Monte Carlo Analysis of Neutron Slowing-Down-Time Spectrometer for Spent Fuel Assay

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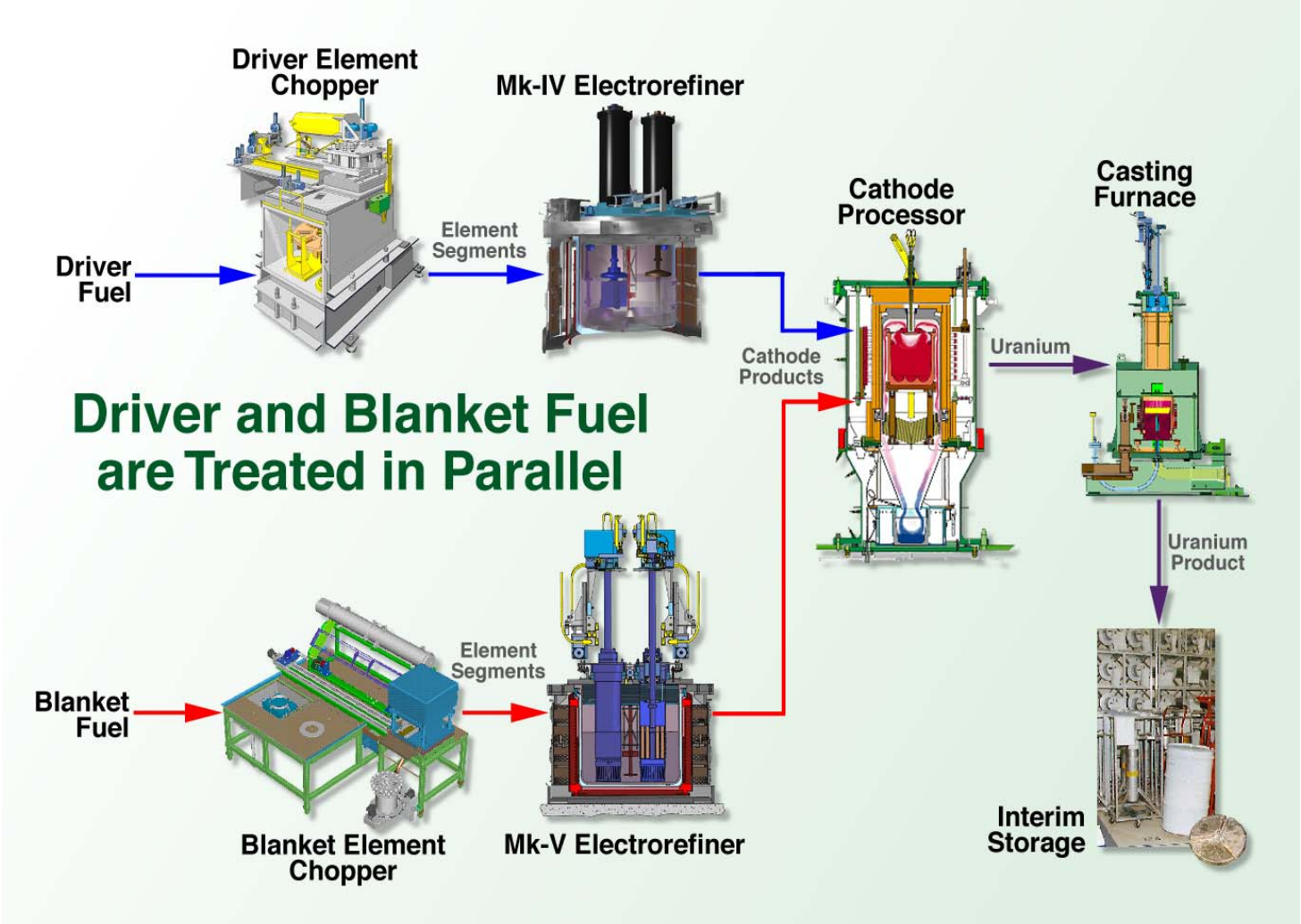
Project Objective

- To investigate the feasibility of using Neutron Slowing Down Time Spectrometer (NSDTS) to improve the input accountancy of Pyro-processing of Advanced Burner Test Reactor (ABTR) Spent Fuel

Outline

- Introduction to pyro-processing, materials accountancy, and non-destructive assay for nuclear spent fuel
- Neutron Slowing Down Method
- Comparison between lead and graphite as slowing down spectrometer material
- Self-shielding effect
- Preliminary experiment results
- Future work

INL Pyro-processing Flow Chart



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Introduction to Materials Accountancy

- Fissile material inputs to any reprocessing facility must be known accurately and precisely
- Demands (DOE, NRC, IAEA) are tough today and will likely be even tougher in a GNEP world
- It's an issue for aqueous and pyro, but especially pyro (no homogeneous sample point)
- Goal is unspecified, but consensus is < 1% uncertainty on fissile material input

Introduction to Materials Accountancy

- Traditional thinking on pyro: use some combination of reactor physics calculations, segment sampling with wet chemistry, and nuclear NDA
- Reactor physics calculations alone can't succeed in a fast burner reactor context – the U238 capture rate is very difficult to calculate accurately
- Sampling is done as a way to normalize the calculations (only relative information, not absolute).
- Also destructive sampling could be not reliable when the spent fuel has poor homogeneity (as for pyro-processing).

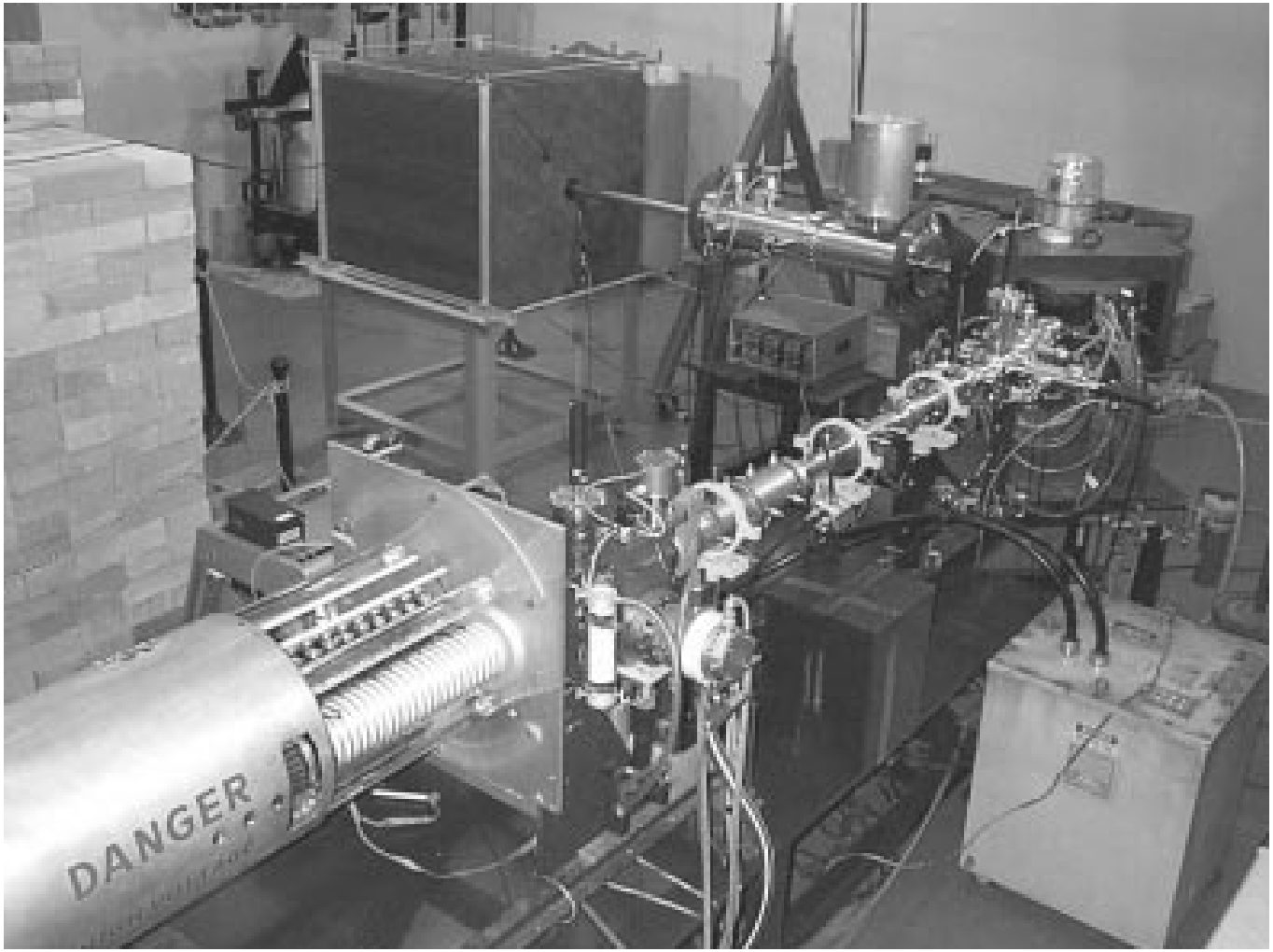
Discussion of NDA Spent Nuclear Fuel Assay Methods

