# Measurement and Covariance Analysis of Reaction Cross Sections for ${}^{58}\text{Ni}(n,p){}^{58}\text{Co}$ Relative to Cross Section for Formation of ${}^{97}\text{Zr}$ Fission Product in Neutron-Induced Fission of ${}^{232}\text{Th}$ and ${}^{238}\text{U}$ at Effective Neutron Energies $E_n = 5.89$ , 10.11, and 15.87 MeV

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**Abstract** – The <sup>58</sup>Ni(n,p)<sup>58</sup>Co reaction cross sections have been measured relative to two monitors: the cross sections for the formation of the <sup>97</sup>Zr fission product in neutron-induced fission of (a) <sup>232</sup>Th and of (b) <sup>238</sup>U. It is demonstrated how to generate and combine covariance matrices (using partial uncertainties and microcorrelations) in relative measurements at various stages like efficiency calibration of the high-purity germanium detector, using the ratio of <sup>58</sup>Ni(n,p)<sup>58</sup>Co reaction cross section relative to monitor cross section, and in the process of normalization. We further illustrate the weighted averaging of equivalent data as

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applicable in relative measurements. We provide the necessary data and the corresponding table of partial uncertainties as required for compilation in the EXchange-FORmat (EXFOR) database. This helps, in principle, anyone to generate and verify the steps in the calculation of the covariance matrices in the present work. We believe that it is important for all nuclear experimental scientists to incorporate a detailed data reduction procedure, reduced data, and partial uncertainties in their publications, to the extent possible, which will be very useful in EXFOR compilation.

# I. INTRODUCTION

Among the various neutron-induced reactions of nickel isotopes, the <sup>58</sup>Ni(n,p)<sup>58</sup>Co reaction is important for various applications, such as fast neutron dosimetry and spectral measurements in nuclear reactors. Moreover, nickel is present in commonly used structural materials for advanced reactors, so an accurate knowledge of the <sup>58</sup>Ni(n,p)<sup>58</sup>Co reaction cross section is required in applications such as the analysis of localized heating, hydrogen gas production, and structural damage in reactor cores. Data on this nuclear reaction also help in validation of nuclear physics model–based computer codes and in systematics studies.

The importance and motivation in generating covariance error matrices in nuclear data have been pointed out, for instance, in Refs. 1 through 4. References 2, 3, and 4 focus mainly in the context of the Indian nuclear power program, on the need for covariances in nuclear data.

In this paper, we present experimental details and covariance analysis of  ${}^{58}Ni(n,p){}^{58}Co$  reaction cross sections, relative to two monitor cross sections [the cross sections for the formation of the <sup>97</sup>Zr fission product in neutron-induced fission of (a)  $^{232}$ Th (monitor 1) and of (b) <sup>238</sup>U (monitor 2)], at three effective incident neutron energies:  $E_n = 5.89$ , 10.11, and 15.87 MeV. At each incident energy, we obtain two relative measurements (ratios) [ratio 1: the  ${}^{58}Ni(n,p){}^{58}Co$  reaction cross section relative to monitor 1, and ratio 2: the  ${}^{58}Ni(n,p){}^{58}Co$ reaction cross section relative to monitor 2]. That is, we obtain three pairs of (equivalent quantities)  $5^{8}$ Ni(n,p)  $5^{8}$ Co reaction cross sections normalized to monitor 1 and monitor 2, respectively, corresponding to the three effective neutron energies. We further collapse each pair of  ${}^{58}\text{Ni}(n,p){}^{58}\text{Co}$  reaction cross sections and summarize the result of the experiment by presenting only a single value for each distinct physical entity  $[^{58}Ni(n,p)^{58}Co$ reaction cross section at each of three effective neutron energies] using least-squares approximation.

## **II. EXPERIMENTAL PROCEDURE**

The experiment was performed using the 14 UD Bhabha Atomic Research Centre and the Tata Institute of Fundamental Research (BARC-TIFR) Pelletron facility at Mumbai, India. The neutron beam was generated using the <sup>7</sup>Li(p,n) reaction from the proton beam main line at 6 m above the analyzing magnet of the pelletron facility to utilize the maximum proton current from the

accelerator. Further, we used a collimator of 6-mm diameter before the target.

The lithium foil was made up of natural lithium with thickness of  $3.2 \text{ mg/cm}^2$ , which was sandwiched between two tantalum foils of different thicknesses. The front tantalum foil facing the proton beam was thin (3.9 mg/cm<sup>2</sup>), in which degradation of proton energy was  $\sim 30 \text{ keV}$ . The back tantalum foil was 0.025 mm thick, which was sufficient to stop the proton beam.

Behind the Ta-Li-Ta stack, we have used natural thorium and uranium metal foil (as flux monitors) and natural nickel foil for the neutron irradiation. The sizes of the U, Th, and Ni square-shaped metal foils were  $1.0 \text{ cm}^2$ . These foils (U, Th, and Ni) were wrapped separately with 0.025-mm-thick aluminium to prevent radioactive contamination from each other during irradiation. They were covered with additional Al foil of the same thickness. The U-Th-Ni stack was mounted at 0 deg with respect to the beam direction at a distance of 2.1 cm from the location of the Ta-Li-Ta stack.

There are three sets of foils (U, Th, and Ni). The foils in set 1 were given tag numbers Ni-1, U-1, and Th-1; set 1 was placed behind the Ta-Li-Ta stack and irradiated at proton energy of 7.8 MeV. Similarly, foils in set 2 were given tag numbers Ni-2, U-2, and Th-2, and foils in set 3 were given tag numbers Ni-3, U-3, and Th-3. The foils in set 2 and set 3 were placed behind the Ta-Li-Ta stack and irradiated at a proton energy of 12 and 18 MeV, respectively.

A schematic diagram of the experimental setup can be found in Ref. 5. Different sets of stacks were made for different irradiations at various neutron energies.

