Measurement of Fission Yields at the National Synchrotron Light Source II

M. Topsakal, A. Mattera, S. Ota, A. A. Sonzogni, E. A. Ricard, S. Gill, C. Morse, M. Jandel

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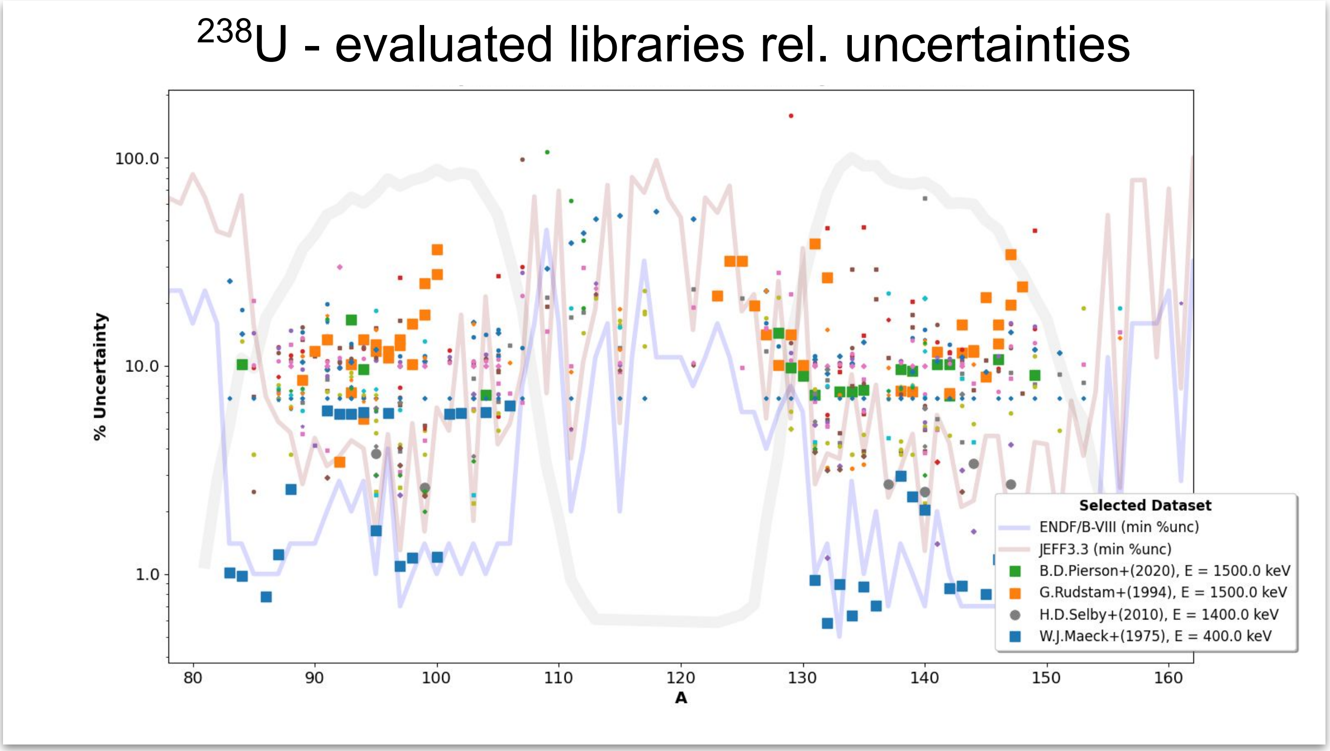
Outline

- Background: an overview of uncertainties in FPY measurements
- A complementary technique to AMS
- First measurements of reaction products at BNL
- Outlook

A brief history of Fission Yields evaluation in ENDF



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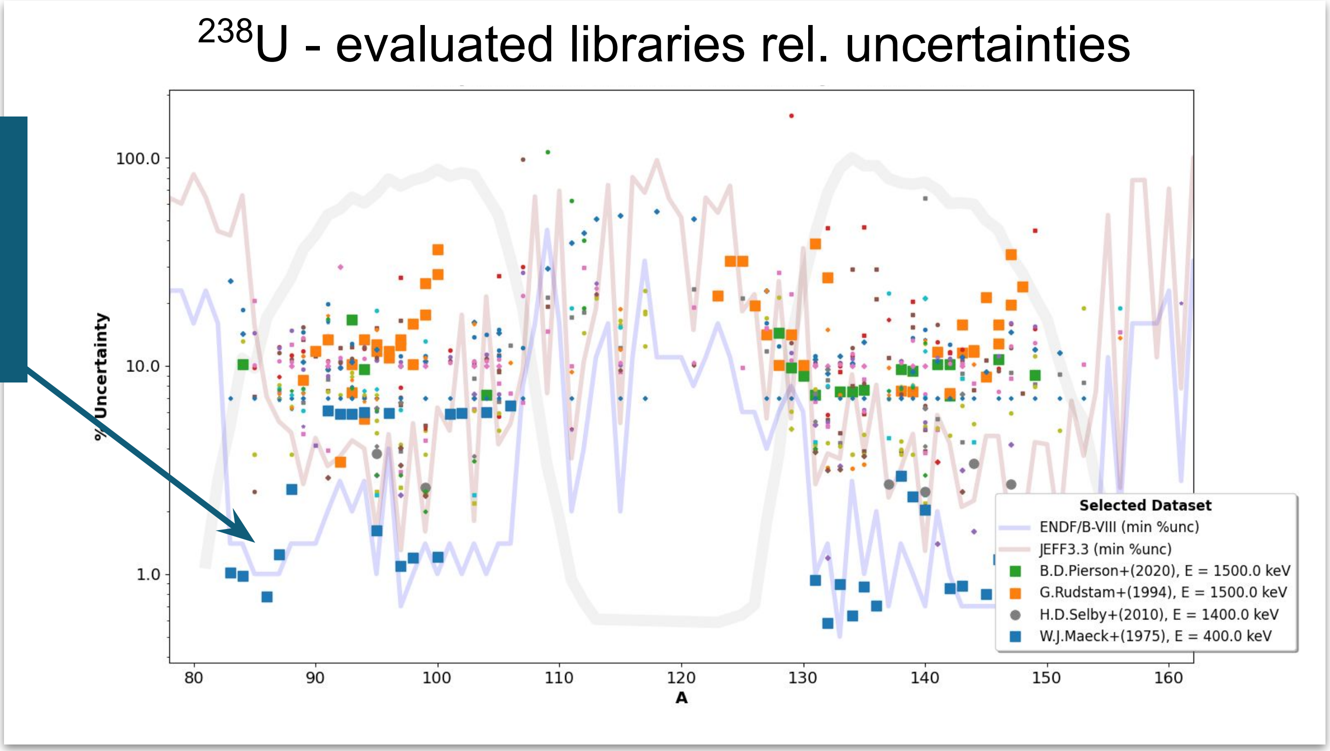
A brief history of Fission Yields evaluation in ENDF

