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Characterization of ^{233}U for Thorium Fuel Cycle Safeguards

Ms. Madeline Lockhart

North Carolina State University, USA

18 December 2023

LA-UR-23-33347

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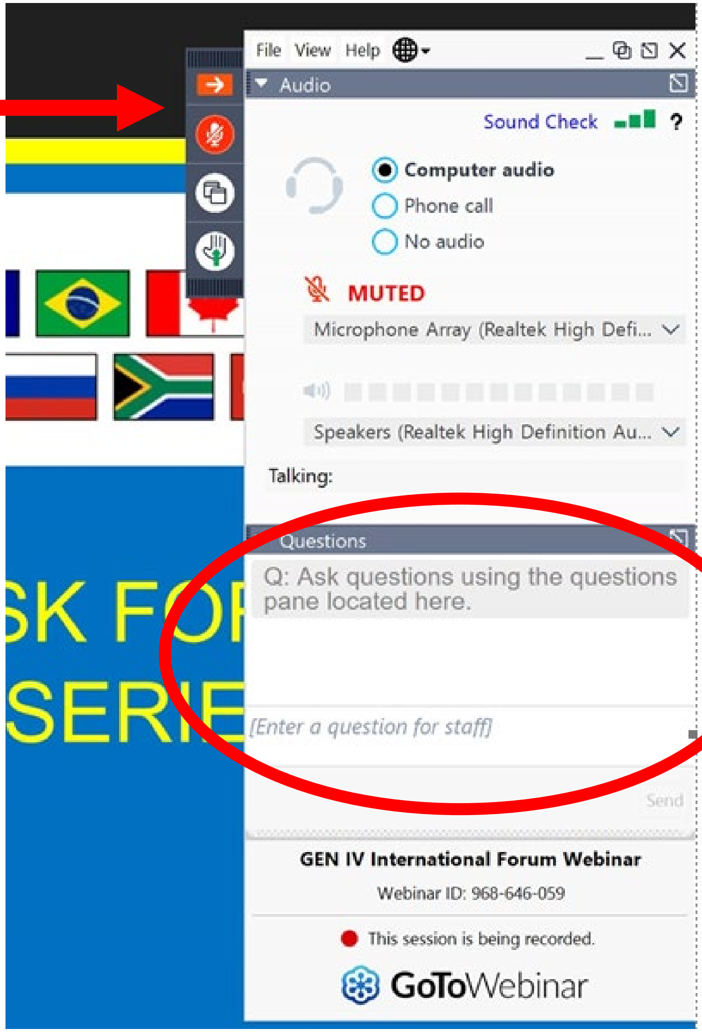

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Meet the Presenter: Madeline Lockhart

- 4th year PhD Student at North Carolina State University
- Nuclear Nonproliferation and International Safeguards (NNIS) Fellow
- Undergraduate and Graduate Research Assistant at Los Alamos National Laboratory (2015 – 2023)
- Visiting scientist at the European Commission Joint Research Centre in Ispra, Italy (October 2023 – April 2024)
- Bachelor’s degree in physics from Texas Tech University

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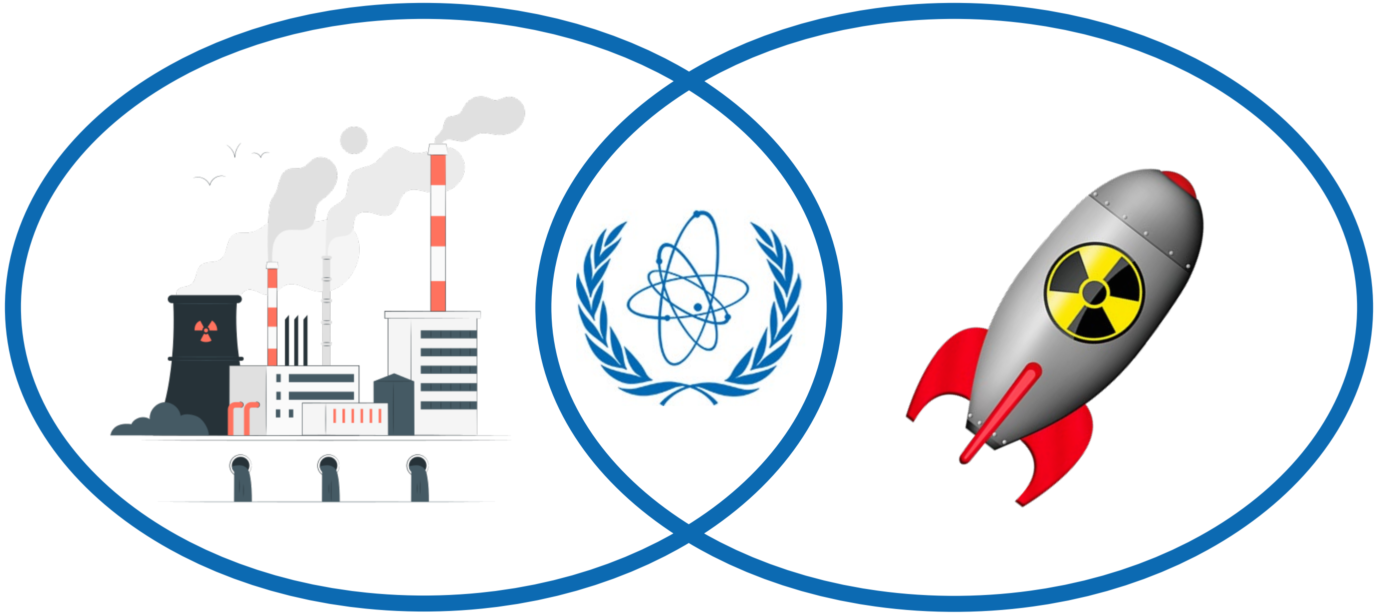
What are Nuclear Safeguards?

