

Material Accounting Challenges for the Advanced Fuel Cycle, Potential Nuclear Measurements and Attendant Data Needs

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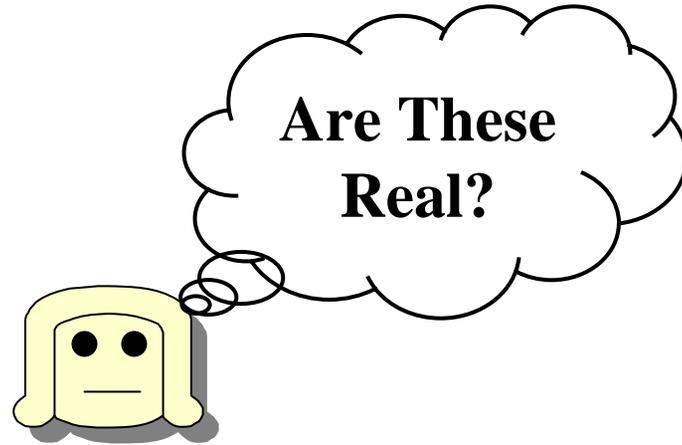
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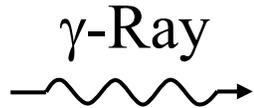
Desire to Detect and Quantify Fissile Materials During Arms Reduction Negotiations



- START I and START II Require Verification
 - Quantify Number and Size
 - Monitor Material Movement In/Out of Facilities
- Passive Radiation Detection Problematic
 - Fissile Materials Not Very Radioactive
 - Easy to Shield Radiation
- Also Developed for Quality Assurance and Waste Assaying

Active Evaluation Techniques Use Radiation as Probe

Radiation



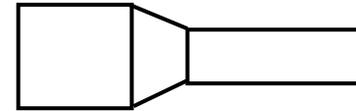
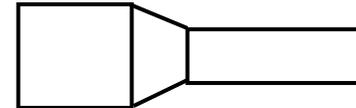
or



Neutron



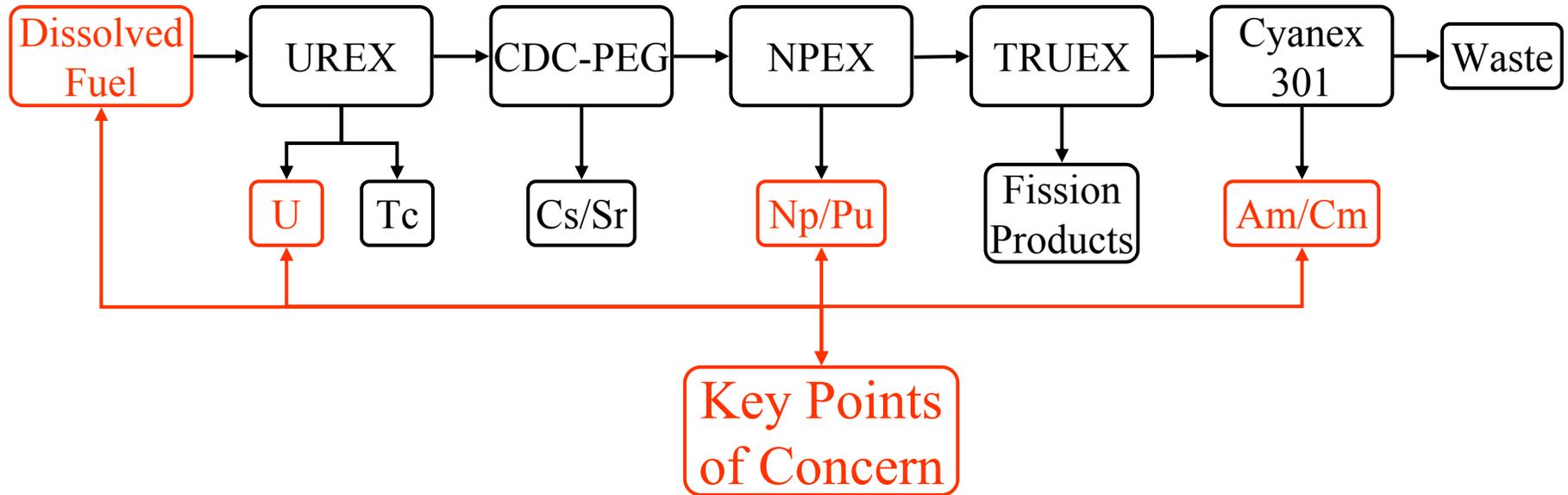
Detectors



- External Radiation Sources
 - Neutron [(n,x) Reactions]
 - Radioisotope
 - Accelerators
 - γ -rays [(γ ,x) Reactions]
 - Accelerators

- Resulting Emission
 - Neutrons
 - Prompt
 - Delayed (Fissionable Materials)
 - High Energy Photons
 - x-rays (XRF)
 - γ -rays (NRF, Capture, Activ.)
- Provide Signature of Material

Active Techniques Applied to Material Accountability



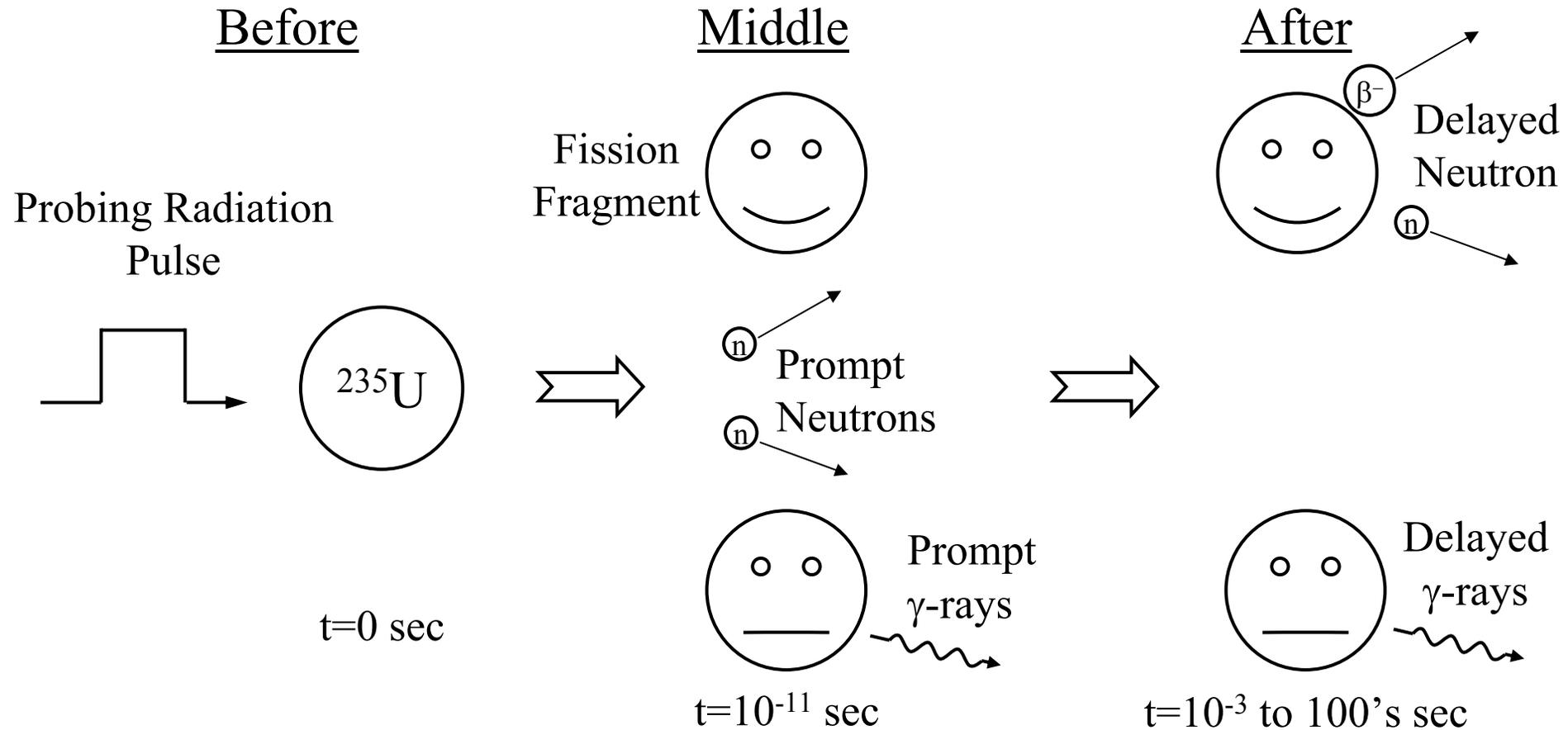
- Reprocessing: Old Fuel → New Fuel
- Techniques to Monitor Fissile Material Quantities
 - Real Time
 - Accurate
 - Sensitive
- Less Separation → More Difficult to Quantify

Active Evaluation Applicable to Homeland Security



- Commerce → Movement of Goods
 - 95% Arrives by Ship
 - 22.5 Million Cargo Containers
 - 118 Million Vehicles
- Very Few Inspected
 - 2% of Containers Inspected
- **Data Needs Overlap with DHS**
- Fissile Material Abundant
 - ~100 Tons in Storage
 - Located at 100's of Sites
 - Security at Some Sites is Lax
 - Some Material is “Missing”
- ~4 kg ^{239}Pu for a Crude Bomb

Plethora of Radiation Emitted from Fission Reactions



- Delayed γ -ray Yield $\sim 400\%$
 - Most Materials Emit
- Delayed Neutron Yield $\sim 2\%$
 - Fissionable Materials Only
- Prompt γ -ray Yield $\sim 800\%$
- Prompt Neutron Yield $\sim 200\%$

