Appendices

Appendix A: Covariance Analysis by ab initio method

Covariance analysis for the $^{100}Mo(n,2n)$ reaction cross-section data

$^{100}Mo(n,2n)^{99}Mo$ reaction cross-section were measured relative to ^{27}Al $(n,\alpha)^{24}Na$ monitor data. The experiment was performed at BARC-TIFR Pelletron accelerator facility. In the following sections detailed calculation for the generation of the covariance matrices of the measured data is provided to present a better insight into the procedure of the analysis. The procedure follows two steps, first is to generate covariance and correlation matrix of the detector efficiencies for the γ-lines used in the calculation. Later in step 2, these correlation and error are used to find the resultant uncertainties in the measured cross-sections using the ratio method.

Step 1: To find the covariance matrix for detector efficiency and fitting parameters following the liner fit method

The detector efficiency at different γ-energies of the ^{152}Eu source used for the present work are given in the following Table 1.

Now using the definitions given in equations 3.41-3.46, the partial derivatives can be obtained as,

$$\frac{\partial \epsilon}{\partial C} = \frac{1}{N_0 I_\gamma e^{-\lambda T} \triangle t} \tag{1}$$

Table 1: Data related to the ^{152}Eu source to obtain the detector efficiency

Energy (keV)	Counts	I_γ	N_0	$T_{1/2}$ (Years)	Efficiency
121.8	439862 ± 3518	0.2853 ± 0.0016	7767.73 ± 88	13.517 ± 0.009	0.101983847
244.7	70828 ± 894	0.0755 ± 0.0004	7767.73 ± 88	13.517 ± 0.009	0.062054709
443.9	18536 ± 296	0.0282 ± 0.0014	7767.73 ± 88	13.517 ± 0.009	0.043059776
964	48429 ± 581	0.1451 ± 0.0007	7767.73 ± 88	13.517 ± 0.009	0.02207775
1112	41140 ± 534	0.1367 ± 0.0008	7767.73 ± 88	13.517 ± 0.009	0.019907306
1408	52162 ± 573	0.2087 ± 0.0009	7767.73 ± 88	13.517 ± 0.009	0.01653288

multiply and divide the above equation by C,

$$\frac{\partial \epsilon}{\partial C} = \frac{1}{N_0 I_\gamma e^{-\lambda T} \triangle t} \times \frac{c}{c} \tag{2}$$

the first term in the above equation can be written as ϵ and the equation can be rearranged for the infinitesimal variance as,

$$\triangle \epsilon = \frac{\epsilon}{C} \triangle C \tag{3}$$

where, $\triangle\epsilon$ and $\triangle C$ are the uncertainties in the efficiency and the counting statistics of the γ-line under consideration. Now $\triangle\epsilon$ can be calculated as,

$$\triangle \epsilon = \frac{\epsilon}{C} \triangle C = \frac{0.101983847}{439862} \times 3518 = 0.000815663 \tag{4}$$

similarly, the partial uncertainties due to I_γ, N_0, and $T_{1/2}$, by omitting the negative sign can be calculated and are given in Table 5.9. The total uncertainty in the efficiency now can be calculated as the quadratic sum of all the individual partial uncertainties from the four attributes, by following the equation 3.45 and also given in Table 2.

Now the diagonal matrix (e_{ir}) for partial uncertainty from each attribute (r) can be written as shown in Table 3.

Similarly, the matrices can be formed for other attributes (r=2,3,4), which will be used to form the covariance matrix for the detector efficiency by using the following sandwich formula,

$$(V_\varepsilon)_{ij} = \sum_r e_{ir} S_{ijr} e_{jr} \tag{5}$$

where, V_ε is the covariance matrix for detector efficiency, e_{ir} is the diagonal matrix for attribute r and the S_{ijr} are the micro correlation matrices defined

Table 2: Partial uncertainties in the efficiency

Energy	Partial uncertainty due to attributes				Total uncertainty
(keV)	$r = 1(C)$	$r = 2(I_\gamma)$	$r = 3(N_0)$	$r = 4(T_{1/2})$	$(\sigma_{\varepsilon_{ii}})$
121.8	0.000815663	0.000571939	0.001155367	4.8321E-06	0.001525554
244.7	0.000783262	0.000328767	0.000703013	2.94022E-06	0.001102644
443.9	0.000687618	0.002137719	0.000487821	2.04022E-06	0.002297964
964	0.000264866	0.000106509	0.000250117	1.04607E-06	0.000379549
1112	0.000258398	0.000116502	0.000225528	9.43229E-07	0.000362224
1408	0.000181614	7.12966E-05	0.0001873	7.83345E-07	0.00027046

Table 3: Diagonal matrix ($\times 10^{-3}$) for partial uncertainty from the attribute C (r=1) using the values given in the Table 2.

$$\begin{vmatrix} 0.815663 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0.783262 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0.687618 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0.264866 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0.258398 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0.181614 \end{vmatrix}$$

for uncorrelated, correlated and partially correlated cases as,

$$\begin{pmatrix} 1 & 0 & \cdots & 0 \\ 0 & 1 & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & 1 \end{pmatrix} \quad \begin{pmatrix} 1 & 1 & \cdots & 1 \\ 1 & 1 & \cdots & 1 \\ \vdots & \vdots & \ddots & \vdots \\ 1 & 1 & \cdots & 1 \end{pmatrix} \quad and \quad \begin{pmatrix} 1 & q & \cdots & q \\ 1 & q & \cdots & q \\ \vdots & \vdots & \ddots & \vdots \\ q & q & \cdots & 1 \end{pmatrix}; 0 < q < 1$$