ENDF/B uncertainties follow closely these experimental data



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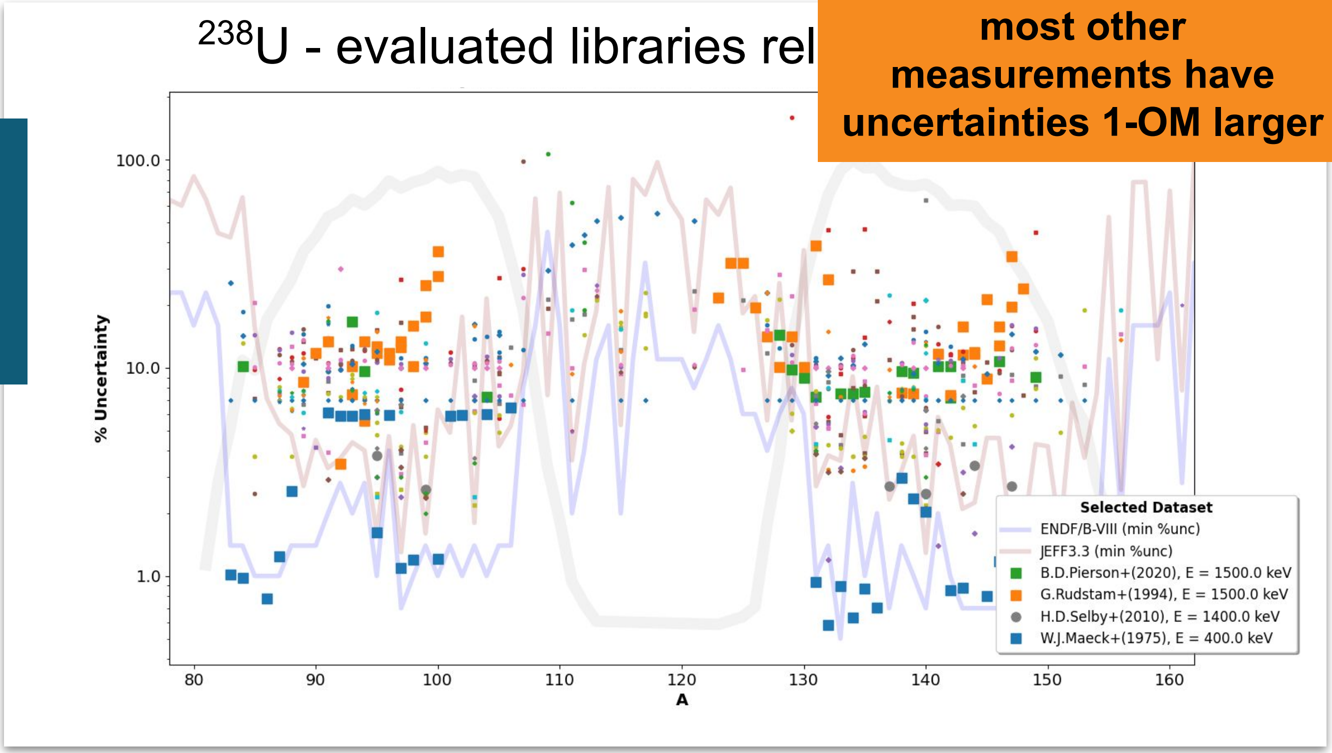

most other measurements have uncertainties 1-OM larger

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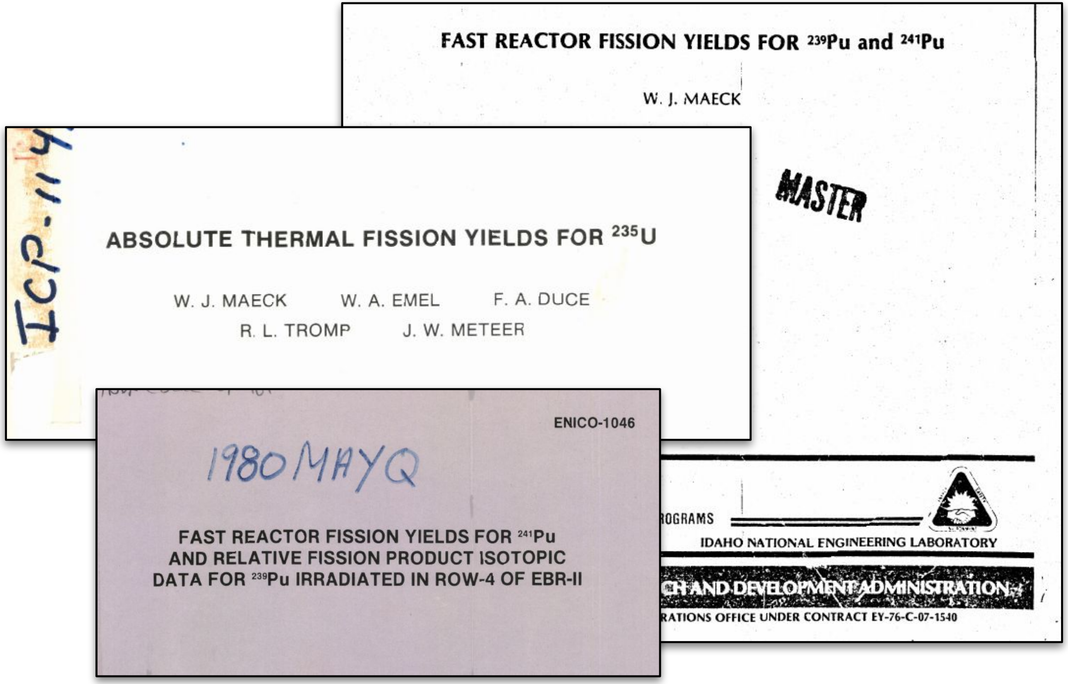
The most precise measurements ever made…

