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Measurement of neutron induced reaction cross sections with covariance analysis

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May 25, Tuesday (14:40 – 15:00) – Oral talk

Abstract of the talk

In the presentation, I am going to talk about

- the experimental measurement details of the neutron activation cross-sections of (n, a) and (n,2n) reactions for copper and potassium at neutron energy 14.92 ± 0.02 MeV, followed by the discussion on covariance analysis which is used for the uncertainty quantification and to propagate the inter-correlation matrix between different reactions cross-section.

- And then in the last, I will talk about the results obtained from the present measurement and will discuss their comparison with literature data, theoretically predicted results (EMPIRE-3.2 and TALYS-1.9), and JEFF-3.1/A, TENDL-2019, JENDL-4.0, and ENDF/B-VIII.0 evaluated data.

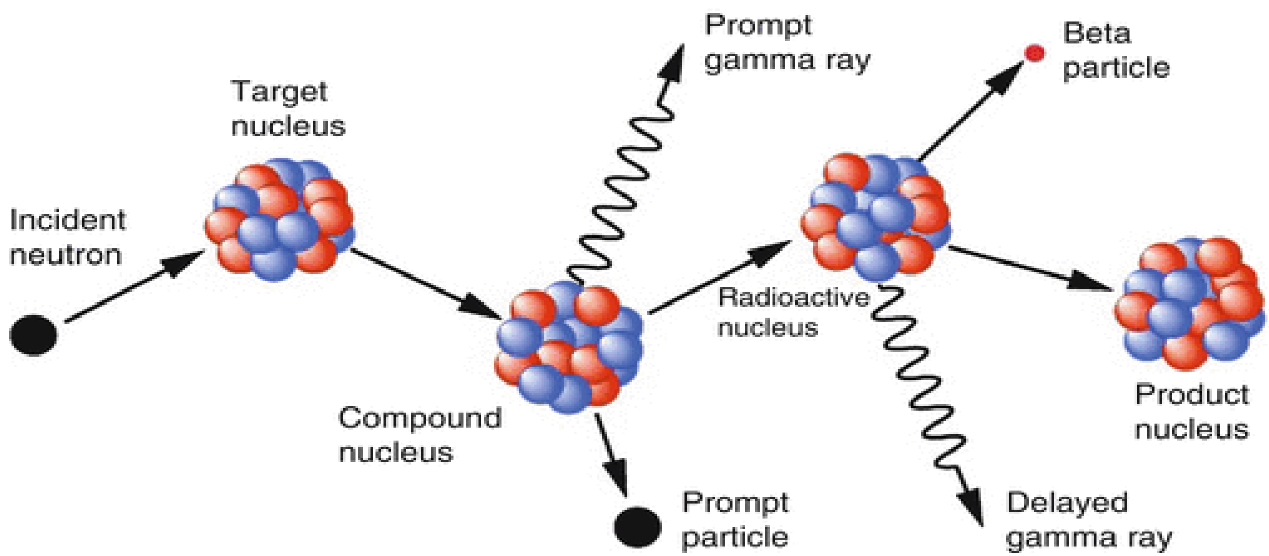
Outline of the talk

- Introduction
- Experimental Details
- Offline Gamma-Ray Spectroscopy
- Cross Section Determination
- Covariance Analysis
- Results and Discussion
- Acknowledgments

Introduction

➢Neutron induced reaction cross section is the quantitative value of interaction probability of neutron with the target nucleus.

➢Neutron activation is the most commonly used experimental technique for the cross section measurement.



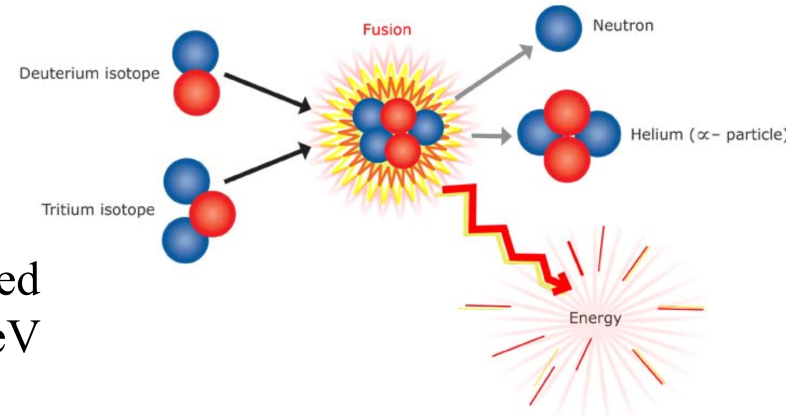
➢Investigation of neutron induced reactions cross section at the energy range about 14 MeV is important for the development of fusion reactor technology from the point of view of activation, radiation damage and mechanical stability of construction materials, etc.

➢Copper and potassium have been chosen for the present study because both the materials are important part of the reactor structural materials.

