Measurement of Short-Lived Fission Product Yields for ^{237}Np via γ-ray Spectroscopy

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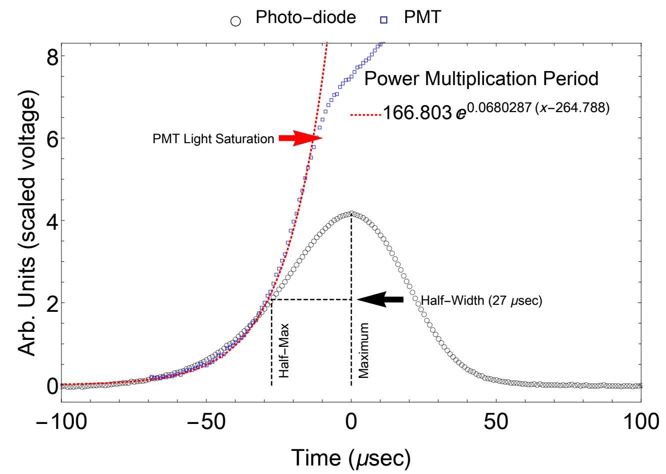
Motivation

- Accurate Fission Product Yields (FPYs) are important for basic and applied science
  - FPYs from a fission neutron spectrum is of particular interest
- Short-lived ($t_{1/2} \sim \text{minutes} - \text{hours}$) FPYs are difficult to measure
  - Separation methods require time
- High resolution γ-ray spectroscopy
  - Count the entire sample
  - Simultaneous measurement of FPYs for many isotopes
  - No need to take time to do chemistry/separation

Experimental Setup

- Four ^{237}Np samples were irradiated in GODIVA critical assembly
  - Neptunium Nitrate ($NpO_2(NO_3)$) sealed in Quartz tubes
  - Masses: 6.0(1), 10.6(7), 20.3(2), and 44.3(5) mg
  - Neutron Flux: $\sim 3 \times 10^{14}$ neutrons/second
  - Irradiation time: $\sim 50\ \mu s$ (FWHM)