Three sets of stacks of foils as mentioned above were irradiated at the proton energies ( $E_p$ ) of 7.8, 12, and 18 MeV. The irradiation times at these three energies were 15, 4, and 5 h, respectively. The proton current during the irradiations varied from 100 to 250 nA. The effective neutron energies hitting the U-Th-Ni stack samples were  $E_n = 5.89$ , 10.11, and 15.87 MeV (see Sec. III.B for details), corresponding to the proton energies of 7.8, 12, and 18 MeV, respectively.

After each irradiation, the samples were cooled for  $\sim 2$  h, and the samples were mounted on different Perspex plates. The  $\gamma$ -ray activities of the reactions and fission products in the irradiated samples of Ni, U, and Th were analyzed by using a precalibrated high-purity germanium (HPGe) detector coupled with a personal computer–based 4K multichannel analyzer. The resolution of the detector system during counting was 2 keV at a 1332-keV gamma line of <sup>60</sup>Co.

# **III. DATA ANALYSIS**

The basics of Bayesian Probability Theory, error propagation, and least-squares approximations needed for this section can be found in Refs. 6 and 7.<sup>a</sup> In the present work, our interest is to obtain mean values<sup>b</sup> and covariance information for the  ${}^{58}Ni(n,p){}^{58}Co$  reaction cross section at effective neutron energies  $E_n = 5.89$ , 10.11, and 15.87 MeV. In this process we obtain covariance information of efficiency calibration of the HPGe detector (Sec. III.A), ratio measurement (Sec. III.C), and normalization (Sec. III.D). Covariance information obtained in efficiency calibration of the HPGe detector is used to obtain covariance information of ratio measurement, which is further used in normalization to obtain covariance information for the  ${}^{58}$ Ni(n,p) ${}^{58}$ Co reaction cross section at three effective neutron energies  $E_n = 5.89$ , 10.11, and 15.87 MeV.

# III.A. Efficiency Calibration of HPGe Detector Using <sup>152</sup>Eu Standard Gamma-Ray Source

The calibration procedure (Refs. 8 and 9) for the HPGe photon detector used in the present work is carried out using a <sup>152</sup>Eu standard point gamma-ray source [source activity (A<sub>0</sub>) is 7767.67  $\pm$  155.35 as of Jan. 10, 1999, and time elapsed *t* is 9.893 years], situated a suitable distance from the detector ( $\approx$  10 cm). The model used to obtain efficiency is

$$\epsilon = \frac{C}{aA_0 e^{-\lambda t}} , \qquad (1)$$

where

- $\epsilon$  = detector efficiency
- C = gamma-ray peak count
- a = branching factor for gamma rays ( $\gamma$  abundance)
- A = source activity at the time of the count
- $A_0$  = source activity at the time of source calibration
- $\lambda = \text{decay constant}$
- t = time elapsed between the source calibration and detector calibration.

Note that *C* is the only measured quantity, whereas auxiliary quantities *a*,  $A_0$ , and  $\lambda$  were known prior to the measurement of *C* and are taken from external sources. Using the mean values of measured quantity *C* and

auxiliary quantities *a*, which are presented in Table I ( $\gamma$  abundances are taken from Ref. 10), we obtain efficiency of the detector for six gamma lines indexed by j = 1, 2, ..., 6 using Eq. (2) (the notation  $\langle ... \rangle$  is used for mean or expectation value<sup>5</sup>):

$$\langle \epsilon_j \rangle = \frac{\langle C_j \rangle}{\langle A_0 \rangle e^{-\langle \lambda \rangle t} \langle a_j \rangle} .$$
 (2)

The *jk*'th element of the covariance matrix for efficiencies  $(\langle \delta \epsilon_j \delta \epsilon_k \rangle)$  is obtained using

$$(V_{\epsilon})_{jk} \equiv \langle \delta \epsilon_j \delta \epsilon_k \rangle = \sum_{r=1}^{2} [(p_r)_j (s_r)_{jk} (p_r)_k] \delta_{jk} + \sum_{r=3}^{4} (p_r)_j (s_r)_{jk} (p_r)_k , \qquad (3)$$

where

- $\delta \epsilon_j$ ,  $\delta \epsilon_k$  = errors in efficiency of the HPGe detector for *j*'th and *k*'th gamma line, respectively
- $(p_r)_j, (p_r)_k =$  partial uncertainties<sup>1</sup> in the *r*'th attribute (the attributes *C*, *a*, *A*<sub>0</sub>, and  $\lambda$  are represented by indexes *r* = 1, 2, 3, and 4, respectively) corresponding to *j*'th and *k*'th gamma line, respectively
  - $(s_r)_{jk}$  = microcorrelation<sup>1</sup> between  $\delta \epsilon_j$  and  $\delta \epsilon_k$  due to *r*'th attribute
    - $\delta_{jk} =$  Kroneker delta  $(\delta_{jk} = 1 \text{ for } j = k \text{ and } \delta_{jk} = 0$ for  $j \neq k$ ) and  $\delta_{jk}$  ensures correlation between  $\delta \epsilon_j$  and  $\delta \epsilon_k$  is due to common errors  $\delta A_0$  and  $\delta \lambda [(s_r)_{jk} = 0 \text{ for } r = 1 \text{ and } 2$ and  $(s_r)_{jk} = 1 \text{ for } r = 3 \text{ and } 4].$

Partial uncertainties in  $\epsilon$  due to attributes C, a,  $A_0$ , and  $\lambda$  required for Eq. (3) are presented in Table II. The last column in Table II refers to uncertainty in efficiency of the HPGe detector for the *j*'th gamma line,  $\Delta \epsilon_j = \sqrt{\langle \delta \epsilon_j \rangle^2} = \sqrt{\sum_{r=1}^4 [(p_r)_j]^2}$ .

Table III presents the results of the covariance analysis for efficiency of the HPGe detector; i.e, the mean values of efficiency of the HPGe detector corresponding to six gamma lines along with covariance matrix  $V_{\epsilon}$  of dimension 6 are presented in Table III.

The characteristic gamma line from the reaction product  ${}^{58}$ Co is 810.77 keV, and the gamma line from the fission product  ${}^{97}$ Zr is 743.36 keV. Both these gamma energies are different from the energies of the gamma lines from  ${}^{152}$ Eu, which is used for calibration of the HPGe detector. But, for the covariance analysis of ratio measurement, the mean values of efficiency of the HPGe detector corresponding to the above-mentioned (810.77 and 743.36 keV) characteristic gamma lines and the

<sup>&</sup>lt;sup>a</sup>Also see references therein for further details.

