Los Alamos
NATIONAL LABORATORY

$^{233}U(n, \gamma)$ DANCE and NEUANCE measurements at LANSCE

Nuclear Data Week(s)-2022

Esther Leal Cidoncha
31 October - 11 November 2022

LA-UR-22-31346

Motivation

- Th-U alternative to U-Pu fuel cycle due to its reduced amount of transuranium elements.
- ^{232}Th is more abundant in nature than uranium.
- In the Th fuel cycle the ^{232}Th, transmutes into the fissile isotope ^{233}U.

$$n +_{90}^{232} Th \rightarrow_{90}^{233} Th \xrightarrow{\beta-}{}_{91}^{233} Pa \xrightarrow{\beta-}{}_{92}^{233} U$$

- ^{233}U(n,f) produces a large rate of emitted neutrons, enough to maintain the chain reaction.
- For this reason, the Th fuel cycle may be the basis of thermal breeder reactors, being also suitable to use in fast reactors.
- Chemical advantages from thorium vs uranium: higher melting point and thermal conductivity.

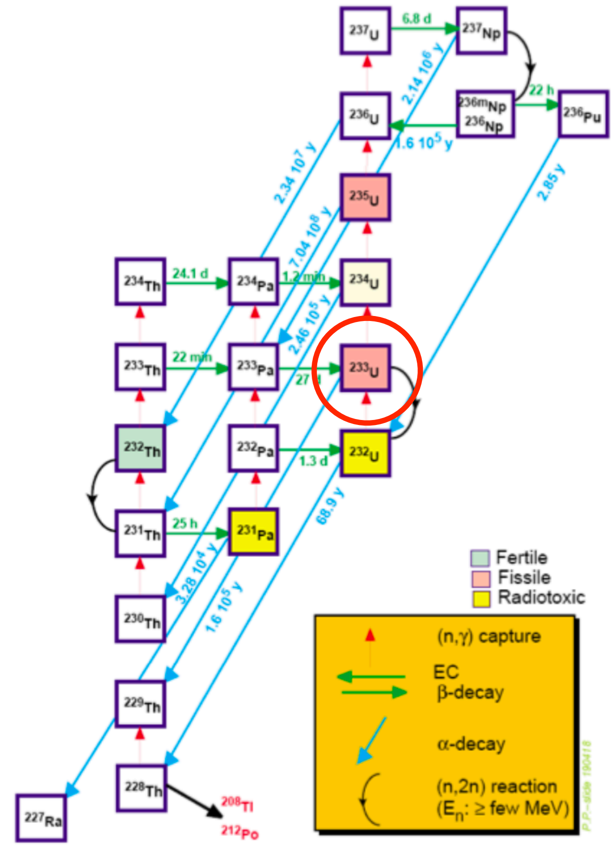
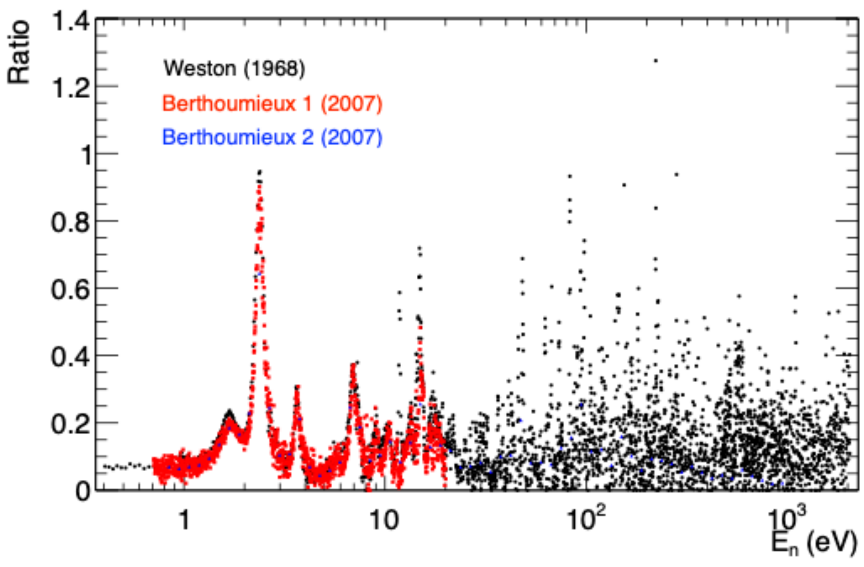
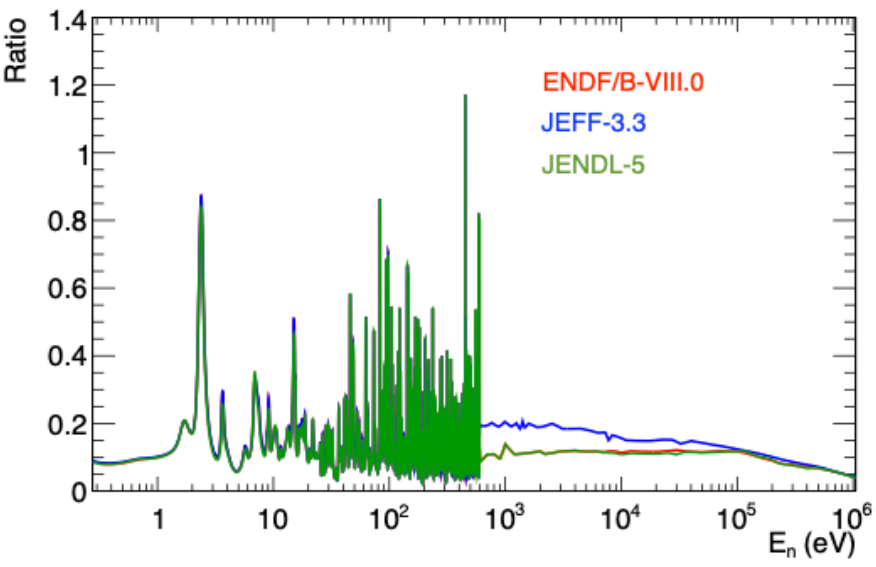


Illustration of the thorium fuel cycle.

Motivation

- Experimental $^{233}U(n,\gamma)$ cross section data in the literature are scarce and were measured decades ago.
- New report [1] suggests that a simultaneous measurement with capture would be useful.

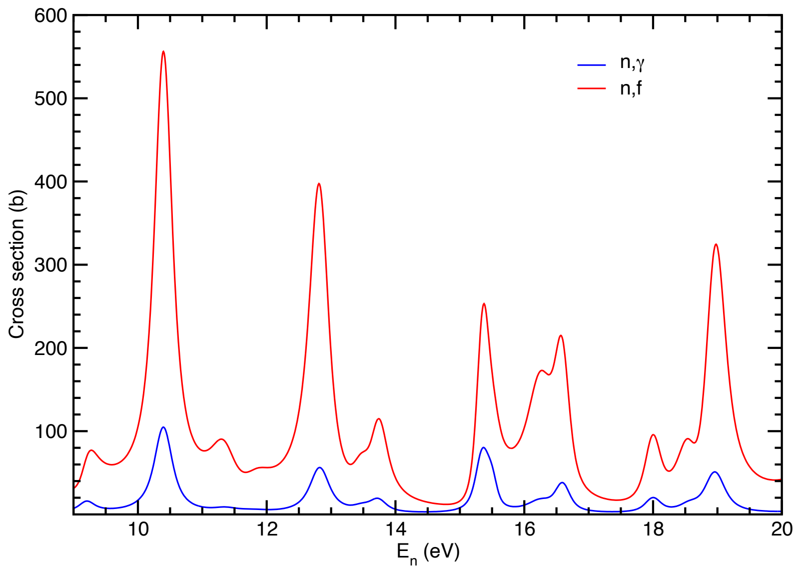




[1] M.T. Pigni, R. Capote and A. Trkov, Annals of Nuclear Energy **163** (2021) 108595.