Results of measurements at Idaho National Lab reactor (ETR and EBR), and only published in a series of INL technical reports

Isotope Dilution Mass Spectrometry on $^{239,241}Pu$ and ^{235}U targets irradiated with thermal & fast neutrons



1981KO45, **1981KO46**
EXFOR: 21155
(recently updated to include uncertainties and additional data)

Someone else did the same

$^{233,235,236,238}U$, ^{237}Np, $^{239,240,241,242}Pu$, ^{241}Am fast fission

Burnup measurement at RAPSODIE fast reactor (Cadarache, FR)

Published in the 80s, it was not included in the very early evaluations (Meek).

E&R included the measurements in their compilation, but labeled them as RC and assign an uncertainty of 10% (maybe because data in 81KO45 were provided without uncertainties? Now included using data from 1981KO46).

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CUMULATIVE FAST REACTOR FISSION YIELDS OF ^{233}U, ^{235}U, ^{236}U, ^{238}U, ^{237}Np, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am

L. KOCH, K. KAMMERICHS, G. COTTONE, D. STEINERT, R. DE MEESTER, J. HEITZ, R. MOLINET, C. RIJKEBOER

COMMISSION OF THE EUROPEAN COMMUNITIES
JOINT RESEARCH CENTRE, KARLSRUHE ESTABLISHMENT
EUROPEAN INSTITUTE FOR TRANSURANIUM ELEMENTS

Why are they so precise?

Uncertainty Budget

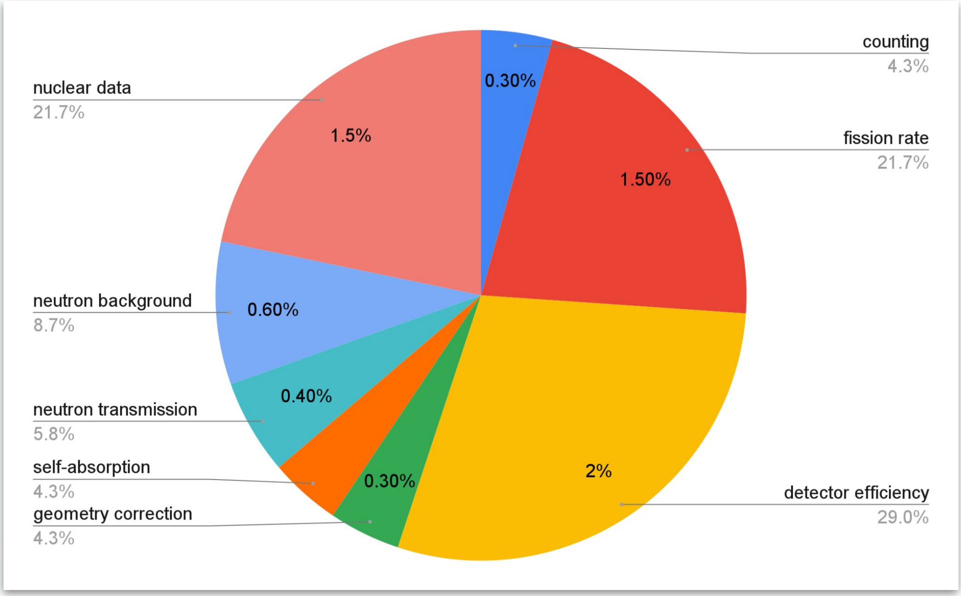

TABLE 6.1.1. UNCERTAINTY SOURCES OF ABSOLUTE FISSION YIELD MEASUREMENT WITH THE γ spectroMETRIC METHOD

error sources	*values (%)*	*remarks*
a. statistical (random) Δy_{sta}	0.3-2.1	depends on measuring conditions [a]
b. fission rate	1.5	
c. detector efficiency calibration	2.0	depends on γ ray energy
d. geometry correction	0.3	
e. γ self-absorption correction	0.3-0.4	
f. neutron transmission correction	0.4	(for thermal neutrons)
g. neutron background correction	0.6-1.0	(for high energy neutrons)
total systematic error	2.6-2.7	$\Delta y_{sys} = \sqrt{\sum_i \Delta y^2_{sys_i}}$
total error	2.6-3.4	$\Delta y_{tot} = \sqrt{\Delta y^2_{sys} + \Delta y^2_{sta}}$

IAEA - TECDOC 1168 (2000)

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Why are they so precise?