<sup>&</sup>lt;sup>b</sup>Mean value or expectation value interpreted in accordance with decision under quadratic loss. The interested reader may consult Sec. 6.11.1, "From Posterior Distribution Function to Estimate," in Ref. 6.

corresponding covariance between the errors of efficiency are needed in the covariance analysis of ratio measurement. This can be accomplished with the method of least squares using the following empirical formula (model) to fit the measured calibration data (details can be found in Ref. 9):

$$Z_i \approx \ln \epsilon_i = \sum_{k=1}^m P_k (\ln E_i)^{k-1} , \qquad (4)$$

where

- $\epsilon_i$  = efficiency of HPGe detector corresponding to gamma line of energy  $E_i$
- k = index that corresponds to number of fitting parameters required

 $P_k = k$ 'th fitting parameter.

We can represent Eq. (4) by the compact matrix expression

# TABLE I

Specification of Gamma-Ray Energy, Measured Gamma Counts, and Gamma Abundance

Line Number	$E_{\gamma}$ (keV)	C (count/s)	a (%)
1	244.6975	$10626 \pm 193$	$7.583 \pm 0.019$
2	411.1163	$1878 \pm 110$	$2.234 \pm 0.004$
3	867.3780	$1617 \pm 95$	$4.245 \pm 0.019$
4	964.0790	$5269 \pm 100$	$14.605 \pm 0.021$
5	1112.074	$4493 \pm 89$	$13.644 \pm 0.021$
6	1299.140	$510 \pm 45$	$1.623 \pm 0.008$
1	1	1	1

TABLE II

Partial Uncertainties, Required in Sec. III.A

Line Number	С	а	$A_0$	λ	Δε
1 2	0.0604 0.1169	0.0083	0.0665	0.0015	0.0903
3	0.0531	0.0040	0.0181	0.0004	0.0563
5 6	0.0155 0.0658	0.0012 0.0037	0.0156 0.0149	0.0003 0.0003	0.0220 0.0676

# TABLE III

Gamma Energy  $E_{\gamma}$ , Efficiency  $\epsilon$ , Total Uncertainty  $\Delta \epsilon$ , and Corresponding Absolute Covariance Matrix  $[(V_{\epsilon})_{ij} = \langle \delta \epsilon_i \delta \epsilon_j \rangle], i, j = 1, 2, ..., 6$ 

i	$E_\gamma$	ε (%)	Δε (%)	$<\!\delta\epsilon_i\delta\epsilon_j>$
1	244.6975	3.3264	0.0903	0.0081           0.0027         0.0153           0.0012         0.0007         0.0032           0.0011         0.0007         0.0003         0.0006           0.0010         0.0006         0.0003         0.0003         0.0005           0.0010         0.0006         0.0003         0.0003         0.00046
2	411.1163	1.9956	0.1236	
3	867.378	0.9042	0.0563	
4	964.079	0.8564	0.0236	
5	1112.074	0.7817	0.0220	
6	1299.140	0.7459	0.0676	

$$\mathbf{Z} \approx \mathbf{A} \mathbf{P} \,, \tag{5}$$

where

 $Z_i = \ln \epsilon_i, i = 1, 2, ..., n =$  elements of the vector **Z P** = matrix containing elements  $P_k, k = 1, 2, ..., m$ **A** = design matrix containing elements  $(\mathbf{A})_{ik} = (\ln E_i)^{k-1}$ .

The least-squares condition states that the best estimate for **P** is the one that minimizes  $\chi^2$  given by

$$\chi^2 = (\mathbf{Z} - \mathbf{A}\mathbf{P})^{\mathrm{T}} \mathbf{V}_{\mathbf{Z}}^{-1} (\mathbf{Z} - \mathbf{A}\mathbf{P}) , \qquad (6)$$

where superscript -1 denotes matrix inversion. The solution **P** can be extracted from the normal equations,  $\frac{\partial \chi^2}{\partial p_k} = 0$ . It is given by the following formulas:

$$\mathbf{P} = \mathbf{V}_{\mathbf{P}} \mathbf{A}^{\mathrm{T}} \mathbf{V}_{\mathbf{Z}}^{-1} \mathbf{Z}$$
(7)

and

$$\mathbf{V}_{\mathbf{P}} = (\mathbf{A}^{\mathrm{T}} \mathbf{V}_{\mathbf{Z}}^{-1} \mathbf{A})^{-1} , \qquad (8)$$

where  $\mathbf{V}_{\mathbf{P}}$  is the covariance matrix for the solution parameters  $\mathbf{P}$  and matrix  $\mathbf{V}_{\mathbf{Z}}$  is obtained using  $(\mathbf{V}_{\mathbf{Z}})_{ij} = \frac{(V_{\epsilon})_{ij}}{\langle \epsilon_i \rangle \langle \epsilon_j \rangle}$ . Substitution of the solution for  $\mathbf{P}$  into Eq. (6) yields a specific value for  $\chi^2$  [which is governed by the  $\chi^2$  distribution with (n - m) degrees of freedom, so its expected value is (n - m); see Ref. 7 for details], thereby providing a means to test the quality of the fit. Using data presented in Table III, we obtain  $\mathbf{P} =$  $(12.7148, -2.9793, 0.1612)^T$  ( $\mathbf{V}_{\mathbf{P}}$  is not presented to save space), and  $\chi^2 = 2.6613$  ( $\approx n - m = 6 - 3$ ) indicates quality of the fit. Using  $\mathbf{P}$  and  $\mathbf{V}_{\mathbf{P}}$  obtained, we get efficiencies  $1.0655 \pm 0.0394$  and  $0.9910 \pm 0.0323$ (in percent) corresponding to the characteristic gamma lines 743.36 and 810.77 keV, respectively, with 99.23% correlation.