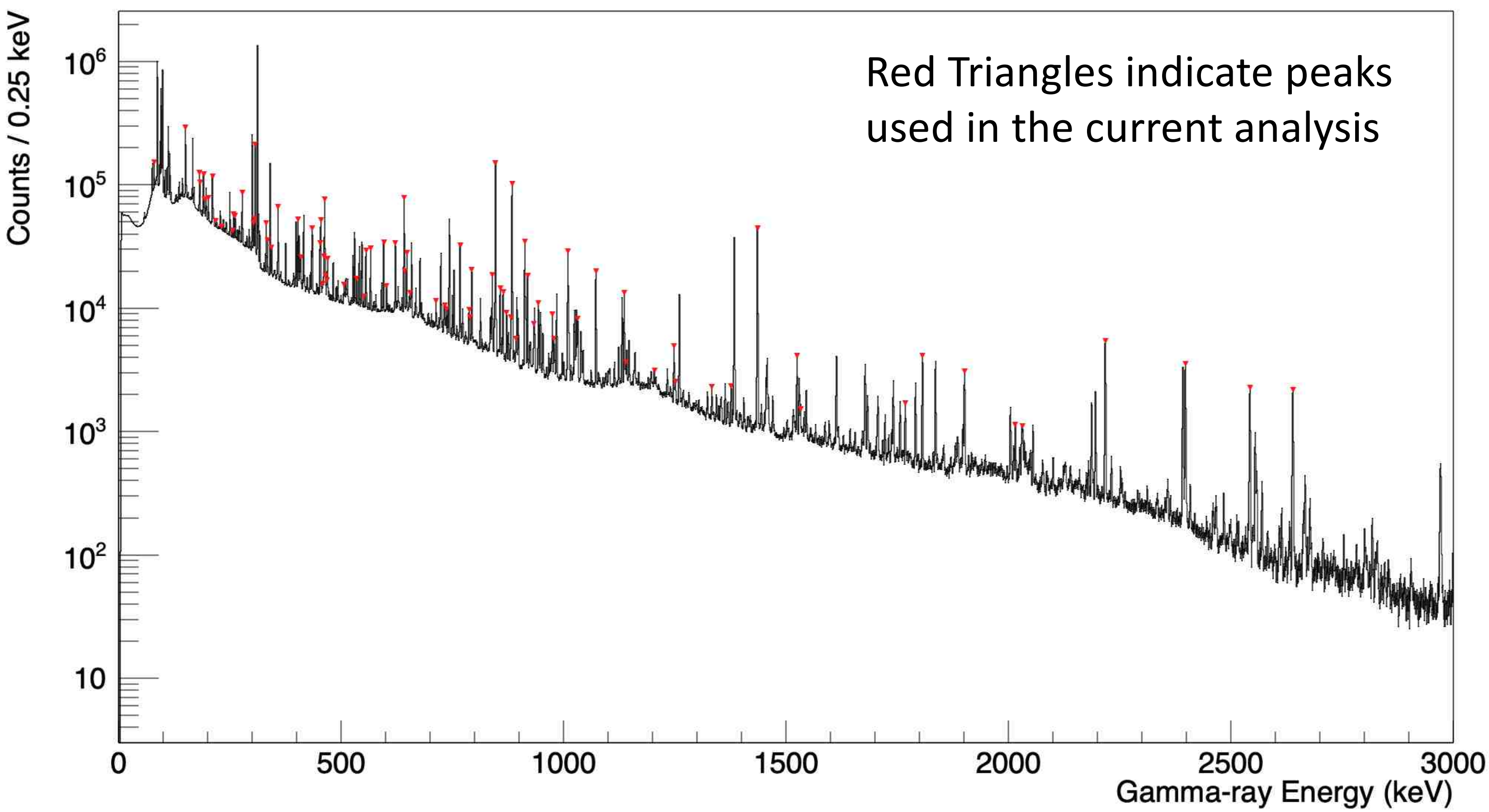


- Samples were retrieved and delivered to γ-ray counting setup $\sim$50 minutes after irradiation

- Counting continued for 7 days following irradiation
  - Short Timescale for first 3 hours
    - 10.6(7) mg sample
  - Long Timescale for next 7 days
    - 26.3(3) mg (6.0 + 20.3) samples

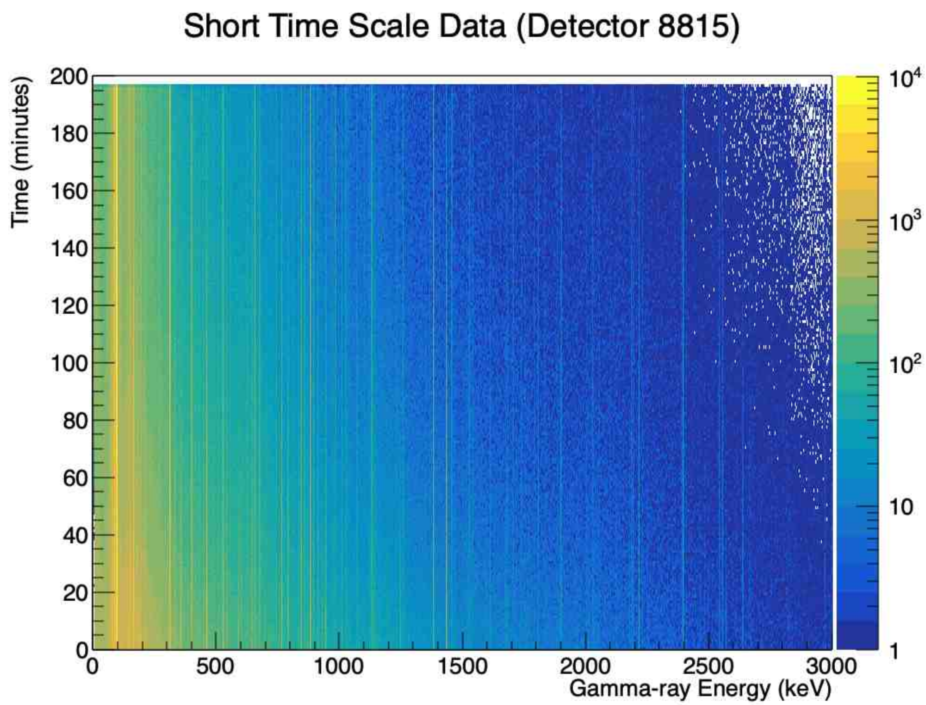
Data Analysis : Full Sum Spectrum

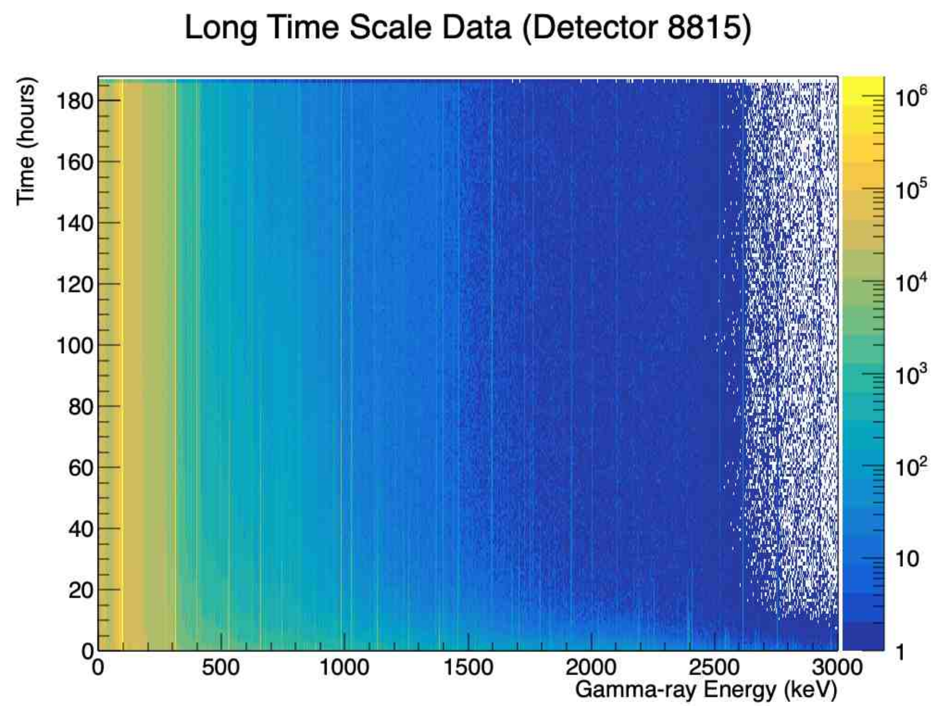
- The entire γ-ray spectrum from the long timescale count



