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Title: Nondestructive Assay for International Safeguards: Context, Science, and Technology

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Nondestructive Assay for International Safeguards

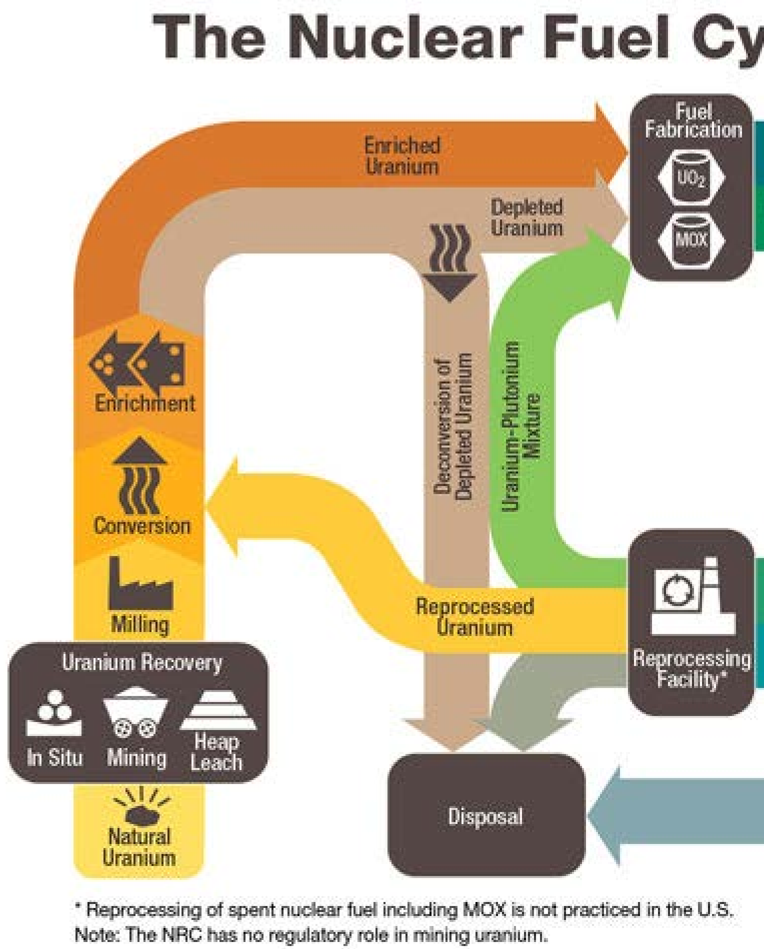
Context, Science, and Technology

Alexis Trahan, Ph.D.

October 23, 2017

International Safeguards

- "Delivering Effective Nuclear Verification for World Peace"
- The objective of safeguards is the timely detection of diversion of significant quantities of nuclear material from peaceful nuclear activities to the manufacture of nuclear weapons
- Inspect nuclear facilities worldwide, monitor amounts of nuclear materials to ensure that it isn't going to illicit uses



NRC.gov

The IAEA Today

Currently, the IAEA is working to achieve….

- **Universal acceptance of the AP**
- **Integrated safeguards**
  - Non-discriminatory and tailored to specific facility types
- **Safeguards-by-design**
  - Integrated within a facility's design, covering safeguards and security
- **Unattended monitoring & data integration**
  - Robust data management systems to reduce on-site inspector presence
- **State-level concept/approach**
  - Assessing each State as a whole

Source(s):
http://www.iaea.org/safeguards/statements-repository/overview.html
http://www.iaea.org/safeguards/documents/LongTerm_Strategic_Plan_%2820122023%29-Summary.pdf

AT THE END OF 2015:

200,110	Significant quantities* of nuclear material were under IAEA safeguards
1,286	Nuclear facilities and locations outside facilities were under IAEA safeguards
2,118	In-field inspections conducted
623	Design information verifications were conducted
64	Complementary accesses were conducted

182 States	IAEA Safeguard Agreements
128 States	Additional Protocols in force

IAEA (2016). IAEA Safeguards 2016: Serving Nuclear Non-Proliferation.

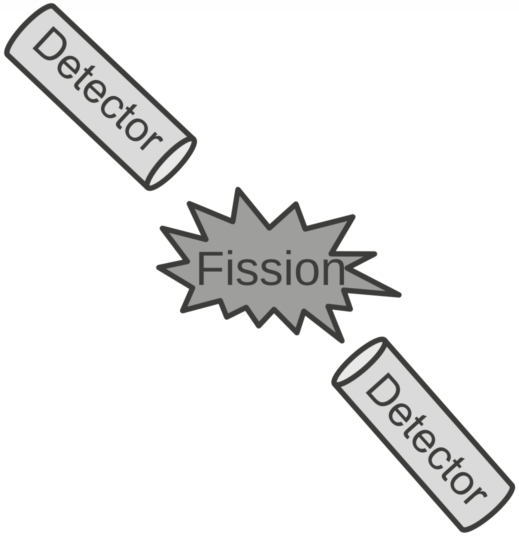
* One significant quantity is the approximate amount of nuclear material for which the possibility of manufacturing a nuclear explosive device cannot be excluded.

Special Nuclear Materials

- **What are we trying to verify?**
  – Special nuclear material (SNM) is where it should be, and in the proper amount

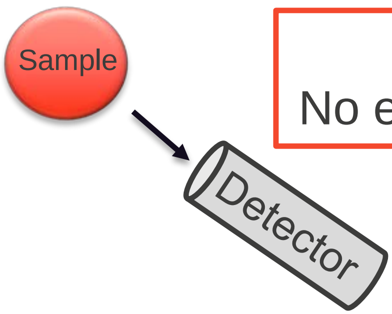

Weapons Grade Pu clad in Stainless Steel and surrounded by a Tungsten Reflector

- **SNM: Nuclear materials that can be used to make a weapon**
  – Highly Enriched Uranium (HEU)
    - Diversion path for HEU: enrichment facilities
    - Certain isotopes of U undergo fission primarily when induced with a neutron source
  – Weapons Grade Plutonium (Pu)
    - Diversion path for Pu: spent fuel (repositories, interim storage, reprocessing facilities)
    - Certain isotopes of Pu undergo fission spontaneously, without any prompting

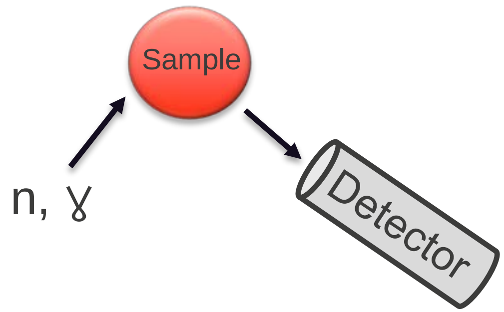


Nondestructive Assay (NDA)

- **NDA is the most commonly employed technique for material accountancy**
- **A series of gamma or neutron detectors are typically used to measure radiation emitted from the sample of interest**
- **Energy, timing, and intensity of radiation may be correlated to isotope type and quantity in the sample**



Passive:
No external source



Active:
Neutrons or gammas irradiate source to magnify signal

- **Passive interrogation requires good signal intrinsic to sample (^{240}Pu, ^{252}Cf)**
- **Active interrogation requires fissile material or material prime for gamma interactions (^{235}U, ^{239}Pu)**

Rail radiation portal monitor (RPM) at the Port of Antwerp, Belgium

Neutrons and Photons

	Neutrons	Photons
Origins	– Spontaneous and induced fission – (α,n) – Cosmic rays – (p,n) – (n,2n) – (γ,n) (Cosmic rays, (p,n), (n,2n), (γ,n): Less common)	– Nucleus (gamma-ray) – Nuclear collision (gamma-ray) – Electron cloud (x-ray)
Signal	**Time and correlations**	**Energy**
Shielding	**Low Z material**	**High Z material**
Detectors	**^{3}He, Scintillators, fission chambers**	**HPGe, Scintillators, NaI**

Neutrons

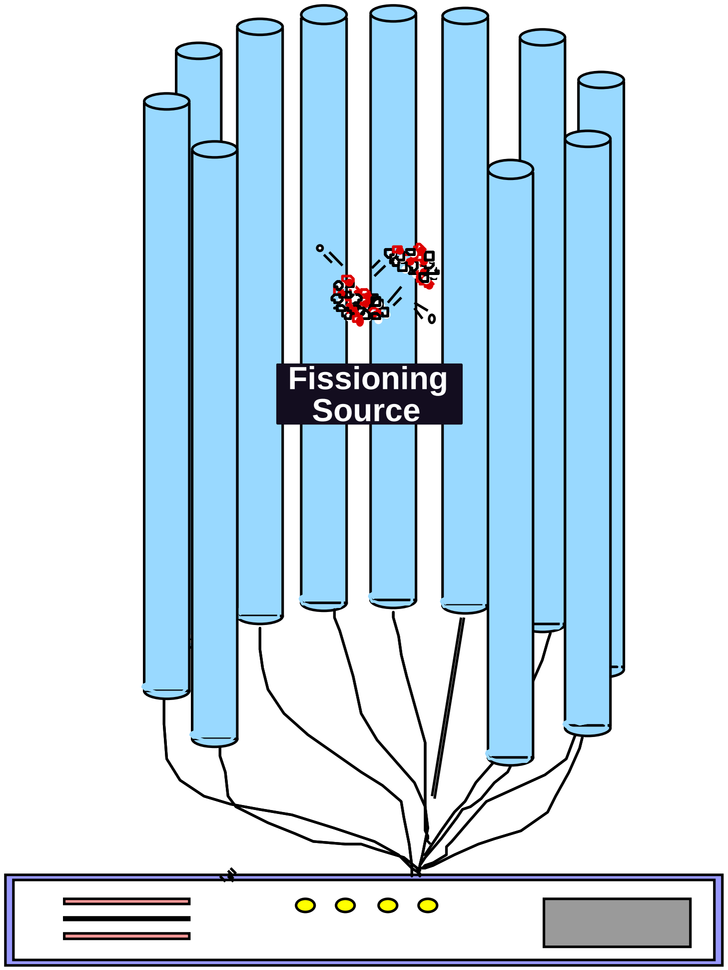
History of Neutron Counting

- **TOTAL NEUTRON**
  - Record the total number of neutrons detected in a certain amount of time
  - Accurate assays can be obtained only for very few types of SNM

- **COINCIDENCE COUNTING**
  - Record the number of times two neutrons arrive within a set time window (gate)
  - Wide application for international safeguards
    - focused on verifying declared materials