#### III.B. Calculation of Effective Neutron Energy $E_n$

Proton energies used in the present work are  $E_p =$  7.8, 12, and 18 MeV. Corrections and uncertainty in  $E_p$  are assigned based on the following information. The spread in the proton beam main line at 6 m above the analyzing magnet of the pelletron facility is of the order of 50 to 90 keV; degradation of the proton energy<sup>11</sup> in the front tantalum foil of thickness 3.9 mg/cm<sup>2</sup> is 84.166, 64.420, and 48.772 keV corresponding to  $E_p =$  7.8, 12, and 18 MeV, respectively; degradation of the proton energy in lithium foil of thickness 3.2 mg/cm<sup>2</sup> is 147.166, 105.675, and 75.710 keV corresponding to  $E_p =$  7.8, 12, and 18 MeV, respectively.

The above-mentioned information was utilized in calculating and assigning uncertainties to  $E_p$ . We obtain  $E_p = 7.664 \pm 0.050$ ,  $11.895 \pm 0.036$ , and  $17.918 \pm 0.029$  MeV. Neutron energy  $E_n^k$  can be obtained using relation  $E_n^k = E_p - E_{\rm Th}$ , where  $E_{\rm Th}$  is the <sup>7</sup>Li(p,n)<sup>7</sup> Be reaction threshold energy ( $E_{\rm Th} = 1880.3558 \pm 0.0812$  keV). Following are the mean values and uncertainties assigned to neutron energy  $E_n^k$ : 5.7840  $\pm$  0.0503, 10.0146  $\pm$  0.0376, and 16.0374  $\pm$  0.0285 MeV.

We obtain neutron energy  $E_n^{sp}$  from neutron spectrums (neutron spectrums corresponding to  $E_p = 7.8$ , 12, and 18 MeV are given in Ref. 5). The mean values of  $E_n^{sp}$ corresponding to the primary group of neutrons (the peak corresponding to the highest neutron energy is due to the primary group of neutrons) are obtained as weighted averages with flux as weight. And, uncertainty assigned to  $E_n^{sp}$  is obtained based on full-width at half-maximum (FWHM) taken from the spectrum corresponding to the primary group of neutrons and then using the following relation: *uncertainty* = FWHM/2.355. Following are the mean values and uncertainty assigned to neutron energy  $E_n^{sp}$ : 5.9926  $\pm$  0.2335, 10.2106  $\pm$  0.1062, and 15.6972  $\pm$ 0.2578 MeV.

The mean values and uncertainty of  $E_n$  quoted in the present work were obtained by taking the average of neutron energies  $E_n^k$  and  $E_n^{sp}$ . Following are the mean values and uncertainty assigned to effective neutron energy  $E_n$  quoted in the present work:  $5.8883 \pm 0.1194$ ,  $10.1126 \pm 0.0563$ , and  $15.8673 \pm 0.1297$  MeV.

#### III.C. Ratio Measurement

Since we used two monitors (cross section for the fission yield of  ${}^{97}$ Zr in  ${}^{232}$ Th and cross section for the fission yield of  ${}^{97}$ Zr in  ${}^{238}$ U), we obtained two ratios at each of the three effective neutron energies along with covariance information, which is further used to obtain the evaluated values of the  ${}^{58}$ Ni(n,p) ${}^{58}$ Co reaction cross section at three effective neutron energies  $E_n = 5.89$ , 10.11, and 15.87 MeV with covariance information. In the present work, a neutron beam is generated using the  ${}^{7}$ Li(p,n) reaction. These neutrons do not form a monoenergetic neutron source at the higher proton energies considered in our experiment. Therefore, a correction factor  $\alpha$ , accounting for low-energy neutron contributions, is incorporated in Eq. (9) for ratio measurement:

$$\frac{\sigma_u(E_n)}{\sigma_m(E_n)} \equiv r_{um} = \frac{Q_u}{Q_m} , \qquad (9)$$

where

# $r_{um}$ = ratio of the unknown *u* cross section $\sigma_u$ to the monitor *m* cross section $\sigma_m$

- $\sigma_u = \text{cross section for the reaction } {}^{58}\text{Ni}(n,p){}^{58}\text{Co at}$ neutron energy  $E_n$
- $\sigma_m$  = cross section for the fission yield of <sup>97</sup>Zr in <sup>232</sup>Th or cross section for the fission yield of <sup>97</sup>Zr in <sup>238</sup>U at neutron energy  $E_n$ ;

$$Q_{i} = \frac{C_{i}\lambda_{i} \left(\frac{CL}{LT}\right)_{i}}{N_{i}a_{i}\epsilon_{i}(1-e^{-\lambda_{i}t_{irr}})e^{-\lambda_{i}t_{icool}}(1-e^{-\lambda_{i}t_{icount}})\alpha_{i}} \quad i = u, m$$

$$(10)$$

and

$$\alpha_{i} = \left(1 + \frac{\beta_{i}}{\Phi(E_{p1})\sigma_{i}(E_{p1})}\right),$$
  
$$\beta_{i} = \Phi(E_{p2})\sigma_{i}(E_{p2}) + \int_{0}^{E_{\max}} \varphi(E)\sigma_{i}(E)dE \quad i = u, m, \quad (11)$$

where

- $C_i$  = gamma-ray peak counts
- $\lambda_i$  = decay constants of product nuclei
- CL, LT = clock and live time of detector
  - $N_i$  = number of sample atoms
  - $a_i =$  gamma abundances
  - $\epsilon_i$  = efficiency of the detector
  - $t_{irr}$  = irradiation time
  - $t_{icool}$  = cooling time
  - $t_{icount}$  = counting time
  - $\Phi$ ,  $\phi$  = flux corresponding to discrete peaks and continuum, respectively, with reference to neutron spectrums given in Ref. 5.

The terms  $E_{p1}$  ( $E_{p1} = \frac{\sum_i E_i \Phi_i}{\sum_i \Phi_i}$  for higher energy peak,

and  $E_n^{sp}$  is used in Sec. III.B for  $E_{p1}$ ) and  $E_{p2}$  are used for higher and lower neutron energy peaks, and E corresponds to much lower neutron energies (continum) with reference to neutron spectrums given in Ref. 5.

