Validation of $(n, n'\gamma)$ data for the GRIN project

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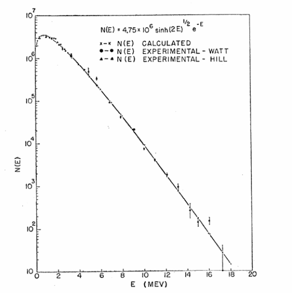
The birth and fate of the neutron

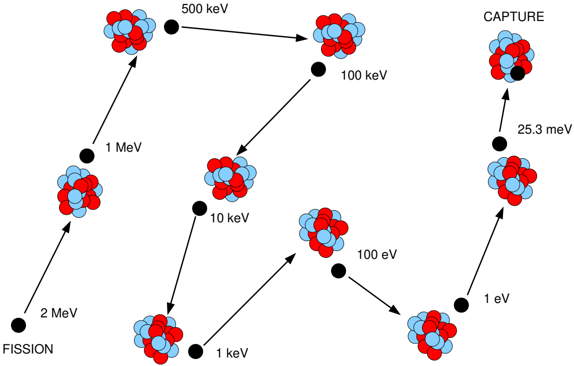


B.E. Watt, Phys. Rev. **87**, 1037 (1952).

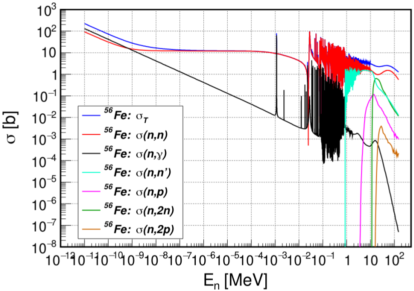
- Neutrons are born fast, e.g., ^{235}U fission.
- Watt proposed empirical relationship (0.075-17 MeV):

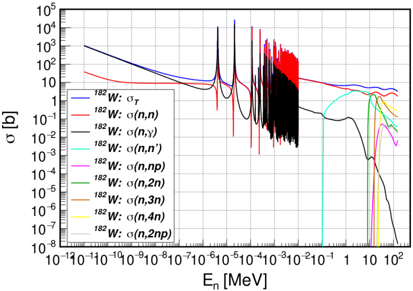
$$N(E_n) = 4.75\times10^6 \sinh\sqrt{(2E_n)}\exp(-E_n).$$

- Negligible energy loss/scattering ($< 1\%$).



Neutron interactions and contributions to σ_F in ^{56}Fe and ^{182}W as function of E_n





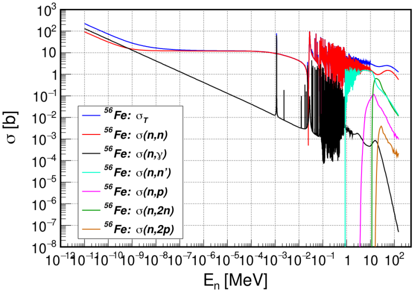
In ^{56}Fe, at $E_n = 25.3$ meV:

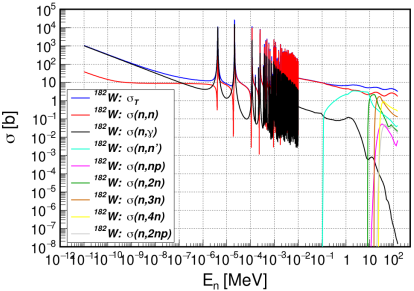
$$\sigma_F(n +^{56}\mathrm{Fe}; E_n) = \sigma(n,\gamma).$$

In ^{182}W, at $E_n = 25.3$ meV:

$$\sigma_F(n +^{182}\mathrm{W}; E_n) = \sigma(n,\gamma).$$

Neutron interactions and contributions to σ_F in ^{56}Fe and ^{182}W as function of E_n





