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Measurement of ^{100}Mo (n, 2n) ^{99}Mo reaction cross section and covariance analysis using extended unscented transformation technique at the incident neutron energy of 13.9 MeV

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In this paper, the measurement and covariance analysis of the cross section of ^{100}Mo (n, 2n) ^{99}Mo reaction, with the ^{197}Au (n, 2n)^{196} Au reaction being used as the monitor, at the incident neutron energy of 13.9 MeV is reported. The ^{3}H (d, n) ^{4}He nuclear reaction is used as the neutron source. The experiment was performed at the Purnima neutron facility, BARC. The method of activation with off-line γ-ray spectrometry is used. The covariance analysis of the ^{100}Mo (n, 2n) ^{99}Mo reaction is also performed, for the first time, using the extended unscented transformation (EUT) technique[1], which is an extension of unscented transformation (UT) technique[2], for the determination of partial uncertainties arising due to attributes in combination with the micro-correlation technique of Geraldo and Smith[3]. The present results obtained for ^{100}Mo (n, 2n) ^{99}Mo reaction cross section are found to be in good agreement with EXFOR data and the theoretically calculated value using the TALYS 1. 8 code. Comparisons with the data in the available basic evaluated nuclear data libraries, such as ENDF/B-VIII.0, JEFF-3.3, JENDL-4.0, ROSFOND-2010, CENDL-3.1 and TENDL 2017 are also presented and discussed.

Keywords: Extended unscented transformation, ^{100}Mo (n, 2n) ^{99}Mo activation reaction cross-section, Off-line γ-ray spectrometry, Covariance analysis, TALYS 1.8, ENDF-B/VIII.0

1 Introduction

The technique of determination of reaction cross sections by the measurement of the activity produced in the sample after irradiating it with a beam of particles is known as activation technique. In this technique, the reaction cross sections are calculated using direct attributes, such as counts, gamma ray intensities, half-life, irradiation time, cooling time, counting time and other auxiliary attributes such as atomic mass, isotopic abundance and many others. Many of the attributes are associated with uncertainties that further propagate through the functional relationship and ultimately lead to the final uncertainties in the reaction cross sections[4].

Among the various uncertainty propagation techniques, the Monte Carlo (MC) method gives the best estimate for the uncertainties propagated through nonlinear transformations. However, it involves propagation of large number of histories through nonlinear transformations resulting in huge amount of computations. The Unscented Transformation (UT) method, is another uncertainty propagation technique which finds extensive applications in error estimation studies. This technique works on the two basic principles, namely that instead of propagating the entire probability density function (pdf) through a nonlinear transformation, it is easier to propagate a set of individual points called sigma points and secondly that it is easier to find these sigma points which have a sample pdf that closely approximates the true pdf of a state vector[5]. So, in the UT technique, sigma points are nonlinearly transformed rather than nonlinearly transforming the entire pdf. Depending on the nature of nonlinear transformation and its application[5], there are different forms of UT techniques based on the principle used in the selection of sigma points. In our earlier paper[6]it has been established that the extended unscented transformation (EUT) technique, which is a form of unscented transformation, gives results, for higher moments, in better agreement with the Monte Carlo method than the unscented transformation technique and so is much better than the UT technique for large nonlinearities and high uncertainties.

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In the present work it is aimed to determine the neutron induced reaction cross-section for the ^{100}Mo (n, 2n) ^{99}Mo reaction at the incident neutron energy of 13.9 MeV and then perform covariance analysis using EUT technique. The ^{100}Mo (n, 2n) ^{99}Mo reaction has been selected for study and analysis as ^{99}Mo is a very useful radionuclide which is involved in the production of medically significant ^{99m}Tc radionuclide. ^{99m}Tc is a nuclear isomer which is metastable and remains in the excited state much longer and eventually de-excites to its ground state by emitting gamma rays. ^{99m}Tc finds extensive applications in various nuclear imaging procedures as a radioactive tracer for functional imaging studies of the heart, brain, thyroid, lungs, kidneys and tumors.