The correction term  $\alpha_i$  in Eq. (9) is obtained following the bootstrap approach adopted from Refs. 12 and 13<sup>c</sup>;  $\alpha_i$  is obtained by using the group flux and group cross sections (see Ref. 13 for details); the group flux data corresponding to  $E_p = 7.8$ , 12, and 18 MeV are obtained from the neutron spectrums of Ref. 5; and the group crosssection data are obtained from the evaluated cross-section database ENDF/B-VII.1 (Ref. 14). The mean values and elements of the relative covariance matrix for the case of two ratios corresponding to two different neutron energies are given by (see Ref. 8 for details)

$$< r_{12} > = \frac{}{}, < r_{34} > = \frac{}{}$$
 (12)

and

$$\frac{\langle \delta r_{12} \delta r_{34} \rangle}{\langle r_{12} \rangle \langle r_{34} \rangle} = \frac{\langle \delta Q_1 \delta Q_3 \rangle}{\langle Q_1 \rangle \langle Q_3 \rangle} + \frac{\langle \delta Q_2 \delta Q_4 \rangle}{\langle Q_2 \rangle \langle Q_4 \rangle} - \frac{\langle \delta Q_1 \delta Q_4 \rangle}{\langle Q_2 \rangle \langle Q_4 \rangle} - \frac{\langle \delta Q_2 \delta Q_3 \rangle}{\langle Q_2 \rangle \langle Q_3 \rangle}.$$
(13)

That is, in order to determine the mean value and relative covariance for the ratio, we need the mean value and relative covariances for Q. Basic data used in the present work to determine  $\langle Q_i \rangle$ ,  $\langle Q_j \rangle$  are presented in Tables IV and V (the half-lives and  $\gamma$  abundances presented in Table V are taken from Ref. 10). In order to save space, instead of presenting raw count data and cooling and counting times, we present  $\langle Q_i \rangle$ ,  $\langle Q_j \rangle$  in Table VI, and the covariance matrix (of dimension 12) in absolute form,

$$<\delta Q_i \delta Q_j > = \sum_{r=1}^3 [(p_r)_i (S_r)_{ij} (p_r)_j] \delta_{ij} + \sum_{r=4}^5 (p_r)_i (S_r)_{ij} (p_r)_j,$$
(14)

is obtained using the table of partial uncertainties (as presented<sup>d</sup> in Table VII), where five attributes, r = 1 through 5, correspond to measured quantity *C* and auxiliary quantities *N*, *a*,  $\epsilon$ , and  $\lambda$ , respectively. Partial errors in *Q* due to attribute 4 (efficiency) are partially correlated, corresponding correlation information is obtained from the calibration process as explained in Sec. III.A, and microcorrelation for attribute 5 (decay constant) is assigned based on the daughter nuclei produced in a reaction (full correlation is assigned for the decay constant of the same daughter nuclei; otherwise, a zero correlation is assigned). Transform the covariance matrix in relative form  $\frac{\langle \delta Q_i \delta Q_j \rangle}{\langle O_i \rangle \langle O_i \rangle}$  to

<sup>&</sup>lt;sup>c</sup>Suggested by D. L. Smith, Nuclear Engineering Division, Argonne National Laboratory.

<sup>&</sup>lt;sup>d</sup>Note that in Table VII, all partial uncertainties presented are multiplied by  $10^{18}$ , for example,  $(p_1)_1 \times 10^{18} = 428.8653$ ; hence,  $(p_1)_1 = 428.8653 \times 10^{-18}$ .

# <sup>58</sup>Ni(*n*,*p*)<sup>58</sup>Co CROSS SECTIONS

# TABLE IV

weight of Natural Ni, Th, and O Samples and Isotope Abundance	Weight of	Natural Ni,	Th, and	U Samples an	nd Isotope Abundance
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Element	Tag Number	Weight (g)	Isotope	Isotope Abundance
Ni	Ni-1 Ni-2 Ni-3	$\begin{array}{c} 0.4262 \pm 0.0085 \\ 0.1813 \pm 0.0036 \\ 0.1260 \pm 0.0025 \end{array}$	<sup>58</sup> Ni	$0.68077 \pm 0.00009$
Th	Th-1 Th-2 Th-3	$\begin{array}{c} 0.2856 \pm 0.0057 \\ 0.3230 \pm 0.0065 \\ 0.3252 \pm 0.0065 \end{array}$	<sup>232</sup> Th	0.999999 ± 0.00001
U	U-1 U-2 U-3	$\begin{array}{c} 0.6970  \pm  0.0139 \\ 0.9917  \pm  0.0198 \\ 0.5795  \pm  0.0116 \end{array}$	<sup>238</sup> U	$0.99275 \pm 0.00006$

# TABLE V

# Decay Data Required for Ratio Measurement

Isotope	$T_{1/2}$	$E_{\gamma}$ (MeV)	Gamma Abundance
<sup>58</sup> Ni	$70.86 \pm 0.07 \ d$	0.81077	$\begin{array}{c} 0.98999  \pm  0.00001 \\ 0.92999  \pm  0.00001 \end{array}$
<sup>97</sup> Zr	16.91 $\pm 0.05 \ h$	0.74336	

# TABLE VI

# Mean Values of Q

	$< Q > \times 10^{-14}$		
$E_n$ (MeV)	Ni	Th	U
$\begin{array}{c} 05.8883  \pm  0.1194 \\ 10.1126  \pm  0.0563 \\ 15.8673  \pm  0.1297 \end{array}$	0.6933 0.8002 0.1563	0.0086 0.0149 0.0146	0.0086 0.0605 0.0536

