



U.S. DEPARTMENT
of ENERGY

Measurement of fission product yields

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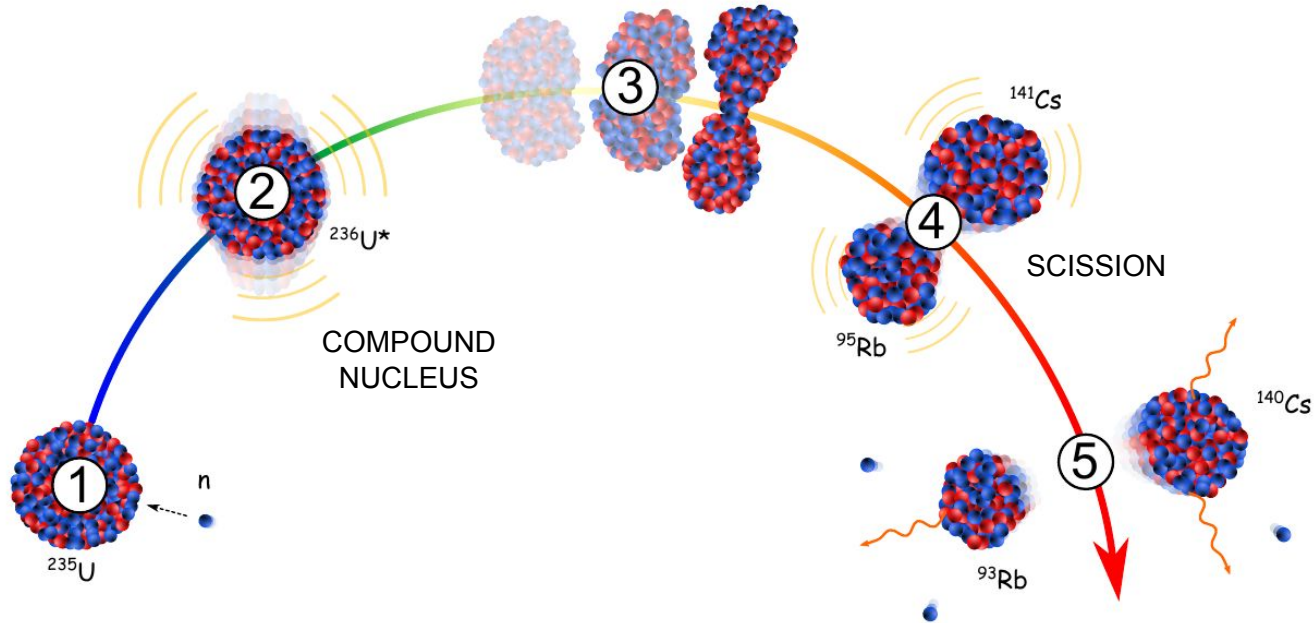
National Nuclear
Data Center



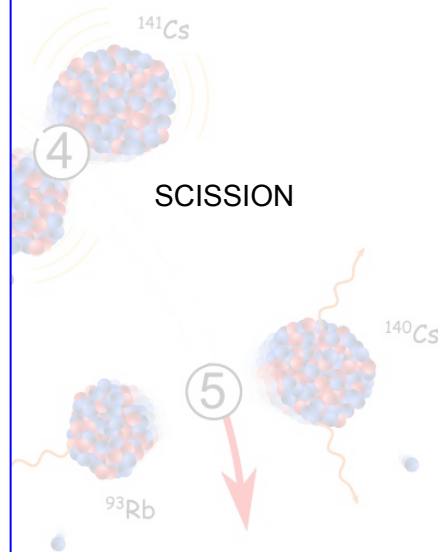
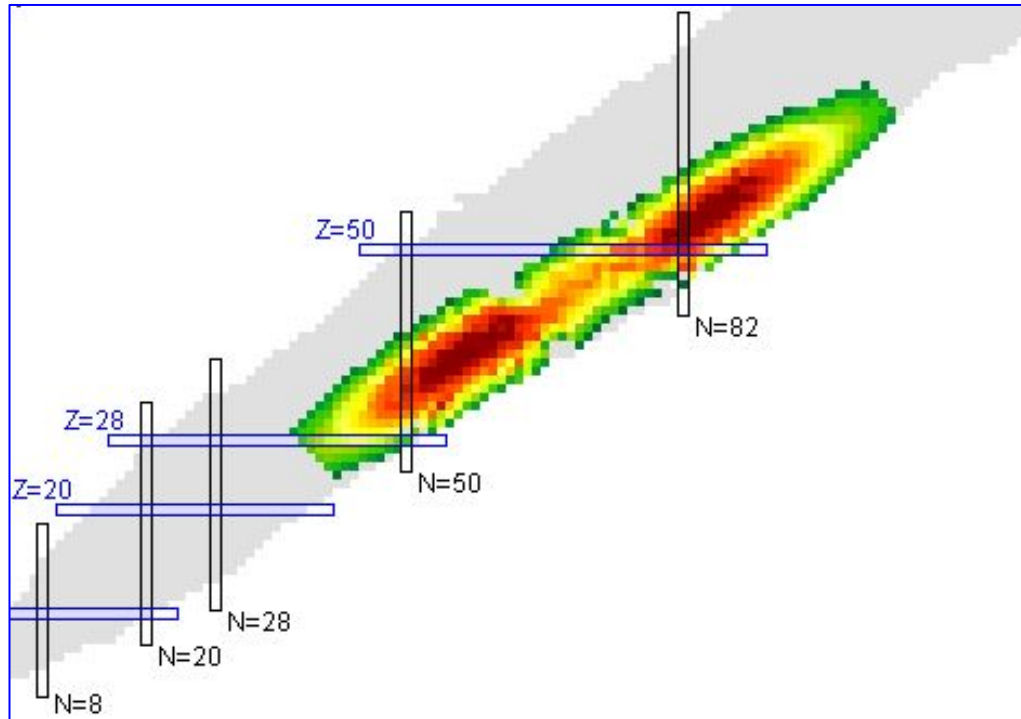
Summary

- Background: fission and fission product yields
- Experimental Setup
- Analysis
- Results
- Lessons Learned