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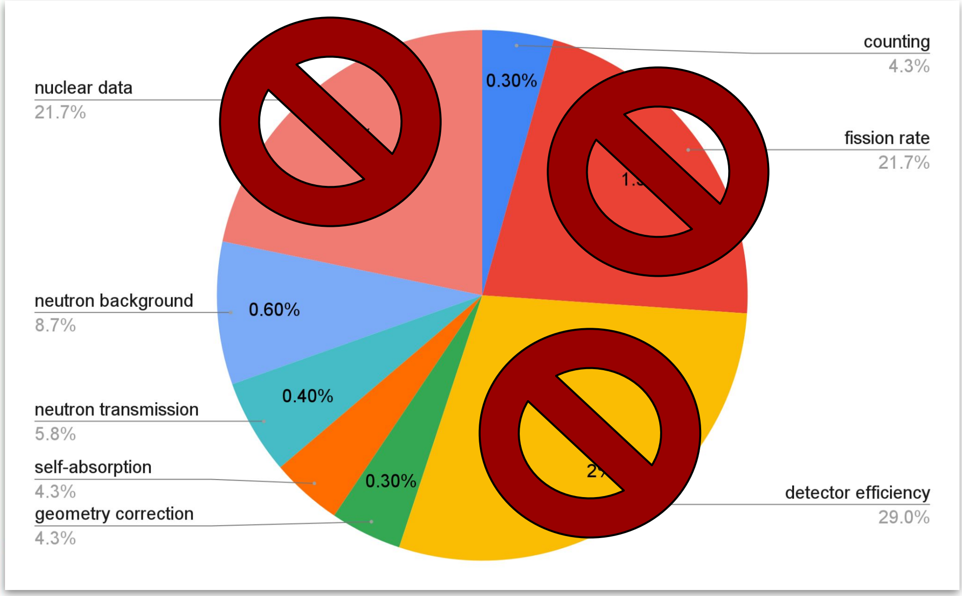

MS measurements make **no use of Nuclear Data** to extract the absolute yields

The **fission rate** does not need to be determined as long as the majority of the yields can be measured (**normalized to 100%** / mass peak)

The uncertainties were estimated as the standard deviation of repeated measurements (possibly not fully considering systematic effects)

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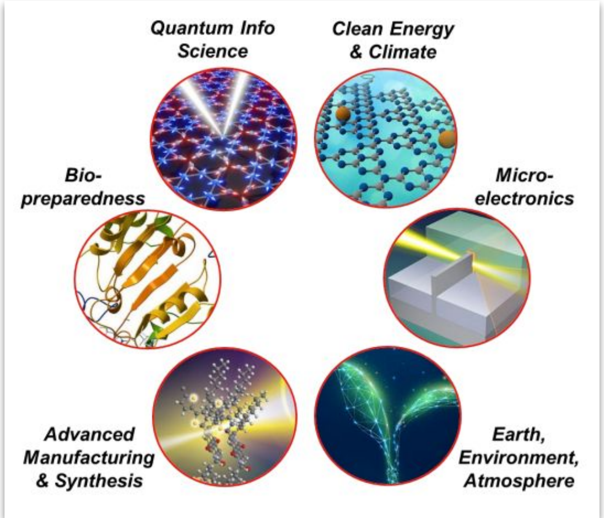
National Synchrotron Light Source II



Vision: as one of the newest, most advanced synchrotron light sources in the world, NSLS-II strives to be an extraordinary hub for the use of synchrotron light to solve the world's most challenging scientific problems.

DOE-SC User Facility

- 29 beam-lines offer the widest spectrum—from infrared to hard x-rays—of synchrotron light among all DOE-SC facilities
- Research focuses on 6 Science Areas

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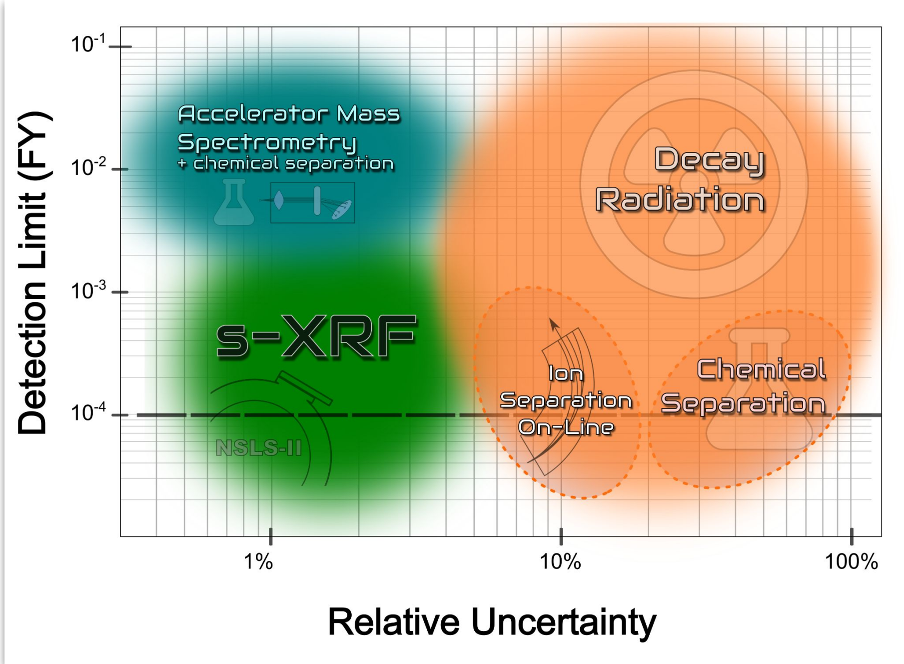
Fission Product Yields at NSLS-II?

detection limit → XRF can reach **ppm-level concentrations** of elements in a substrate: this would allow us to get to **low-yield fission products** and a **full normalization** (no fission rate), with no limitations on the measured elements

A calibration of the setup with standard samples would let us not rely on decay data for determination of the absolute yields