DEFINITION

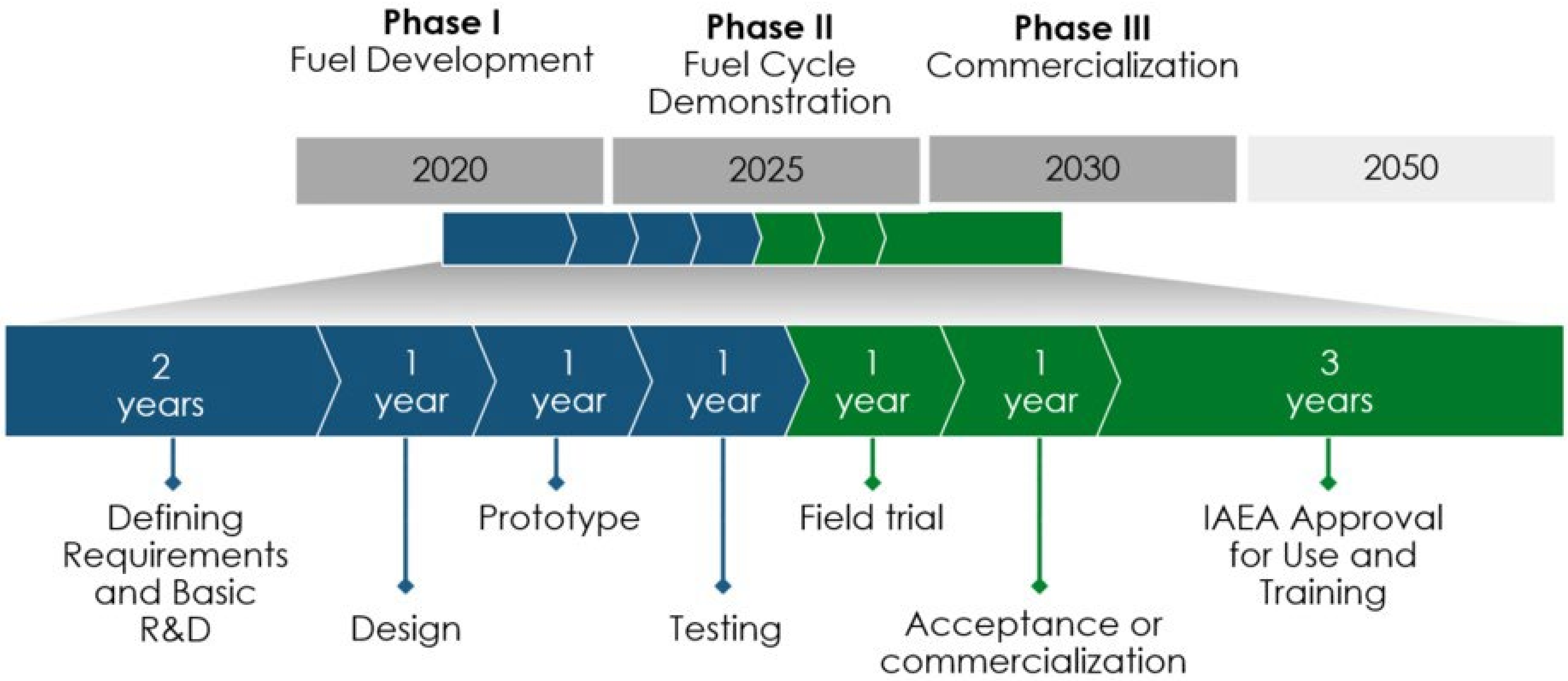
a set of technical measures applied by the IAEA on nuclear material and activities

OBJECTIVE

to deter the spread of nuclear weapons by the early detection of the misuse of nuclear material or technology



Safeguards Development Timeline



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Worrall, Louise G., et al. *Safeguards Technology for Thorium Fuel Cycles: Research and Development Needs Assessment and Recommendations*. United States: N. p., 2021. Web. doi:10.2172/1818724.

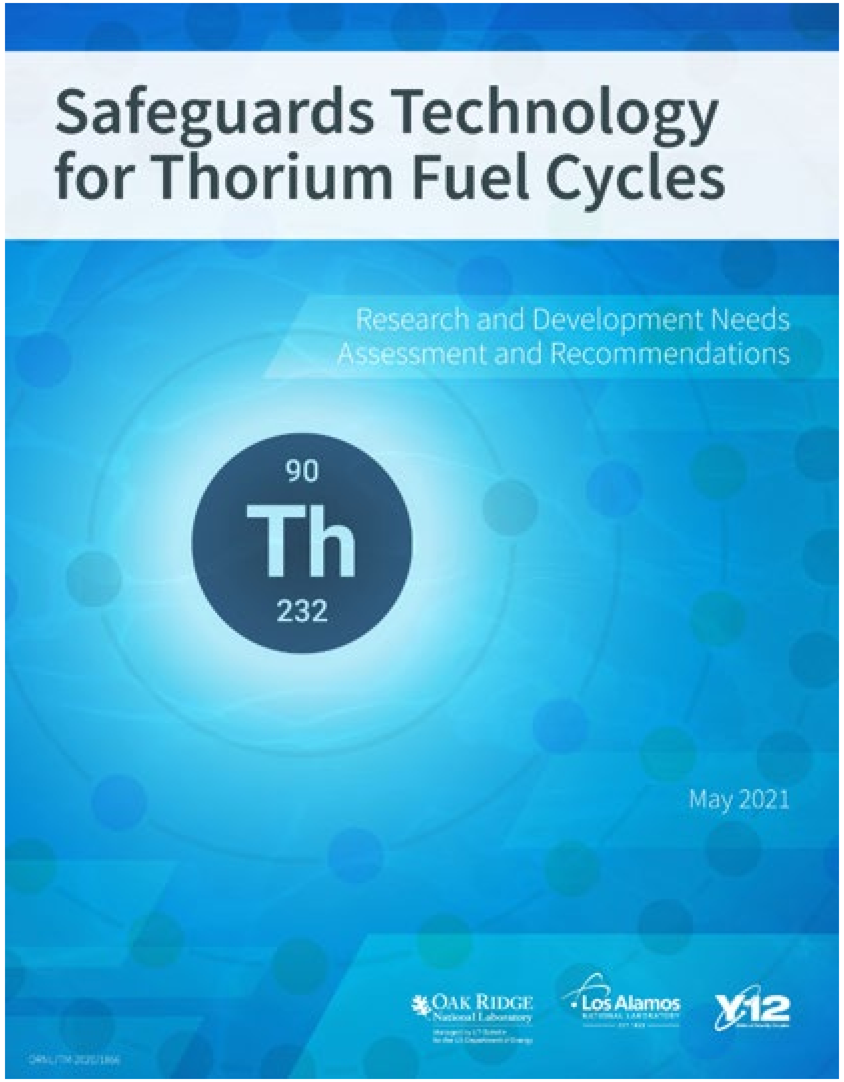
Safeguards for the Thorium Fuel Cycle

- Proliferation detection R&D is needed so that the detection toolkit (safeguards, remote detection, etc.) is ready to monitor thorium fuel cycle activities
- Advanced reactor designer needs to think about safeguards during the design process, not only when they are looking to export
- If a material is “self-shielding” or “proliferation resistant”, it is not exempted from safeguards
- Characterization of ^{233}U is important for safeguards and nuclear material accounting and control (NMAC)

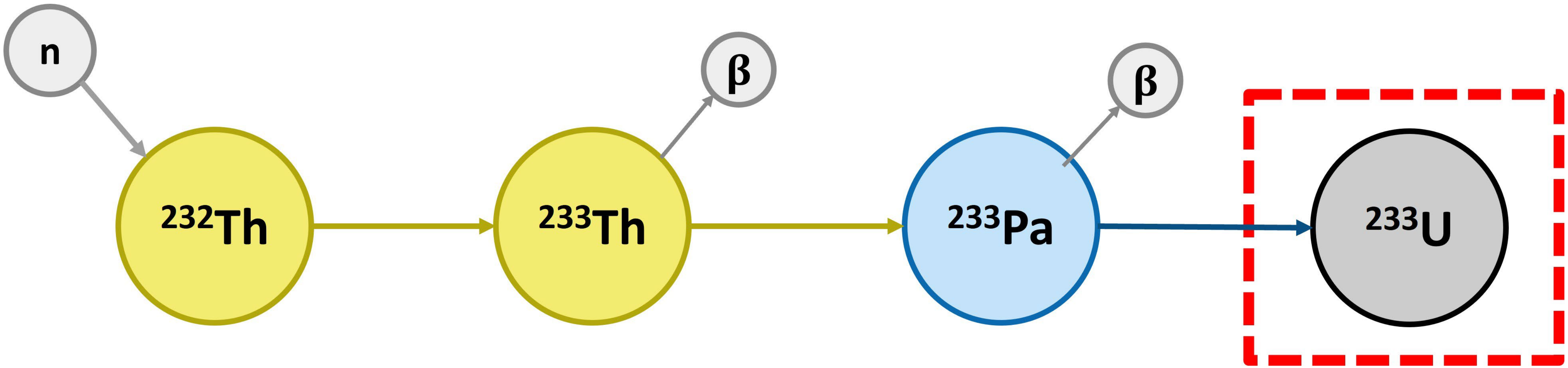
R&D Needs Assessment

Understand the R&D that is necessary to transition the current safeguards technology toolkit to meet the verification needs of thorium fuel cycles

- Identify leading candidate thorium fuel cycles and their characteristics that impact safeguards technology
- Provides the scientific basis for **strengthening existing instrumentation capabilities** or **developing new instrumentation** that may be needed to fill any potential capability gaps within the international nuclear safeguards community to properly verify declarations of any ^{232}Th and ^{233}U bearing materials



How is ^{233}U produced?