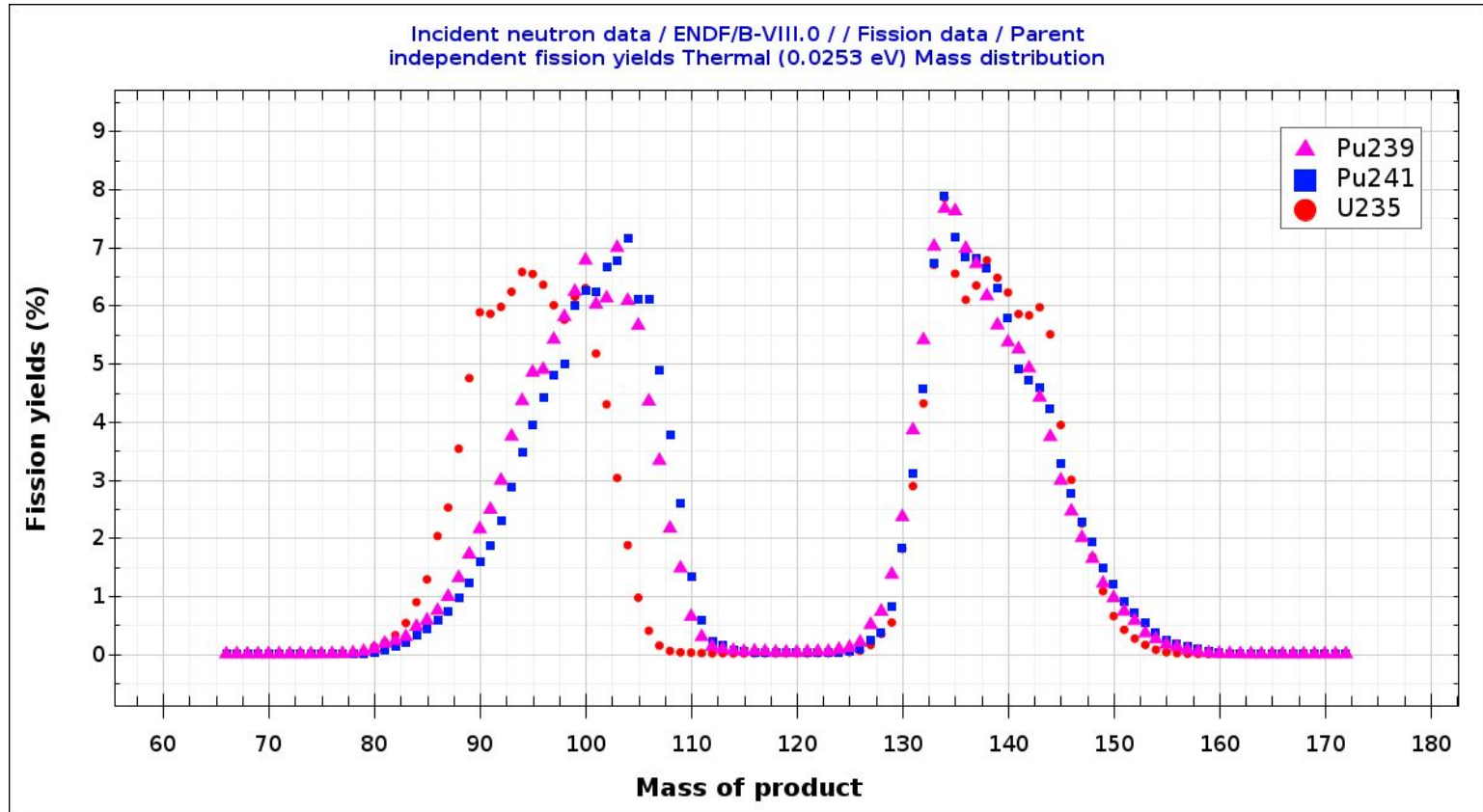
Fission



Fission



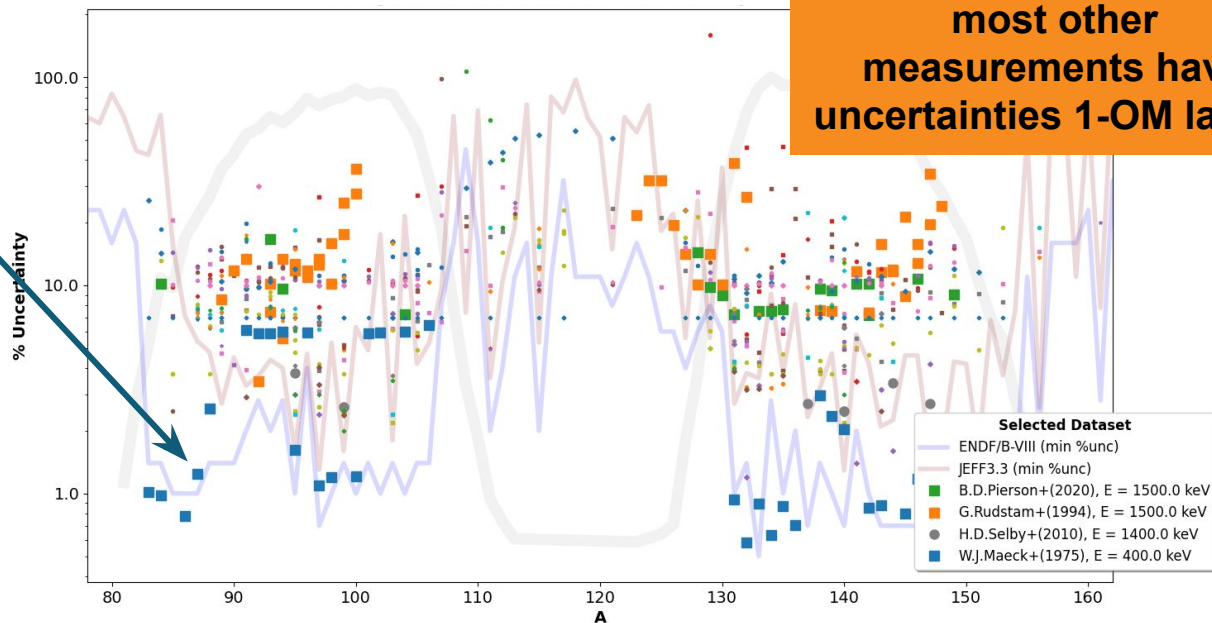
Fission Product distributions



Long-lived fission products in evaluations

^{238}U - evaluated libraries rel. uncertainties

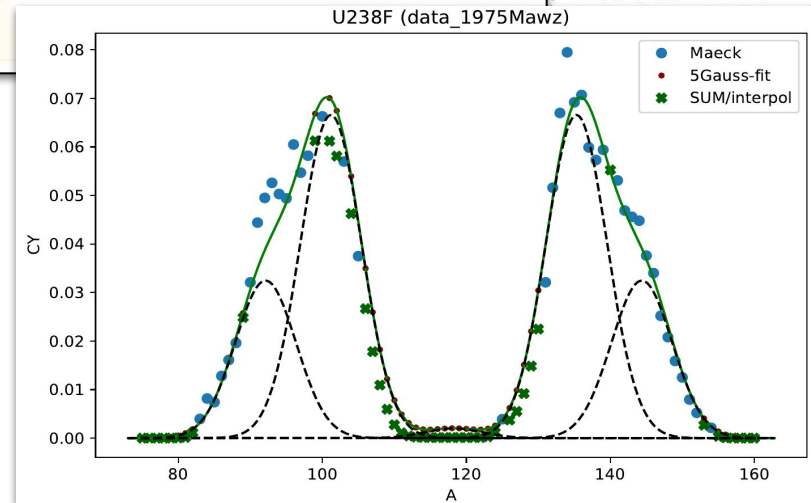
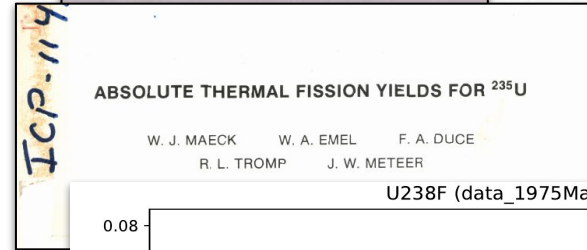
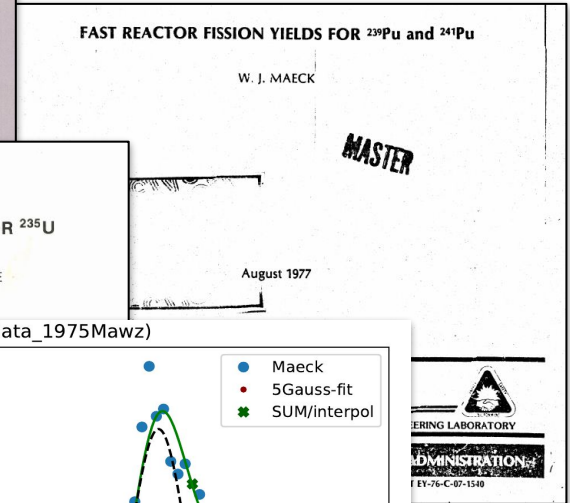
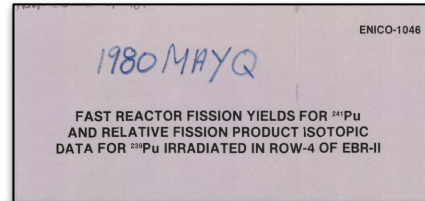
ENDF/B uncertainties follow closely these experimental data



Measurement campaign at EBR-II - 1970s-80s

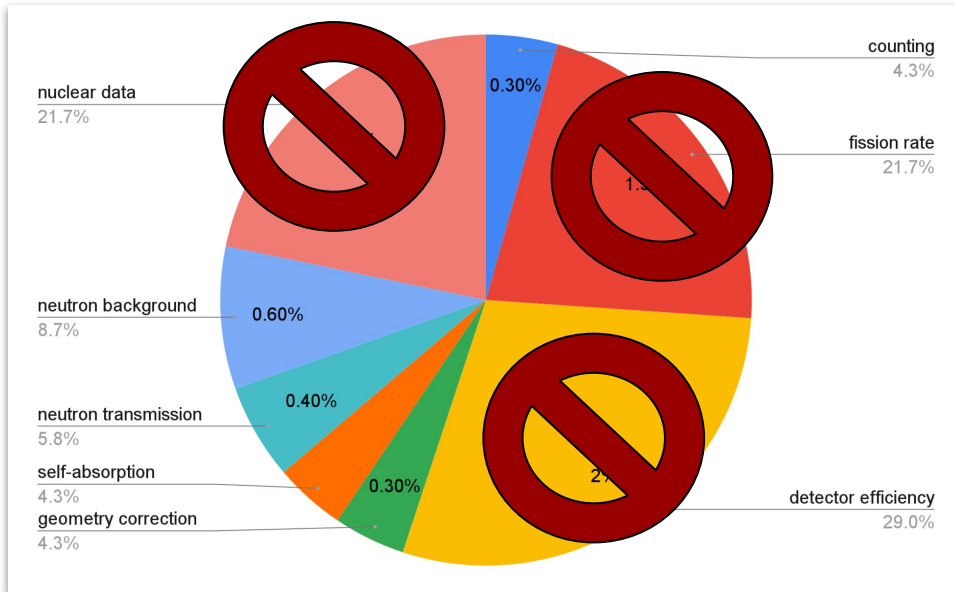
Results of measurements at Idaho National Lab reactor (ETR and EBR), and only published in a series of INL technical reports

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



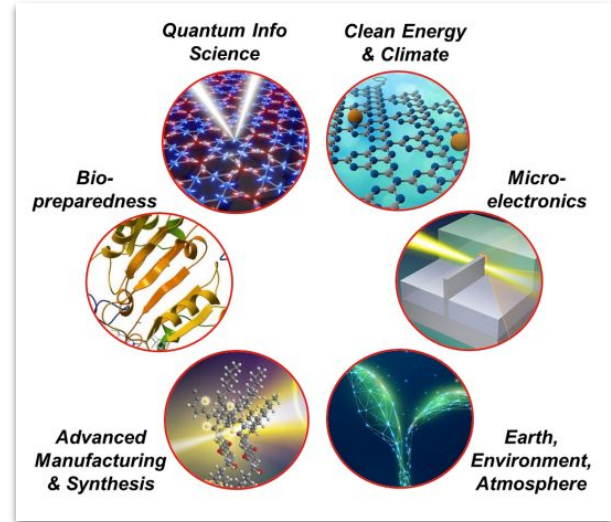
Why are they so precise?