Experimental Details

➢The neutron-source use for the present work is

$$^{2}_{1}D + ^{3}_{1}T \rightarrow ^{4}_{2}He + ^{1}_{0}n,\ Q \sim +17.589\ MeV\ ;$$



This [d-t] reaction based neutron source is most commonly used neutron source to produce the mono energetic neutron at 14 MeV energy range.

➢The experiment has been performed using the Purnima neutron generator facility, BARC, Mumbai, India.

➢The D+ ions were accelerated to the 140 ± 5 keV and bombarded on the Ti-T target producing the neutrons in the forward direction with the flux value 9.419E+07 n/cm^2.s. The average deuteron beam current was 60 μA during sample irradiation. The energy of neutron and its uncertainty were calculated using the two-body kinematics.

➢For the normalization of the neutron flux, $^{27}Al(n,\alpha)^{24}Na$ reference reaction was used and its cross section retrieved from the IRDFF-1.05 library. The activation samples details of the present measurement are given below:

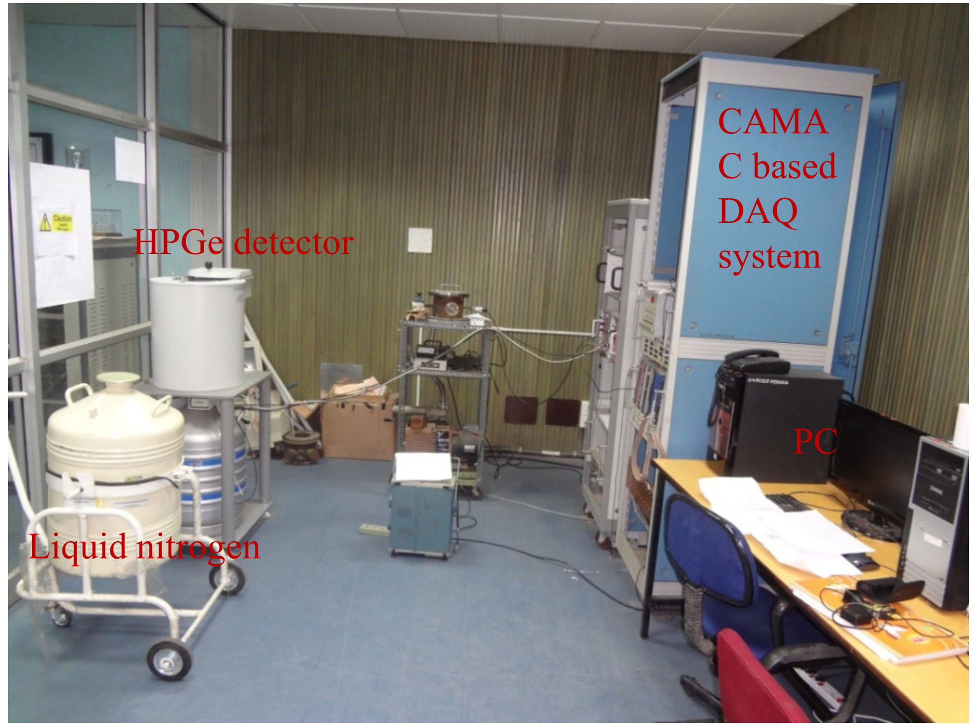
Isotope	Sample irradiated	Isotope abundance (%)	Thickness (cm)	density (g/cm^3)	Isotope weight in the sample (mg)	Number of target atoms (10^{-4} atoms/b)
^{41}K	K_2SO_4 powder	6.7302 ± 0.0044	0.2	2.66	370.5 ± 0.1	1.612
^{65}Cu	Cu metal sheet	30.85 ± 0.15	0.0125	8.96	116.5 ± 0.1	3.330
^{27}Al	Al foil	100	0.0025	2.70	22.5 ± 0.1	5.019

Offline Gamma-Ray Spectroscopy

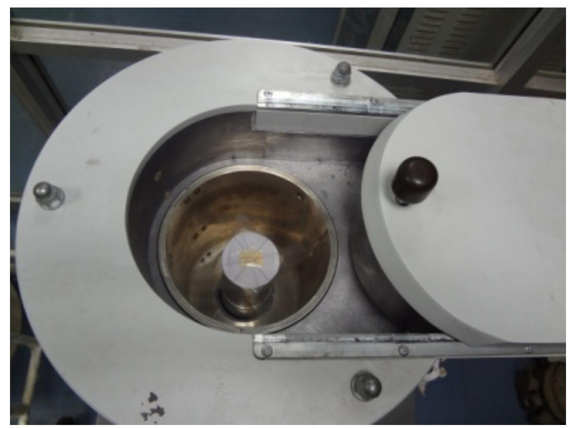
➢The induced activity of the irradiated samples were measured using a pre-calibrated lead-shielded 185-cc high purity germanium detector (HPGe) which having 30% relative efficiency and 1.8 keV energy resolution for 1.33 MeV γ-ray energy.

➢The data acquisition was carried out using the CAMAC-based Linux Advanced Multi-parameter System (LAMPS) Software.

