Delayed Neutron and Gamma Measurements of Special Nuclear Materials at the Royal Military College of Canada

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Delayed Neutron & Gamma Measurements at RMCC

- DNC system was designed and built in 2010 for the analysis of DN emissions from ^{233}U, ^{235}U & ^{239}Pu.
- Upgraded in 2012 to accommodate gamma measurements from SNM -> DNGC system.
- This system analyzes the temporal behaviour of the delayed neutrons and gammas to discern which fissile isotope(s) is(are) present .
- MCNP6 simulations of the experimental process are compared to simple measurements to characterize system behaviour.
- MCNP6 simulations will be used to identify useful delayed neutron and gamma signatures for the characterization of these SNM samples.
  - This includes gamma line pairs, the ratios of which can be used to characterize SNM.
  - Delayed neutron magnitudes and temporal behavior, activation products.
  - Will also aid in the determination of irradiation, decay, and counting times.

Application of the DNGC System

- Canada will evaluate its nuclear forensics capabilities in an upcoming exercise organized by the Nuclear Forensics International Technical Working Group (ITWG).

- RMCC is a participant in this exercise planned for the fall of 2014.
  - Site is licenced to handle and receive uranium and plutonium material.
  - Research at RMCC aims to characterize special nuclear material content in a variety of matrices.

- We will receive LEU samples of unknown enrichment and origin for characterization.

Separate from the ITWG exercise, the LEU sample, along with other SNM samples will be analyzed using RMCC's delayed neutron and gamma counting system.

- Will also characterize certified reference materials including ^{233}U, nat. and depleted U, ^{239}Pu to do a full assessment of the DNGC system's capabilities and limitations.

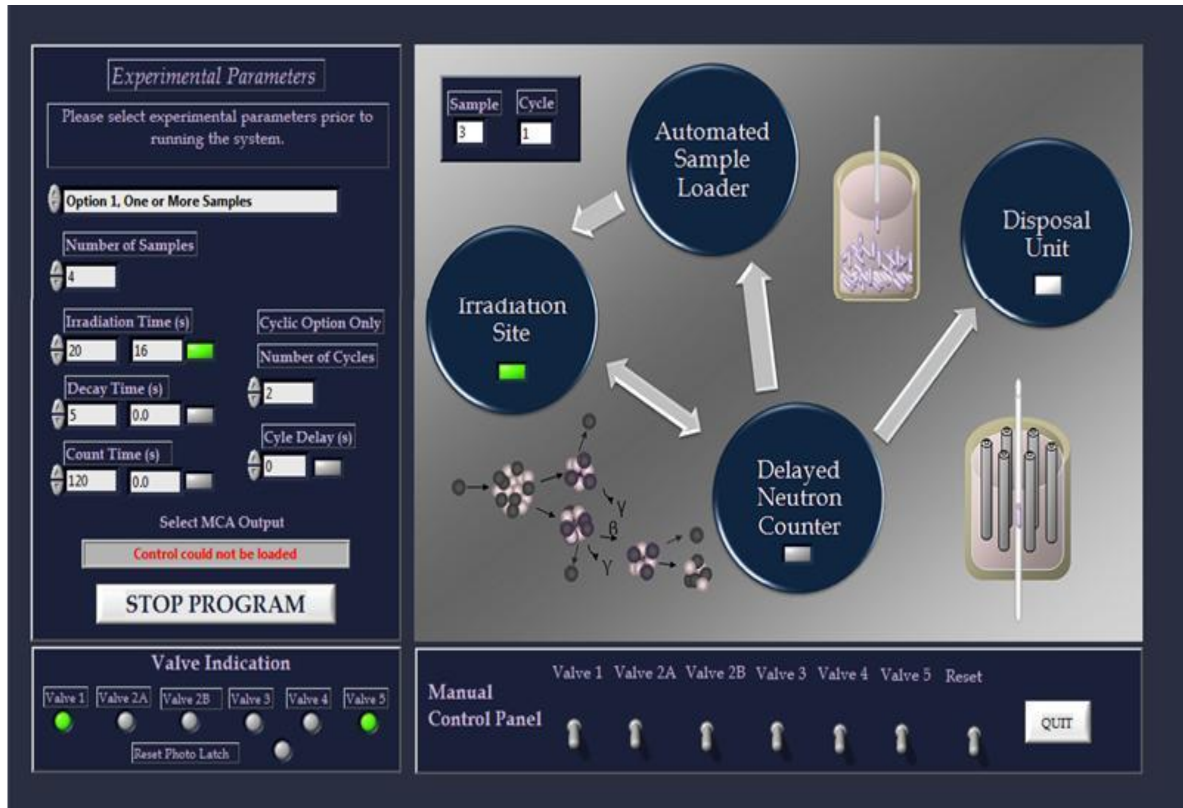
- Preparation for this evaluation includes the characterization of the DNGC system and a determination of relevant delayed particle signatures for SNM identification and measurement.

Delayed Neutron & Gamma Counting System

Aqueous samples containing fissile content is prepared from certified reference materials

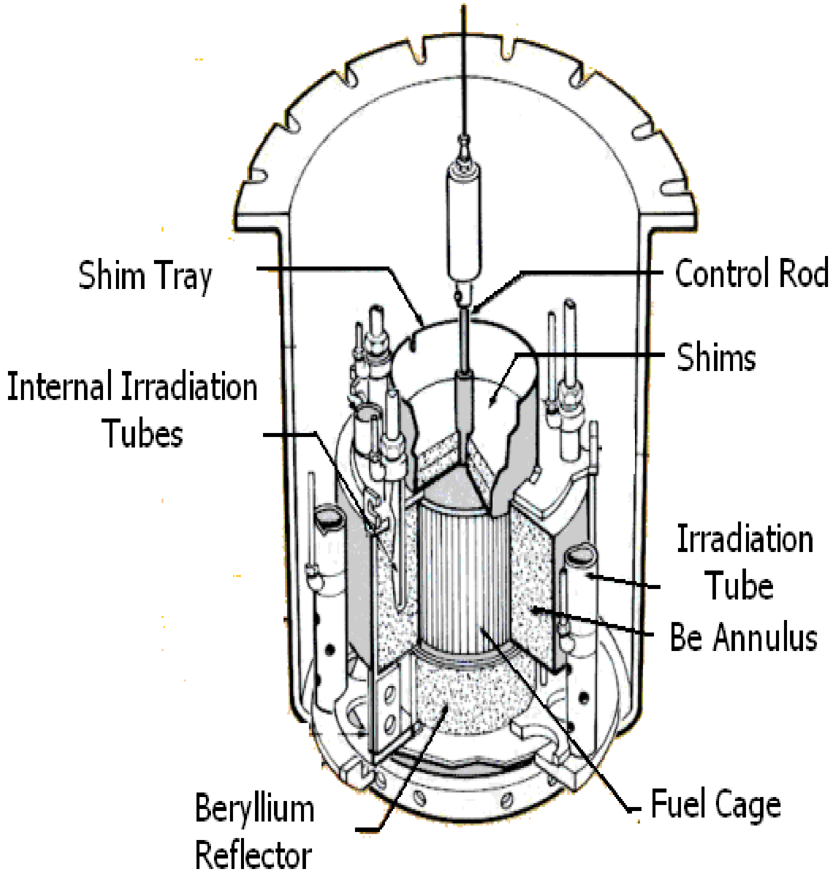
LabVIEW software controls data acquisition and hardware components.

