TUNL Photofission: Measurements of Prompt and Delayed Neutrons from Photofission of $^{235}\text{U}$, $^{238}\text{U}$ and $^{239}\text{Pu}$

**Photofission at HIGS:**
- Prompt Fission Neutron Energy Spectra (PFNS) at $E_\gamma > 10$ MeV
- Prompt Fission Neutron Energy Spectra (PFNS) at $E_\gamma < 10$ MeV
- Delayed Fission Neutron Energy Spectra at $E_\gamma < 10$ MeV
- Cumulative Fission Product Yields (FPY) with $T_{1/2} > 1$ s
- Independent FPY mass distribution measurements

Presentation by: Calvin R. Howell
Duke University and TUNL
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Active Interrogation of Cargo with Photons: neutron and gamma detection

A. Prompt neutron emission from nuclei from actinide nuclei, e.g., $^{238}$U, $^{235}$U, $^{239}$Pu

Inclusive ($\gamma$, n) double differential cross-section measurements:

$$\sigma(\gamma,\text{tot}) = \sigma(\gamma,\text{fn}) + \sigma(\gamma,\text{n}) + \sigma(\gamma,\text{2n}) + \sigma(\gamma,\text{pn})$$

B. Delayed neutron and gamma emission from fission of $^{238}$U, $^{235}$U, $^{239}$Pu

Tunable $\gamma$-ray source

Courtesy LLNL

During the Primary Scan, EZ-3D™ generates a series of slices that reconstructs the contents of the cargo in 3D

“Nuclear Photonics” Scanner

Concept by Passport
Photofission: Differential Measurements of Prompt and Delayed Neutrons

Program: Countering Weapons of Mass Destruction (CWMD) Research and Development Division
Funding Opportunity Title: Countering Weapons of Mass Destruction: Academic Research Initiative (ARI)
Funding Opportunity Number: DHS-20- CWMD-077-001

(1) **Photofission Nuclear Data for Active Interrogation**
Collect data on photofission for Uranium-235, Uranium-238, and Plutonium-239 at relevant x-ray energies to improve nuclear data for photofission cross sections, angular distribution, and energy spectra. Submit the improved data to relevant databases to improve modeling capabilities for DHS application and to provide better nuclear data for broader nuclear community.

(2) **Near Real-Time Biothreat Sensor Using Fluid-Based Mechanisms**

(3) **Alternative Signatures for Biological Weapons of Mass Destruction**

**Grant Number:** 20CWDARI00035-01-00
PI: Calvin R. Howell, Duke University
Co-PI: Mohammad W. Ahmed, North Carolina Central University
Co-PI: Jack A. Silano, Lawrence Livermore National Laboratory
Project Period: September 1, 2020 – August 31, 2025
Project Focus:
- Photon-induced fission of $^{235}\text{U}$, $^{238}\text{U}$ and $^{239}\text{Pu}$
- $E_\gamma = 6 - 10 \text{ MeV}$

A. Data Products and publications

A.1. Prompt total neutron ($\gamma, fn$) + ($\gamma, xn$) emission measurements: Energy and angle double differential neutron yield data at 4 photon beam energies: $E_\gamma = 10, 8, 7$ and 6 MeV

A.2. Prompt fission neutron emission measurements: Energy and angle double differential neutron yield data at one photon beam energy: $E_\gamma = 10 \text{ MeV}$

A.3. Delayed fission neutron emission measurements: Energy differential neutron yield data at one photon beam energy: $E_\gamma = 8, 9$ or $10 \text{ MeV}$

B. Education of young physicists

- Provide research opportunities for graduate and undergraduate students
- Emphasis on enabling postdocs to develop into independent scientists
- Give special attention to recruiting students from underrepresented ethnic groups and women

Lessons

1st round measurements and data analysis

2nd round measurements
Research Team

Senior Investigators

Calvin Howell, PI
Duke University
Prompt Fission Neutron Spectrum (PFNS)

Werner Tornow, SI
Duke University
Delayed Neutron Emission

Forrest Friesen, Postdoc
Duke University
PFNS

Mohammad Ahmed, Co-PI
NC Central University
PFNS and Delayed Neutron Emission

Jack Silano, Co-PI
LLNL
Delayed Neutron Emission

Ronald Malone(a),
Postdoc
Duke University
PFNS

Krishichayan(b), Scientist
Duke University
PFNS

GE Hitachi Nuclear Energy

Sean Finch, Scientist
Duke University
PFNS and Delayed Neutron Emission

Collin Malone(c)
Duke University
PFNS

(c) Postdoc, SRNL

Ethan Mancil
Duke University
PFNS

Postdocs and Research Scientists

Graduate Students

Postdoc at LLNL

(c) Postdoc, SRNL

Delayed Neutron Emission
High Intensity Gamma-ray Source (HIGS)