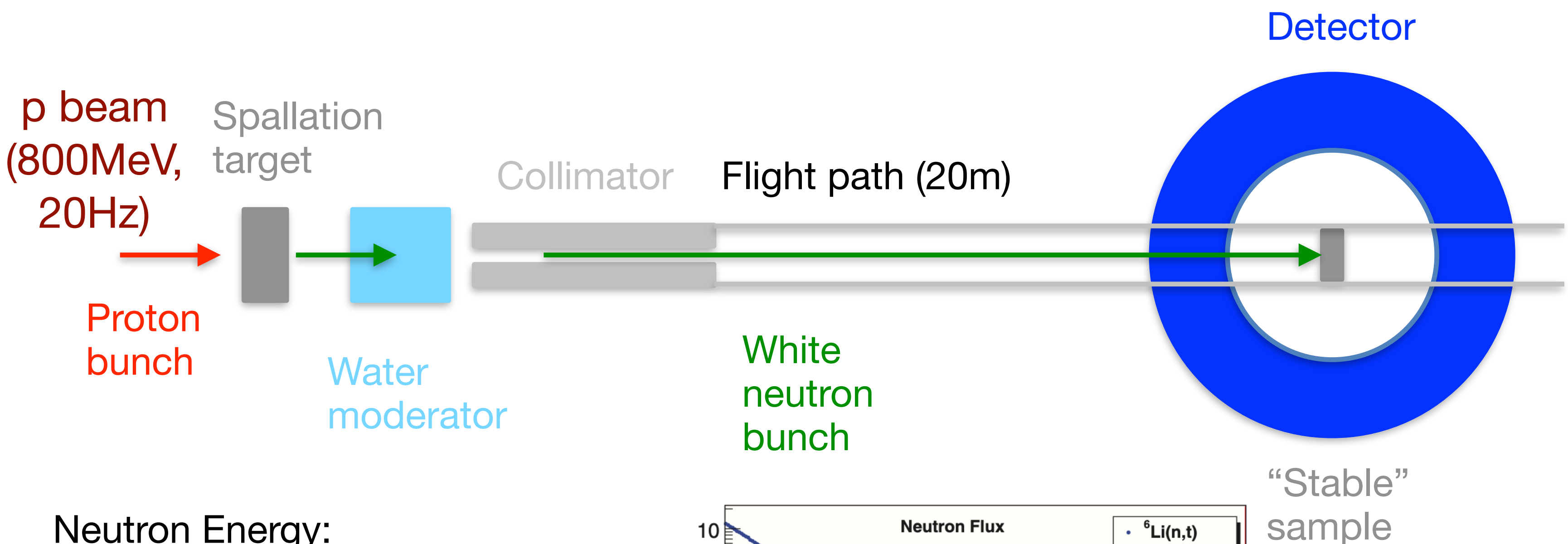
Motivation

- For ^{233}U fission is around one order of magnitude more likely than capture.
  - Good discrimination between gammas coming from capture and fission is required.
  - New measurement proposed at LANL combining NEUANCE and DANCE.



$^{233}U(n,\gamma)$ and $^{233}U(n,f)$ cross sections from ENDF/B-VIII.

Time-of-flight measurements

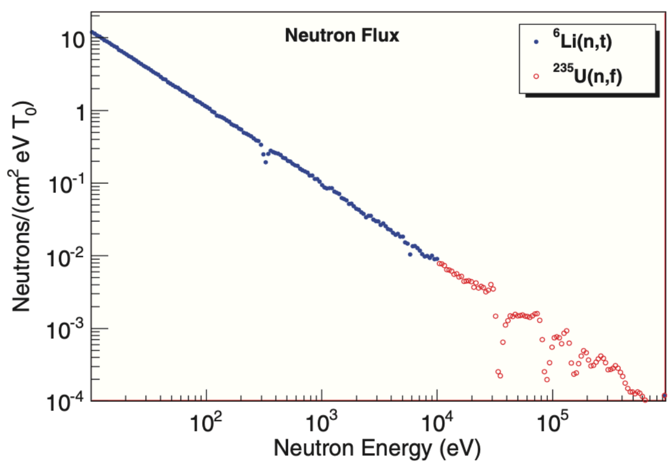


Neutron Energy:

$$E_n = m_n c^2 \frac{1}{\sqrt{1-(\frac{v}{c})^2}} - 1$$

with:

$$v = L/T$$



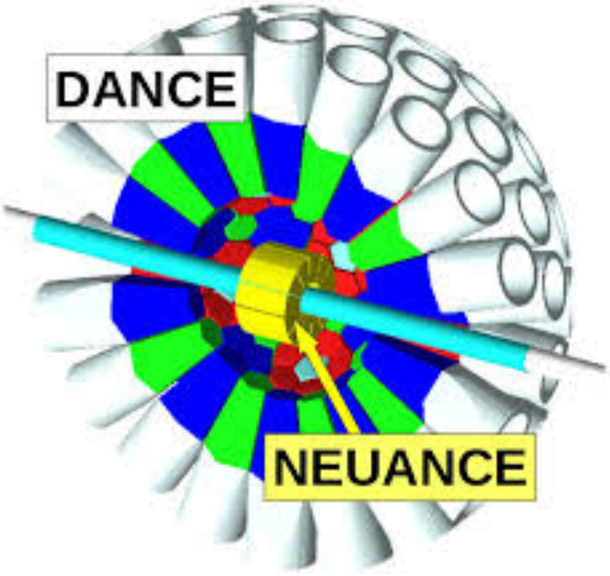
Flux$_n$ = 3 * 10^5 n/s/cm^2/dec

Los Alamos NATIONAL LABORATORY

Detectors

DANCE (**D**etector for **A**dvanced **N**eutron **C**apture **E**xperiments)

- $4\pi BaF_2$ **γ**-ray calorimeter composed by 160 crystals with an inner cavity of 17 cm radius [2].
- Used to measure neutron capture cross section data on small quantities of radioactive isotopes.
- We can measure En, Esum, Ecl, and Mcl, providing more information than with C6D6 detectors.



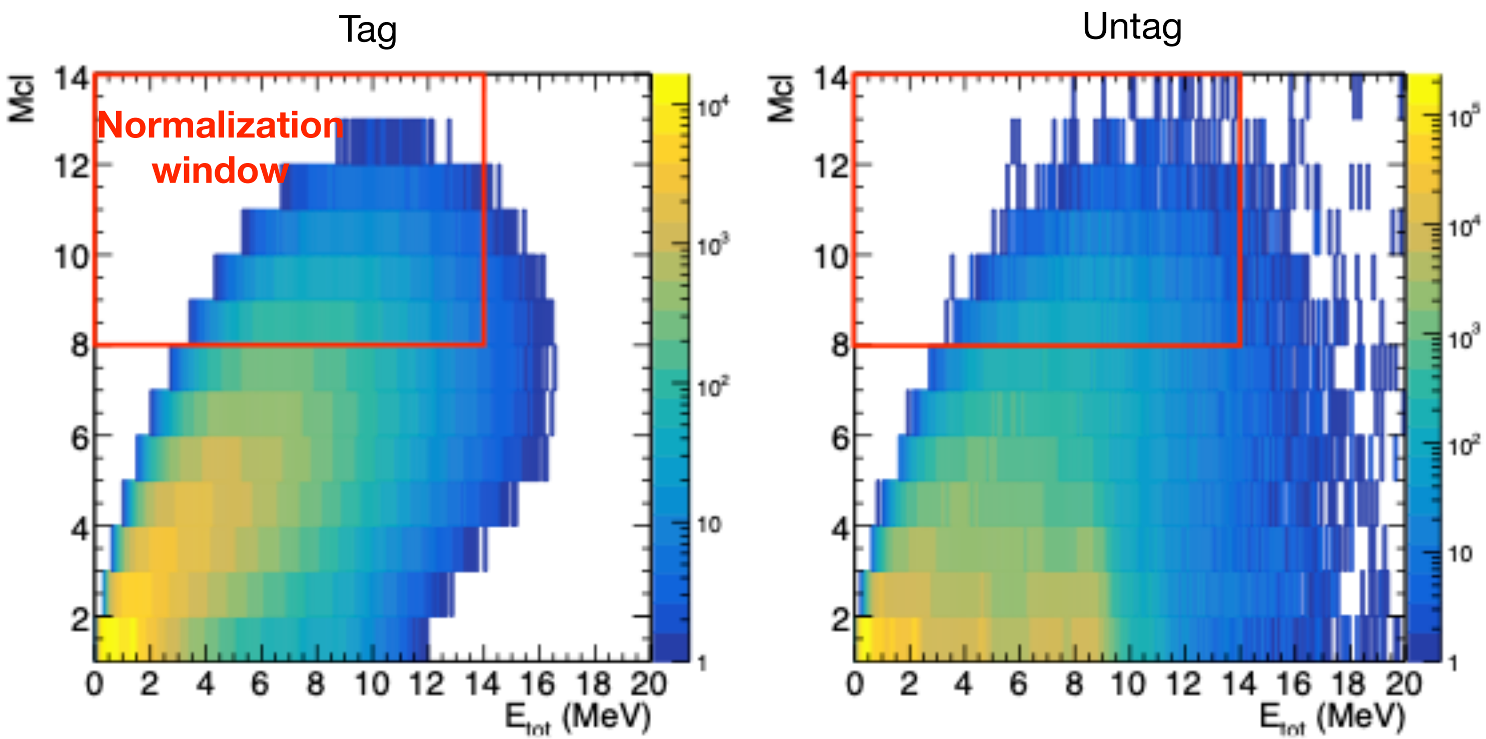
NEUANCE (**NEU**tron detector array at d**ANCE**)

- Neutron detector array that consists in 21 stilbene crystals arranged in a cylindrical geometry around the beam pipe [3].
- Used to detect neutrons coming from fission and determine by coincidence with DANCE, the gammas coming from fission.
- NEUANCE detects neutrons with energies above 200 keV (fission neutrons have these energies), therefore **low energy scattered neutrons** that are below this threshold are **discriminated**.
- Possibility to use a thick target.
- NEUANCE can also detect gammas.