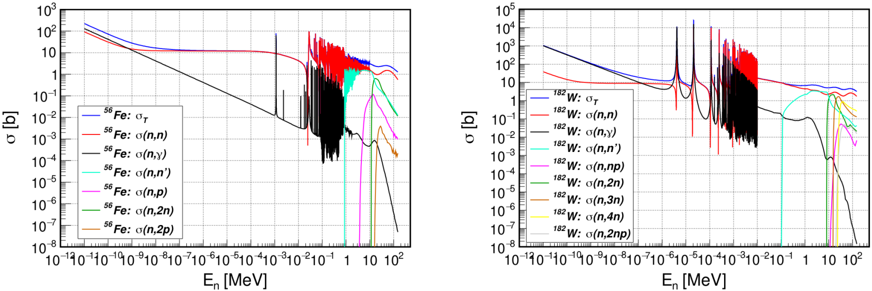
In ^{56}Fe, at $E_n = 14$ MeV:

$$\sigma_F(n +^{56}\text{Fe}; E_n) = \sigma(n,\gamma) + \sigma(n,n') + \sigma(n,p) + \sigma(n,2n) + \sigma(n,2p).$$

In ^{182}W, at $E_n = 14$ MeV:

$$\begin{aligned} \sigma_F(n +^{182}\text{W}; E_n) &= \sigma(n,\gamma) + \sigma(n,n') + \sigma(n,np) + \sigma(n,2n) + \sigma(n,3n) \\ &+ \sigma(n,4n) + \sigma(n,2np). \end{aligned}$$

Neutron interactions and contributions to σ_F in ^{56}Fe and ^{182}W as function of E_n



In ^{56}Fe, at $E_n = 0.86 \sim 10$ MeV:

$$\sigma_F(n + ^{56}\text{Fe}; E_n) = \sigma(n,\gamma) + \sigma(n,n') + \sigma(n,p).$$

In ^{182}W, at $E_n = 0.10 \sim 10$ MeV:

$$\sigma_F(n + ^{182}\text{W}; E_n) = \sigma(n,\gamma) + \sigma(n,n') + \sigma(n,2n).$$

Reactor neutrons: ^{235}U fission neutron spectrum

Different empirical relations have been proposed to describe the energy spectrum of reactor neutrons:

- **Watt:**

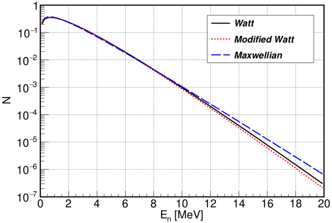
$$N(E_n) = \sqrt{\frac{2}{\pi e}} \exp(-E_n) \sinh \sqrt{(2E_n)}$$

- **Modified Watt:**

$$N(E_n) = A \exp(-bE_n) \sinh \sqrt{(cE_n)}$$

- **Maxwellian:**

$$N(E_n) = 2\sqrt{\frac{E_n}{\pi k T^3}} \exp\left(\frac{-E_n}{kT}\right)$$



Best-fit parametrizations deduced for ^{235}U:

- Modified Watt: $A = 0.4527$, $b = 1.036$, $c = 2.29$.
- Maxwellian: $kT = 1.290$ MeV.

The Baghdad IRT-M Reactor and $(n, n'\gamma)$ data

https://nucleardata.berkeley.edu/atlas/

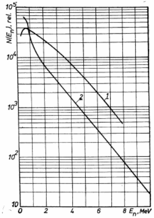


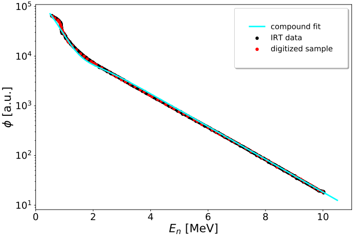
Рис. 5
Спектр нейтронов деления (*1*) и нейтронов от реактора ИРТ (*2*)

Fig. 5
Fission neutron spectrum (*1*) and the IRT reactor neutron spectrum (*2*)

- Compilation of energy-integrated inelastic neutron-scattering $(n, n'\gamma)$ data disseminated in book format.
- $\sim$ 7000 γ rays (E_γ and BR) from 105 samples: 76 natural and 29 isotopically-enriched targets.
- Ge(Li) viewing filtered fast-neutron beam line at the IRT-M Reactor: NRI, Baghdad, Iraq.
- Unique ^{56}Fe 847-keV $2_1^+ \rightarrow 0_{\mathrm{gs}}^+$ γ-ray normalization.
- Out-of-print book (*out-of-print* reactor!).
- Now Digitized database, open source dissemination.
- Project maintained on GitHub:

  github.com/AaronMHurst/baghdad_atlas
- A.M. Hurst *et al.*, NIMA **995**, 165095 (2021).