- Extensive NDA methods were investigated to assay spent fuel and radioactive solutions
- Eight methods are reported to be able to measure the total fissile content, but only two are reported to be able to determine Pu-239 and U-235 respectively
- Neutron Resonance Transmission: Use filtered reactor neutron beam, similarly it can be done using time-of-flight method with pulsed neutron from accelerator. Too low neutron flux cause too long assay time.
- Multiple Detector Coincidence System: use multiplicity method to distinguish spontaneous fission neutrons from (α, n) neutrons, Pu-239 amount is not measured directly, but inferred from Cm-244 content.
- NDA has not been explored as part of pyro development, because up to now, we didn't see a technique of sufficient promise

Neutron Slowing Down Spectrometer

- Pulsed neutron slowing down technique
  - High energy neutron pulse
  - Low absorption cross section, high elastic cross section medium
  - Neutron pulse slowing down mean energy correlates to slowing down time

A Slowing Down Spectrometer Setup using Graphite (Hawari, 1999)



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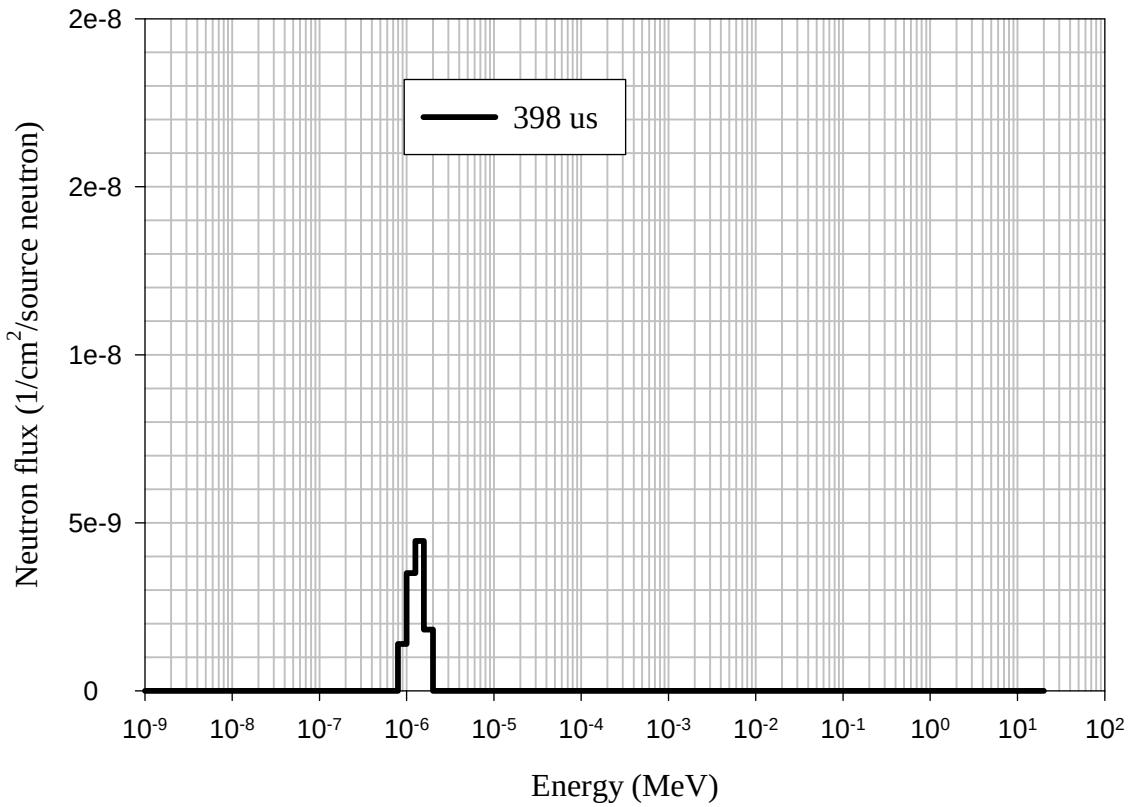
Pulse Neutron Slowing Down



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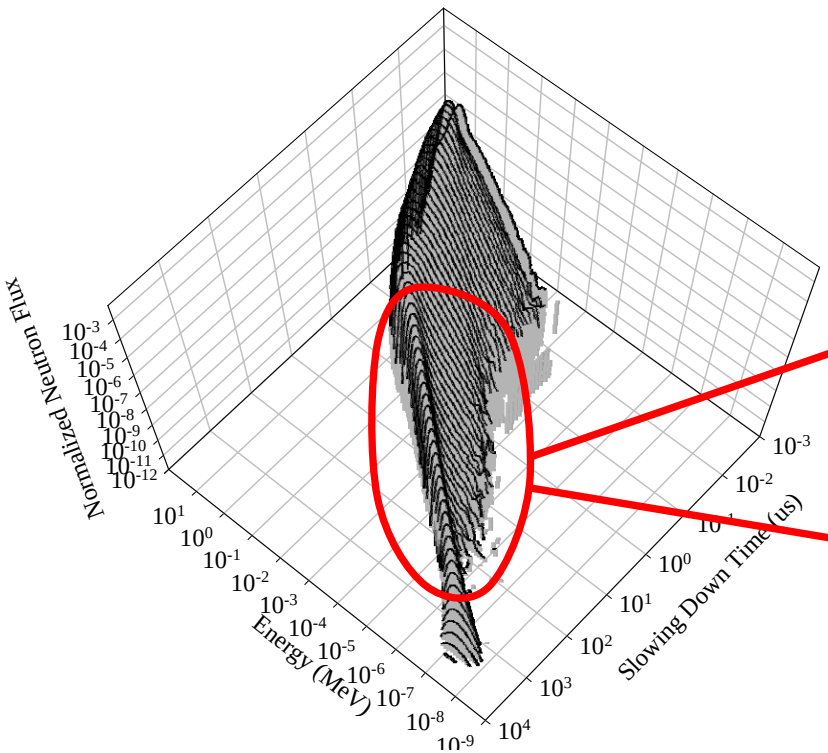
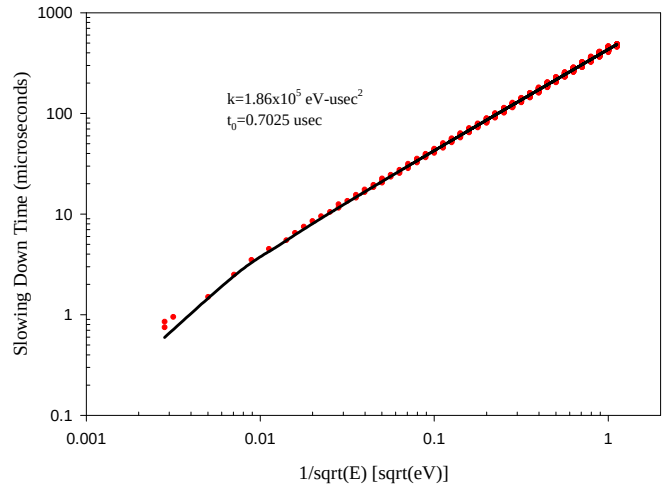
Neutron Slowing Down Energy-Time Correlation





$$\overline{E} = \frac{k}{(t+t_0)^2}$$

- E is the mean energy of the neutron slowing down pulse
- k and t_0 are constants determined by moderating medium and STDS geometry