[2] M. Heil et al., Nucl. Instrum. Methods Phys. Res. A **459**, 229 (2001).
[3] M. Jandel et al. Nuclear Inst. and Methods in Physics Research, A **882** (2018) 105-113.

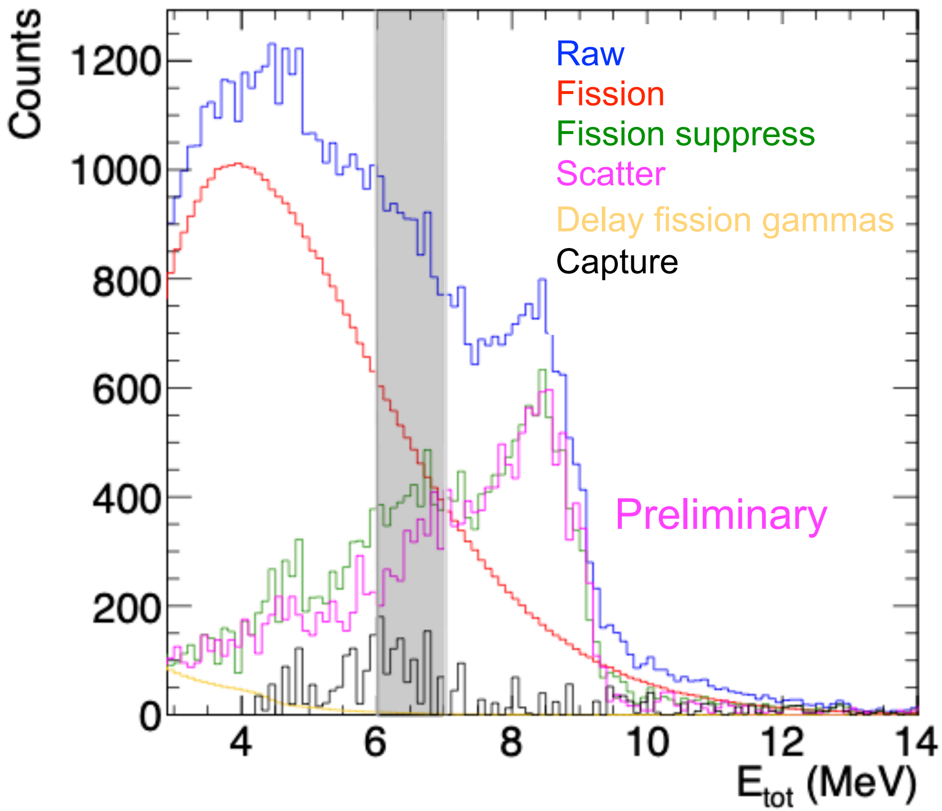
Fission tagging process

- Search for coincidences between the two detectors.
- The DANCE gammas in coincidence with the NEUANCE neutrons are tagged as fission gammas.
- The purpose of tagging is to define the shape of the fission γ-ray spectrum that can be subtracted from the untagged spectrum.



Background studies

- The background varies with the neutron energy, therefore it is subtracted per En bin.



Mcl=(4,6)

En = 300eV

Q value peak = 6.845 MeV

Los Alamos NATIONAL LABORATORY

Capture to fission ratio

The capture to fission ratio is given by:

$$\alpha(E_n) \equiv \frac{\sigma_\gamma(E_n)}{\sigma_f(E_n)}$$

Capture to fission ratio

The capture to fission ratio is given by:

$$\alpha(E_n) \equiv \frac{\sigma_\gamma(E_n)}{\sigma_f(E_n)}$$

We measure a number of events Ci as a function of the neutron energy associated with process i (fission or capture):

$$C_i(E_n) = \varepsilon_i Y_i(E_n)$$

Capture to fission ratio

The capture to fission ratio is given by:

$$\alpha(E_n) \equiv \frac{\sigma_\gamma(E_n)}{\sigma_f(E_n)}$$

We measure a number of events Ci as a function of the neutron energy associated with process i (fission or capture):

$$C_i(E_n) = \varepsilon_i Y_i(E_n)$$

and

$$Y_i(E_n) = \sigma_i(E_n) N_{^{233}U} \Phi_n(E_n)$$

Capture to fission ratio

The capture to fission ratio is given by:

$$\alpha(E_n) \equiv \frac{\sigma_\gamma(E_n)}{\sigma_f(E_n)}$$

We measure a number of events Ci as a function of the neutron energy associated with process i (fission or capture):

$$C_i(E_n) = \varepsilon_i Y_i(E_n)$$

and

$$Y_i(E_n) = \sigma_i(E_n) N_{^{233}U} \Phi_n(E_n)$$

Therefore, the capture to fission ratio can be expressed as:

$$\frac{C_\gamma(E_n)}{C_f(E_n)} = \frac{\varepsilon_\gamma Y_\gamma(E_n)}{\varepsilon_f Y_f(E_n)}$$

Capture to fission ratio

The capture to fission ratio is given by:

$$\alpha(E_n) \equiv \frac{\sigma_\gamma(E_n)}{\sigma_f(E_n)}$$

We measure a number of events Ci as a function of the neutron energy associated with process i (fission or capture):

$$C_i(E_n) = \varepsilon_i Y_i(E_n)$$

and

$$Y_i(E_n) = \sigma_i(E_n) N_{^{233}U} \Phi_n(E_n)$$

Therefore, the capture to fission ratio can be expressed as:

$$\begin{aligned} \frac{C_\gamma(E_n)}{C_f(E_n)} &= \frac{\varepsilon_\gamma Y_\gamma(E_n)}{\varepsilon_f Y_f(E_n)} \\ &= \frac{\varepsilon_\gamma \sigma_\gamma(E_n) N_{^{233}U} \Phi_n(E_n)}{\varepsilon_f \sigma_f(E_n) N_{^{233}U} \Phi_n(E_n)} \end{aligned}$$

Capture to fission ratio

The capture to fission ratio is given by:

$$\alpha(E_n) \equiv \frac{\sigma_\gamma(E_n)}{\sigma_f(E_n)}$$

We measure a number of events Ci as a function of the neutron energy associated with process i (fission or capture):

$$C_i(E_n) = \varepsilon_i Y_i(E_n)$$

and

$$Y_i(E_n) = \sigma_i(E_n) N_{^{233}U} \Phi_n(E_n)$$

Therefore, the capture to fission ratio can be expressed as:

$$\frac{C_\gamma(E_n)}{C_f(E_n)} = \frac{\varepsilon_\gamma Y_\gamma(E_n)}{\varepsilon_f Y_f(E_n)} = \frac{\varepsilon_\gamma \sigma_\gamma(E_n) \cancel{N_{^{233}U}} \cancel{\Phi_n}(E_n)}{\varepsilon_f \sigma_f(E_n) \cancel{N_{^{233}U}} \cancel{\Phi_n}(E_n)}$$