Characterization the IRT-M Baghdad Reactor neutron flux



$E_n < 1.5$ MeV (Maxwellian):

$$\phi_1(E_n) = 2A_1\sqrt{\left(\frac{E_n}{\pi k T^3}\right)}\exp\left(\frac{E_n}{kT}\right).$$

$E_n \geq 1.5$ MeV (Exponential):

$$\phi_2(E_n) = A_2\exp(-\beta E_n).$$

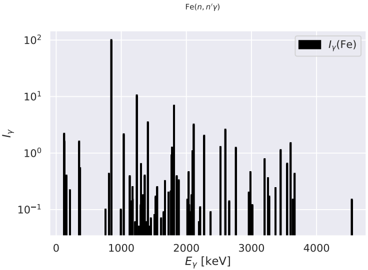
Overall fit according to parametrization of IRT-M data:

$$\phi(E_n) = \phi_1(E_n) + \left[\frac{1+\tanh[K(E_n-1.5)]}{2}\right](\phi_2(E_n) - \phi_1(E_n)).$$

$$\phi(E_n \ll 1.5\text{ MeV}) \to \phi_1(E_n);$$
$$\phi(E_n \gg 1.5\text{ MeV}) \to \phi_2(E_n).$$

Experimental Fe$(n, n'\gamma)$ γ-ray spectrum at the IRT-M

https://github.com/AaronMHurst/baghdad_atlas



Most important γ rays in ^{56}Fe from integral fission-neutron spectrum:

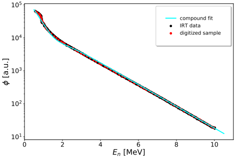
E_γ [keV]	$J_i^{\pi_i} \to J_f^{\pi_f}$	I_γ	B_A
846.8	$2_1^+ \to 0_{gs}^+$	100	1.0
1238.3	$4_1^+ \to 2_1^+$	10.5(5)	0.105(5)
1810.8	$2_2^+ \to 2_1^+$	6.9(4)	0.069(4)

$$B_A = \frac{I_\gamma(E_\gamma)}{I_\gamma(E_\gamma = 846.8)} = \frac{I_\gamma(E_\gamma)}{100}.$$

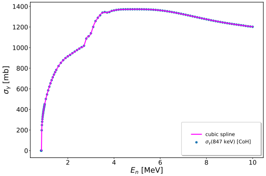
- Well-characterized flux should reproduce measured experimental data.
- Determine flux-weighted cross section ($\langle\sigma_\gamma\rangle$) by convolving $\sigma_\gamma(E_n)$ with $\phi(E_n)$ and compare to corresponding *Baghdad Atlas* branching ratios.
- Parameterized Baghdad Reactor neutron-flux distribution yields $\langle\sigma_\gamma\rangle$ values that reproduces measured integral B_A to within $\sim 1.5\sigma$.

Determination of $\langle\sigma_\gamma(E_\gamma = 846.8\text{ keV})\rangle$ in ^{56}Fe

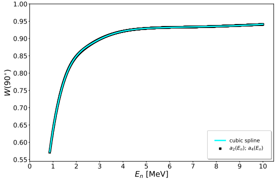
Fast-reactor flux: $\phi(E_n)$



Cross section: $\sigma_\gamma(E_n)$



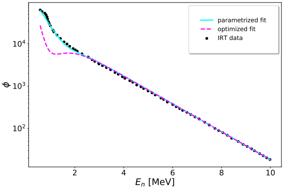
Angular distribution: $W_\gamma(E_n)$



$$\langle\sigma_\gamma(E_\gamma)\rangle = \frac{\int_{E_n=0.862}^{E_n=10} \phi(E_n)\sigma_\gamma(E_n)W_\gamma(\theta = 90^\circ; E_n)dE_n}{\int_{E_n=0}^{E_n=+\infty} \phi(E_n)dE_n}$$

- $\phi(E_n)$: Parameterized and adjusted for kT ($\phi(E_n \ll 1.5\text{ MeV}) \rightarrow$ Maxwellian; $\phi(E_n \gg 1.5\text{ MeV}) \rightarrow$ exponential).
- $\sigma_\gamma(E_n)$: γ-ray production data as function of E_n from reaction model, e.g., CoH$_3$, EMPIRE, or a nuclear data library, e.g., ENDF.
- $W_\gamma(\theta = 90^\circ; E_n)$: Experimental anisotropy-attenuation coefficients, a_2 and a_4 (not always available!).