Most intense Compton γ-ray source in the world

Features that enable basic and applied research

• Wide beam energy range: 1 to 100 MeV
• Selectable beam energy spread (by collimation)
• High beam intensity on target (>10⁷ γ/s @ ΔE/E = 5%)
• E < 20 MeV; ~10³ γ/s/eV
• >95% beam polarization (linear and circular)

1.2 GeV Storage Ring FEL

Beam time structure:
T = 180 ns and Δt = 300 ps
Prompt Neutron Emission ($\gamma$, fn) + ($\gamma$, xn): 1st round measurements

Gamma-ray Energy Selection

Bremsstrahlung distribution at 10 and 9 MeV

Target = 3-mm thick tungsten

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>$S_n$ (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{238}\text{U}$</td>
<td>6.2</td>
</tr>
<tr>
<td>$^{239}\text{Pu}$</td>
<td>5.6</td>
</tr>
<tr>
<td>$^{235}\text{U}$</td>
<td>5.3</td>
</tr>
</tbody>
</table>

Runs (2020 and 2021): E$_\gamma$ (MeV)

(1) Oct. 26 – Nov. 6: beam time = 106 hrs
- PFNS: E$_\gamma$ = 10, 8, and 7 MeV
- Delayed neutron measurement R&D: E$_\gamma$ = 7 MeV

(2) Apr. 28 – May 6: beam time = 115 hrs
- PFNS: E$_\gamma$ = 6, 5.5, and 5.3 MeV
Experimental Setup

- Gamma-ray beam properties
  - Energy Resolution ~7% FWHM
  - Circular beam polarization
  - Pulse period 179.2 ns
- Detect neutrons with an array of liquid scintillator (BC501A/EJ301) neutron detectors capable of pulse-shape-discrimination (PSD)
  - Shield with ~3mm of lead on front faces
  - Neutron energy by time-of-flight (TOF)
  - Digitize all signals
- Measure incident beam flux with the dual fission chamber mounted upstream of the target
Experiment Technique: Signal-to-background

**Neutron Detector Setup:**
- An array of 28 liquid organic scintillators with PSD capability
- Detectors in two planes: horizontal and vertical, 12 each with 6 at other $\phi$ angles
- Detector dimensions: 5” dia. x 2” length
- $\theta = 22.5^\circ$ to $157.5^\circ$, $\Delta \theta = 22.5^\circ$
- Flight path (target to detector center) = 100 cm
- Data analysis pulse-height threshold = 60 keVee

**Gamma-ray Beam Parameters:**
- Circular beam polarization
- Beam collimator diameter = 0.75”; $\Delta E_g/E_g \approx 4\%$ (FWHM)
- Beam pulse width = 0.3 ns; Beam pulse period = 179 ns

<table>
<thead>
<tr>
<th>Target</th>
<th>Diameter (mm)</th>
<th>Mass (mg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{235}\text{U}$</td>
<td>12.35</td>
<td>223</td>
</tr>
<tr>
<td>$^{238}\text{U}$</td>
<td>12.42</td>
<td>442</td>
</tr>
<tr>
<td>$^{239}\text{Pu}$</td>
<td>12.40</td>
<td>467</td>
</tr>
</tbody>
</table>

![Graph showing pulse height and PSD](image-url)
- The detectors have an energy and threshold-dependent efficiency which must be applied to determine the cross section measured counts.
- The energy spectrum from each detector is processed independently and fit with a maxwellian spectrum.
  - Depending on the \((\gamma, n)\) contribution, a gaussian may be added to the fit.
- We present neutron yields in units of cross section.
  - For prompt photofission neutrons this is \(\langle \nu \rangle \sigma_{(\gamma,f)}\).
  - For photoneutrons this is \(\sigma_{(\gamma,n)}\).

### Efficiency

- **GEANT4**
  - 100 keVee
- **PTB**
  - 60 keVee

### PRELIMINARY

**Efficiency**

- **E_n (MeV)**

**Efficiency Data**

- **\(E_{g} = 10.0 \text{ MeV}\)**
  - \(q = 90 \degree\)
  - \(f = 90 \degree\)

**235U(\gamma, xn)**

- \(E_{\gamma} = 10.0 \text{ MeV}\)
- \(\theta = 90 \degree\)
- \(\phi = 90 \degree\)
Example: Prompt Neutron Energy Spectrum, $^{238}$U at $E_\gamma = 7.0$ MeV

$^{238}$U, $S_n = 6.15$ MeV
$E_\gamma = 7.0$ MeV
60 keVee
$0 = 112.5^\circ$
$\phi = 0^\circ$
PSD $n/\gamma$ cut applied
$E_{\text{thr}} = 60$ keVee
($E_n \gg 0.5$ MeV)

$N(E) = A \frac{4E}{\pi T^3} e^{-E/T}$

PRELIMINARY
Example: Prompt Neutron Energy Spectrum, $^{239}$Pu at $E_\gamma = 7.0$ MeV