Data Analysis : Time Dependent Spectra

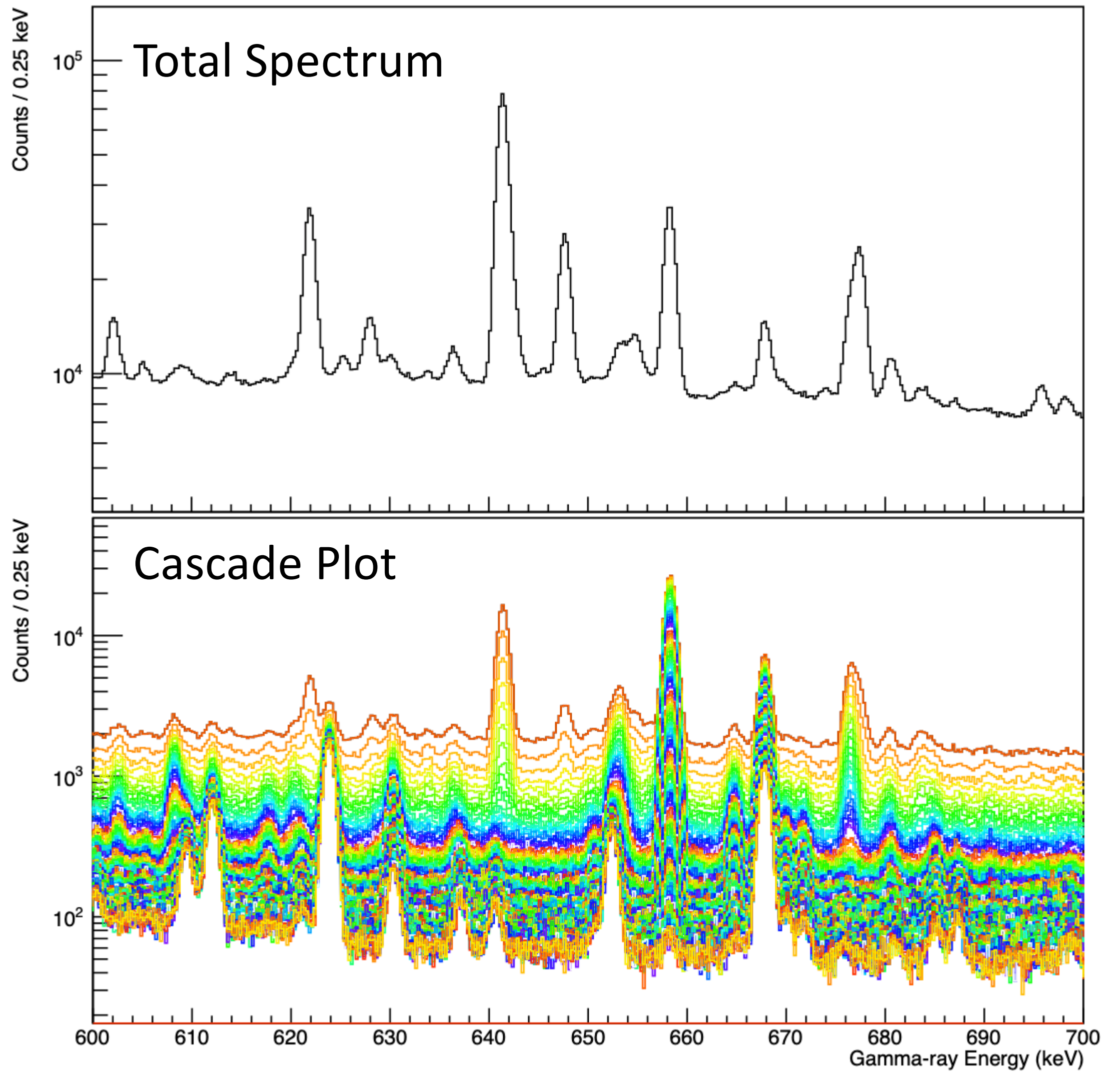
- Parse data into time-binned matices
- Fit a peak in each in time bin and plot the intensity versus time (decay curve)
  - Use half-life and γ-ray energy to identify isotope
- Use decay curve to extract the activity of the isotope immediately after irradiation





Data Analysis : Cascade Plots

- Used to build γ-ray 'input deck'
  - List of expected γ-rays with energy and time windows
- Easily identify time dependence of γ-ray spectrum
- Helps identify interfering γ-rays