Neutron-flux optimization: kT-adjustment to ^{56}Fe data



Reminder: the uncorrelated χ^2:

$$\chi^2 = \sum_{i=1}^{N} \frac{[y_i - f(x_i)]^2}{\sigma_i^2}.$$

Adjust kT in χ^2 minimization to find optimal flux:

$$\sum_{i=1}^{N}\sum_{j=1}^{N}[B_{A_i} - B_{kT_i}][V_{ij}^{-1}][B_{A_j} - B_{kT_j}].$$

or in matrix notation:

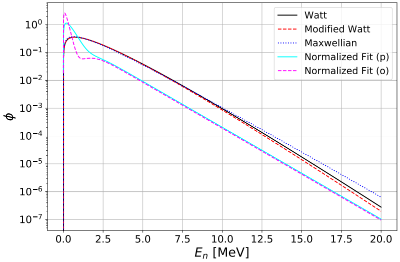
$$\chi^2 = (\boldsymbol{B_A} - \boldsymbol{B_{kT}})\boldsymbol{V}^{-1}(\widetilde{\boldsymbol{B_A}} - \widetilde{\boldsymbol{B_{kT}}}).$$

- $V \Rightarrow$ covariance matrix $\because I_\gamma$ are correlated.
- $N = 3 \Rightarrow$ number of γ rays.
- $\text{ndf} = 3 - 1 = 2$.
- $\chi^2/\text{ndf} \approx 0.35$ for $\text{ndf} = 2$.
- Correlation coefficient in range $0 < \rho_{ij} \lesssim 0.75$ reproduces expected χ^2/ndf consistent with $kT = 0.155(30)$ MeV (cf. $kT = 1.290$ MeV for pure Maxwellian ^{235}U fission.)

Normalized fits cf. standard ^{235}U neutron-flux distributions

Flux must satisfy normalization condition:

$$\int_{-\infty}^{+\infty} \phi^*(E_n)\phi(E_n)dE_n = 1$$



Expectation energies:

- Watt $\langle E_n \rangle = 2.00$ MeV.
- Modified Watt $\langle E_n \rangle = 1.98$ MeV.
- Maxwellian $\langle E_n \rangle = 1.94$ MeV.
- Parameterized Flux at IRT-M $\langle E_n \rangle = 0.88$ MeV.
- Optimized Flux at IRT-M $\langle E_n \rangle = 0.63$ MeV.

kT-adjusted optimized neutron flux used to deduce flux-weighted quantities.

Optimized flux-weighted cross sections

Values for $\langle\sigma_\gamma\rangle$ and $\langle\sigma_\gamma\rangle_W$ deduced according to fitted flux using compound function with optimized $kT = 0.155$ MeV:

E_γ [keV]	B_A	B_{kT}	$\langle\sigma_\gamma\rangle$ [mb]	$\langle\sigma_\gamma\rangle_W$ [mb]	$\langle\sigma_\gamma\rangle_{\mathrm{FRM}}$ [mb]	$\langle\sigma_\gamma\rangle_S$ [mb]
846.8	1.0	1.0	166(34)	143(29)	586(41)	521(106)
1238.3	0.105(5)	0.096(27)	15.9(31)	13.7(27)	58(5)	49.9(98)
1810.8	0.069(4)	0.061(18)	10.0(21)	8.7(18)	37(3)	31.7(66)

Our cross sections for ^{56}Fe are consistent with the recent FRM-II measurement* upon scaling by $\langle E_n\rangle$ at the two facilities.

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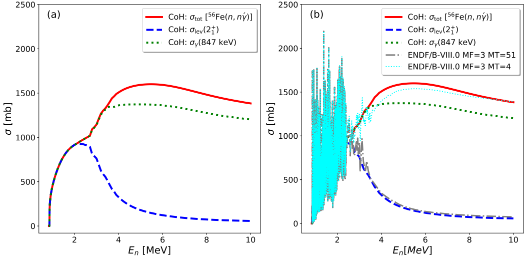
*Z. Ilic *et al.*, J. Radioanal. Nucl. Chem. **325**, 641 (2020).