2 Details of the Experiment

An experiment was performed using the Cockcroft-Walton voltage multiplier accelerator housed at the Purnima reactor at Bhabha Atomic Research Center (BARC), Mumbai. In this neutron generator, an RF ion source is used for generation of the D^+ ions which are accelerated up to 300 kV. These D^+ ions are then focused on the target, which is a titanium–tritium (TiT) target maintained at ground potential. This resulted in the production of quasi-mono-energetic neutrons of 13.9 MeV through the ^{3}H(d,n)^{4}He nuclear reaction[8].

The arrangement of the various sample and monitor foils are shown in Fig. 1. The neutron beam energy is almost constant in the forward direction up to $\pm 10^0$ of the laboratory frame. The molybdenum sample irradiated was approximately of 165.8 mg weight and a square of area 2.0×2.0 cm^2. During neutron irradiation both the sample and monitor were wrapped with 0.011 mm thick aluminum foil to avoid the radioactive contamination from one foil to the other. The stack of sample-monitor foils was mounted at zero-degree angle relative to the beam direction[9] and were irradiated for 1.5 hours and then shifted to the off-line γ-ray spectroscopy setup for measurement of gamma-ray counts. The irradiated sample foil was mounted separately in a Perspex plate and gamma-ray counting was performed after 53.44 and 100.97 hours of cooling of the foil. Then the monitor foil was mounted in another Perspex plate and its gamma-ray counting was done after 26.07 h of cooling of the foil. For neutron flux measurement the γ-ray activity of ^{196}Au produced from the ^{197}Au (n, 2n) monitor reaction was used.

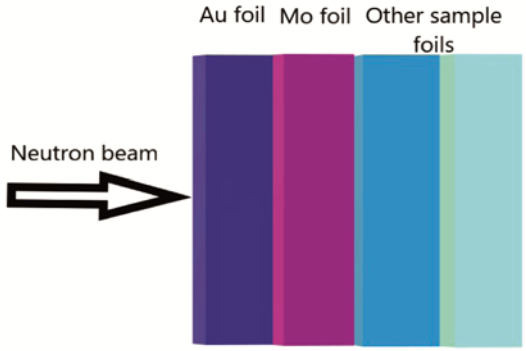

Fig. 1 — Experimental setup of sample and monitor foils.

The dead time of the γ-ray spectrometry was kept lower than 5% by keeping the distance between the detector end cap and the foils of sample and monitor at a distance of 1 cm. The counts of γ-rays emitted from the irradiated foils of sample ^{99}Mo and monitor ^{196}Au were calculated using a high-purity germanium (HPGe) detector and the details of the technique are already reported in our earlier paper[7]. The data of sample and monitor counts were acquired using Computer Automated Measurement and Control (CAMAC) based Linux Advanced Multi-parameter System (LAMPS) software.

Before the actual measurement of counts, the first step was to perform the calibration of efficiency of high purity germanium (HPGe) detector using various standard radioactive γ-ray sources such as ^{152}Eu and ^{133}Ba whose activity is accurately known[10]. The mean and uncertainty of efficiency of HPGe detector is determined for application in the covariance analysis of reaction cross section. This was performed using EUT technique, for the first time and the procedure was reported in our earlier paper[7]. Also the comparison of the results of the mean and uncertainty determined using Sandwich formula, MC, UT and EUT techniques were reported in[7] and it was seen that the EUT method estimated both mean and uncertainty in better agreement with MC method in comparison to the other two methods, which makes it superior for nonlinear studies.

3 Determination of Reaction Cross Section of ^{100}mo (N, 2n) ^{99}mo Reaction

3.1 Estimation of ^{100}Mo (n, 2n) ^{99}Mo reaction cross section

To estimate the reaction cross section of ^{100} Mo (n, 2n) ^{99} Mo reaction, the γ-ray counts were periodically recorded for different irradiation, cooling and counting time, for performing the decay curve analysis of the γ ray of interest. The produced