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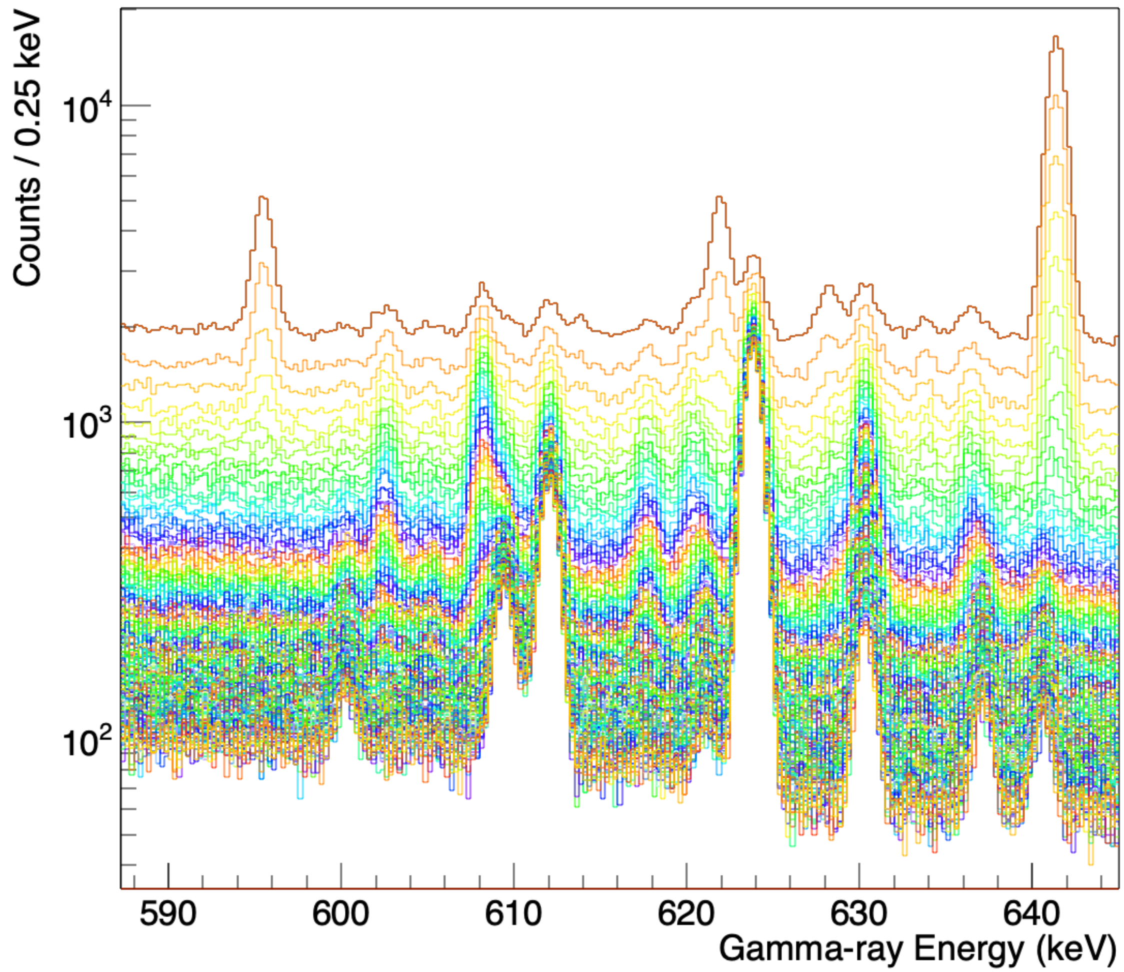


Results : Example ^{93}Y

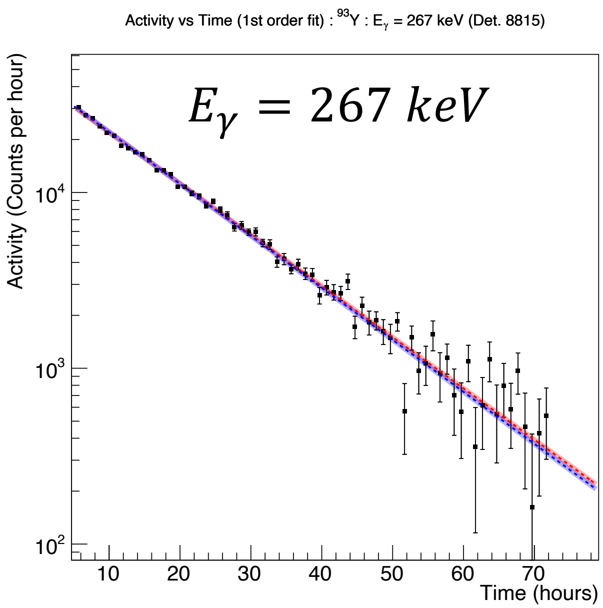
- Observed 3 'clean' γ-rays from the decay of ^{93}Y
- Extrapolate Decay Curve fits back to irradiation time : A_0
  - Correct for DAQ livetime, detection efficiency, and self-attenuation

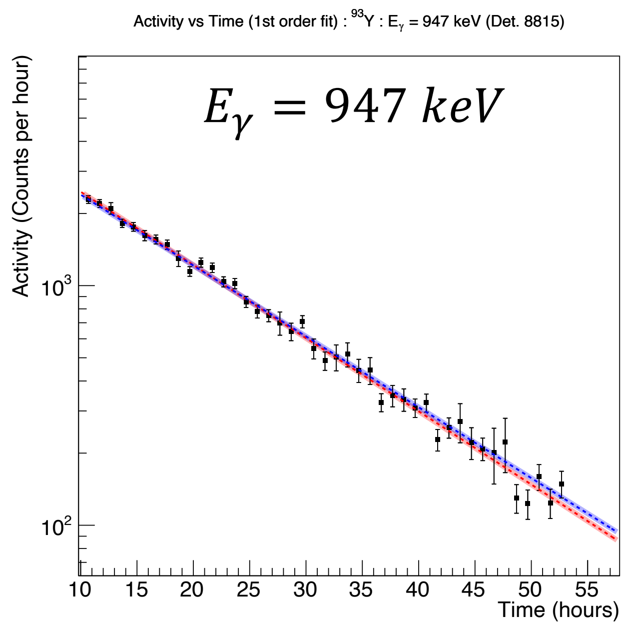
$$Y = \frac{A_0\, t_{1/2}}{\ln(2)\,\Gamma\, N_f}$$