➢The details of the nuclear decay data and their uncertainties used in the present experiment are given in Table below:

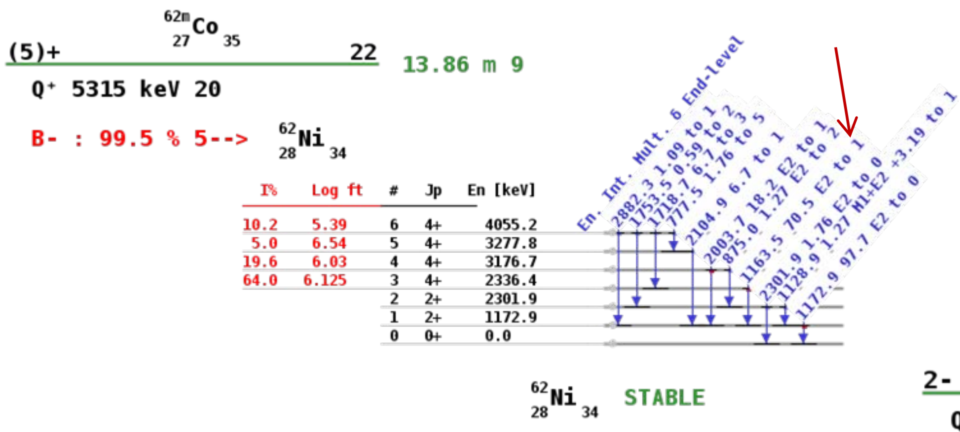


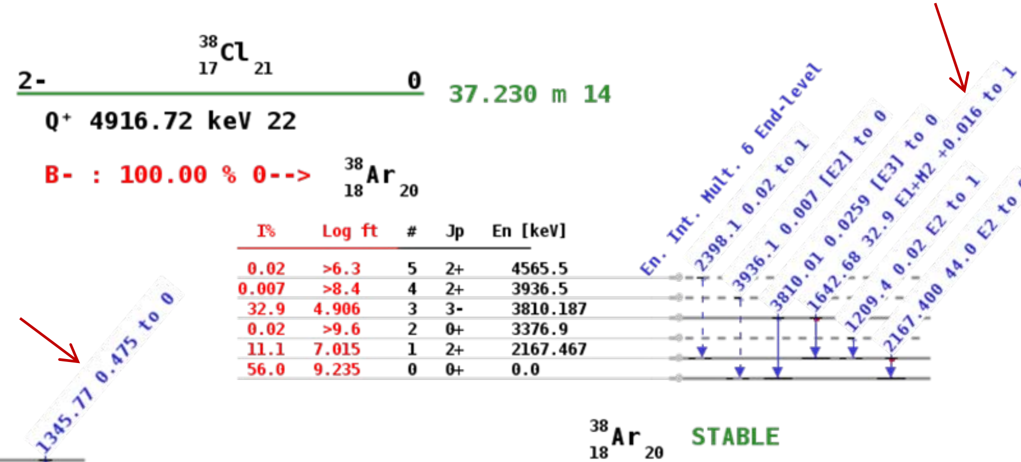
Reaction	Residue product	Half-life ($t_{1/2}$)	E_γ (keV)	I_γ (%)
$^{65}Cu(n,\alpha)$	^{62m}Co	13.86 ± 0.09 min	1163.50	70.5 ± 1.4
$^{41}K(n,\alpha)$	^{38}Cl	37.230 ± 0.014 min	1642.68	32.9 ± 0.5
$^{65}Cu(n,2n)$	^{64}Cu	12.701 ± 0.002 h	1345.77	0.475 ± 0.011
$^{27}Al(n,\alpha)$	^{24}Na	14.997 ± 0.012 h	1368.62	99.9936 ± 0.0015