Uncertainty Budget



- 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)

National Synchrotron Light Source II



DOE-SC User Facility

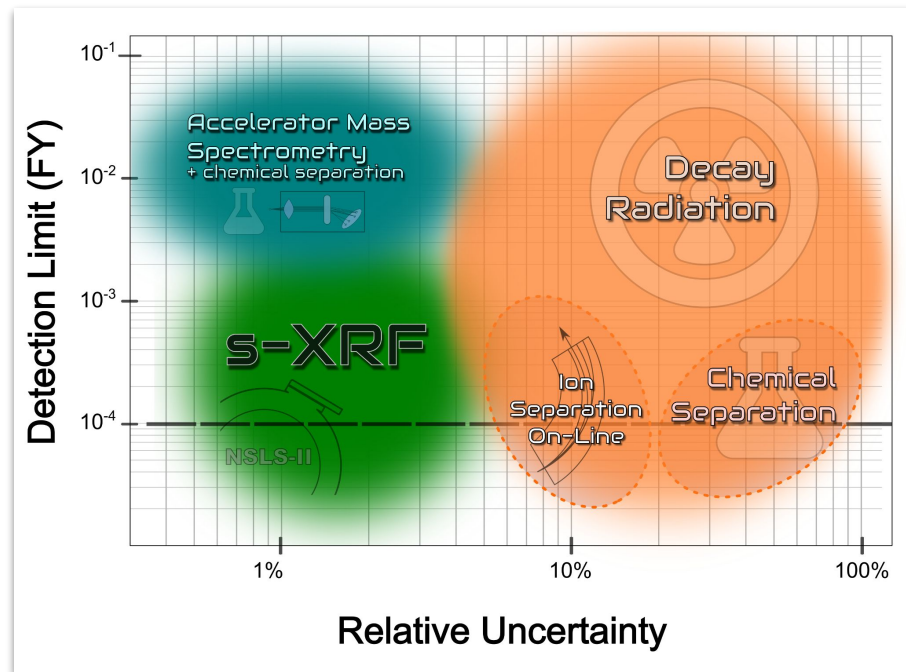
Vision: as one of the newest, most advanced synchrotron light sources in the world, NSLS-II strives to be an extraordinary hub for the use of synchrotron light to solve the world's most challenging scientific problems.

- 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

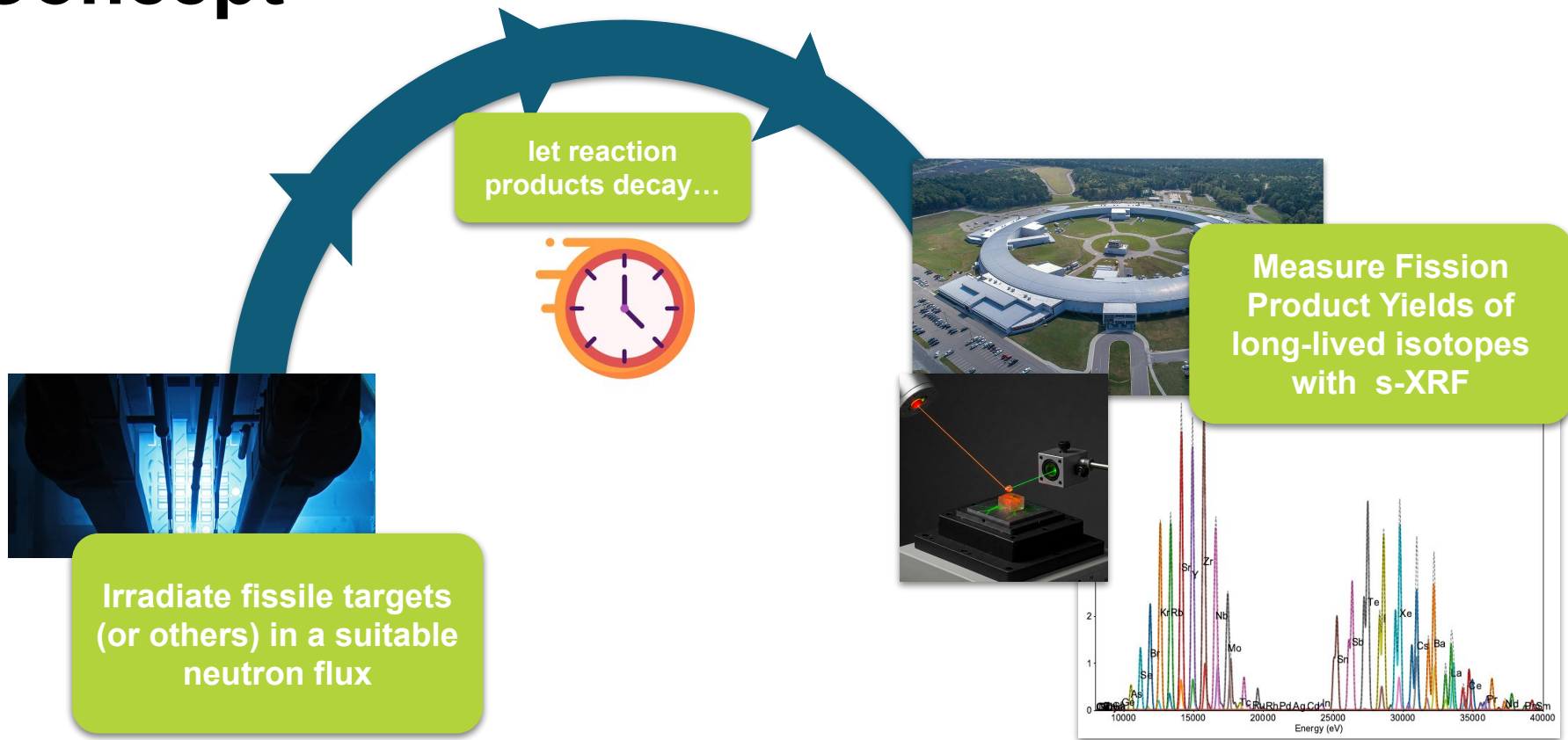
Concept

XRF can measure **ppm-level concentrations** of elements in a matrix: low-yield fission products and full normalization (no fission rate), with no limitations on the measured elements