# TABLE VII

# Partial Uncertainties, Required in Sec. III.C

		Partial Uncertainties $\times 10^{18}$ due to Attributes				
$E_n$ (MeV)	Tag	С	N	a	E	λ
$\begin{array}{c} 5.8883 \pm 0.1194 \\ 5.8883 \pm 0.1194 \\ 5.8883 \pm 0.1194 \\ 10.1126 \pm 0.0563 \\ 10.1126 \pm 0.0563 \\ 10.1126 \pm 0.0563 \\ 10.1126 \pm 0.0563 \\ 15.8673 \pm 0.1297 \\ 15.8673 \pm 0.1297 \end{array}$	Ni-1 Th-1 U-1 Ni-2 Th-2 U-2 Ni-3 Th-3	428.8653 6.1420 15.0747 405.7781 9.2834 7.2366 104.2130 7.1843	138.6695 1.7214 8.0167 160.0501 2.9778 12.1093 31.2646 2.9178	0.0700 0.0009 0.0043 0.0808 0.0016 0.0065 0.0158 0.0016	226.1246 3.1797 14.8085 260.9894 5.5006 22.3684 50.9823 5.3899	6.8492 0.2545 1.1852 7.9052 0.4402 1.7903 1.5442 0.4314
$15.8673 \pm 0.1297$	U-3	6.9953	10.7181	0.0058	19.7985	1.5846

obtain the covariance matrix (of dimension 6) for the ratio in relative form<sup>e</sup>  $\frac{\langle \delta r_{ij} \delta r_{kl} \rangle}{\langle r_{ij} \rangle \langle r_{kl} \rangle}$  using Eq. (13) as presented in Table VIII.

# III.D. Normalization

After obtaining the mean values  $\langle r_{ij} \rangle$  and relative covariance  $R_r$  for the ratio as presented in Table VIII, the next step is to obtain the mean value and covariances for the <sup>58</sup>Ni(*n*,*p*)<sup>58</sup>Co reaction cross section, normalized to the cross section for formation of the <sup>97</sup>Zr fission product in neutron-induced fission of <sup>232</sup>Th and <sup>238</sup>U, at effective neutron energies  $E_n = 5.89$ , 10.11, and 15.87 MeV, respectively, using

$$<\sigma_i> = < r_{ij}> < \sigma_j> = < r_{ij}> < \sigma_{fj}> , (15)$$

$$\langle \sigma_k \rangle = \langle r_{kl} \rangle \langle \sigma_l \rangle = \langle r_{kl} \rangle \langle \sigma_{fl} \rangle$$
, (16)

and

<sup>e</sup>Elements of the covariance matrix for the ratio in relative form  $R_r$ .

$$\frac{\langle \delta \sigma_i \delta \sigma_k \rangle}{\langle \sigma_i \rangle \langle \sigma_k \rangle} = \frac{\langle \delta r_{ij} \delta r_{kl} \rangle}{\langle r_{ij} \rangle \langle r_{kl} \rangle} + \frac{\langle \delta \sigma_{fj} \delta \sigma_{fl} \rangle}{\langle \sigma_{fj} \rangle \langle \sigma_{fl} \rangle} + \frac{\langle \delta Y_{fj} \delta Y_{fl} \rangle}{\langle Y_{fj} \rangle \langle Y_{fl} \rangle} .$$
(17)

The mean values and uncertainties of the fission cross sections  $\sigma_{fi}$  and fission product yields  $Y_{fi}$  required for Eqs. (15), (16), and (17) are presented in Table IX. The fission cross sections taken from Refs. 14 and 15 were linearized using the PREPRO linear module,<sup>16</sup> and the fission cross sections used in the present work (column 3 of Table IX) were obtained using linear-linear interpolation.<sup>16</sup> The fission product yield was taken from Ref. 17 for 14-MeV neutron-induced fission, and the fission product yield is assumed constant at three effective neutron energies.

In order to obtain the covariance matrix in relative form for the normalized cross section  $\frac{\langle \delta \sigma_i \delta \sigma_k \rangle}{\langle \sigma_i \rangle \langle \sigma_k \rangle}$  [see Eq.(17)], we need  $\frac{\langle \delta r_{ij} \delta r_{kl} \rangle}{\langle r_{ij} \rangle \langle r_{kl} \rangle}$  (relative covariance for ratios as presented in Table VIII),  $\frac{\langle \delta \sigma_{fj} \delta \sigma_{fl} \rangle}{\langle \sigma_{fj} \rangle \langle \sigma_{fl} \rangle}$ (relative covariance for fission cross sections), and  $\frac{\langle \delta Y_{fj} \delta Y_{fl} \rangle}{\langle Y_{fj} \rangle \langle Y_{fl} \rangle}$  (relative covariance for fission yields). Since only the mean values and uncertainties of the fission cross sections were considered, the correlations between

#### TABLE VIII

We all values $\langle T_{ii} \rangle$ and Relative Covariance Matrix $K_r$ for Ka	Mean	Values	$\langle r_{ii} \rangle$	and	Relative	Covariance	Matrix	$R_r$	for	Ra
--	------	--------	--------------------------	-----	----------	------------	--------	-------	-----	----

$E_n$ (MeV)	< <i>r</i> <sub>ij</sub> >	$R_r \times 100$
$\begin{array}{c} 5.8883 \pm 0.1194 \\ 10.1126 \pm 0.0563 \\ 15.8673 \pm 0.1297 \\ 5.8883 \pm 0.1194 \\ 10.1126 \pm 0.0563 \\ 15.8673 \pm 0.1297 \end{array}$	80.5667 53.7473 10.7147 17.2973 13.2169 2.9170	0.9766         0.0047       0.7306         0.0047       0.0047       0.7716         0.4273       0.0047       0.0047       0.6087         0.0047       0.3018       0.0047       0.3561         0.0047       0.0047       0.4891       0.0047       0.5462