Neutron and γ -Rays Probes Complimentary

Neutron

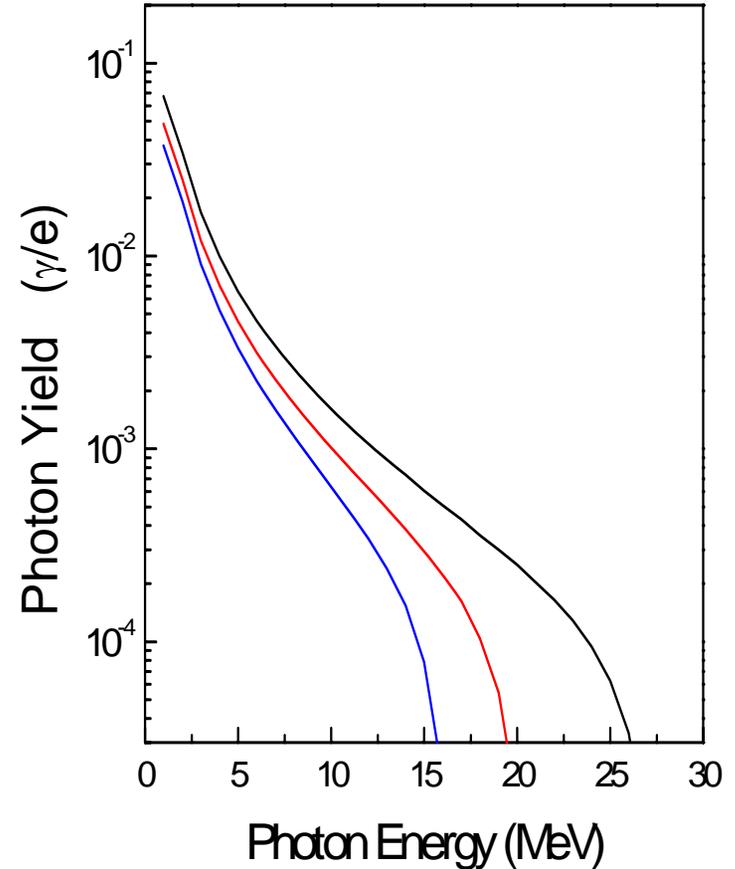
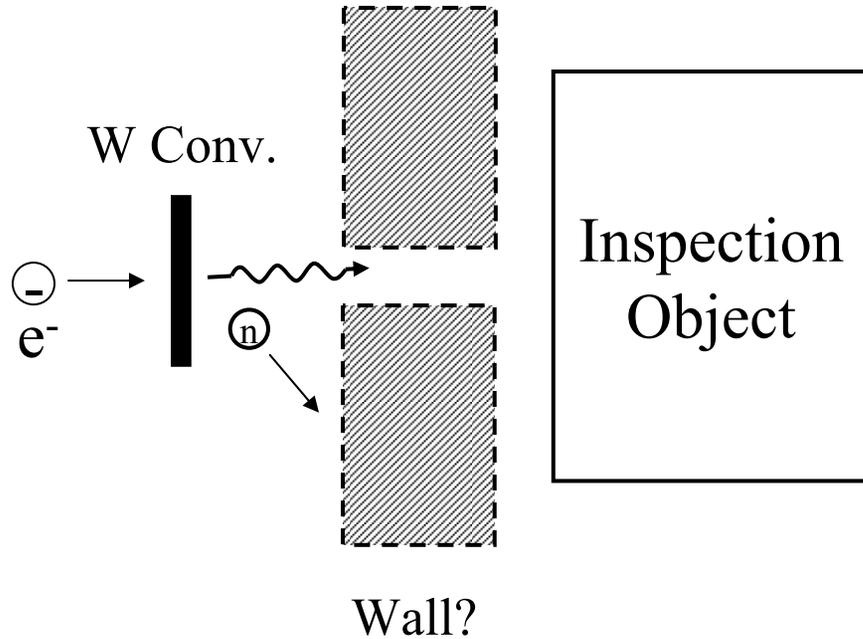
- Advantages
 - Highly Penetrating in High-Z
 - Easier to Detect γ -rays
 - Capture γ -rays Emitted
 - Better Known Cross Sections
- Disadvantages
 - Shielded by Low-Z
 - Hard to Detect Neutrons
 - Omni Directional

γ -Ray

- Advantages
 - Highly Penetrating in Low-Z
 - Easier to Detect Neutrons
 - Forward Directed
 - Accelerators Very Robust
- Disadvantages
 - Shielded by High-Z
 - Hard to Detect γ -Rays
 - Poorly Known Cross Sections

At Idaho Accelerator Center Only Tried Bremsstrahlung Beam

Photons or Neutrons Created by High-Z Converter

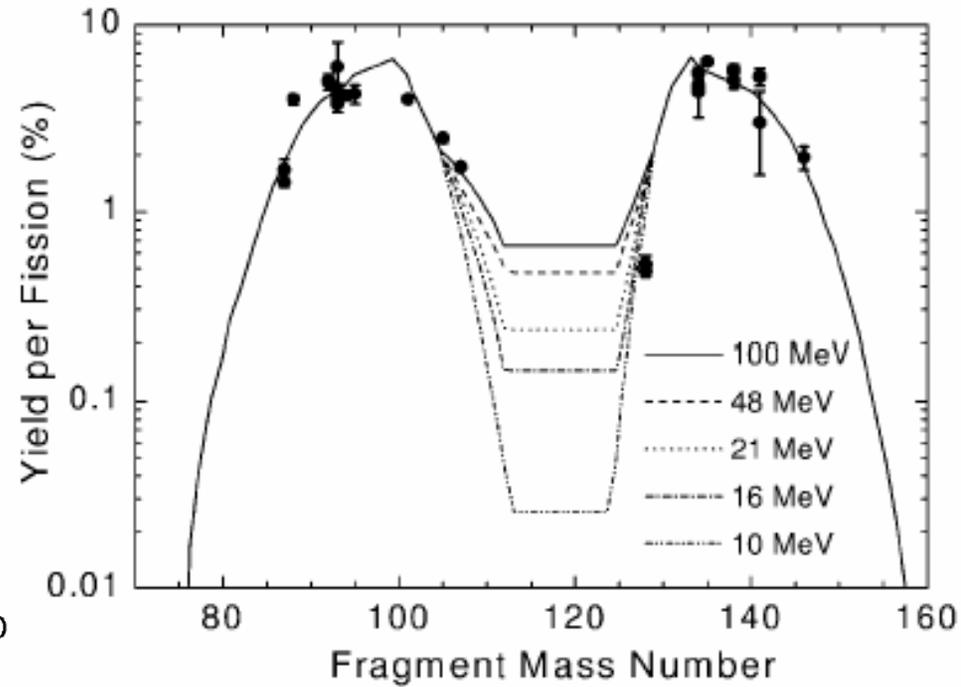
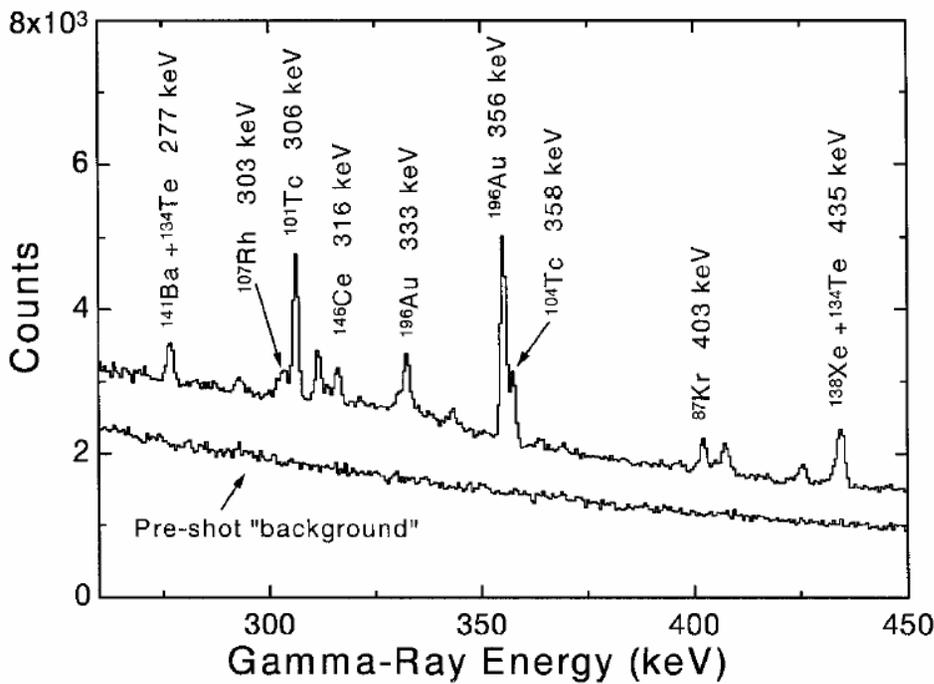


- γ -rays \rightarrow Thin Converter (~ 1 mm)
- Neutrons \rightarrow Thick Converter (~ 5 cm)
- Extensive Shielding May Be Required for Converter Emissions
- **New High-Energy γ -Ray Sources of Great Interest**
 - **Monoenergetic!**

γ -Ray Emissions

Fissile Material Detection Techniques
Using γ -Ray Emissions

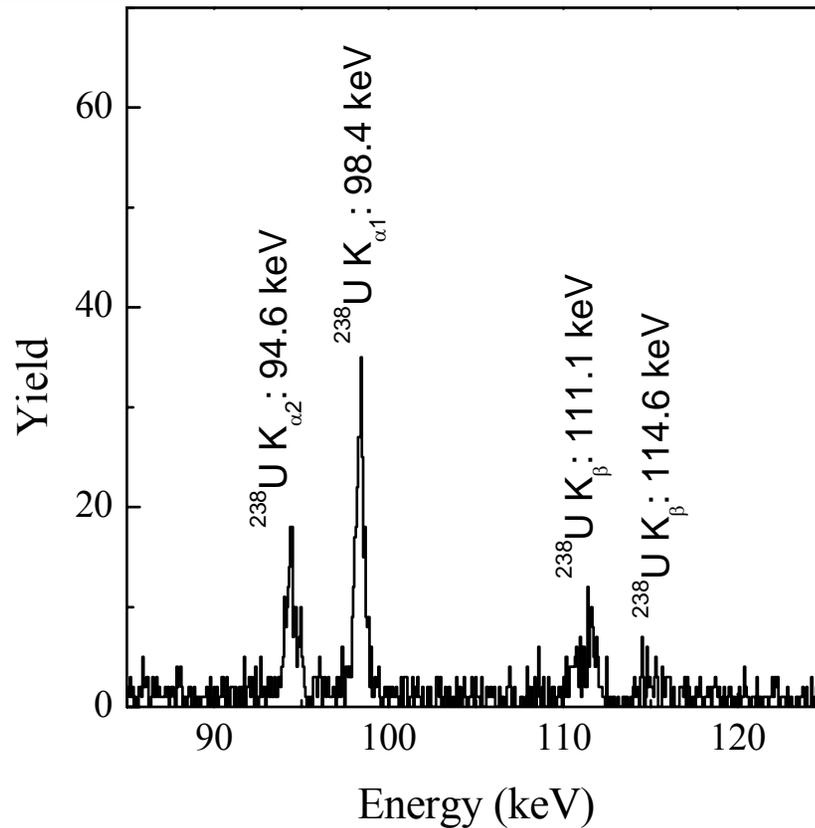
Activation Analysis Identifies Fissionable Material