- **NEUTRON MULTIPLICITY COUNTING**
  - Extension of neutron coincidence counting
  - Record the number of times we detect 2, 3, 4, etc. neutrons within a gate
  - It improves neutron assay accuracy dramatically by adding more measured information

Passive Neutron Counter



Pulse-processing Electronics

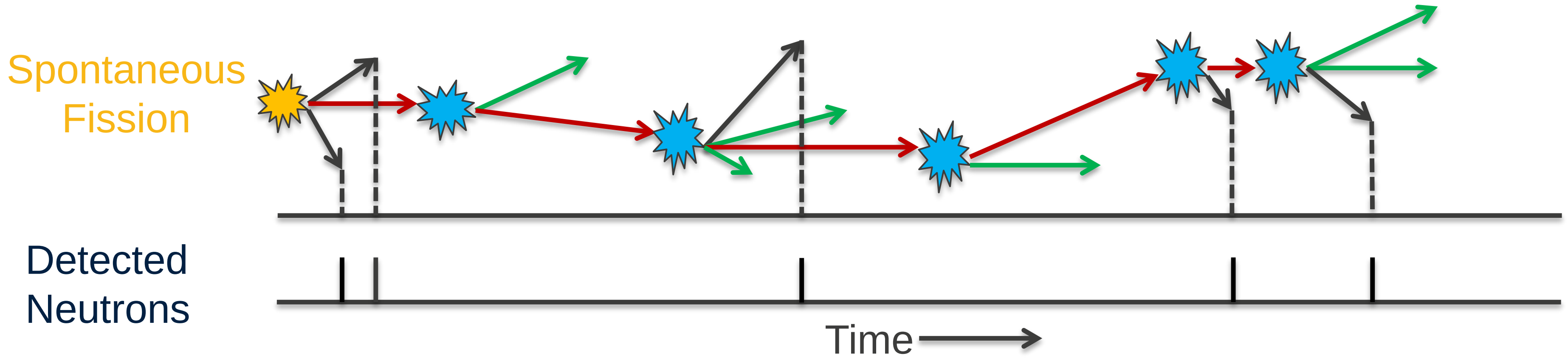
^{3}He neutron detectors

Fissioning source surrounded by neutron detectors

Emission of multiple **prompt** neutrons from fission detected as coincident neutron events

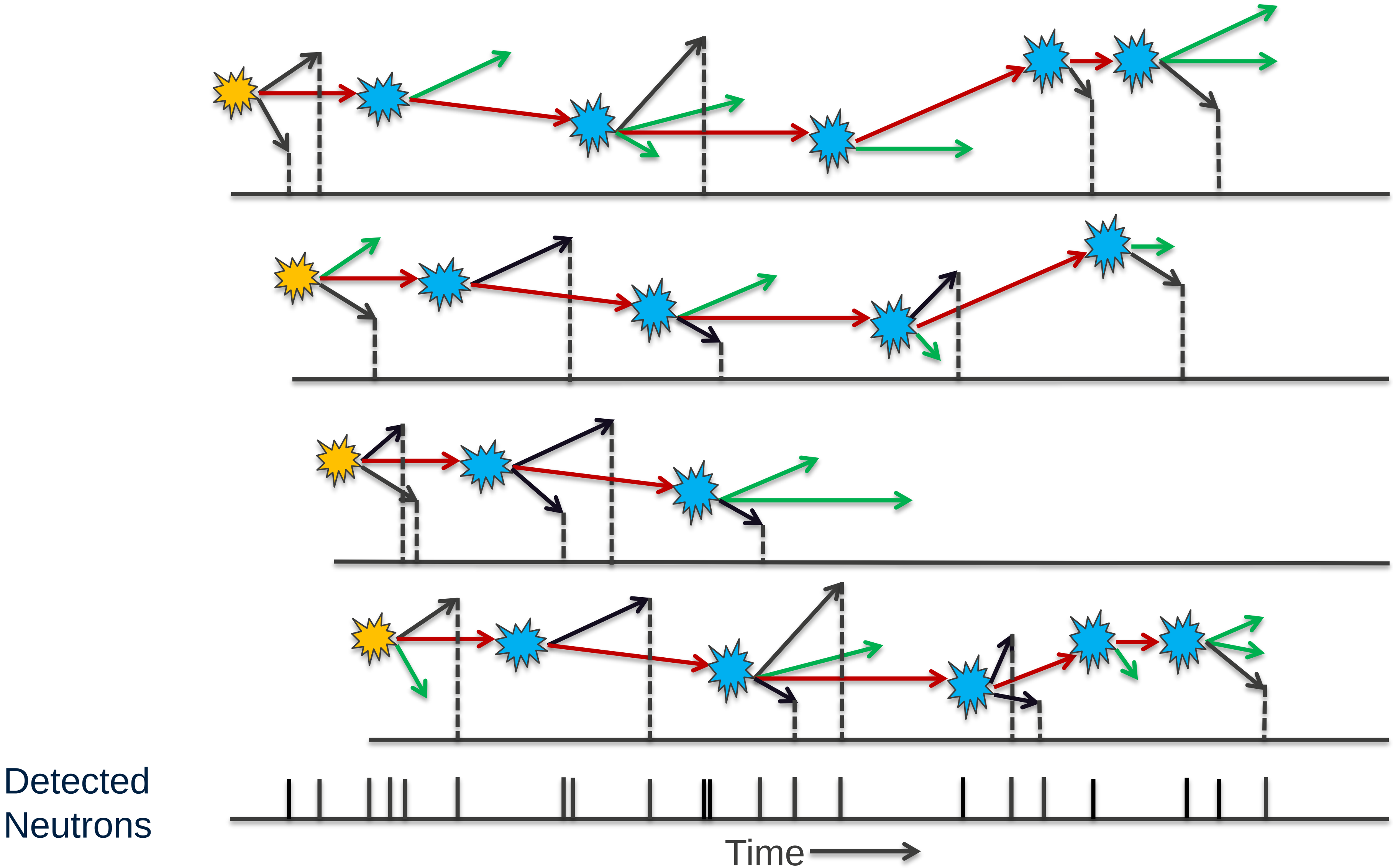
Multiplicity information is used to calculate the mass of fissioning isotopes

Neutron Coincidence Counting

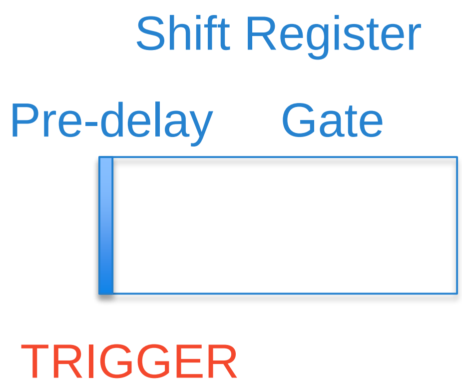


- As neutrons are detected, they trigger the shift register and neutron coincidence counting is performed

Neutron Coincidence Counting



Neutron Coincidence Counting

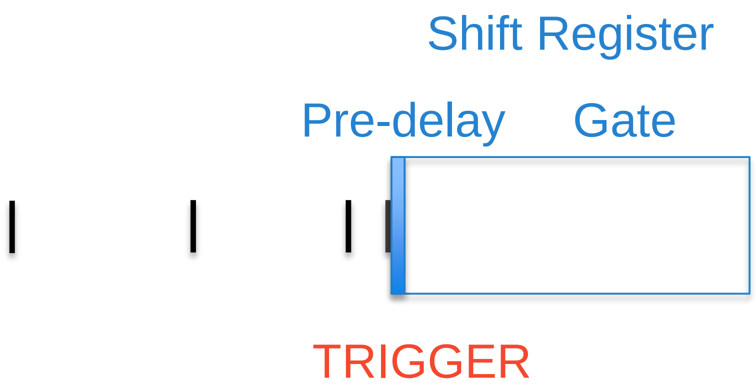


Singles: |

Doubles:

- **First neutron detection triggers the shift register**

Neutron Coincidence Counting

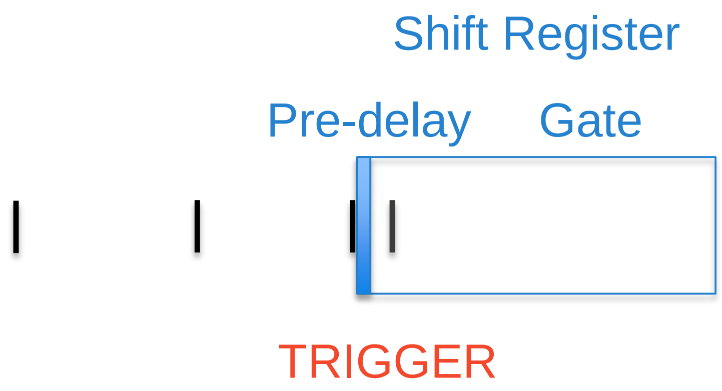


Singles: ||

Doubles: |

- **First neutron detection triggers the shift register**
- **Next neutron detection triggers the shift register again; this time, one neutron is already in the shift register, so we have one coincidence**

Neutron Coincidence Counting



Singles:

Doubles:

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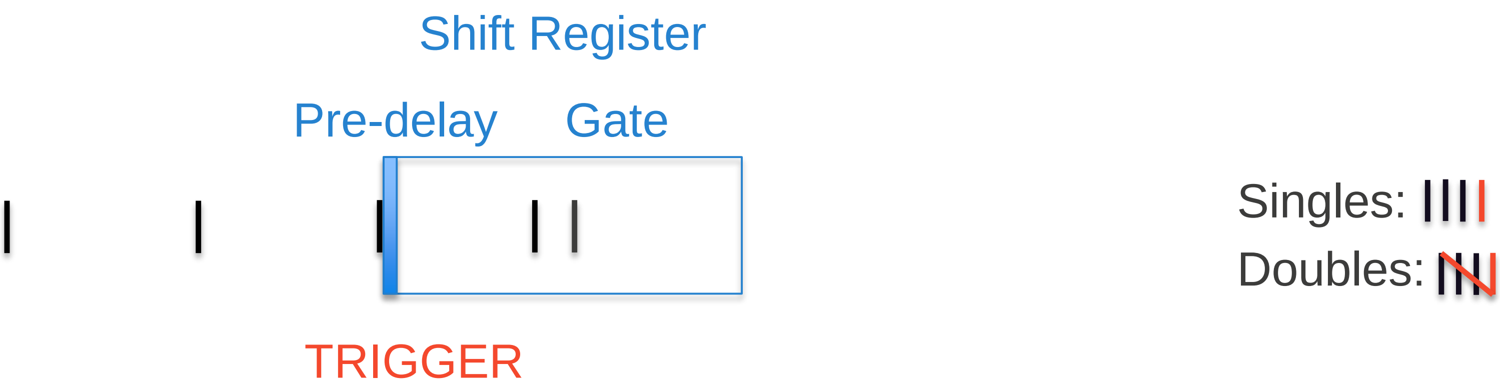
- **Next neutron detection triggers the shift register again; this time, two neutrons are already in the shift register, so we have two coincidences**