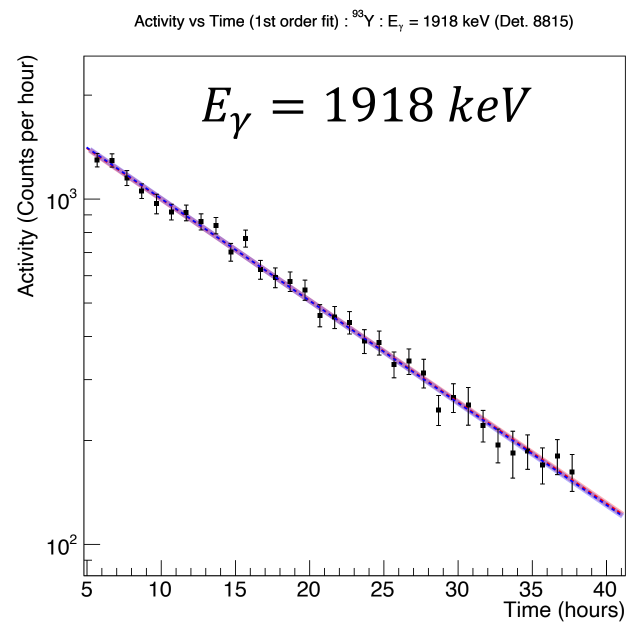
Γ : *Branching Ratio*

N_f : *Number of Fissions*

$$C(t_1, t_2) = \int_{t_1}^{t_2} A(t)dt = \frac{A_0}{\lambda} e^{-\lambda t_2}(e^{\lambda \Delta t} - 1)$$







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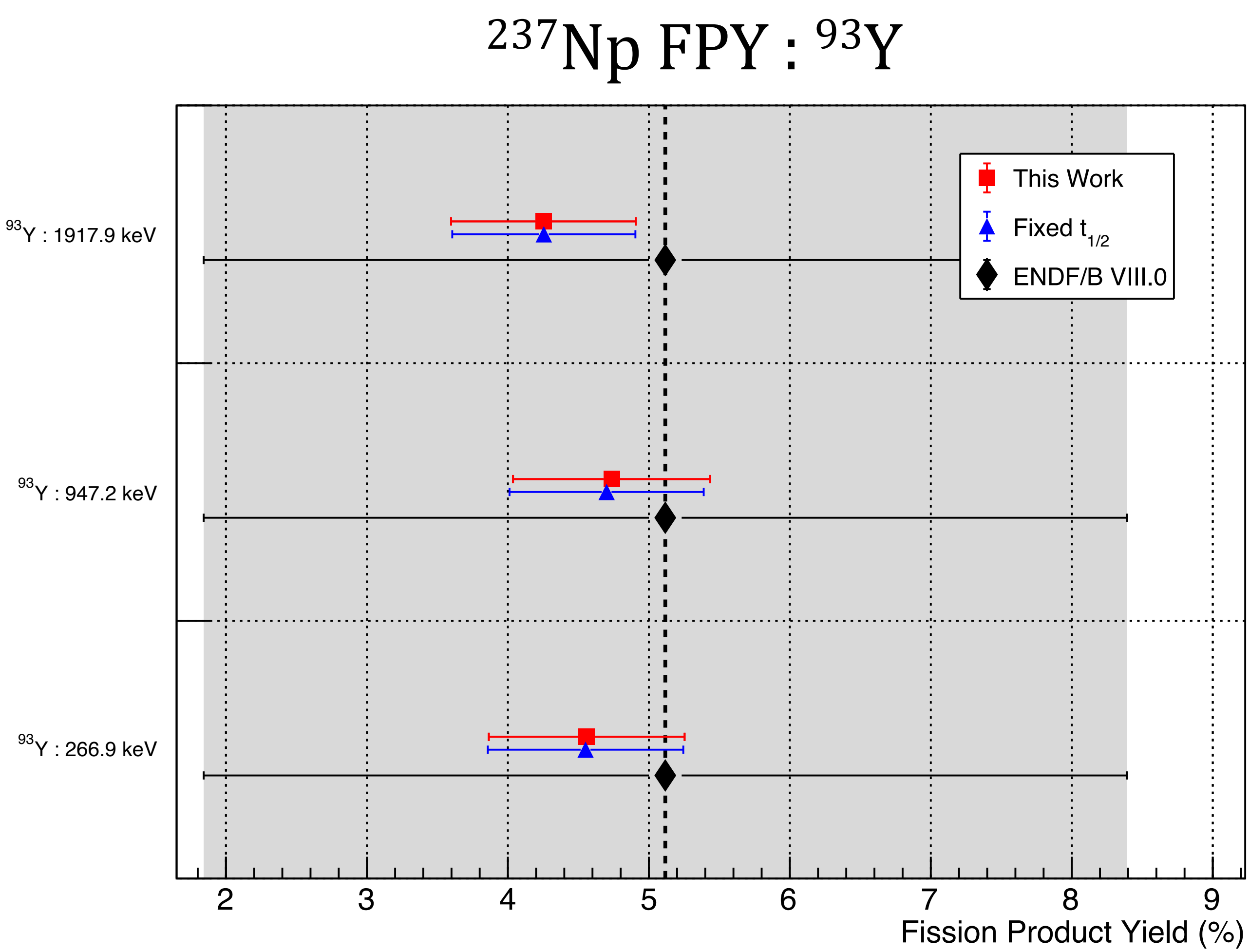