T. E. Cowan, A. W. Hunt et. al.

- Fission Products “Unique”
 - Neutron Rich
 - Short Lived $t_{1/2} \approx 10$ min.
- Could be Significant Contamination in Reprocessing
- High Radiation Fields Difficult for HPGe Detectors
- **Need Accurate Fission Cross Section and Fragment Distributions**

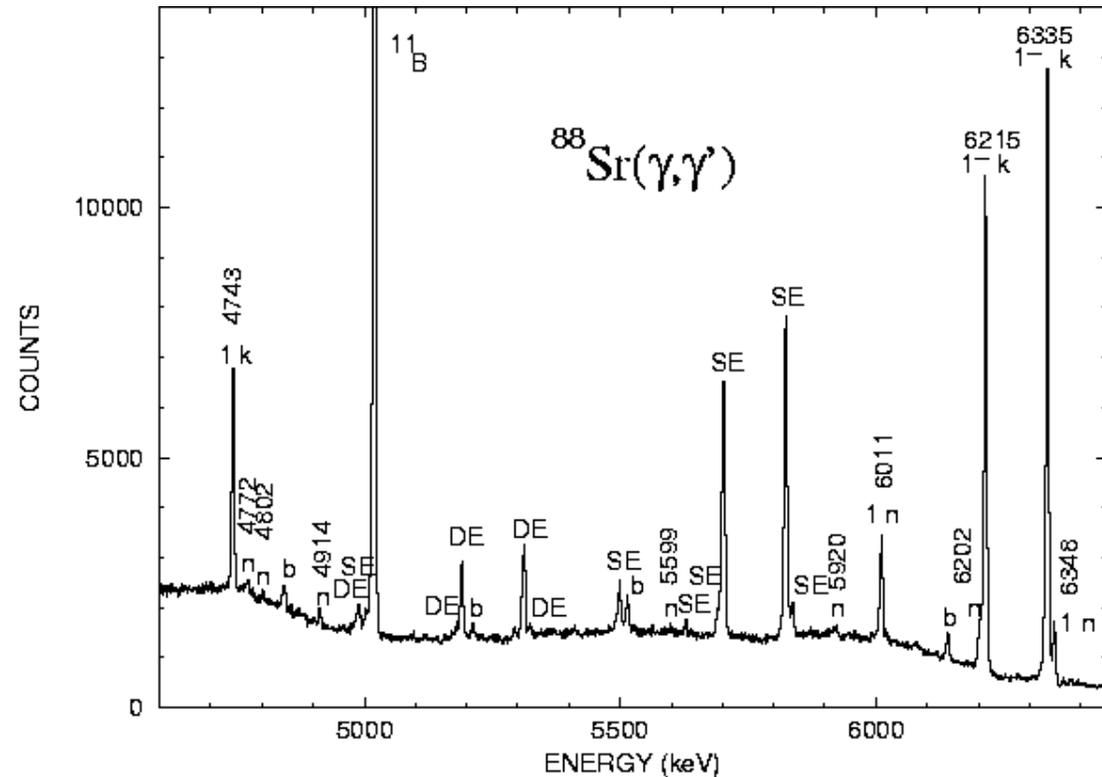
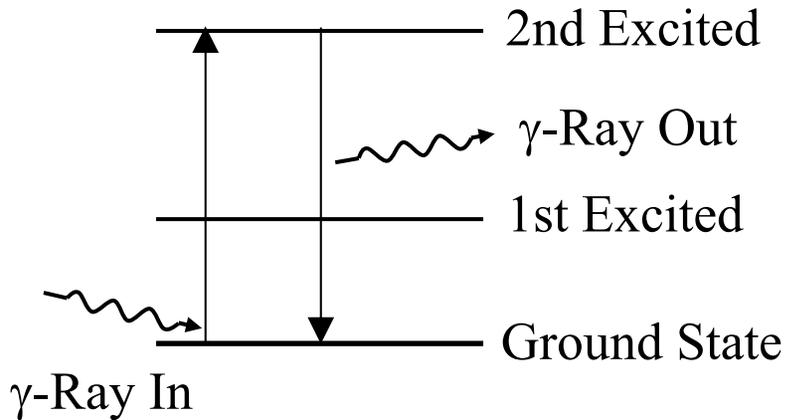
X-Ray Fluorescence Identifies Elements



^{238}U ~5 g
5 MeV Brem.
325 sec.
60 Hz

- X-Ray Lines Provides Elemental Fingerprint
- Sensitivity ~ppm
- Not Isotope Specific
- Extensively Use in Literature (Waste Assay, Environmental and Medical)
- May Not Need Probe (e.g. Enough Natural Activity)
- High Radiation Fields Difficult for HPGe and Si Detectors

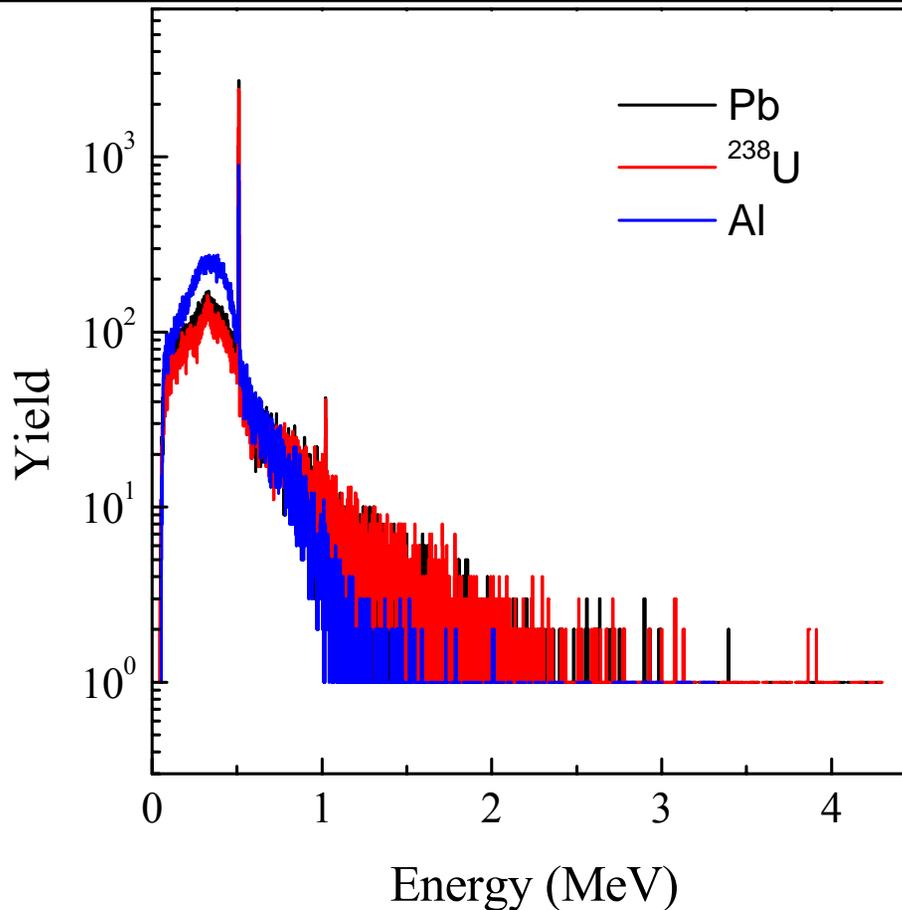
Nuclear Resonance Fluorescence Provides Finger Print



L. Kaeuble et. al.

- Photons Excite Nucleus
 - Decays by Photon Emission
 - Analogous to XRF
 - Isotope Identification
- Physics is Proven
 - **Levels Not Measured for ^{235}U , ^{239}Pu etc...**
- Technology Not Developed
 - Evaluation Time Could be Long
 - Requires High Resolution Detectors (Problematic in High Radiation Fields)

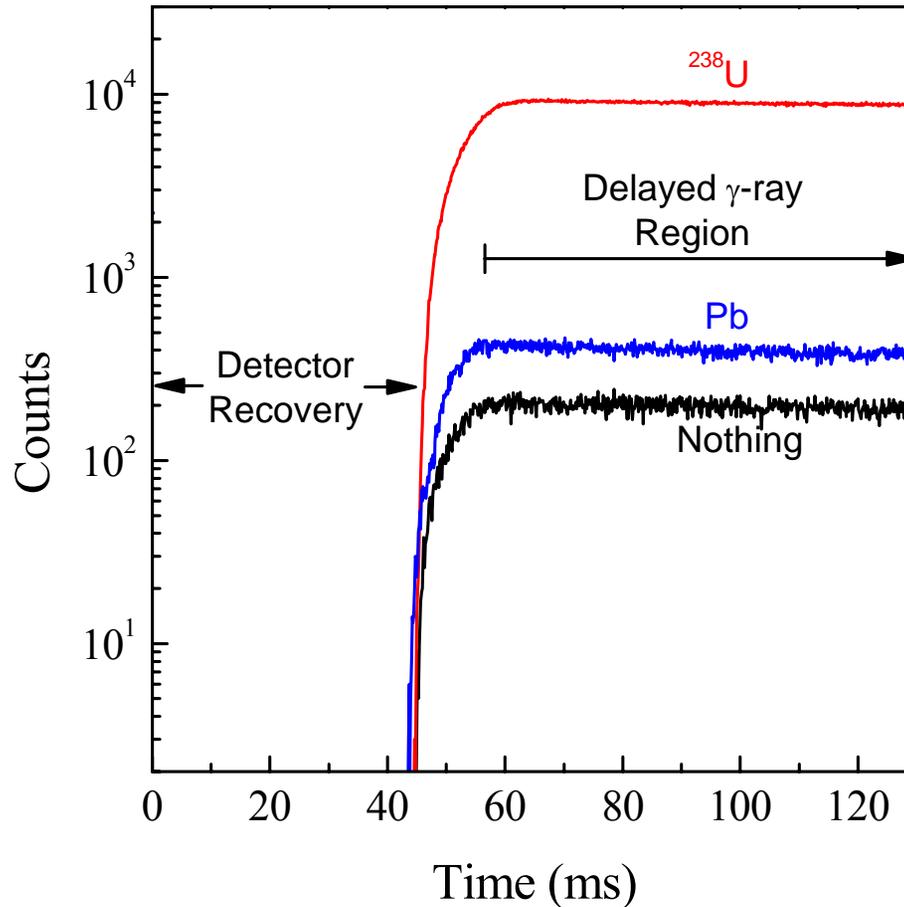
Prompt γ -Rays Difficult to Distinguish Using Bremsstrahlung