Neutron Coincidence Counting



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- **Next neutron detection triggers the shift register again; this time, two neutrons are already in the shift register, so we have two coincidences**
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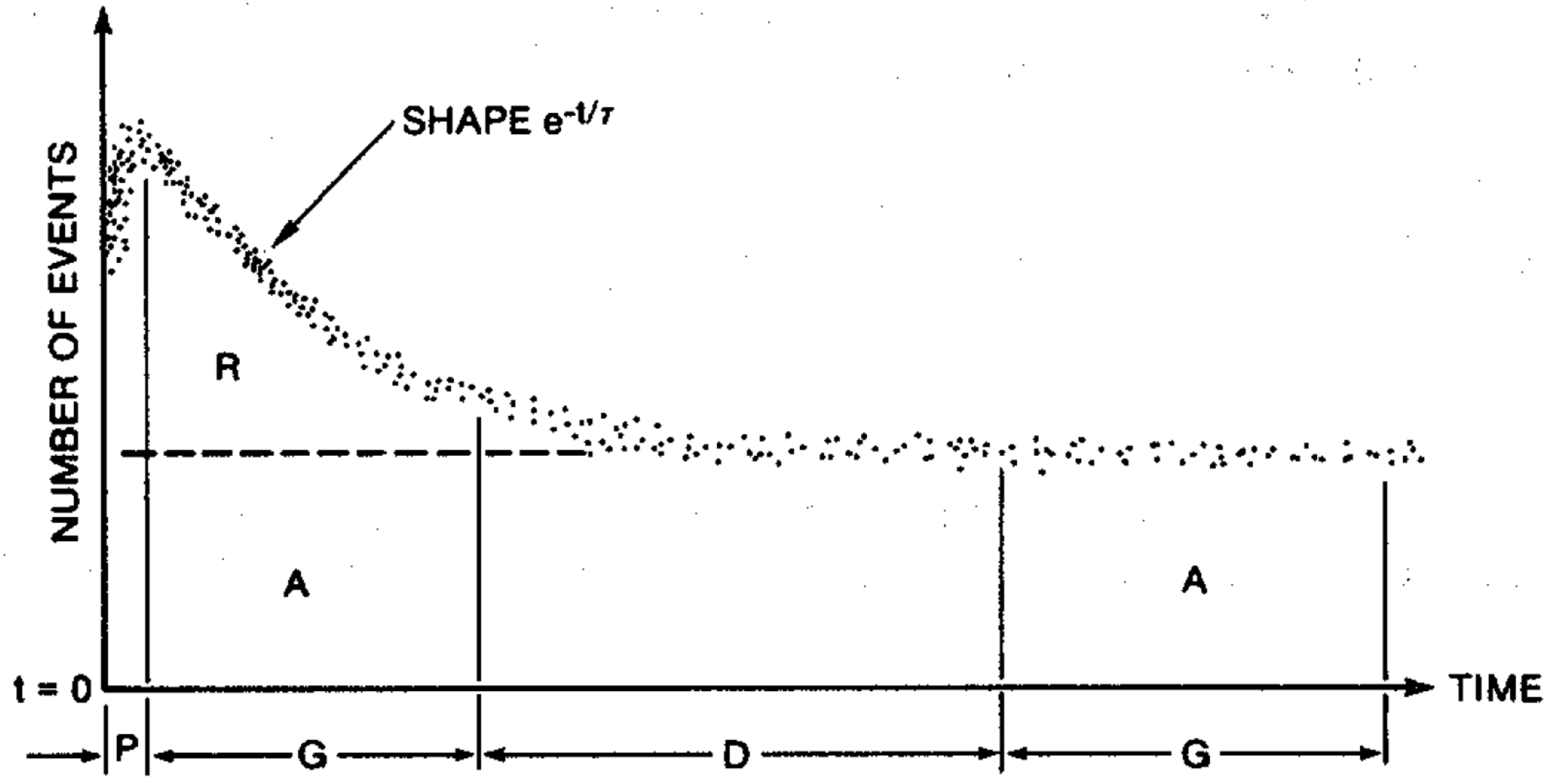
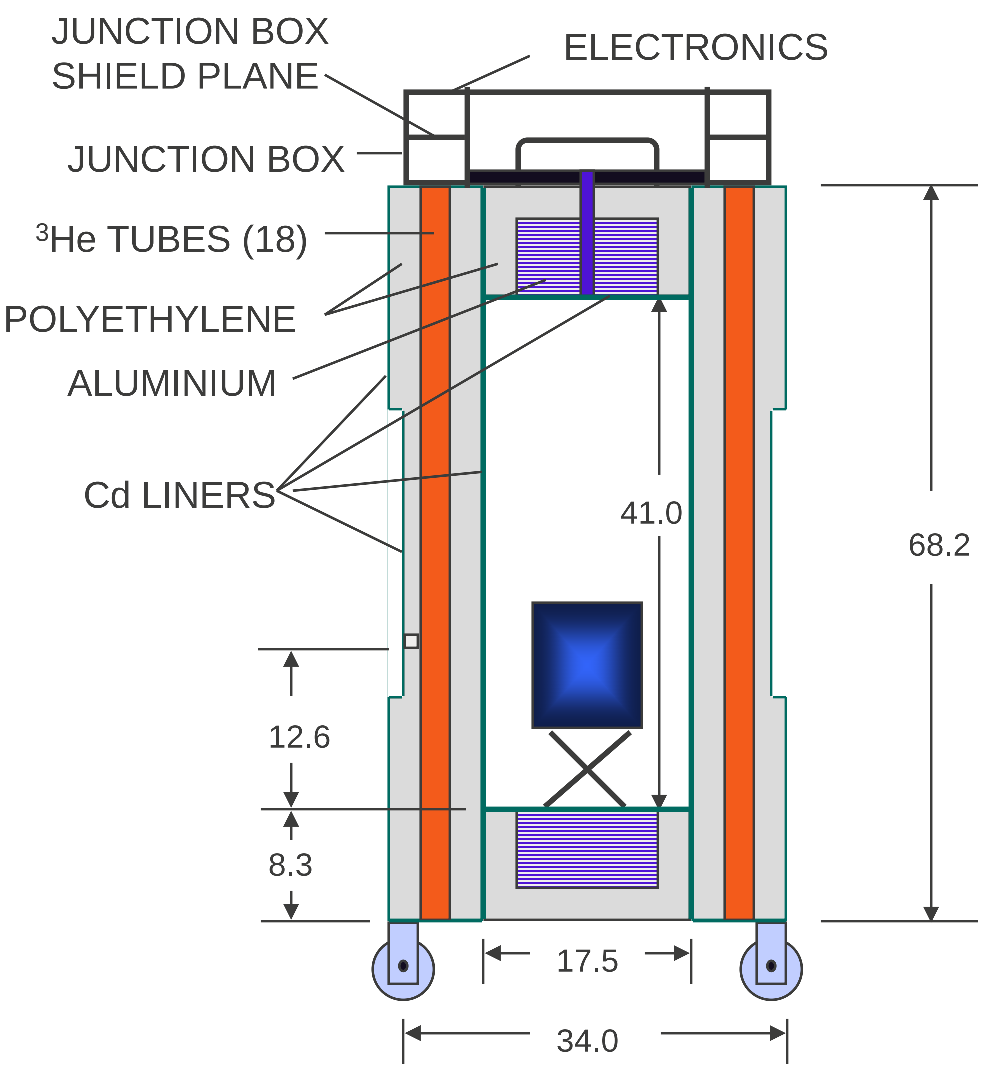
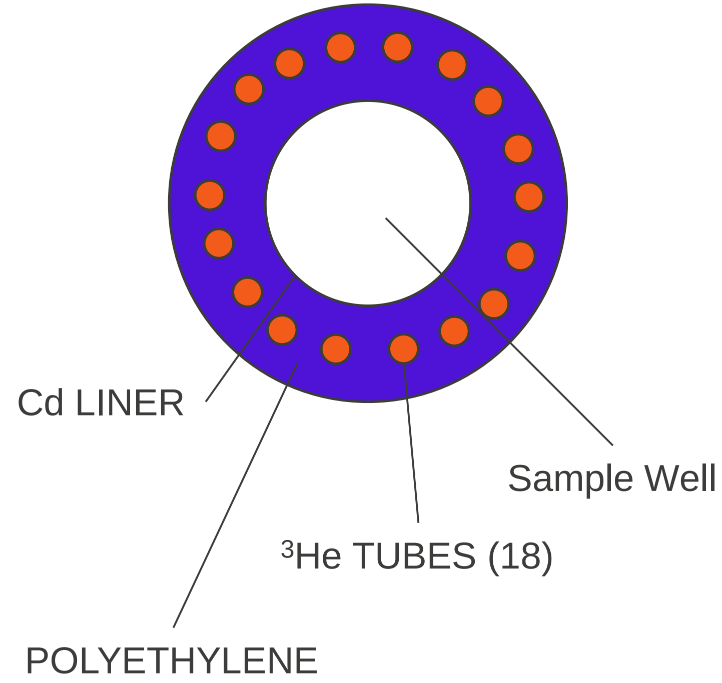
Rossi-Alpha Distribution



High-Level Neutron Coincidence Counter (HLNC)