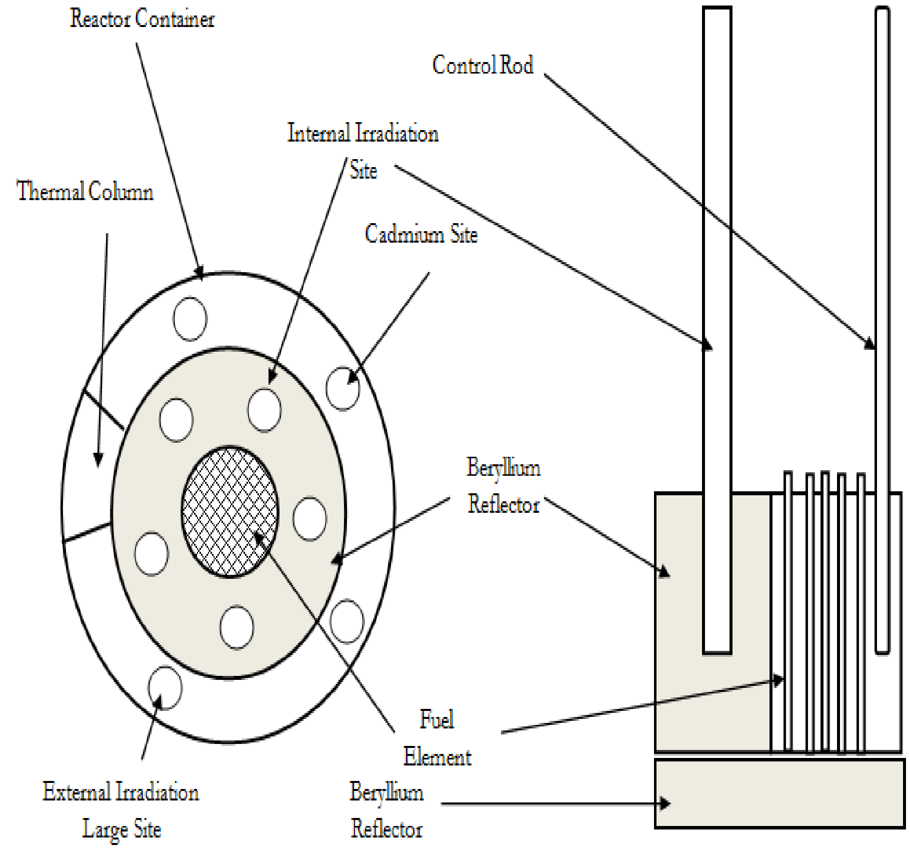
User specifies: Irradiation, decay, count time, count intervals and # of samples.



M.T. Sellers, D.G. Kelly, E.C. Corcoran, J. Radioanal. Nucl. Chem **291** *2* (2012).
M.T. Sellers, E.C. Corcoran, D.G. Kelly, J. Radioanal. Nucl. Chem **295** *2* (2013).