8 MeV Bremsstrahlung
500 Hz Rep. Rate
 $\frac{1}{4}$ " ^{238}U
20 min Data Collection

- Atomic Cross Sections Dominate
- Fission γ -Rays Broad Energy Spectrum
- **Need Prompt γ -Ray Energy Distribution**
- Not Sure if Isotope Specific
- Want Higher Duty Factor Accelerator (New One Coming on Line)

Ubiquitous Delayed γ -Rays from Fissionable Material



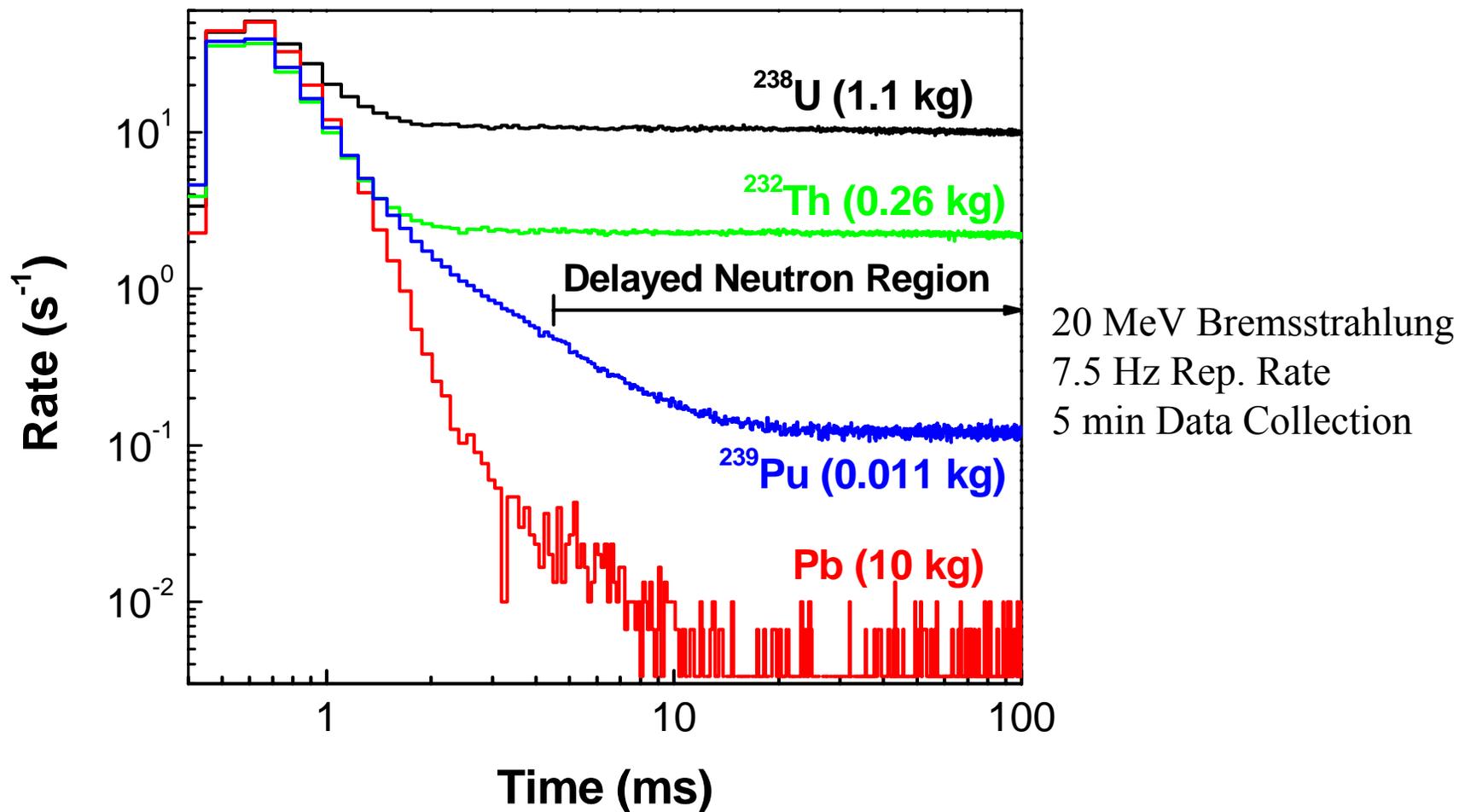
20 MeV Bremsstrahlung
7.5 Hz Rep. Rate
1.1 kg ^{238}U
10 kg Pb
5 min Data Collection

- ~100 Times More γ -rays for ^{238}U in ~100's ms
- Broad Energy Distribution (Careful Measurement Planned)
- Are There Short Lived Lines for Isotope Identification
- **Need Delayed γ -Ray Energy Distribution**

Neutron Emissions

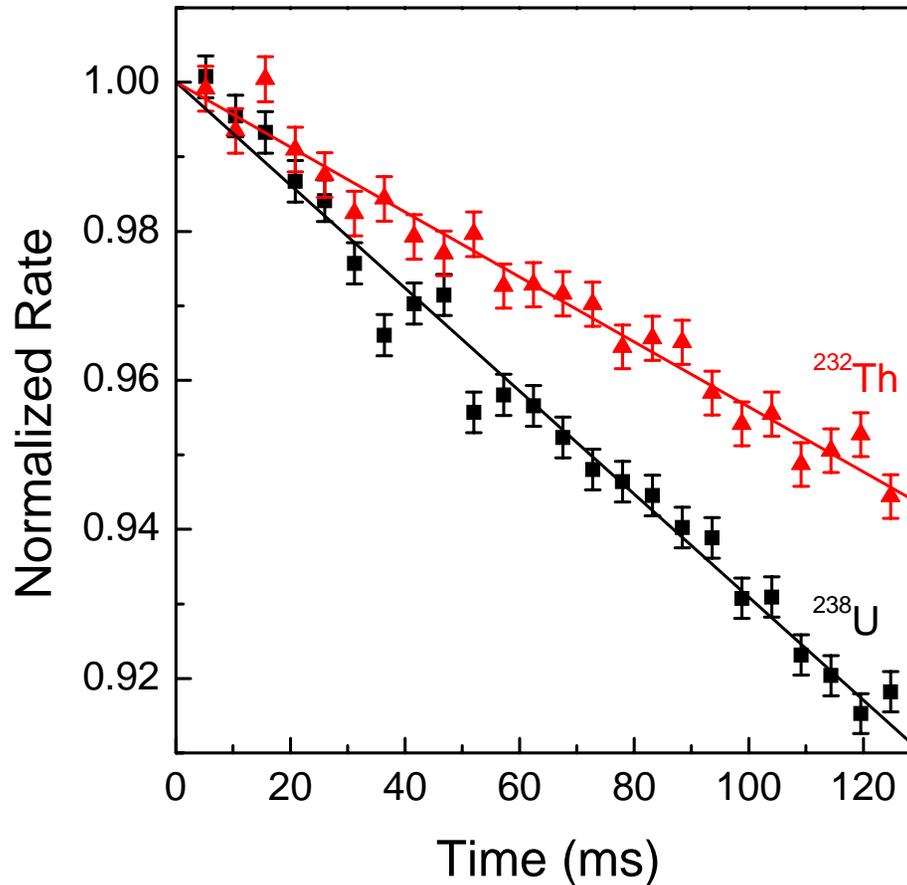
Fissile Material Detection Techniques
Using Neutron Emissions

Delayed Neutrons Excellent Signature of Fissionable Material



- Delayed Neutron Region beyond 5 ms
 - 2×10^6 More Neutrons Detected for ^{238}U

Decay of Delayed Neutron Emission Allows Discrimination between Fissionable Material