TABLE IX

ield	
	ield

Reaction	Neutron Energy (MeV)	Fission Cross Section (b)	Fission Product	Fission Product Yield
$^{232}$ Th( <i>n</i> , <i>f</i> )	5.8883 10.1126 15.8673	$\begin{array}{c} 0.1507 \pm 0.0036 \\ 0.3169 \pm 0.0073 \\ 0.4502 \pm 0.0145 \end{array}$	<sup>97</sup> Zr	0.0340 ± 0.0014
<sup>238</sup> U( <i>n</i> , <i>f</i> )	5.8883 10.1126 15.8673	$\begin{array}{r} 0.5860  \pm  0.0057 \\ 1.0014  \pm  0.0096 \\ 1.2985  \pm  0.0170 \end{array}$	<sup>97</sup> Zr	0.0537 ± 0.0012

the errors of the fission cross sections are assigned

zero; hence, 
$$\frac{\langle \delta \sigma_{fj} \delta \sigma_{fl} \rangle}{\langle \sigma_{fj} \rangle \langle \sigma_{fl} \rangle} = 0$$
 for  $j \neq l$  and  $\langle \delta \sigma_{r} \delta \sigma_{r} \rangle = (\Delta \sigma_{rr})^2$ 

$$\frac{\langle \delta\sigma_{fj} \delta\sigma_{fl} \rangle}{\langle \sigma_{fj} \rangle \langle \sigma_{fl} \rangle} = \left(\frac{\Delta\sigma_{fj}}{\langle \sigma_{fj} \rangle}\right)^2 \text{ for } j = l \ (\langle \sigma_f \rangle \pm \Delta\sigma_f)$$

are presented in Table IX). The relative covariance for the fission yields is generated based on the following discussion. We have considered the 97Zr fission yield in the  $^{232}$ Th(*n*,*f*) reaction and the  $^{97}$ Zr fission yield in the <sup>238</sup>U(n,f) reaction, respectively, for 14-MeV neutroninduced fission, and the fission product yield is assumed constant at three effective neutron energies. Hence, the <sup>97</sup>Zr fission yield [in <sup>232</sup>Th(n, f)] error is common at three effective neutron energies and fully correlated; similarly, the  ${}^{97}$ Zr fission yield [in  ${}^{238}$ U(*n*,*f*)] error is common at three effective neutron energies and fully correlated, whereas the correlation between the error in the <sup>97</sup>Zr fission yield corresponding to the  $^{232}$ Th(*n*,*f*) reaction and the error in the <sup>97</sup>Zr fission yield corresponding to the  $^{238}$ U(*n*,*f*) reaction is assigned zero. The mean values and covariance matrix in relative form for the normalized cross sections are presented<sup>f</sup> in Table X.

#### III.E. Weighted Averaging of Equivalent Data Points

As can be observed in Table X, we have mean values  $(6 \times 1 \text{ column vector})$ 

$$\sigma_n = [(\langle \sigma_{n1} \rangle, \langle \sigma_{n2} \rangle), (\langle \sigma_{n3} \rangle, \langle \sigma_{n4} \rangle), (\langle \sigma_{n5} \rangle, \langle \sigma_{n6} \rangle)]^T \equiv [\sigma_{\alpha_1}, \sigma_{\alpha_2}, \sigma_{\alpha_3}]^T,$$
(18)

<sup>f</sup>Note that in Table X the <sup>58</sup>Ni(n,p)<sup>58</sup>Co cross sections normalized to the cross sections for the formation of the <sup>97</sup>Zr fission yield in the <sup>232</sup>Th(n,f) reaction are 0.4128 ± 0.0453, 0.5792 ± 0.0566, and 0.1640 ± 0.0168 at effective neutron energies 5.8883, 10.1126, and 15.8673 MeV, respectively. where

$$\sigma_{\alpha_i} \equiv [\langle \sigma_{nk} \rangle, \langle \sigma_{nl} \rangle]^I \tag{19}$$

represents the pair of normalized cross sections of the same physical quantity [ ${}^{58}Ni(n,p)$  reaction cross section at energy  $E_{ni}$ , which has a definite value] and covariance matrix (of dimension 6)

$$\mathbf{V}_{\sigma_{\mathbf{n}}} = [\langle \delta \sigma_{\alpha_i} \delta \sigma_{\alpha_j} \rangle], i, j = 1, 2, 3, \qquad (20)$$

where

$$\mathbf{V}_{\alpha_{ij}} \equiv \langle \delta \sigma_{\alpha_i} \delta \sigma_{\alpha_j} \rangle \equiv [\langle \delta \sigma_{nk} \delta \sigma_{nk} \rangle, \langle \delta \sigma_{nk} \delta \sigma_{nl} \rangle;$$
  
$$\langle \delta \sigma_{nl} \delta \sigma_{nk} \rangle, \langle \delta \sigma_{nl} \delta \sigma_{nl} \rangle]^T . \tag{21}$$

The problem is to obtain the evaluated value  $\sigma_e = [\langle \sigma_{e1} \rangle, \langle \sigma_{e2} \rangle, \langle \sigma_{e3} \rangle]^T$  at energy  $E_n = [5.89, 10.11, 15.87]^T$  MeV using the following approximation [see Eq. (19)]:

$$\sigma_{\alpha_i} \equiv [\langle \sigma_{nk} \rangle, \langle \sigma_{nl} \rangle]^T \approx [\langle \sigma_{ei} \rangle, \langle \sigma_{ei} \rangle]^T = [1, 1]^T \langle \sigma_{ei} \rangle = \mathbf{A}_{\alpha \mathbf{i}} \langle \sigma_{ei} \rangle .$$
(22)

The least-squares approach to obtain  $\langle \sigma_{ei} \rangle$  is to minimize  $\chi^2_{\alpha i} \left( \frac{\partial \chi^2_{\alpha i}}{\partial \langle \sigma_{ei} \rangle} = 0 \right)$  given by

$$\chi_{\alpha i}^{2} = (\sigma_{\alpha_{i}} - \mathbf{A}_{\alpha \mathbf{i}} < \sigma_{ei} >)^{T} V_{\alpha_{ij}}^{-1} (\sigma_{\alpha_{i}} - \mathbf{A}_{\alpha \mathbf{i}} < \sigma_{ei} >) . \quad (23)$$

The mean value  $\langle \sigma_{ei} \rangle$ , which corresponds to the least-squares solution,<sup>18,19</sup> is obtained using

$$<\sigma_{ei}>=\mathbf{B}_{\alpha \mathbf{i}}^{\mathbf{T}}\sigma_{\alpha_i}$$
, (24)

where

$$\mathbf{B}_{\alpha \mathbf{i}} = (\mathbf{C}_{\alpha \mathbf{i}} \mathbf{A}_{\alpha \mathbf{i}} \mathbf{V}_{\alpha_{\mathbf{i}\mathbf{i}}}^{-1})^T$$
(25)