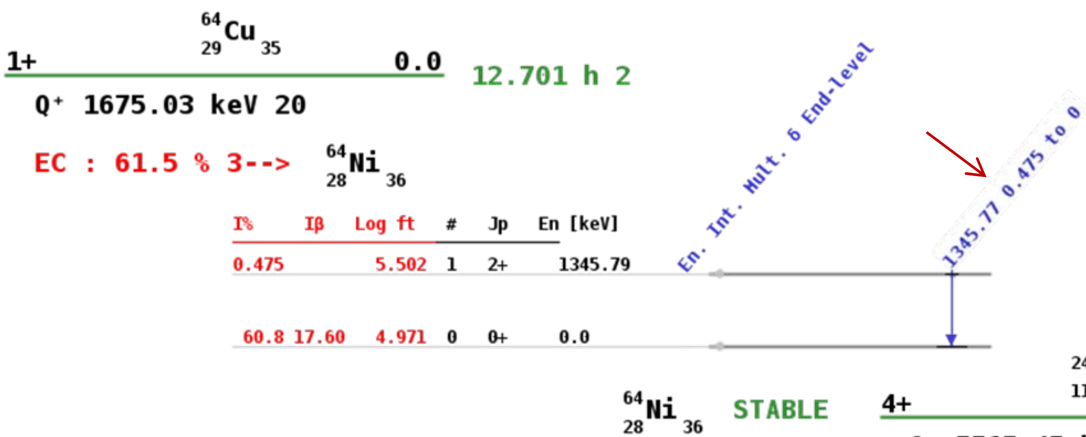


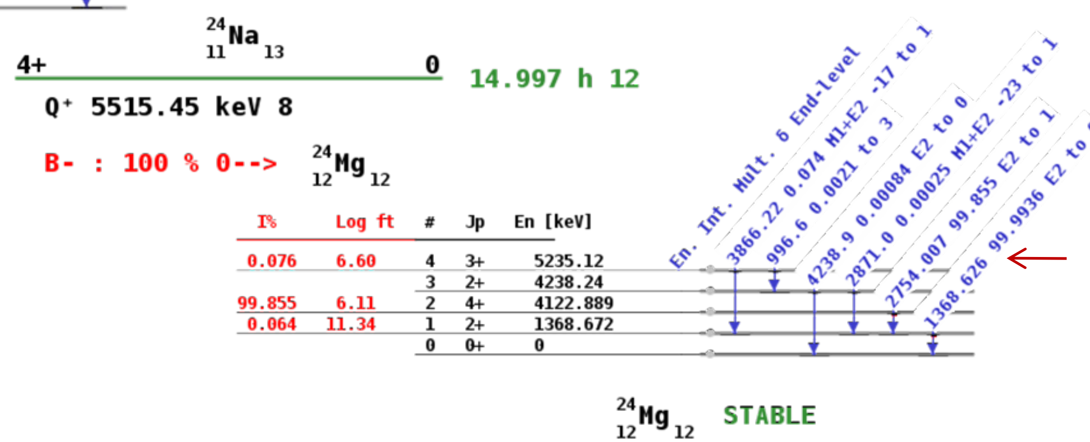
Irradiated sample was placed at top of the lead-shielded HPGe detector to get high count rate