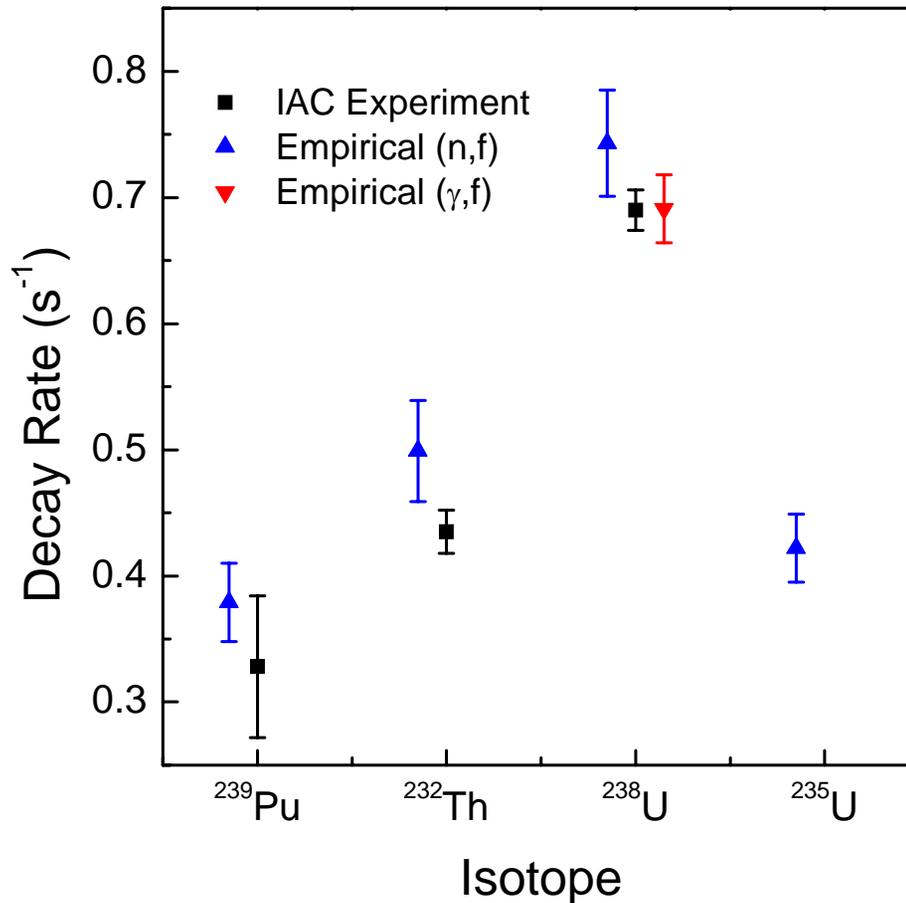


20 MeV Bremsstrahlung
7.5 Hz Rep. Rate
1.1 kg ^{238}U
0.26 kg ^{232}Th
5 min Data Collection

- Decay Dictated by β^- Emission and Precursor Yield

- $$R = N_o \sum_i \frac{\lambda_i \alpha_i}{1 - e^{-\lambda_i \Delta t}} e^{-\lambda_i t} \quad \lambda_i; \alpha_i \text{ Decay Constant and Yield of Precursors}$$

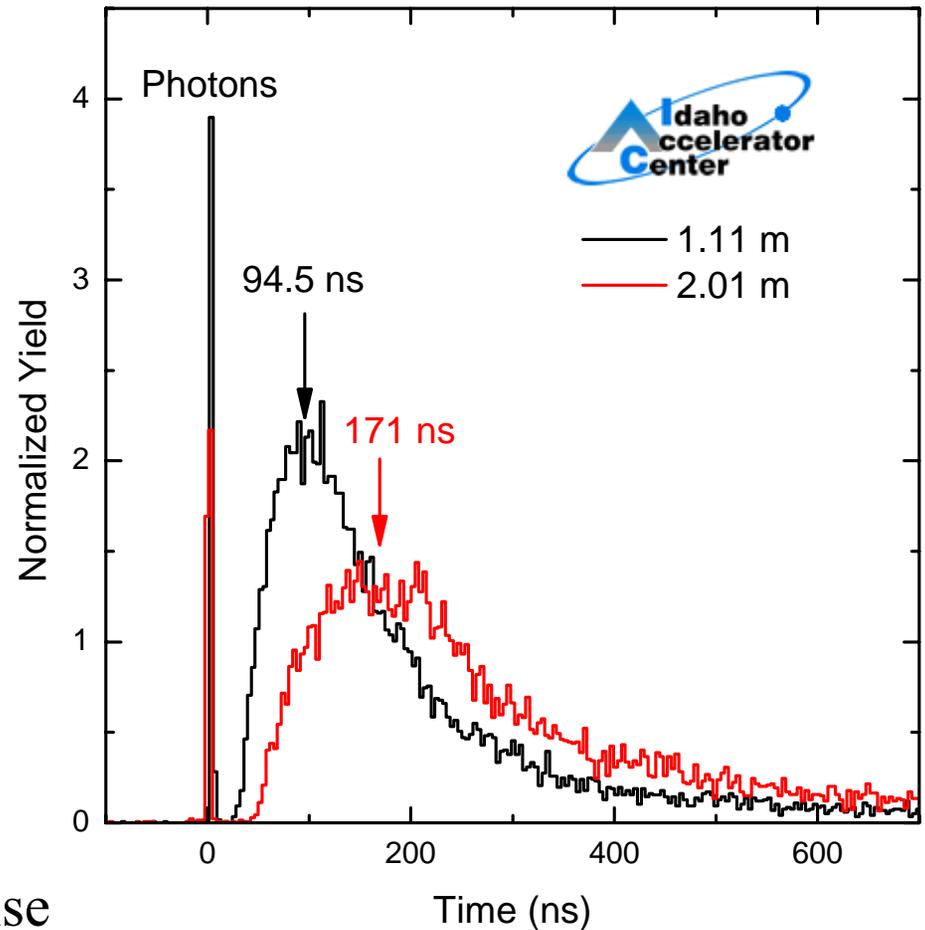
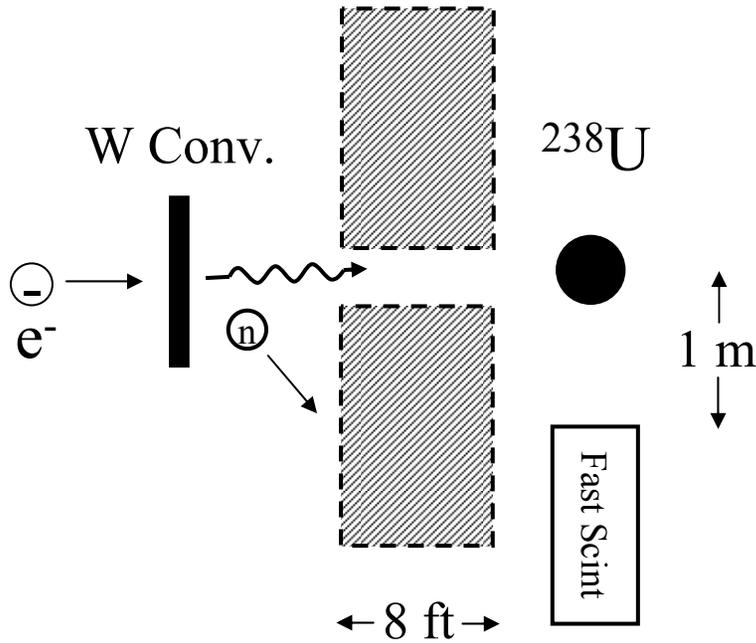
Decay of Delayed Neutron Emission Identifies Fissionable Material



20 MeV Bremsstrahlung
7.5 Hz Rep. Rate
1.1 kg ^{238}U
0.26 kg ^{232}Th
11 g ^{239}Pu

- 10 σ Difference Between ^{238}U and ^{232}Th
- Limited Data for (γ ,f) to Make Prediction for Other Isotopes
- **Need Accurate Measurement of Delayed Group Parameters**

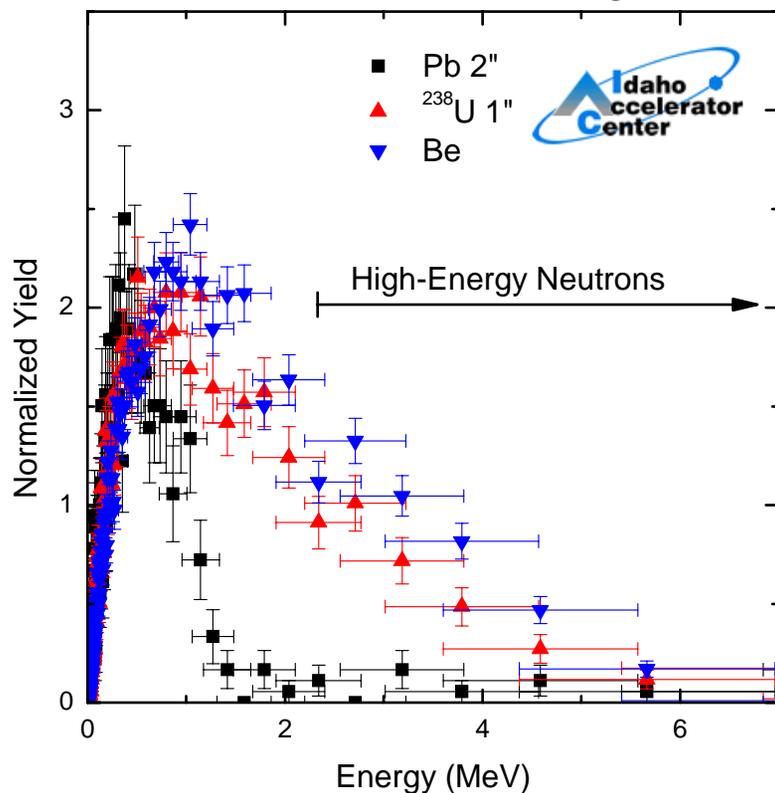
Neutron Time of Flight to Detect Prompt Neutrons



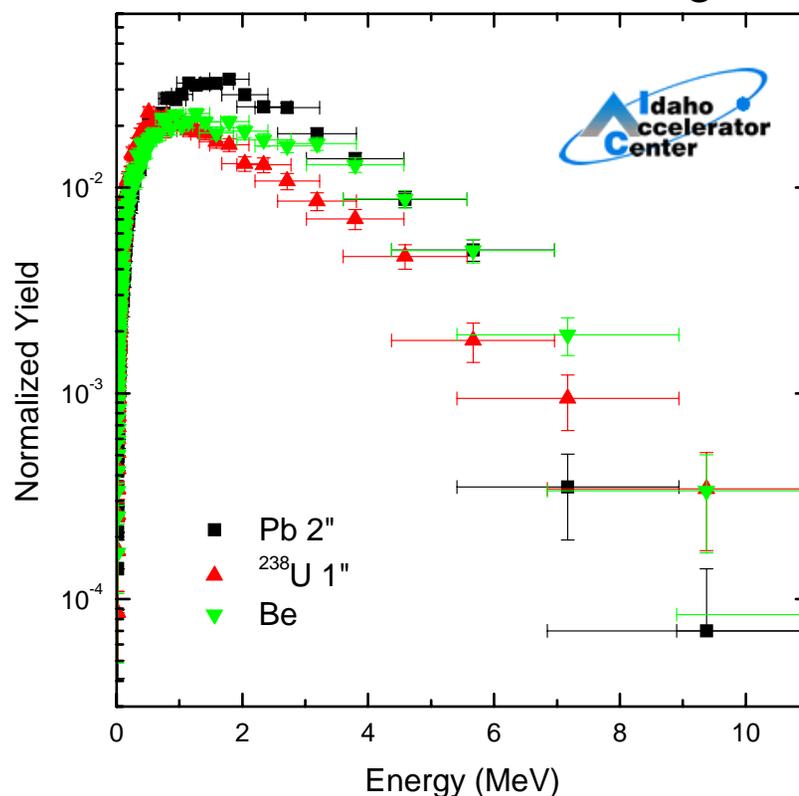
- Time Spectrum
 - TAC Started by Accelerator Pulse
 - TAC Stopped by Fast Scintillator (Proton Recoil Detector)
 - Shield by $\sim 8''$ of Pb
- ~ 1.8 ns Bremsstrahlung Pulse
- 100 to 1000 More Prompt Compared to Delayed