1 Thorium-232 captures a neutron, becoming Thorium-233

2 Thorium-233 undergoes beta decay (**$t_{1/2}$=22 min**) into Protactinium-233

3 Protactinium-233 undergoes beta decay (**$t_{1/2}$=27 d**) into Uranium-233

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Why do we care about ^{233}U?

SIGNIFICANT QUANTITY (SQ)

"the approximate amount of nuclear material for which the possibility of manufacturing a nuclear explosive device cannot be excluded"
-IAEA Safeguards Glossary 2022

Pu (<80% ^{238}Pu)	8 kg
^{233}U	8 kg ^{233}U
HEU (>20% ^{235}U)	25 kg ^{235}U

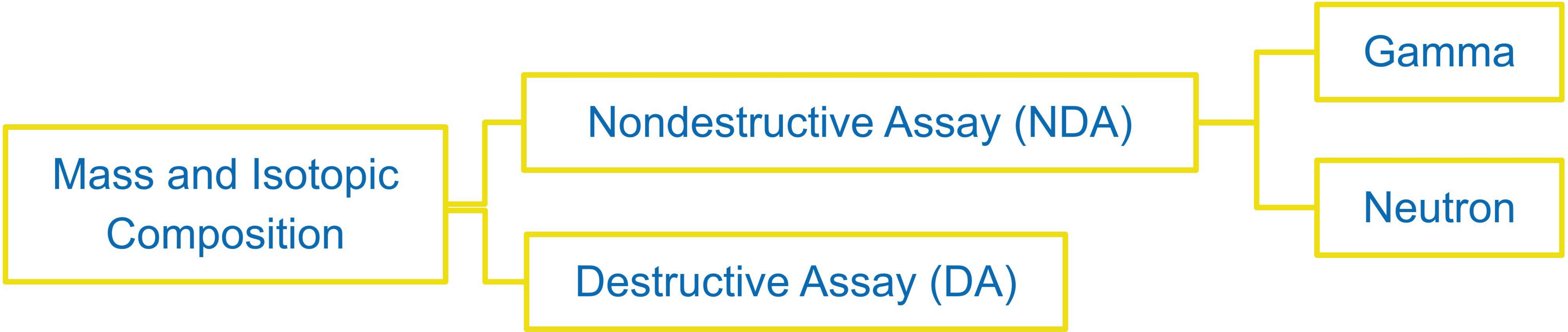
DIRECT USE MATERIALS

"nuclear material that can be used for the manufacture of nuclear explosive devices without transmutation or further enrichment"
-IAEA Safeguards Glossary 2022

How is SNM characterized?

SPECIAL NUCLEAR MATERIAL (SNM)

plutonium, **uranium-233**, or uranium enriched in the isotopes uranium-233 or uranium-235, but does not include source material

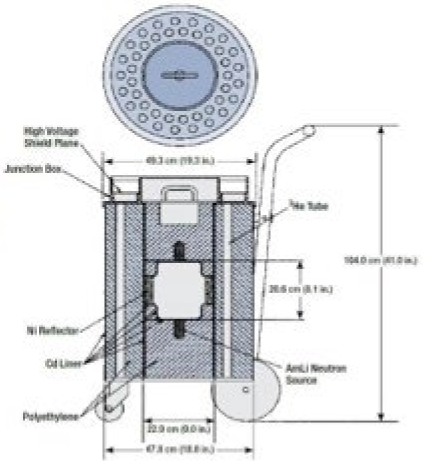


Nondestructive Assay (NDA) Methods

Gamma X

^{232}U contamination dominates the gamma spectra

Sources are often in lead shielding



Neutron ✓

In oxide form, ^{233}U has measurable neutrons from (α,n) reactions.

Active interrogation, used to measure ^{235}U, also works with ^{233}U

Neutron NDA

- Coincidence counting methods
  - Passive neutron coincidence counting
  - Active neutron coincidence counting using AmLi neutron source
- Time and Energy based signatures – **Oskar Searfus, University of Michigan**
  - Delayed Neutron (DN), differential die away (DDA), passive neutron spectroscopy

Nuclide	Half-life	SF multiplicity	SF yield [n·s^{-1}·g^{-1}]	Alpha yield [α·s^{-1}·g^{-1}]	(α,n) yield in oxide [n·s^{-1}·g^{-1}]
^{232}Th	1.41×10^{10} years	2.14	$>6\times10^{-8}$	4.1×10^{3}	2.2×10^{-5}
^{232}U	71.7 years	1.71	1.3×10^{0}	8.0×10^{11}	1.5×10^{4}
^{233}U	1.59×10^{5} years	1.76	8.6×10^{-4}	3.5×10^{8}	4.8
^{234}U	2.45×10^{5} years	1.81	$5.02\text{x}10^{-3}$	$2.3\text{x}10^{8}$	3.0
^{235}U	7.04×10^{8} years	1.86	2.99×10^{-4}	7.9×10^{4}	7.1×10^{-4}
^{238}U	4.468×10^{9} years	2.01	1.36×10^{-2}	1.2×10^{4}	8.3×10^{-5}

Instrumentation

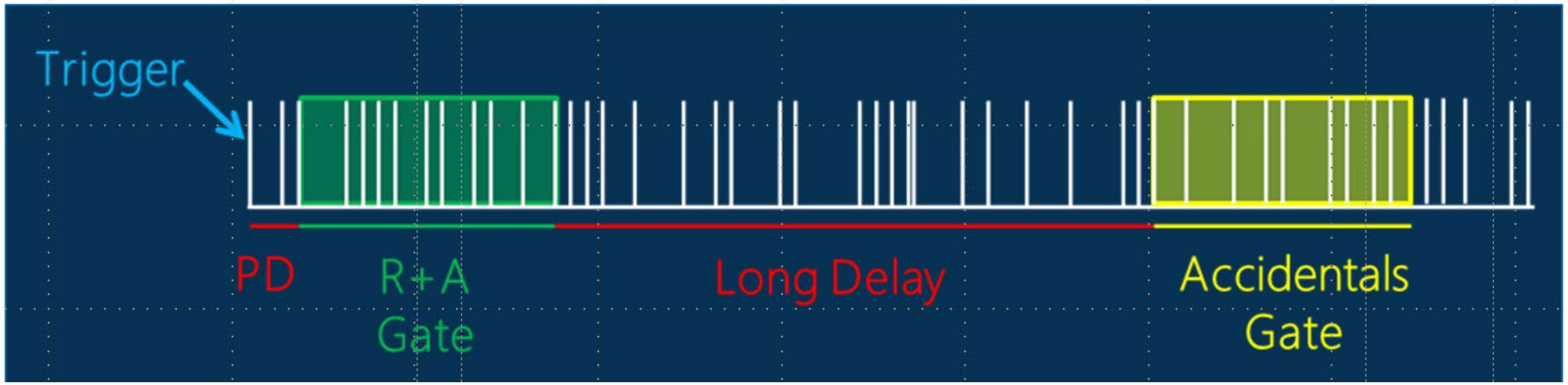
Active Well Coincidence Counter (AWCC)