The equation 5 can now be used to find the covariance matrix for r=1 as,

$$= \begin{pmatrix} 0.815663 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0.783262 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0.687618 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0.264866 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0.258398 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0.181614 \end{pmatrix} \times \begin{pmatrix} 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 \end{pmatrix}$$

$$\times \begin{pmatrix} 0.815663 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0.783262 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0.687618 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0.264866 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0.258398 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0.181614 \end{pmatrix} \times 10^{-6}$$

$$V_{r=1} = \begin{pmatrix} 6.653E-07 & 0 & 0 & 0 & 0 & 0 \\ 0 & 6.135E-07 & 0 & 0 & 0 & 0 \\ 0 & 0 & 4.728E-07 & 0 & 0 & 0 \\ 0 & 0 & 0 & 7.015E-08 & 0 & 0 \\ 0 & 0 & 0 & 0 & 6.676E-08 & 0 \\ 0 & 0 & 0 & 0 & 0 & 3.298E-08 \end{pmatrix}$$

Similarly the covariance matrices for r=2, 3 and 4 can be calculated and were found to be,

$$V_{r=2} = \begin{pmatrix} 3.271E-07 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1.080E-07 & 0 & 0 & 0 & 0 \\ 0 & 0 & 4.569E-06 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1.134E-08 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1.357E-08 & 0 \\ 0 & 0 & 0 & 0 & 0 & 5.083E-09 \end{pmatrix}$$

The covariance matrix for r=3 and 4 will be calculated using the correlated case for S_{ijr} matrix and the result are as follows,

$$V_{r=3} \begin{pmatrix} 1.334E-06 & 8.122E-07 & 5.636E-07 & 2.889E-07 & 2.605E-07 & 2.164E-07 \\ 8.122E-07 & 4.942E-07 & 3.429E-07 & 1.758E-07 & 1.585E-07 & 1.316E-07 \\ 5.636E-07 & 3.429E-07 & 2.379E-07 & 1.220E-07 & 1.100E-07 & 9.136E-08 \\ 2.889E-07 & 1.758E-07 & 1.220E-07 & 6.255E-08 & 5.640E-08 & 4.684E-08 \\ 2.605E-07 & 1.585E-07 & 1.100E-07 & 5.640E-08 & 5.0863E-08 & 4.224E-08 \\ 2.164E-07 & 1.316E-07 & 9.136E-08 & 4.684E-08 & 4.2241E-08 & 3.508E-08 \end{pmatrix}$$

$$V_{r=4} \begin{pmatrix} 2.334E-11 & 1.420E-11 & 9.858E-12 & 5.054E-12 & 4.557E-12 & 3.785E-12 \\ 1.420E-11 & 8.644E-12 & 5.998E-12 & 3.075E-12 & 2.773E-12 & 2.303E-12 \\ 9.858E-12 & 5.998E-12 & 4.162E-12 & 2.134E-12 & 1.924E-12 & 1.598E-12 \\ 5.054E-12 & 3.075E-12 & 2.134E-12 & 1.094E-12 & 9.866E-13 & 8.194E-13 \\ 4.557E-12 & 2.773E-12 & 1.924E-12 & 9.866E-13 & 8.896E-13 & 7.388E-13 \\ 3.785E-12 & 2.303E-12 & 1.598E-12 & 8.194E-13 & 7.388E-13 & 6.136E-13 \end{pmatrix}$$

The final covariance matrix is calculated by adding the four matrices $V_{r=1,2,3,4}$ and is given in Table 4.

Table 4: Co-variance matrix $(V_\varepsilon \times 100)$ for the detector efficiency

$$(V_\epsilon)_{ij} = \begin{vmatrix} 0.0030938 & & & & & \\ 0.0008305 & 0.0015126 & & & & \\ 0.0005763 & 0.0003507 & 0.0009057 & & & \\ 0.0002955 & 0.0001798 & 0.0001248 & 0.0001840 & & \\ 0.0002664 & 0.0001621 & 0.0001125 & 0.0000577 & 0.0001593 & \\ 0.0001346 & 0.0001346 & 0.000934 & 0.0000479 & 0.0000432 & 0.0000968 \end{vmatrix}$$

The uncertainties in the efficiency of the γ-lines used, can be calculated by taking the square root of the diagonal elements of the covariance matrix given in Table 4. The correlation factors among the γ-lines of ^{152}Eu can be calculated by using the equation,

$$\mathbf{Corr}(E_{\gamma_i}, E_{\gamma_j}) = \frac{\mathbf{Cov}(E_{\gamma_i}, E_{\gamma_j})}{\sqrt{\mathbf{Var}(E_{\gamma_i})}\sqrt{\mathbf{Var}(E_{\gamma_j})}} \tag{6}$$

where, $E_{\gamma_{i,j}}$ are the i, j^{th} element of the matrix $(V_\epsilon)_{ij}$.