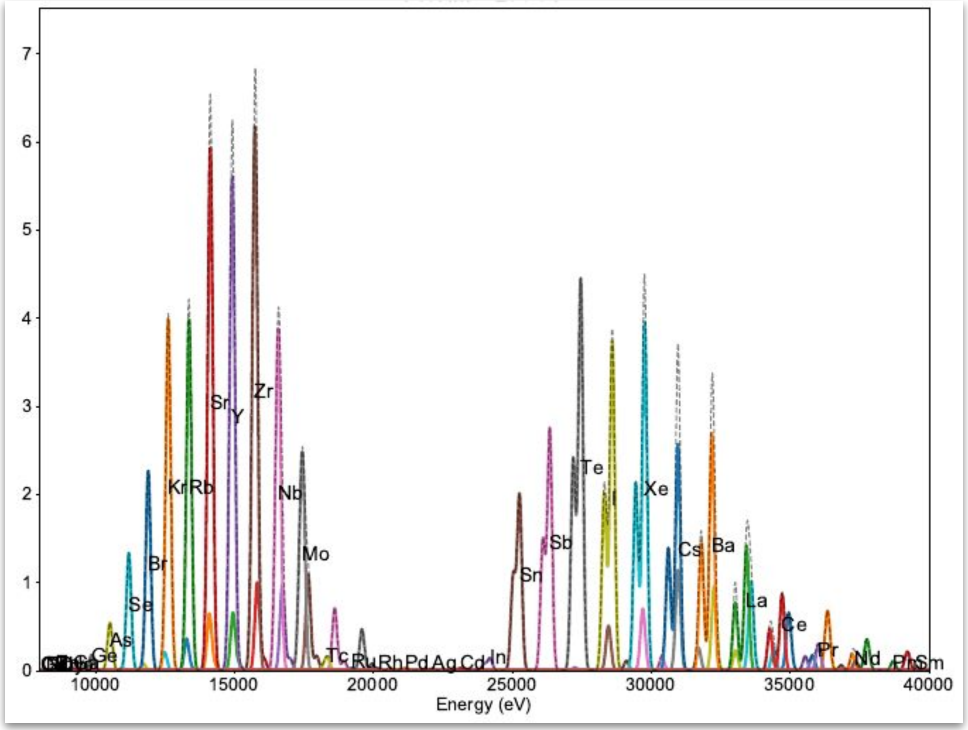


Fission Product Yields at NSLS-II

Long-term goal: use the Light Source to **quantify** the amount of fission products in a sample

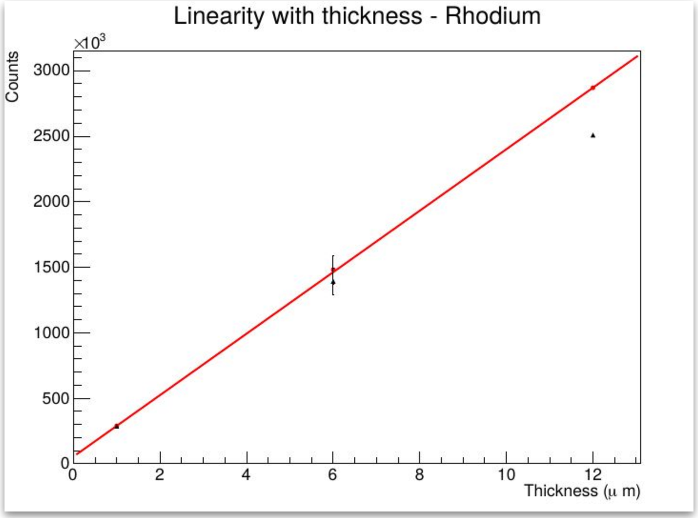
NSLS-II combined with the new Ge-XRF detector at NE can allow us to reach low-yield fission products

Measure Z-distribution (XRF) of long-lived or stable fission products *à-la* Maeck (but looking at elements instead of masses) - it provides different information, but still valuable for an evaluation as an ‘anchor point’

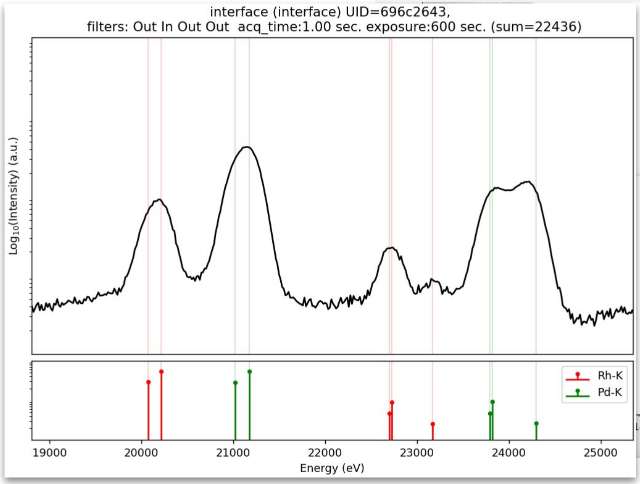


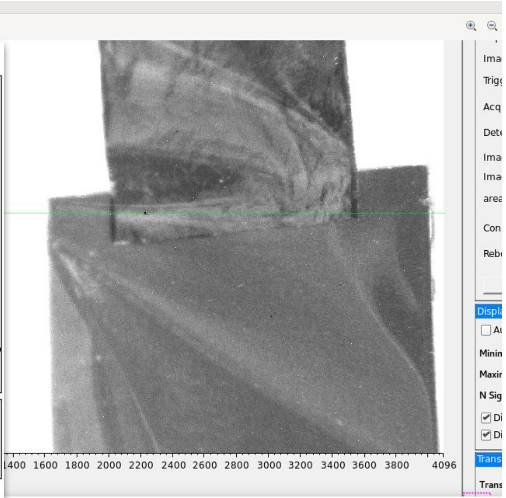
calculated X-ray peaks from the fission products in a ^{238}U sample irradiated in a fast neutron spectrum