### TABLE X

<sup>58</sup>Ni(*n*,*p*)<sup>58</sup>Co Reaction Cross Section  $\langle \sigma_n \rangle$  Obtained by Normalizing with Respect to <sup>232</sup>Th(*n*,*f*) and <sup>238</sup>U(*n*,*f*) Monitor Cross Section and <sup>97</sup>Zr Fission Yield with Covariance  $V_{\sigma_n}$  Matrix in Absolute Form

$E_n$ (MeV)	$<\sigma_n>$ (b)	$V_{\sigma_n}  imes 100$		
$5.8883 \pm 0.1194$	$\begin{array}{c} 0.4128 \pm 0.0453 \\ 0.5447 \pm 0.0445 \end{array}$	0.2054 0.0961 0.1982		
$10.1126 \pm 0.0563$	$\begin{array}{c} 0.5792 \pm 0.0566 \\ 0.7108 \pm 0.0458 \end{array}$	$\begin{array}{cccccccccccccccccccccccccccccccccccc$		
$15.8673 \pm 0.1297$	$\begin{array}{c} 0.1640 \pm 0.0168 \\ 0.2034 \pm 0.0159 \end{array}$	0.0119 0.0004 0.0167 0.0005 0.0282 0.0004 0.0060 0.0006 0.0079 0.0163 0.0254		

#### SHIVASHANKAR et al.

#### TABLE XI

Evaluated Values of  ${}^{58}$ Ni(*n*,*p*) ${}^{58}$ Co Reaction Cross Sections  $<\sigma_e>$  with Absolute Covariance Matrix and Chi-Square Values

$E_n$ (MeV)	$<\sigma_e>$ (b)	$V_{\sigma_e} \times 100$	$\chi^2$
$5.8883 \pm 0.1194 \\ 10.1126 \pm 0.0563 \\ 15.8673 \pm 0.1297$	$\begin{array}{c} 0.4810 \pm 0.0386 \\ 0.6709 \pm 0.0429 \\ 0.1864 \pm 0.0146 \end{array}$	0.1489 0.0145 0.1838 0.0045 0.0056 0.0215	8.2173 6.1466 7.4185

and

$$\mathbf{C}_{\alpha \mathbf{i}} = (\mathbf{A}_{\alpha \mathbf{i}}^{\mathbf{T}} \mathbf{V}_{\alpha_{\mathbf{i}\mathbf{i}}}^{-1} \mathbf{A}_{\alpha \mathbf{i}})^{-1}$$
(26)

and covariance  $\langle \delta \sigma_{ei} \delta \sigma_{ej} \rangle$  is obtained using Eq. (24):

$$<\!\delta\sigma_{ei}\delta\sigma_{ej}\!>=\!\mathbf{B}_{\alpha\mathbf{i}}^{\mathbf{T}}V_{\alpha_{ij}}\mathbf{B}_{\alpha\mathbf{j}}\;.$$
(27)

By substituting  $\langle \sigma_{ei} \rangle$  from Eq. (24) in Eq. (23),  $\chi^{2ai}_{\alpha i}$  can be obtained. For the present work,  $\langle \sigma_{ei} \rangle$  along with uncertainty (elements of vector  $\sigma_e$ ),  $\langle \delta \sigma_{ei} \delta \sigma_{ej} \rangle$  (elements of  $\mathbf{V}_{\sigma_e}$ ), and  $\chi^2 \equiv [\chi^2_{\alpha 1}, \chi^2_{\alpha 2}, \chi^2_{\alpha 3}]^T$  obtained are presented in Table XI. It can be observed in Table XI that  $\chi^2 \equiv [\chi^2_{\alpha 1}, \chi^2_{\alpha 2}, \chi^2_{\alpha 3}]^T = [8.2173, 6.1466, 7.4185]^T$ , which is greater than the required  $\frac{\chi^2_{\alpha i}}{n-f} = 1$  (n = 2, f = 1) for consistency; this is due to discrepant data ( $|\langle \sigma_{ei} \rangle - \langle \sigma_{ej} \rangle | \rangle > |\Delta \sigma_{ei} + \Delta \sigma_{ej}|$ ) (see column 2 of Table X). An ad hoc method to resolve the problem of discrepancy is scaling up<sup>19,20</sup> the elements of covariance matrix  $V_{\alpha_{ij}}$  by scaling factor  $\frac{\chi^2_{\alpha i}}{n-f}$ . An advanced method to deal with discrepant data can be found<sup>g</sup> in Refs. 20 and 21. Discussion of the issue of discrepant data is beyond the scope of the present investigation.

#### **IV. CONCLUSIONS**

The following conclusions may be made:

1. In the present work, the reaction cross section of  ${}^{58}\text{Ni}(n,p){}^{58}\text{Co}$  at effective neutron energies  $E_n = 5.89$ , 10.11, and 15.87 MeV are determined using activation and off-line gamma-ray spectrometry along with covariance analysis.

2. Table XI presents the evaluated mean values and covariances of the <sup>58</sup>Ni(*n*,*p*)<sup>58</sup>Co reaction cross section at effective neutron energies  $E_n = 5.89$ , 10.11, and 15.87 MeV.

3. We provide the measured cross sections with their partial uncertainties and correlation properties in a computer-readable form through the EXchange FORmat (EXFOR) library<sup>22</sup> (entry number 33076) following the new format rule introduced in Ref. 1. This helps, in principle, for anyone to generate the covariance matrix for the present work.

4. We believe that it is important for all nuclear experimental scientists to incorporate a detailed data reduction procedure, reduced data, and partial uncertainties in their publications, to the extent possible.

5. A detailed report presenting all data for the intermediate steps, not presented herein to save space, is available with the author.<sup>23</sup>

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NUCLEAR SCIENCE AND ENGINEERING VOL. 179 APR. 2015

<sup>&</sup>lt;sup>g</sup>See Sec. 2.6 of Ref. 20.

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