Setup calibration: we don't rely on decay data to determine absolute yields



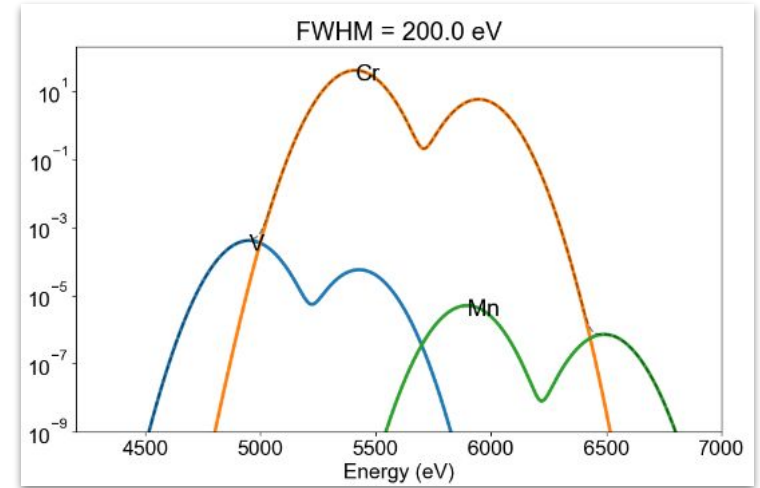
Concept



Spin-off

measurement of capture cross sections for low-abundance isotopes

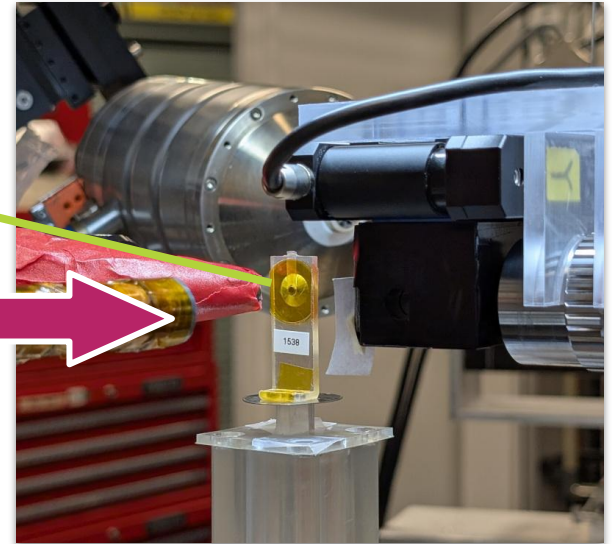
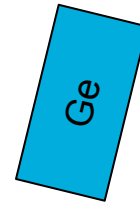
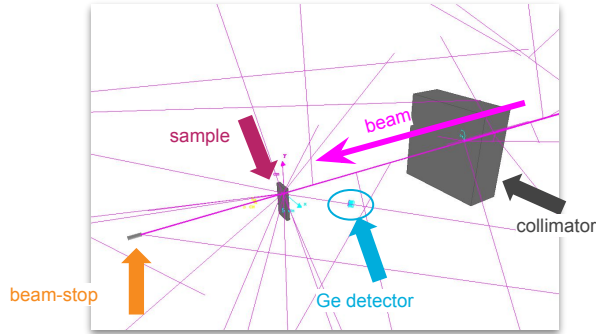
⁵² Fe 275 h 100.00%	⁵³ Fe 8.51 min $\epsilon = 100.00\%$	⁵⁴ Fe STABLE 5.845% 2.252	⁵⁵ Fe 2.744 y $\epsilon = 100.00\%$	⁵⁶ Fe STABLE 91.754% 2.589	⁵⁷ Fe STABLE 2.119% 2.426	⁵⁸ Fe STABLE 0.282% 1.149	$\beta^- =$
⁵¹ Mn .2 min 100.00%	⁵² Mn 5.591 d $\epsilon = 100.00\%$	⁵³ Mn 3.74E+6 y $\epsilon = 100.00\%$	⁵⁴ Mn 312.20 d $\epsilon = 100.00\%$ $\beta^- = 9.3E-5\%$	⁵⁵ Mn STABLE 100% 1.341E+1	⁵⁶ Mn 2.5789 h $\beta^- = 100.00\%$	⁵⁷ Mn 85.4 s $\beta^- = 100.00\%$	$\beta^- =$
⁵⁰ Cr 3E+18 y 3.45% 2e 592E+1	⁵¹ Cr 27.7025 d $\epsilon = 100.00\%$	⁵² Cr STABLE 83.789% 7.455E-1	⁵³ Cr STABLE 9.501% 1.806E+1	⁵⁴ Cr STABLE 2.365% 3.600E-1	⁵⁵ Cr 3.497 min $\beta^- = 100.00\%$	⁵⁶ Cr 5.94 min $\beta^- = 100.00\%$	$\beta^- =$
⁴⁹ V 30 d 100.00%	⁵⁰ V > 2.1E+17 y 0.250% $\epsilon \approx 92.90\%$	⁵¹ V STABLE 99.750%	⁵² V 3.743 min $\beta^- = 100.00\%$	⁵³ V 1.543 min $\beta^- = 100.00\%$	⁵⁴ V 49.8 min $\beta^- = 100.00\%$		



technique is sensitive to any decay/reaction that changes the atomic number of the products

Feasibility study: Experimental setup

NSLS-II 28ID-2 beamline



$$E_{\text{x-ray}} = 68 \text{ keV}$$

Feasibility Study - samples

- Samples from ATR at Idaho National Lab (NSUF): test the method on irradiated samples
- Thin samples manufactured at Argonne National Lab: test sensitivity to fission products signal on '*thick*' actinide backing
- Standard samples: estimate linearity & calibrate system for quantitative analysis



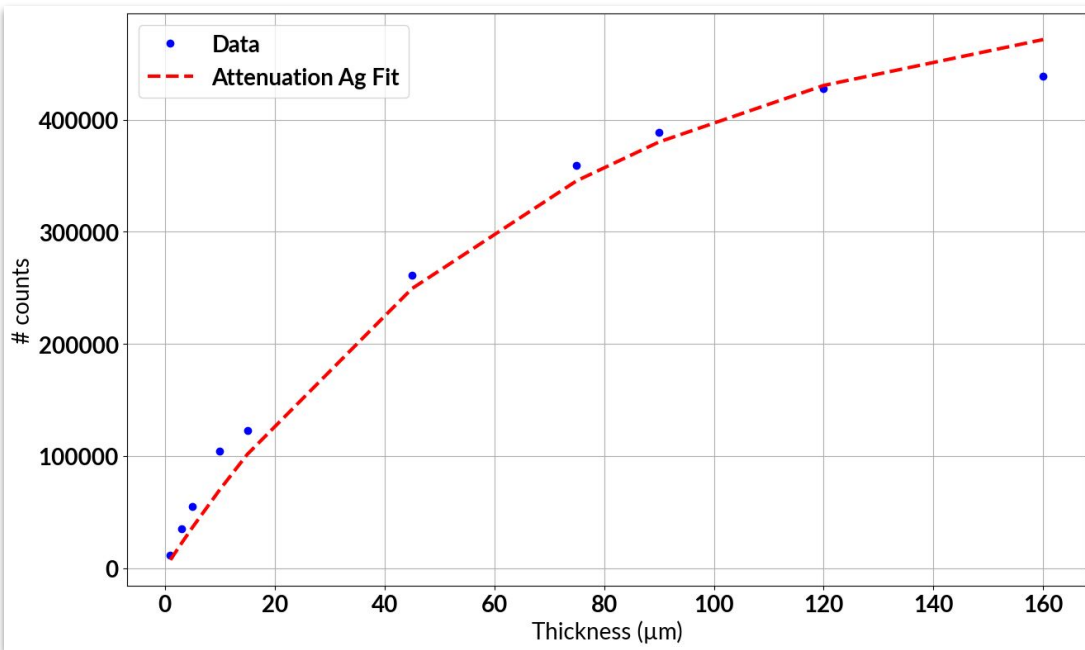
Identifier		Sample Description	Total Fluence n/cm ²
KGT	Sample ID		
1538	09-157-033	Hafnium Aluminide	1.29 × 10 ²²
1539	09-157-034	Hafnium Aluminide	1.3 × 10 ²²
4785-1	034-08-331	Silver (99.95%)	1.47 × 10 ²²
4785-2	034-08-331	Silver (99.95%)	1.47 × 10 ²²
4785-3	034-08-331	Silver (99.95%)	1.47 × 10 ²²
4785-4	034-08-331	Silver (99.95%)	1.47 × 10 ²²
4942	149-08-331	Silver (99.95%)	2.9 × 10 ²²
5101	148-08-331	Tungsten (99.95%)	2.94 × 10 ²²
5108	109-08-331	Tungsten (99.95%)	1.58 × 10 ²²
5128	052-08-331	Tungsten (99.95%)	1.05 × 10 ²²

	Material	Nominal Thickness	
		μm	μg/cm ²
K	Kapton (C ₂₂ H ₁₀ N ₂ O ₄)	50	≈7100
C	Carbon (C)	≈ 0.09	20
U	Uranium Tetrafluoride (UF ₄)	≈ 0.65	310
Sn	Tin (Sn)	≈ 0.51	370
Te	Tellurium (Te)	≈ 0.54	340

Id	Layers
I	K + K
II	K + C + U + K
III	K + C + U + Sn + K
IV	K + C + U + Te + K
V	K + C + U + Sn + Te + K
VI	K + C + U + Sn + C + U + Sn + Te + K
VII	K + C + U + Te + C + U + Sn + Te + K
VIII	K + C + U + Sn + Te + C + U + Sn + Te + K



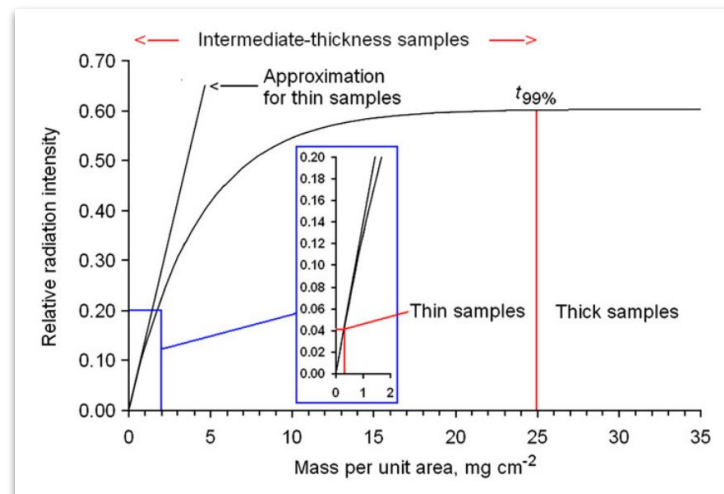
Results - studies of attenuation



Fit of the attenuation of X-rays in Ag

In bulk materials (“thick sample”), the X-ray intensity is “(~!!) proportional” to the material concentration

Beckhoff, B., Kanngießner, B., Langhoff, N., Wedell, R., & Wolff, H. (Eds.). (2007). *Handbook of practical X-ray fluorescence analysis*.