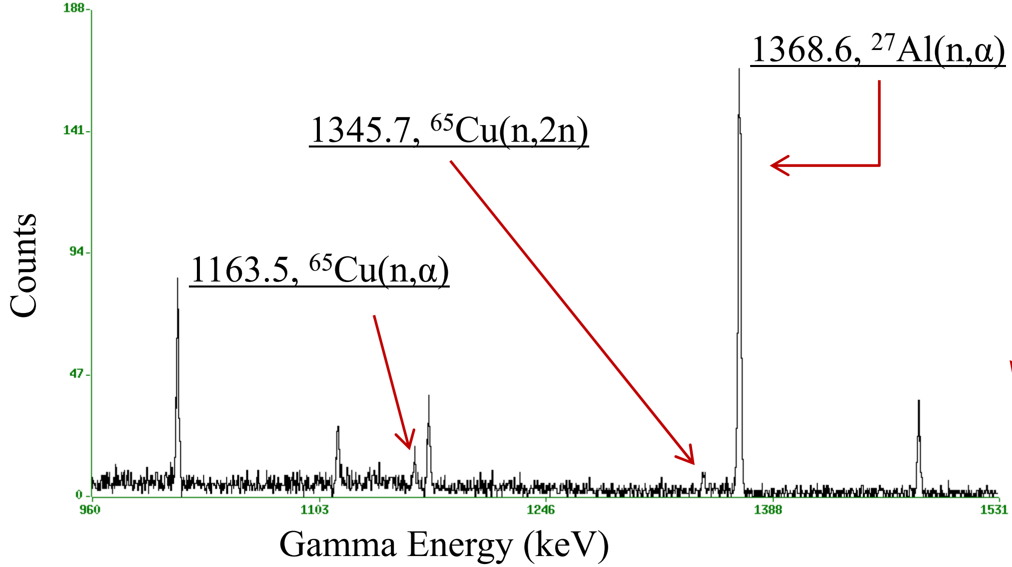
Decay scheme of radioactive nucleus



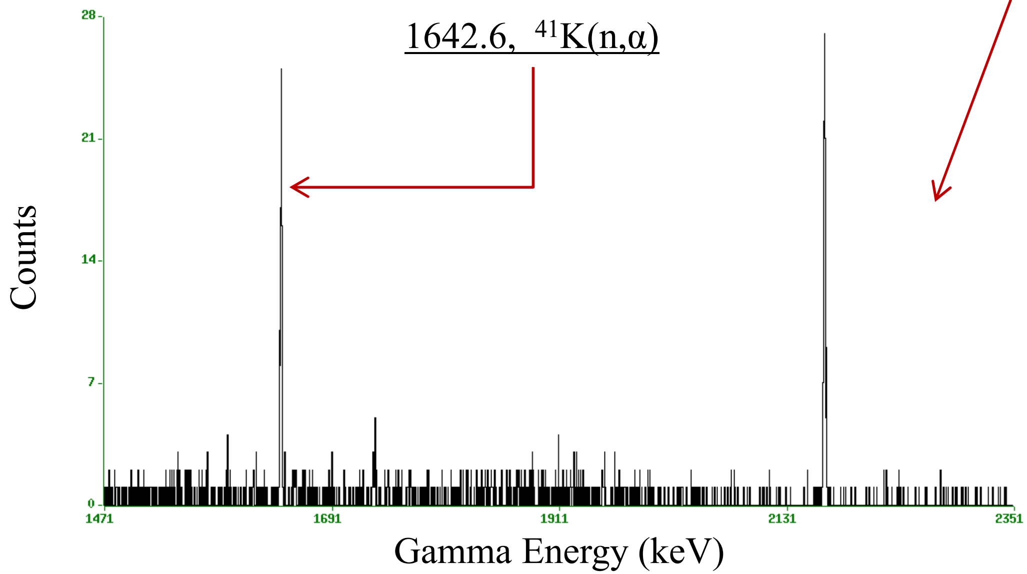








Copper + Aluminium gamma-ray spectra



Potassium gamma-ray spectra

- HPGe detected photo-peaks of characteristics gamma-ray produced from the residues of the ^{65}Cu(n,α)^{62m}Co, ^{65}Cu(n,2n)^{64}Cu, ^{27}Al(n,α)^{24}Na, and ^{41}K(n,α)^{38}Cl reactions .

HPGe detector efficiency calibration

➢The efficiency calibration of the HPGe detector has been determined using a standard ^{152}Eu point source ($T_{1/2}$ = 13.517 ±0.009 y, of known activity (A_0 = 6659.21 ± 81.60 Bq as on 1 Oct. 1999).

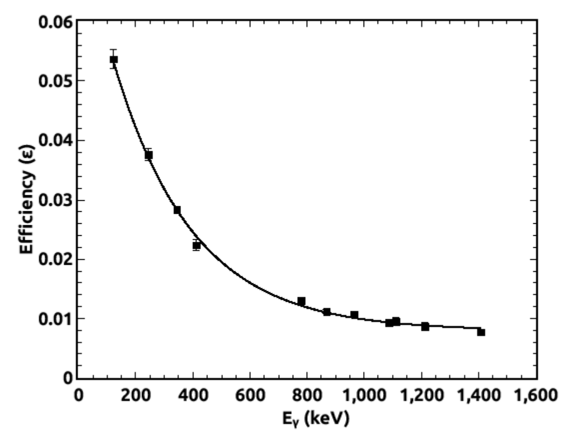
➢The efficiency (ε_p) of the point source placed at a distance of 2 mm from the detector absorber was determined by

$$\varepsilon_p = \varepsilon_I \varepsilon_G = \frac{C K_c \varepsilon_G}{A_0 e^{-\lambda t} \Delta t I_\gamma}$$

➢Since our samples have a finite area, therefore the efficiency for the point source geometry (ε_p) was transferred to the efficiency for sample geometry (ε) by using the Monte Carlo simulation code EFFTRAN and the same code also has been used to calculate the gamma-rays coincidence-summing correction factor (K_c).

➢The obtained efficiency values and uncertainty for each gamma-ray are given in table below and the value of (K_c) also has been given in table.

E_γ (keV)	I_γ	Counts (C)	K_c	ε_p	ε
121.78	0.2853 ± 0.0016	186555.4 ± 5118.3	1.165	0.053694	0.053318 ± 0.001630
244.69	0.0755 ± 0.0004	32729.2 ± 710.5	1.230	0.037583	0.037320 ± 0.000951
344.27	0.2659 ± 0.0020	95780.1 ± 1417.3	1.113	0.028258	0.028060 ± 0.000579
411.11	0.02238 ± 0.00013	5512.2 ± 231.0	1.288	0.022360	0.022203 ± 0.000978
778.90	0.1293 ± 0.0008	20438.3 ± 308.7	1.165	0.012979	0.012888 ± 0.000263
867.38	0.0423 ± 0.0003	5259.4 ± 178.7	1.274	0.011165	0.011086 ± 0.000408
964.05	0.1451 ± 0.0007	20001.3 ± 317.4	1.099	0.010677	0.010602 ± 0.000218
1085.83	0.1011 ± 0.0005	14414.6 ± 761.5	0.925	0.009295	0.009230 ± 0.000502
1112.94	0.1367 ± 0.0008	17657.6 ± 1248.9	1.045	0.009514	0.009447 ± 0.000680
1212.94	0.01415 ± 0.00008	1362.8 ± 79.1	1.265	0.008587	0.008527 ± 0.000508
1408.01	0.2087 ± 0.0009	21291.8 ± 391.1	1.069	0.007687	0.007633 ± 0.000171



Cross section Determination

➢the neutron activation cross sections were derived with respect to the $^{27}Al(n,\alpha)^{24}Na$ reference monitor cross section using the

$$\sigma_s = \sigma_{\rm Al} \frac{A_s \lambda_s a_{\rm Al} N_{\rm Al} I_{\gamma({\rm Al})} \varepsilon_{\rm Al} f_{\rm Al}}{A_{\rm Al} \lambda_{\rm Al} a_s N_s I_{\gamma(s)} \varepsilon_s f_s} \times \frac{C_{\rm attn.(s)}}{C_{\rm attn.(Al)}},$$

where
(f) is the timing factor calculated given by using equation

$$f = (1 - e^{-\lambda t_{\rm irr}}) e^{-\lambda t_{\rm cool}} (1 - e^{-\lambda t_{\rm count}}),$$