The next step is to calculate the matrix $(V_z)_{ij}$ 3.52, whose inverse will be used to calculate the fitting parameters and their covariance matrix by using the equations 3.49, 3.50 and 3.51. To calculate matrix $(V_z)_{ij}$ with the equation 3.52, first we need to form a diagonal matrix of detector efficiencies as,

$$1/\epsilon_i = \begin{pmatrix} 9.69686 & 0 & 0 & 0 & 0 & 0 \\ 0 & 15.93632 & 0 & 0 & 0 & 0 \\ 0 & 0 & 22.96630 & 0 & 0 & 0 \\ 0 & 0 & 0 & 44.79279 & 0 & 0 \\ 0 & 0 & 0 & 0 & 49.67643 & 0 \\ 0 & 0 & 0 & 0 & 0 & 59.81559 \end{pmatrix}$$

Now the matrix $(V_z)_{ij}$ can be calculated as,

$$(V_z)_{ij} = \begin{pmatrix} 9.69686 & 0 & 0 & 0 & 0 & 0 \\ 0 & 15.93632 & 0 & 0 & 0 & 0 \\ 0 & 0 & 22.96630 & 0 & 0 & 0 \\ 0 & 0 & 0 & 44.79279 & 0 & 0 \\ 0 & 0 & 0 & 0 & 49.67643 & 0 \\ 0 & 0 & 0 & 0 & 0 & 59.81559 \end{pmatrix} \times$$

$$\begin{pmatrix} 3.09377E-06 & 8.30548E-07 & 5.76318E-07 & 2.95492E-07 & 2.66442E-07 & 2.21278E-07 \\ 8.30548E-07 & 1.51262E-06 & 3.50675E-07 & 1.79799E-07 & 1.62124E-07 & 1.34642E-07 \\ 5.76318E-07 & 3.50675E-07 & 9.05744E-07 & 1.24763E-07 & 1.12498E-07 & 9.34284E-08 \\ 2.95492E-07 & 1.79799E-07 & 1.24763E-07 & 1.83963E-07 & 5.76801E-08 & 4.79029E-08 \\ 2.66442E-07 & 1.62124E-07 & 1.12498E-07 & 4.76801E-08 & 1.59302E-07 & 4.31936E-08 \\ 2.21278E-07 & 1.34642E-07 & 9.34284E-08 & 4.79029E-08 & 4.31936E-08 & 9.68049E-08 \end{pmatrix} \times$$

$$\begin{pmatrix} 9.69686 & 0 & 0 & 0 & 0 & 0 \\ 0 & 15.93632 & 0 & 0 & 0 & 0 \\ 0 & 0 & 22.96630 & 0 & 0 & 0 \\ 0 & 0 & 0 & 44.79279 & 0 & 0 \\ 0 & 0 & 0 & 0 & 49.67643 & 0 \\ 0 & 0 & 0 & 0 & 0 & 59.81559 \end{pmatrix}$$

$$= \begin{pmatrix} 0.290905 & 0.128347 & 0.128347 & 0.128347 & 0.128347 & 0.128347 \\ 0.128347 & 0.384155 & 0.128347 & 0.128347 & 0.128347 & 0.128347 \\ 0.128347 & 0.128347 & 0.477736 & 0.128347 & 0.128347 & 0.128347 \\ 0.128347 & 0.128347 & 0.128347 & 0.369103 & 0.128347 & 0.128347 \\ 0.128347 & 0.128347 & 0.128347 & 0.128347 & 0.393117 & 0.128347 \\ 0.128347 & 0.128347 & 0.128347 & 0.128347 & 0.128347 & 0.346359 \end{pmatrix}$$

Now the inverse of the matrix $(V_z)_{ij}$ can be easily given by,

$$(V_z)^{-1}_{ij} = \begin{pmatrix} 5012.8877 & & & & & \\ -723.6418 & 3449.3234 & & & & \\ -529.8212 & -336.6848 & 2615.6325 & & & \\ -768.8848 & -488.6022 & -357.7347 & 3634.4313 & & \\ -699.1473 & -444.2863 & -325.2883 & -472.0636 & 3347.6061 & \\ -849.0981 & -539.5753 & -395.0551 & -573.3102 & -521.3112 & 3953.7803 \end{pmatrix}$$

Now to fit the efficiencies an interpolation model is used as given in equation 3.49 with the simple liner solution, Z=AP, where Z ($z_i = ln\varepsilon_i$) is a column matrix, A ($A_{im} = (lnE_i)^{m-1}$) is the design matrix with elements, and P is the matrix having parameters p_m, which p_m can be estimated using the least square method. Now we form Z and A matrices for the order m=5[1] by the given definitions as,

$$Z = ln(\varepsilon_i) = \begin{pmatrix} -2.28294 \\ -2.77973 \\ -3.14516 \\ -3.81318 \\ -3.91666 \\ -4.10240 \end{pmatrix};$$

$$A = (lnE_i)^{m-1} = \begin{pmatrix} 1 & -2.105375 & 4.432604 & -9.332292 & 19.647974 \\ 1 & -1.407722 & 1.981682 & -2.789658 & 3.927064 \\ 1 & -0.812156 & 0.659597 & -0.535696 & 0.435069 \\ 1 & -0.036664 & 0.001344 & -0.000049 & 0.000002 \\ 1 & 0.106160 & 0.011270 & 0.001196 & 0.000127 \\ 1 & 0.342170 & 0.117080 & 0.040061 & 0.013708 \end{pmatrix}$$

Now, the covariance matrix for parameters P, can be calculated by the following equation using the matrix A, Transpose of A and V_z^{-1} as,

$$= V_{\hat{p}} = (A' V_z^{-1} A)^{-1}$$

$$= \begin{pmatrix} 0.268541 & -0.10068 & -0.763573 & -0.673544 & -0.165564 \\ -0.10068 & 1.224617 & 0.301311 & -1.1193217 & -0.499049 \\ -0.763573 & 0.301311 & 8.935453 & 9.790935 & 2.70637 \\ -0.673544 & -1.193217 & 9.790935 & 12.987068 & 3.869357 \\ -0.165564 & -0.499049 & 2.70637 & 3.869357 & 1.183991 \end{pmatrix} \times 10^{-3}$$