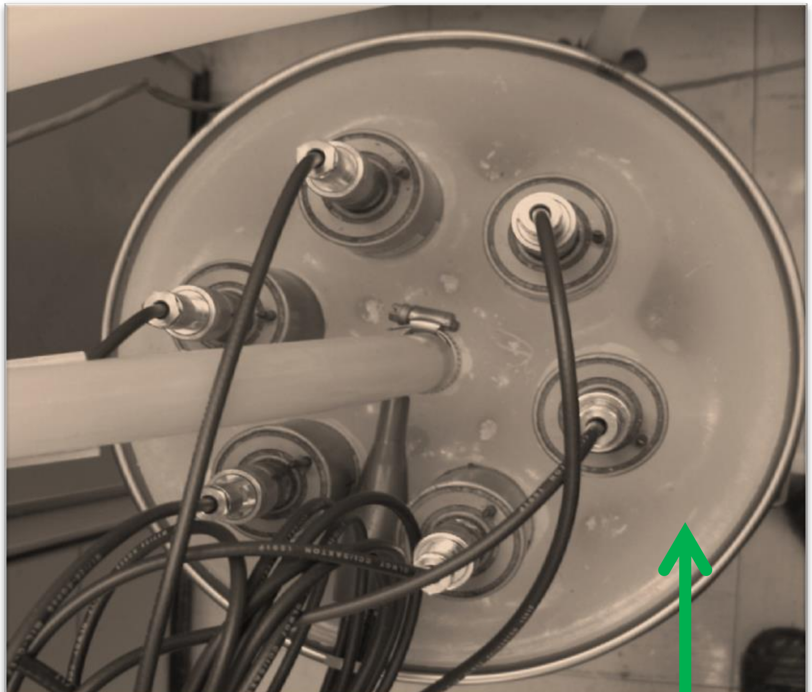
Irradiation in the SLOWPOKE-2 Reactor

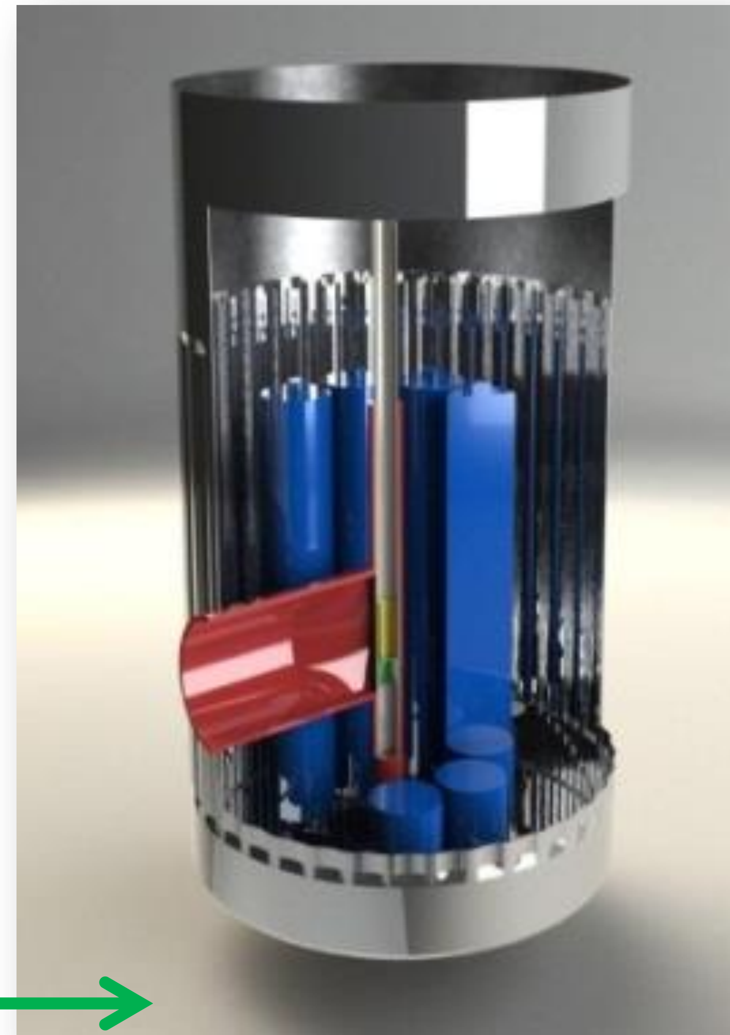




20 kW research reactor enriched to 19.89 % ^{235}U

Counting Arrangements

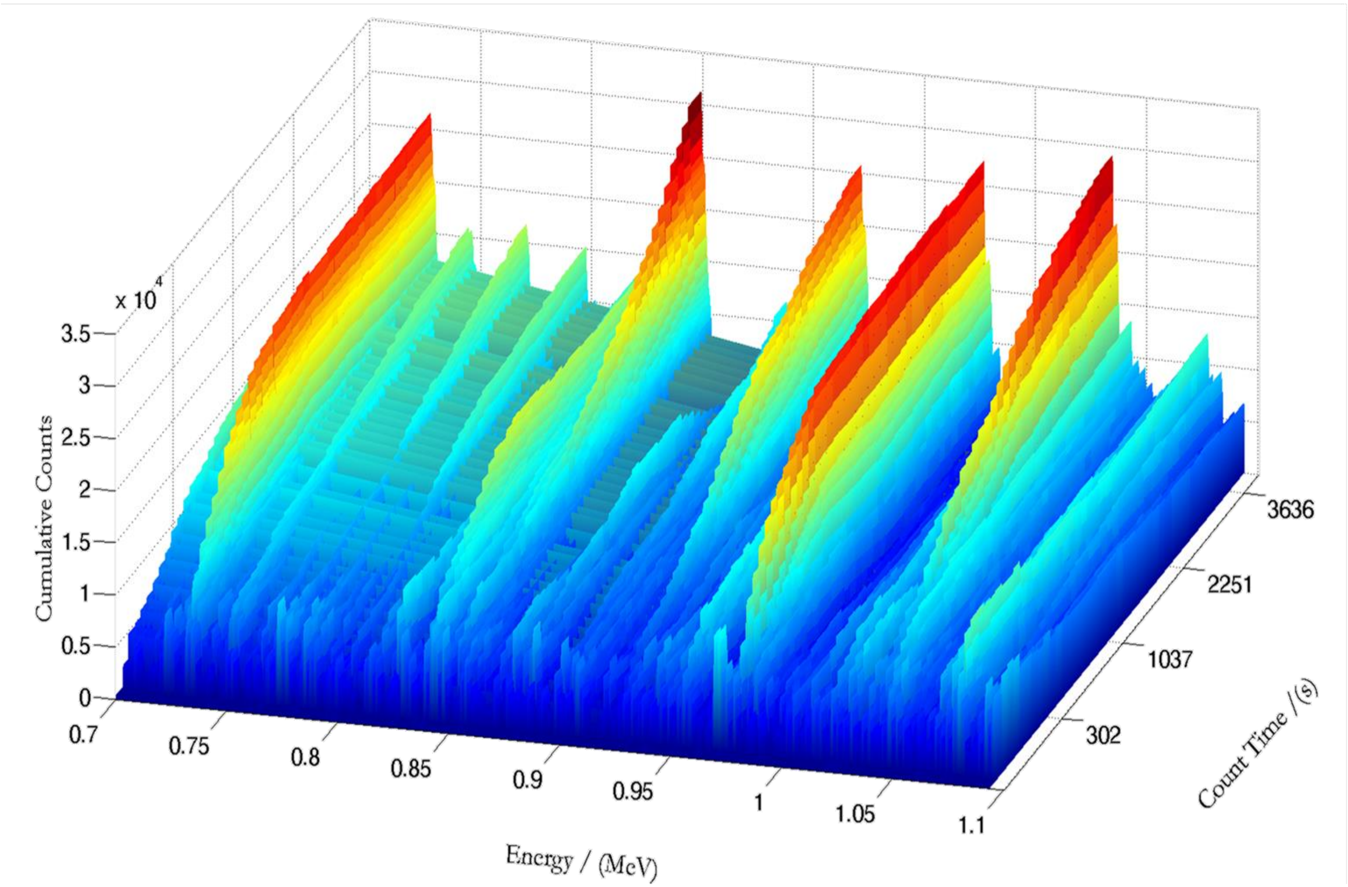




- Delayed Neutron Counter (2010-2012)
- Delayed Neutron & Gamma Counter (2012 – present)

Delayed Gamma Measurements

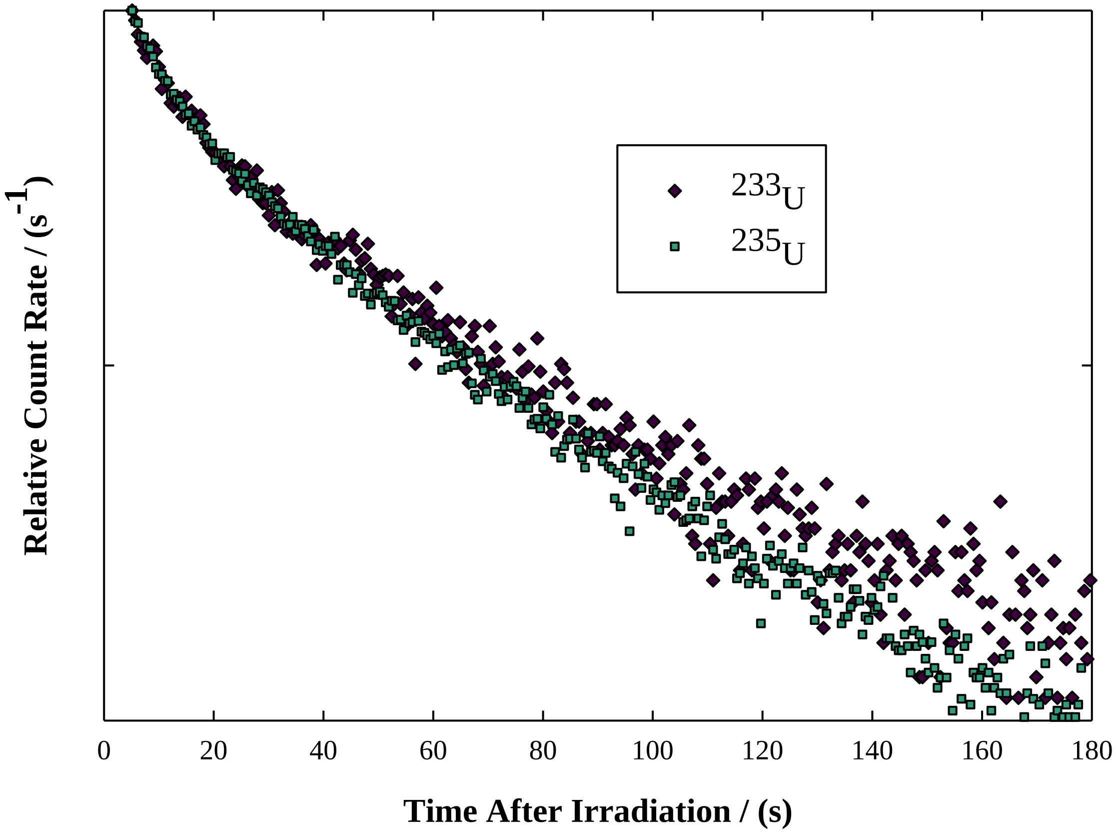
- DNGC system records neutron and gamma emissions as a function of time and energy.
- **Ratios of fission product gamma lines can be used to identify SNM.**