- Example for $^{239}$Pu with 7 MeV incident photons:
  - $^{239}$Pu can be moved into $\frac{1}{2}$ or $\frac{3}{2}$ excited state
  - Allowing for neutrons up to $l=2$, all possible neutrons above detection threshold are shown at right
    - Peak near 1.3 MeV is from decay(s) into band 1
    - Peak near 700 keV is from decay(s) into band 2

$^{239}$Pu, $S_n = 5.65$ MeV

$E_\gamma = 7.0$ MeV

60 keVee

PRELIMINARY
### Preliminary Results of 1st round measurements (tabular)

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Energy (MeV)</th>
<th>Mean ($\gamma$,fn) (mb)</th>
<th>+/- stat (mb)</th>
<th>Maxwellian Temp (MeV)</th>
<th>+/- stat (MeV)</th>
<th>other ($\gamma$,n) (mb)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{239}\text{Pu}$</td>
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<td>0.6</td>
<td>0.2</td>
<td>3.2</td>
<td>1.9</td>
<td>0.7</td>
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<tr>
<td>$^{239}\text{Pu}$</td>
<td>5.5</td>
<td>12.0</td>
<td>3.4</td>
<td>2.2</td>
<td>0.8</td>
<td>7.9</td>
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<td>4.7</td>
<td>1.5</td>
<td>0.1</td>
<td>33.0</td>
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<td>105.0</td>
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<td>1.3</td>
<td>0.1</td>
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<tr>
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<td>0.0</td>
<td>72.4</td>
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<tr>
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<td>19.0</td>
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<td>1.7</td>
<td>0.4</td>
<td>12.9</td>
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<td>21.3</td>
<td>2.6</td>
<td>1.6</td>
<td>0.2</td>
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<td>32.4</td>
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<tr>
<td>$^{235}\text{U}$</td>
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<td>7.8</td>
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<tr>
<td>$^{235}\text{U}$</td>
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<td>23.3</td>
<td>8.2</td>
<td>2.0</td>
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<td>23.7</td>
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<tr>
<td>$^{235}\text{U}$</td>
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<td>34.9</td>
<td>9.3</td>
<td>1.8</td>
<td>0.4</td>
<td>74.3</td>
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<tr>
<td>$^{235}\text{U}$</td>
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<td>64.6</td>
<td>15.0</td>
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<td>0.3</td>
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<tr>
<td>$^{235}\text{U}$</td>
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<td>441.0</td>
<td>36.0</td>
<td>1.2</td>
<td>0.0</td>
<td>139.3</td>
</tr>
</tbody>
</table>
Preliminary results of 1st round measurements (graphical)

- We have measured differential cross sections for prompt neutron production from $^{238}\text{U}$, $^{239}\text{Pu}$, and $^{235}\text{U}$ at low energies
- Simulation work is in progress for target self shielding
- Push analysis of the differential neutron energy spectrum below 1.0 MeV
- The 2nd round measurement campaign will use a neutron detector array with ~x6 larger solid angle coverage

![Estimated Neutrons from Photofission](image1)

![Lower Bounds on Neutrons not from Photofission](image2)
Scintillator light output measurements at proton recoil energies below 1.0 MeV

Setup for scintillator light output measurements in the tandem lab at TUNL:
- Pulsed neutron beam
- Nearly mono-energetic neutron beams: $^2$H(d,n), $^7$Li(p, n)
- Collimated beam
Upgraded Neutron Detector Setup for PFNS Measurements

- Soccer ball design: up to 30 detectors
- Flight path = 30 – 60 cm (L = 42 cm)
- At L = 42 cm, Array dΩ = π sr
- Beam time at HIGS scheduled for May 25 – June 13, 2023
Summary

• First round of measurements of Prompt Fission Neutron Spectra (PFNS) completed
  • Double differential cross-section measurements of $E_n$ and angle made on $^{235}\text{U}$, $^{238}\text{U}$, $^{239}\text{Pu}$ at $E_\gamma = 5.3 - 10.0$ MeV
  • Differential cross-section data, $d\sigma(\theta, E_n)/d\Omega dE_n$, for the ($\gamma$, fn) reaction measured for each isotope at several energies below 10.0 MeV
  • Differential cross-section data, $d\sigma(\theta, E_n)/d\Omega dE_n$, for the ($\gamma$, fn) reaction measured for each isotope at several energies below 10.0 MeV
  • Determined lower limit of the differential cross section, $d\sigma(\theta, E_n)/d\Omega dE_n$, for the ($\gamma$,n) reaction for each isotope at several energies below 10.0 MeV

• Challenge is to push PFNS data analysis down to a minimum $E_n$ of about 0.5 MeV
• Preparing manuscript reporting results of 1st round measurements
• Second round of PFNS measurements scheduled for May 2023
• Preparation of the experiment for the double differential (time and $E_n$) delayed neutron energy spectrum is underway with goal for performing measurements by end of 2023
QUESTIONS