Evaluation of neutron-induced reactions on medium-mass nuclei and actinides

Ionel Stetcu
Los Alamos National Laboratory (T-2)

February 22, 2024

LA-UR-24-21100

Technical Program Review
February 20 -- 22, 2024

Outline

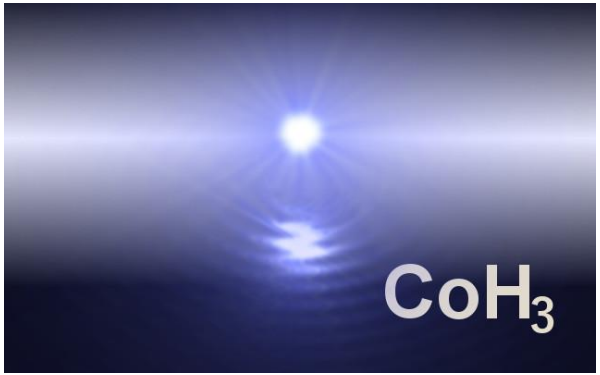
- CoH_3: the main tool for evaluations
- Measurement and evaluation of ^{35}Cl (not funded by, but relevant to NCSP)
- Evaluation of neutron-induced reactions on 139La
- ^{181}Ta evaluation (finalized)
- $^{238,240\text{-}242}Pu(n,f)$: evaluation of nubar
- $^{233}U(n,g)$: exploring new evaluation
- Summary

CoH_3: Coupled-Channels Hauser-Feshbach code

❑ Hauser-Feshbach-Moldauer theory for compound nuclear reactions
- 45,000 lines C++ code (~ 140 C++ source files, ~60 headers, ~80 classes)
- maintain by GNU Autotools package

❑ Modules and Models employed
- spherical and deformed optical models
- DWBA for direct inelastic scattering
- Moldauer's width fluctuation correction with LANL parametrization
- Gilbert-Cameron level density with updated parameters
- pre-equilibrium 2-component exciton model
- Madland-Nix prompt fission neutron spectrum including pre-fission emission
- direct/semidirect capture model
- mean-field models (FRDM and Hartree-Fock BCS)

Consistent evaluations in all channels



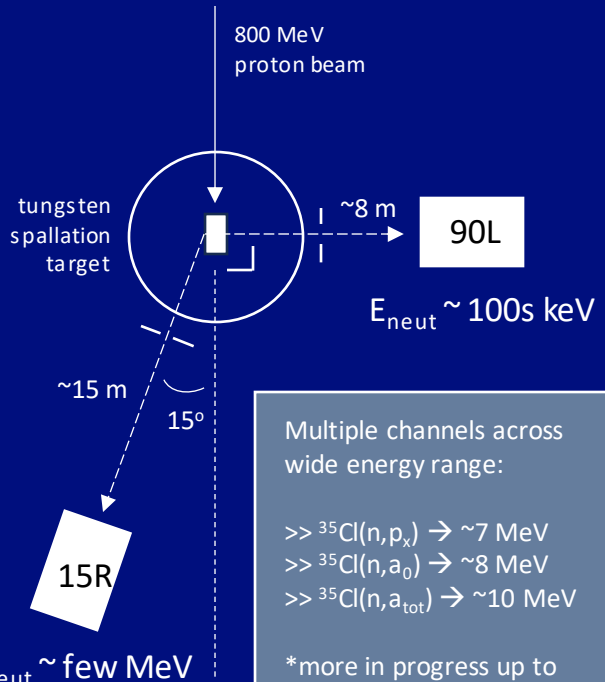
Main developer: Toshihiko Kawano

LENZ Experiment: ^{35}Cl(n,Z)

(Funded by GAIN voucher + CRADA with TerraPower LLC)

Courtesy K. Hanselman (P-3)

LANSCE

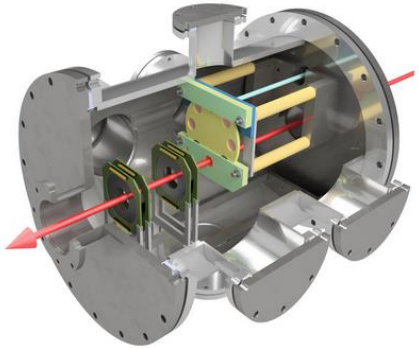


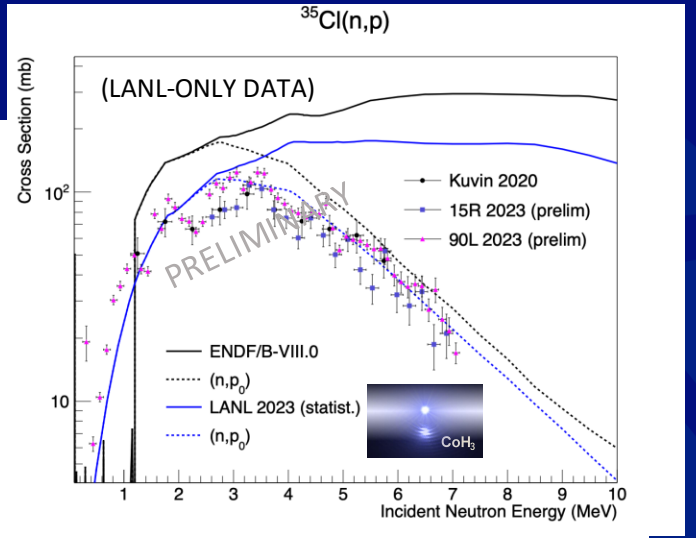
Multiple channels across wide energy range:

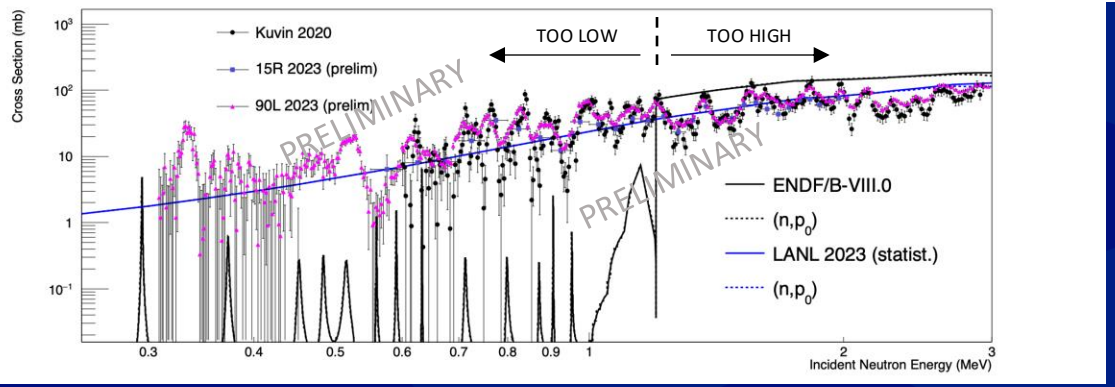
>> ^{35}Cl(n,p_x) → ~7 MeV
>> ^{35}Cl(n,a_0) → ~8 MeV
>> ^{35}Cl(n,a_{tot}) → ~10 MeV

*more in progress up to ~14 MeV...

LENZ: Low-Energy (n,Z)







→ PRELIMINARY evaluation ongoing for future ENDF release...

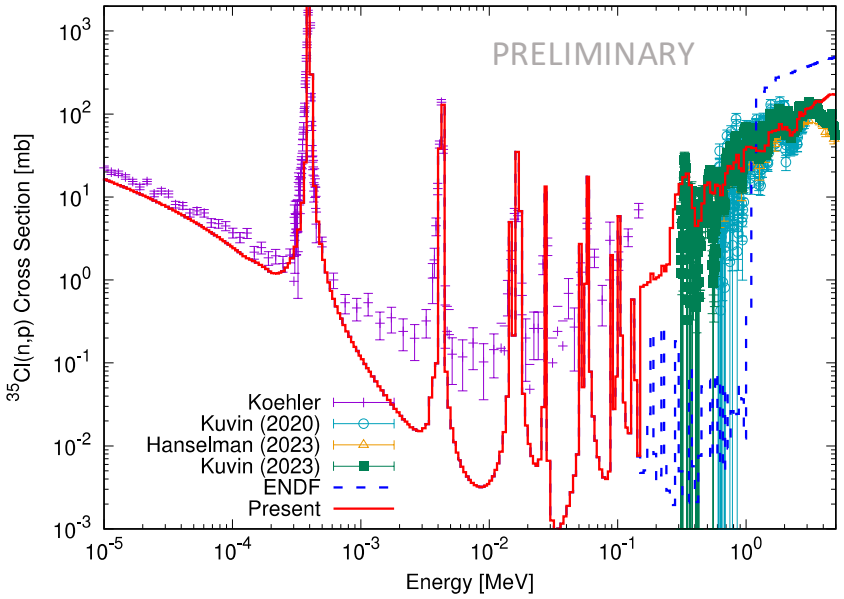
Current Working Evaluation

Courtesy K. Hanselman (P-3)

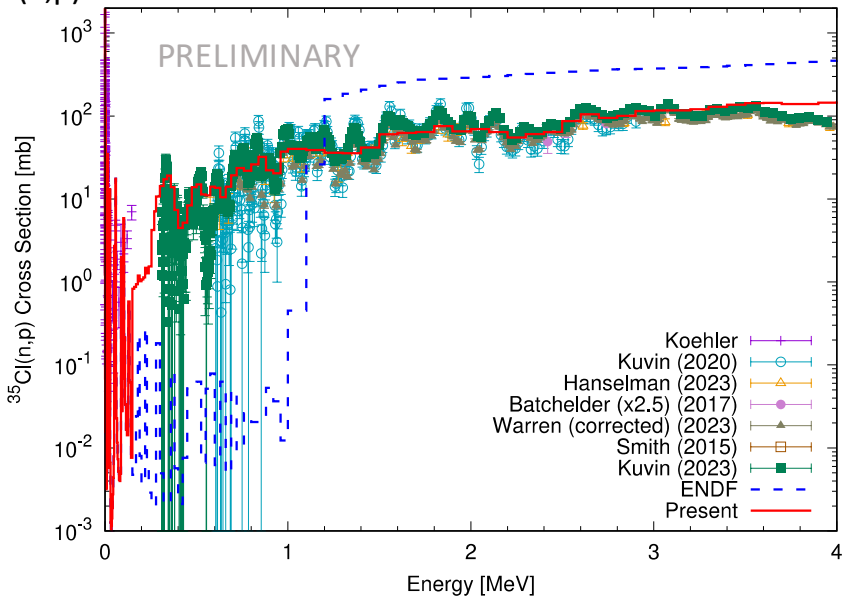
- → on ALL available data (some tweaks)
- → including higher-energy (>10 MeV) channels – e.g. (n,2n)
- → blend current RRR analysis with new statistical