Capture to fission ratio

The capture to fission ratio is given by:

$$\alpha(E_n) \equiv \frac{\sigma_\gamma(E_n)}{\sigma_f(E_n)}$$

We measure a number of events Ci as a function of the neutron energy associated with process i (fission or capture):

$$C_i(E_n) = \varepsilon_i Y_i(E_n)$$

and

$$Y_i(E_n) = \sigma_i(E_n) N_{^{233}U} \Phi_n(E_n)$$

Therefore, the capture to fission ratio can be expressed as:

$$\begin{aligned}
\frac{C_\gamma(E_n)}{C_f(E_n)} &= \frac{\varepsilon_\gamma Y_\gamma(E_n)}{\varepsilon_f Y_f(E_n)} \\
&= \frac{\varepsilon_\gamma \sigma_\gamma(E_n) \cancel{N_{^{233}U}} \cancel{\Phi_n}(E_n)}{\varepsilon_f \sigma_f(E_n) \cancel{N_{^{233}U}} \cancel{\Phi_n}(E_n)} \\
&= \frac{\varepsilon_\gamma \sigma_\gamma(E_n)}{\varepsilon_f \sigma_f(E_n)}
\end{aligned}$$

Capture to fission ratio

The capture to fission ratio is given by:

$$\alpha(E_n) \equiv \frac{\sigma_\gamma(E_n)}{\sigma_f(E_n)}$$

We measure a number of events Ci as a function of the neutron energy associated with process i (fission or capture):

$$C_i(E_n) = \varepsilon_i Y_i(E_n)$$

and

$$Y_i(E_n) = \sigma_i(E_n) N_{^{233}U} \Phi_n(E_n)$$

Therefore, the capture to fission ratio can be expressed as:

$$\begin{aligned}\frac{C_\gamma(E_n)}{C_f(E_n)} &= \frac{\varepsilon_\gamma Y_\gamma(E_n)}{\varepsilon_f Y_f(E_n)} \\ &= \frac{\varepsilon_\gamma \sigma_\gamma(E_n) \cancel{N_{^{233}U}} \cancel{\Phi_n}(E_n)}{\varepsilon_f \sigma_f(E_n) \cancel{N_{^{233}U}} \cancel{\Phi_n}(E_n)} \\ &= \frac{\varepsilon_\gamma \sigma_\gamma(E_n)}{\varepsilon_f \sigma_f(E_n)} \\ &= k \frac{\sigma_\gamma(E_n)}{\sigma_f(E_n)}\end{aligned}$$

Los Alamos NATIONAL LABORATORY

Capture to fission ratio

The capture to fission ratio is given by:

$$\alpha(E_n) \equiv \frac{\sigma_\gamma(E_n)}{\sigma_f(E_n)}$$

We measure a number of events Ci as a function of the neutron energy associated with process i (fission or capture):

$$C_i(E_n) = \varepsilon_i Y_i(E_n)$$

and

$$Y_i(E_n) = \sigma_i(E_n) N_{^{233}U} \Phi_n(E_n)$$

Therefore, the capture to fission ratio can be expressed as:

$$\begin{aligned}
\frac{C_\gamma(E_n)}{C_f(E_n)} &= \frac{\varepsilon_\gamma Y_\gamma(E_n)}{\varepsilon_f Y_f(E_n)} \\
&= \frac{\varepsilon_\gamma \sigma_\gamma(E_n) \cancel{N_{^{233}U}} \cancel{\Phi_n}(E_n)}{\varepsilon_f \sigma_f(E_n) \cancel{N_{^{233}U}} \cancel{\Phi_n}(E_n)} \\
&= \frac{\varepsilon_\gamma \sigma_\gamma(E_n)}{\varepsilon_f \sigma_f(E_n)} \\
&= k \frac{\sigma_\gamma(E_n)}{\sigma_f(E_n)}
\end{aligned}$$

Hence:

$$\alpha(E_n) \equiv \frac{1}{k} \frac{C_\gamma(E_n)}{C_f(E_n)}$$

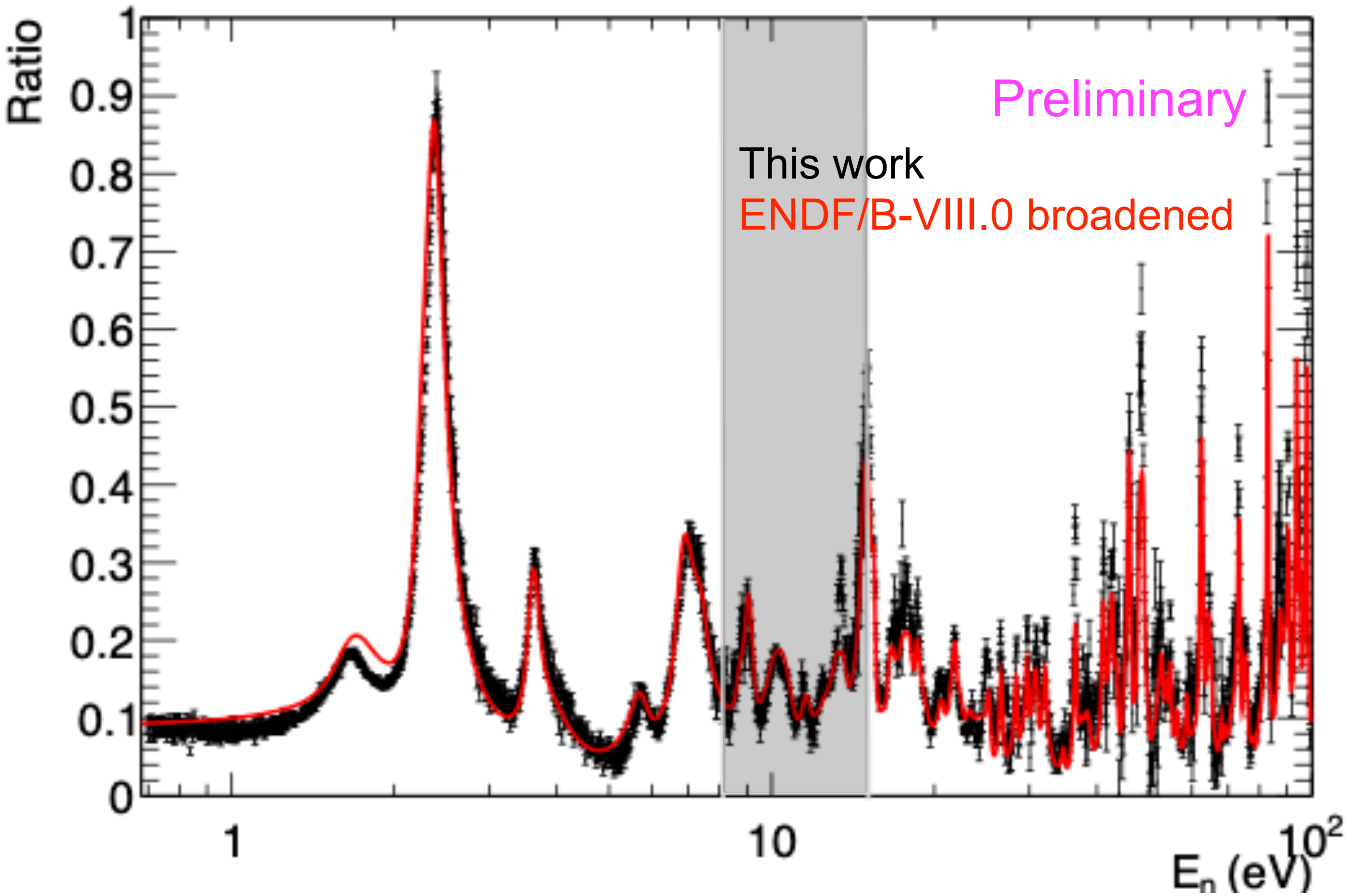
Capture to fission ratio

- Experimental advantages of the capture to fission ratio:
  - It is much simpler and more reliable to determine experimentally as many of the systematic questions:
    - Sample mass
    - Self-shielding
    - Neutron exposure

  will cancel out in an appropriately designed experiment.