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Determining the Number of Fissions

- Four γ-rays from isotopes with well-known FPYs were selected as reference yields
  - Chosen to span a range of:
    - γ-ray energy
    - Isotope half-life
    - Isotope atomic number (Z)
    - Solid and Gas

STAYSL PNNL:
$7.2(3)\times10^{11}$ fissions/gram
(based on witness foil analysis performed by B. Pierson PNNL)

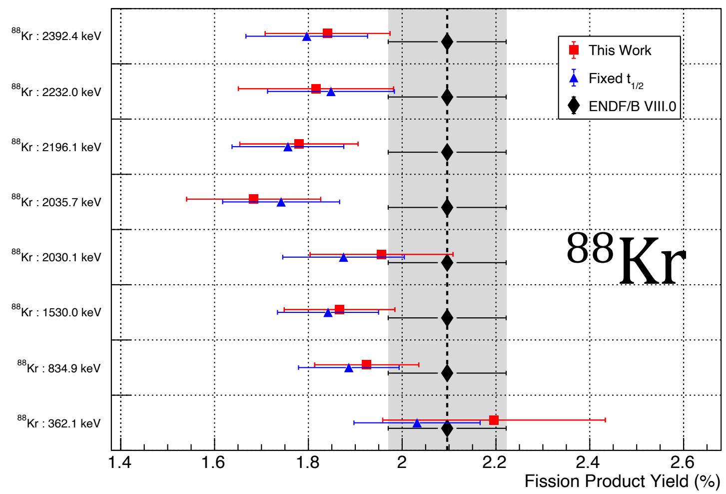
Number of Fissions in the Sample 2 (Short time-scale count)

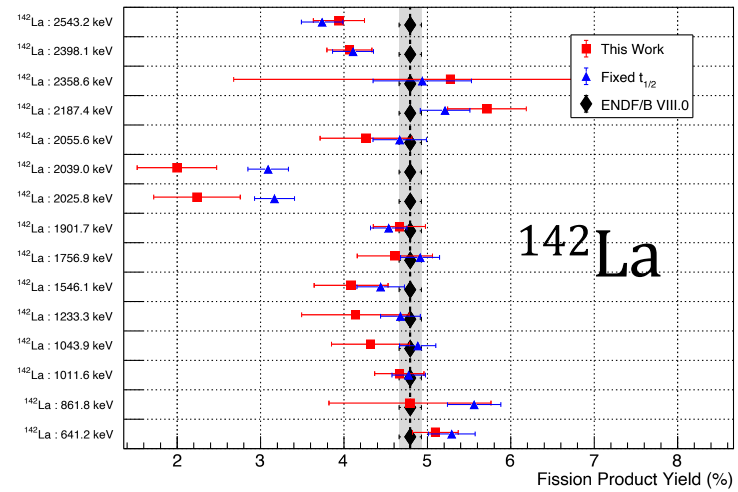
Isotope	E_γ (keV)	Γ	unc.	$t_{1/2}$ (m)	unc.	Ref. Y	unc.	N_f	unc.
^{87}Kr	402.6	0.50	0.03	76.3	0.5	1.74	0.05	7.75E+09	5.22E+08
^{92}Sr	1384.0	0.90	0.06	156.66	1.02	4.37	0.17	8.01E+09	4.33E+08
^{134}Te	566.1	0.186	0.01	41.8	0.8	4.41	0.18	8.08E+09	5.54E+08
^{135}I	1260.6	0.287	0.009	394.2	1.2	6.71	0.19	7.93E+09	3.74E+08
						Total Fissions =		7.94E+09	2.30E+08
						Fissions/gram =		7.49E+11	5.40E+10

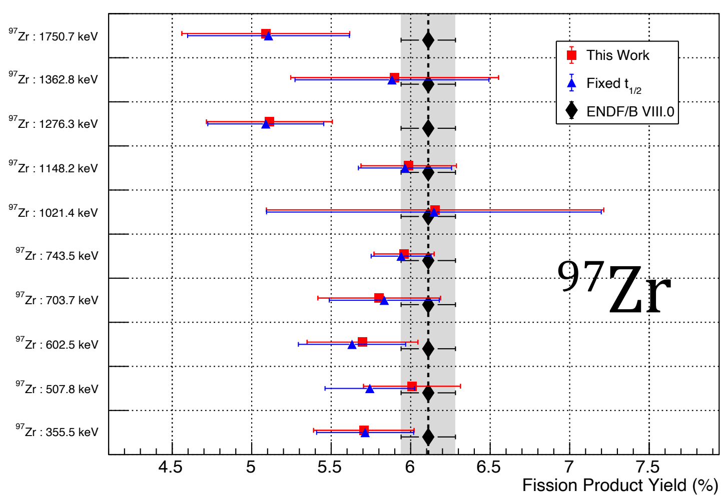
Number of Fissions in the Sample 1+3 (Long time-scale count)