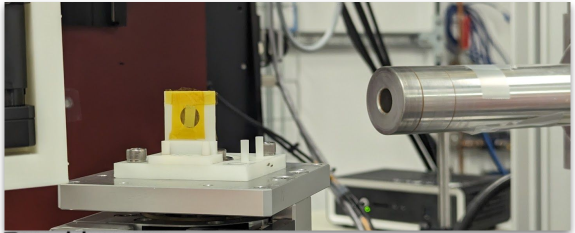
First tests



check of linearity & attenuation corrections







28-ID-2 XPD
beamline

70 keV X-ray beam

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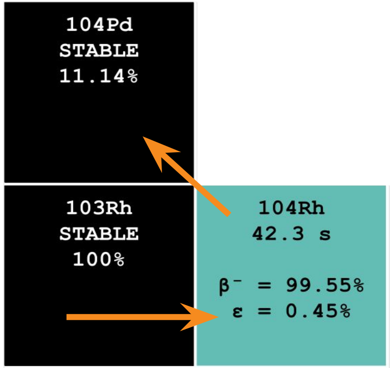
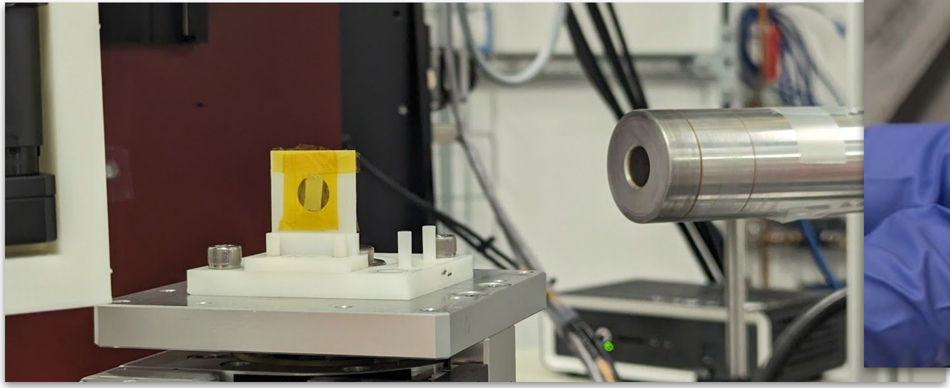
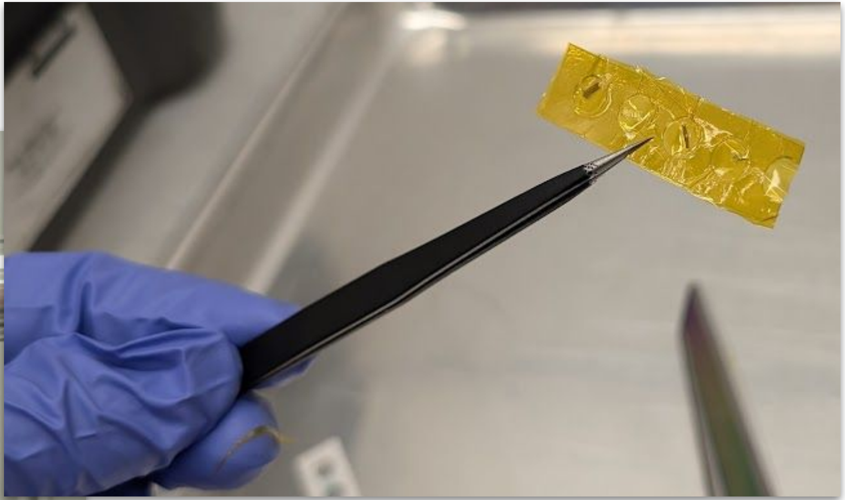
Step 1: Feasibility Study

test the method in a more simple experiment: neutron capture on ^{103}Rh

Rh samples irradiated at Lowell Research Reactor (thermal capture)







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UMASS LOWELL

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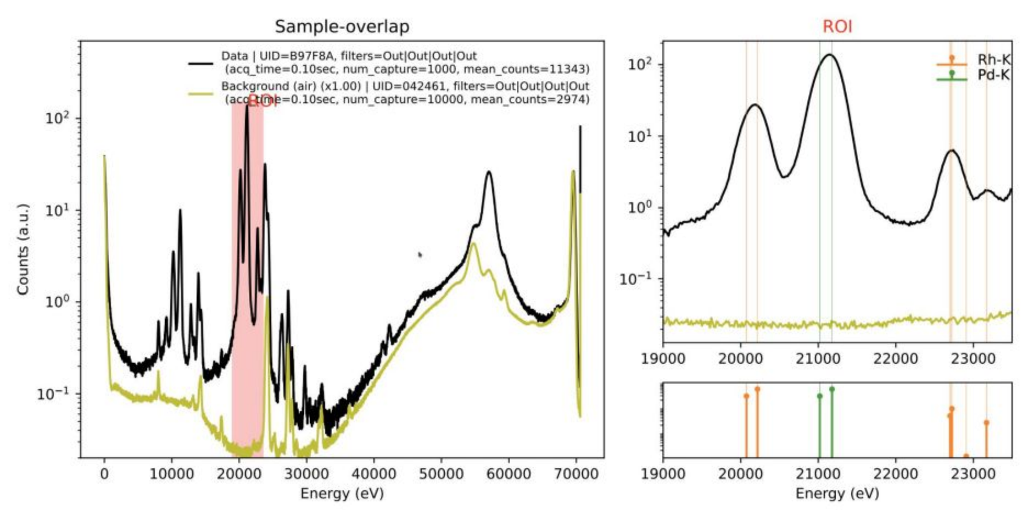
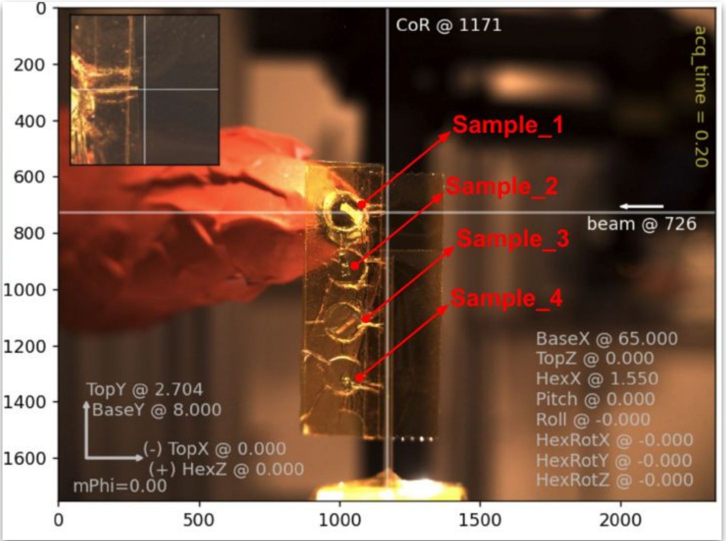
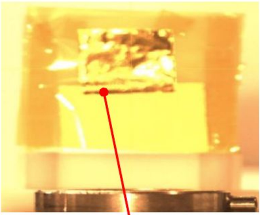
Step 1: Feasibility Study