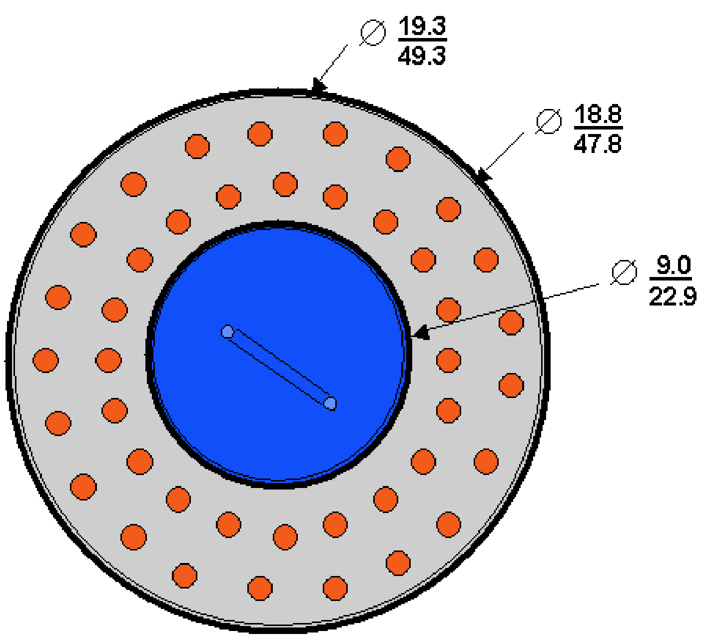


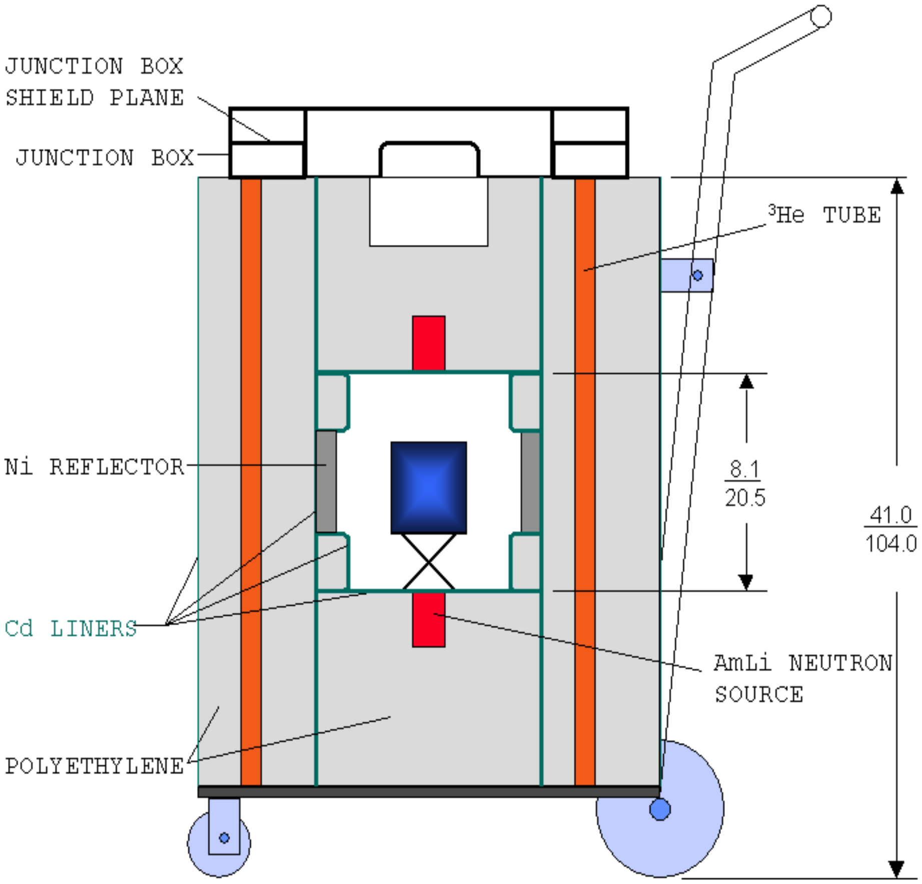
- ε = 17.5%
- τ = 43 μs
- 18 detector tubes: 4-atm ^{3}He, 50.8-cm active-length, ϕ2.54cm
- 6 Amptek preamplifiers



Active Well Coincidence Counter (AWCC)

- $\varepsilon = 33\%$
- $\tau = 51\ \mu\text{sec}$
- 42 tubes: 4-atm ^{3}He, 50.8-cm active-length, ϕ2.54cm
- 6 Amptek preamplifiers





Photons

Photons

- **Generic Assay Equation**

$$M_{SNM} = \frac{R_{Rad} \times CF}{Cal}$$

M_{SNM} = Mass of special nuclear material
R_{Rad} = Measured radiation rate (counts per unit time) from SNM item
CF = Correction for losses due to:

- **item self absorption**
- **container absorption**
- **measurement system electronics**

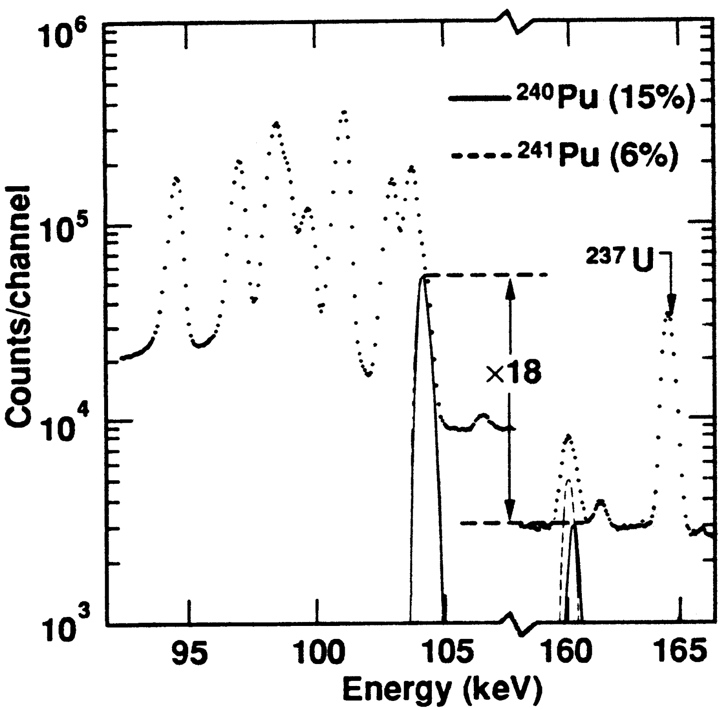
Cal = Calibration constant

MGA

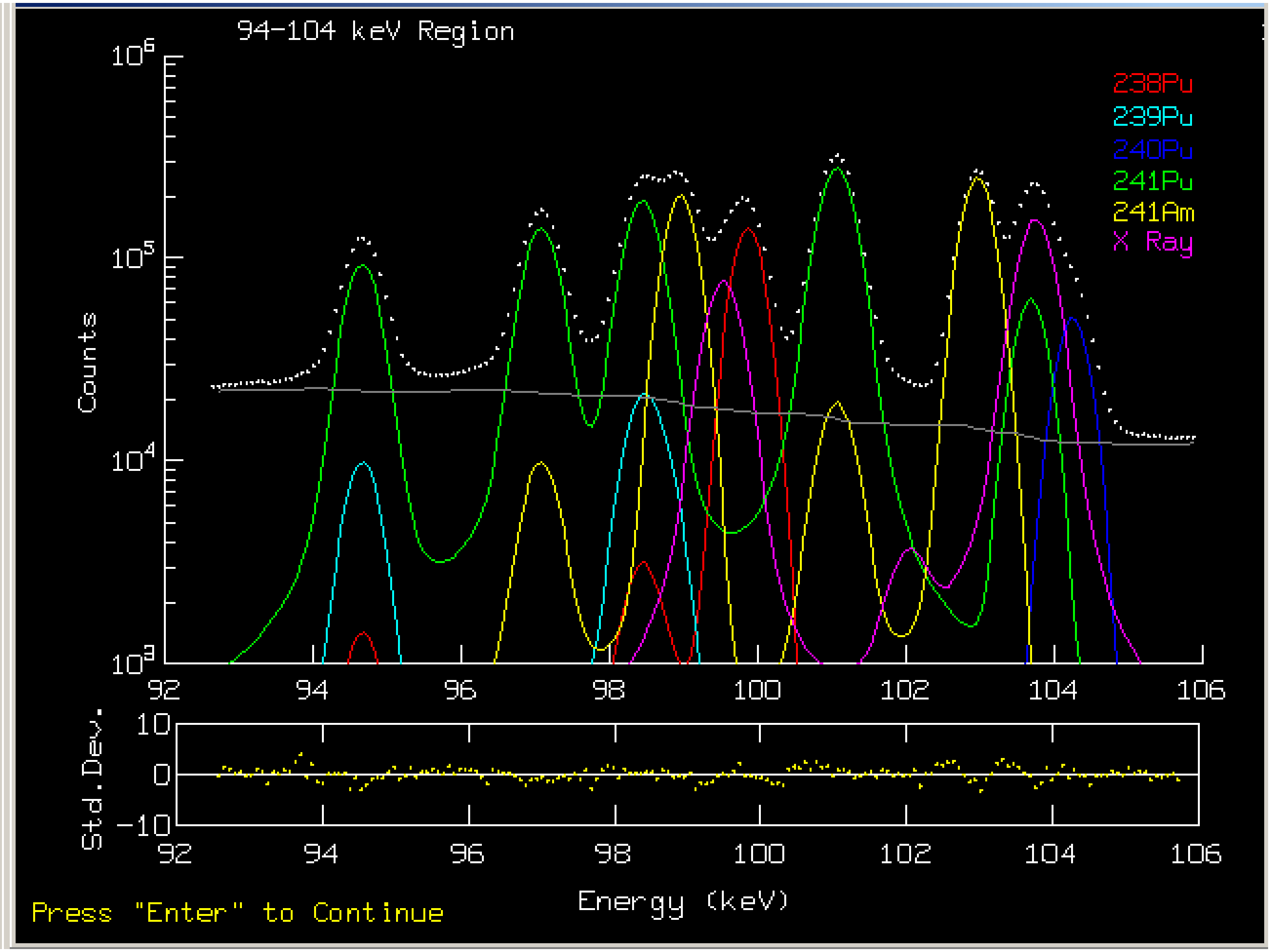
- Multi-Group Analysis of 100 keV x-ray and gamma-ray region
- Plutonium isotopics determination
- Uses multiple peaks to create an efficiency curve, eliminating the need for a separate efficiency measurement
- High-resolution Ge detector needed

^{240}Pu Peak Intensity:

Compare the intensity of the peak at 104 keV with that of the one at 160 keV



MGA



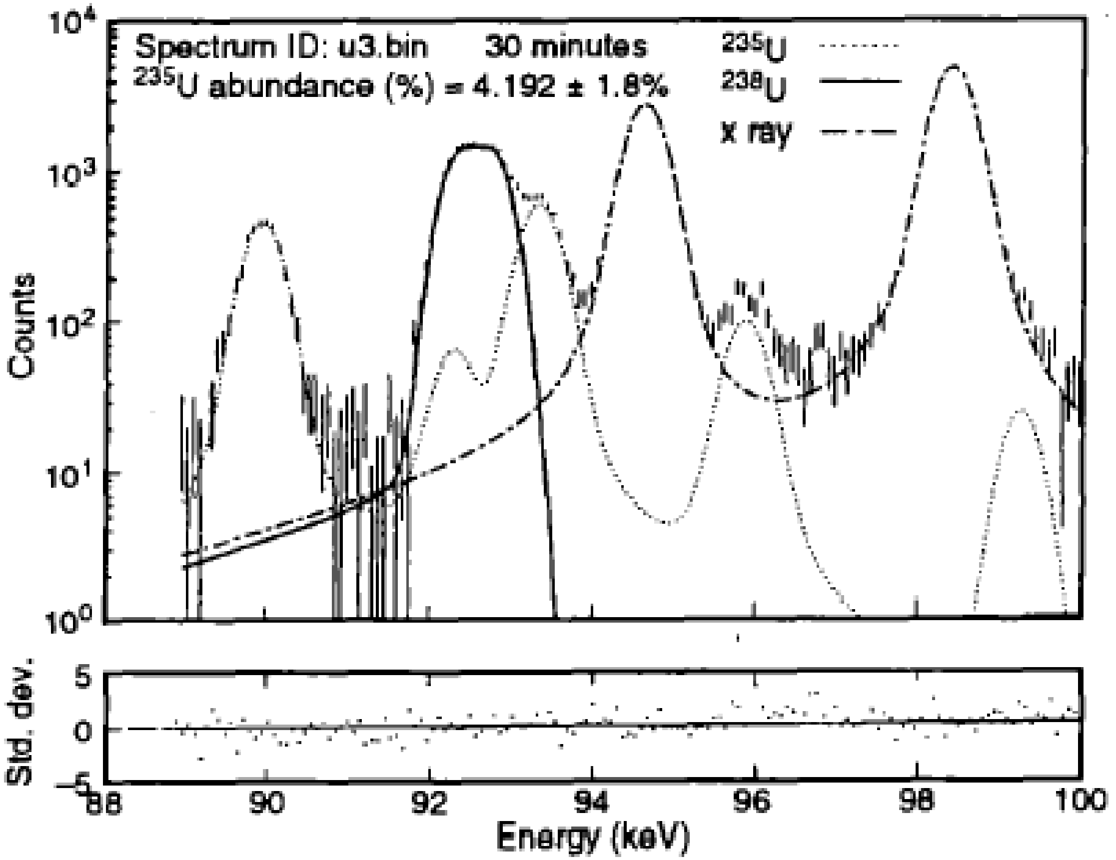
94-104 keV Region
238Pu
239Pu
240Pu
241Pu
241Am
X Ray
Counts
Std. Dev.
Press "Enter" to Continue
Energy (keV)

MGA

```
Report generated  1-Feb-2013 09:18:33
Spec. ID: CBNM61L.CNF                                   LT:  38.9 Mins  DT:44%
                   Measurement Date: 8-Sep-1992  Declared Date: 8-Sep-1992
Sample ID:

PU g/cm2 = 2.7056   CD thickness = 1.35 mm        FWHM at 122 keV = 531 eV
QFIT = 1.60         Gain = 75.154 eV/ch                at 208 keV = 663 eV
NQFIT= 1.007                      ISOTOPIC ANALYSIS AT
        RELATIVE    %    %*      MEAS. DATE      DECLARED DATE  SPECIFIC POWER
        ABUNDANCE ERR   ERR     WT.PCT.  %ERR     WT.PCT.  %ERR (MILLIWATTS/GM)
Pu238 = 0.018208  0.2   0.7     1.16741  0.65     1.16741  0.65   6.62589
Pu239 = 1.000000  0.6   0.0    64.11562  0.51    64.11562  0.51   1.23666
Pu240 = 0.407647  0.5   0.8    26.13654  0.69    26.13654  0.69   1.85109
Pu241 = 0.078905  0.1   0.7     5.05902  0.62     5.05902  0.62   0.17261
Pu242 = (Default Algorithm)     3.5214   (10)     3.5214   (10)   0.00408
Am241 = 0.050407  0.2   0.6     3.23185  0.61     3.23185  0.61   3.69077
                      *=Error in Ratio (1 Sigma Error) TOTAL= 13.581 +/-0.51%
241Am Separated About 10.227 +/-0.069 Years before Measurement.

Pu-240 effective = 34.99 (at meas. date) 34.99(at Decl. date) +/-  1.83%

Press "Enter" to Continue
```

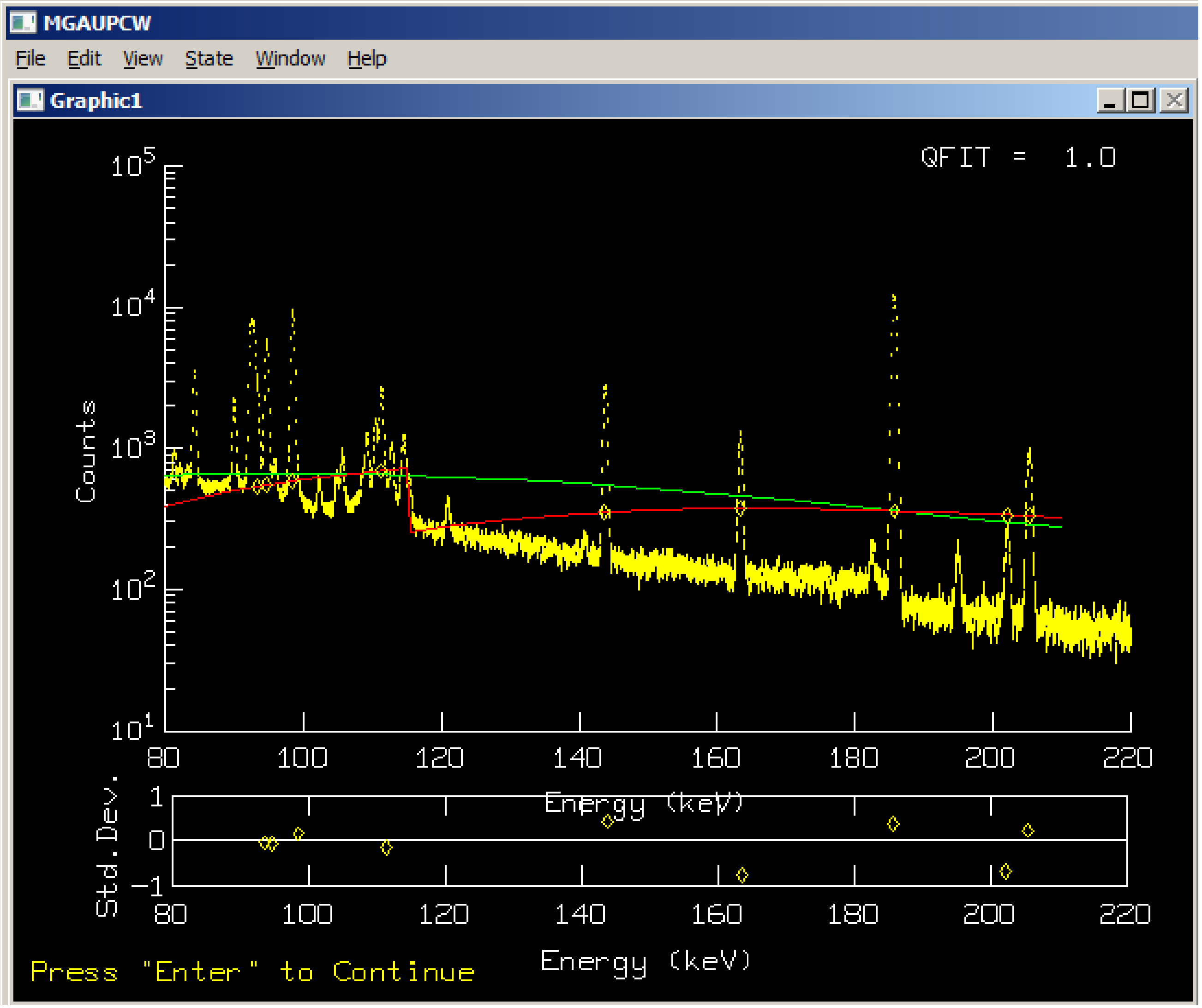
MGAU

- **Multi-Group Analysis for Uranium**
- **Uranium enrichment measurement method**
  - Uses 89-120 keV region of uranium gamma-ray spectrum