Reaction	t_{irr} (s)	t_{cool} (s)	t_{count} (s)
$^{65}Cu(n,\alpha)^{62m}Co$	8525	1468	250
$^{41}K(n,\alpha)^{38}Cl$	8525	2208	356
$^{65}Cu(n,2n)^{64}Cu$	8525	6620	693
$^{27}Al(n,\alpha)^{24}Na$	8525	11268	2030

➢(Cattn.) is the correction factor for γ -ray self-attenuation applied to the measured cross section. The mass attenuation coefficient was retrieved from the XMuDat version 1.0.1

$$C_{\rm attn.} = \frac{\mu_m d}{1 - \exp(-\mu_m d)},$$

Sample	E_γ (keV)	$C_{attn.}$
Cu	1163.50	1.0030
	1345.77	1.0028
K_2SO_4	1642.68	1.0013
Al	1368.62	1.0002

Covariance Analysis

- The covariance analysis is a mathematical tool based on the error estimation which provides the best estimation of the uncertainty along with the cross-correlations among the measured quantities, which in this present case, are the reaction cross-sections.

- In the present work, different reactions cross section have been measured at a neutron energy 14.92 ± 0.02 MeV, and as the counting of all the irradiated samples has been done with the same detector system and same monitor reaction cross section, therefore all the reaction cross sections are correlated with the efficiency and monitor cross section uncertainties.

- And in this case, the covariance analysis plays a vital role as it transfers the errors from each quantity i.e., efficiency, monitor cross section and other parameters such as Half-life, abundance, timing factor etc used in the calculations into the final uncertainties.

- The first part of the covariance analysis to obtain the total uncertainty and inter-correlation matrix between the different reactions cross sections is :

→HPGe detector efficiency uncertainty and the correlation coefficients:

- To obtain the efficiency for the gamma-rays of residues, we did the fitting of the measured efficiency by using the exponential fitting function. We further then used the values of the fitting parameters and its covariance matrix to calculated the residues gamma-rays efficiency value with its uncertainty and the covariance matrix.

$$\varepsilon(E_\gamma) = \varepsilon_o \exp(-E_\gamma/E_0) + \varepsilon_c,$$

Parameters	Value	Uncertainty	Correlation matrix		
ε_c	0.00792	3.67958×10^{-4}	1.0000		
ε_0	0.07047	0.0029	0.4765	1.0000	
E_0 (keV)	277.11562	14.62125	−0.7741	−0.8365	1.0000

➢The covariance between the two interpolated efficiencies $\varepsilon_{(i)}$ and $\varepsilon_{(j)}$ are propagated by using the covariance's of three fitting parameters following the prescription by Mannhart

$$\begin{aligned}\mathrm{Cov}(\epsilon(E_i), \epsilon(E_j)) &= e^{-\frac{E_i+E_j}{E_0}}(\Delta\epsilon_0)^2 + \frac{\epsilon_0^2 E_i E_j}{E_0^4} e^{-\frac{E_i+E_j}{E_0}}(\Delta E_0)^2 + (\Delta\epsilon_c)^2 \\ &+ \epsilon_0 \frac{E_i + E_j}{E_0^2} e^{-\frac{E_i+E_j}{E_0}} \mathrm{Cov}(\epsilon_0, E_0) \\ &+ \left(e^{-\frac{E_i}{E_0}} + e^{-\frac{E_j}{E_0}}\right)\mathrm{Cov}(\epsilon_0, \epsilon_c) \\ &+ \frac{\epsilon_0}{E_0^2}\left(E_i e^{-\frac{E_i}{E_0}} + E_j e^{-\frac{E_j}{E_0}}\right)\mathrm{Cov}(E_0, \epsilon_c),\end{aligned}$$

Where the uncertainty in the detection efficiency is

$$(\Delta\epsilon_i)^2 = \mathrm{Cov}(\epsilon(E_i), \epsilon(E_i)).$$

And the correlation coefficient is:

$$\mathrm{Cor}(\varepsilon(E_i),\varepsilon(E_j)) = \mathrm{Cov}(\varepsilon(E_i),\varepsilon(E_j))/(\Delta\varepsilon_i).(\Delta\varepsilon_j)),$$

Table: Interpolated efficiency of the HPGe detector for the corresponding gamma-ray energy of the samples and monitor reactions with their uncertainty and their correlation matrix.