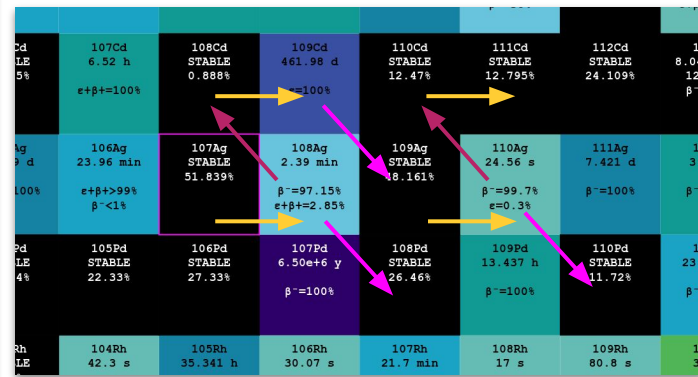
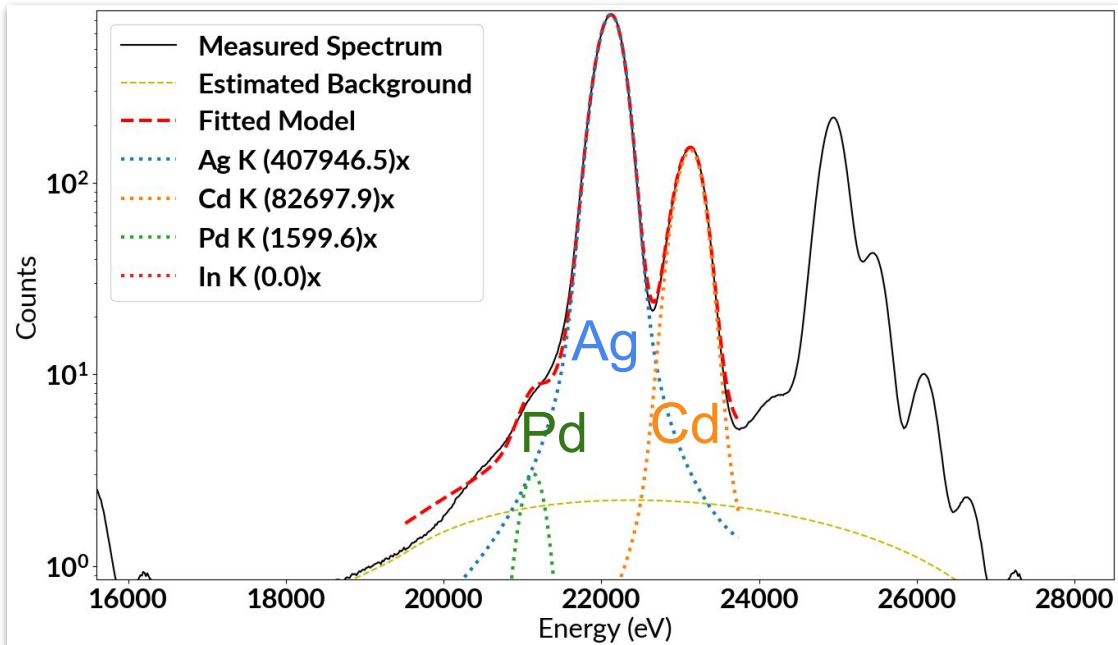


Sitko, Rafal. "Quantitative X-ray fluorescence analysis of samples of less than 'infinite thickness': difficulties and possibilities." *Spect Acta B* 64, 11-12 (2009): 1161-1172.

Results - NSUF samples

Ag - high flux

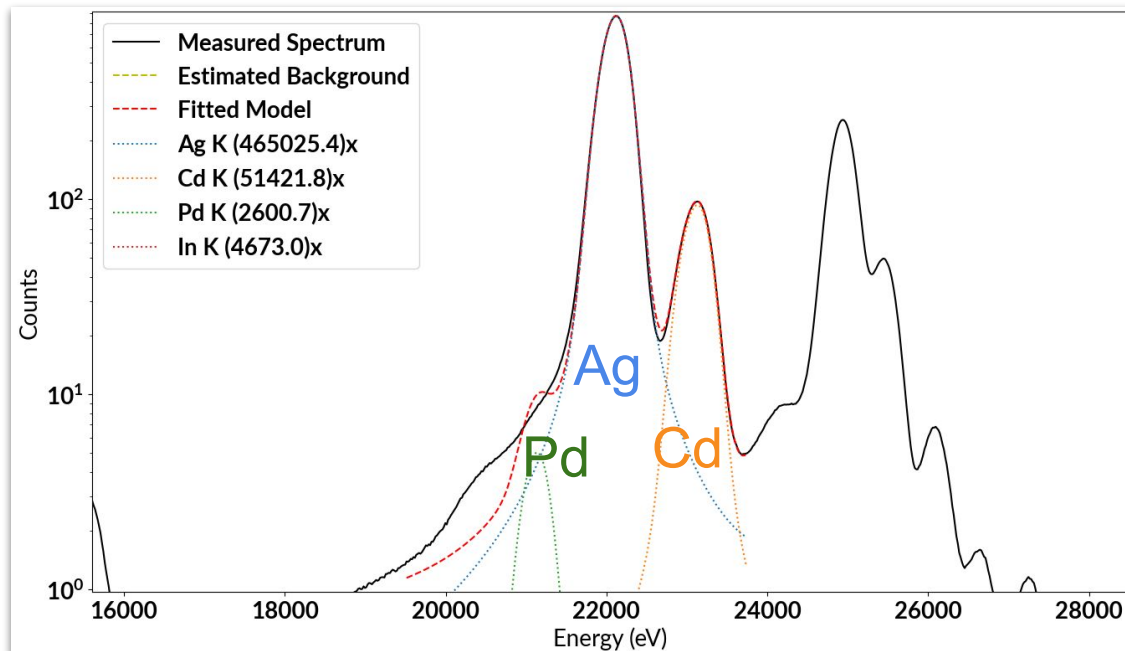
4942 149-08-331 Silver (99.95%) 2.9×10^{22}



measured fraction (Cd) = $(20.27 \pm 0.10)\%$

Results - NSUF samples

Ag - low flux
(2-4)



4785-2	034-08-331	Silver (99.95%)	1.47×10^{22}
4785-3	034-08-331	Silver (99.95%)	1.47×10^{22}
4785-4	034-08-331	Silver (99.95%)	1.47×10^{22}

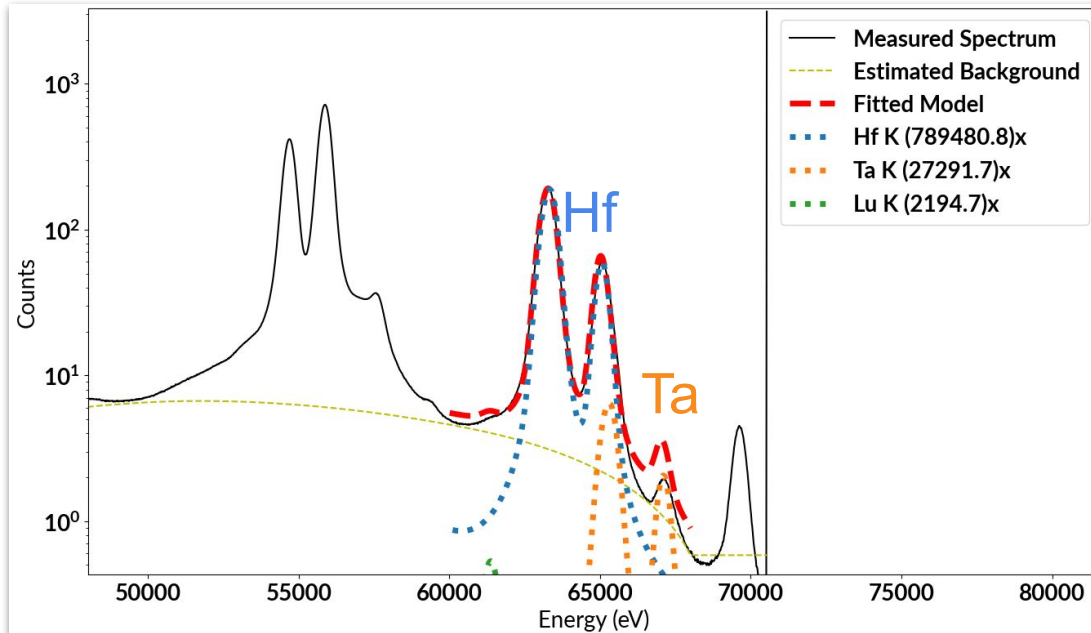
measured fraction = $(10.92 \pm 0.10) \%$
measured fraction = $(11.01 \pm 0.09) \%$
measured fraction = $(12.89 \pm 0.10) \%$