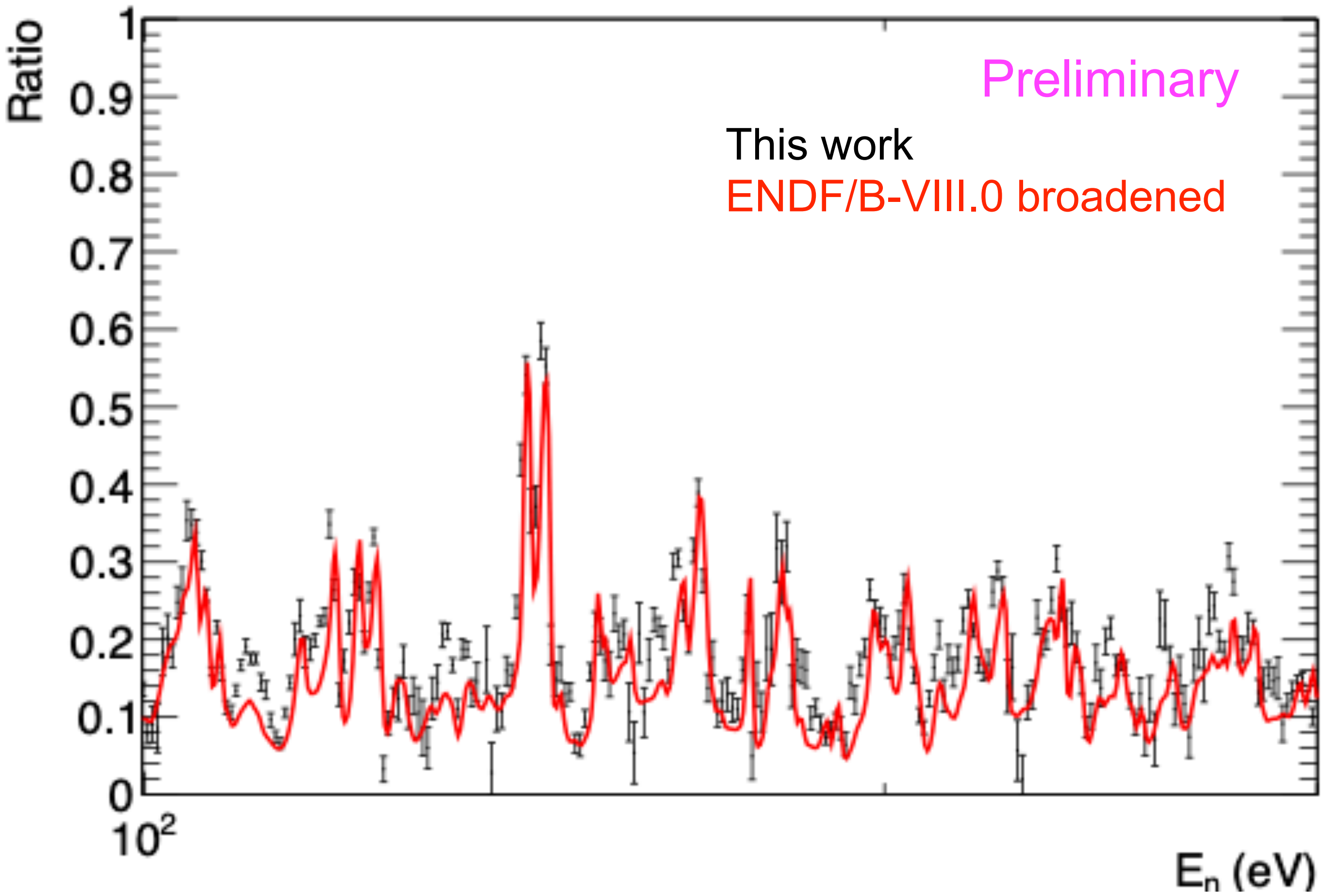
Capture to fission ratio

- Normalization to **ENDF/B-VIII.0** broadened cross section ratio in the neutron energy region suggested by the Evaluators (8.1-14.7) eV:



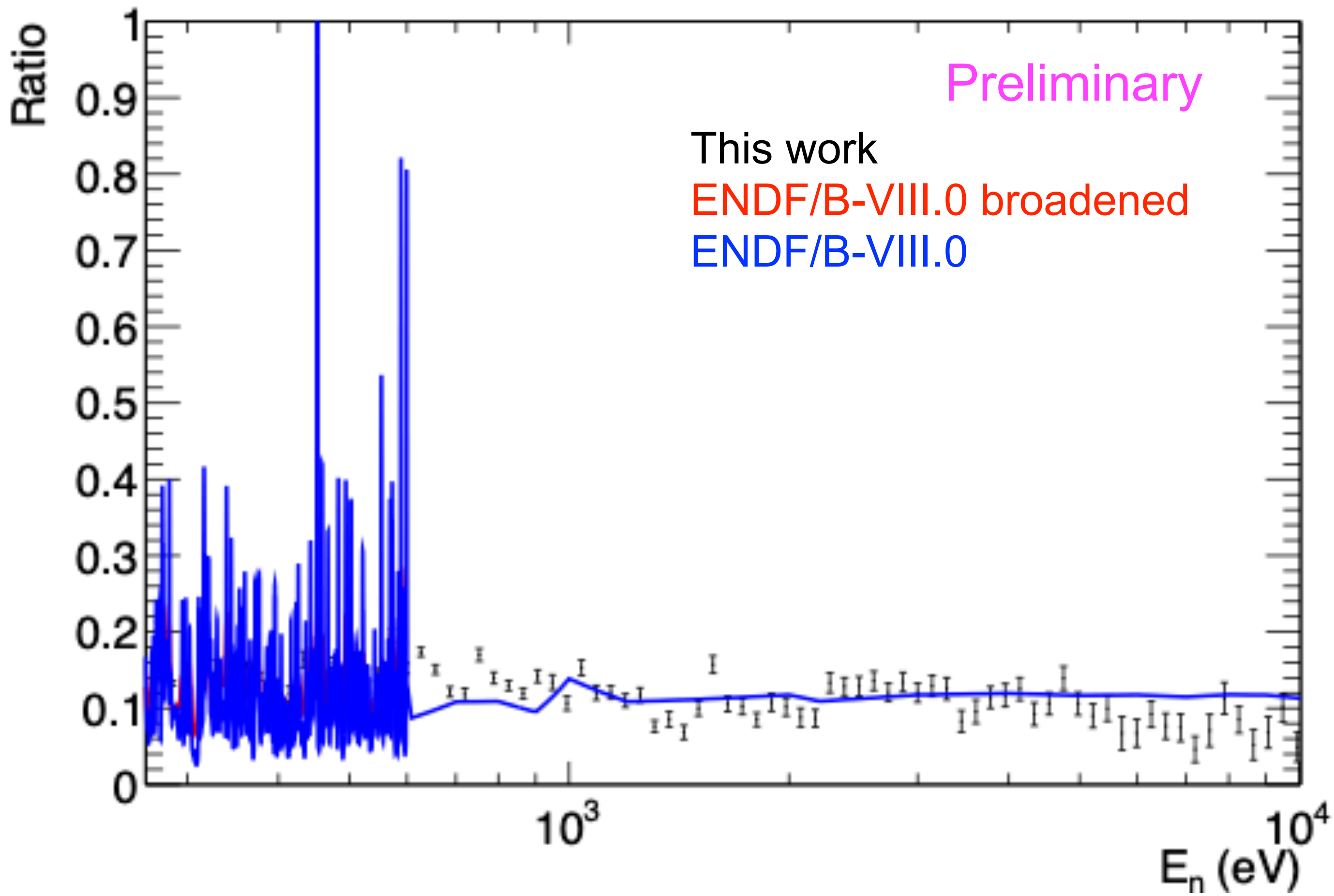
Capture to fission ratio

- Normalization to **ENDF/B-VIII.0** broadened cross section ratio in the neutron energy region suggested by the Evaluators (8.1-14.7) eV:



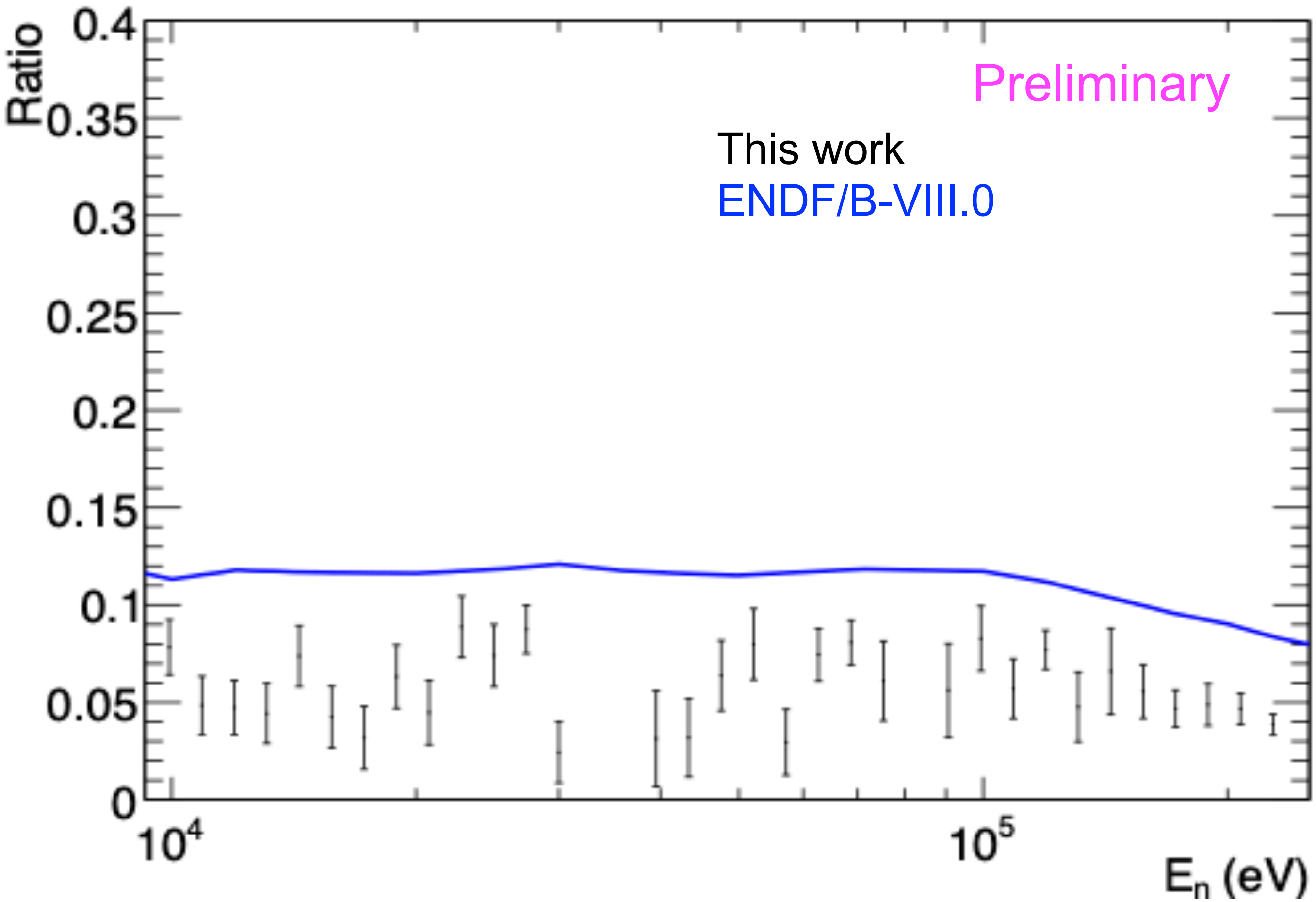
Capture to fission ratio

- Normalization to **ENDF/B-VIII.0** broadened cross section ratio in the neutron energy region suggested by the Evaluators (8.1-14.7) eV:

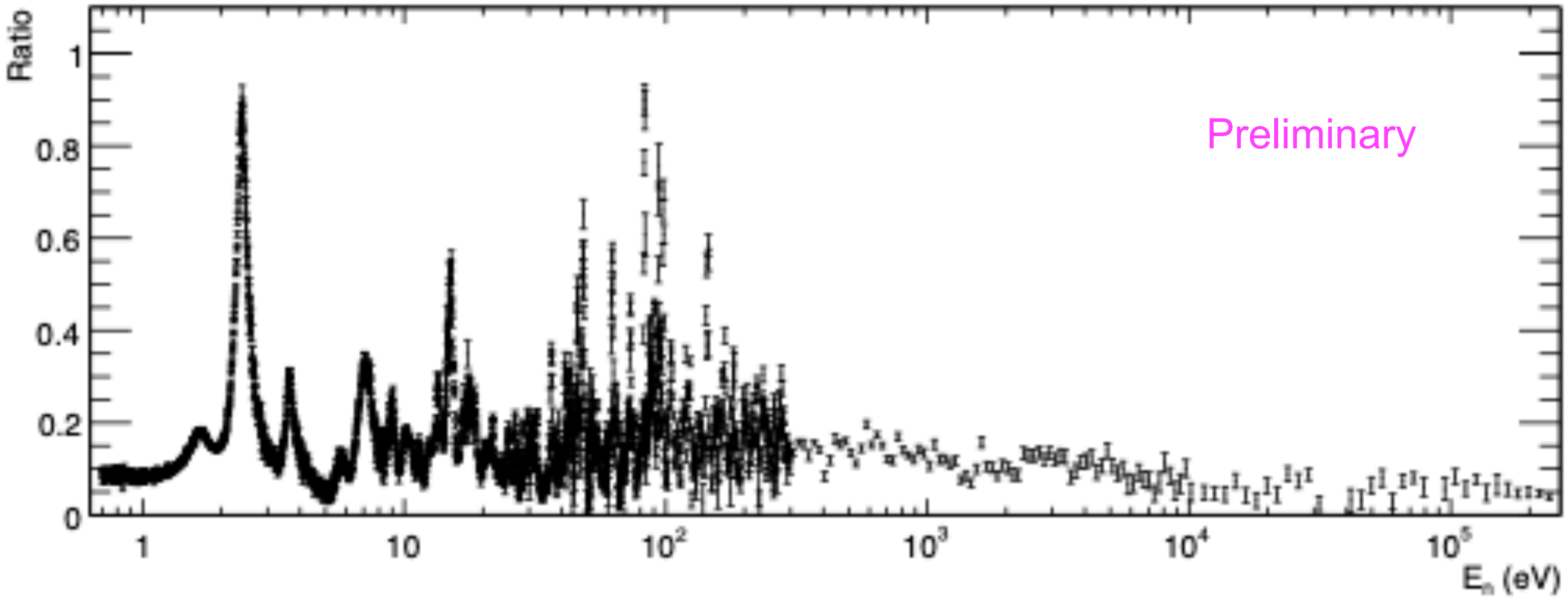


Capture to fission ratio

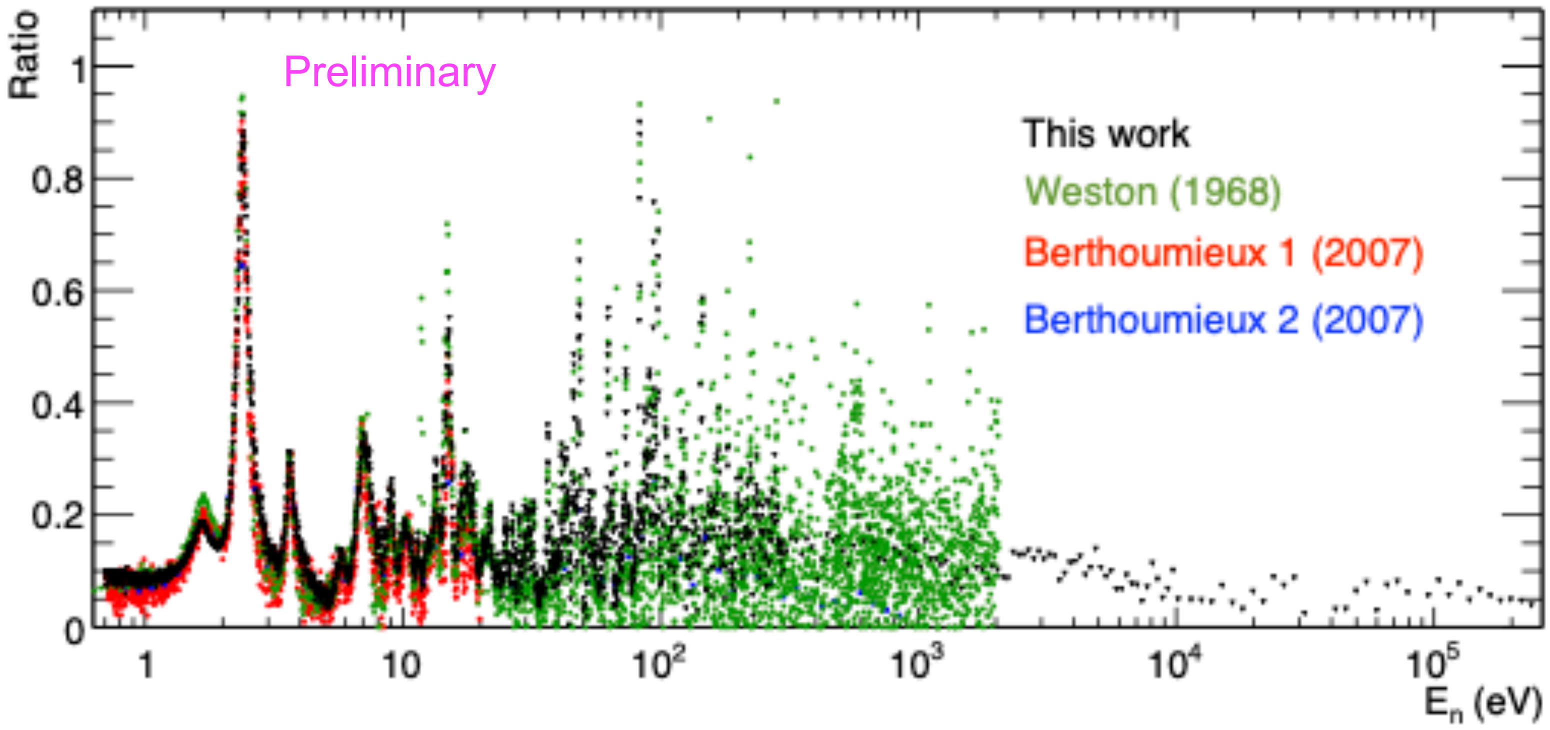
- Normalization to **ENDF/B-VIII.0** broadened cross section ratio in the neutron energy region suggested by the Evaluators (8.1-14.7) eV:



Capture to fission ratio



Capture to fission ratio



Conclusions

- New measurement at LANSCE combining DANCE and NEUANCE at the end of 2020 and 2021.
- The ^{233}U material was provided by Oak Ridge National Laboratory (December 2020).
- Two samples of 10 mg and 20 mg of ^{233}U have been prepared at LANL by Evelyn M. Bond (December 2020).
- Data analysis has been finished and results of the capture to fission ratio on ^{233}U in the neutron energy region from 0.7 eV to 250 keV have been provided.
- The focus was to provide data from 1-300 keV. We are providing data from 0.7 eV to 1 keV in addition.
- The result has been normalized to the ENDF/B-VIII.0 broadened capture to fission cross section ratio in the neutron energy region recommended by the Evaluators, between 8.1 and 14.7 eV.
- This is the first measurement of the capture to fission ratio above 2 keV.
- The data show some small differences in the RRR with the evaluation though the general trend is consistent.
- In the URR this data show a smaller capture to fission ratio than the evaluation.
- We are working with Luis Leal and Marco Pigni to get the data into evaluation.
- Ionel Stetcu expressed interest in the high energy data.

Acknowledgements

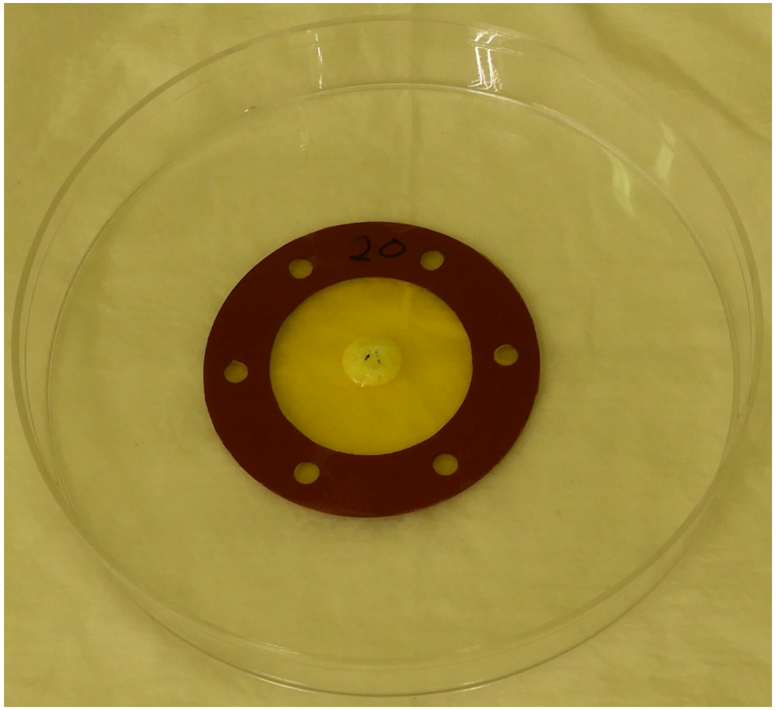
This work was supported by the Nuclear Criticality Safety Program, funded and managed by the National Nuclear Security Administration for the Department of Energy.

The ^{233}U was supplied by DOE/SC Isotope Program.
Thanks to our collaborators John Ullmann (P-3), Cathleen Fry (P-3) and Todd A. Bredeweg (C-NR) and Evelyn M. Bond.

^{233}U targets

- The 30 mg of ^{233}U were supplied from Oak Ridge National Laboratory (ORNL).
- Material composition:

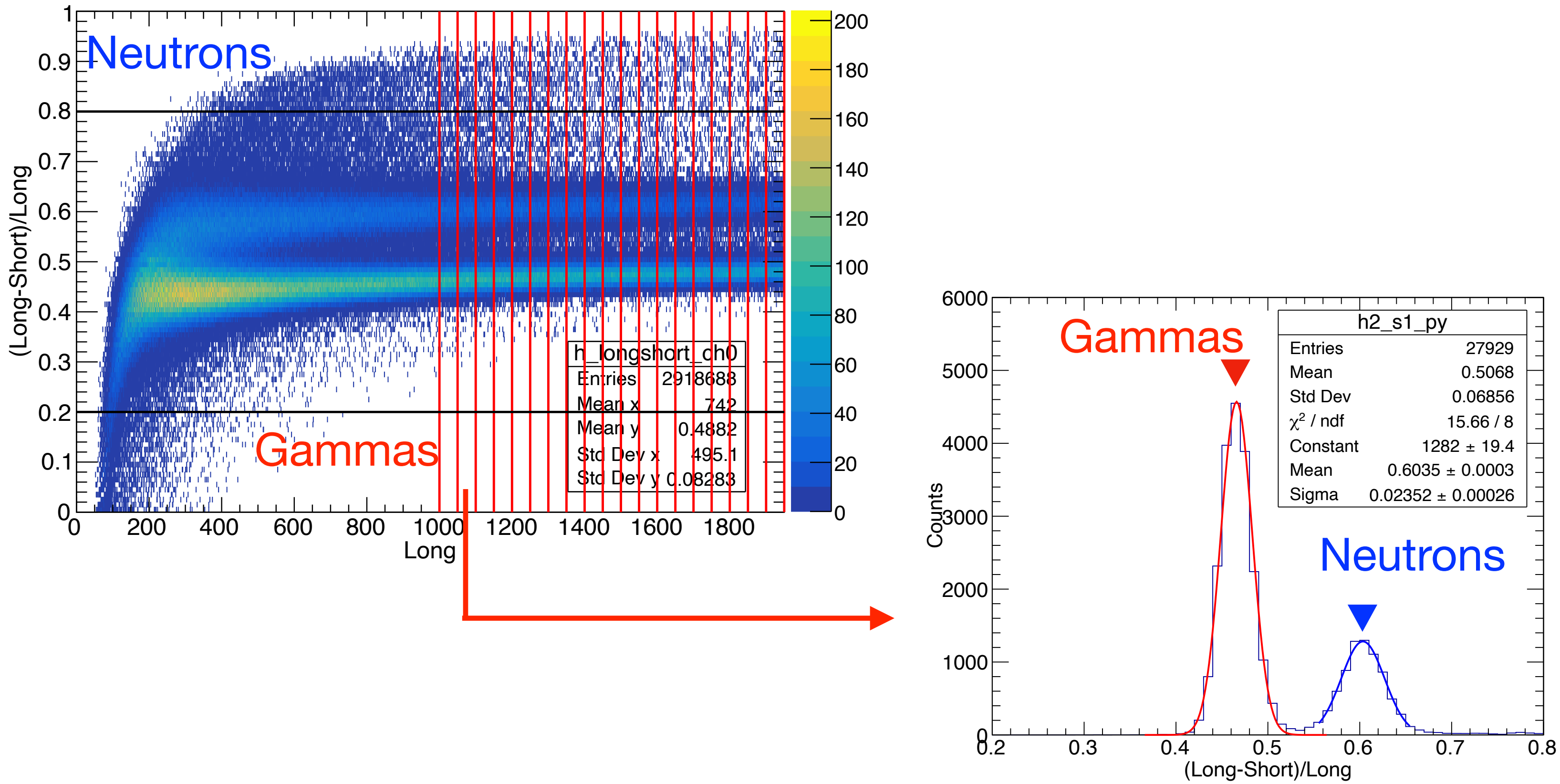
Isotope	Atom (%)
^{233}U	99.9843
^{234}U	<0.0002
^{235}U	0.0017
^{236}U	0.0004
^{238}U	0.0134