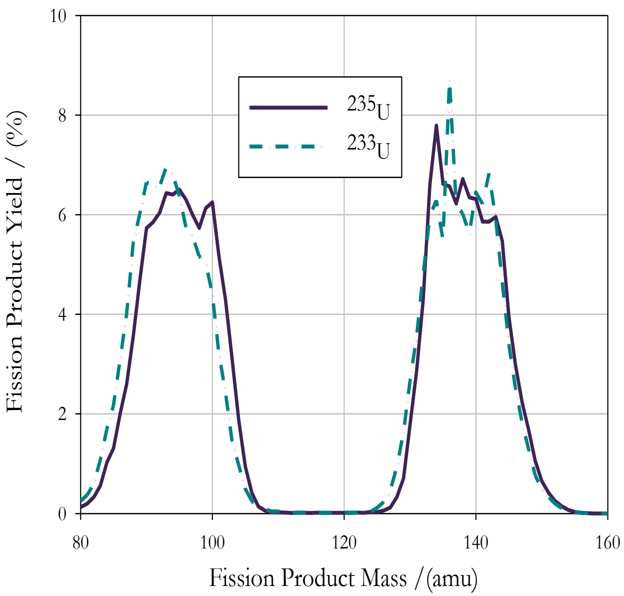


M.T. Andrews, *et al.* "A System for the Measurement of Delayed Neutrons and Gammas from Special Nuclear Materials" *submitted to J. Radioanal. Nucl. Chem.* September 2014.

Delayed Neutron Measurements

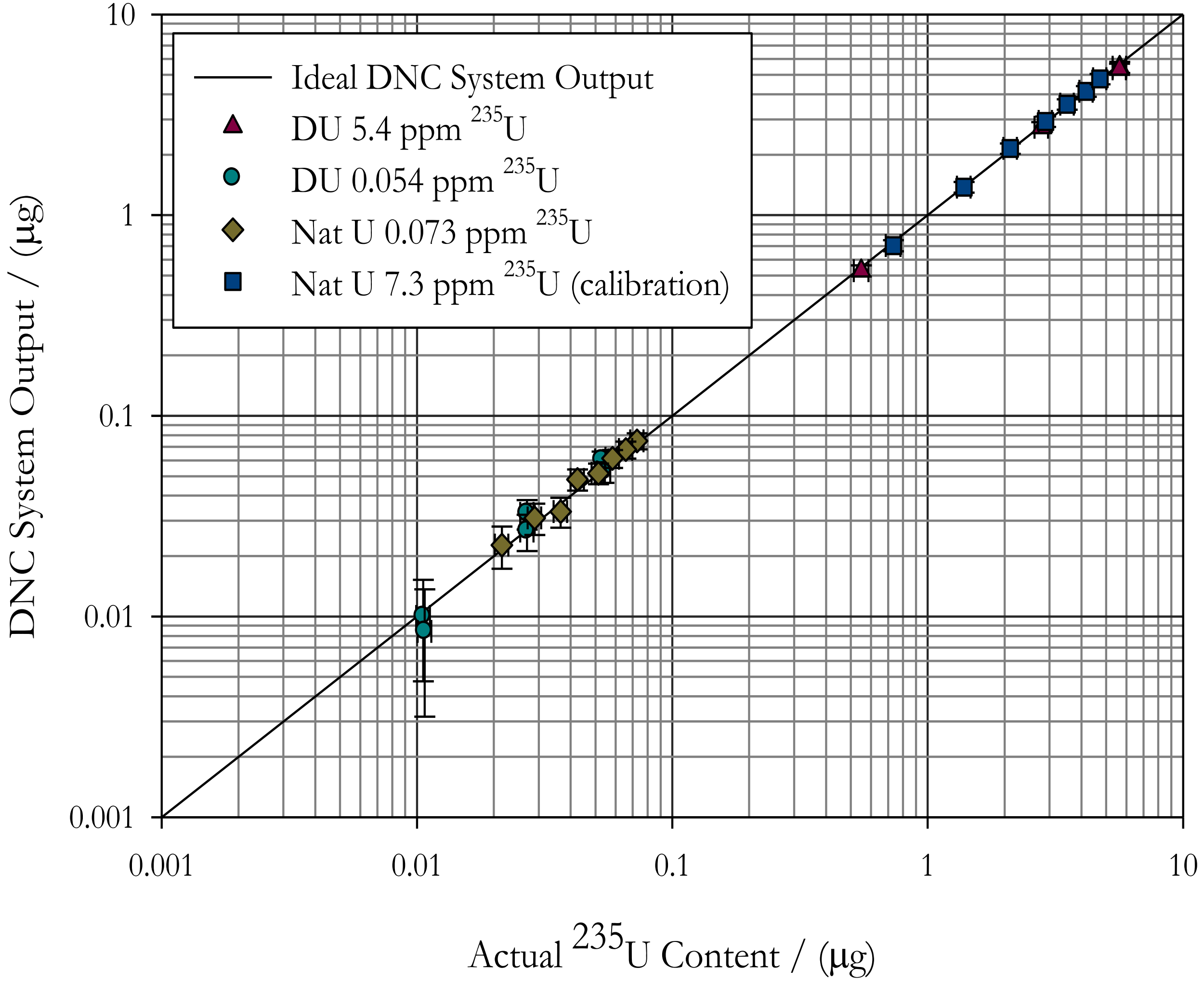
- Delayed neutron magnitude and temporal behavior is dependent on the fissile isotope.
- The temporal behavior can be used to characterize mixtures of SNM.
- RMCC uses the cumulative counts of pre-identified SNM to determine their magnitudes.