[1]The order m is achieved by checking the value condition for χ^2 with all the possible values of m and the best among all is adopted

Now, the fitting parameters $\hat{P}$ can be calculated by the following equation,

$$\hat{P} = V_{\hat{P}}(A'V_z^{-1}Z) = \begin{pmatrix} -3.83706 \\ -0.86929 \\ 0.16981 \\ 0.32085 \\ 0.10003 \end{pmatrix}$$

Now the value of χ^2_m can be tested as,

$$\frac{\chi^2_m}{n-m} = \frac{(Z - AP)'V_z^{-1}(Z - AP)}{n-m} = \frac{0.72}{6-5} = 0.72$$

where, n is the number of γ-lines used for the calculation and m is order of the fitting. Since, the condition $\chi^2_m/(n-m) \approx 1$ fulfills, therefore, the parameters $\hat{P}$ can be used to find the efficiencies of the γ-rays of the sample and monitor reactions.

Step 2: To find the efficiencies, their covariance & correlation matrix for the sample/monitor γ-lines by using the fitting parameters

Now we follow the similar procedure again in order to find the covariance and correlation matrices for the sample and monitor reaction γ-lines. To do so, first we find the efficiencies for the desired γ-lines by using the fitting parameters into the equation 3.49,

$$ln\varepsilon_i = \sum_m p_m(lnE_i)^{m-1} \tag{7}$$

which can be expended for the 0.1405 MeV γ-line of ^{99}Mo as,

$$ln\varepsilon_1 = p_1(lnE_1)^0 + p_2(lnE_1)^1 + p_3(lnE_1)^2 + p_4(lnE_1)^3 + p_5(lnE_1)^4 \tag{8}$$

$$ln\varepsilon_1 = -3.83706 - 0.86929(-1.966112856) + 0.16981(3.865599764) +$$
$$0.32085(-7.600205394) + 0.10003(14.94286154)$$
$$ln\varepsilon_1 = -2.415270015$$
$$\varepsilon_1 = exp(-2.415270015) = 0.089343$$

similarly, the efficiency for the 1.368 MeV γ-line of the $^{27}Al(n,\alpha)^{24}Na$ monitor reaction can be find to be,

$$\varepsilon_2 = exp(-4.082272134) = 0.016875$$

now the covariance/correlation matrix for the efficiencies ϵ_1 and ϵ_2 can be calculated by following the method of liner fitting and reconstructing the matrices Z, A and using $V_{\hat{p}}$. The matrices Z and A for ϵ_1 and ϵ_2 can now be written as,

$$Z_{\epsilon_1} = \begin{pmatrix} -2.415270 \\ -4.082272 \end{pmatrix};$$

$$A = \begin{pmatrix} 1 & -1.966112856 & 3.865599764 & -7.600205394 & 14.94286154 \\ 1 & 0.313349819 & 0.098188109 & 0.030767226 & 0.009640905 \end{pmatrix}$$

$$A' = \begin{pmatrix} 1 & 1 \\ -1.966112856 & 0.313349819 \\ 3.865599764 & 0.098188109 \\ -7.600205394 & 0.030767226 \\ 14.94286154 & 0.009640905 \end{pmatrix}$$

Now, the covariance matrix ($V_{z_{\epsilon_i}}$) for the column matrix Z is computed as,

$$V_{z_{\epsilon_i}} = A' V_{\hat{p}} A \tag{9}$$

$$V_{z_{\epsilon_i}} = \begin{pmatrix} 0.000412906 & 0.000120279 \\ 0.000120279 & 0.000288745 \end{pmatrix}$$

and the covariance matrix for efficiencies ϵ_1 and ϵ_2 is now calculated by the equation,

$$V_{\epsilon_{ij}} = \epsilon_i V_{z_{\epsilon_{ij}}} \epsilon_j \tag{10}$$

where, $\epsilon_{i,j}$ is the diagonal matrix,

$$\begin{pmatrix} 0.089343 & 0 \\ 0 & 0.016875 \end{pmatrix}$$

and the final covariance ($V_{\epsilon_{ij}}$) and correlation ($Corr[V_{\epsilon_{ij}}]$) matrix 6 for ϵ_1 and ϵ_2 is,

$$V_{\epsilon_{ij}} = \begin{pmatrix} 3.29591E-06 & \\ 1.81336E-07 & 8.22208E-08 \end{pmatrix}; Corr[V_{\epsilon_{ij}}] = \begin{pmatrix} 1 & \\ 0.3483 & 1 \end{pmatrix}$$

The uncertainties in the efficiencies ϵ_1 and ϵ_2 can be calculated by taking the square root of the diagonal elements of the covariance matrix ($V_{\epsilon_{ij}}$). The efficiencies of the γ-lines with their uncertainties and correlation coefficients are given in Table 5.