radioisotope ^{99}Mo from the ^{100}Mo (n, 2n) ^{99}Mo reaction has a half-life of 65.976 hours. For further analysis, the primary γ-line of 739.5 keV of the sample ^{99}Mo with good branching intensity of 12.26% and the other γ-line of 181.06 keV with good branching intensity of 6.16 % were considered for determination of reaction cross section. Similarly, the primary γ-line of 355.7 keV, of monitor ^{196}Au with very good branching factor of 87.0 % was considered for the ratio measurement of reaction cross section. The calculation of reaction cross section σ_r for the characteristic γ-line of the sample ^{99}Mo is determined using the ratio method. This method of determining reaction cross-section is found to be better than the activation method, in which the neutron flux should be known accurately, thereby causing difficulties to determine. So, a monitor element, namely, gold is mounted and irradiated along with the sample such that both the foils encounter the same neutron flux. Then the ratio of their cross sections is determined using Eq. (1)

$$\frac{\sigma_S}{\sigma_M} = \frac{C_S \lambda_S A_S W_M a_M I_{\gamma_M} \varepsilon_M \left(1 - e^{-\lambda_M t_{irrM}}\right) e^{-\lambda_M t_{coolM}} \left(1 - e^{-\lambda_M t_{countM}}\right) \Gamma_{attnM}}{C_M \lambda_M A_M W_S a_S I_{\gamma_S} \varepsilon_S \left(1 - e^{-\lambda_S t_{irrS}}\right) e^{-\lambda_S t_{coolS}} \left(1 - e^{-\lambda_S t_{countS}}\right) \Gamma_{attnS}} \prod_k \frac{(C_k)_M}{(C_k)_S} \quad \ldots (1)$$

In which, the subscripts S and M denote respectively, the sample and monitor.

- $\sigma_S(E_n)$ and $\sigma_M(E_n)$are reaction cross section at the neutron energy E_n,
- C_S and C_M are the observed γ-ray counts of ^{99}Mo and ^{196}Au,
- λ_S and λ_M are decay constants,
- W_S and W_M are weights of foils,
- a_S and a_M are isotopic abundances,
- $I_{\gamma_S}$and I_{γ_M} are the γ-ray abundances,
- A_S and A_M are the average atomic masses,
- t_{irr}, t_{cool} and t_{count}denote irradiation, cooling and counting time,
- $\varepsilon_S$and ε_M are the efficiencies of HPGe detector at the energies of the characteristics γ-rays of sample ^{99}Mo and monitor ^{196}Au,determined using the regression method. In order to obtain the detector efficiencies at the characteristic 'γ-ray' energies of the sample ^{99}Mo (E_{Mo} = 181.06 and 739.5 keV) and monitor ^{196}Au (E_{Au} = 355.7 keV), the point wise efficiencies determined using EUT technique are regressed, through the fitting function given in Eq. (2):

$$\varepsilon = e^{p_0 + p_1 \ln E + p_2 (\ln E)^2 + p_3 (\ln E)^3 + p_4 (\ln E)^4 + p_5 (\ln E)^5} \quad \ldots (2)$$

in which p_0, p_1, p_2, p_3, p_4 *and*are coefficients of the fitting function. This function was selected based on the best goodness of fit parameter χ^2. The detector efficiencies at the characteristic 'γ' energies of the sample ^{99}Mo and monitor ^{196}Auare given in Table 1. The uncertainties in detector efficiency at these characteristic 'γ' energies are determined using the propagation technique followed in Refs.[14,15].

- $(C_k)_S$ and $(C_k)_M$ are the correction factors for the k th attribute, where k indicates the dead time of HPGe detector ($\frac{Clocktime}{Livetime}$) and
- Γ_{attnS} and Γ_{attnM} are the γ-ray self-attenuation factors of sample and monitor.

The information regarding the attributes such as half-life, isotopic abundances, γ-ray abundances and average atomic mass were retrieved from NuD at 2.7 database[12] and the details are given in Table 2. The self-attenuation factor (Γ_{attn}) for the irradiated foils were estimated using the relation $\Gamma_{attn} = \frac{(1-e^{-\mu l})}{\mu l}$, in which l is the thickness of each foil and μ is the mass attenuation coefficient retrieved from XMuD at ver.

Table 1 — Estimated HPGe detector efficiencies with uncertainty at the characteristic 'γ' energies of sample^{99}Mo and monitor ^{196}Au.

