Fission Product Chain Yield Measurements

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Fission Product Yield Measurements Supporting National Security

Objectives:

Provide improved measurements of energy integral and differential fission product yields and related activation cross sections in relevant neutron fields for major and minor actinides.

Relevance:

Fission product yields (FPY) represent an important nuclear fission observable for basic science and numerous applications. This and related work provides important experimental data to validate and improve differential nuclear data, nuclear physics modeling and application tools.

Current plans in the international nuclear data community are to produce a new fission product yield evaluation within the next five years (ENDF/B-VIII.1). The data collected under these efforts will directly feed into the new evaluation.

Collaborations:

Most of these experiments have been and will continue to be jointly conducted with PNNL and/or LLNL. We also have a new technical collaboration with researchers at Bruyères-le-Châtel under the NNSA-CEA Joint Agreement.

Approach:

Make use of critical assemblies and other neutron sources to irradiate well characterized samples. Samples are then analyzed by

- Radiochemical analysis of irradiated samples to determine relative ($R_i^{j,k}$) cumulative FPYs and associated reaction rates.

and/or

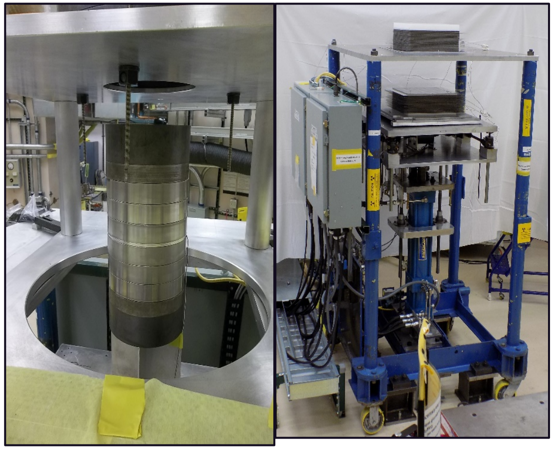
- Direct γ-ray counting of the irradiated samples to determine relative ($R_i^{j,k}$) cumulative FPYs and associated reaction rates.

In both cases we use fission chambers to convert relative FPYs to absolute ($Y_i^{j,k}$) FPYs.

Accomplishments/Results:

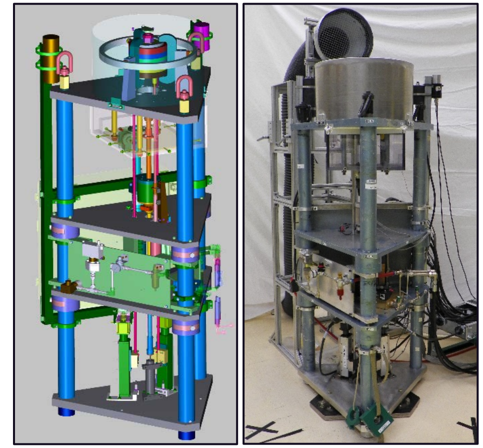
- Fission spectrum irradiations of ^{233}U, ^{235}U, ^{238}U, ^{237}Np and ^{239}Pu at NCERC. For each actinide, relative fission product yields were determined by radiochemical analysis.
- Monoenergetic irradiations of ^{235}U, ^{238}U and ^{239}Pu with collocated fission chambers at TUNL. Relative and absolute FPYs were determined by γ-counting.

Critical Assemblies at NCERC



Planet

- Light Capacity Vertical Lift Assembly
- 10^{13} fissions/g on samples, Variable Spectrum

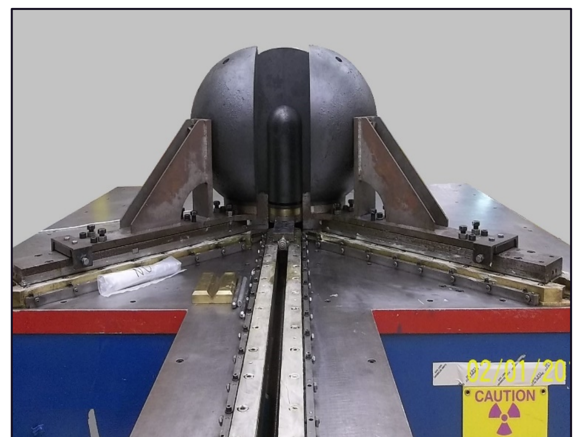


Godiva-IV

- 65.5 kg HEU (1.5% Mo by weight)
- Super-Prompt Critical Operations
- Short-Lived Fission Products
- 1-4 x 10^{16} Total Fissions, Hard Spectrum

Comet

- High Capacity Vertical Lift Assembly
- Supports Large Experiments
- 10^{13} fissions/g on samples, Variable Spectrum