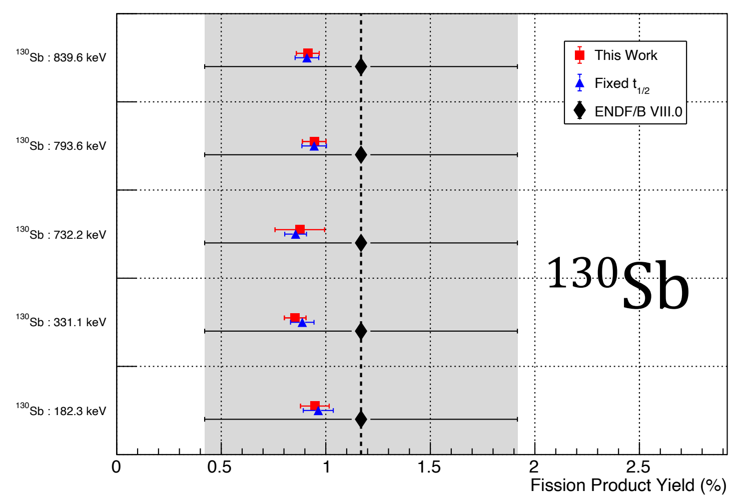
Isotope	E_γ (keV)	Γ	unc.	$t_{1/2}$ (h)	unc.	Ref. Y	unc.	N_f	unc.
^{87}Kr	402.7	0.50	0.03	1.2717	0.0083	1.74	0.05	1.99E+10	1.37E+09
^{92}Sr	1384.0	0.90	0.06	2.611	0.017	4.37	0.17	1.92E+10	1.51E+09
^{99}Mo	739.6	0.122	0.005	65.976	0.024	6.12	0.24	2.02E+10	1.18E+09
^{135}I	1260.6	0.287	0.009	6.57	0.02	6.71	0.19	1.91E+10	8.25E+08
						Total Fissions =		1.96E+10	5.80E+08
						Fissions/gram =		7.44E+11	2.36E+10

Results: 45 Isotopes/Isomers : 191 γ-rays



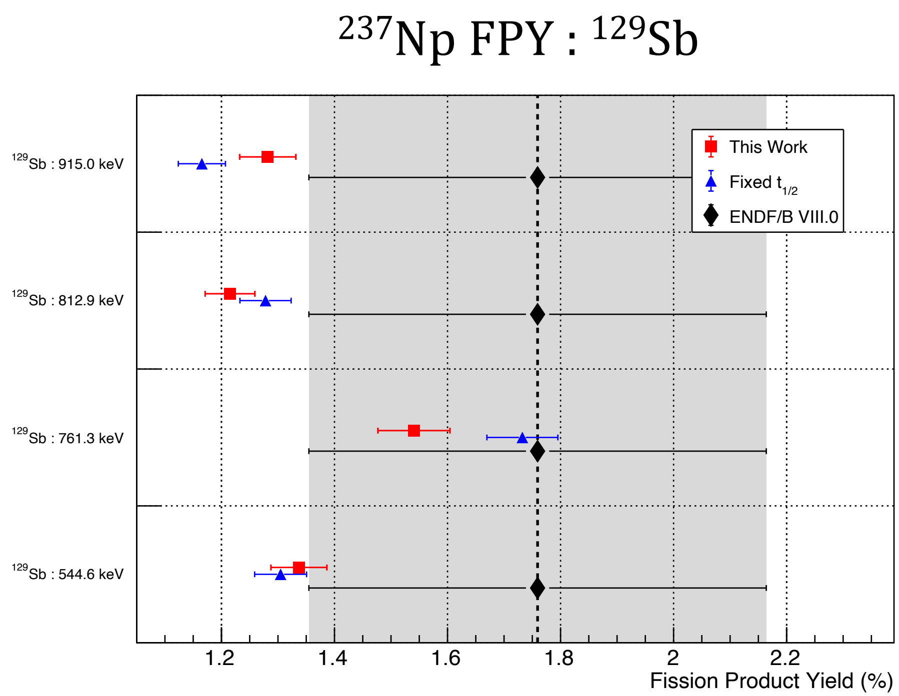






Results: Investigating Branching Ratios

- Example ^{129}Sb: Four γ-rays were observed
  - Three γ-rays give consistent result
  - One is a clear outlier -> Potential issue with branching ratio for this γ-ray?
- Requires further investigation
  - What is ENSDF FPY value based on?
    - Ex. Was it the 761 keV γ-ray?
  - What is the ENSDF branching ratio value based on?
- Potential to improve accuracy of nuclear data
- Highlight areas where future experiments could be focused
  - Investigate ^{129}Sb branching ratios