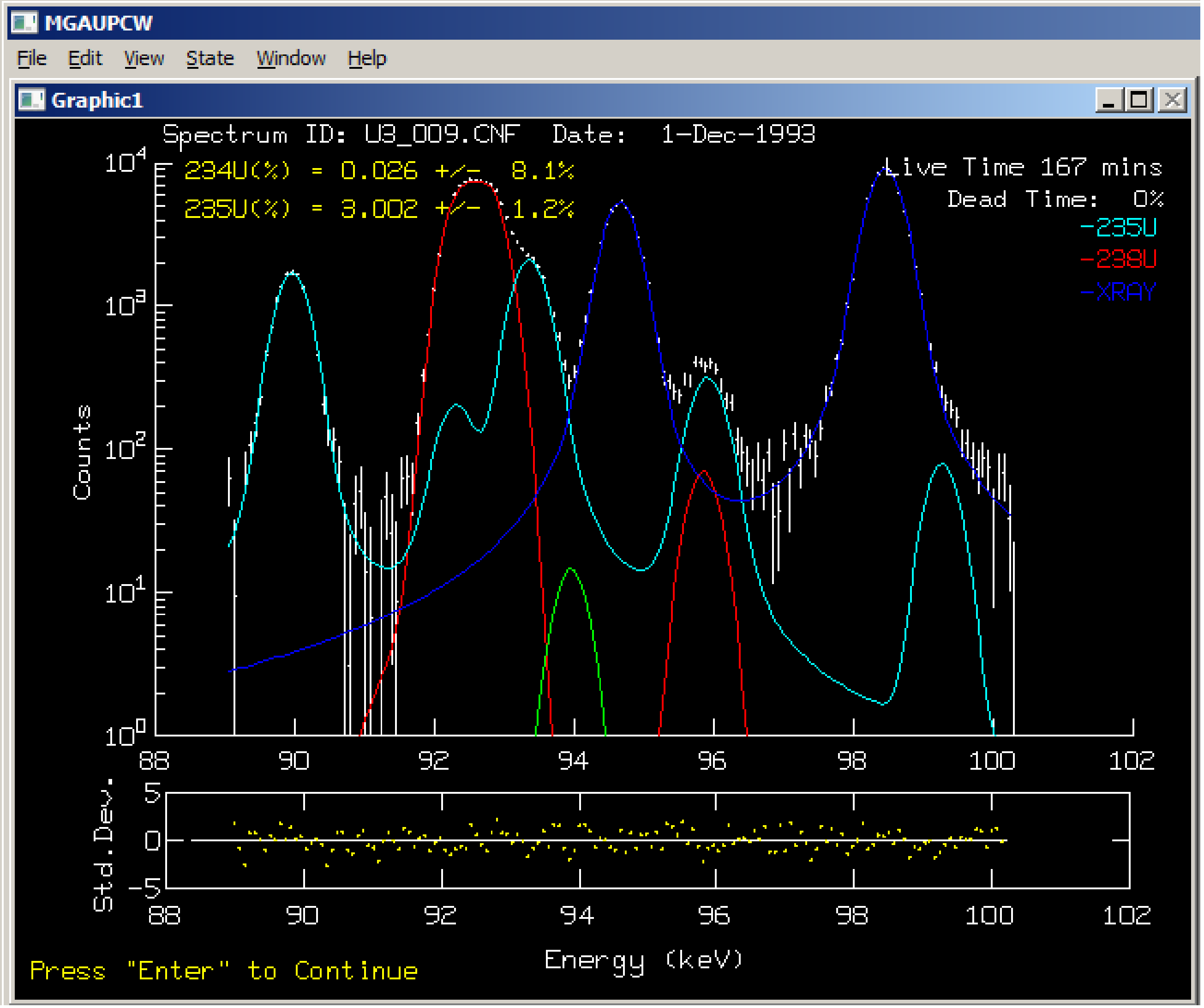
- **The low energy region of the spectrum is very complex with many overlapping X-ray and gamma ray peaks**

MGAU



MGAUPCW
File Edit View State Window Help
Graphic1
QFIT = 1.0
Counts
10^5
10^4
10^3
10^2
10^1
80 100 120 140 160 180 200 220
Energy (keV)
Std. Dev.
1
0
-1
80 100 120 140 160 180 200 220
Energy (keV)
Press "Enter" to Continue

MGAU



MGAUPCW
File Edit View State Window Help
Graphic1
Spectrum ID: U3_009.CNF Date: 1-Dec-1993
234U(%) = 0.026 +/- 8.1%
235U(%) = 3.002 +/- 1.2%
Live Time 167 mins
Dead Time: 0%
-235U
-238U
-XRAY
Counts
Std.Dev.
Energy (keV)
Press "Enter" to Continue

Heat

Calorimetry

- Well-established, precise method of NDA
- Uses thermal power generated by radioactive decay in the sample to determine the mass of special nuclear material
- Heat flow calorimetry is most commonly used for safeguards NDA
- 60 Wheatstone bridge calorimeters currently being used for Pu and tritium measurements at LANL
- Bulk measurements can be taken without issues from absorption or self-shielding
- Takes much longer than other NDA techniques

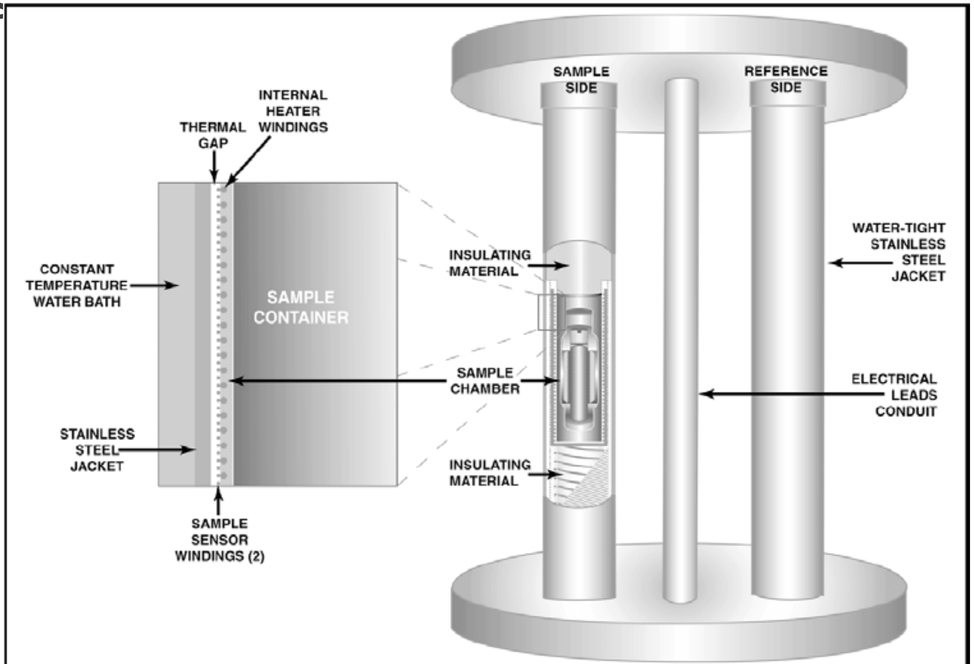


Figure 3. Schematic of a twin-bridge heat-flow calorimeter with basic components labeled.

Spent Fuel NDA

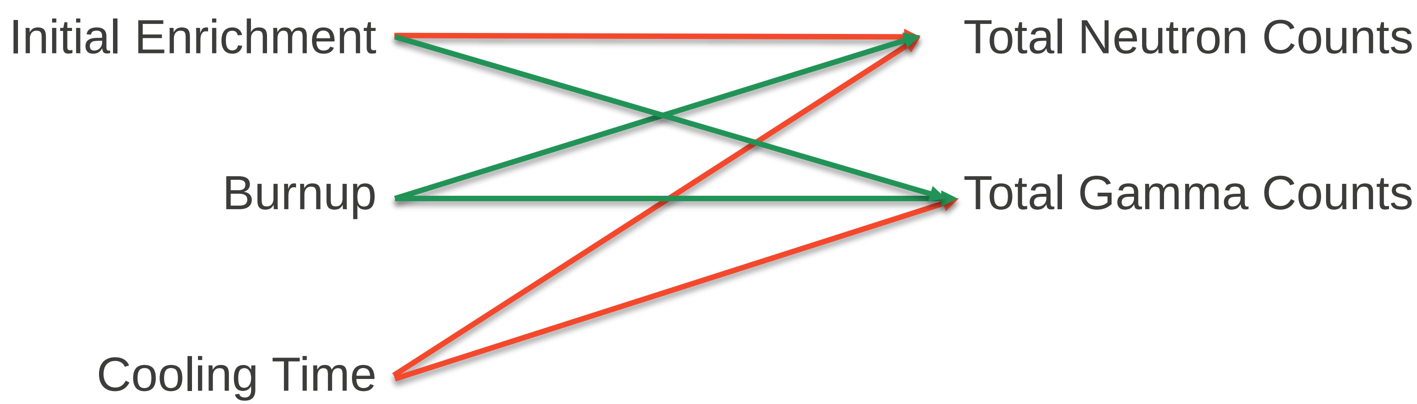
Spent Fuel NDA: Objectives

- **Verify operator declaration of residual uranium, and buildup of plutonium**
  - Burnup
  - Initial enrichment
- **Verify cooling time of assembly to assist with other parameters**
- **Verify completeness of assemblies**

- **IAEA R&D plan lists a milestone requesting "…more sensitive and less intrusive alternates to existing nondestructive assay (NDA) instruments"**
  - IAEA Department of Safeguards Long-Term R&D Plan, 2012-2023, Vienna, January (2013).

Spent Fuel NDA: Challenges

- **Interruptions to reactor operations**
  - Nuclear facilities have a standard way of operating and large disruptions (i.e. long measurements, drastic fuel movement) are not acceptable
- **Fuel inhomogeneity**
  - Both axially and radially, neutron flux in the reactor affects burnup, resulting in inhomogeneous fuel assemblies
- **Competing parameters**



- **Very difficult to accurately model**
  - Burnup codes are highly dependent on the accuracy of nuclear data and reactor operating history

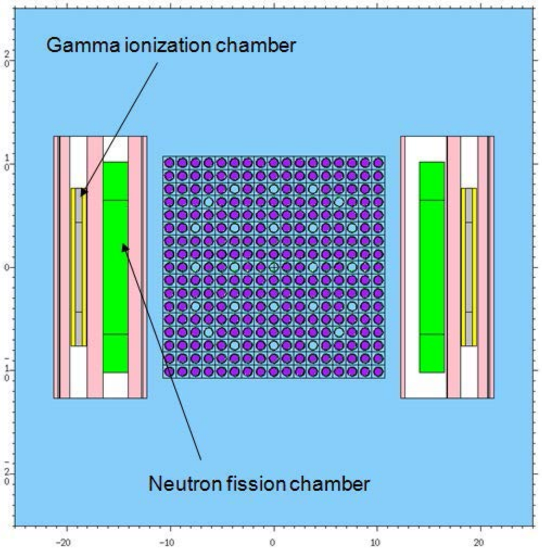
Fork Detector

- **NDA technique widely used by the IAEA and EURATOM**
- **Detector system straddles light water reactor fuel assemblies with four fission chambers (neutrons) and two ion chambers (gammas)**
  - Total gamma and neutron intensities as well as ratios of intensities give information about fuel assembly such as cooling time and burnup
  - One of the fission chambers is wrapped in cadmium to provide a means for estimating multiplication
- **Other versions of the Fork detector exist with ^{3}He tubes instead of fission chambers, etc.**

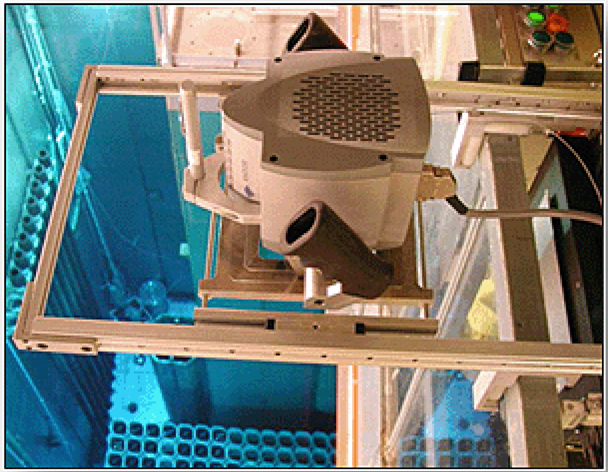
Fork Detector

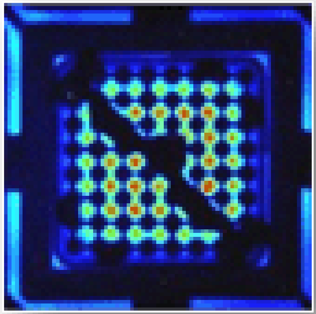
- **Benefits**
  - Rugged, reliable, validated and verified, easy to use
  - Requires minimal fuel movement