More "continuous" now between 0.1-1 MeV region

(group cross sections)



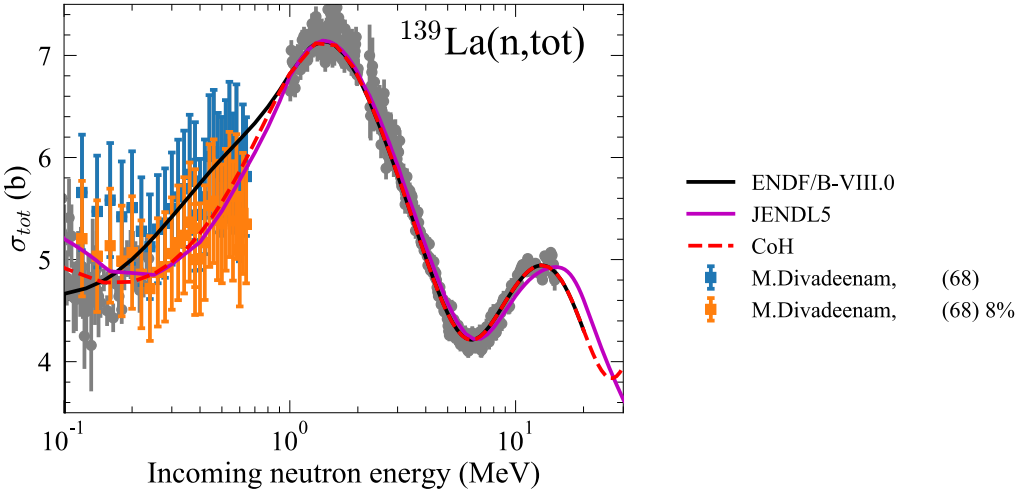
^{35}Cl(n,p)

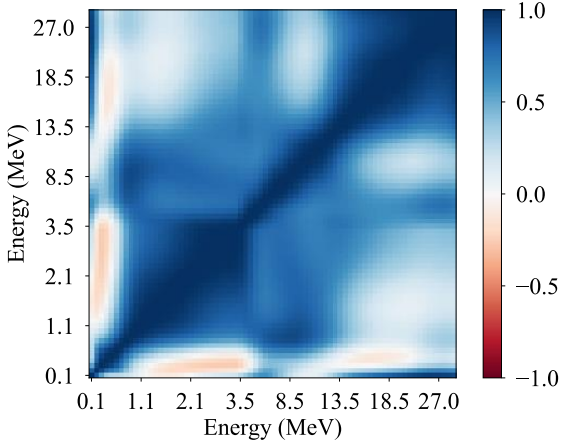


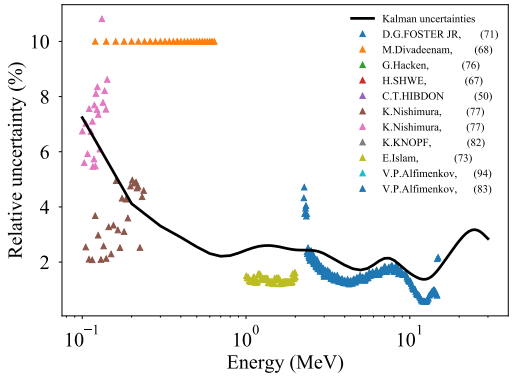
Latest development: covariances

^{139}La(n,tot) evaluation

- Good agreement with ENDF/B-VIII.0/CENDL-3 and JENDL5 above 1 MeV
- ENDF/B-VIII.0 follows Divadeenam data below 1 MeV
- Could there be problem with the Divadeenam data? Not enough information
- New evaluation: better matching of the low-energy data
- Waiting for the resonance analysis from ORNL to see whether we should adjust the low-energy cross section

