Table 2: 197 Au (n, γ) 198g Au and 115 In (n, γ) 116m In reactioncross-sections at differentneutron energies.



Fig. 4: Neutron spectrum from the ${}^{6}Li(p,n)$ reaction for the proton energy of 6 MeV.

rameterizations. It calculates nuclear reactions involving targets with mass larger than 12 amu and projectiles like photon, neutron, proton, ²H, ³H, ³He and alpha particles in the energy range from 1 keV to 200 MeV. In the present work, we did the TALYS calculation by using the default parameters for the neutron energies of 10 keV to 20 MeV with ¹¹⁵In and ¹⁹⁷Au as targets. All possible outgoing channels for a given projectile (neutron) energy were considered including (n, γ) and inelastic reactions. However, the (n, v) reaction cross-sections within the neutron energy of 10 keV to 20 MeV were collected. The theoretical reaction cross-sections from TALYS are for the mono-energetic neutrons. Thus the flux-weighted average ${}^{115}In(n, \gamma){}^{116m}In$ and ${}^{197}Au(n, \gamma){}^{198g}Au$ reaction cross-sections ($\langle \sigma_{\rm R} \rangle$) were calculated using Eq. (2) as given in ref. [21].

$$(\langle \sigma_{\rm R} \rangle) = \sum \sigma_{\rm R} \cdot \varphi / \sum \varphi$$
 (2)

Using the neutron spectrum of Figure 4 and theoretical values from TALYS, the flux-weighted average ¹¹⁵In(n, γ)^{116m}In and ¹⁹⁷Au(n, γ)^{198g}Au reaction crosssections for the neutron energy of 4.12 MeV were obtained as 53.141 and 45.692 mb, respectively. Based on the theoretical flux-weighted average ¹¹⁵In(n, γ)^{116m}In reaction cross-section of 53.141 mb and the photo-peak activity of 412.8 keV γ -ray of ^{198g}Au, the ¹⁹⁷Au(n, γ)^{198g}Au reaction cross-section was obtained as 47.506 mb. This is close to the theoretical flux-weighted average ¹⁹⁷Au(n, γ)^{198g}Au reaction cross-section of 45.692 mb. Thus for the neutron energy of 4.12 MeV, the experimental ¹¹⁵In(n, γ)^{116m}In reaction cross-section of 16.7 mb [32] and the photo-peak activity of 412.8 keV γ -ray of ^{198g}Au, the ¹⁹⁷Au(n, γ)^{198g}Au reaction cross-section obtained as 14.926 mb is reasonable. This is because both the ${}^{115}In(n, \gamma){}^{116m}In$ and 197 Au(n, γ) 198 Au reaction cross-sections decrease in parallel with increase of neutron energy. Thus the contribution to the ¹⁹⁷Au(n, γ)^{198g}Au reaction cross-section due to the tail part of the neutron spectrum has been avoided by using the ${}^{115}In(n, \gamma){}^{116m}In$ reaction cross-section as the neutron flux monitor. Based on the above argument, the 197 Au $(n, \gamma)^{198g}$ Au reaction cross-section shown in Table 2 using the experimental ¹¹⁵In(n, γ)^{116m}In reaction crosssection at the neutron energies of 1.12-4.12 MeV are reasonable. For the proton energies of 3, 4 and 5 MeV, the neutron spectra have no tail part of the neutrons. Thus the 197 Au(n, γ) 198g Au reaction cross-section at the neutron energies of 1.12, 2.12 and 3.12 MeV does not need the corrections.

The 197 Au(n, γ) 198g Au reaction cross-sections at the neutron energies of 1.12, 2.12, 3.12 and 4.12 MeV were determined in the present work for the first time. There is no data available in literature [5-16] within the neutron energy range of 3-4.5 MeV. However, data at the neutron energy of 0.025 eV [1-3], within 0.2-3 MeV [7-13, 16] and at 14.7 MeV [14, 15] are available in literature. The experimental data of present work at the neutron energies of 1.12 and 2.12 MeV lie in between the values within the neutron energies of 1 to 1.2 MeV and 2 to 2.2 MeV, respectively. However, for all four neutron energies, the 197 Au(n, γ) 198g Au reaction cross-sections based on the evaluated 115 In(n, γ) 116m In reaction cross-sections [35] are lower than the experimental values. In view of this, the theoretical 197 Au(n, γ) 198g Au reaction cross-sections from TALYS 1.6 at the neutron energies of 1.12, 2.12, 3.12 and 4.12 MeV are shown in Table 2 for comparison.

It can be seen from Table 2 that the experimentally determined reaction cross-sections from the present work at the neutron energies of 1.12 and 4.12 are in general agreement with the theoretical values of TALYS 1.6 [17]. However, the data at the neutron energies of 2.12 and 3.12 MeV are lower than the theoretical values of TALYS 1.6. It can be also seen from Table 2 that the 197 Au $(n, \gamma)^{198g}$ Au reaction cross-sections based on the evaluated ¹¹⁵In (n, γ) ^{116m}In reaction cross-sections are very much lower than the theoretical values of TALYS 1.6. The experimental data from the present work at the four neutron energies and literature data [3–16] at other neutron energies along with the theoretical values from TALYS 1.6 within the 10 keV to 20 MeV are plotted in Figure 5. It can be seen that the experimental data from the present work and the literature data [7-13] within the neutron energies of 2.12-4.12 MeV are slightly lower than the theoretical values but follow a similar trend closely. The $^{197}Au(n, \gamma)^{198g}Au$ reaction



Fig. 5: Plot of the experimental and theoretical ¹⁹⁷Au(n, γ)^{198g}Au reaction cross-section as a function of the neutron energy from 10 keV to 20 MeV. Experimental values from the present work and from Refs. [3–16] are in different symbols and colour, whereas the theoretical values from TALYS are in solid square.

cross-section at different neutron energies is important from the point of view of its use for the determination of neutron flux during other reaction cross-section measurements. It is a well-known fact that the ¹⁹⁷Au(n, γ)^{198g}Au reaction cross-section as a function of neutron energy is one among the nine of the IAEA standard reaction monitors. Based on the IAEA reaction cross-section standards, the uncertainties for the neutron capture cross-section of Au should be within 1.4%–2.2%. Since we did the measurements of Au(n, γ) reaction cross-section based on the experimental ¹¹⁵In(n, γ)^{116m}In reaction cross section, the uncertainty is on the higher side. This can be mentioned by the evaluator. Besides this, the experimentally determined ¹⁹⁷Au(n, γ)^{198g}Au reaction cross-section is important for the test of TALYS model.

4 Conclusions

We determined the ¹⁹⁷Au(n, γ)^{198g}Au reaction crosssection at the neutron energies of 1.12, 2.12, 3.12 and 4.12 MeV by using an activation and off-line γ -ray spectrometric technique. The values for the latter two energies are reported for the first time. Our data at the neutron energies of 1.12 and 2.12 MeV are in agreement with the literature data. The ¹⁹⁷Au(n, γ)^{198g}Au reaction cross-section as a function of neutron energy was also calculated theoretically using the computer code TALYS 1.6 version. The present data at four neutron energies and the literature data at other neutron energies are systematically lower than the theoretical values.

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