Results

175Ta 10.5 h e+β=100%	176Ta 8.08 h e+β=100%	177Ta 56.56 h e+β=100%	178Ta 2.41 h e+β=100%	179Ta 1.815 y e=100%	180Ta 8.154 h 0.01201% e=85% β=15%	181Ta STABLE 99.98799%	182Ta 114.804 d β=100%
174Hf 7.00e+16 y 0.16% α=100%	175Hf 70.67 d e=100%	176Hf STABLE 5.26%	177Hf STABLE 18.6%	178Hf STABLE 27.28%	179Hf STABLE 13.62%	180Hf STABLE 35.08%	181Hf 42.39 d β=100%
173Lu 1.37 y e=100%	174Lu 3.31 y e+β=100%	175Lu STABLE 97.401%	176Lu 3.71e+10 y 2.599% β=100%	177Lu 6.65 d β=100%	178Lu 28.4 min β=100%	179Lu 4.59 h β=100%	180Lu 5.6 min β=100%
172Yb STABLE 21.686%	173Yb STABLE 16.103%	174Yb STABLE 32.025%	175Yb 4.16 d	176Yb STABLE 12.985%	177Yb 1.911 h	178Yb 74 min	179Yb 8 min

Hf

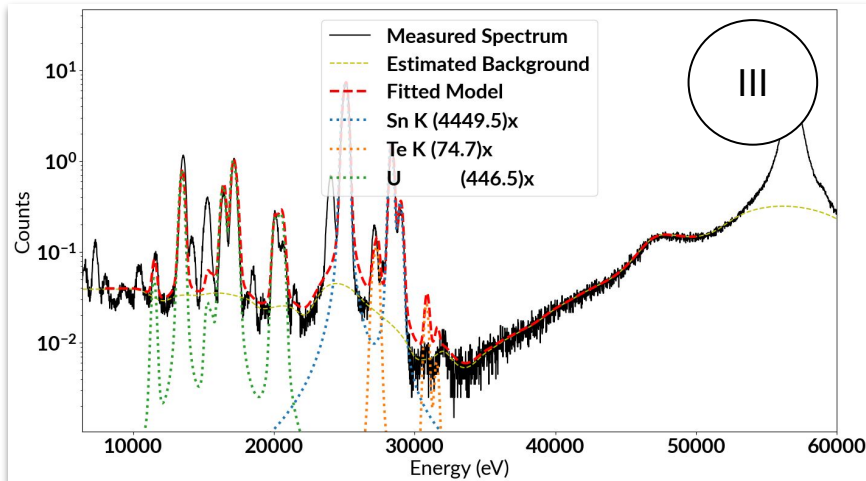
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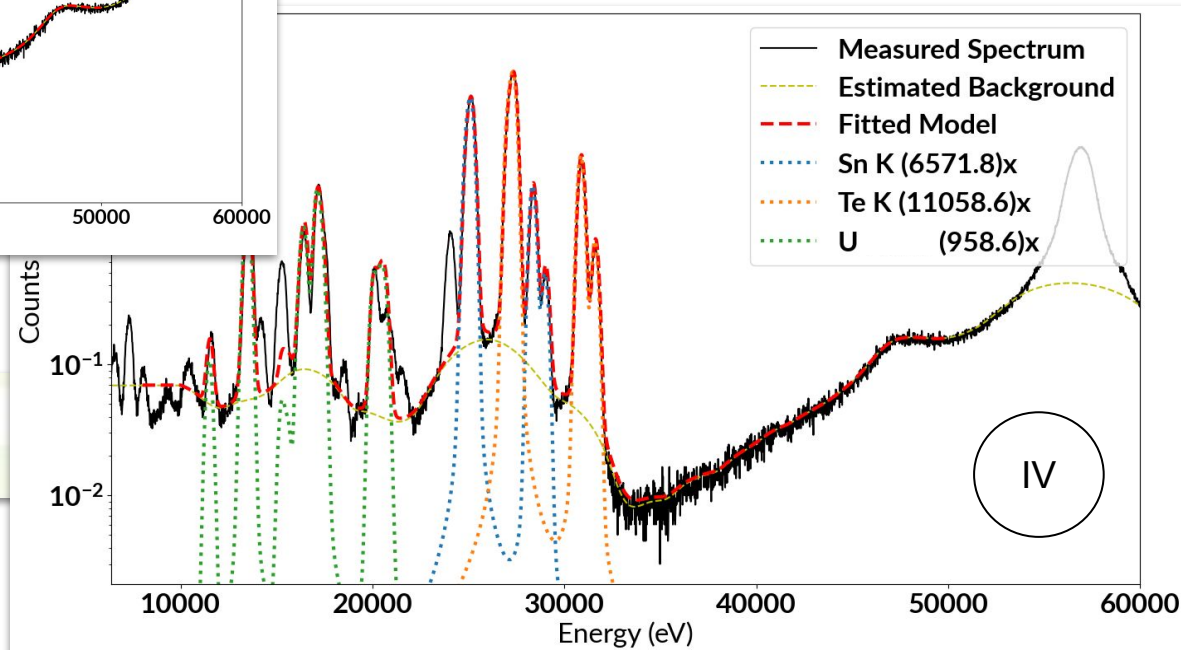
measured (Ta) = $(3.79 \pm 0.12) \%$
 measured (Ta) = $(4.27 \pm 0.14) \%$

measured (Lu) = $(1.09 \pm 0.13) \%$
 measured (Lu) = $(0.74 \pm 0.15) \%$

Results - ANL samples



multiple thickness
(thin-targets)



III K + C + U + Sn + K
 IV K + C + U + Te + K

Results - ANL samples

Id	Sn	Te	U	Layers
I	335 ± 80	0 ± 63	4 ± 7	K + K
II	8 ± 8	59 ± 8	392 ± 7	K + C + U + K
III	4449 ± 22	75 ± 11	447 ± 6	K + C + U + Sn + K
IV	6572 ± 27	$11\,059 \pm 36$	959 ± 10	K + C + U + Te + K
V	4672 ± 19	4983 ± 20	498 ± 7	K + C + U + Sn + Te + K
VI	$10\,549 \pm 39$	5600 ± 26	962 ± 11	K + C + U + Sn + C + U + Sn + Te + K
VII	9 ± 10	5911 ± 23	477 ± 6	K + C + U + Te + C + U + Sn + Te + K
VIII	$10\,715 \pm 39$	$11\,605 \pm 40$	1028 ± 13	K + C + U + Sn + Te + C + U + Sn + Te + K

Summary: Lessons Learned

INL samples:

- the ratio between “high” and “low” irradiation follows expectations
- even if we get the order of magnitude, the amount seems lower than what was estimated (non-thermal reactions and full neutron-spectrum simulation needed)
- Relative intensities of different clusters doesn't follow a standard 'efficiency' curve (\sim HPGe) \rightarrow MATRIX EFFECTS (i.e., sample-dependent attenuation/enhancement of X-ray lines)

standard Ag samples:

- attenuation follows expected non-linear curve.
- With INL samples we are in the 'bulk' / 'thick-target' regime

ANL samples:

- we can distinguish signal from FPs over Actinides 'background'

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ANL samples:

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BACKGROUND

**ATTENUATION
(MATRIX EFFECT)**

Outlook

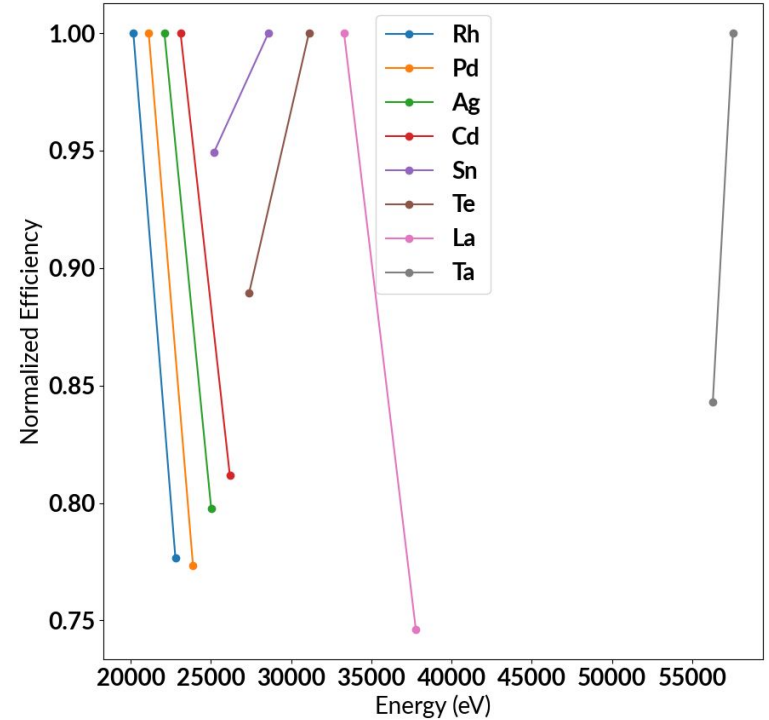
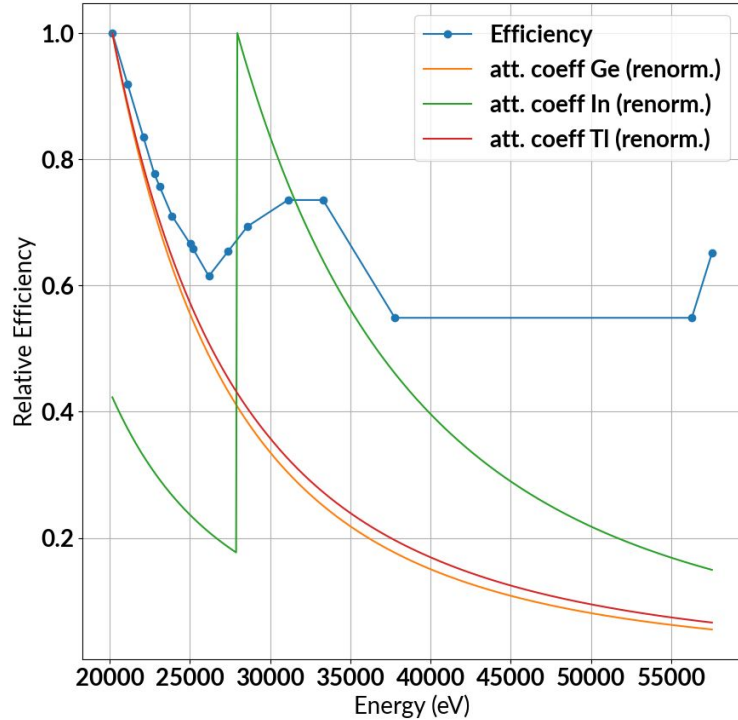
- Geant4 simulation of the experimental setup to study the matrix effects on samples
- MCNP simulation to estimate expected concentrations in irradiated samples
- Efficiency calibration with standard sources
- Test of measurement of fission products