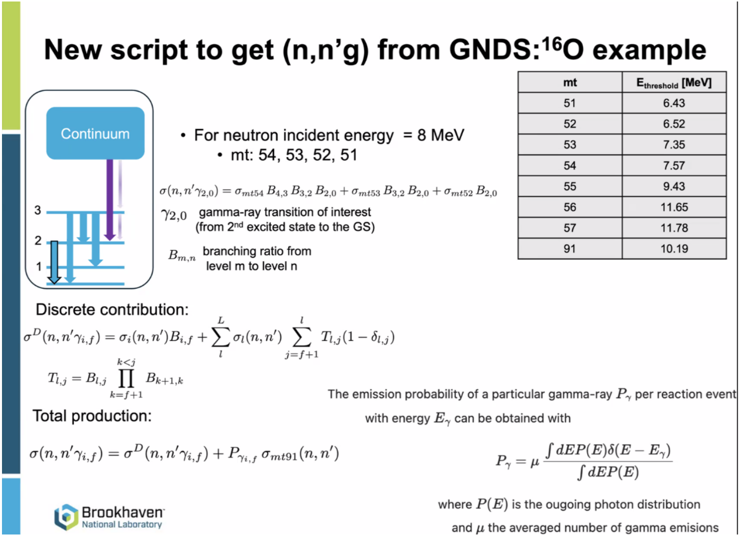
Why not take σ_γ directly from ENDF rather than a model?



	CoH	ENDF
$\langle\sigma_{\rm lev}\rangle$	113.0	101.6
$\langle\sigma_{\rm tot}\rangle$	171.8	163.6
$\frac{\langle\sigma_{\rm lev}\rangle}{\langle\sigma_{\rm tot}\rangle}$	0.658	0.621
$\langle\sigma_\gamma\rangle$	165.7	157.8

- σ_γ cannot be extracted directly from ENDF.
- Results indicate: $\frac{\langle\sigma_{\rm lev}^{\rm CoH}\rangle}{\langle\sigma_{\rm tot}^{\rm CoH}\rangle} \approx \frac{\langle\sigma_{\rm lev}^{\rm ENDF}\rangle}{\langle\sigma_{\rm tot}^{\rm ENDF}\rangle}$.
- Reasonable to expect ratios of partial γ-ray production cross section to total inelastic cross section to also be in agreement, i.e., $\langle\sigma_\gamma^{\rm ENDF}\rangle \approx \langle\sigma_{\rm tot}^{\rm ENDF}\rangle \frac{\langle\sigma_\gamma^{\rm CoH}\rangle}{\langle\sigma_{\rm tot}^{\rm CoH}\rangle}$.

Emanuel Chimanski's method for σ_γ extraction from ENDF



New script to get (n,n'g) from GNDS:^{16}O example

- For neutron incident energy = 8 MeV
  - mt: 54, 53, 52, 51

$$\sigma(n,n'\gamma_{2,0}) = \sigma_{mt54}\,B_{4,3}\,B_{3,2}\,B_{2,0} + \sigma_{mt53}\,B_{3,2}\,B_{2,0} + \sigma_{mt52}\,B_{2,0}$$

$\gamma_{2,0}$ gamma-ray transition of interest (from 2nd excited state to the GS)

$B_{m,n}$ branching ratio from level m to level n

mt	$E_{threshold}$ [MeV]
51	6.43
52	6.52
53	7.35
54	7.57
55	9.43
56	11.65
57	11.78
91	10.19

Discrete contribution:

$$\sigma^D(n,n'\gamma_{i,f}) = \sigma_i(n,n')B_{i,f} + \sum_{l}^{L}\sigma_l(n,n')\sum_{j=f+1}^{l}T_{l,j}(1-\delta_{l,j})$$

$$T_{l,j} = B_{l,j}\prod_{k=f+1}^{k<j}B_{k+1,k}$$

Total production:

$$\sigma(n,n'\gamma_{i,f}) = \sigma^D(n,n'\gamma_{i,f}) + P_{\gamma_{i,f}}\,\sigma_{mt91}(n,n')$$

The emission probability of a particular gamma-ray P_γ per reaction event with energy E_γ can be obtained with