High-Energy Neutrons Distinguish Fissionable Isotope

9 Mev Bremsstrahlung

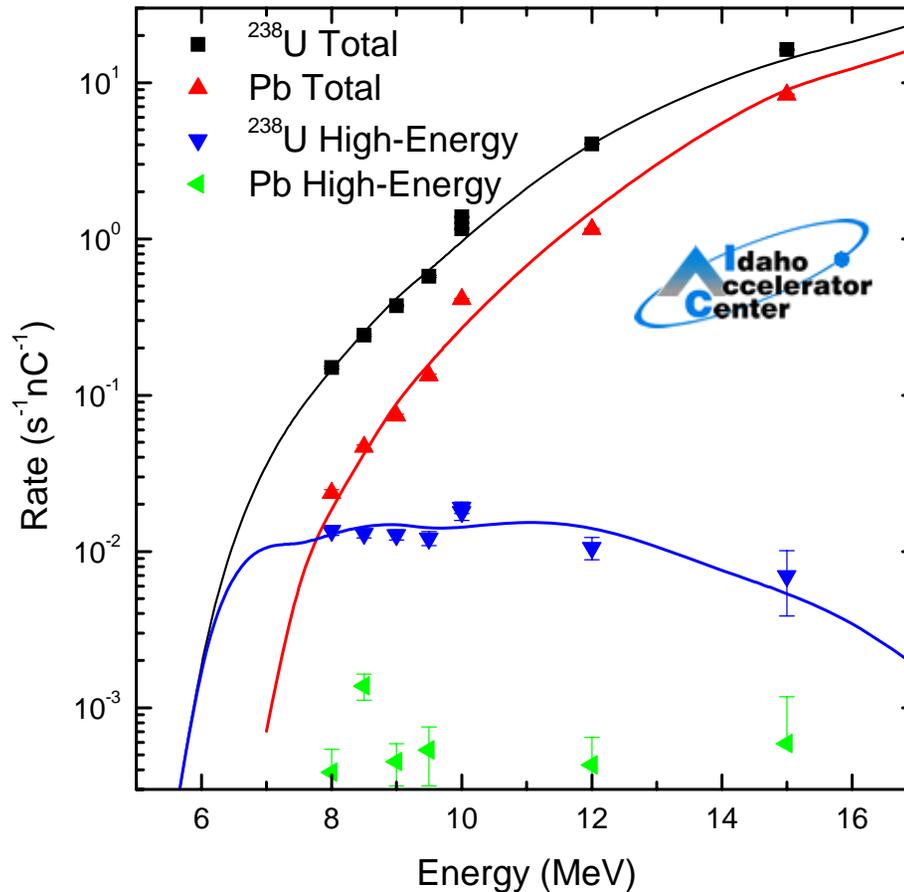


15 Mev Bremsstrahlung



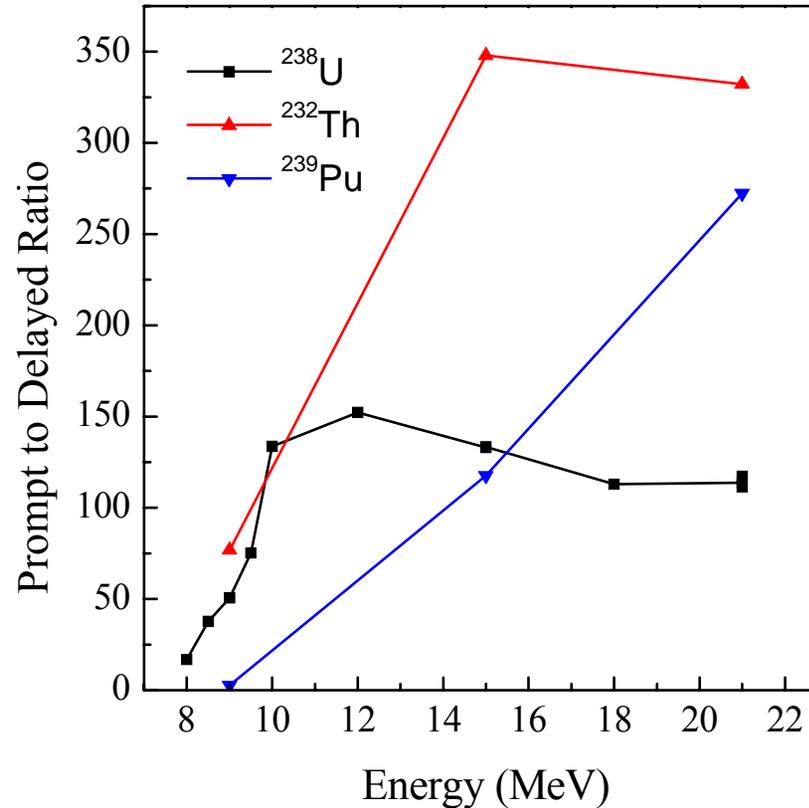
- Lower Energy Bremsstrahlung
 - $(\gamma, f) \rightarrow$ High-Energy Neutrons (i.e. Watts Spectrum)
 - $(\gamma, n) \rightarrow$ Neutron Energy Limited; $E_n < E_{\max} - E_{\text{Thr}}$
- Isotopic Specificity?

High-Energy Neutrons Signify Fissionable Material



- Significant High-Energy Neutrons from ²³⁸U
- Lower Energies (~7 MeV) May Be Advantageous
 - Allows Simpler Detectors?
- **Need Accurate Measurement of Neutron Energy Distribution**

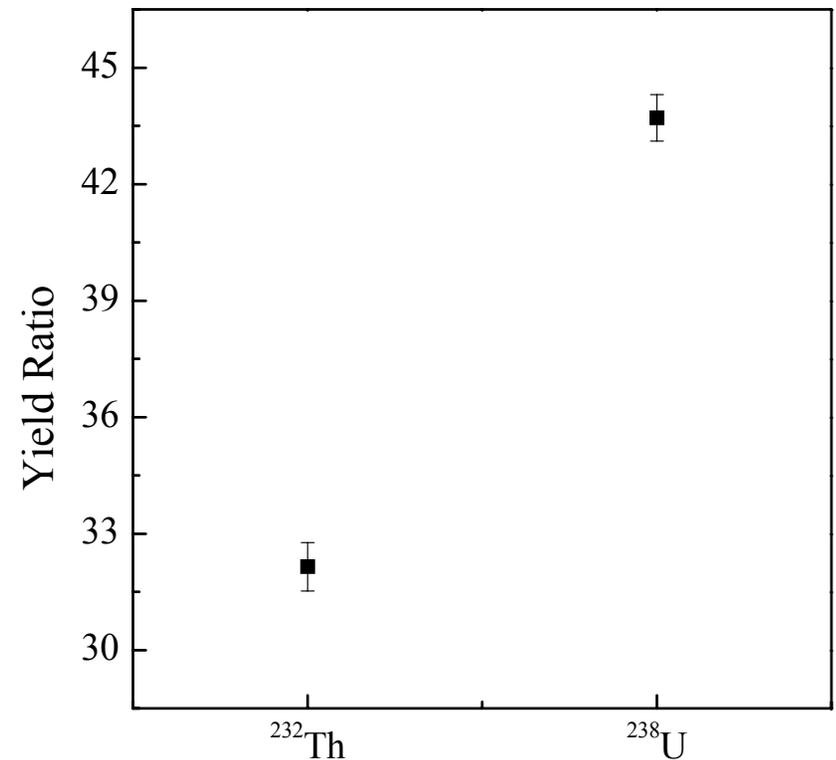
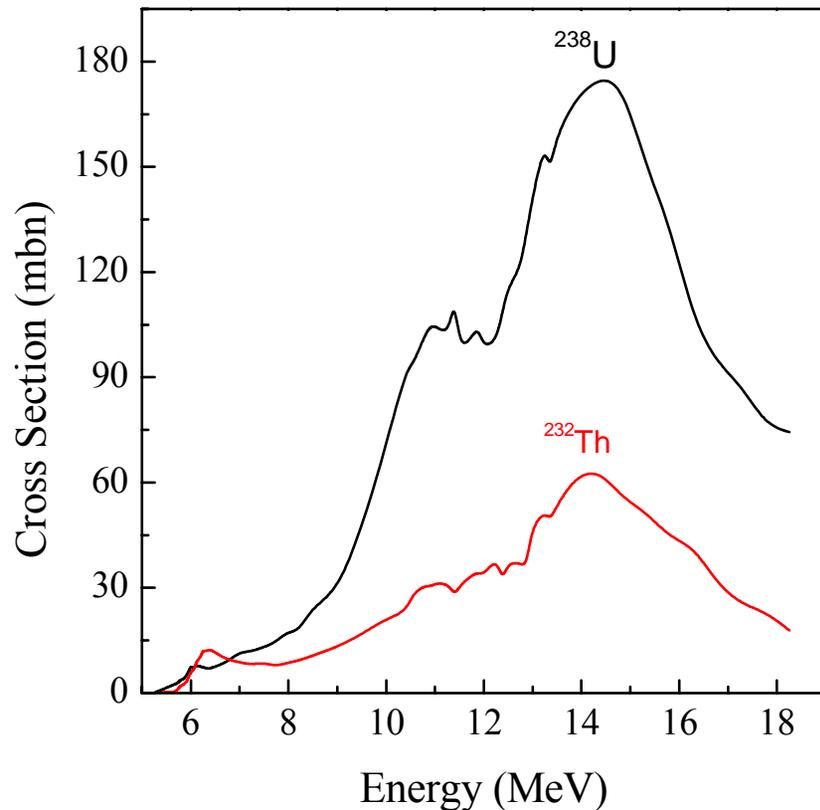
Prompt to Delayed Neutron Ratio Identifies Fissionable Material