The calculated uncertainties and correlation coefficient in Table 5 will be used in the calculation for the uncertainty in the measured cross-section in the following steps. It can be noticed from the Table 5 that the uncertainty in the efficiencies are $\approx$ 2% and 1.6%, which can be reduced further by introducing a relative parameter $\eta_{m,s} = \epsilon_m/\epsilon_s$, with the partial uncertainties

Table 5: Measured efficiencies with correlation matrix for the sample and the monitor reaction

E_γ (keV)	Efficiency	Correlation Matrix	
140.5	0.089343 ± 0.001815	1	
1368.68	0.016874 ± 0.000287	0.348	1

given as,

$$\frac{\triangle\eta_{m,s}}{\eta_{m,s}} = Var(\epsilon_m) + Var(\epsilon_s) - 2cov(\epsilon_m, \epsilon_s)$$

where, 'm' and 's' are used for monitor and sample reactions. Therefore, using the definition above, calculations can be performed as,

$$\eta_{m,s} = \epsilon_m/\epsilon_s = 0.089343/0.016875 = 0.188874$$

$$\triangle\eta_{m,s} = \sqrt{3.29591E-06+8.22208E-08-2\times 1.81336E-07}$$

$$= 0.001736506$$

$$\triangle\eta_{m,s}/\eta_{m,s} = 0.001736506/0.188874 = 0.00919$$

The value 0.00919 can now be used for the ratio ϵ_m/ϵ_s present in the equation 3.57 for the relative measurement of the uncertainties for measured cross-sections[2].

Step 3: Covariance analysis for the measured cross-sections

to calculate the covariance matrix and correlations among the different measured cross-sections we follow the procedure described in section 3.2.3. In the relative measurement technique, the sample reaction cross-section are measured relative to a monitor reaction. The equation for NAA takes the form,

$$<\sigma_s> = <\sigma_m> \frac{C_s N_{0_m} \varepsilon_m I_{\gamma_m} f_{\lambda_m}}{C_m N_{0_s} \varepsilon_s I_{\gamma_s} f_{\lambda_s}} \tag{11}$$

with the time factor f defined as,

$$f = (1-e^{-\lambda t_i})(e^{-\lambda t_c})(1-e^{-\lambda T_L})/\lambda \tag{12}$$

Equation 11 is differentiated with respect to different attribute in order to calculate the partial uncertainties present in each measured/used quantity.

[2]The details for relative measurement are provided in section 3.2.3

The uncertainties present in each attribute of the equation 11 is listed in Table 6.

Table 6: Fractional uncertainties in various parameters used to obtain $^{100}Mo(n,2n)^{99}Mo$ cross-sections

E_n (MeV)	Partial uncertainty (%)												
	C_s	C_m	$I_{\gamma s}$	$I_{\gamma m}$	$\eta_{m,s}$	f_{λ_s}	f_{λ_m}	M_s	M_m	a_s	A_s	A_m	σ_W
10.95 ± 0.45	8.687	8.152	0.257	0.002	0.919	0.035	0.078	1.996	1.884	3.157	0.010	2.59E-06	0.973
13.97 ± 0.68	10.591	6.949	0.257	0.002	0.919	0.036	0.078	1.970	2.026	3.157	0.010	2.59E-06	0.462
16.99 ± 0.53	7.213	10.161	0.257	0.002	0.919	0.036	0.075	1.981	1.823	3.157	0.010	2.59E-06	0.826
20.00 ± 0.58	8.025	8.908	0.257	0.002	0.919	0.036	0.079	1.931	1.879	3.157	0.010	2.59E-06	1.301
Corr	0	0	1	1	1	1	1	0	0	1	1	1	0

The correlation assigned at the bottom of the Table 6 provides the essential knowledge to introduce the micro correlation matrix in the sandwich formula 5. Similar definitions applies to the step 3 as used in step 1 for generating covariance matrix. Now covariance matrices can be calculated for each attribute (let's take C_s) as,

$$V_{C_s} = \begin{pmatrix} 0.08687 & 0 & 0 & 0 \\ 0 & 0.10591 & 0 & 0 \\ 0 & 0 & 0.07213 & 0 \\ 0 & 0 & 0 & 0.08025 \end{pmatrix} \times \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}$$

$$\times \begin{pmatrix} 0.08687 & 0 & 0 & 0 \\ 0 & 0.10591 & 0 & 0 \\ 0 & 0 & 0.07213 & 0 \\ 0 & 0 & 0 & 0.08025 \end{pmatrix}$$

$$= \begin{pmatrix} 0.007547163 & 0 & 0 & 0 \\ 0 & 0.011217521 & 0 & 0 \\ 0 & 0 & 0.00520337 & 0 \\ 0 & 0 & 0 & 0.00644046 \end{pmatrix}$$

Similarly for $\eta_{m,s}$, the micro correlation matrix will take the form for the full correlation case, as,

$$V_{\eta_{m,s}} = \begin{pmatrix} 0.00919 & 0 & 0 & 0 \\ 0 & 0.00919 & 0 & 0 \\ 0 & 0 & 0.00919 & 0 \\ 0 & 0 & 0 & 0.00919 \end{pmatrix} \times \begin{pmatrix} 1 & 1 & 1 & 1 \\ 1 & 1 & 1 & 1 \\ 1 & 1 & 1 & 1 \\ 1 & 1 & 1 & 1 \end{pmatrix}$$

$$\times \begin{pmatrix} 0.00919 & 0 & 0 & 0 \\ 0 & 0.00919 & 0 & 0 \\ 0 & 0 & 0.00919 & 0 \\ 0 & 0 & 0 & 0.00919 \end{pmatrix} = 8.45297E-05 \begin{pmatrix} 1 & 1 & 1 & 1 \\ 1 & 1 & 1 & 1 \\ 1 & 1 & 1 & 1 \\ 1 & 1 & 1 & 1 \end{pmatrix}$$

Similarly, the covariance matrices for other attributes can be calculated and add up to get the final covariance $[V_{cs_{ij}}]$ and the corresponding correlation matrix, as given in Table 7.