Acknowledgements

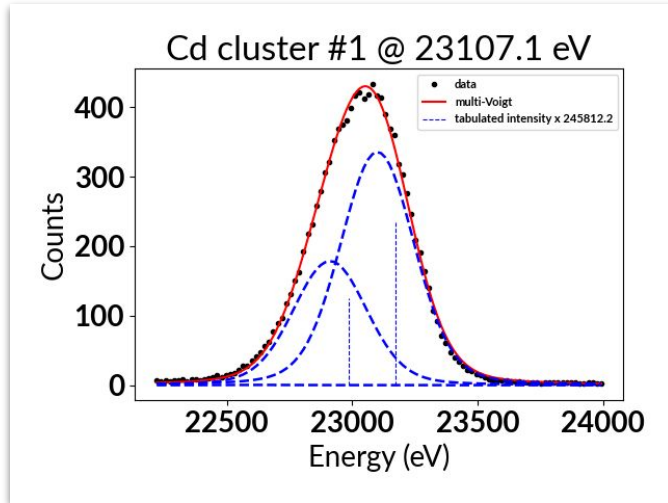
*This work is supported by BNL under the **LDRD project No. 24026**. The work at Brookhaven National Laboratory was sponsored by the **Office of Nuclear Physics, Office of Science of the U.S. Department of Energy** under Contract No. DE-SC0012704 with Brookhaven Science Associates, LLC. This work was also supported by the U.S. Department of Energy, **Office of Nuclear Energy** under DOE Idaho Operations Office Contract DE-AC07-05ID14517 as part of **Nuclear Science User Facilities award #4941** and by the **National Nuclear Security Administration, Defense Nuclear Nonproliferation Research and Development**.*

*This project was supported in part by the U.S. Department of Energy, Office of Science, **Office of Workforce Development for Teachers and Scientists** under the **Community College Internships Program (CCI)** and the **Science Undergraduate Laboratory Internships Program (SULI)**.*

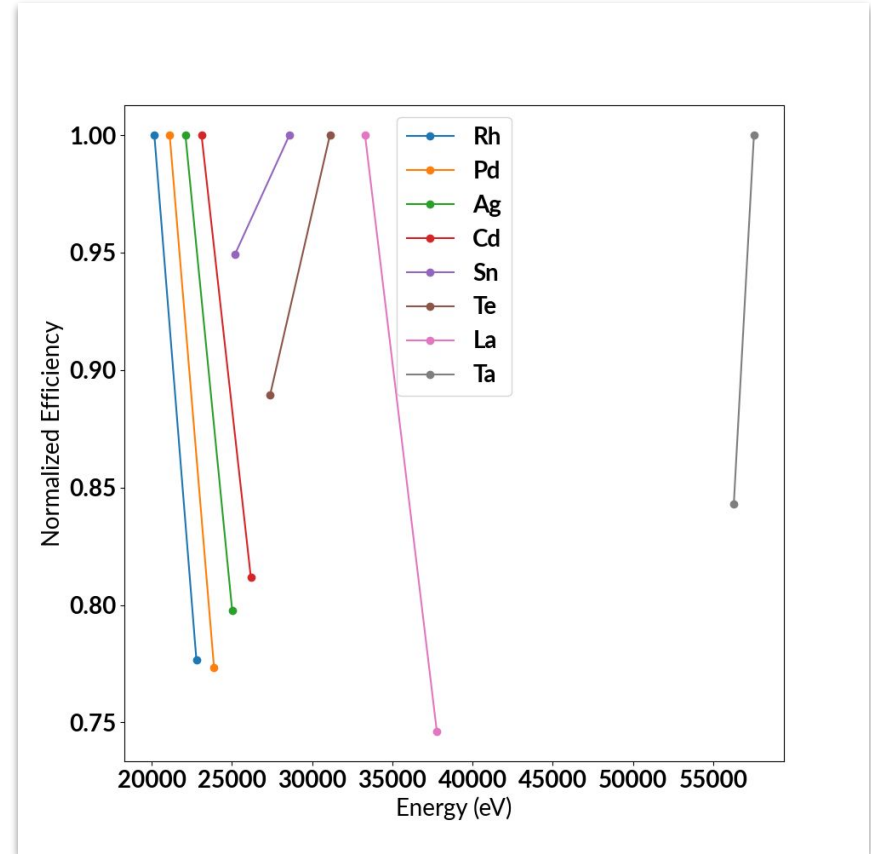
Results (2) - efficiency calibration



Results (2) - efficiency calibration



- estimate energy-dependent photopeak efficiency:
- > cluster overlapping peaks
 - > estimate change in intensity wrt expected BR



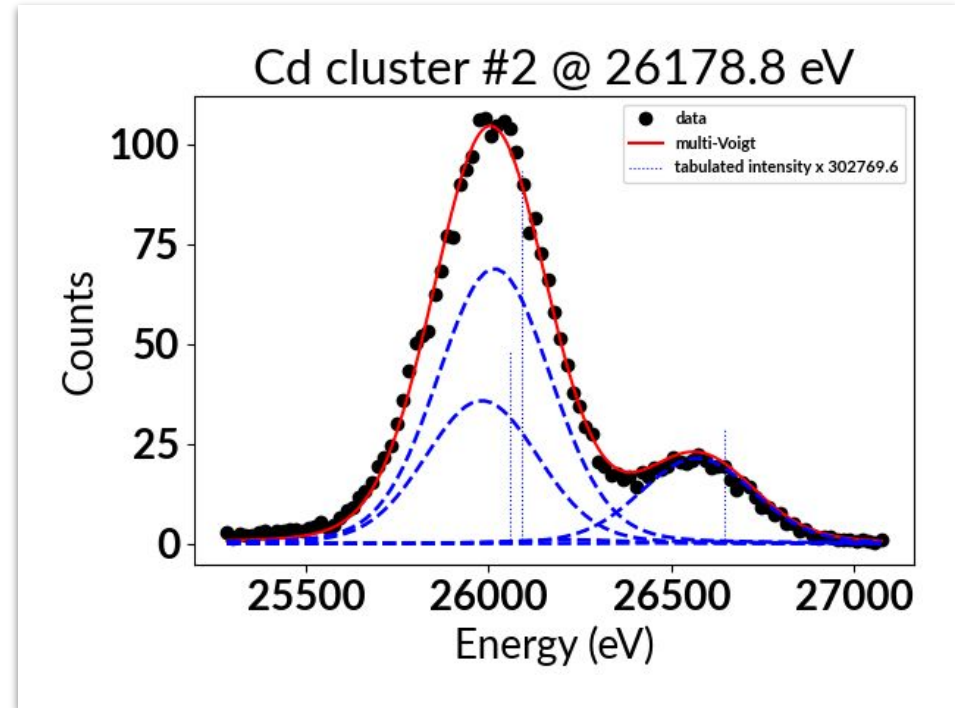
Analysis (1): Fitting procedure

X-ray intensities and energies from literature¹.