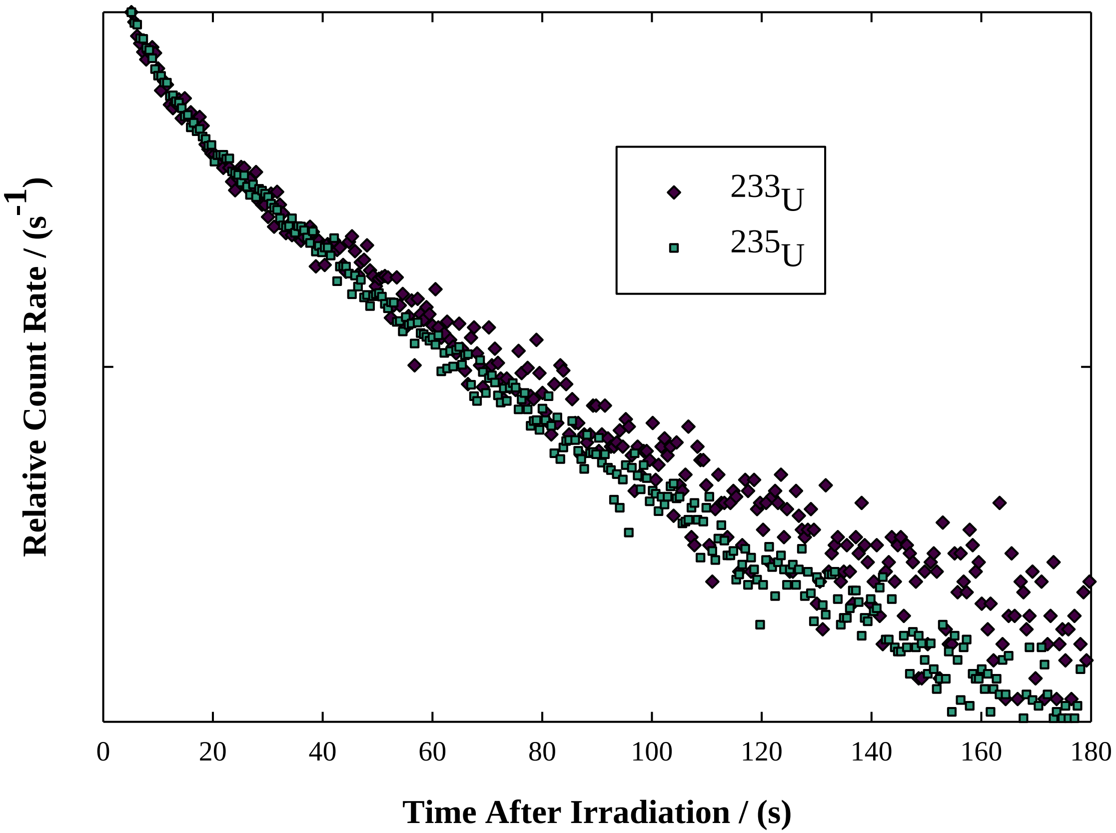
M.T. Sellers, *et al.* "Simultaneous ^{233}U and ^{235}U Characterization through the Assay of Delayed Neutron Temporal Behavior" *PHYSOR 2012.*.

Detection and Mass Determinations of Special Nuclear Material Quantities



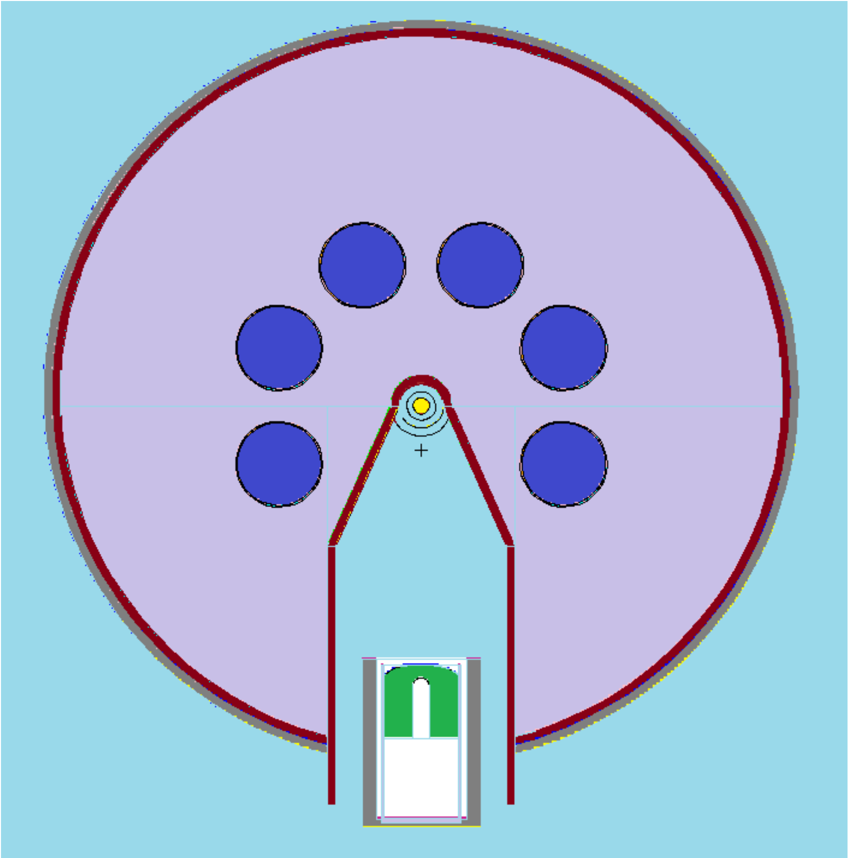
- Determined ^{235}U content in aqueous matrices ranging from 10 *ng* to 10 µg with average absolute error of 3.6 % using delayed neutron counts.
- Use of delayed neutron and gamma counts determined ^{233}U content with an average absolute error of 1.5 %.

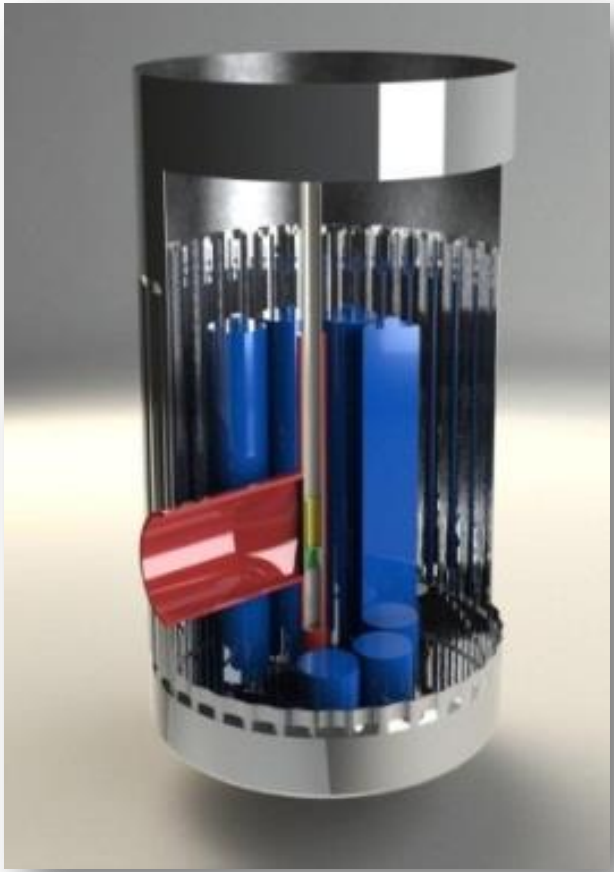
Detection and Mass Determinations of Special Nuclear Material Quantities



- Analyzed mixtures of ^{233}U and ^{235}U in triplicate.
- Predicted the ratio of ^{233}U to ^{235}U with an average absolute error of ± 4 %.