Reaction	E_γ (keV)	Efficiency	Correlation matrix			
^{65}Cu$(n,\alpha)^{62m}$Co	1163.50	0.00897 ± 0.00023	1.0000			
^{41}K$(n,\alpha)^{38}$Cl	1642.68	0.00810 ± 0.00032	0.9125	1.0000		
^{65}Cu$(n,2n)^{64}$Cu	1345.77	0.00846 ± 0.00027	0.9679	0.9860	1.0000	
^{27}Al$(n,\alpha)^{24}$Na	1368.62	0.00842 ± 0.00028	0.9626	0.9892	0.9997	1.0000

→ Uncertainity in timing factor Δf/f

- For the timing factor (f) as discussed previous given by equation

$$f = (1 - e^{-\lambda t_{\text{irr}}}) e^{-\lambda t_{\text{cool}}} (1 - e^{-\lambda t_{\text{count}}}),$$

And the uncertainties in this timing factors for samples and monitor reactions were propagated from the uncertainties in the decay constants (λ) by

$$(\Delta f/f)^2 = s_{f\lambda}^2 (\Delta\lambda/\lambda)^2$$

($f = f_x$ or f_r, and $\lambda = \lambda_x$ or λ_r) with the relative sensitivity $s_{f\lambda}$

$$s_{f\lambda} = \frac{\lambda}{f}\frac{\partial f}{\partial \lambda} = \left(\frac{\lambda t_i e^{-\lambda t_i}}{1 - e^{-\lambda t_i}} - \lambda t_c + \frac{\lambda t_m e^{-\lambda t_m}}{1 - e^{-\lambda t_m}} - 1 \right).$$

Where the uncertainty in the decay constant

$$\Delta\lambda = (\ln 2 \Delta T_{1/2}) / T_{1/2}^2$$

can be obtained from $\Delta T_{1/2}$ in the ENSDF Library.

Table: Fractional uncertainties (%) of all attributes associated with the different reactions cross section measured at neutron energy 14.92 ± 0.02 MeV.

attributes	fractional uncertainties (%)		
	$^{65}Cu(n,\alpha)^{62m}Co$	$^{41}K(n,\alpha)^{38}Cl$	$^{65}Cu(n,2n)^{64}Cu$
(x)	(1)	(2)	(3)
A_s	13.7361	8.7706	10.1015
A_m	1.1311	1.1311	1.1311
I_s	1.9858	1.5197	2.3157
I_m	0.0015	0.0015	0.0015
N_s	0.0858	0.0270	0.0858
N_m	0.4444	0.4444	0.4444
a_s	0.4862	0.0653	0.4862
ε_s	2.5641	3.9506	3.1914
ε_m	3.3254	3.3254	3.3254
f_s	0.2526	0.0134	0.0004
f_m	0.0054	0.0054	0.0054
σ_m	0.3644	0.3644	0.3644
Total error (%)	14.57	10.37	11.42

→ Correlation between the different reactions cross section

➢The correlation coefficient of two parameters (x_1, x_2) is represented as uncorrelated [Cor(x_1, x_2) = 0] and fully correlated coefficient [Cor(x_1, x_2) = 1]. However, the numerical value of the correlation coefficient must be between $-1 \leq$ Corr(x_1, x_2) ≤ 1 and it occurs when x_1 and x_2 are determined not independently, but still x_2 is not automatically determined from x_1 (in present case, we have efficiency uncertainty).

Table: Correlation coefficient between the different attributes associated with the different reactions cross section measured at neutron energy 14.92 ± 0.02 MeV.

Correlation coefficient													
(x,x)	A_s	A_m	I_s	I_m	N_s	N_m	a_s	ε_s	ε_m	f_s	f_m	σ_m	
(1,1)	1	1	1	1	1	1	1	1	1	1	1	1	1.0000
(1,2)	0	1	0	1	0	1	0	0.9125	1	0	1	1	0.1451
(1,3)	0	1	0	1	1	1	1	0.9679	1	0	1	1	0.1237
(2,2)	1	1	1	1	1	1	1	1	1	1	1	1	1.0000
(2,3)	0	1	0	1	0	1	0	0.9860	1	0	1	1	0.2119
(3,3)	1	1	1	1	1	1	1	1	1	1	1	1	1.0000

➢The fractional variance and covariance and hence the correlation coefficients between each reactions cross section are constructed using the following equations

$$\mathrm{cov}(\Delta x_i, \Delta x_j) = \sum_i \sum_j \Delta x_i \Delta x_j cor(\Delta x_i, \Delta x_j)$$

$$\mathrm{var}(\Delta x_i) = \sum_i (\Delta x_i)^2$$

$$\text{Correlation} = \mathrm{Cov}(\Delta x_i, \Delta x_j)/[\mathrm{Var}(\Delta x_i).\ \mathrm{Var}(\Delta x_j)]^{1/2}$$

Results and Discussion

➢The experimentally measured cross sections result for all the three reactions at neutron energy 14.92 ± 0.02 MeV along with their total uncertainty and the correlation matrix are summarized in table below and presented in Fig. 1-3

Reaction	Present data $[\sigma_s]$	Correlation matrix		
$^{65}Cu(n,\alpha)^{62m}Cu$	0.00404 ± 0.00059	1.0000		
$^{41}K(n,\alpha)^{38}Cl$	0.02060 ± 0.00214	0.1451	1.0000	
$^{65}Cu(n,2n)^{64}Cu$	1.03082 ± 0.11776	0.1237	0.2119	1.0000