Radionuclide	Eγ (keV)	$\varepsilon \pm \Delta\varepsilon$	Correlation Matrix		
	739.5	0.03029±0.00087	1	0.21	0.57
^{99}Mo	181.06	0.10570 ± 0.00128	0.21	1	0.50
^{196}Au	355.7	0.06234±0.00093	0.57	0.50	1

Table 2 — Decay data adopted in the present work taken from NuDat 2.7 database [12].

Nuclide	Half-life (h)	Eγ (keV)	Iγ (%)	a	A
^{99}Mo	65.97	739.50	12.20	9.74	98.9077
	65.97	181.06	6.05	9.74	98.9077
^{196}Au	148.0	355.7	87.0	100	195.967

1.0.1[11] and its details are given in Table 3. The irradiation time of the experiment was 1.5 hours. The monitor reaction cross section at the neuron energy of 13.9 MeV, was found using the Talys-1.8 Code. Based on the details given in Table 2, Table 3 and other experimental details of sample and monitor given above, the reaction cross section of ^{100}Mo (n, 2n) ^{99}Mo reaction was determined using Eq. (1) for the data sets measured at regular time intervals corresponding to the first and second half-life of the sample of interest. Then the weighted average of the reaction cross section based on variance was computed and is given in Table 4.

3.2 Covariance analysis of ^{100} Mo (n, 2n) ^{99} Mo reaction cross-section

In our earlier paper[13] it was shown that the unscented transformation technique was good in estimating mean, however for large uncertainties the estimated variance was compromised to the order of linearity. So, the extended unscented transformation (EUT) technique was applied and established to be a good option in determining the mean and variance of propagated uncertainties for nonlinear cases of study[6]. Also, for the first time in literature, the partial uncertainty of the efficiency of HPGe detector was determined using EUT technique and then combined with the micro-correlation method given by Geraldo and Smith 1990[2] for determining the covariance matrix of HPGe detector efficiency (see Ref. [7]).

3.2.1 Estimation of partial uncertainties in reaction cross section

In this Section, the method of determining the partial uncertainties of attributes of the reaction cross section of ^{100} Mo (n, 2n) ^{99} More action using the EUT technique is presented. The reaction cross section given in Eq. (1) can be considered as a nonlinear function, as given in Eq. (3):

$$y = \sigma_S = f(\boldsymbol{x}) \quad \ldots (3)$$

In this, $\boldsymbol{x}$ represents a $n \times 1$ vector that denotes the various attributes of reaction cross section, such as counts, half-life, atomic mass, detector efficiency, etc. According to the EUT technique given in earlier studies[7], 3 sigma points are determined for each of the attribute of the $\boldsymbol{x}$ vector and then propagated through Eq. (1) to determine the propagated sigma points $\left(y^{(i)}\right)$. Then the partial uncertainty in cross section due to each of the attributes can be determined from the square root of the weighted ($W^{(i)}$) sum of the difference between $y^{(i)}$and the mean ($\bar{y}$) as reported in our earlier paper [7] and given here for continuity purpose in Eq. (4).

$$\sigma_y^2 = \sum_{i=0}^{n} W^{(i)}(y^{(i)} - \bar{y})\,(y^{(i)} - \bar{y})^T \approx \left(\frac{\partial f}{\partial x}\Big|_{\bar{x}}\right)^2 \sigma_x^{\ 2} \quad \ldots (4)$$

Firstly, the attributes whose uncertainties contribute to the uncertainty in the reaction cross section are identified. For this study, 12 attributes were considered with uncertainties namely, σ_M, C_S, C_M, λ_S, λ_M, W_S, W_M, A_S, A_M, $I_{\gamma M}$, $\varepsilon(E_\gamma)_S$, $\varepsilon(E_\gamma)_M$, and other attributes like a_S, a_M, t_{irr}, t_{cool} and t_{count} were observed without error. Also, in this particular case of sample ^{99}Mo, it is found that $I_{\gamma S}$ has been reported with no error in NuD at 2.7[12]. In addition, the γ-ray self-attenuation factor (Γ_{attn}) of sample and monitor was determined after retrieving μ which is mass attenuation coefficient from XMuD at ver. 1.0.1[11]. It can be seen that the ratio of Γ_{attn} of monitor to sample is found to be very close to '1' (0.999) as given in Table 3 and so they have not been considered as attributes in this particular study of covariance determination.