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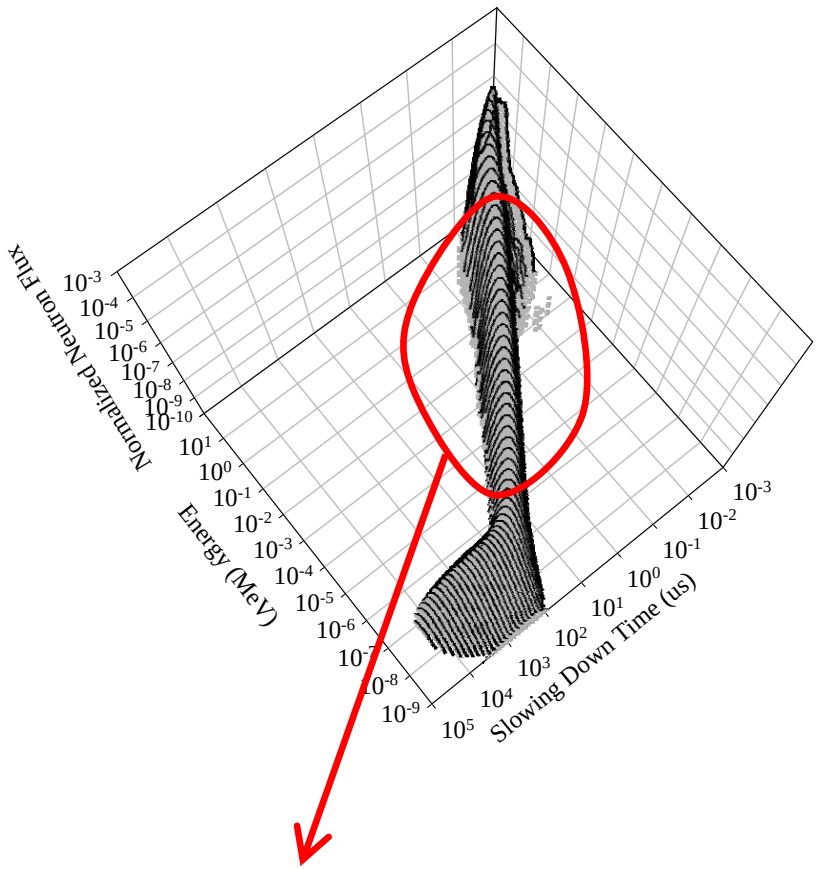
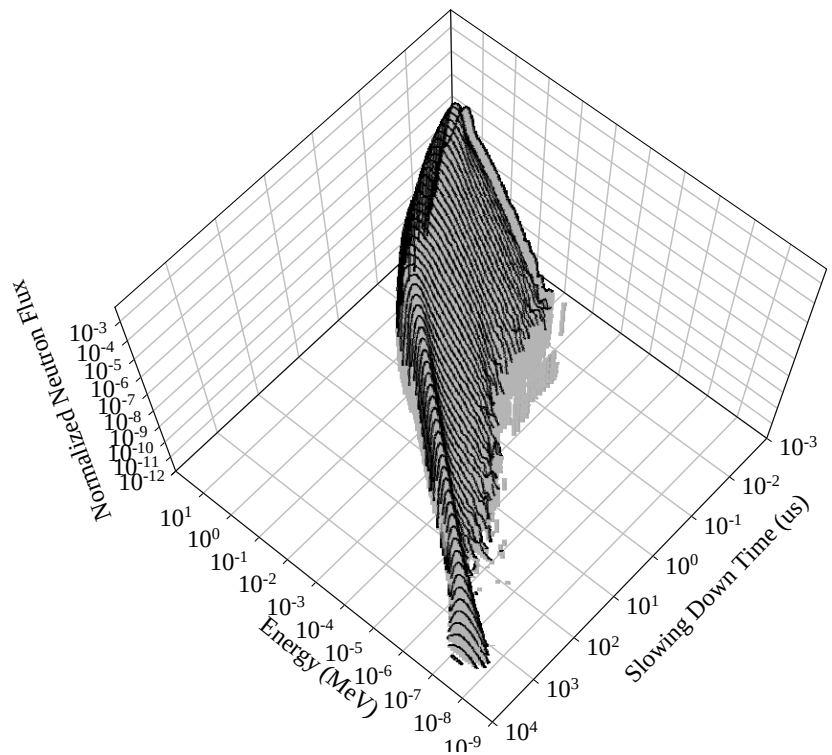
Neutron Slowing Down Comparison Between Graphite and Lead





Graphite: k = 0.498 keV*μs^2, t_0 = 0.8428μs

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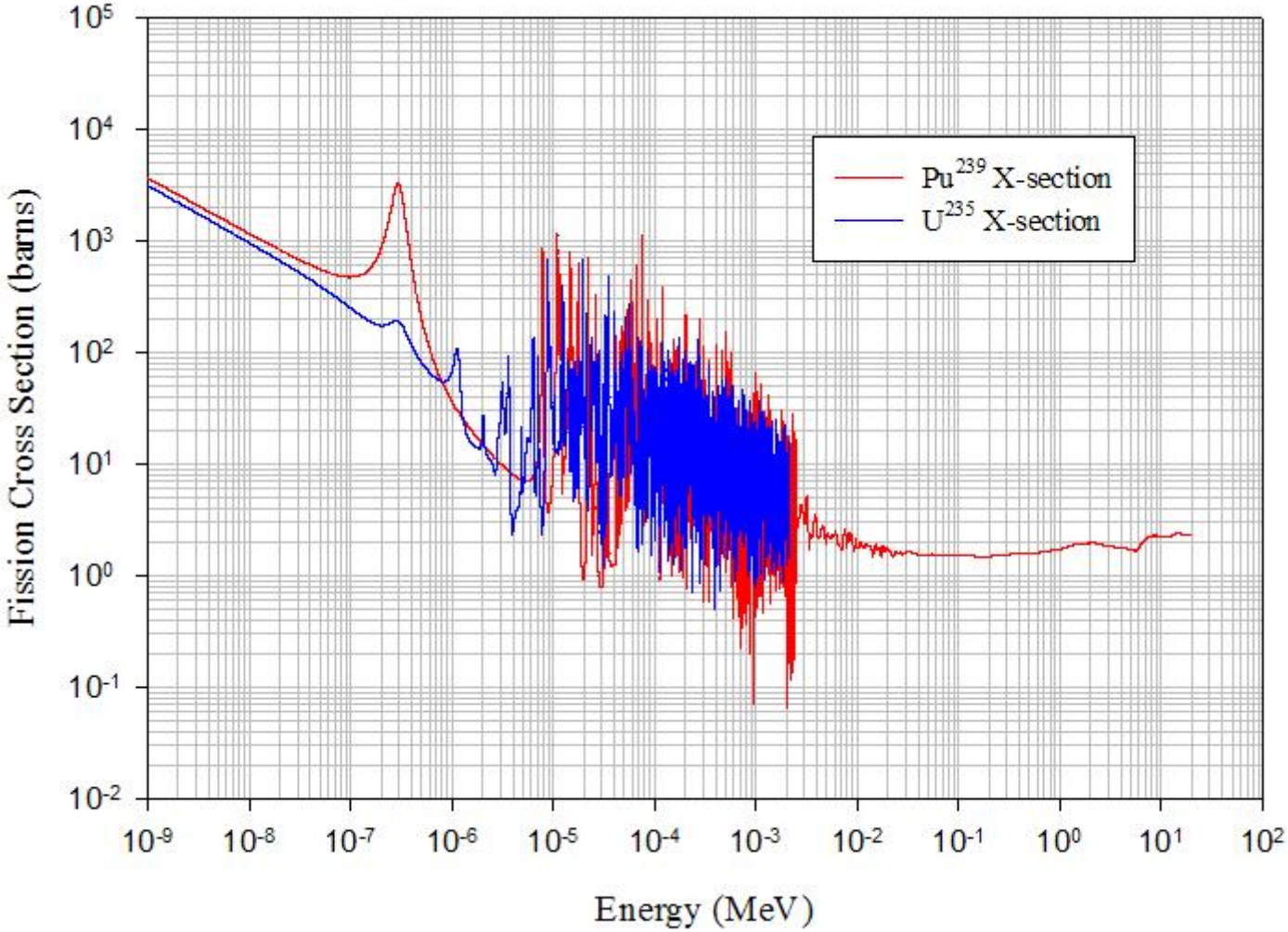
Neutron Slowing Down Spectrometer