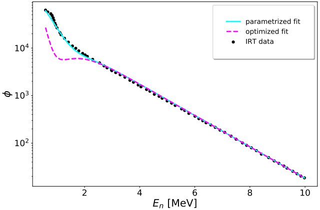
$$P_\gamma = \mu\frac{\int dEP(E)\delta(E-E_\gamma)}{\int dEP(E)}$$

where $P(E)$ is the ougoing photon distribution and μ the averaged number of gamma emisions

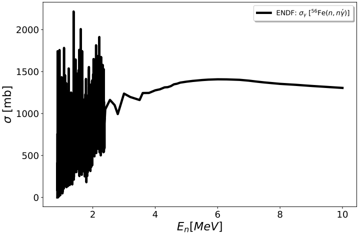
Brookhaven National Laboratory

Determination of $\langle\sigma_\gamma\rangle$ derived from ENDF for ^{56}Fe γ rays

Fast-reactor flux: $\phi(E_n)$



ENDF cross section: $\sigma_\gamma(E_n)$



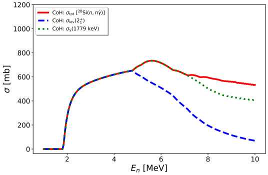
$$\langle\sigma_\gamma(E_\gamma)\rangle = \frac{\int_{E_n=0.862}^{E_n=10} \phi(E_n)\sigma_\gamma(E_n)dE_n}{\int_{E_n=0}^{E_n=+\infty} \phi(E_n)dE_n}$$

E_γ [keV]	$\langle\sigma_\gamma\rangle$ [mb] CoH	$\langle\sigma_\gamma\rangle$ [mb] ENDF
846.8	166(34)	160.3
1238.3	15.9(31)	13.5
1810.8	10.0(21)	9.4

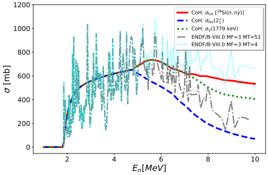
Values of $\langle\sigma_\gamma\rangle$ deduced directly from ENDF are consistent with value from well-tuned CoH$_3$ reaction-model calculation.

Flux-weighted validation for ^{28}Si$(n, n'\gamma)$: 1779 keV

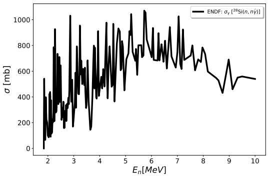
CoH$_3$



CoH$_3$ cf. ENDF



Derived σ_γ(1779) from ENDF

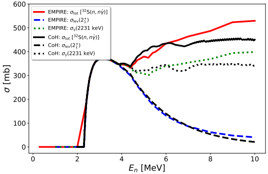


Source	$\langle\sigma_\gamma\rangle$ [mb]	$\langle\sigma_{\rm lev}\rangle$ [mb]	$\frac{\langle\sigma_{\rm lev}\rangle}{\langle\sigma_{\rm tot}\rangle}$
CoH	55.69	53.45	0.957
ENDF (estimate)	48.17	46.59	0.965
ENDF (derived)	49.40	—	—
Baghdad Atlas	**47.1(94)**	—	—

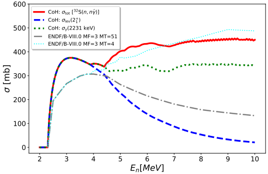
Integral measurement in agreement with 3 different validation results.

Flux-weighted validation for $^{32}\mathrm{S}(n, n'\gamma)$: 2231 keV

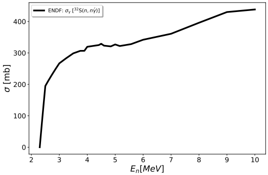
CoH$_3$ cf. EMPIRE



CoH$_3$ cf. ENDF



Derived $\sigma_\gamma(2231)$ from ENDF

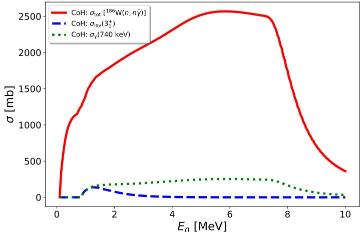


Source	$\langle\sigma_\gamma\rangle$ [mb]	$\langle\sigma_{\mathrm{tot}}\rangle$ [mb]	$\langle\sigma_{\mathrm{lev}}\rangle$ [mb]	$\frac{\langle\sigma_{\mathrm{lev}}\rangle}{\langle\sigma_{\mathrm{tot}}\rangle}$
CoH	26.57	27.91	23.95	0.858
EMPIRE	26.49	28.80	23.78	0.890
ENDF (estimate)	21.67	22.76	19.98	0.878
ENDF (derived)	21.79	–	–	–
Baghdad Atlas	**25.0(60)**	–	–	–