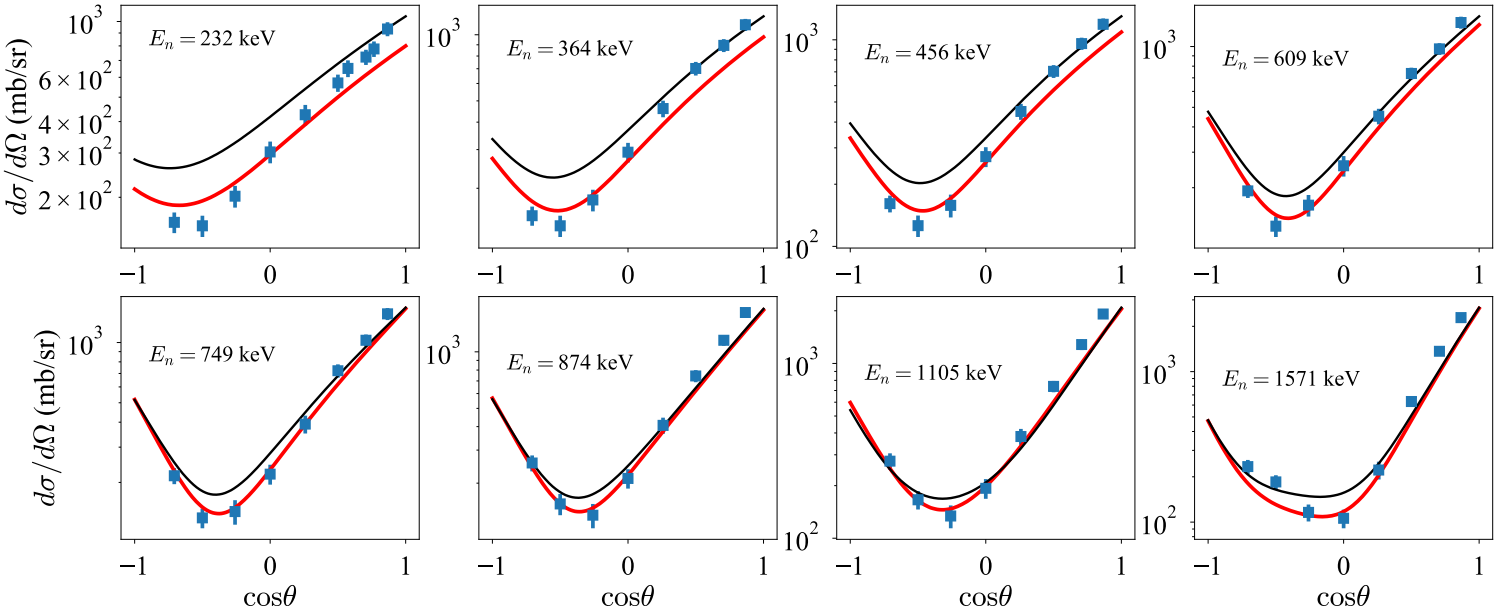






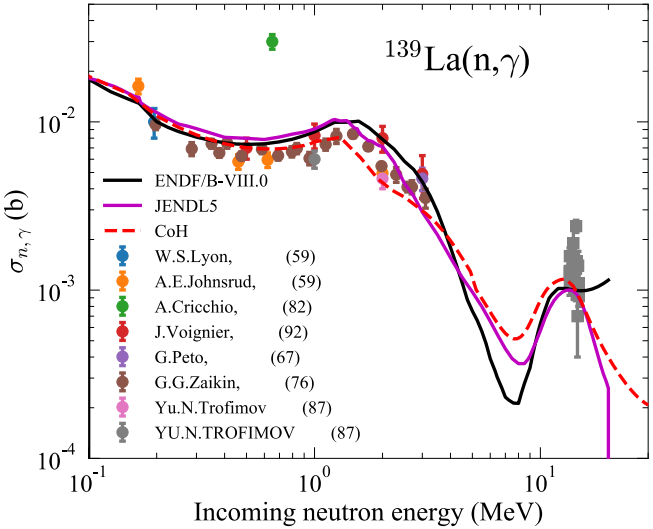
Documented in the paper for ICNC 2023

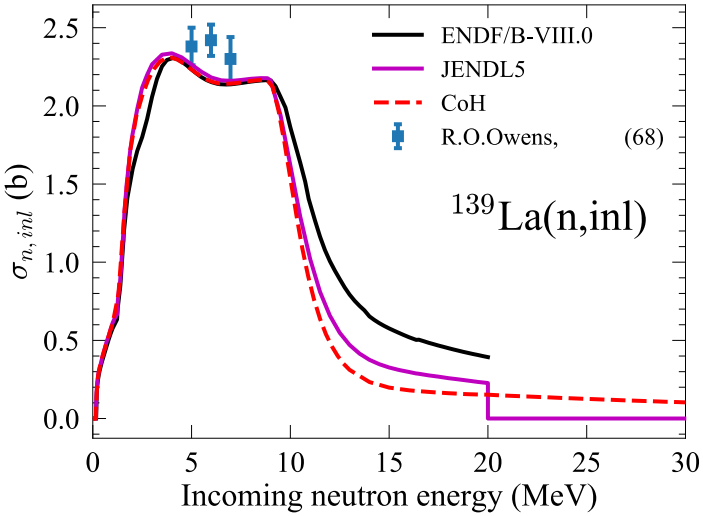
^{139}La angular dependence



Fairly good reproduction of experimental data (no adjustments)
(black: ENDF/B-VIII.0, red: new evaluation based on CoH)

^{139}La(n,g) and ^{139}La(n,inl)





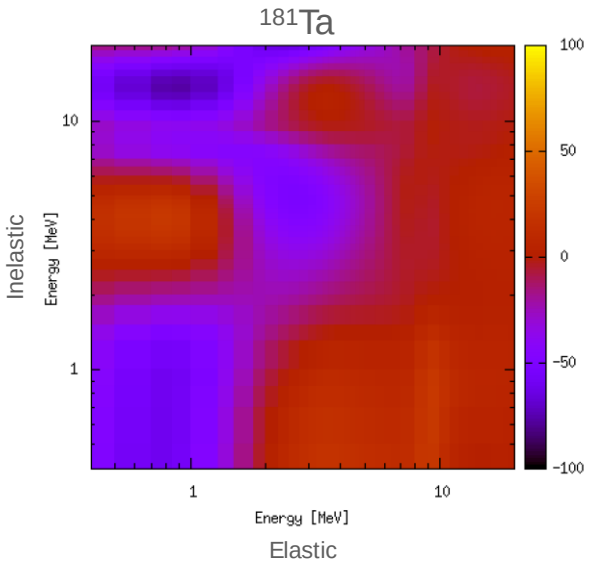
- At energies above 1 MeV, the capture data can be less reliable
- Inelastic channel very close to the previous evaluation

^{181}Ta evaluation

Empire code used in evaluation:

- Coupled-Channels with adjusted dispersive Optical Model potential
- Multistep Direct (MSD) model for pre-equilibrium neutron emission.
- Heidelberg formulation of the Multistep Compound (MSC) model for pre-equilibrium neutron and γ-emission.
- Exciton model for pre-equilibrium proton emission.
- Exciton model with Iwamoto-Harada extension for pre-equilibrium cluster emission.
- Gilbert-Cameron model for level densities.
- Hauser-Feshbach with Moldauer width correction and Blatt-Biedenharn angular distributions for compound nucleus decay.