60 Hz Rep. Rate
1'' ^{238}U

- Preliminary Data
- Expect Differences in Ratios
- Heavily Dependent on Non-Fissionable Constituents
- **Need Accurate Prompt and Delayed Yield Measurements**

Differences in (γ, f) Cross Sections Identifies Fissionable Material



- Measure Relative (γ, f) Yield (e.g. Yield at Two Energies)
- Relative Yield Difference for Different Isotopes
- **Need Accurate Fission Cross Sections**
- **Need Accurate # Neutrons per Fission**

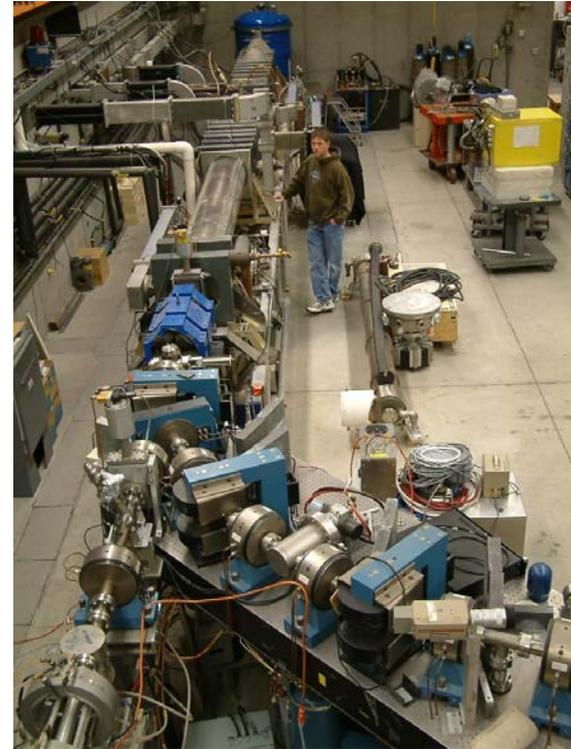
Data Needs for Materials Accountability

- **Fission Cross Sections**
 - γ -Ray Emission Data
 - **Prompt Energy Distribution**
 - Delayed Energy Distribution
 - NRF Level in ^{235}U , ^{239}Pu etc...
 - Neutron Emission Data
 - **Delayed Group Parameters**
 - Prompt Energy Distribution
 - **Number of Neutrons per Fission**
 - Fragment Distribution
 - **Photonuclear Cross Sections Poorly Known**
 - **Significant Overlap with DHS Data Needs**
- Data Needs**
- Mentioned Yesterday**
-
- ```
graph LR; DN[Data Needs] --> FCS[Fission Cross Sections]; DN --> PED[Prompt Energy Distribution]; MTD[Mentioned Yesterday] --> DGP[Delayed Group Parameters]; MTD --> NPF[Number of Neutrons per Fission];
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# Idaho Accelerator Center

- Electrons
  - 1.2 MeV DC Accelerator
  - 4 MeV LINACs
  - 10 MeV Induction Accel. (~10 kA)
  - 25 MeV LINAC
  - 45 MeV Short Pulse LINAC
- X-Rays
  - X-Ray Tubes: ~450 keV
  - Monoenergetic LCS: Energy ~30 keV
  - $^{137}\text{Cs}$  Sheppard Source: 13 Ci
- Ions
  - 2 MeV Van de Graff
- Neutrons
  - Electron LINACS
  - Variety of Sources

L Band Traveling Wave Linac



- Infrastructure
  - ~35,000 sq. feet Lab Space
  - Machine Shop
  - Electronics Shop

# Physics Department Starts New Ph.D. Program

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- Physics and Health Physics
- Doctor Of Philosophy in Applied Physics
  - Program Started in August 2005
- Student Population
  - Graduate: 67
  - Undergraduate: 100 (Doubled Since 2002)
- Faculty Positions 17
  - Almost Exclusively Nuclear

# Identification of Fissionable Material is Also Critical

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- Nonproliferation
  - Verify Contents of Stored Weapons Material
  - Prevent Weapons Material Theft
- Fuel Reprocessing
  - Prevent Material Diversion
  - Quality Assurance of Material Streams
- Waste
  - Allows Handling Decisions of Legacy Waste
- Security
  - $^{238}\text{U}$  and  $^{232}\text{Th}$  are Naturally Occurring and Used in Industry
  - Imperative to Eliminate Positives that are not a Security Concern

# Recent Paper in Applied Physics Letters

APPLIED PHYSICS LETTERS 86, 254104 (2005)

## Time dependence of delayed neutron emission for fissionable isotope identification

M. T. Kinlaw and A. W. Hunt<sup>a)</sup>

*Idaho Accelerator Center, Idaho State University, Pocatello, Idaho 83209-8263 and Department of Physics, Idaho State University, Pocatello, Idaho 83209-8106*

(Received 9 March 2005; accepted 17 May 2005; published online 17 June 2005)

The time dependence of delayed neutron emission was examined as a method of fissionable isotope identification. A pulsed bremsstrahlung photon beam was used to induce photofission reactions in  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{239}\text{Pu}$  targets. The resulting delayed neutron emission was recorded between irradiating pulses and is a well-known technique for fissionable material detection. Monitoring the decay of delayed neutron emission between irradiating pulses demonstrates the ability to not only detect the presence of fissionable materials, but also to identify which fissionable isotope is present. © 2005 American Institute of Physics. [DOI: 10.1063/1.1953874]