- Two samples have been prepared by Evelyn M. Bond at LANL.
  - 20 mg
  - 10 mg

PSD NEUANCE

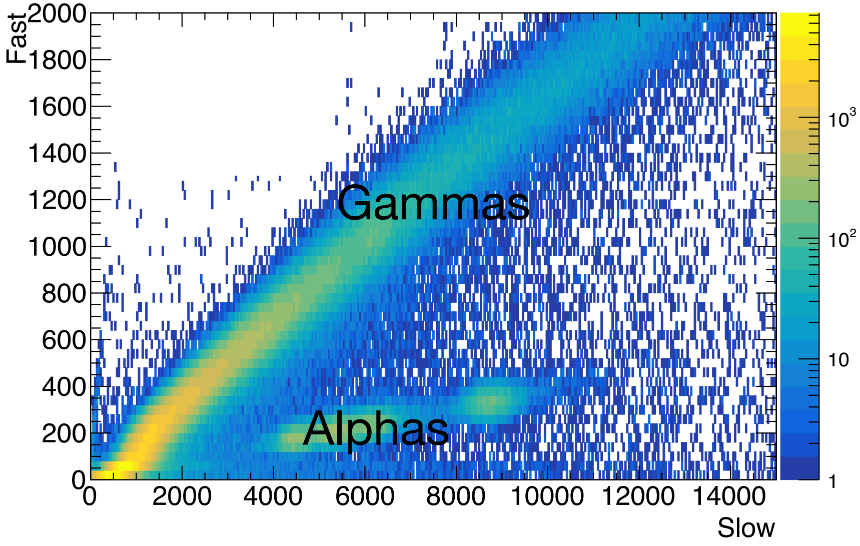
- Neutrons & gammas separation using the plot (long-short)/long vs long.

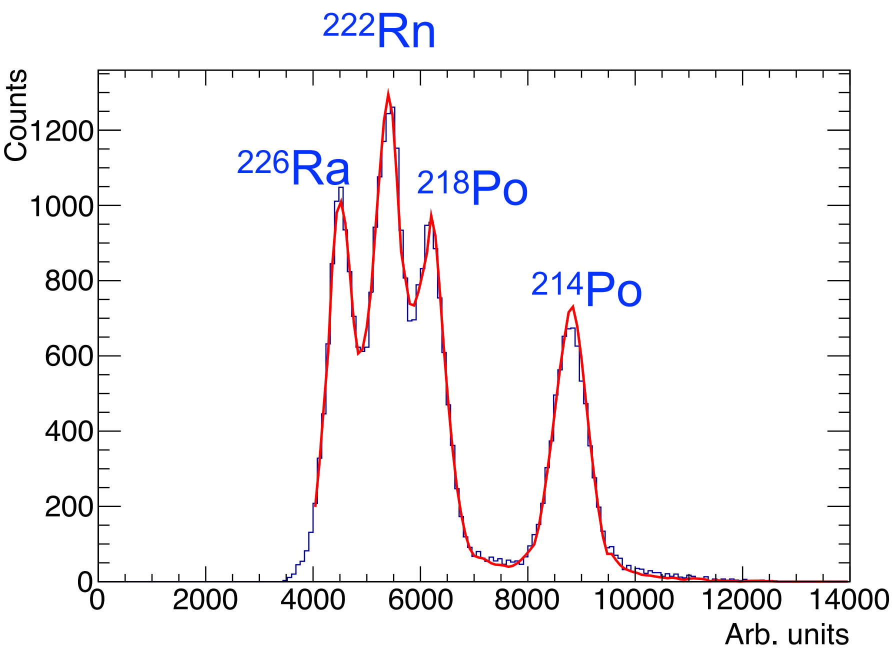


- Clear discrimination between fission neutrons and γ-rays.

DANCE calibrations

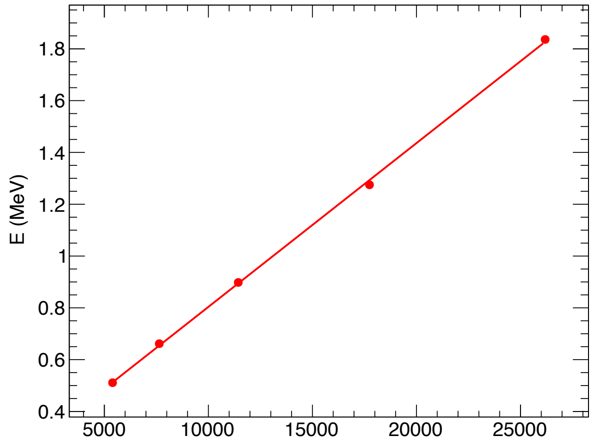
- Intrinsic radioactivity of BaF_2 used to calibrate the DANCE crystals.
- Using the Alpha-decay chain of the ^{226}Ra present in the BaF_2.
  - ^{226}Ra (4.8 MeV)
  - ^{222}Rn (5.5 MeV)
  - ^{218}Po (6.0 MeV)
  - ^{214}Po (7.7 MeV)

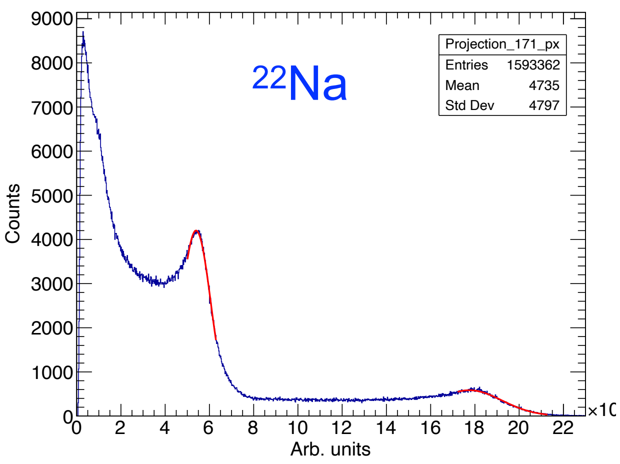


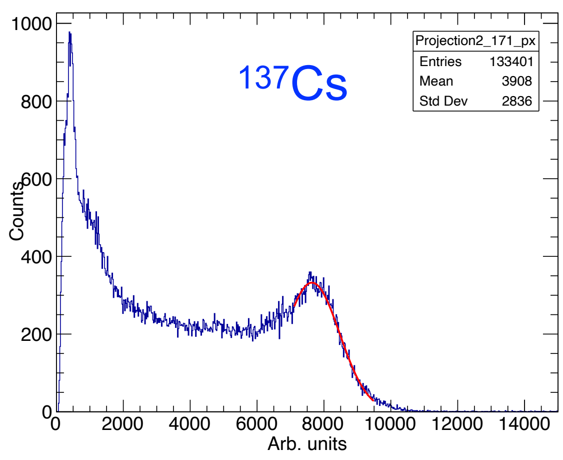


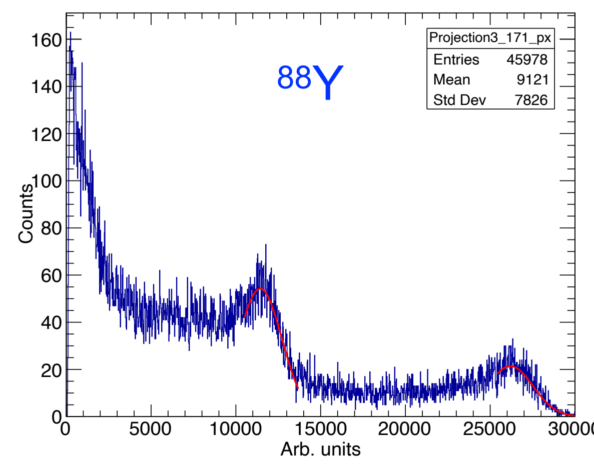
NEUANCE calibrations

- Calibration using gamma sources:
  - ^{22}Na (511 keV and 1274.537 keV).
  - ^{137}Cs (661.657 keV).
  - ^{88}Y (898.047 keV and 1836.090 keV).









Los Alamos
NATIONAL LABORATORY