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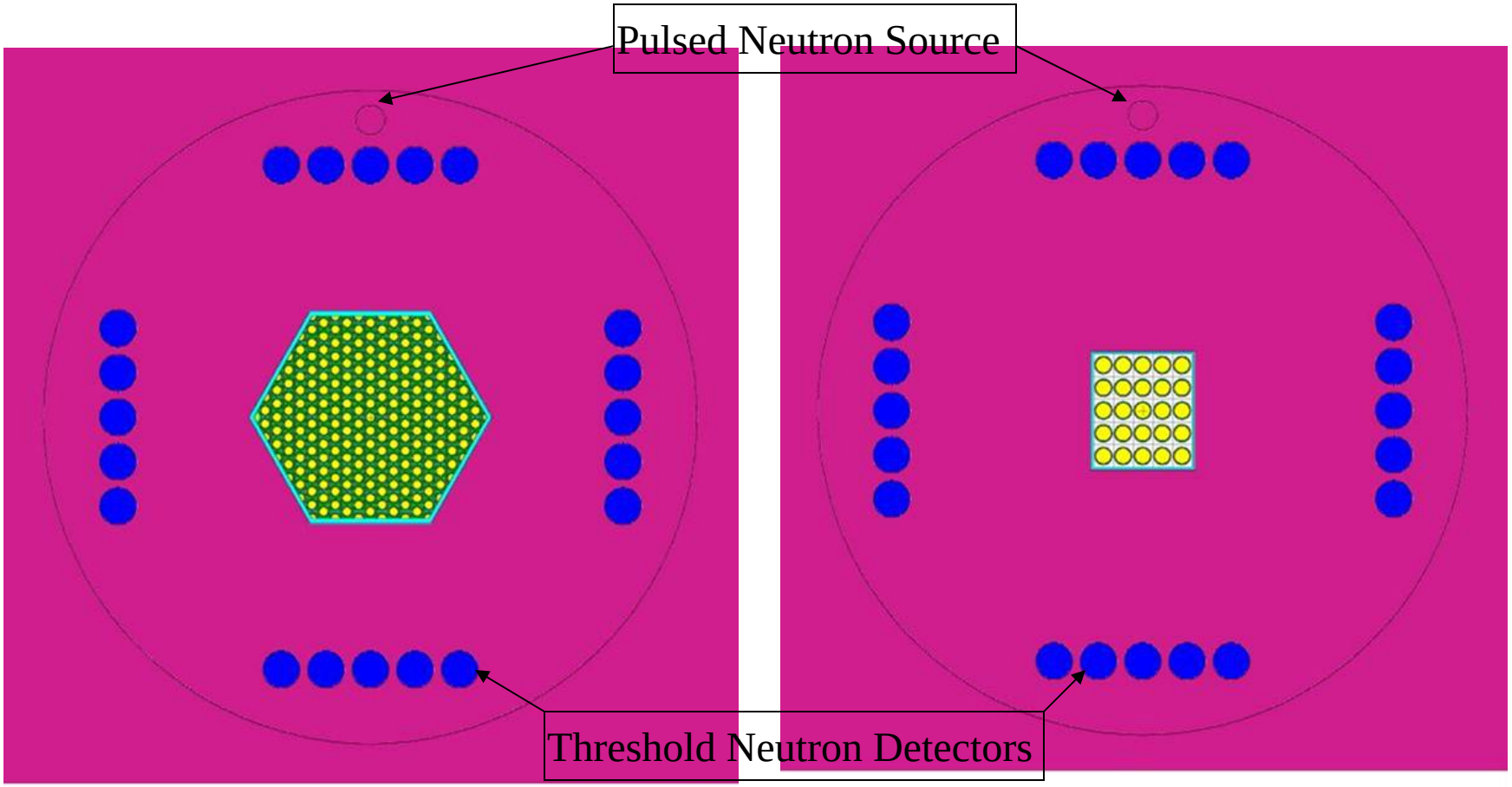
Neutron Slowing Down System



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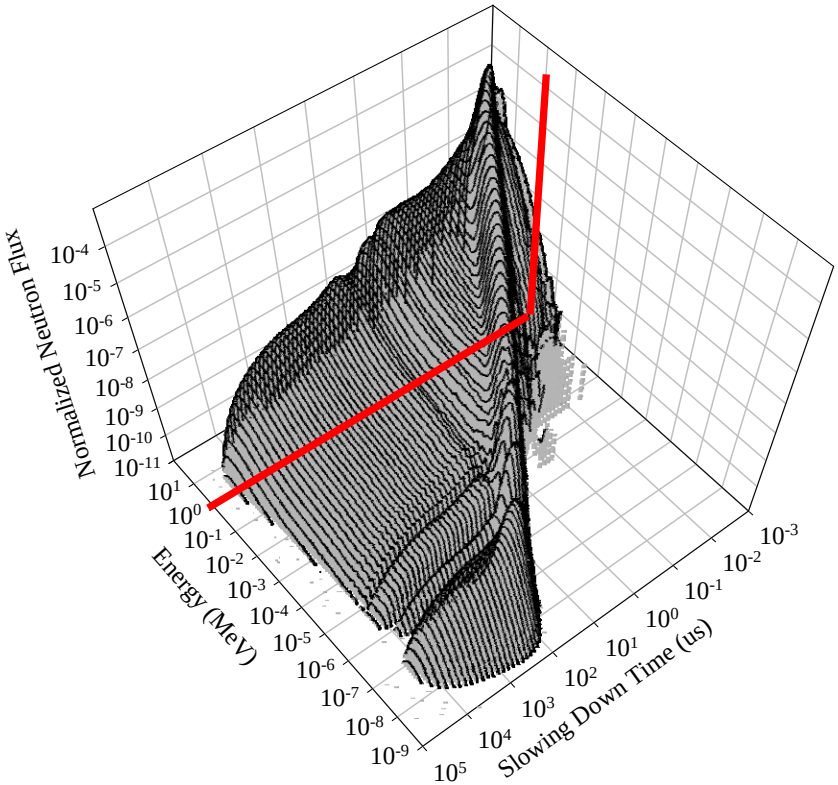
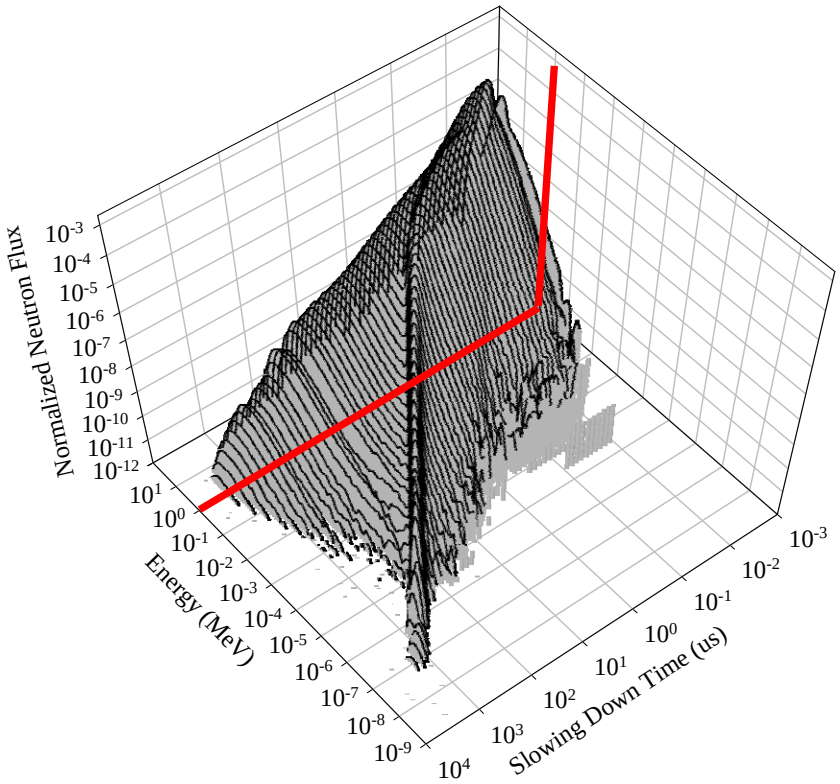
Heavy Metal Inventories of ABTR fuel at discharge burnup

Isotope	Mass (kg)	Isotope	Mass (kg)	Isotope	Mass (kg)	Isotope	Mass (kg)
U-234	0.01	Np-237	0.88	Pu-241	2.26	Am-243	0.20
U-235	0.84	Pu-238	0.56	Pu-242	1.03	Cm-242	0.04
U-236	0.14	Pu-239	121.80	Am-241	0.90	Cm-244	0.06
U-238	710.29	Pu-240	22.29	Am-242	0.05	Cm-245	0.01

Neutron Fluxes inside Fuel Assembly in Slowing Down Spectrometer





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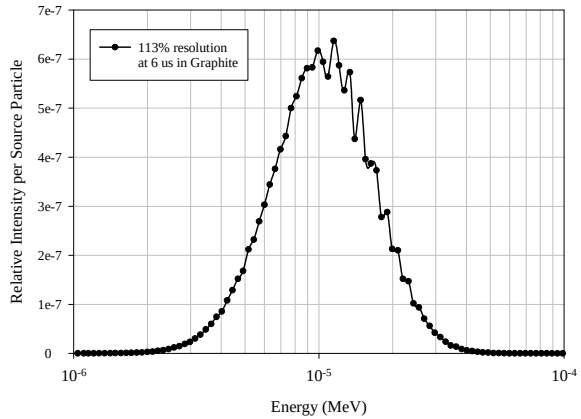
Neutron Slowing Down Pulse Resolution

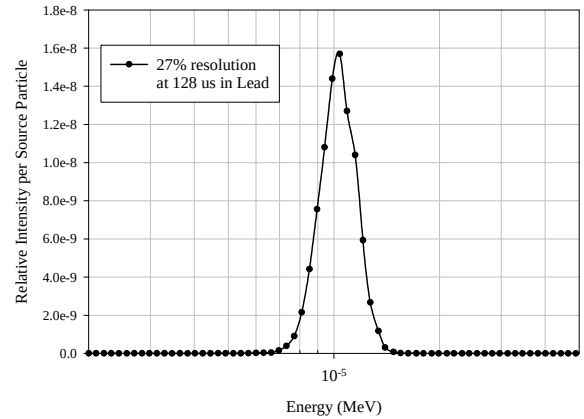
- Slowing Down Neutron Pulse resolution is determined by the atomic mass A of the slowing down medium:

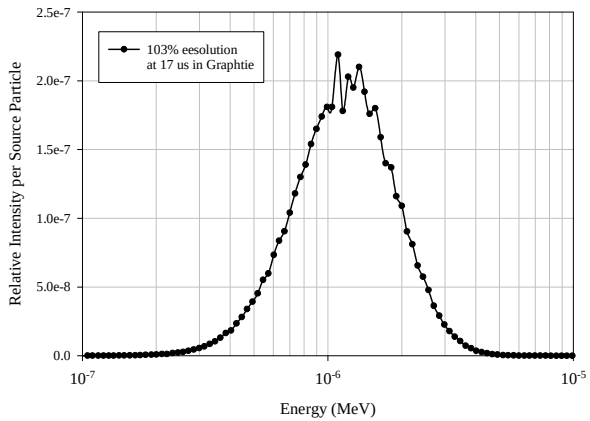
$$\frac{FWHM}{\overline{E}} \propto \sqrt{\frac{8}{3A}}$$

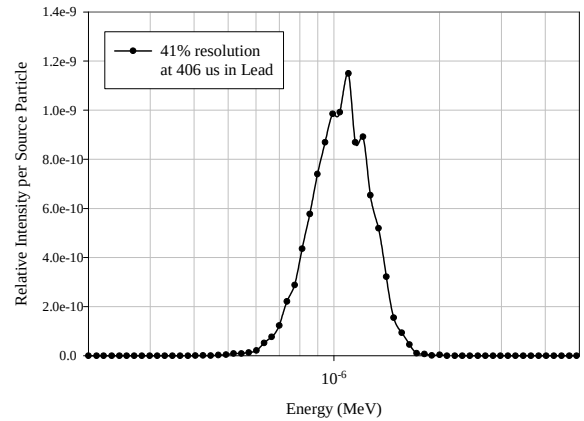
- For graphite: 103%
- For Lead: 27%

Neutron Slowing Down Pulse Resolution









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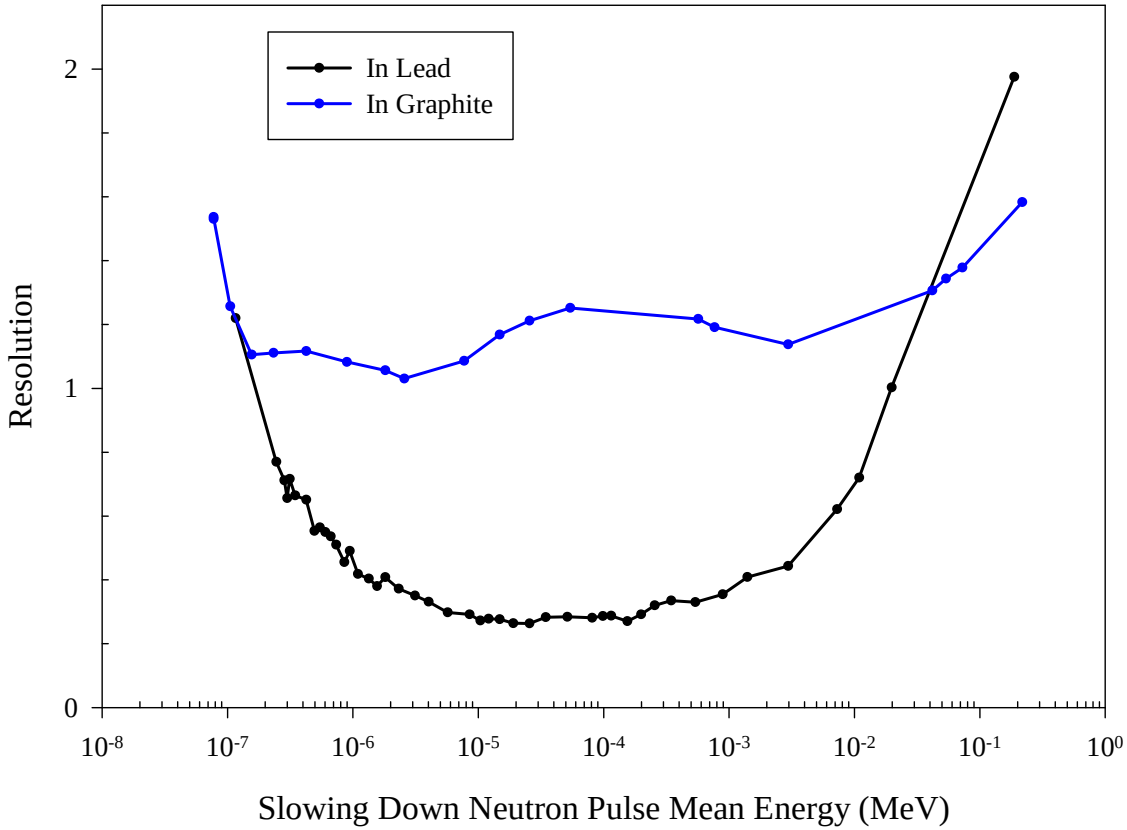
Neutron Slowing Down Pulse Resolution



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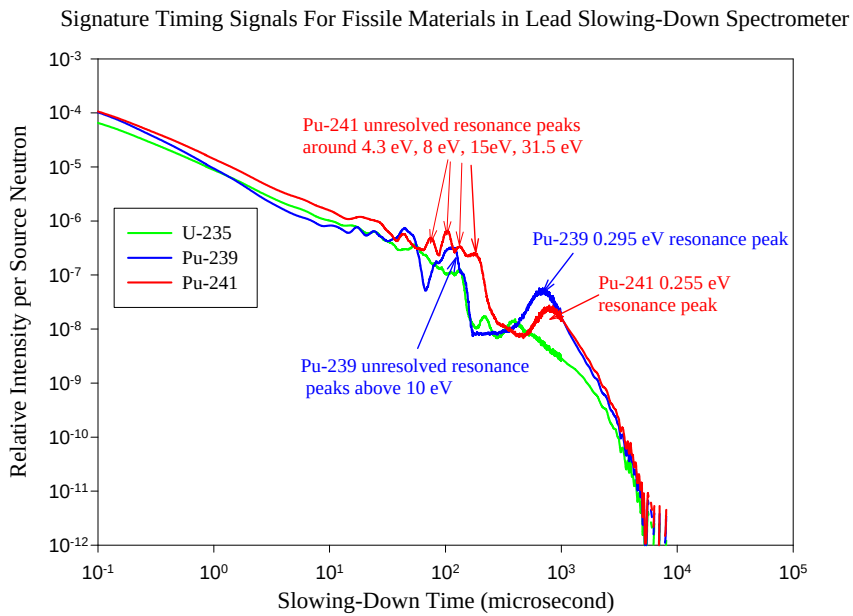
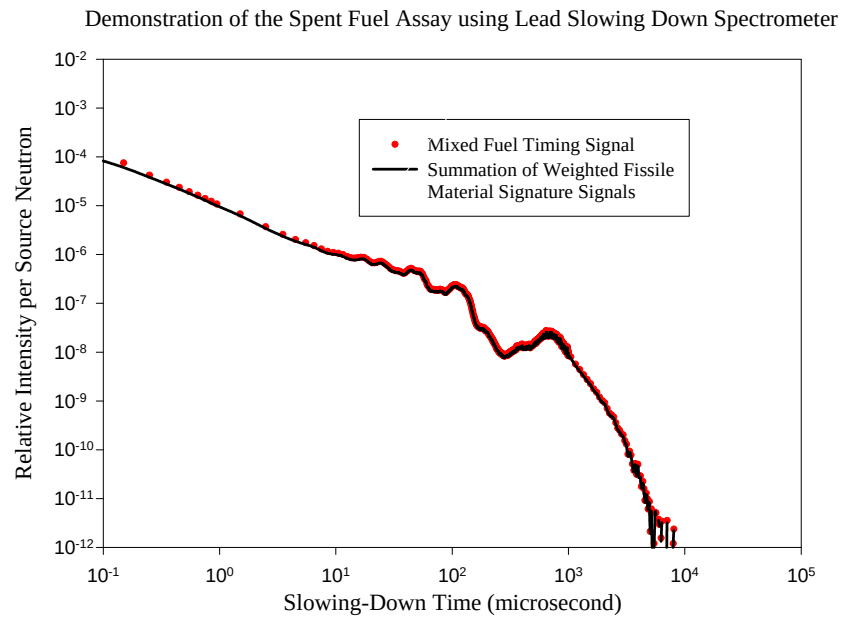
Neutron Slowing Down using Lead





This technique has been applied to LWR spent fuel assemblies, ~1% uncertainty has been claimed.