Table 7: Covariance matrix and corresponding correlation coefficients for the measured $^{100}Mo(n,2n)^{99}Mo$ cross-sections

E_n (MeV)	Covariance matrix ($V_{cs_{ij}}$)				Correlation matrix			
10.95 ± 0.45	0.01611726				1			
13.97 ± 0.68	0.00108844	0.01795559			0.064	1		
16.99 ± 0.53	0.00108842	0.00108842	0.01740994		0.065	0.061	1	
20.00 ± 0.58	0.00108845	0.00108846	0.00108843	0.01635372	0.067	0.063	0.064	1

The uncertainty at a given incident neutron energy can now be calculated by taking the square root fo the diagonal element of the $V_{cs_{ij}}$ matrix and are given in Table 8 and the results for the $^{100}Mo(n,2n)^{99}Mo$ reaction cross-section are listed in Table 9,

Table 8: Uncertainty (%) present in the measured $^{100}Mo(n,2n)^{99}Mo$ cross-sections at respective neutron energy

E_n (MeV)	Diagonal element ($V_{cs_{ii}}$)	Uncertainty (%)
10.95 ± 0.45	0.01611726	12.69
13.97 ± 0.68	0.01795559	13.39
16.99 ± 0.53	0.01740994	13.19
20.00 ± 0.58	0.01635372	12.78

Table 9: $^{100}Mo(n,2n)^{99}Mo$ reaction cross-sections compare with ENDF-B/VII.1, JENDL-4.0 and TALYS-1.9 data

Neutron	Flux	Cross-Section (mb)			
Energy (MeV)	($n\ cm^{-2}s^{-1}$)	Corrected	ENDF	JENDL0	TALYS
10.95 ± 0.45	7.27×10^5	1243.89 ± 156.73	1120.28	1092.62	1289.20
13.97 ± 0.68	1.48×10^6	1422.53 ± 190.61	1478.30	1375.23	1533.38
16.99 ± 0.53	2.68×10^6	1216.82 ± 159.40	1393.71	1326.21	1286.85
20.00 ± 0.58	3.09×10^6	774.96 ± 98.41	612.44	925.11	671.88

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Covariance analysis for the $^{58}Ni(n,x)$ reaction cross-section data

The experimental measurements for the $^{58}Ni(n,x)$ reaction cross-sections at desired energies were measured in two parts as mentioned in Tables 5.1 and 5.2. The covariance analysis for this case has been carried out in these steps keeping the correlations of different sample monitor combinations under consideration. In set 1, $^{58}Ni(n,p)^{58}Co$ reaction cross-sections were measured at 2.97 and 3.37 MeV incident neutron energies using FOTIA relative to $^{197}Au(n,\gamma)^{198}Au$ monitor reaction data.The other measurement, for both $^{58}Ni(n,p)^{58}Co$ and $^{58}Ni(n,2n)^{57}Ni$ reactions relative to the $^{115}In(n,n')^{115m}In$ and $^{27}Al(n,\alpha)^{24}Na$ monitor reactions, respectively, was carried out at 5.99, 13.97, and 16.99 MeV incident energies using Pelletron. Therefore, it is clear that the correlation may exist, separately, among the data measured at FOTIA and Pelletron. The matrices used in the following section will be assigned with subscript 1 for FOTIA and 2 for Pelletron experiment. Since, a complete description with step by step calculation for each matrix is shown previously for the $^{100}Mo(n,2n)$ reaction data, therefore, in this section the calculations will be more straight forward to avoid the repetition.

The covariance matrices for the detector efficiencies are calculated using the technique described in the previous calculations and are given in Tables 10 and 11. The covariance matrix for Pelletron experiment set is similar to the Mo(n, 2n) reaction case as both the measurements were carried out using similar calibration source.

Table 10: Co-variance matrix $(V_\varepsilon \times 10^6)$ for the efficiencies of the detector used at FOTIA.

0.009328					
0.001132	0.004273				
0.000976	0.000602	0.002746			
0.000588	0.000363	0.000313	0.001181		
0.000433	0.000267	0.000023	0.000139	0.000661	
0.000378	0.000233	0.000201	0.000121	0.000089	0.000485

Table 11: Co-variance matrix ($V_\varepsilon \times 100$) for the efficiencies of the detector used at Pelletron.

$$
\begin{vmatrix}
0.0030938 & & & & & \\
0.0008305 & 0.0015126 & & & & \\
0.0005763 & 0.0003507 & 0.0009057 & & & \\
0.0002955 & 0.0001798 & 0.0001248 & 0.0001840 & & \\
0.0002664 & 0.0001621 & 0.0001125 & 0.0000577 & 0.0001593 & \\
0.0001346 & 0.0001346 & 0.000934 & 0.0000479 & 0.0000432 & 0.0000968
\end{vmatrix}
$$