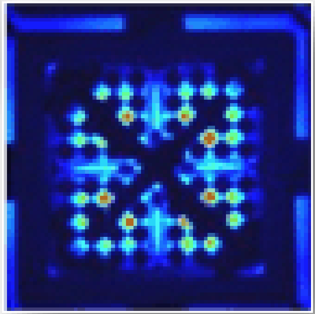
- **Drawbacks**
  - Asymmetric burnup could affect gamma signal
  - Assumptions about how neutron and gamma counts trend with burnup and cooling time fall apart under irregular burning history
  - Results rely heavily upon data provided by operator
  - May not be able to detect pin removal under 50%



Cerenkov Viewing Device



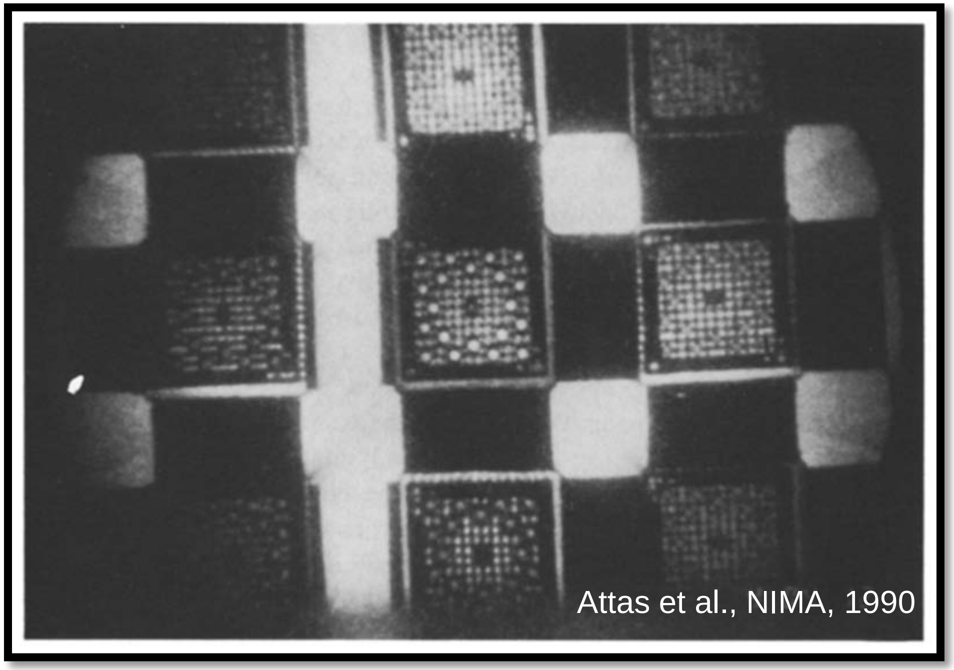




- Viewing device sensitive to ultraviolet radiation in the water surrounding spent fuel
- Cerenkov radiation provides the UV light and is derived from the intense gamma radiation in spent fuel
- Electrons may exceed the speed of light in water and therefore must lose energy by emitting Cerenkov radiation. β particles contribute as well
- Glow patterns above fuel rods used to distinguish fuel from non-fuel

Cerenkov Viewing Device

- **Benefits:**
  - Tested, validated method with reliable history of use
  - Readily detects missing fuel rods
  - Burnup and cooling time verification
  - Indirect measurement method, meaning fuel assemblies may remain in storage positions

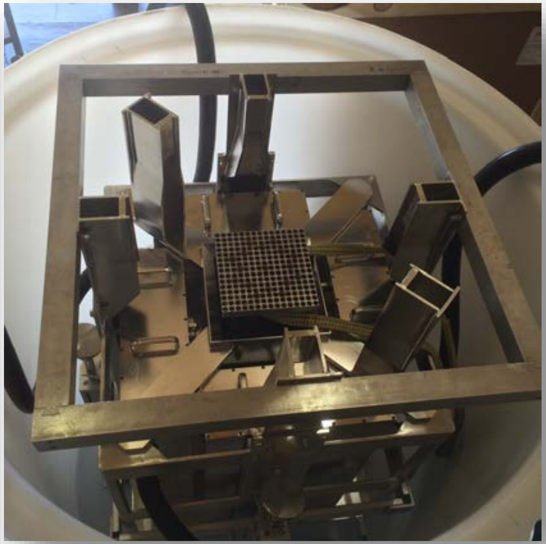


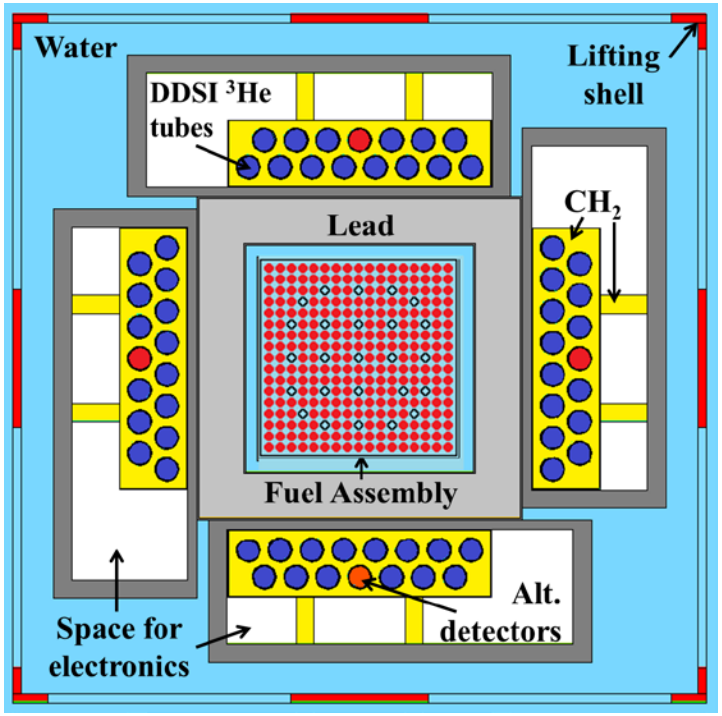
Attas et al., NIMA, 1990

- **Drawbacks:**
  - Murky water or weak Cerenkov signals can inhibit ability to use CVDs
  - Neighboring assemblies in pool can confuse measurement
  - Limited to certain burnups and cooling times due to required signal strength
  - Potentially easy to fool with cutoff pins or fake fuel rods

Differential Die-Away Self-Interrogation

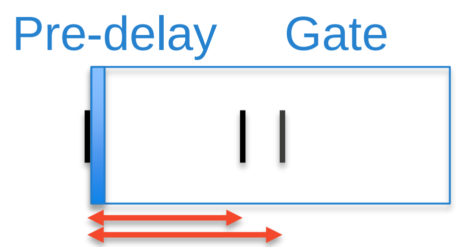
- **Spontaneous fission neutrons from ^{244}Cm, ^{240}Pu in spent fuel thermalize in water and interrogate fuel pins**
- **Neutron coincidence counting: aim to detect two neutrons that are temporally correlated**
  - Same fission event, same fission chain
- **Record times of neutron detections**
  - ➢ *list-mode data*