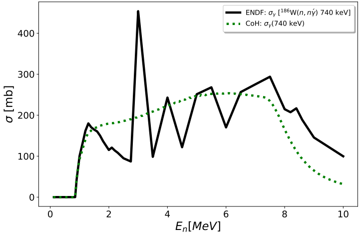
Integral measurement in agreement with 4 different validation results.

Flux-weighted validation for ^{186}W$(n, n'\gamma)$: 740 keV

CoH$_3$



CoH$_3$ cf. Derived $\sigma_\gamma(740)$ from ENDF



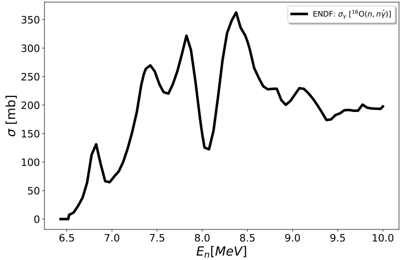
	CoH	ENDF (derived)	Baghdad Atlas
$\langle\sigma_\gamma\rangle$ [mb]	30.60	28.06	**23.9(56)**

- 740-keV γ ray resolved from doublet.
- 740-keV γ ray deexcites 862-keV level.

Integral measurement in agreement with 2 different validation results.

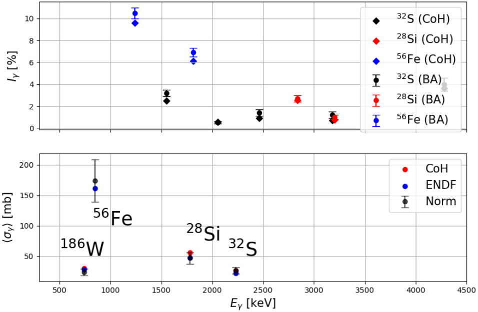
Flux-weighted validation for ^{16}O$(n, n'\gamma)$: 6129 keV

Derived $\sigma_\gamma(6129)$ from ENDF



- $\langle\sigma_\gamma\rangle = 1.09(50)$ mb (Baghdad Atlas)
- $\langle\sigma_\gamma\rangle = 0.533$ mb (ENDF)
- ^{16}O γ rays hard to measure.
- Flux at energy needed is low.
- ENDF-derived σ_γ especially needed for cases like ^{16}O which are also quite difficult to model cf. statistical Hauser-Feshbach approach.

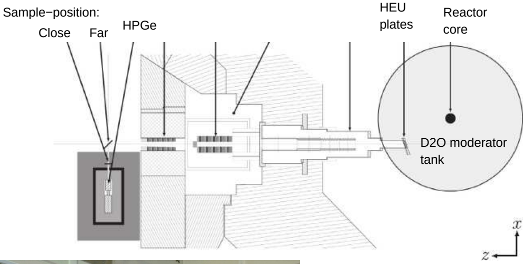
Validation using integral data for ^{28}Si, ^{32}S, ^{56}Fe, and ^{186}W



- Absolute flux-weighted quantities from CoH and ENDF reproduce known absolute integral I_γ data from the Baghdad Atlas.
- Normalization transitions and *weaker* transitions.
- Explore additional isotopes covering broader energy range.

Neutron flux at the Baghdad Research IRT-M Reactor is *well characterized* in region $0.862 \leq E_n \leq 5.0$ MeV.