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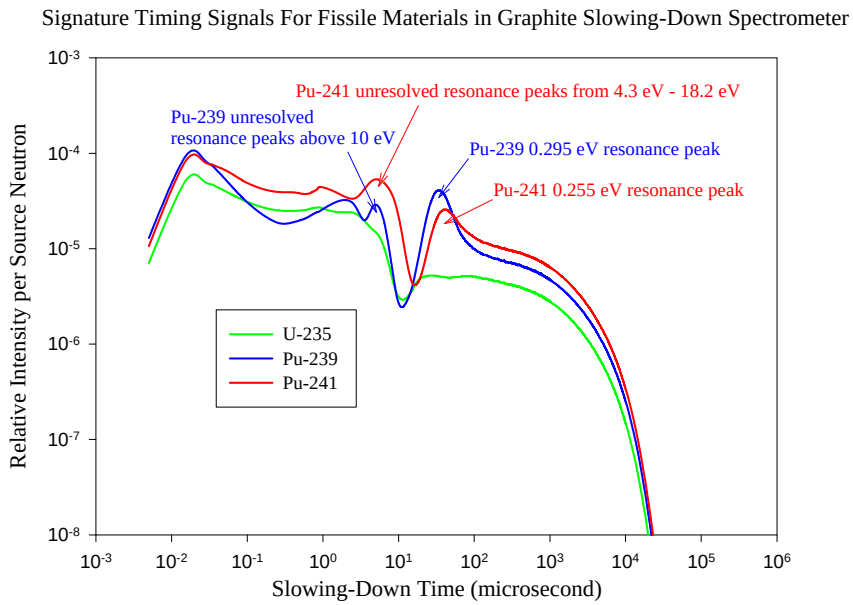
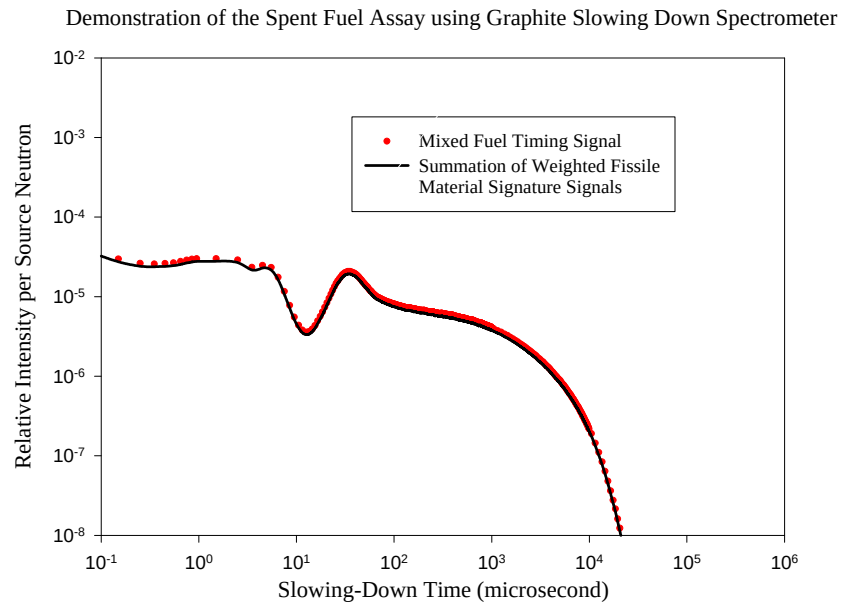
Neutron Slowing Down using Graphite





Using graphite SDTS has some challenge in the instrumentation and data acquisition system (too short slowing down time assay range), but it has some advantages such as lighter, easier to access, non-poisonous, most importantly, it has much higher neutron flux than lead slowing down spectrometer.

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Regression Analysis (N. Abdurrahman, 1993)

$$Y_i = \kappa\left[a_0 + a^{U235} X_i^{U235} + a^{Pu239} X_i^{Pu239} + a^{Pu241} X_i^{Pu241}\right]$$

κ = normalization constant

a_0 is the non-zero intercept, introduced to obtain better fitting, it is very close to zero compared to other parameters

$a_{fissile}$ is the atomic fraction of the corresponding fissile material, Yi is the tally in the *i*-th channel of the assay signal.

$Xi_{fissile}$ is the tally in the *i*-th channel of the corresponding fissile material signature signal.

The weight fraction of each fissile material can then be calculated as:

$$w^i = \frac{a^i A^i}{a^{U235} \cdot 235 + a^{Pu239} \cdot 239 + a^{Pu241} \cdot 241}$$

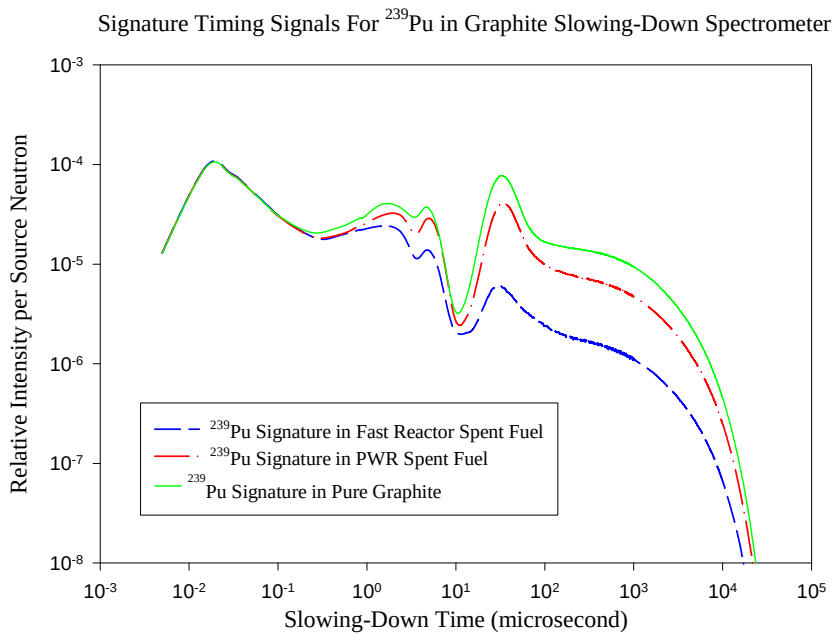
Fissile Contents Assay Results Using Least Squares Fitting Weighted by Inverse Variance

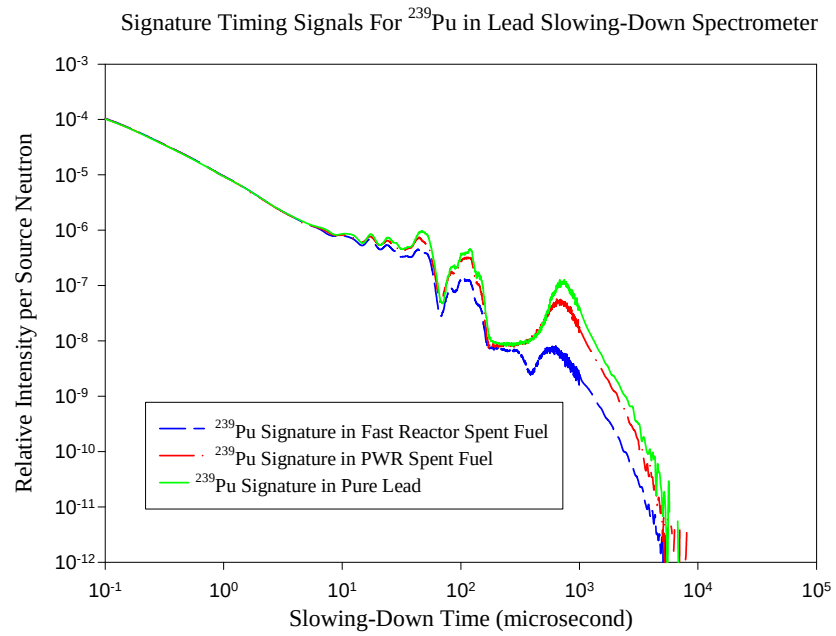
Energy (eV)	LSDTS (normalized weight fraction)			GSDTS (normalized weight fraction)		
	U-235	Pu-239	Pu-241	U-235	Pu-239	Pu-241
100 – 0.1	0.0082 ± 0.0013	0.9751 ± 0.0023	0.0186 ± 0.0002	0.0125 ± 0.0040	0.9701 ± 0.0030	0.0180 ± 0.0008
1 – 0.1	0.0173 ± 0.0124	0.9723 ± 0.0097	0.0172 ± 0.0266	0.0233 ± 0.1812	0.9721 ± 0.0095	0.0200 ± 0.0539
10 - 1	0.0082 ± 0.0043	0.9791 ± 0.0082	0.0185 ± 0.0002	0.0086 ± 0.0262	0.9697 ± 0.0105	0.0183 ± 0.0022
100 -10	0.0080 ± 0.0036	0.9743 ± 0.0030	0.0181 ± 0.0023	0.0156 ± 0.1270	0.9869 ± 0.0368	0.0130 ± 0.1022
True value	0.0067	0.9752	0.0181	0.0067	0.9752	0.0181