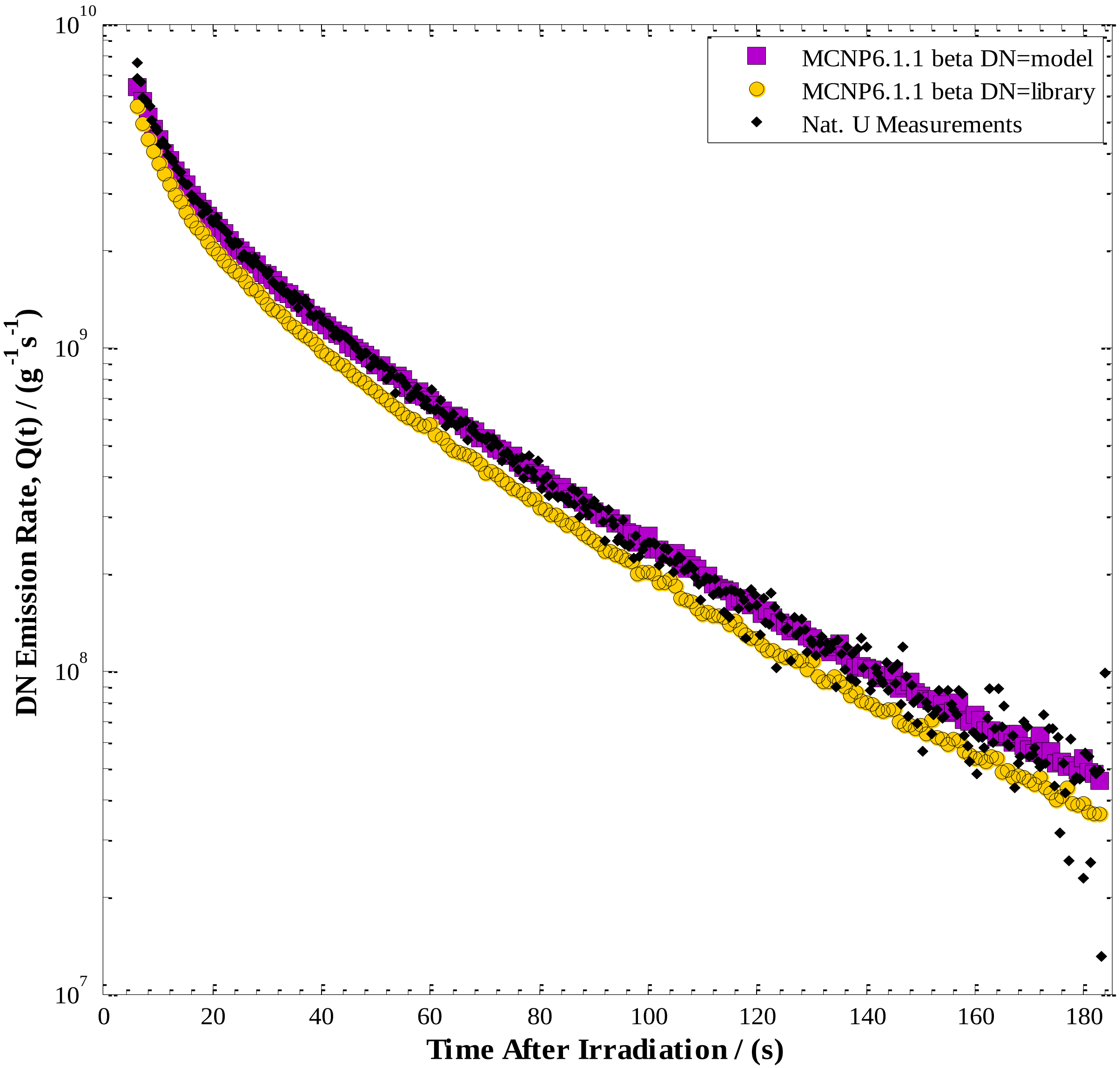
MCNP6 Simulations





- Used to characterize and confirm system behaviour.
- Aids in the identification of gamma and neutron emissions useful for fissile material identification.

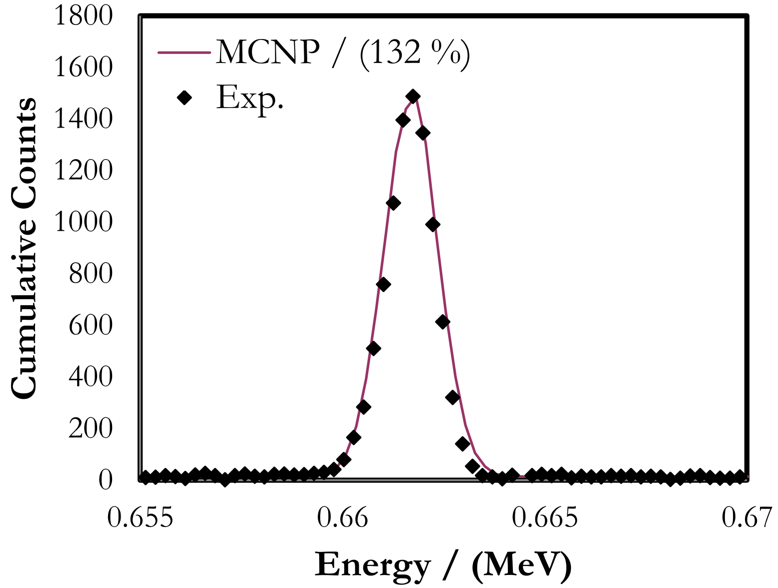
MCNP6 Delayed Neutron Simulations



- Evaluated three options available for DN simulations in MCNP6.1.1 β
- DN=model for MCNP6.1.1β had the best agreement with RMCC measurements for ^{233}U, nat. U, ^{239}Pu comparisons.