- LV-AWCC
- Traditional ^{3}He Well Counter
- 48 ^{3}He tubes in 2 rings
- Passive and Active
- Removable Cd liner
- Thermal and Fast mode

Predelay	3 µs
Gate length	64 µs
High voltage	1700 V
Die away time	50 µs
Efficiency	32%

JSR-15

- Traditional Shift Register
- Used by the IAEA for verification measurements
- Predetermined analysis parameters
- International Neutron Coincidence Counting (INCC)



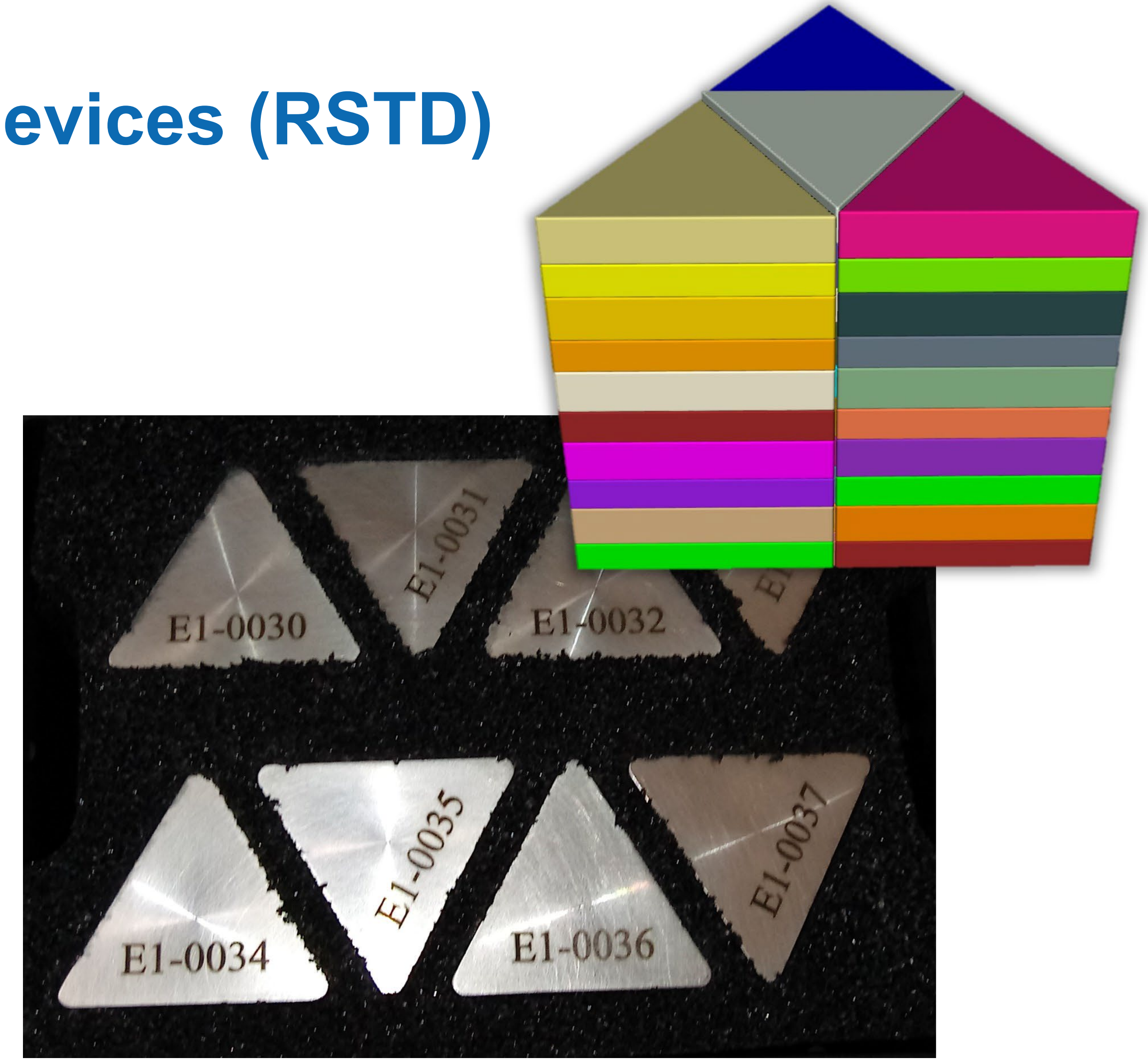
Advanced List Mode Module (ALMM)

- List mode data acquisition
- Record the pulse train
  - Time and channel for each detection event
- Allows for additional analysis methods & techniques
- Two channels: inner and outer rings of the detector

^{233}U Sources

Radiation Signature Training Devices (RSTD)

- Made for DHS
- Individual source ‘tiles’, ~2 g each
- Total of 40 individual ^{233}U oxide sources
- HEU sources also available for simulations of ^{233}U/^{235}U ratios



Traditional Mass Verification for ^{235}U

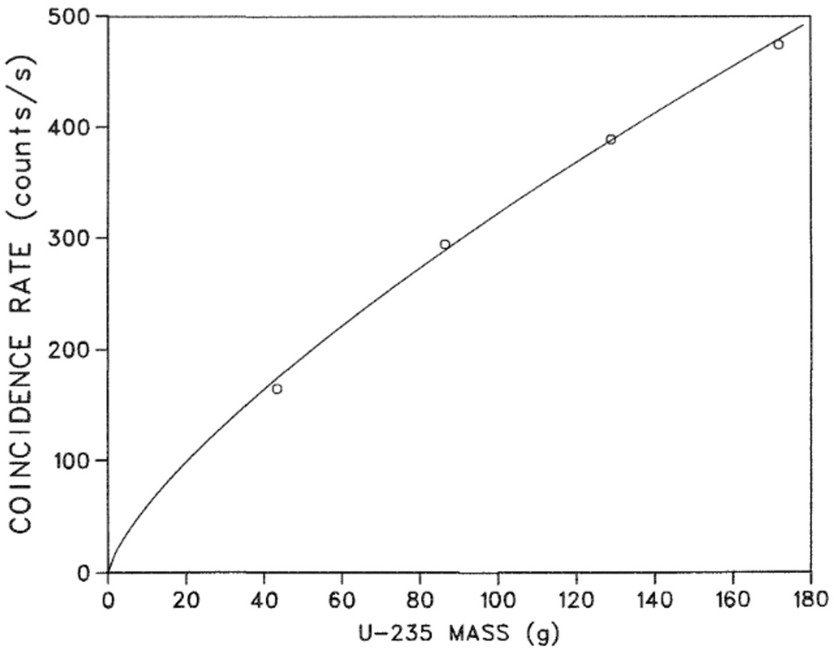
1. Perform active measurements with AmLi sources
2. Build a calibration curve with representative samples
3. Use curve and doubles count rate to determine mass