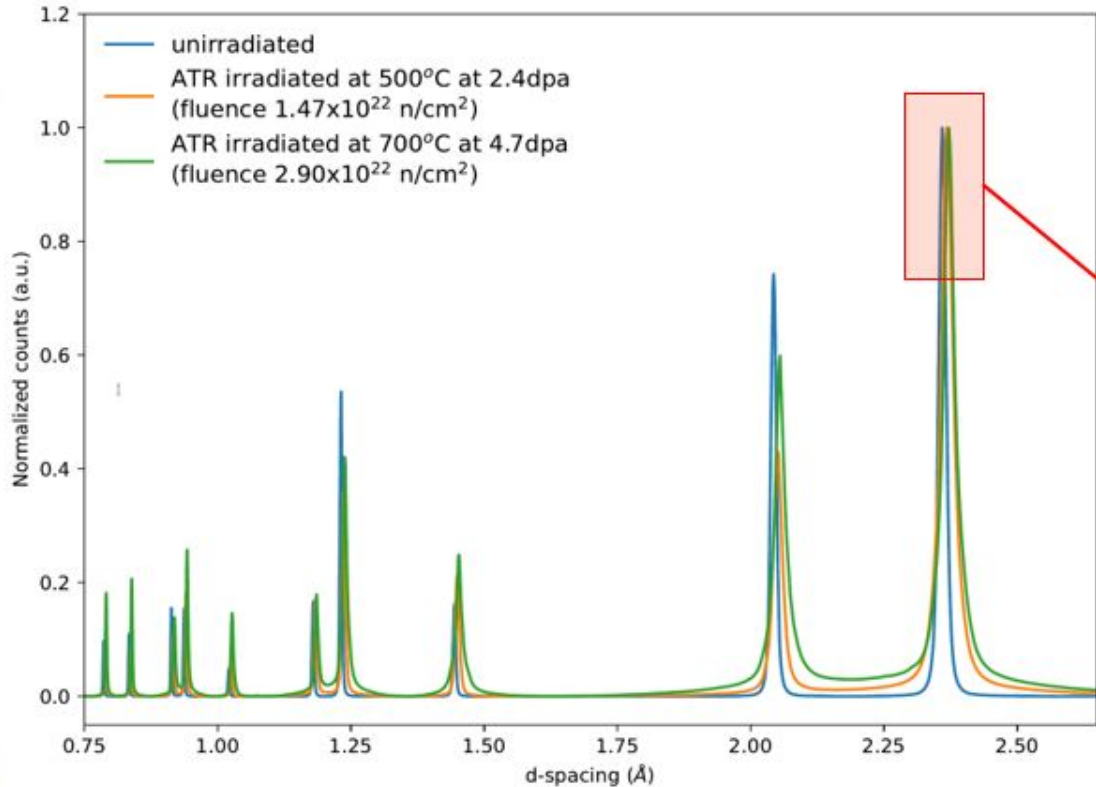
Rigid fit of a multi-Voigt distribution scaled to match data. Background estimated and included in the fit.

5 free parameters: σ , γ , S , background scaling and small energy offset.

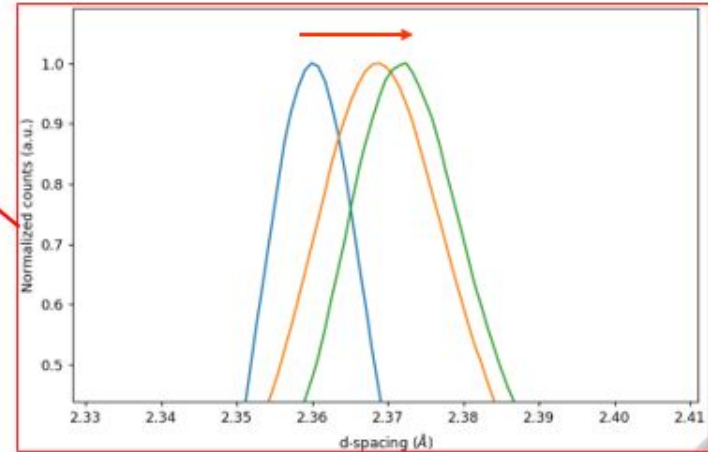
¹Elam, William Timothy, Bruce D. Ravel, and J. R. Sieber. "A new atomic database for X-ray spectroscopic calculations." *Radiation Physics and Chemistry* 63.2 (2002): 121-128.



Results (5) - X-ray Diffraction



Dramatic changes with ATR irradiation!!!



X-Ray Diffraction also shows effect of irradiation

Analysis -- Estimate expected amounts

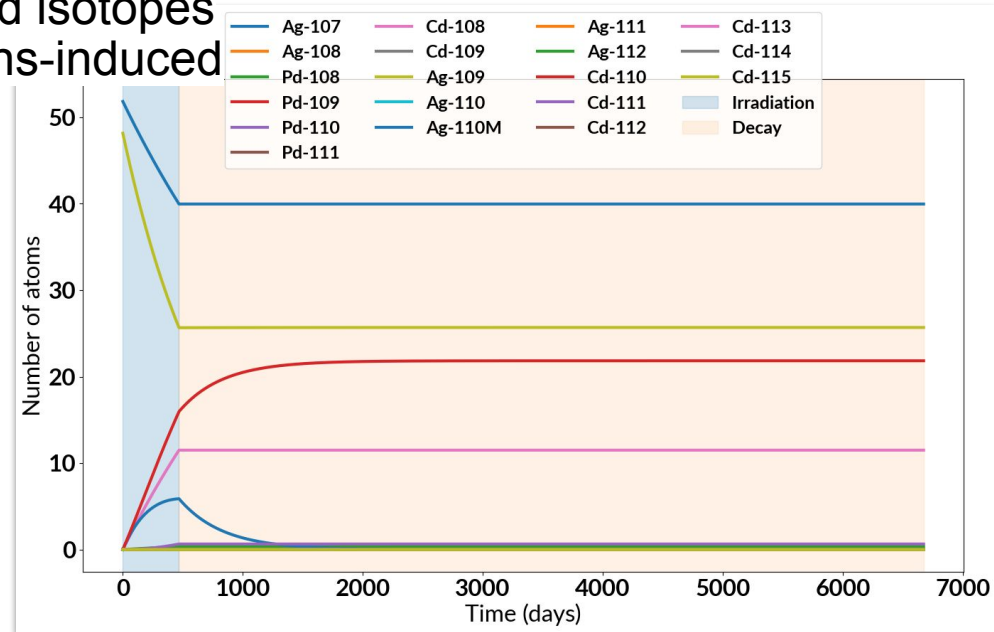
Considered

- irradiation history and total fluence from NSUF / INL
- create a simplified network of capture/decay
- assume $\frac{1}{2}$ thermal neutrons²

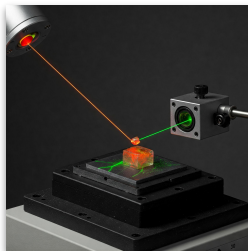
Ignored

- irradiation cycles
- capture from shorter-lived isotopes
- fast-neutrons-induced reactions

² Idaho National Laboratory, Advanced Test Reactor User Guide, Rev. 24. Tec. Rep. INL/EXT-20-50097, INL, 2023.



Concept

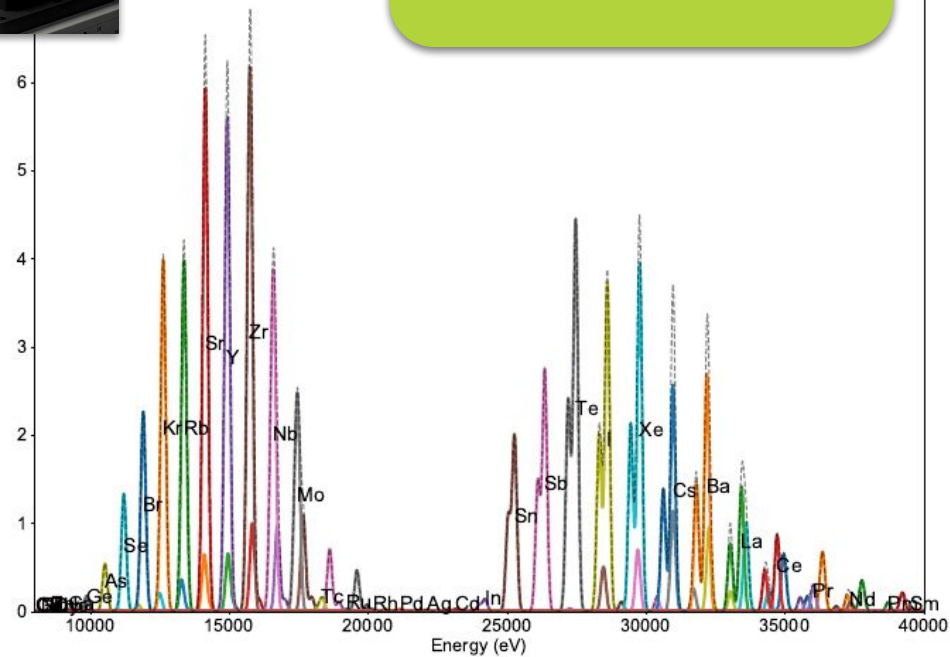


Measurement of Fission Product Yields using X-Ray Fluorescence

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

NSLS-II combined with the 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 - it provides 'anchor point' used in a FPY evaluation.



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

Acknowledgements

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