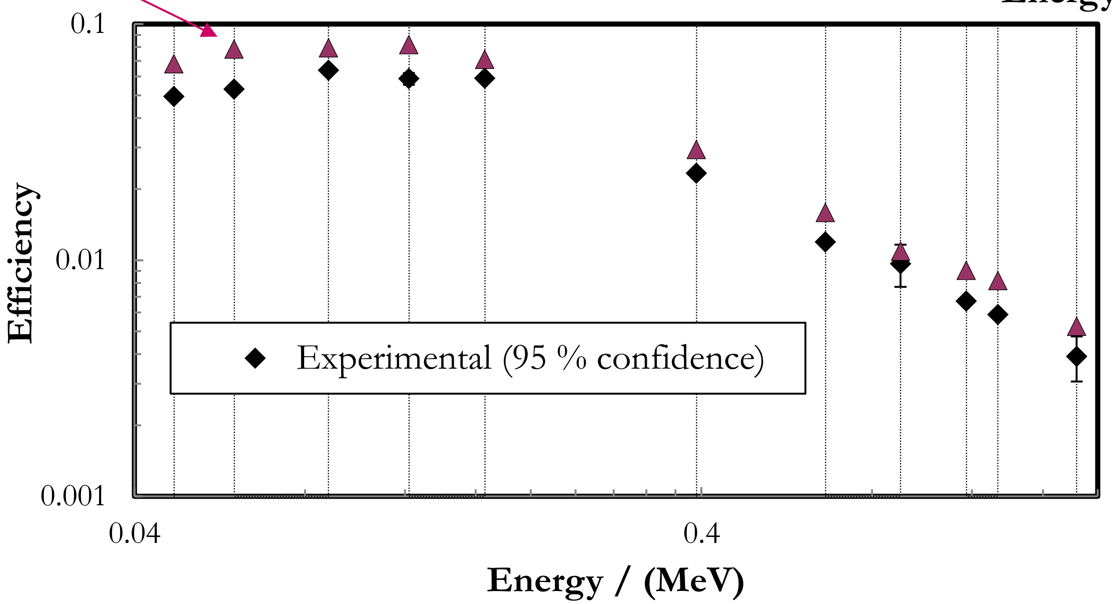
Multinuclide HPGe Detector Response in MCNP6

- Comparisons of MCNP & exp. FWHM and efficiencies were performed.
- *Gaussian Energy Broadening* card with pulse height tally reproduces energy resolutions.