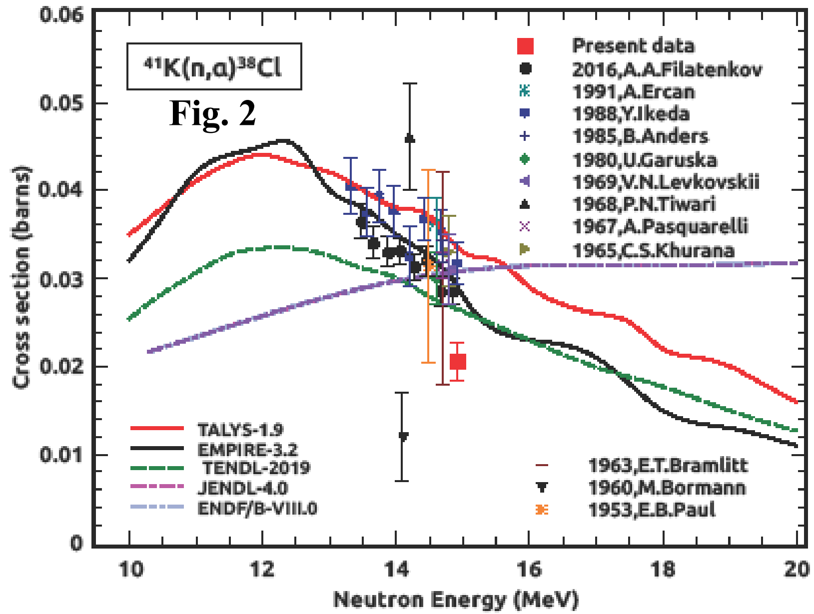


Fig. 2

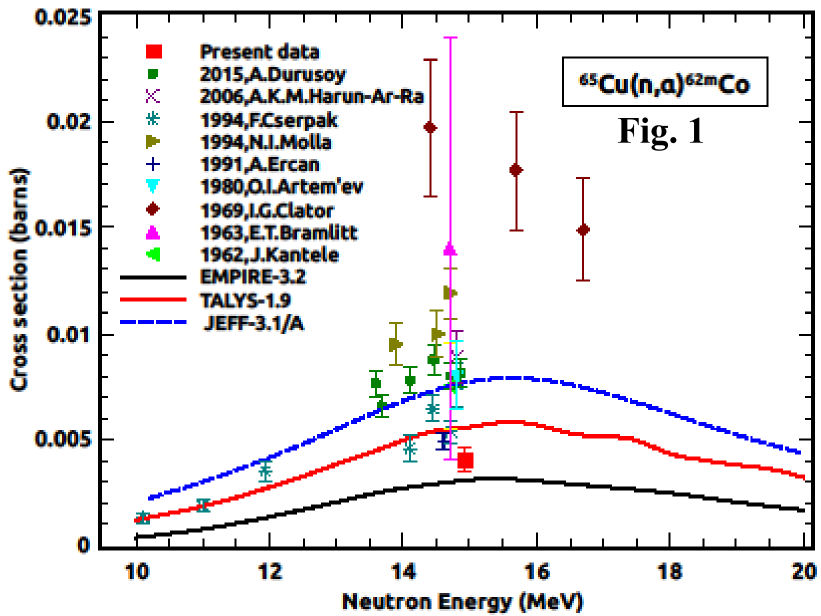


Fig. 1

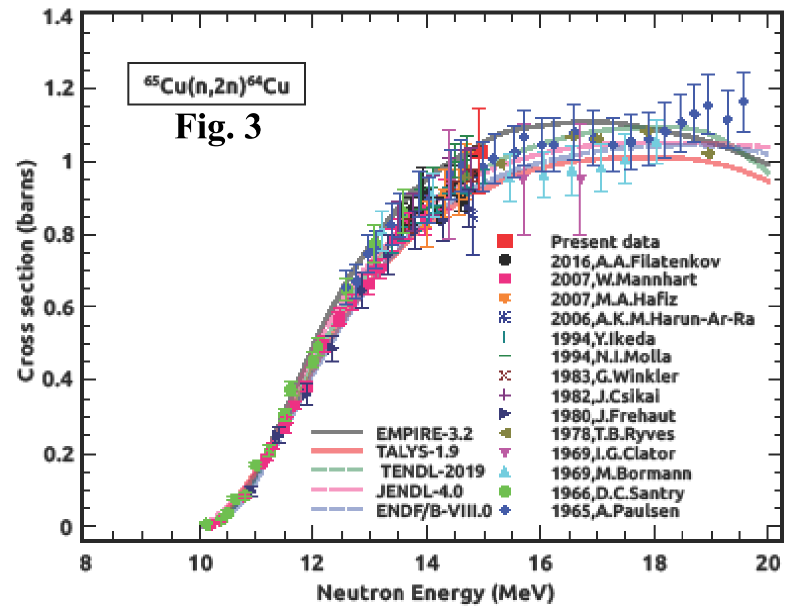


Fig. 3

Acknowledgements

➢ I would like to thanks all the collaborators who helped in making this work possible.

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Thank you
very much
for
your kind
attention