Self Shielding Effect

- The signature signals for Pu-239 and U-235 can be obtained by measuring the individual fission counting spectra of Pu-239 and U-235 fission chamber inside the NSDTS before start the fuel assay
- This measurement takes much longer counting time than usual fuel assembly assay measurement to obtain very small uncertainty in the signature spectra for fissile materials, which used to fit the measured spent fuel spectrum
- This measurement can be taken with fission chambers inside a spent fuel assembly to account for self-shielding effect from spent fuel.
- However, the other fuel assemblies could have different fissile material fractions, also, the location of the fission chambers inside the assembly could be different from the average self-shielding effect over all volume of assembly

Self Shielding Effect





Fast reactor spent fuel assay could be very different from PWR fuel assay due to self-shielding induced by high content of fissile materials. This phenomenon can be investigated using surface tally and volume tally in MCNP

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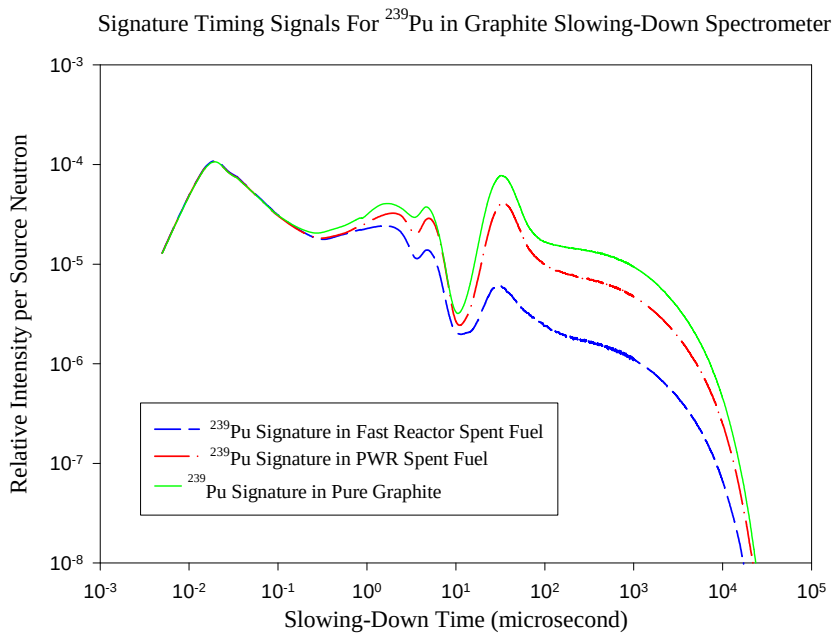
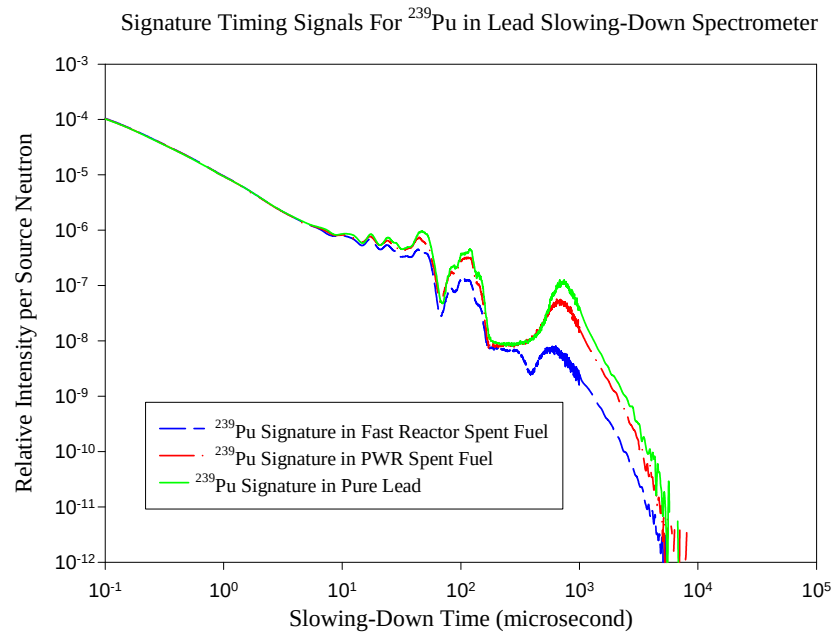
Self-Shielding Effect (PWR Fuel Pin, surface tally for signature spectra, volume tally for spent fuel spectrum)

Energy (eV)	LSDTS (normalized weight fraction)			GSDTS (normalized weight fraction)		
	U-235	Pu-239	Pu-241	U-235	Pu-239	Pu-241
100 – 0.1	0.5235 ± 0.0101	0.3924 ± 0.0089	0.0840 ± 0.0046	0.5393 ± 0.0124	0.3587 ± 0.0098	0.1019 ± 0.0067
1 – 0.1	-5.2913 ± 9.4349	8.4311 ± 13.2	-2.1398 ± 3.8523	0.0036 ± 1.2409	0.7756 ± 0.9677	0.2207 ± 0.2809
10 - 1	0.7583 ± 0.0486	0.1691 ± 0.0514	0.0725 ± 0.0080	0.4304 ± 0.1279	0.4463 ± 0.1012	0.1232 ± 0.0297
100 -10	0.5642 ± 0.0239	0.3593 ± 0.0207	0.0764 ± 0.0192	0.5305 ± 0.1111	0.3277 ± 0.0941	0.1417 ± 0.0766
True value	0.563	0.3529	0.084	0.563	0.3529	0.084

Self Shielding Effect





Which region is the best for the fitting?

- There should be a resonance peak to fit
- The largest peak might not be the best due to self-shielding

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Self Shielding Effect

- How do we find the best fitting region?
- A search algorithm is implemented as below:
  1. Start with the beginning channel as the first channel in the spectrum
  2. Ending channel of fit region to be beginning channel plus 5 channels
  3. For this region, perform LSF to obtain fissile materials fractions
  4. Calculate the sum square of the difference between true fissile material fractions and those obtained from regression analysis
  5. Then advance the ending channel by one, repeat step 3 and 4
  6. Once the ending channel reaches the last channel of the spectrum, beginning channel advance by one, and repeat step 2 – 5, until exhaust the whole spectrum
  7. The spectrum regions produce the least square difference of all three fissile material fractions are selected

Self-Shielding Effect (PWR Fuel Pin, surface tally for signature spectra, volume tally for spent fuel spectrum)

Energy (eV)	LSDTS (normalized weight fraction)			GSDTS (normalized weight fraction)		
	U-235	Pu-239	Pu-241	U-235	Pu-239	Pu-241
38.2 – 0.602 (66 – 554 us)	0.5631 ± 0.0033	0.3530 ± 0.0032	0.0838 ± 0.0009	N/A	N/A	N/A
3.46 – 0.245 (222 – 932 us)	0.5631 ± 0.0062	0.3528 ± 0.0050	0.0842 ± 0.0045	N/A	N/A	N/A
7.71 – 0.0546 (7 – 198 us)	N/A	N/A	N/A	0.5628 ± 0.0031	0.3517 ± 0.0025	0.0855 ± 0.0013
True value	0.563	0.3529	0.084	0.563	0.3529	0.084

Further investigation needs to be done to validate these region for physics meaning, and sensitivity to data variance, fissile contents variation, etc.

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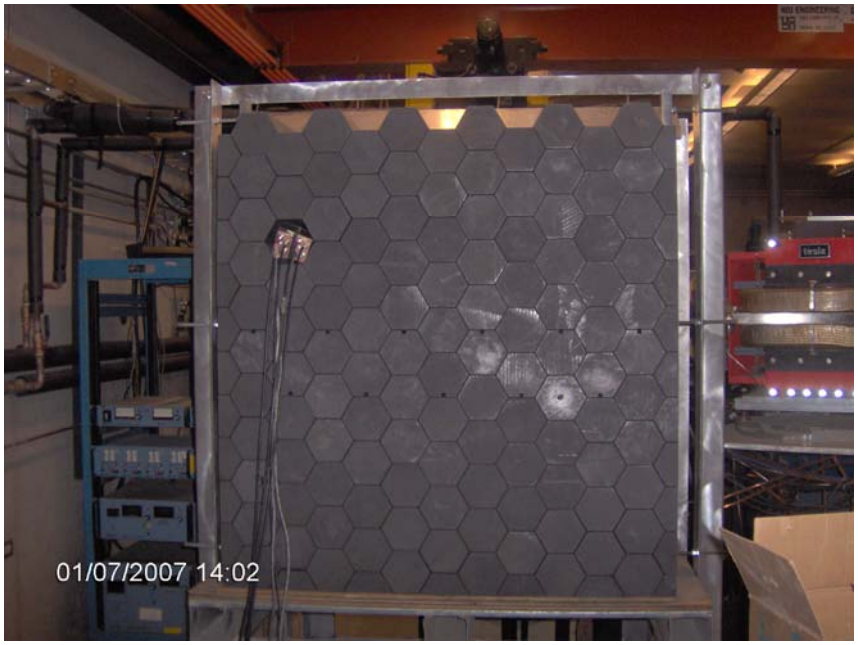
Preliminary Experiment Setup