Table 3 — Self-attenuation coefficient of sample and monitor.

Energy (keV)	Density (g/cm^3)	Weight (gm)	Area (cm^2)	μ	Thickness (cm)	Γ_{attn}
739.5	10.28	0.1658	4	0.06913	0.004864	0.999832
355.7	19.32	0.3343	0.9	0.05023	0.041891	0.998949

Table 4 — Cross section σ_r at the characteristic γ-lines of ^{100}Mo (n, 2n)^{99}Mo reaction.

γ-energy (keV)	Data set collected during	Cooling time (sec)	Count time (sec)	Counts	σ_r (barns)
739.5	First half-life	192390.98	3635.9	263.0	1.36
	Second half-life	363497.00	1507.5	72.6	1.49
181.06	First half-life	192390.98	3635.9	444.7	1.40
	Second half-life	363497.00	1507.5	133.5	1.67
Weighted average cross-section of ^{100}Mo(n,2n)^{99}Mo reaction					**1.48**

Then the sigma points of the 12 attributes are propagated through the cross-section formula given in Eq. (1) to obtain 36 propagated sigma points. These propagated sigma points are then applied in Eq. (4) to determine the values of the partial uncertainties for the 12 attributes of the reaction cross section at the two characteristic γ-lines as given in Table 5, along with the details of the correlation between the attributes for the two characteristic γ-lines.

3.2.2 Micro-correlation between attributes in reaction cross section

The correlations between the values of each attribute, called micro-correlation matrices[3], are used to determine the covariance matrix of the reaction cross section. According to method suggested by Geraldo and Smith[3], if the measurement setup and procedure is the same for a particular attribute across all the energies, then that attribute's micro-correlation matrix is a matrix with rank one and all values equal to one. Otherwise the micro-correlation matrix is an identity matrix indicating there is no correlation within the attribute. From Table 5 it can be seen that in this study, 8 of the attributes are correlated, 2 attributes are partially correlated according to the correlation matrix given in Table 1 and the remaining 2 attributes are uncorrelated.

The advantage of using micro-correlation is that the effect of an attribute being common to all the measurements is accounted for, in the micro-correlation matrix and thereby in covariance matrix. This is especially significant in this study where 8 attributes out of the 12 attributes are fully correlated and their correlation is accounted in the micro-correlation matrix and so in the covariance matrix, as against the standard method of covariance matrix determination given elsewhere[16].

3.2.3 Determination of covariance matrix of reaction cross section

The covariance matrix of reaction cross section can be obtained using partial uncertainties determined through EUT technique and the micro-correlation method[3] as given in Table 4. The covariance matrix so obtained provides the complete information on uncertainties without the assumption that is usually considered in the case of partial derivatives method that errors are small and that there exists a linear approximation of measurement with reference to attributes under consideration.

It can be seen clearly from the above that the procedure adopted, is much simpler and the computational efforts in determining the covariance matrix using EUT technique for the 12 attributes of reaction cross section, is much less than that of the partial derivative method given in literature[3].

4 Results and Discussion

The ^{100}Mo (n, 2n) ^{99}Mo reaction cross section at the incident neutron energy of 13.9 MeV was determined from the yield of two characteristic γ-lines of 739.5 and 181.06 keV energy. The data was then combined using weighted average method to provide a single estimate of the reaction cross section which was found to be 1.471 ± 0.305 barns. For the first time, the covariance analysis was also performed using EUT technique for determination of partial uncertainties due to the 12 attributes and then combined with the micro-correlation method[3] to determine the covariance matrix. Table 6 gives the covariance

Table 5 — Partial uncertainties and correlation of the 12 attributes of cross section