Conclusion

- Fission product yields have been measured for 45 unique isotopes/isomers
  - Using 191 γ-rays
  - More to come!
  - Currently investigating discrepancies in results (ex. ^{129}Sb)
    - Update branching ratios?
    - Take another look at experiments included in the ENSDF evaluation

- Full Results to be published in future Nuclear Data Sheet Article

- Results from this experiment will be compared to FPYs of ^{235}U, ^{238}U, ^{239}Pu, and ^{233}U
  - ^{235}U, ^{238}U, and ^{239}Pu irradiations have been completed
  - ^{233}U irradiation planned for early/mid 2021
  - Self-consistent FPY results for 5 actinides
    - All irradiations utilized GODIVA
    - All γ-ray count utilized the same experimental setup
    - All data will be analyzed/re-analyzed with the codes developed in this work

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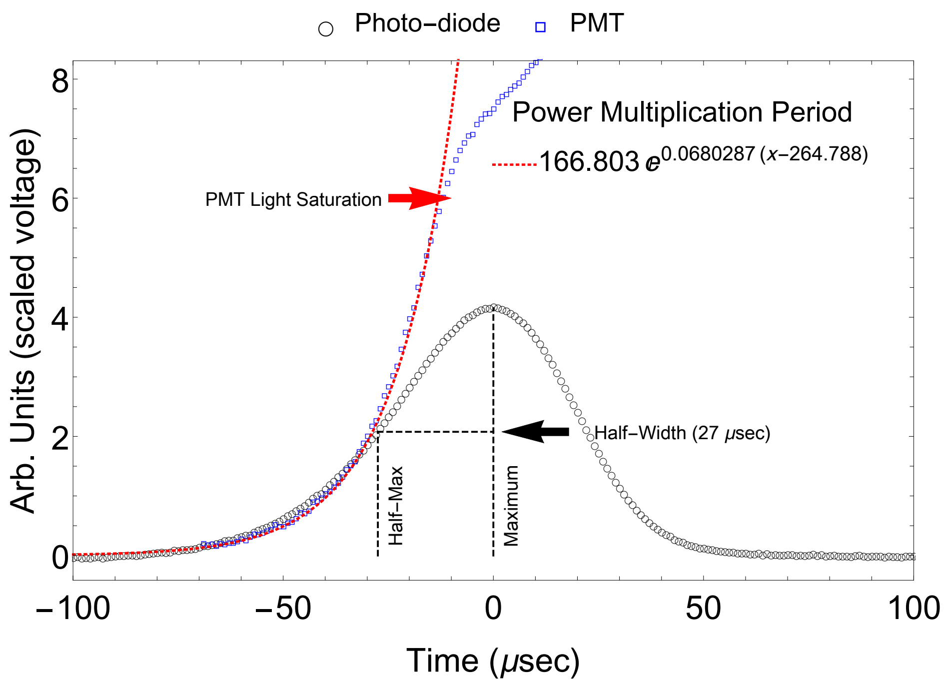
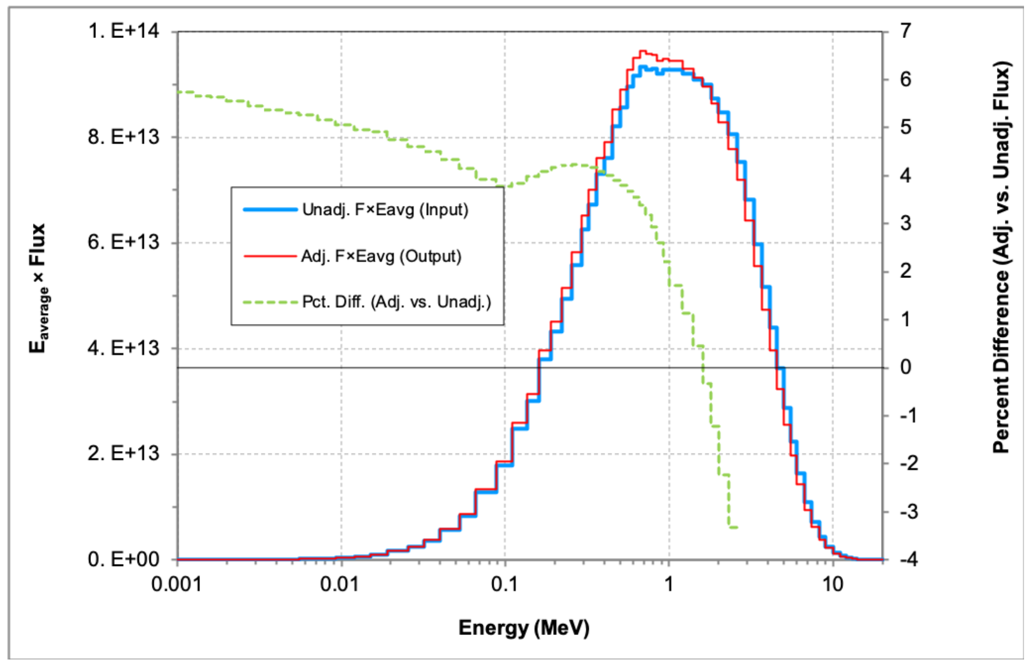
GODIVA Burst



Neutron Flux determined by witness foil analysis and STAYSL PNNL code



Figures generated by Bruce Pierson