Evaluation already performed FY2021-2022 (TPR-2023), but extra work on obtaining the cross-channel covariances



Courtesy M. Herman (T-2)

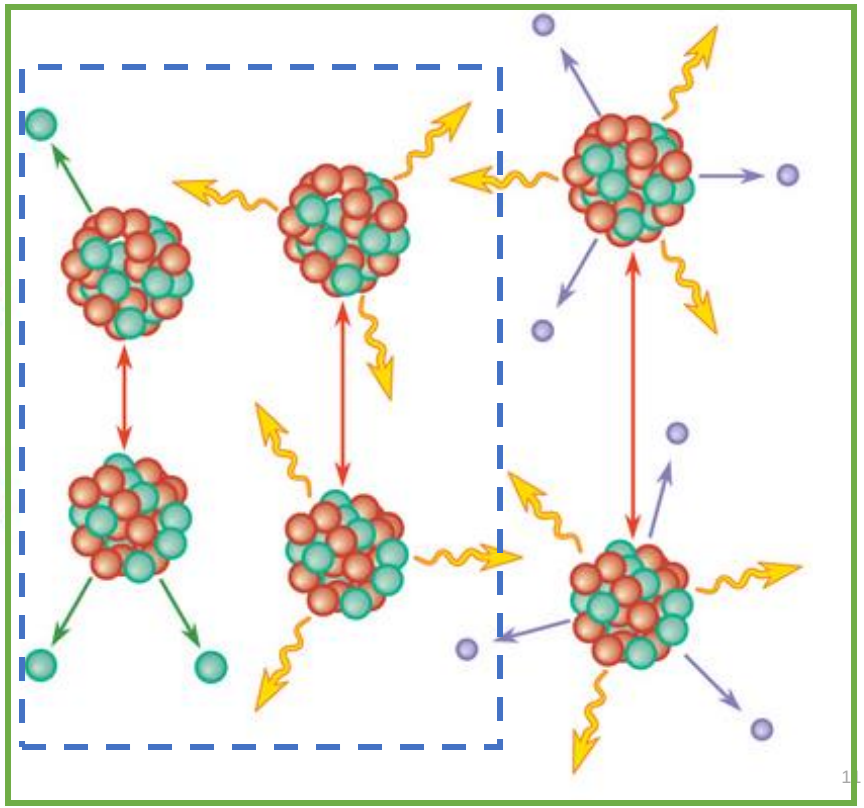
Evaluation and Theory FY23Q4 milestone:

- $^{238,240\text{-}242}$Pu: “Attempt a consistent nu-bar evaluation supported by a model code to provide better evaluated nu-bar for minor Pu-isotopes”.

Work by D. Neudecker (XCP-5) and A. Lovell (T-2)

LANL fission models consistently calculate prompt and delayed fission observables

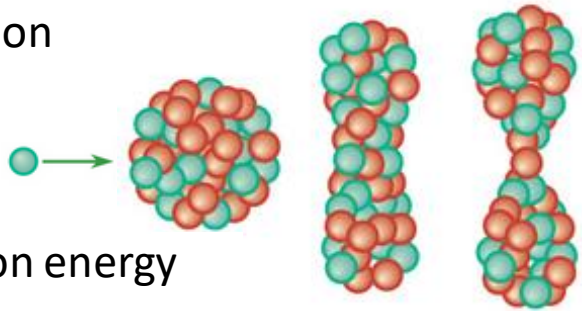
Deterministic: BeoH (prompt and delayed)



Input needed from theory and experiment:

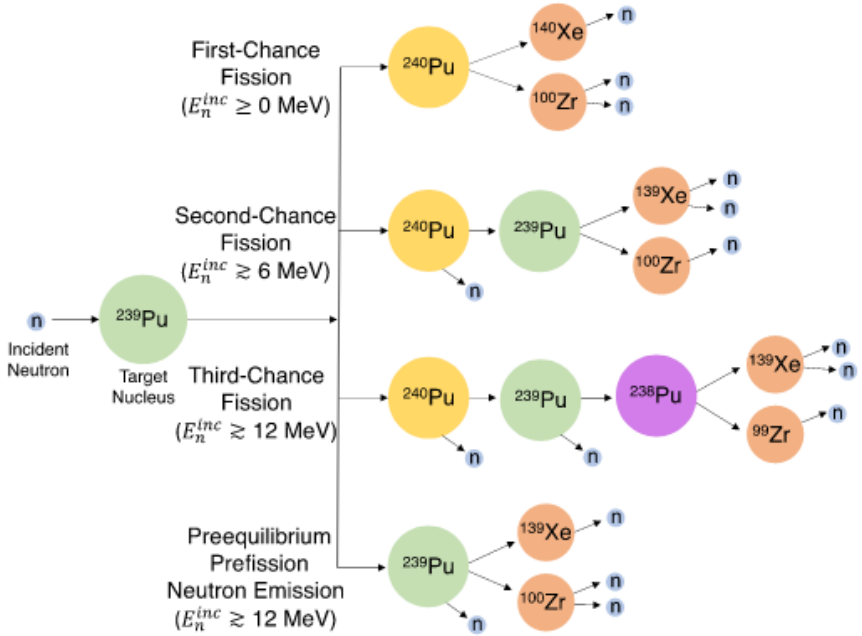
- First-principle calculations of fission yields
- Multi-chance fission probabilities
- Pre-fission neutron energy spectra
- Mass, charge, and kinetic energy of fission yields