The $(V_Z)^{-1}$ matrices for both the covariance matrices given above can be calculated by following the equation 3.52, with the help of diagonalized $1/\epsilon_i$ matrices as,

$$
[(V_z)^{-1}_{ij}]_1 = \begin{pmatrix}
118.4834 & 0 & 0 & 0 & 0 & 0 \\
0 & 191.9385 & 0 & 0 & 0 & 0 \\
0 & 0 & 222.7171 & 0 & 0 & 0 \\
0 & 0 & 0 & 369.8361 & 0 & 0 \\
0 & 0 & 0 & 0 & 502.2912 & 0 \\
0 & 0 & 0 & 0 & 0 & 574.7126
\end{pmatrix} \times
$$

$$
\begin{pmatrix}
0.009328 & & & & & \\
0.001132 & 0.004273 & & & & \\
0.000976 & 0.000602 & 0.002746 & & & \\
0.000588 & 0.000363 & 0.000313 & 0.001181 & & \\
0.000433 & 0.000267 & 0.000023 & 0.000139 & 0.000661 & \\
0.000378 & 0.000233 & 0.000201 & 0.000121 & 0.000089 & 0.000485
\end{pmatrix} \times
$$

$$
\begin{pmatrix}
118.4834 & 0 & 0 & 0 & 0 & 0 \\
0 & 191.9385 & 0 & 0 & 0 & 0 \\
0 & 0 & 222.7171 & 0 & 0 & 0 \\
0 & 0 & 0 & 369.8361 & 0 & 0 \\
0 & 0 & 0 & 0 & 502.2912 & 0 \\
0 & 0 & 0 & 0 & 0 & 574.7126
\end{pmatrix}
$$

$$
[(V_z)^{-1}_{ij}]_1 = \begin{pmatrix}
8465.8173 & & & & & \\
-830.8127 & 6930.2913 & & & & \\
-990.4599 & -791.2620 & 8109.9541 & & & \\
-806.0586 & -643.9469 & -767.6863 & 6742.9896 & & \\
-775.7072 & -619.6996 & -738.7798 & -601.2357 & 6511.7275 & \\
-814.7673 & -650.9041 & -775.9804 & -631.5104 & -607.73149 & 6809.018326
\end{pmatrix}
$$

$$
[(V_z)^{-1}_{ij}]_2 = \begin{pmatrix}
5012.8877 & & & & & \\
-723.6418 & 3449.3234 & & & & \\
-529.8212 & -336.6848 & 2615.6325 & & & \\
-768.8848 & -488.6022 & -357.7347 & 3634.4313 & & \\
-699.1473 & -444.2863 & -325.2883 & -472.0636 & 3347.6061 & \\
-849.0981 & -539.5753 & -395.0551 & -573.3102 & -521.3112 & 3953.7803
\end{pmatrix}
$$

Following the method described previously, the parameters $[\hat{P}]_1$ and $[\hat{P}]_2$ were found to be,

$$[\hat{P}]_1 = \begin{pmatrix} -6.12540 \\ -0.79281 \\ 0.17388 \\ 0.40937 \\ 0.13875 \end{pmatrix} \text{with } \frac{\chi^2}{(n-m)} = 2.35$$

$$[\hat{P}]_2 = \begin{pmatrix} -3.8370 \\ -0.8694 \\ 0.1698 \\ 0.3208 \\ 0.1 \end{pmatrix} \text{with } \frac{\chi^2}{(n-m)} = 0.72$$

using the covariance matrices $V_{\hat{P}}$ as,

$$V_{\hat{P}_1} = \begin{pmatrix} 0.118096 & -0.010794 & -0.552945 & -0.581947 & -0.157487 \\ -0.010794 & 0.634335 & -0.051975 & -0.864767 & -0.330606 \\ -0.552944 & -0.051975 & 6.182727 & 7.234424 & 2.0635353 \\ -0.581947 & -0.864767 & 7.234424 & 9.638681 & 2.8835398 \\ -0.157487 & -0.330606 & 2.063535 & 2.883539 & 0.8772604 \end{pmatrix} \times 10^{-3}$$

$$V_{\hat{P}_2} = \begin{pmatrix} 0.268541 & -0.10068 & -0.763573 & -0.673544 & -0.165564 \\ -0.10068 & 1.224617 & 0.301311 & -1.1193217 & -0.499049 \\ -0.763573 & 0.301311 & 8.935453 & 9.790935 & 2.70637 \\ -0.673544 & -1.193217 & 9.790935 & 12.987068 & 3.869357 \\ -0.165564 & -0.499049 & 2.70637 & 3.869357 & 1.183991 \end{pmatrix} \times 10^{-3}$$

Using the set of fitting parameters above, the γ-ray efficiencies for both the $^{58}Ni(n,p)$ and $^{58}Ni(n,2n)$ reactions were measured with the relative uncertainties and correlations. The details for set 1 and 2 are provided in Tables 12, 13, and 14, respectively.

Table 12: Measured efficiencies with correlation matrix for the sample and the monitor reaction measured for set 1

E_γ (keV)	Efficiency	Covariance Matrix		Correlation Matrix	
411.80	0.004148 ± 0.000055	3.10329E-09		1	
810.77	0.002592 ± 0.000028	6.90224E-10	7.87024E-10	0.441	1

Table 13: Measured efficiencies with covariance matrix for the sample and the monitor reaction measured for set 2

E_γ (keV)	Efficiency	Covariance Matrix ($\times 10^7$)			
336.241	0.051708 ± 0.000963	9.28271			
810.77	0.025991 ± 0.000457	2.42891	2.09595		
1368.68	0.016869 ± 0.000287	1.14452	0.451411	0.823715	
1377.63	0.016797 ± 0.000290	1.1674	0.429814	0.833721	0.844832

Table 14: Measured efficiencies with correlation matrix for the sample and the monitor reaction measured for set 2

E_γ (keV)	Efficiency	Correlation Matrix			
336.241	0.051708 ± 0.000963	1			
810.77	0.025991 ± 0.000457	0.551	1		
1368.68	0.016869 ± 0.000287	0.413	0.343	1	
1377.63	0.016797 ± 0.000290	0.416	0.323	0.99	1