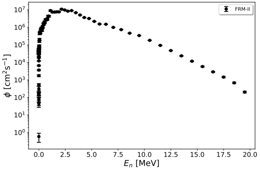
Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM-II)



Fission neutrons:

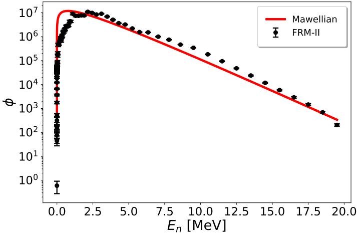
$\sim 1.4 \times 10^{8}/\mathrm{cm}^2/\mathrm{s}$

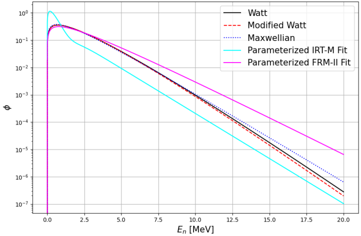
$\langle E_n \rangle \approx 2.3$ **MeV**



Fast **N**eutron **Ga**mma **S**pectroscopy (FaNGaS) instrument with neutron collimator, gamma counting and sample positioning system setup at the fission-neutron beamline (SR10) of the FRM-II.

Validation using FRM-II Reactor flux

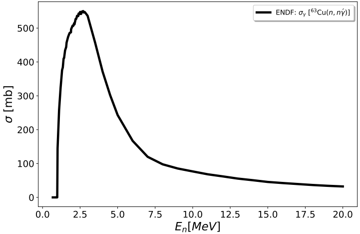


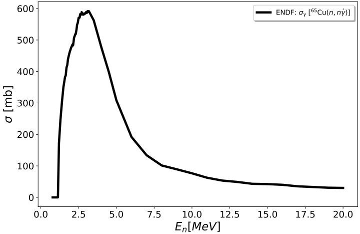


Source	$\langle\sigma_\gamma\rangle_{W_\gamma}$ [mb]	$\langle\sigma_\gamma\rangle$ [mb]	$\langle\sigma_{\rm tot}\rangle$ [mb]	$\langle\sigma_{\rm lev}\rangle$ [mb]	$\frac{\langle\sigma_{\rm lev}\rangle}{\langle\sigma_{\rm tot}\rangle}$
CoH	622.5	759.4	797.3	468.8	0.59
ENDF (estimate)	590.7	720.7	756.6	461.3	0.61
ENDF (derived)	552.6	673.9	–	–	–
FRM-II	**586(41)**	**715(50)**	–	–	–

- FRM-II: $\langle E_n\rangle = 2.32$ MeV from fit cf. 2.3 MeV [Ilic 2020].
- Validation results for ^{56}Fe$(n,n'\gamma)$; $E_\gamma = 846.8$ keV $(2_1^+ \to 0_{\rm gs}^+)$.
- Additional validation datasets from FRM-II: Al, Ti, Cu, In, Ca.

Comparison with ^{63}Cu (962 keV) and ^{65}Cu (1115 keV) [PRELIMINARY]





- $^{63}\mathrm{Cu}(n, n'\gamma)$
- $E_\gamma = 962$ keV
- $\langle\sigma_\gamma\rangle$(FRM-II) = 410(41) mb
- $\langle\sigma_\gamma\rangle$(ENDF) = 297 mb

- $^{65}\mathrm{Cu}(n, n'\gamma)$
- $E_\gamma = 1115$ keV
- $\langle\sigma_\gamma\rangle$(FRM-II) = 404(39) mb
- $\langle\sigma_\gamma\rangle$(ENDF) = 288 mb

Summary and outlook

- Two different fission-based neutron sources considered in validation of $(n, n'\gamma)$ data from ENDF libraries for the GRIN project: Baghdad Reactor and FRM-II.
- Flux at Baghdad Reactor appears to be well-characterized in region $0.862 \leq E_n \leq 5.0$ MeV.
- Validation approach utilizes γ-ray production data from both reaction-model calculations as well as ENDF-derived γ-ray data to deduce: (i) calculated; (ii) estimated; and (iii) library-derived flux-weighted γ-ray production cross sections that can be compared to integral measurements from reactor facilities.
- Additional low-energy data needs to be considered to help constrain low-E_n tail to better describe the flux in the region $E_n < 862$ keV at the Baghdad Reactor.
- Preliminary assumption of Maxwellian ($kT = 1.55$ MeV; $\langle E_n \rangle = 2.32$) does reasonable job of reproducing measured ^{56}Fe data at FRM-II.
- More work needed for other isotopes (e.g., ^{63,65}Cu) and better description of flux.
- Emanuel developed script to extract γ-production data from ENDF libraries $\Rightarrow$ disseminate as part of the FUDGE library in future?