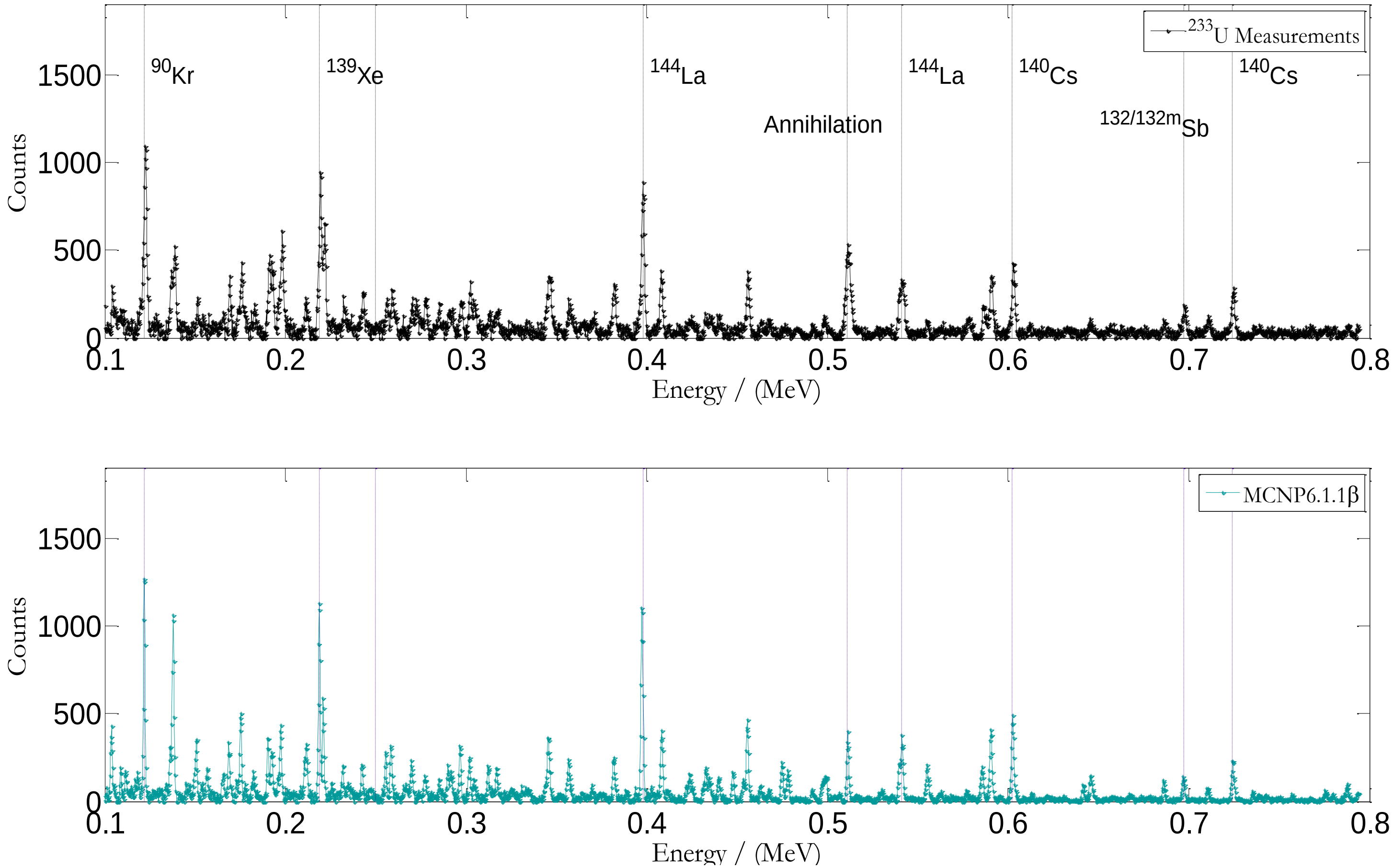


$$\left(\frac{\varepsilon_{MCNP}}{\varepsilon_{EXP}}\right) = 132 \pm 10\ \%$$

$$FWHM = a + b\sqrt{E + cE^2}$$



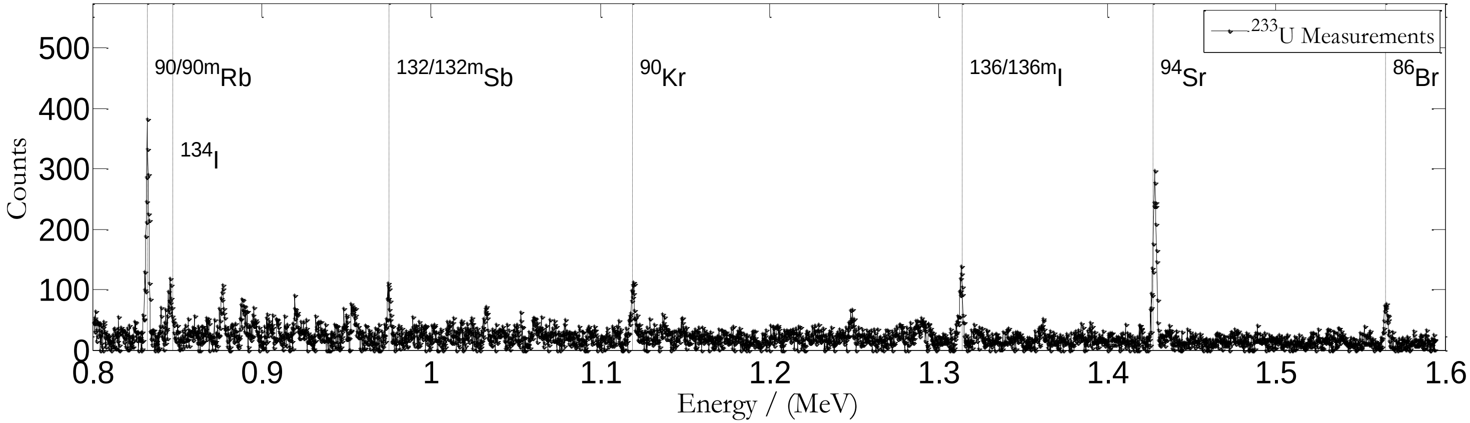
Measured and MCNP6 Peak Predictions

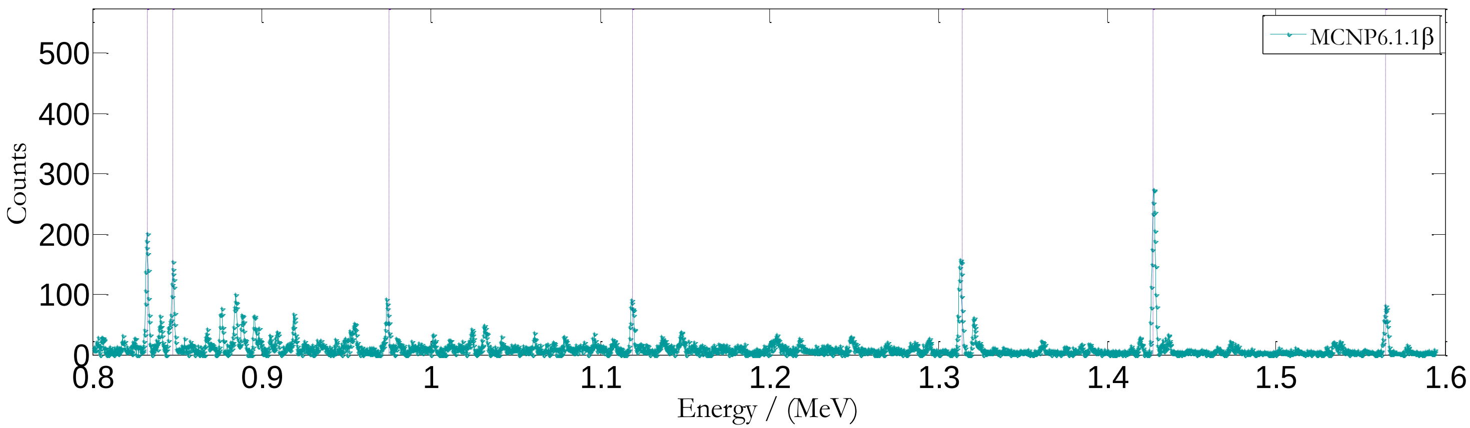


- MCNP6 peak prediction capabilities first were assessed by a qualitative comparison of the 25 most prominent measured peaks from 0.1 – 1.6 MeV for each solution.

- Every prominent measured peak had a corresponding MCNP6 peak.

Measured and MCNP6 Peak Predictions





- $> 0.8\ MeV$ peaks were selected for a more detailed comparison.
- Identified fission product peaks which were in good agreement with measurements.

MCNP6 Simulations – Delayed Gamma Signatures

- MCNP6 simulations were compared to measurements at RMCC to select gamma line pairs appropriate to use for fissile isotope identification.
- *Marrs et al**. also identified gamma line pairs useful for fissile material identification over a wide range of counting times.

MCNP6 & Measured Expected Delayed Gamma Intensities in the DNGC System – 18 min decay, 15 min count

Fission Products	Line Pair (keV)	^{233}U	^{235}U	^{239}Pu
^{89}Rb/^{138}Cs	1032/1010	2.5 ± 0.2	1.45 ± 0.06	0.62 ± 0.04
		2.45 ± 0.04	1.55 ± 0.02	N/A

*Marrs, R. E., et al. "Fission-product gamma-ray line pairs sensitive to fissile material and neutron energy." *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 592.3 (2008): 463-471.

Conclusions & Acknowledgements

- Ability to detect and quantify small amounts of SNM non-destructively and rapidly via delayed neutron and gamma counting is established at RMCC.
- MCNP6 was used to identify gamma line pairs useful for fissile material identification.
- Preparation is underway for the analysis of LEU, ^{239}Pu, ^{235}U, and ^{233}U.
  - MCNP6 simulations of the DNGC system have focused on the identification of characteristic gamma lines from prominent fission products.
  - Neutron and gamma signatures will be used in complement for SNM characterization.

Project Funding

- CNSC/NSERC Doctoral Award
- Canadian DND
- ASC program at LANL (for MCNP comparisons)

Thank you.

1. M.T. Sellers, D.G. Kelly, E.C. Corcoran, "An Automated Delayed Neutron Counting System for the Mass Determinations of Fissile Isotopes" *Journal of Radioanalytical and Nuclear Chemistry* **291** 2 (2012), 281-285.

2. M.T. Andrews, J.T. Goorley, E.C. Corcoran, D.G. Kelly, "Modeling the Detection of Delayed Neutron Signatures in MCNP6 and Comparisons with Measurements of ^{233}U, ^{235}U and ^{239}Pu" *Journal of Nuclear Technology* **187** *3* (2014) 235-242.

3. M.T. Sellers, E.C. Corcoran, D.G. Kelly, "Simultaneous ^{233}U and ^{235}U characterization through the assay of delayed neutron temporal behavior" *PHYSOR 2012*, Knoxville, TN, April (2012).

4. M.T. Andrews, E.C. Corcoran, D.G. Kelly, J.T. Goorley, "Fission Product γ-ray Measurements of ^{235}U and MCNP6 Predictions" *Transactions of the American Nuclear Society*, **109** (2013) 995 – 998.

5. M.T. Sellers, E.C. Corcoran, D.G. Kelly, "The analysis and attribution of the time-dependent neutron background resultant from sample irradiation in a SLOWPOKE-2 reactor" *Journal of Radioanalytical and Nuclear Chemistry* **295** 2 (2013) 1221-1228.