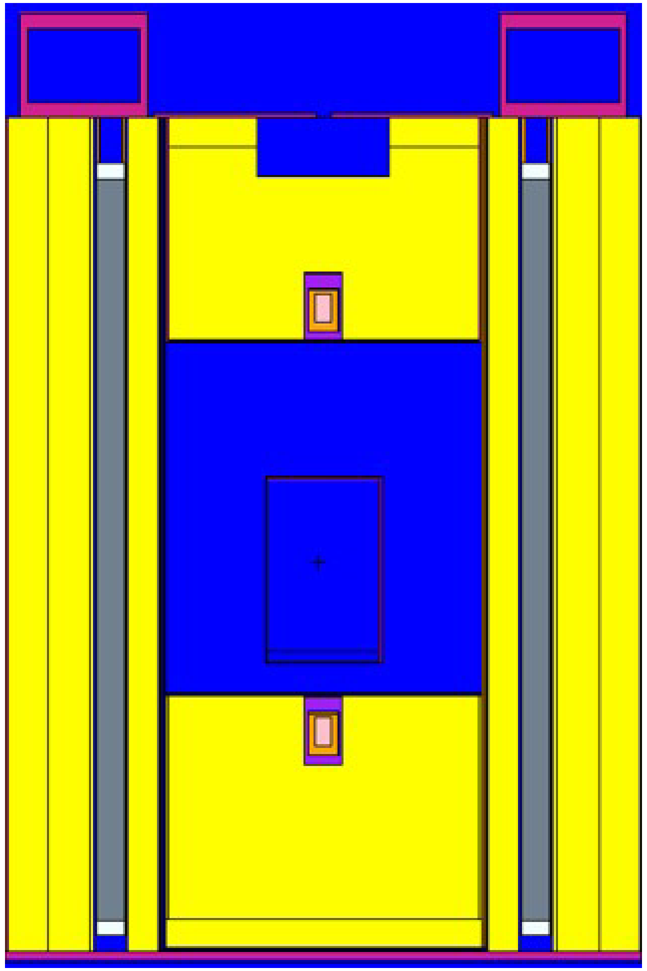
$$D = \frac{Am}{1+Bm}$$

$$\sigma_D = \frac{Am}{1+Bm}\sqrt{\left(\frac{\sigma_A}{A}\right)^2 + \left(\frac{\sigma_B m}{1+Bm}\right)^2 - 2\frac{m^2}{Am(1+Bm)}\sigma_{AB}}$$

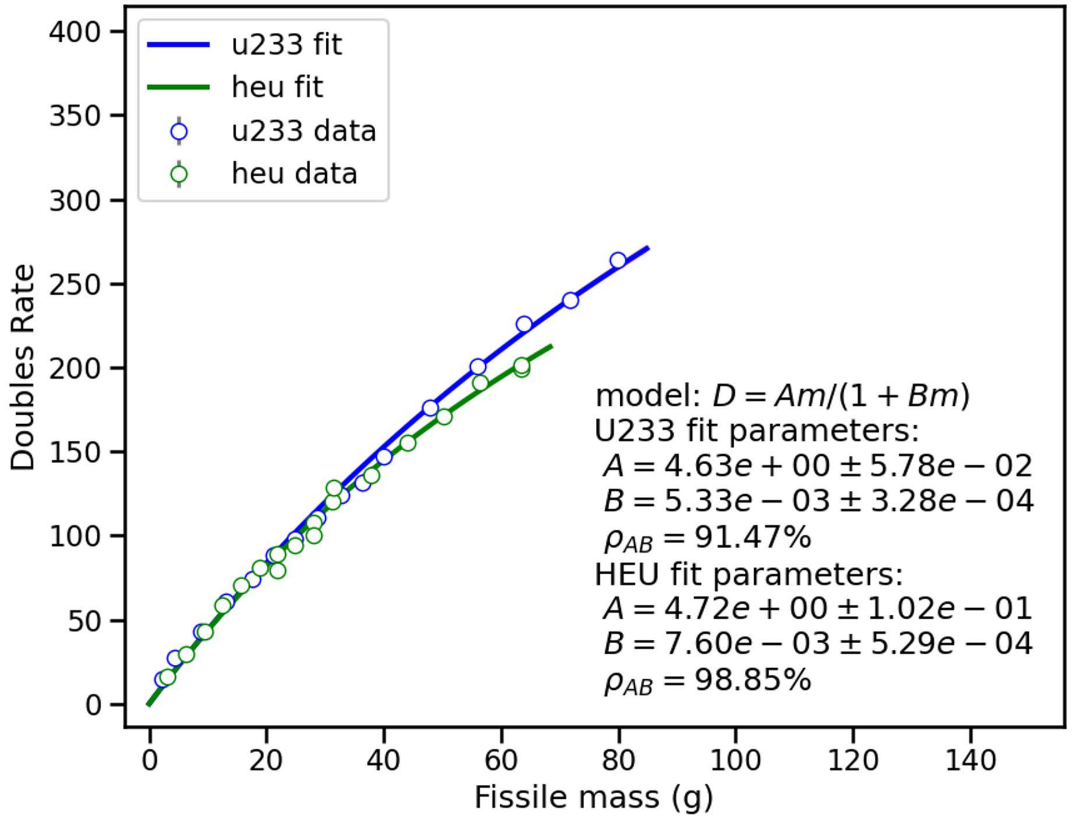
$$m = \frac{D}{A-DB}$$

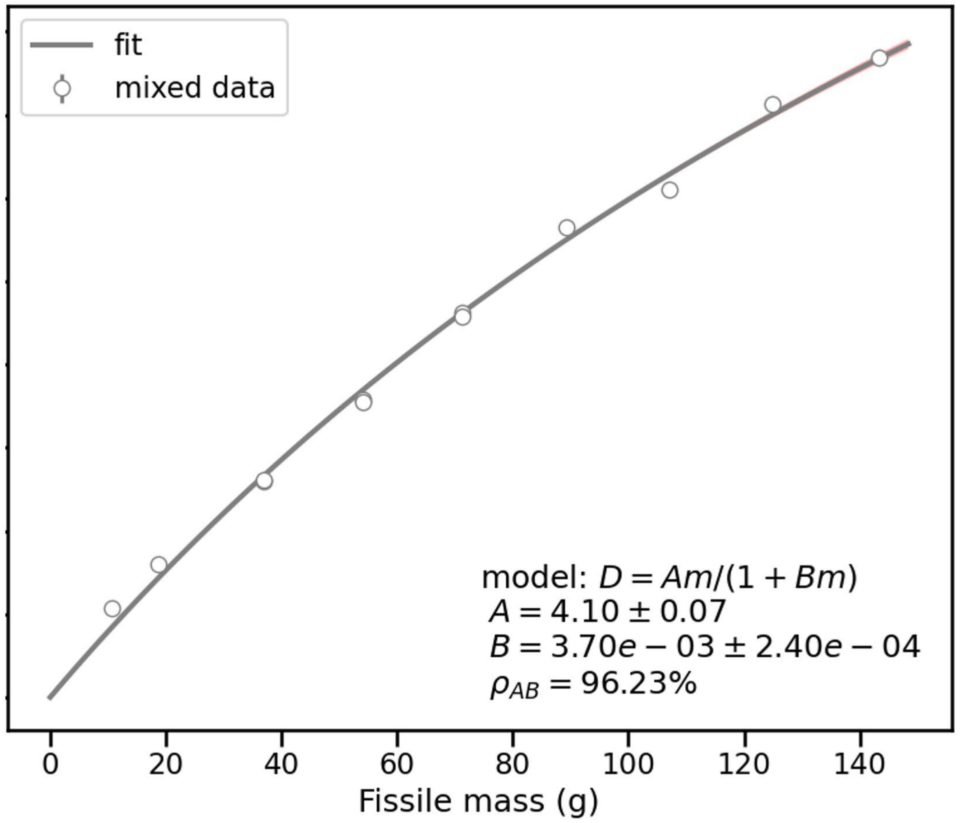
$$\sigma_D = m^2\sqrt{\frac{A^2}{D^4}\sigma_D{}^2 + \frac{1}{D^2}\sigma_A{}^2 + \sigma_B{}^2 - \frac{2}{D}\sigma_{AB}}$$