Now with the generated covariances/correlations of the γ-lines of sample and monitor, we can proceed for the covariance analysis of the measured data. The measured cross-sections are again treated separately as per the experiment setup. Since the two reactions and the corresponding monitors in two sets of experiment have different thresholds and only minor correlation exists dew to the detector efficiencies, therefore, the measured (n, p) and (n, 2n) reaction data were assumed to be independent. On behalf of the above discussion, the covariance analysis was performed in the following combinations,

- $^{58}Ni(n,p)^{58}Co$ sample data with $^{197}Au(n,\gamma)$ monitor reaction at 2.97 and 3.37 MeV (experiment performed at FOTIA, BARC)
- $^{58}Ni(n,p)^{58}Co$ sample data with $^{115}In(n,n')$ monitor reaction at 5.99, 13.97, and 16.99 MeV (experiment performed at Pelletron, TIFR)
- $^{58}Ni(n,2n)^{57}Ni$ sample data with $^{27}Al(n,\alpha)$ monitor reaction at 13.97, and 16.99 MeV (experiment performed at Pelletron, TIFR)

The partial uncertainties in the attributes of the equation 11 for the given three sample monitor combinations are listed in Tables 15, 16, and 17. The covariance and correlation matrices can be calculated easily with the help of the procedure discussed for the previous case. The calculated covariances and correlation matrices for each case are provided in Tables 18, 19, and 20, respectively.

Table 15: Fractional uncertainties in various parameters used to obtain $^{58}Ni(n,p)^{58}Co$ reaction cross-sections

E_n (MeV)	Partial uncertainty (%)												
	C_s	C_m	I_{γ_s}	ε_s	ε_m	f_{λ_s}	f_{λ_m}	M_s	M_m	a_s	A_r	A_m	σ_W
2.97 ± 0.19	10.874	2.932	0.00905	1.0819	1.342	0.0962	0.0054	1.881	2.595	0.0132	1.208E-06	3.030E-07	1.007
3.37 ± 0.23	8.962	9.433	0.00905	1.0891	1.342	0.0923	0.0071	1.562	2.149	0.0132	1.208E-06	3.030E-0	0.862
Corr	0	0	1	1	1	1	1	0	0	1	1	1	0

Table 16: Fractional uncertainties in various parameters used to obtain $^{58}Ni(n,p)^{58}Co$ reaction cross-sections

E_n (MeV)	Partial uncertainty (%)												
	C_s	C_m	I_{γ_s}	I_{γ_m}	$\eta_{m,s}$	f_{λ_s}	f_{λ_m}	M_s	M_m	a_s	A_r	A_m	σ_W
5.99 ± 0.48	8.747	8.932	0.00905	0.217	0.0405	0.0950	0.000186	1.641	1.595	0.0132	1.20E-06	4.35E-06	5.67
13.97 ± 0.68	9.962	7.433	0.00905	0.217	0.0405	0.0968	0.000793	1.462	2.489	0.0132	1.20E-06	4.35E-06	5.67
16.99 ± 0.53	11.923	9.934	0.00905	0.217	0.0405	0.0973	0.000601	1.280	1.995	0.0132	1.20E-06	4.35E-06	5.67
Corr	0	0	1	1	1	1	1	0	0	1	1	1	0

Table 17: Fractional uncertainties in various parameters used to obtain $^{58}Ni(n,2n)^{57}Ni$ reaction cross-sections

E_n (MeV)	Partial uncertainty (%)												
	C_s	C_m	I_{γ_s}	I_{γ_m}	$\eta_{m,s}$	f_{λ_s}	f_{λ_m}	M_s	M_m	a_s	A_r	A_m	σ_W
13.97 ± 0.68	9.248	8.849	0.293	0.0015	0.00105	0.00058	0.0015	1.28	1.495	0.0132	1.21E-06	4.45E-07	0.37
16.99 ± 0.53	10.122	10.161	0.293	0.0015	0.00105	0.0021	0.0018	1.162	1.049	0.0132	1.21E-06	4.45E-07	2.03
Corr	0	0	1	1	1	1	1	0	0	1	1	1	0

Table 18: Covariance matrix (%) and corresponding correlation coefficients for the measured $^{58}Ni(n,p)^{58}Co$ reaction cross-sections using the FOTIA at BARC

E_n (MeV)	Covariance matrix $(V_{cs_{ij}})$		Correlation matrix	
2.97 ± 0.19	1.4113		1	
3.37 ± 0.23	0.0298	1.8011	0.0187	1

Table 19: Covariance matrix (%) and corresponding correlation coefficients for the measured $^{58}Ni(n,p)^{58}Co$ reaction cross-sections using the BARC-TIFR Pelletron facility

E_n (MeV)	Covariance matrix $(V_{cs_{ij}})$			Correlation matrix		
5.99 ± 0.48	1.9375			1		
13.97 ± 0.68	0.3221	1.9698		0.164	1	
16.99 ± 0.53	0.3221	0.3221	2.79614	0.138	0.1371	1

Table 20: Covariance matrix (%) and corresponding correlation coefficients for the measured $^{58}Ni(n,2n)^{57}Ni$ reaction cross-sections using the BARC-TIFR Pelletron facility

E_n (MeV)	Covariance matrix $(V_{cs_{ij}})$		Correlation matrix	
13.97 ± 0.68	1.7774	1		
16.99 ± 0.53	8.65×10^4	2.3594	0.0004	1

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