Oct 2024 run







we confirmed that we can easily quantify elements when concentrations are similar

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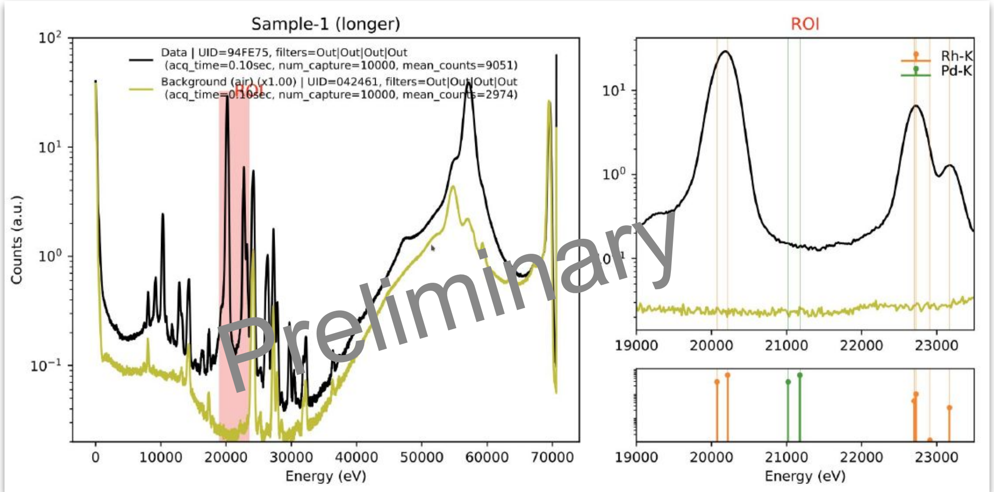
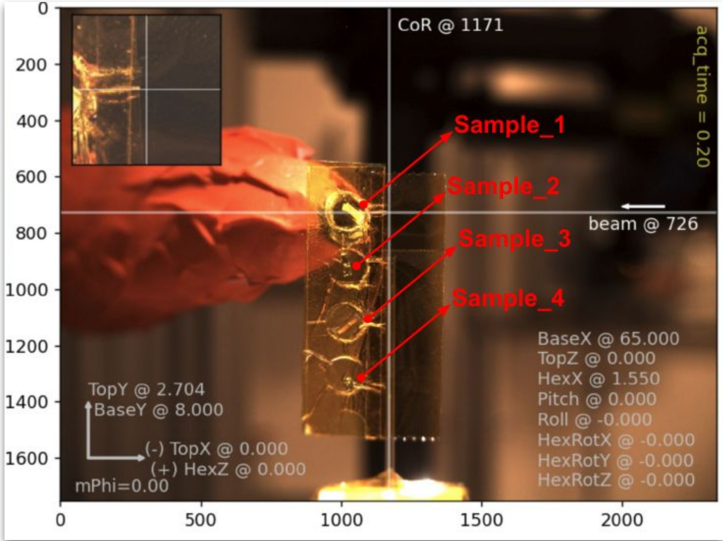
Step 1: Feasibility Study

Oct 2024 run





the Rh peak seems to drown the Pd at product concentrations of ~100 ppm

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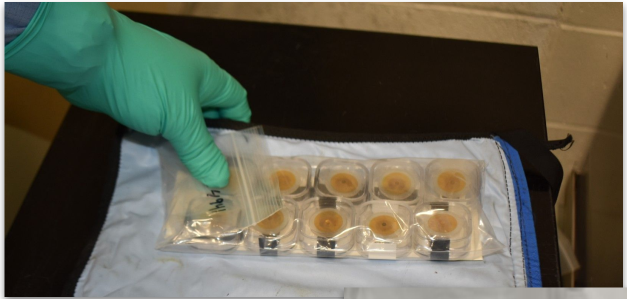
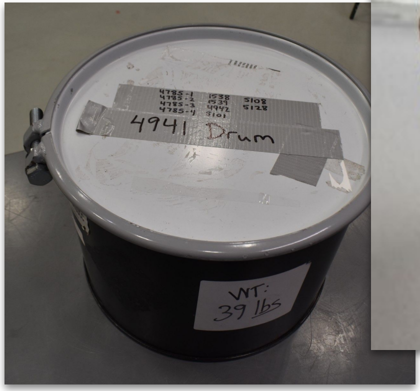
Beamtime with high-dose targets from NSUF

Nuclear Science
User Facilities

Sample	Elem.	Integrated Fluence (n/cm²)	approx. product concentration	Applic.
034-08-331 149-08-331	Ag	1.47×10^{22} 2.9×10^{22}	2-4%	FY
052-08-331 109-08-331 148-08-331	W	1.05×10^{22} 1.58×10^{22} 2.94×10^{22}	5-100‰✠	IF
09-157-033 09-157-034 09-157-035	Hf, Al	1.29×10^{22} 1.3×10^{22} 1.3×10^{22}	100-600 ppm✠	IF
10-242-0011	Mo	1.05×10^{21}	20-30 ppm	FY





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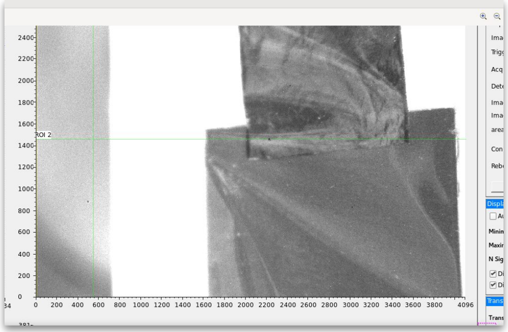
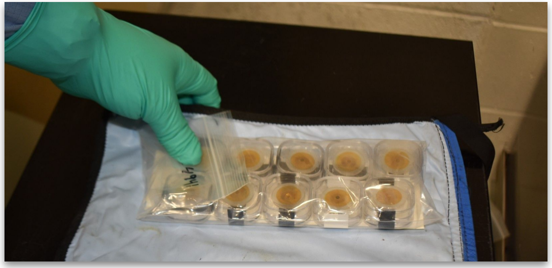
Next steps