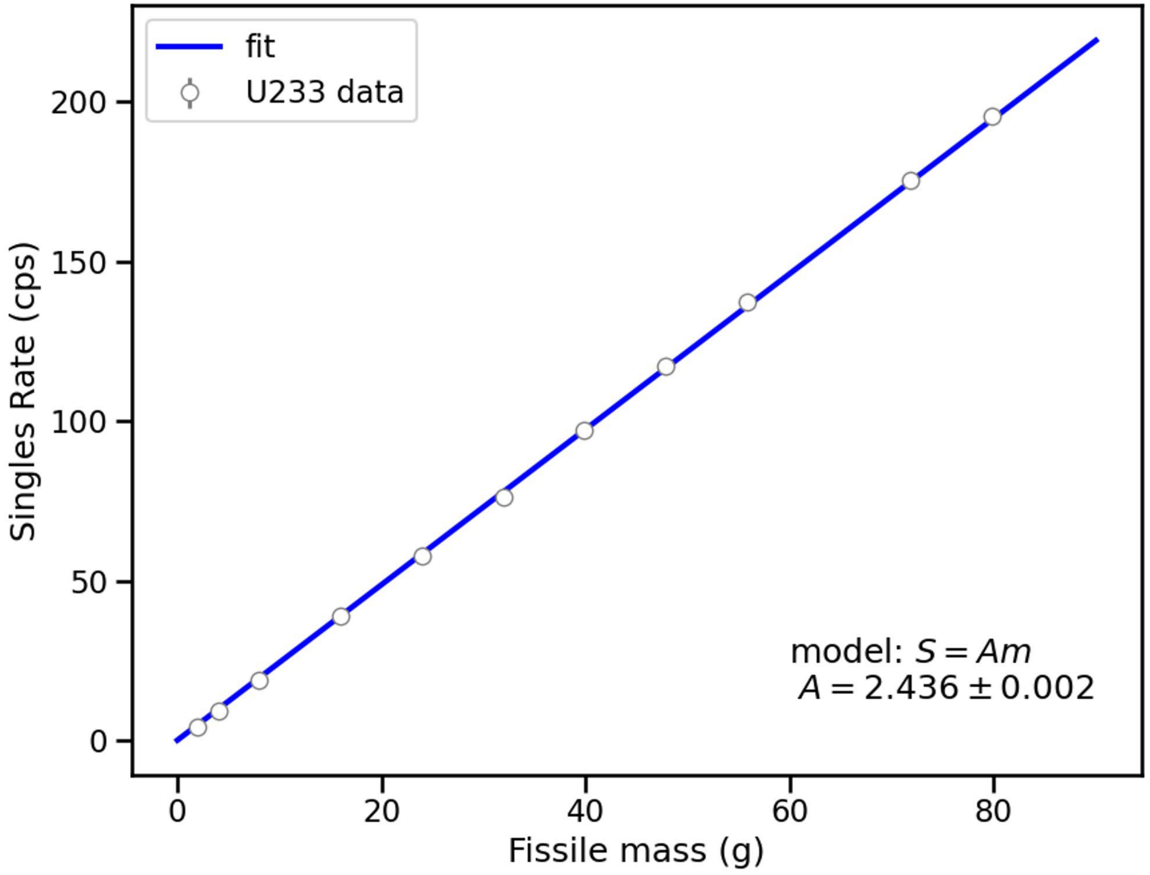


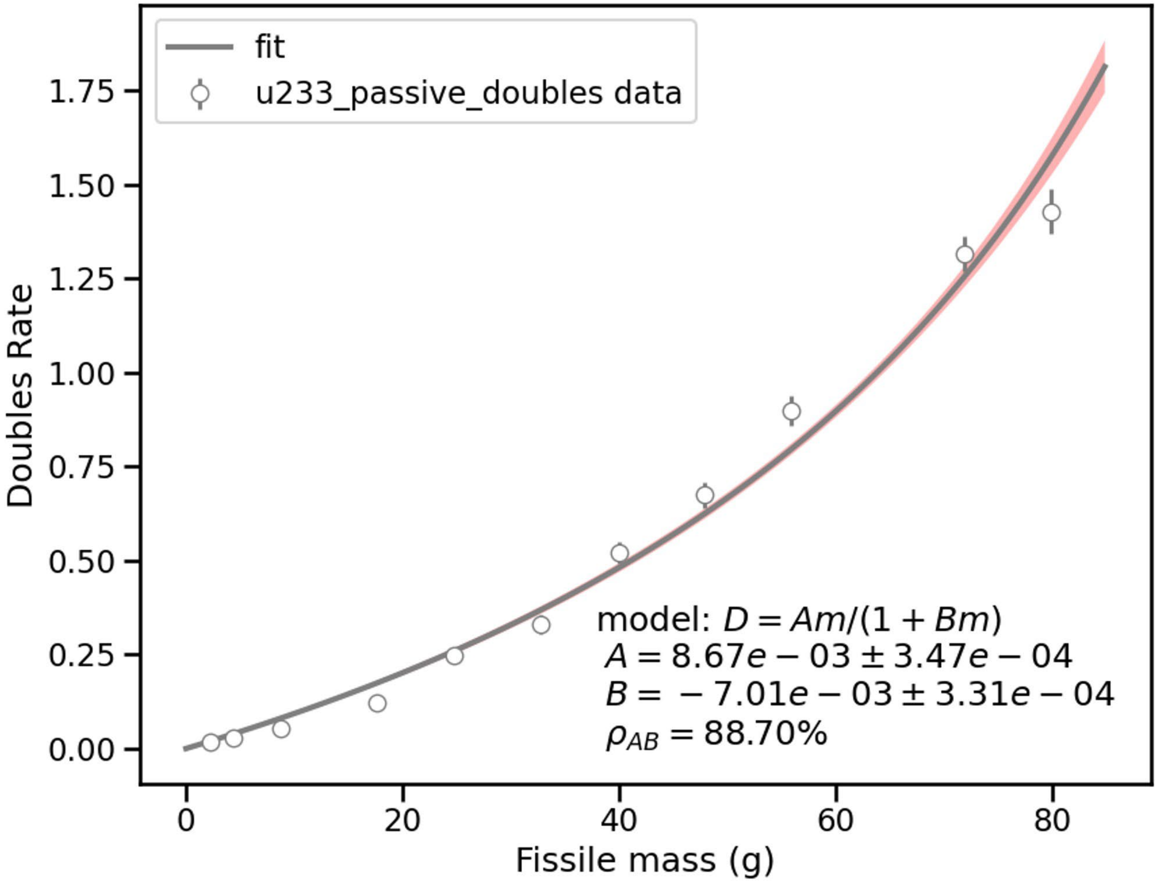
Active Doubles Calibration – Thermal Mode (no Cd)