Monte Carlo: CGMF (prompt)



Los Alamos NATIONAL LABORATORY

CGMF models multi-chance fission explicitly, so parameter inputs connect between fission reactions



K.J. Kelly, *et al.*, PRL **112**, 072503 (2019)

We calculate average prompt neutron multiplicities for ^{238}Pu(n,f), ^{239}Pu(n,f), ^{240}Pu(n,f), ^{241}Pu(n,f), and ^{242}Pu(n,f), which includes parametrizations for the compound nuclei ^{236}Pu, ^{237}Pu, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{243}Pu.

Parametrizing the CGMF input as a function of the mass of the compound nucleus, A_c, allows us to perform a consistent evaluation across all reactions.

We get evaluated data and covariances with GLLS using model data as a prior and updating with exp. info.

- GLLS combines:
- **Model ("M") parameters and covariances by CGMF (from Amy),**
- **Experimental mean values ("x")** and covariances (from Denise),
- **To evaluated mean values and covariances ("post") for a ND file** using,
- Design matrix S (by Amy) that transforms from model parameter to observable space.

$$\underline{\phi}^{post} = \underline{\phi}^{M} + \mathbf{Cov}^{post}\mathbf{S}^{+}\left(\mathbf{Cov}^{x}\right)^{-1}\left(\underline{\phi}^{x} - \mathbf{S}\underline{\phi}^{M}\right),$$

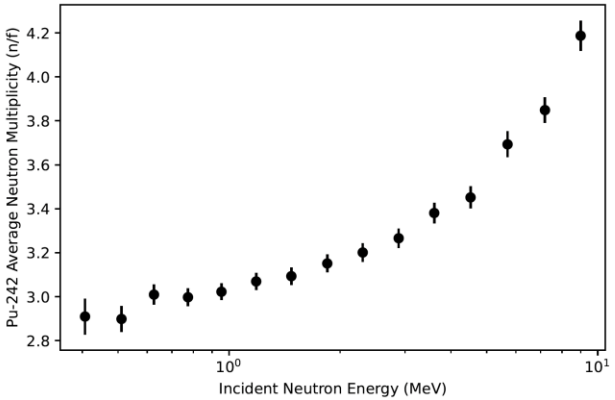
$$\mathbf{Cov}^{post} = \mathbf{Cov}^{M} - \mathbf{Cov}^{M}\mathbf{S}^{+}\left(\mathbf{S}\mathbf{Cov}^{M}\mathbf{S}^{+} + \mathbf{Cov}^{x}\right)^{-1}\mathbf{S}\mathbf{Cov}^{M}$$

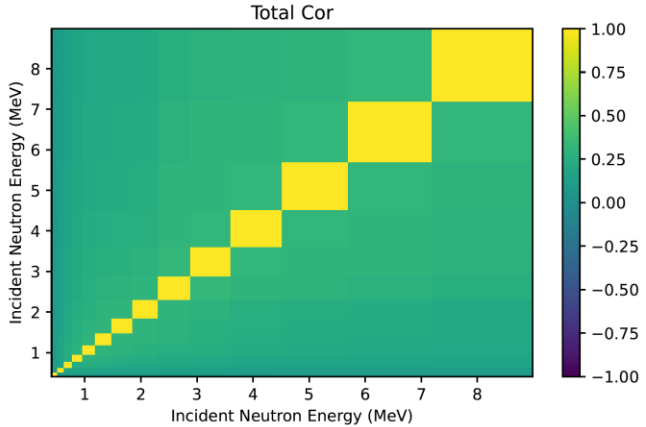
The evaluation was undertaken with the code *ARIADNE* that was extended to evaluate multiple nu-bar isotopes.

Experimental $^{238,240\text{-}242}$Pu nu-bar values were reviewed carefully and UQ was undertaken with *ARIADNE*.

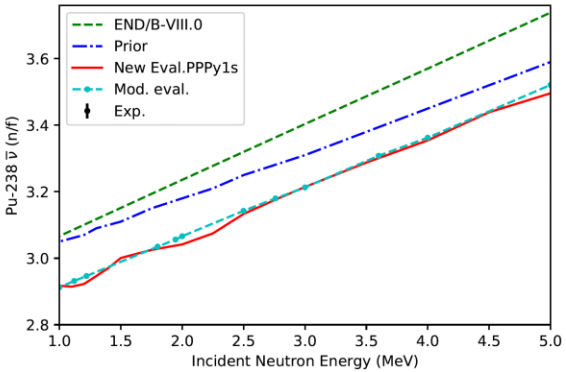
Exp. data for ^{238}Pu are only available at thermal. Only one set exists for ^{242}Pu. That highlights why a cross-isotope evaluation is key for obtaining consistent nu-bar.