Experiment Conditions:

Electron beam energy: 25 MeV

Pulse width: 2 ns

Peak beam current: 400 mA

Repetition rate: 60 Hz

Using U-235 fission chambers, DAQ triggered by each pulse, and extends to 200 μs

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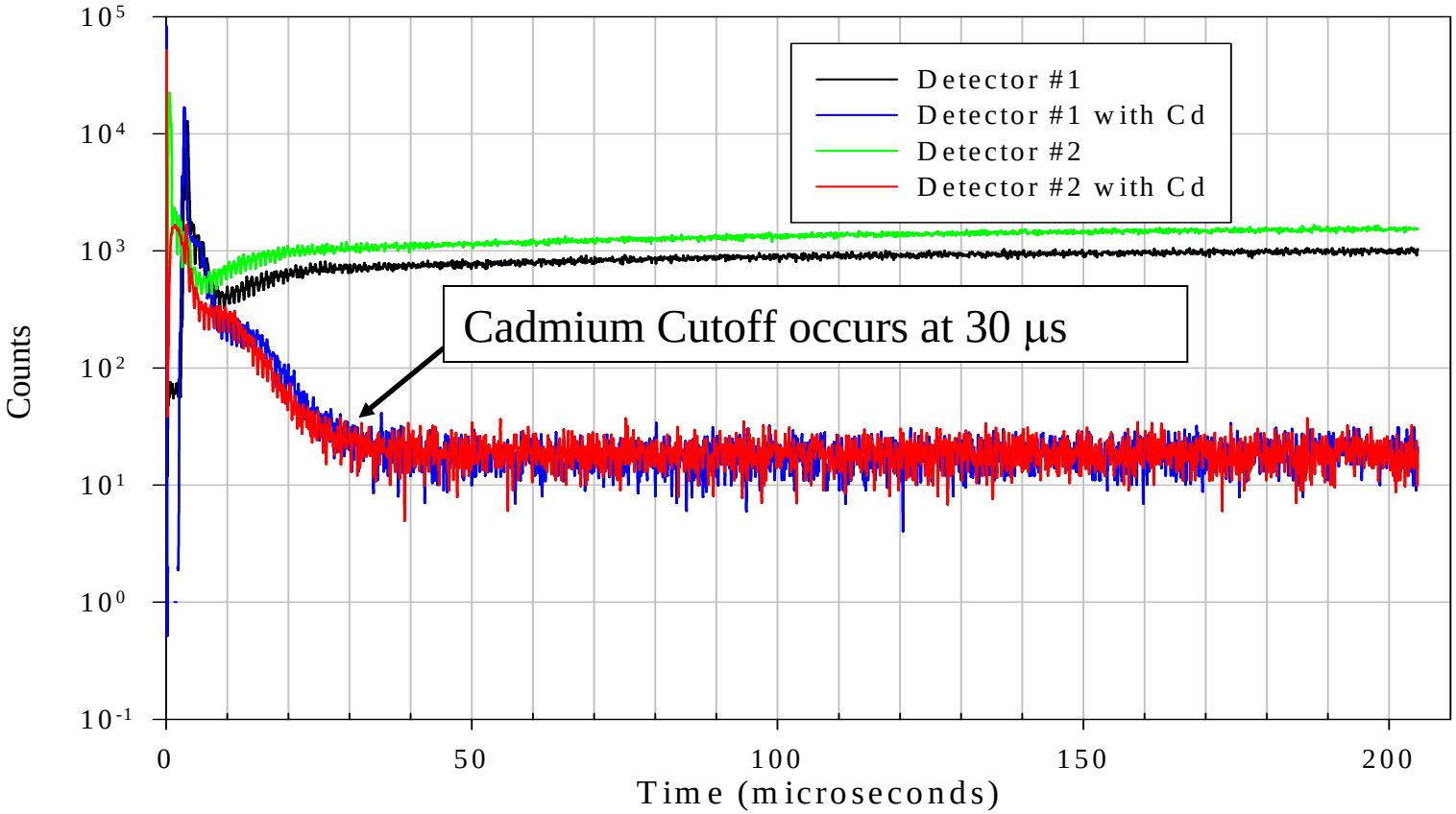
Preliminary Experiment Results



$$\overline{E} = \frac{k}{(t + t_0)^2}, k = 0.498\,\text{keV} * \mu s^2, t_0 = 0.8428\mu s \text{ for graphite}$$

MCNP simulation results show neutrons slow down to 0.5 eV 30 microseconds after the injection of neutron pulse

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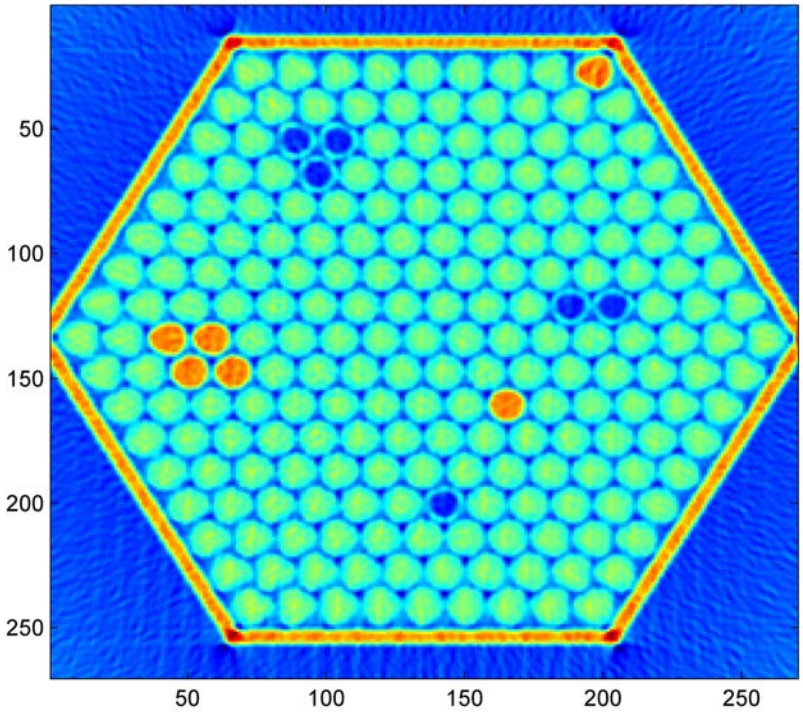
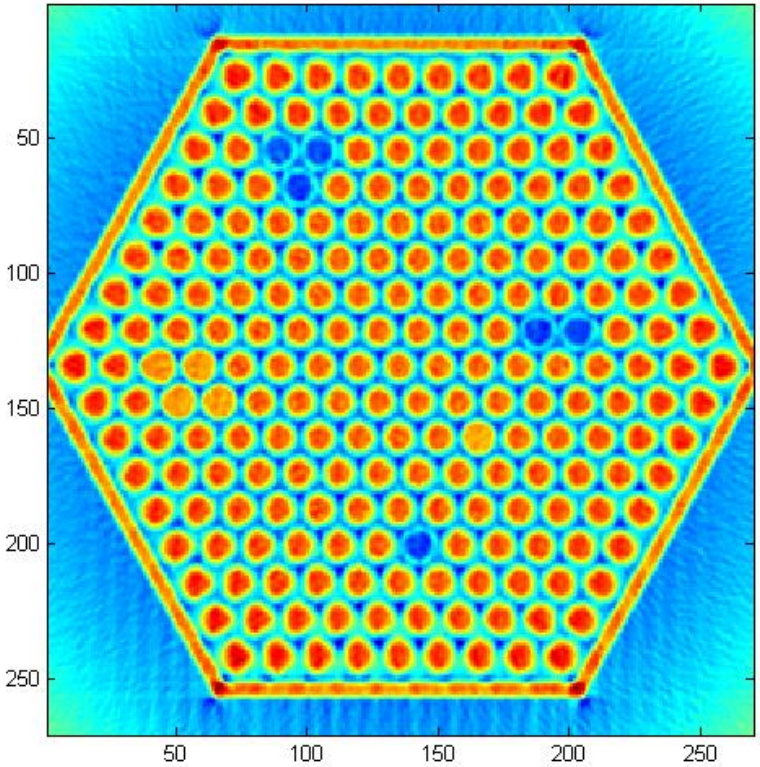
Future Work for NSDTS

- Pu-239 assay measurement
- MCNP calculations
  - Continue self-shielding effect investigation
  - System optimization
  - Assay procedure design

Future Work for NSDTS

Neutron tomographic fissile material assay





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