Passive Calibration Curves





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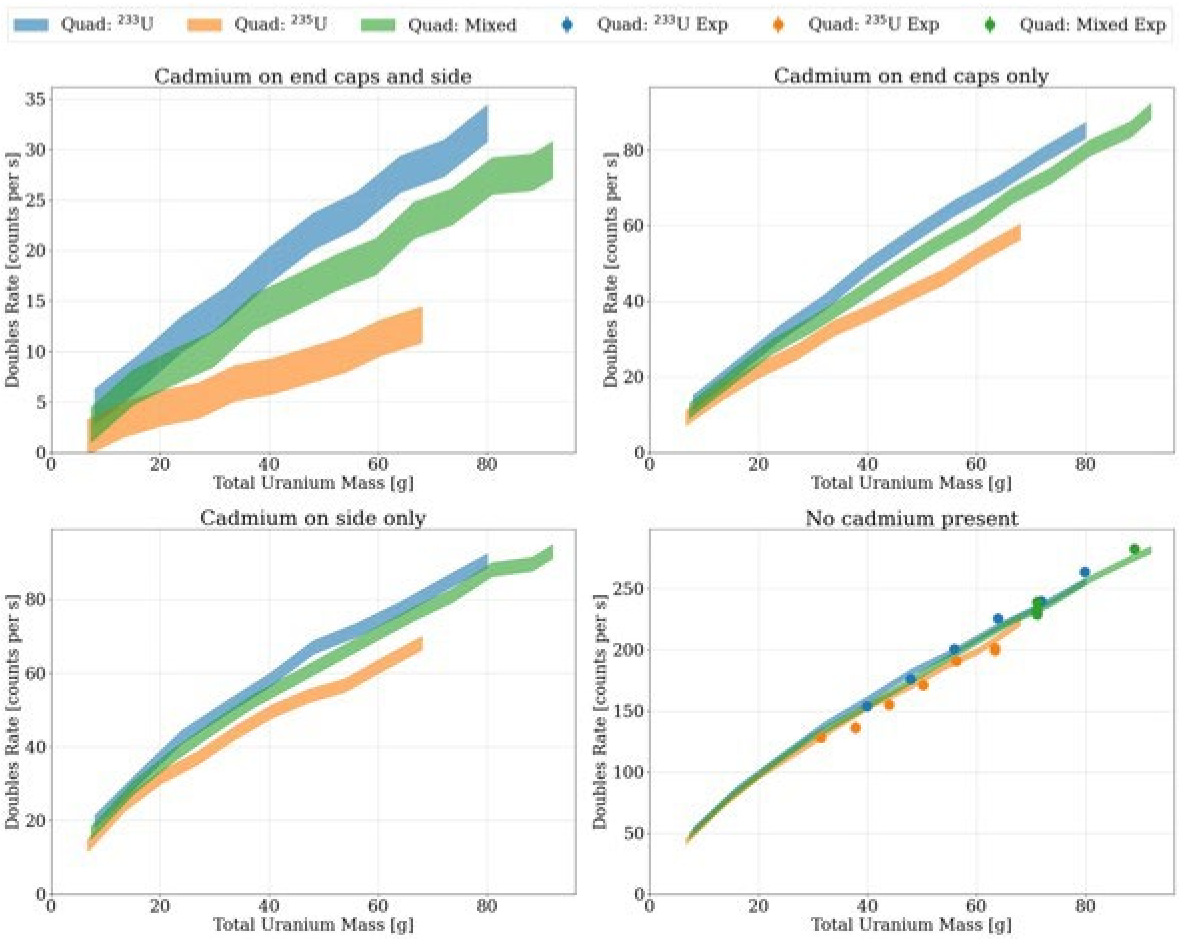
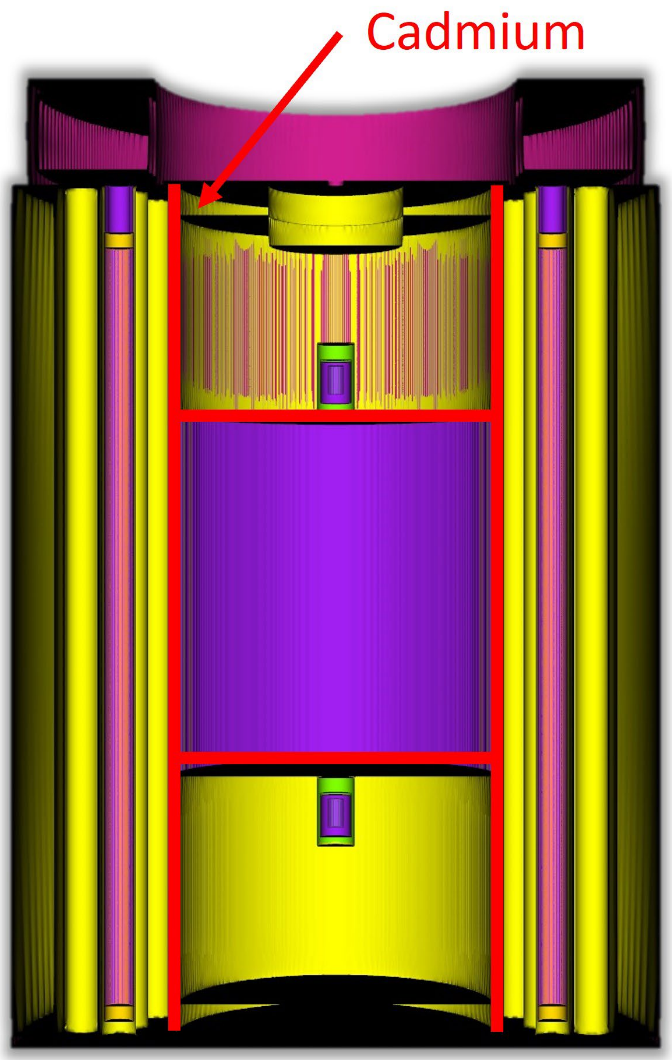
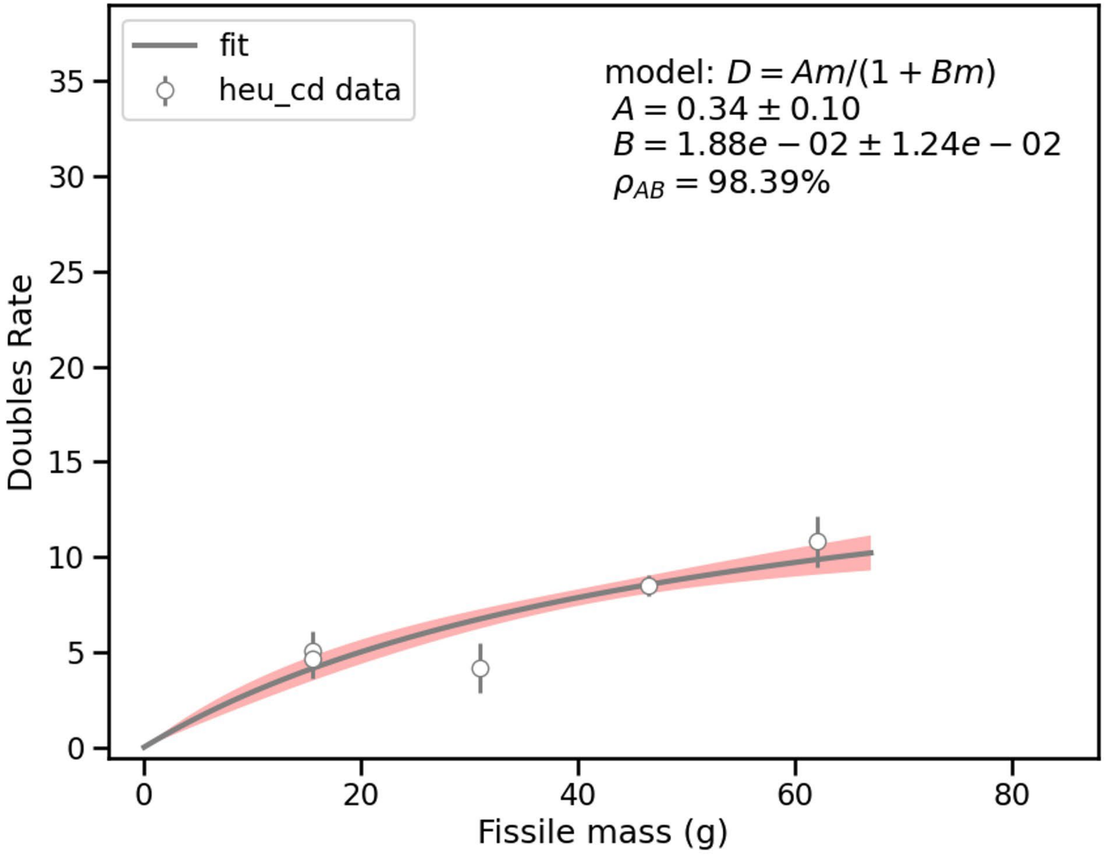
Active Doubles Simulation