- manufacture ad-hoc non-irradiated targets to study the effect of a strong substrate signal on the detection limit
- simulation of signal and full X-ray measurement setup
- using AMS to complement the Z-distributions with A-distributions for specific fission products



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Summary

- Exploratory study to test measurement of reaction rates (and Fission Yields) using s-XRF
- Samples irradiated at UMass Lowell measured at NSLS-2 at the end of October -- no apparent signal from 100 ppm of Pd on the strong Rh background
- Two additional beamtimes scheduled for early 2025 to assay high-dose samples from NSUF and ad-hoc samples fabricated to replicate

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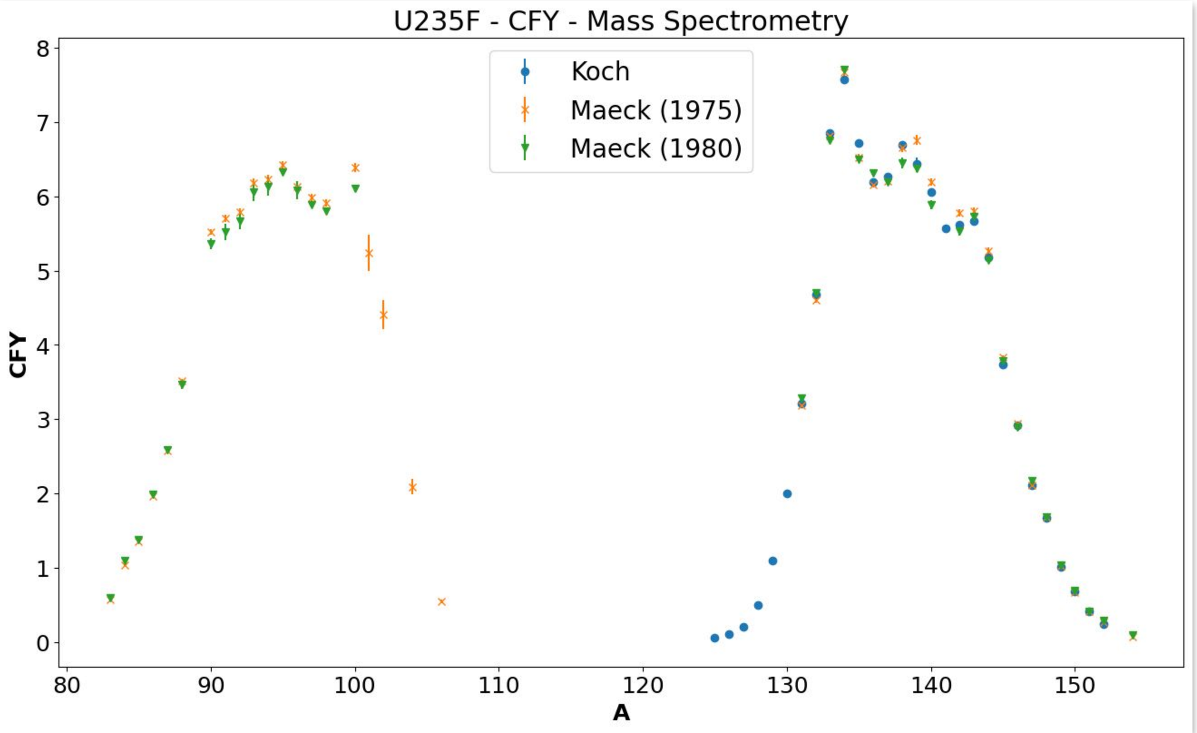
What can be improved?



Ba and Ce elements could not be measured (by Koch) because of **heavy contamination of natural-occurring isotopes** - and Maeck's results were found to be up to 14% too low in a later measurement by the same group (several times the quoted error).

Maeck also had difficulties measuring **low-yield fission products (< 0.1 %)** and isotopes of Iodine, Tellurium and Ruthenium ~70% of the fission products in the heavy peak could measured by Koch; ~90% by Maeck

Maeck **never published** final results in a peer-reviewed publication

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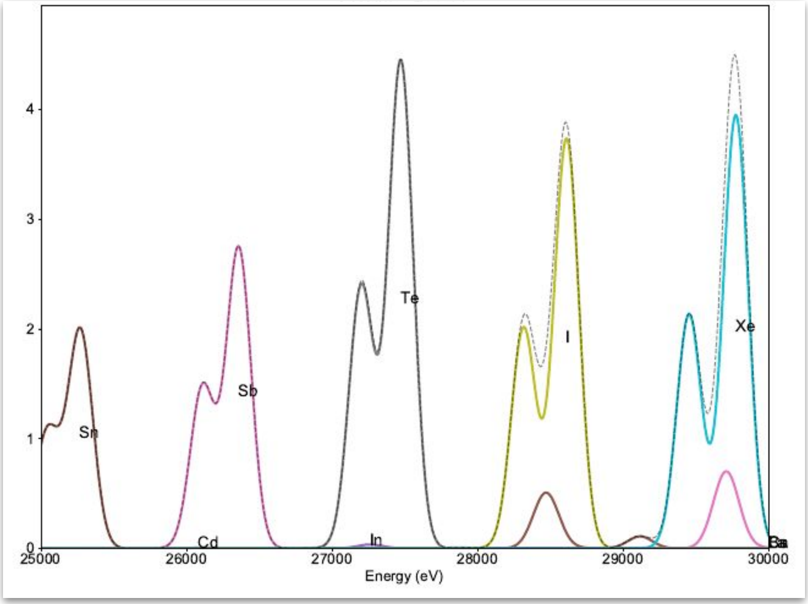
Challenges

activation level / contamination of samples (NSLS-II can measure only low-activity radioactive samples) - it may take too long to irradiate and let samples decay

ability of the X-ray detector to resolve contributions in a complex spectrum (fission yields)

long irradiations → correction for n-capture in fission products



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