The ability to detect and identify fissionable materials has become increasingly important for nonproliferation and homeland security applications.<sup>1-3</sup> Several active evaluation techniques have been developed that focus on the detection of delayed neutrons that are emitted after fission reactions, which are induced by an external pulsed high-energy photon or neutron source.<sup>1,2,4,5</sup> The timescale for the emission of these delayed neutrons ranges from hundreds of milliseconds to over 50 s.<sup>6-10</sup> Since the fission reaction is essentially the only available reaction that emits neutrons on these time scales, the detection of delayed neutrons is a clear and reliable indication that fissionable isotopes are present, making it possible to detect concealed fissionable materials. How

$$R = N_0 \sum_{i=1}^6 \frac{\lambda_i \alpha_i}{1 - e^{-\lambda_i \Delta t}} e^{-\lambda_i t}. \quad (1)$$

Here,  $N_0$  is the total precursor yield per irradiation pulse and  $\Delta t$  is the period between these pulses. Hence, the decay rate of the resulting neutron emission gives a signature of the fissionable isotope, if the group parameters are sufficiently different for the isotopes of interest. The group parameters have been measured by many researchers for a variety of fissionable isotopes. Tuttle<sup>9</sup> evaluated the data available prior to 1975 and compiled a list of recommended group parameter values. Due to the importance of delayed neutrons for

# System Being Developed for Field Deployment



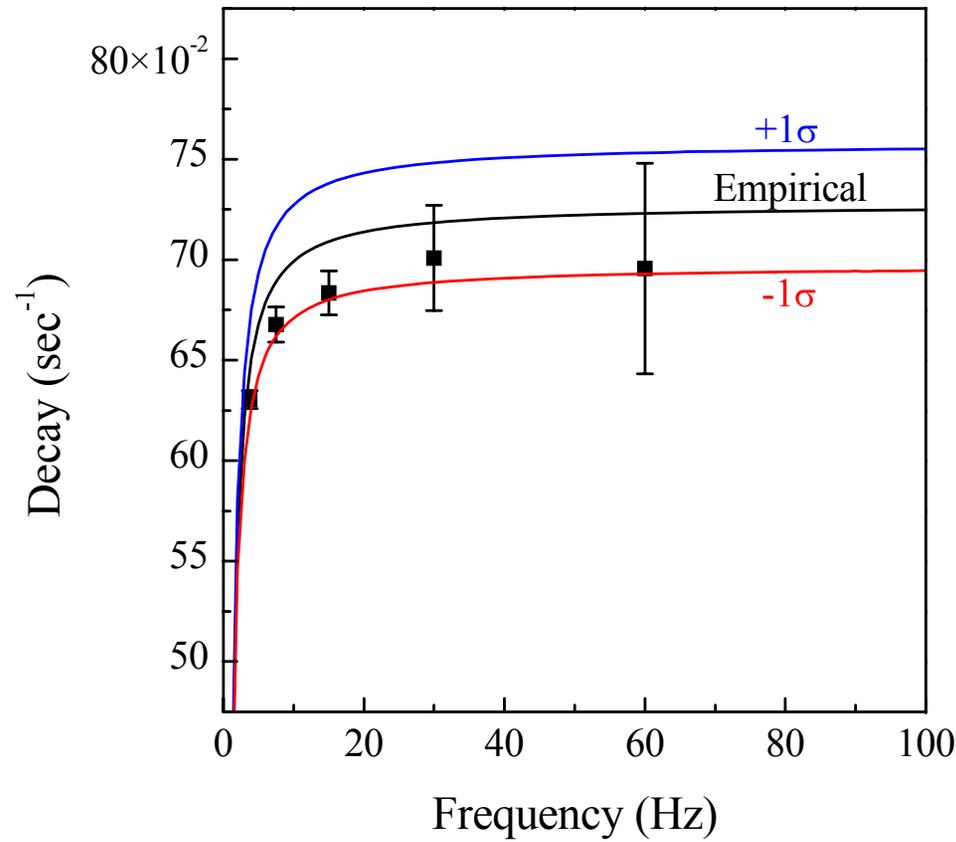
- Collaboration between PACECO, Casper-Philips and ISU
  - Scan Entire 40 ft. Container in ~60 sec.
  - Construct True Prototype System
  - Can Easily Add Identification Technology

# Integrated Detection of $\gamma$ -rays and Neutrons Needed

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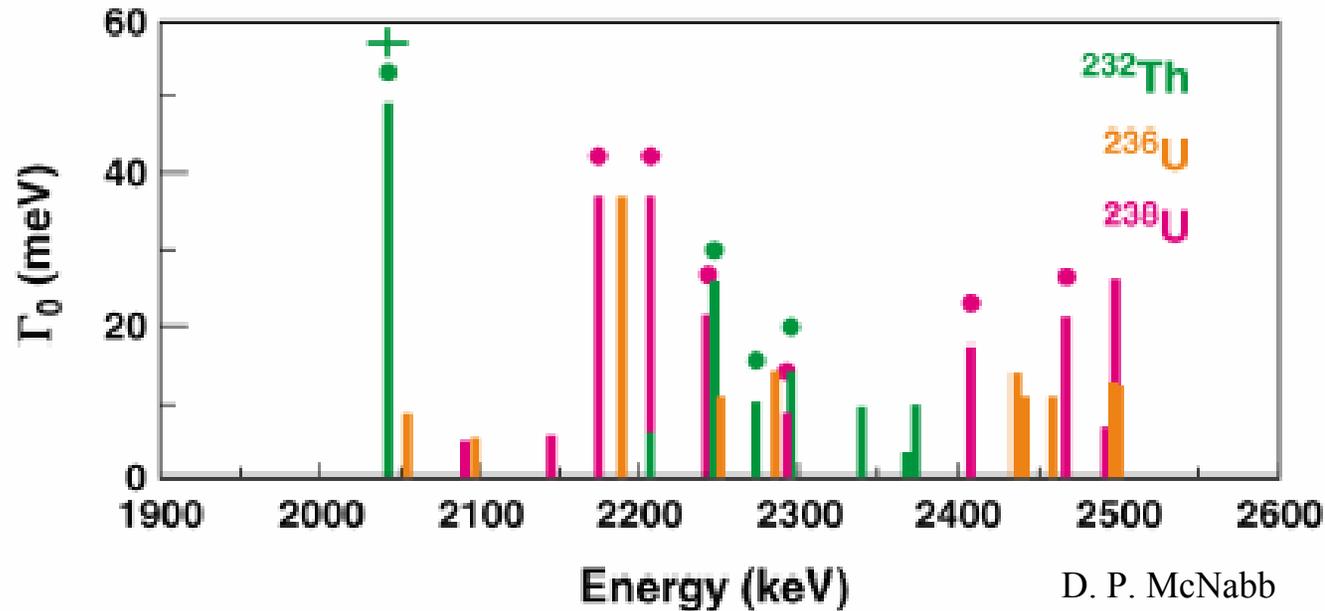
- Neutrons
  - Advantages
    - Low Background
    - “Easy” to Detect in Accelerator Environments
    - Fissionable Isotopes “Only” Delayed Neutron Emitters
    - Highly Penetrating through High-Z Materials
  - Disadvantages
    - Shielded/Absorbed by Low-Z Materials (e.g. Hydrogen)
    - Material Specificity Lacking (i.e. Difficult to Identify Material)
- $\gamma$ -rays
  - Advantages
    - Excellent Material Specificity
    - Highly Penetrating through Low-Z Materials
    - Abundant Production
  - Disadvantages
    - High Background
    - Difficult to Detect in Accelerator Environments
    - Easily Shielded/Absorbed by High-Z Materials (e.g. Lead)

# Identify at “Any” Accelerator Repetition Rate



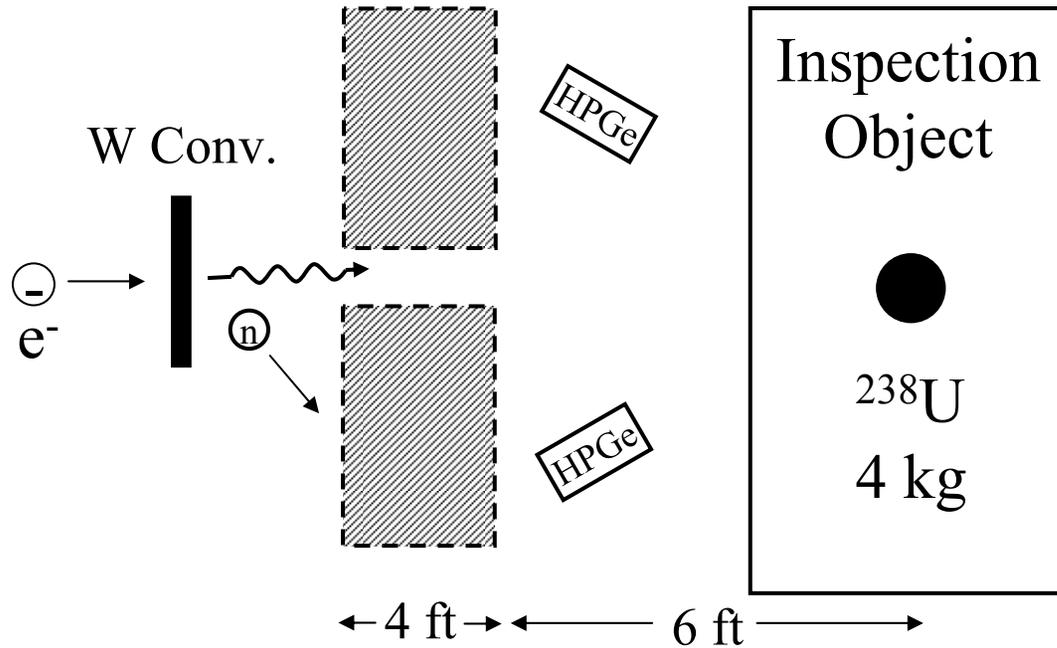
20 MeV Bremsstrahlung  
7.5 to 60 Hz Rep. Rate  
1.1 kg <sup>238</sup>U  
5 min Data Collection

# NRF Integrated Cross Sections are Small



- Largest Width in  $^{238}\text{U}$   $\Gamma_0 \approx 30$  meV
  - Integrated Cross Section  $\sim 7 \times 10^{-5}$  bn·MeV
  - Integrated Cross Section Proportional to Width
- Widths Smaller in  $^{235}\text{U}$   $\Gamma_0 \approx 8$  meV (Estimate Only)
  - Strengths Fragmented in Odd Isotopes (Larger Level Density)
- Fission Width in  $^{238}\text{U}$   $\sim 7$  MeV
  - Maximum Cross Section  $\sim 180$  mbn
  - Integrated Cross Section  $\sim 1$  bn·MeV

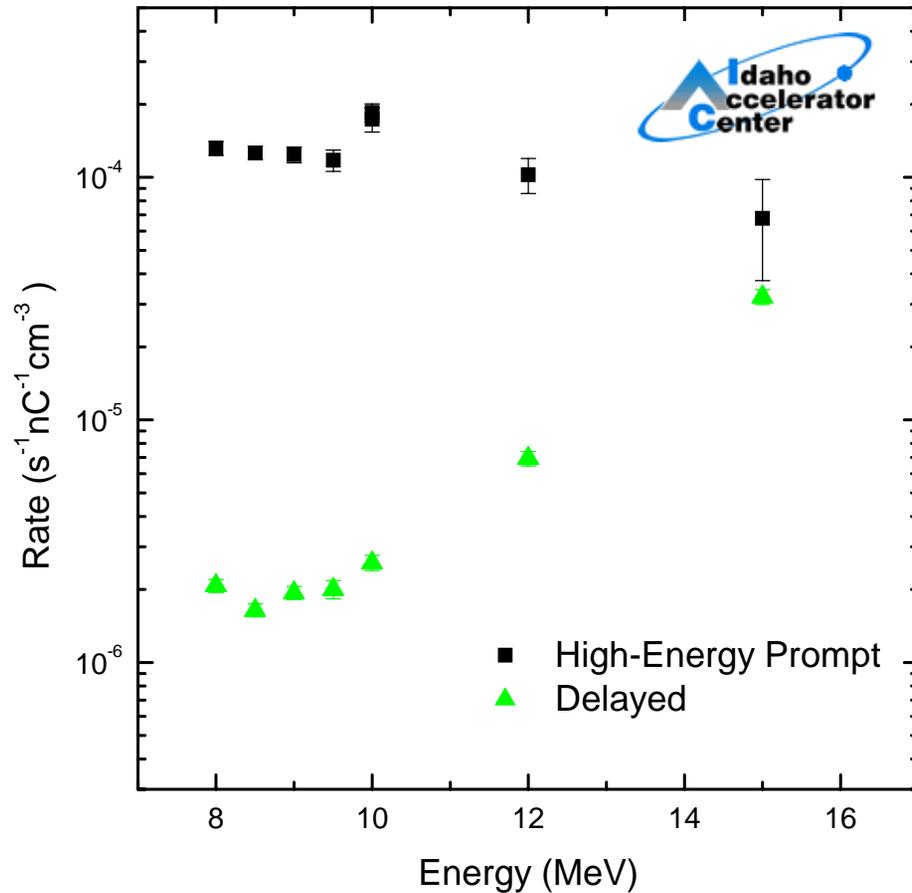
# Simulations Suggest NRF Reaction Rates Are Small



- Photon Spectrum
  - Calculated in MCNPX
  - No  $(\gamma, \gamma')$  Absorption
- Over Estimate  $(\gamma, \gamma')$  Rates

- Reaction Rate for Assumed  $^{235}\text{U}$  Line  $\Gamma \approx 8$  meV
  - $0.16 \text{ s}^{-1} \cdot \mu\text{A}^{-1} \cdot \text{g}^{-1}$  @ 5 MeV
  - $2 \times 10^{-5} \text{ s}^{-1} \cdot \mu\text{A}^{-1} \cdot \text{g}^{-1}$  after Solid Angle for 7 cm HPGe Detector
- Reaction Rate for  $(\gamma, f)$  in  $^{238}\text{U}$  Line
  - $1.6 \times 10^6 \text{ s}^{-1} \cdot \mu\text{A}^{-1}$  @ 5.5 MeV
  - $7.4 \times 10^9 \text{ s}^{-1} \cdot \mu\text{A}^{-1}$  @ 10 MeV

# Prompt Neutrons Will be More Sensitive



- Rate Normalized to Detector Volume

- 100 Times More High-Energy Neutrons
- Lower-Energies Better for Prompt (Beneficial for Rad Safety?)
- Higher Energies Better for Delayed