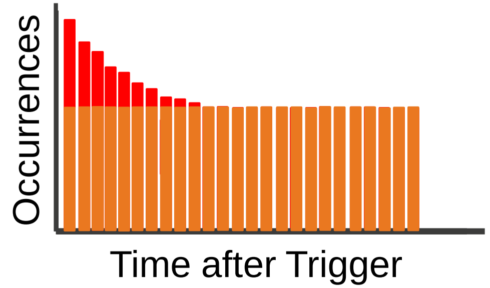


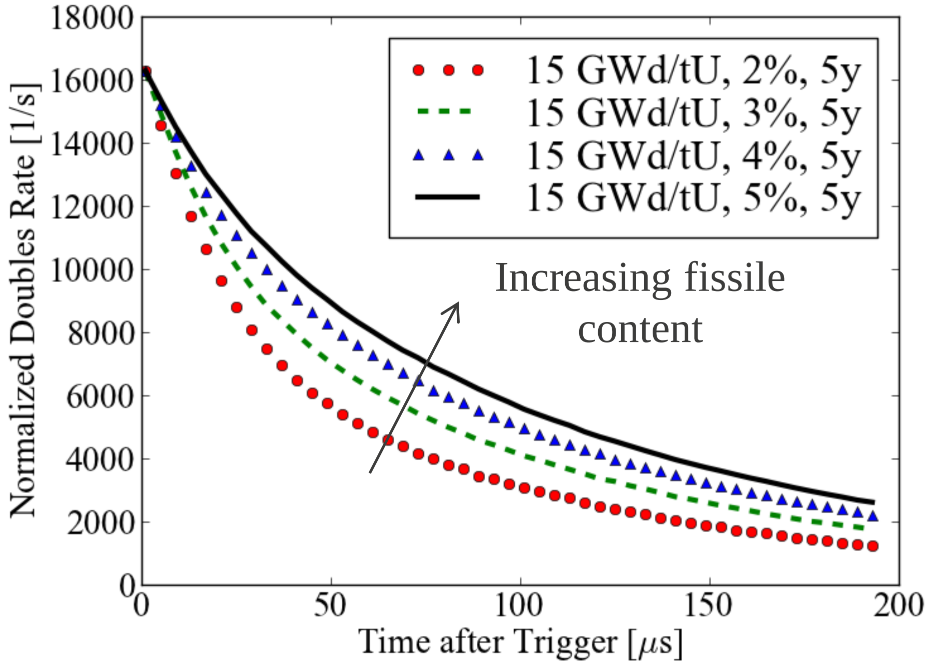


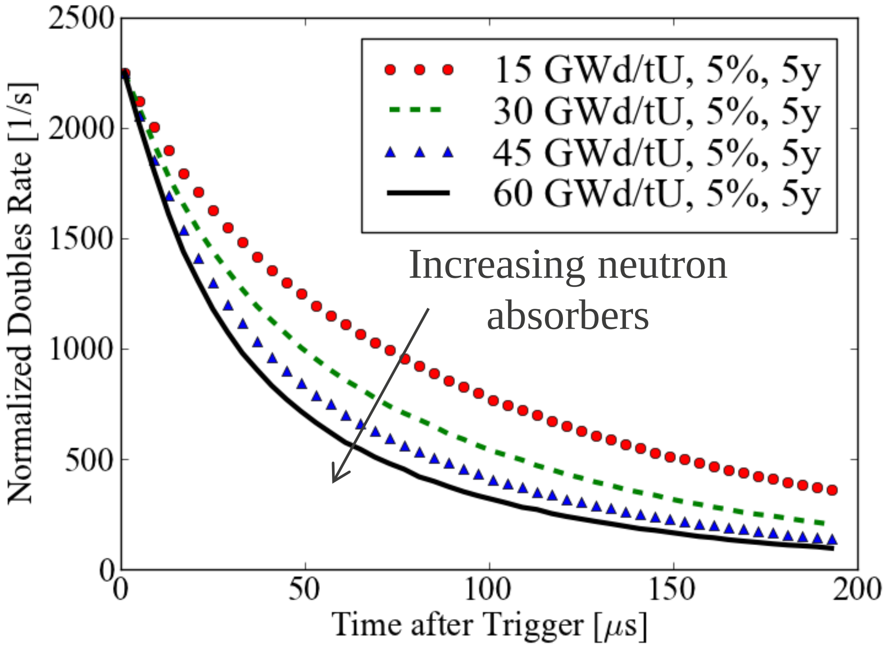
Differential Die-Away Self-Interrogation

- Rossi-alpha distribution is a histogram of the times between the trigger and each neutron in the gate



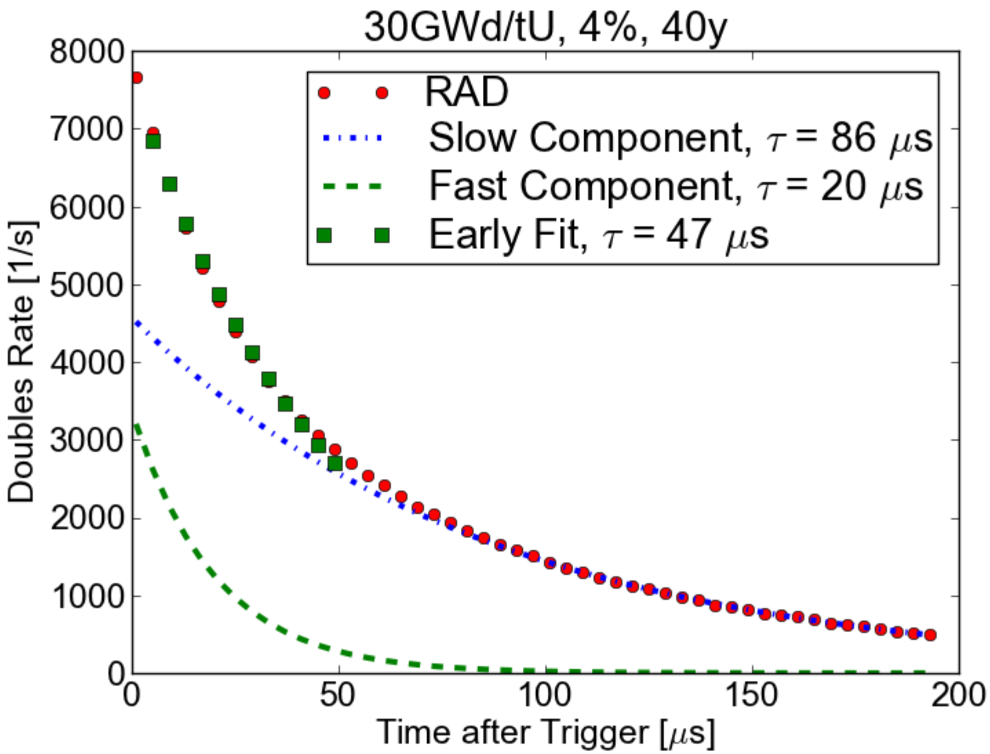






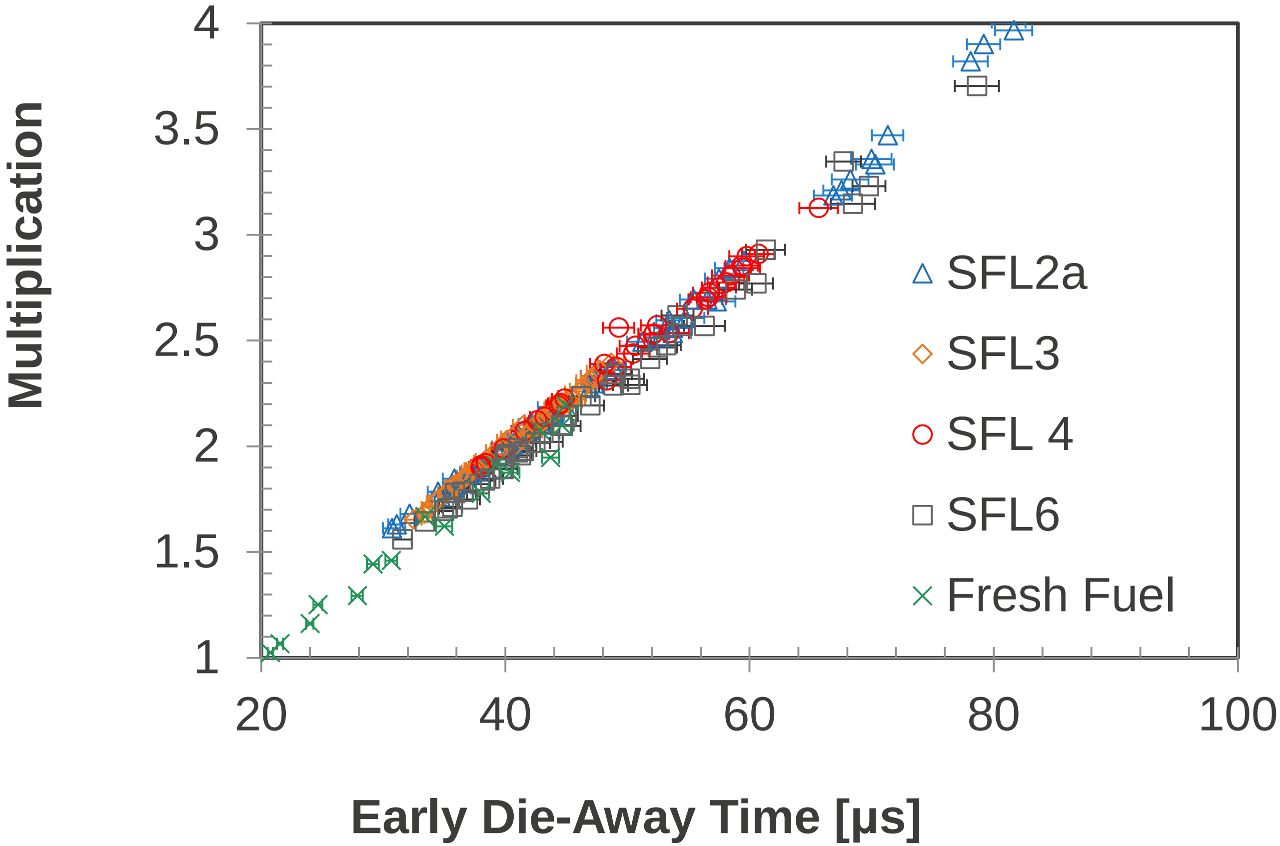
DDSI Analysis

- **RAD can be broken down into fast and slow components**
  – Fast component comes from detector die-away time
  – Slow component comes from neutron lifetime in the fuel
- **Early time domain of RAD can be fit with single exponential to yield early die-away time**
  – $y=Ae^{-t/\tau}$



DDSI Analysis

- **Early die-away time is nearly linearly proportional to assembly multiplication**
  - Using this, one can determine whether pins have been removed



Advanced Experimental Fuel Counter

- **Designed for research reactors only**
- **System uses:**
  - Active and passive neutron coincidence counting;
  - An ion chamber for gross gamma-ray counting.
- **Measurement objective is to verify residual fissile mass (i.e., ^{235}U + ^{239}Pu) using active neutron interrogation**
- **Extended analysis of passive neutron and gamma-ray count rates helps verify declared burnup, cooling time, and initial enrichment**
- **Field trials have occurred as follows:**
  - 2006 High Flux Australian Reactor (HIFAR), Australia,
  - 2011 Institute of Nuclear Physics (INP), Uzbekistan, and
  - 2014 Institute of Nuclear Physics (INP), Uzbekistan.

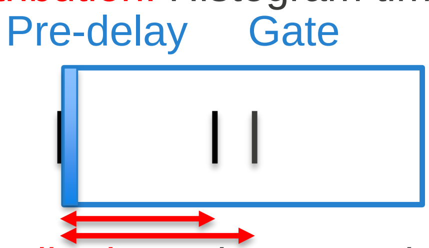
By K. Miller, H. Menlove

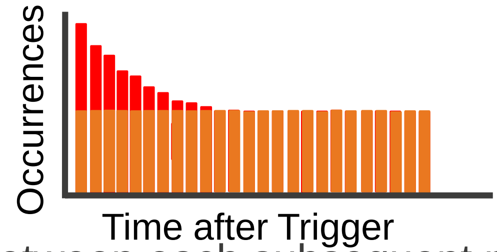
Safeguards at LANL

Thank you!

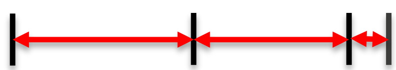
Neutron Counting: Distributions

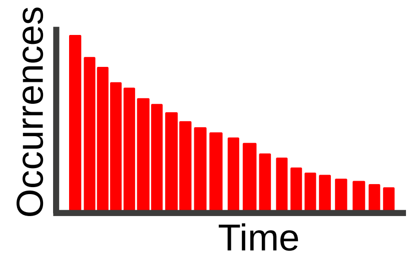
- **If you record the time of arrival of every neutron (list mode), several distributions can be produced:**
  - Rossi-Alpha Distribution: Histogram times between neutron arrivals within a certain gate





  - Time-Interval Distribution: Histogram times between each subsequent neutron





  - Multiplicity Distribution: Count number of neutrons in gate after trigger