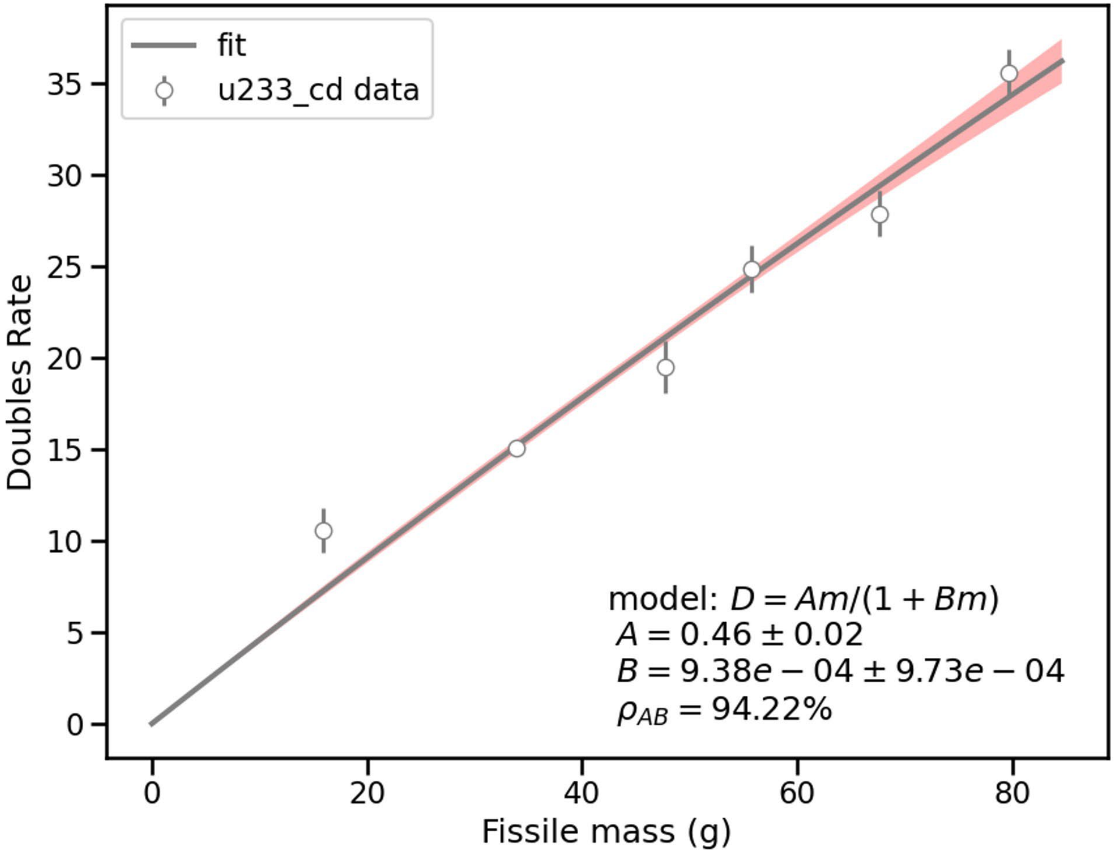




simulations performed by Richard Reed at ORNL

Active Doubles Calibration – Fast Mode (with Cd)





Quantification of an "unknown" source

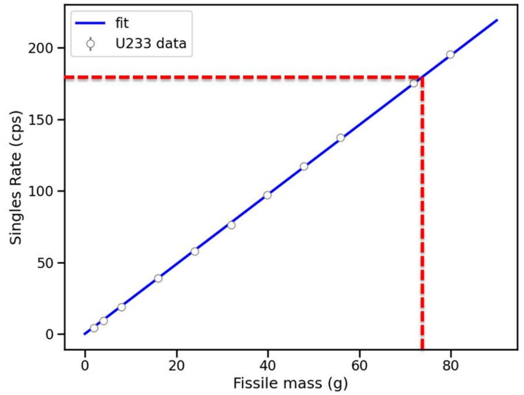
36 ^{233}U + 24 HEU triangles

1 Perform passive and active measurements of the item

2 Use calibration curves to determine the fissile mass and ^{233}U mass

3 Propagate uncertainty from measurements and fits for final mass estimate

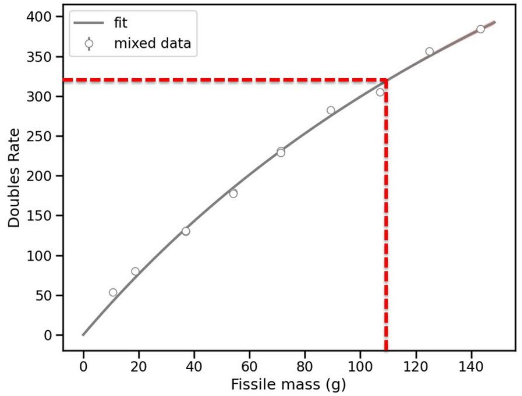
Passive measurement w/out Cd (thermal mode)
Singles Rate: **178.42 ± 0.41**



Known ^{233}U mass = **71.85 g**
Measured ^{233}U mass = **73.24 ± 0.18 g**

evidence of additional induced fission in ^{235}U

Active measurement w/out Cd (thermal mode)
Doubles Rate: **317.07 ± 3.94**



Known total fissile mass = **109.34 g**
Calculated fissile mass = **108.44 ± 2.04 g**

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Conclusion

- Development of safeguards techniques for ^{233}U is needed
- Neutron NDA techniques show promise to address this need
- Methods are under development to utilize combinations of neutron signatures to determine the composition and mass of materials containing ^{233}U and ^{235}U
- Characterization of materials containing ^{233}U requires the extension of current models and methods used for Pu and ^{235}U in traditional safeguards

Thank you!

Upcoming Webinars

Date	Title	Presenter
31 January 2024	Revolutionizing Nuclear Engineering Education: Developing Virtual Labs for Neutron Detection, Geiger Counter, and Reactor Experiments	Jonah Lau, Purdue University, USA
28 February 2024	The Analysis of the Reactivity Loss of the Phenix Core Cycles for the Experimental Validation of the DARWIN-FR Code Package	Victor Viallon, CEA, France
20 March 2024	Overview of Canadian R&D Capabilities to Support Advanced Reactors	Lori Walters, CNL, Canada