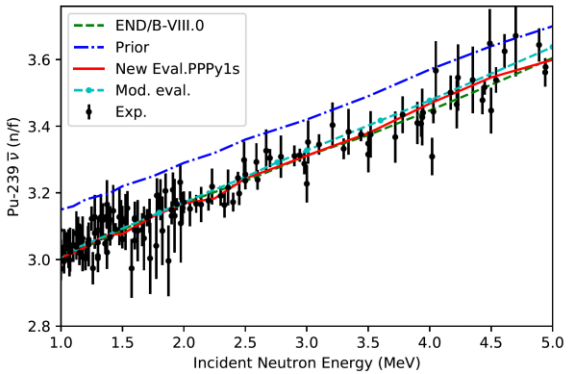
Isotope	# exp. found	Accepted
^{238}Pu	3	2
^{240}Pu	7	3
^{241}Pu	14	7
^{242}Pu	1	1

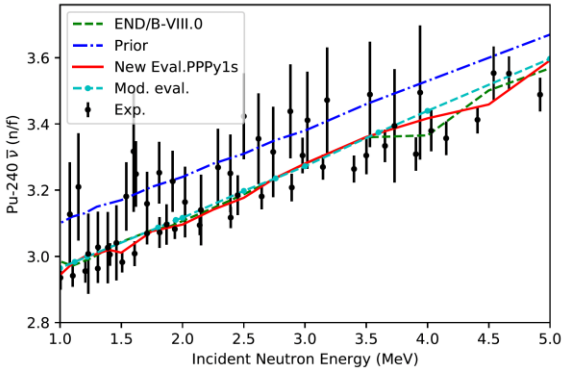


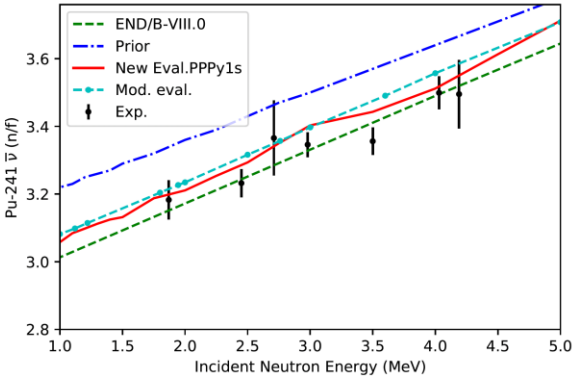


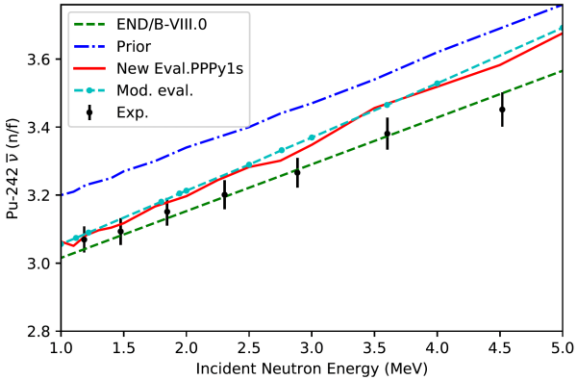
Evaluated $^{239\text{-}241}$Pu nu-bar close to VIII.0, new ^{238}Pu nu-bar lower and new ^{242}Pu nu-bar higher than VIII.0.





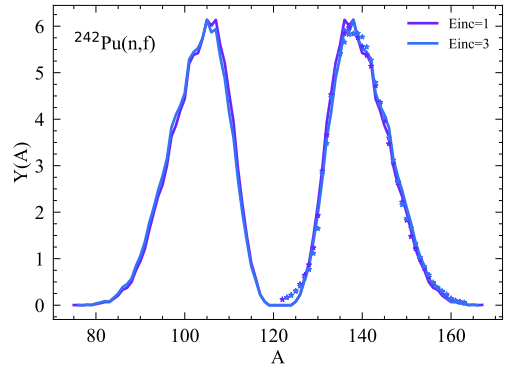


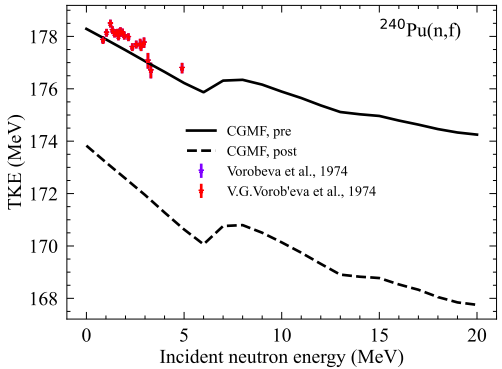


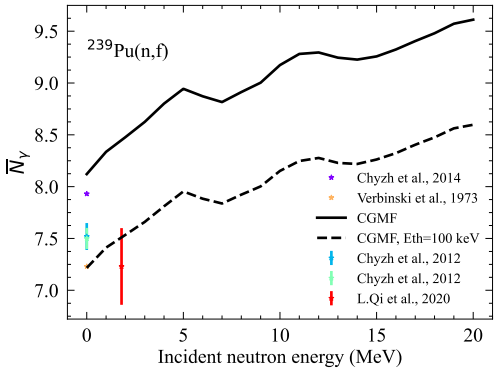


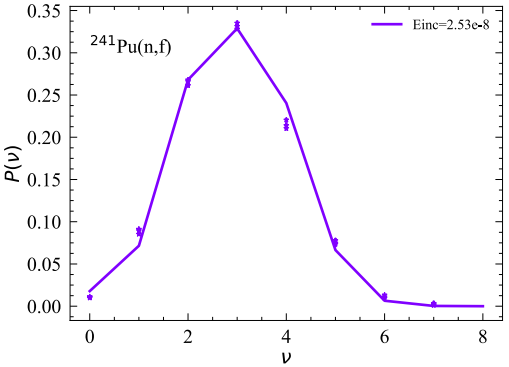
The evaluation gives values at thermal and from 100 keV-20 MeV for all five isotopes.