S. No.	Attribute	E_γ= 739.5 keV	E_γ = 181.06keV	Correlation
1	C_s	12.95e-02	19.15e-02	Uncorrelated
2	$t_{1/2_s}$	13.57e-04	16.25e-04	Fully correlated
3	A_s	3.99e-09	4.46e-09	Fully correlated
4	W_s	5.20e-04	5.81e-04	Fully correlated
5	ε_s	4.18e-02	1.42e-02	Partiallycorrelated
6	C_m	2.44e-02	2.74e-02	Uncorrelated
7	$t_{1/2_m}$	14.52e-04	16.25e-04	Fully correlated
8	A_m	2.29e-08	2.56e-08	Fully correlated
9	W_m	2.58e04	2.89e-04	Fully correlated
10	I_{γ_m}	5.14e-02	5.76e-02	Fully correlated
11	ε_m	2.21e-02	2.48e-02	Partially correlated
12	σ_M	1.08e-02	1.20e-02	Fully correlated

Table 6 — Covariance matrix of reaction cross section for ^{100}Mo (n, 2n) ^{99}Mo reaction at the two characteristic γ–ray energies using EUT method

Energy (keV)	Absolute covariance matrix	
739.5	0.1589	0.0042
181.06	0.0042	0.2231

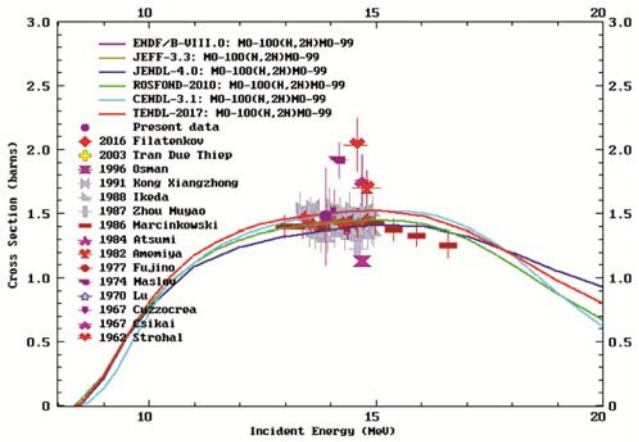

Fig. 2 — Comparison of ^{100}Mo (n, 2n)^{99}Mo reaction cross section data of our present work with the EXFOR data and the evaluated data in different evaluated libraries such as ENDF/B-VIII, JEFF-3.3, JENDL-4.0, ROSFOND-2010, CENDL-3.1, TENDL-2017.

matrix of the cross section of ^{100}Mo (n, 2n) ^{99}Mo reaction for the two characteristic γ-rays of the sample ^{99}Mo. It can be seen that the resultant variances in the reaction cross section at the characteristic γ-energy lines of 739.5 and 181.06 keV are 11.13% and 14.51%, respectively.

Figure 2 shows the comparison of the present data with the evaluated data from the various evaluated libraries such as ENDF/VIII-B[17], JEFF-3.3[18], JENDL-4.0[19], ROSFOND-2010[20], CENDL-3.1[21], TENDL-2017[22] and the literature data of the other experiments reported in EXFOR[23]. It can be observed that the present result is in good agreement with the evaluated and literature data. Also, the theoretical cross section of ^{100}Mo (n, 2n) ^{99}Mo reaction for the neutron energies in the range of (8.13-20.5) MeV was estimated using Talys-1.8 code and found to be 1.49 barns at the neutron energy of 13.9 MeV which is in close agreement with our finding.

5 Conclusions

In this paper, for the first time the extended unscented transformation (EUT) technique is applied for the determination of nuclear reaction cross section of the ^{100}Mo (n, 2n) ^{99}Mo reaction at the incident neutron energy of 13.9 MeV for the experiment performed at Purnima reactor, BARC. The EUT technique is also used for propagating uncertainties in attributes to compute the partial uncertainties and then combining it with the micro-correlation method of Geraldo and Smith[3] to determine the covariance matrix of reaction cross section of ^{100}Mo (n, 2n) ^{99}Mo reaction, for the first time. The results of the reaction cross section determined using the EUT method, is found to be in good agreement with the theoretical value of reaction cross section computed using TALYS-1.8 code[24] with default parameters. Also, the comparison of the present result with the evaluated data given in ENDF/B-VIII.0[17], JEFF-3.3[18], JENDL-4.0[19], ROSFOND-2010[20], CENDL-3.1[21], and TENDL-2017[22]libraries as well as with the other experimenters' data given in EXFOR[23], show good agreement.

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