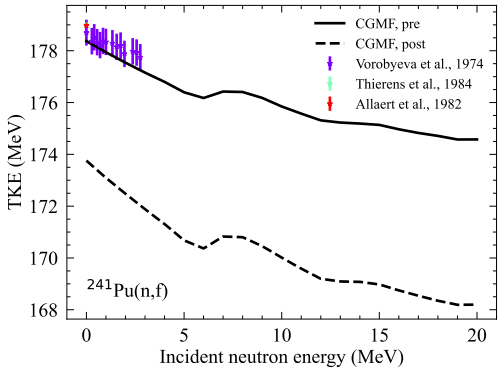
Neutron multiplicities in CGMF are consistent with other prompt data, where data are available (mostly sparse)

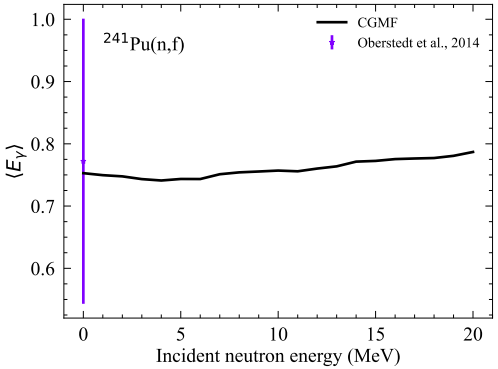






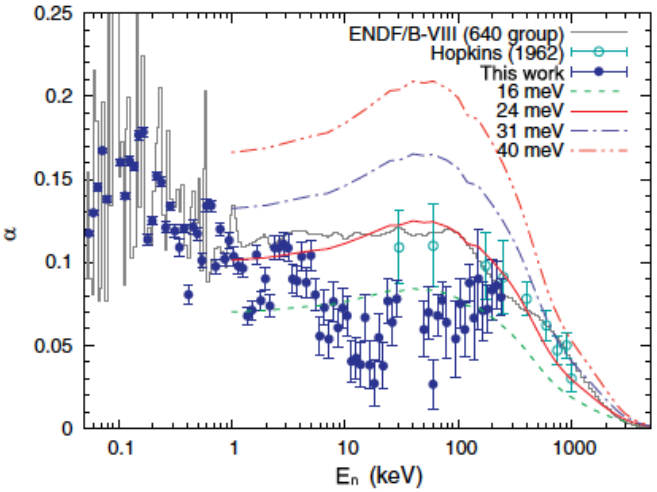


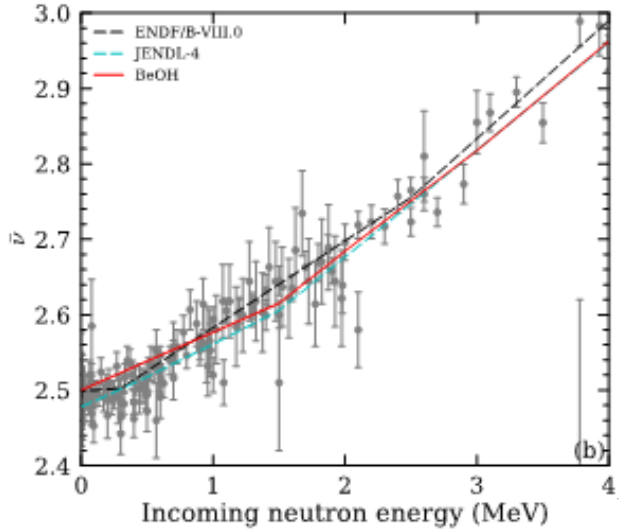




^{233}U(n,γ): challenge to model consistently

- Fast region has become an even bigger problem for the modeling codes
- Can affect criticality benchmarks
- Freedom: tweaks of nubar within uncertainties
- Evaluation work in progress





Summary

- ^{35}Cl(n,Z): new measurements with LENZ funded by GAIN and TerraPower
  - Produced candidate evaluations, preliminary tests successful
- ^{139}La: finished the evaluation, including covariances
  - Waiting for ORNL to make final adjustments and finalize the file for submission to NNDC
- ^{181}Ta: evaluation complete
  - Included cross-correlation covariances in a new file that will be submitted to NNDC
- We performed the first consistent evaluation of average prompt neutron multiplicities for $^{238\text{-}242}$Pu(n,f) reactions using CGMF
  - Moving forward, we are exploring an increased parameter space to take into account model stiffness and correlations
  - Covariances
- ^{233}U(n,g): new DANCE measurement provides challenges for the evaluation
  - Continue to investigate different scenarios for a new evaluation, with and without DANCE data

This work was supported by the Nuclear Criticality Safety Program, funded and managed by the National Nuclear Security Administration for the Department of Energy.