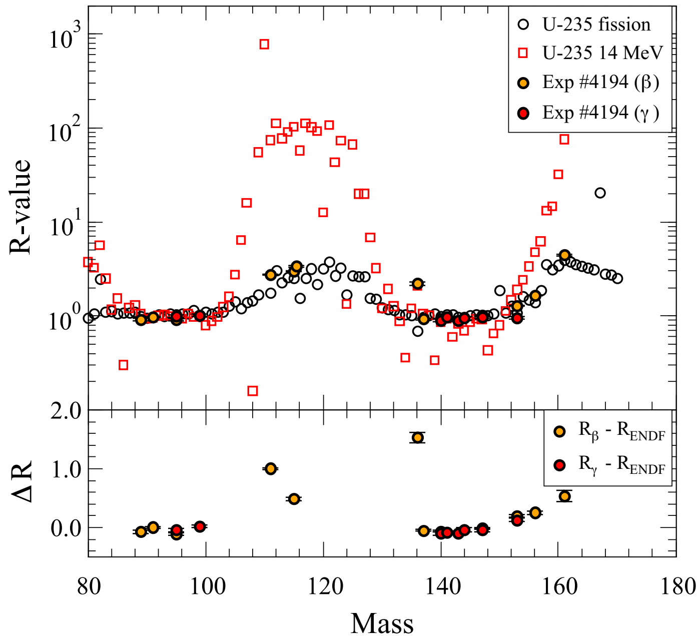
Flat-Top

- Assembly with either a 17.7 kg HEU core or a 6 kg WG Pu core. Reflected by ~1000 kg NU.
- 10^{13} fissions/g on samples, Hard Spectrum

NCERC Experiments to Date (and in the future)

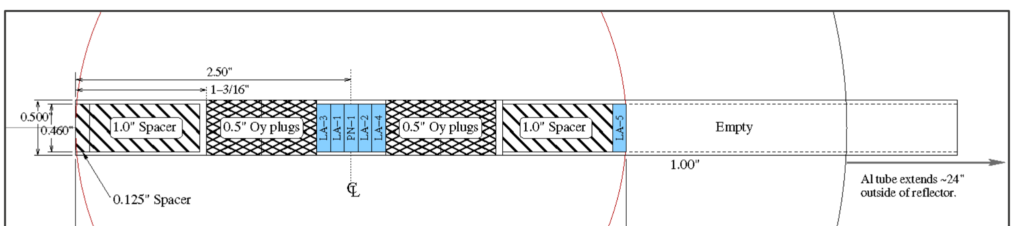
Assembly	Date	^{233}U	^{235}U	^{238}U	^{237}Np	^{239}Pu	R-values	Actinides	FC	PNNL	LLNL	Sponsor(s)
Comet/ZEUS	Sep-11		✓	✓		✓	✓	✓		✓		NA-10/80
Comet/ZEUS	Sep-12		✓	✓			✓	✓		✓		NA-22
Flattop (half)	Jun-13	✓	✓	✓			✓	✓		✓		NA-22
Flattop (half)	Aug-14		✓	✓	✓		✓	✓		✓		NA-22
Flattop (full)	Apr-15		✓	✓		✓	✓	✓		✓		NA-22
Planet (special)	Jun-16		✓	✓			✓	✓			✓	DTRA
Flattop (full)	Mar-17		✓	✓	✓		✓	✓			✓	DTRA
Planet (special)	Jul-17		✓	✓			✓	✓			✓	DTRA
Flattop (full)	Apr-17		✓	✓			✓	✓	✓	✓		NA-22
Flattop (full)	Apr-18		✓	✓	✓		✓	✓	✓	✓		NA-22
Planet (special)	Jun-18		✓	✓			✓	✓			✓	DTRA
Planet (special)	2019		✓	✓			✓	✓	?	✓	✓	NA-10/NCSP
Flattop (full)	2020		✓	✓			✓	✓	✓	✓		NA-22
Flattop (full)	2022		✓	✓		✓	✓	✓	✓	✓		NA-22

Results For Experiment #4194 (HEU)



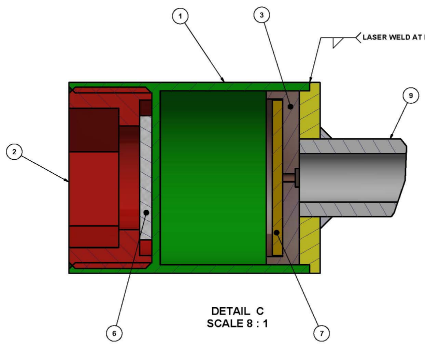
Experimentally determined R-values for ^{235}U in the center position of Flattop compared to R-values calculated from ENDF/B-VII.1 fission product yields for 0.5 and 14 MeV incident neutrons.

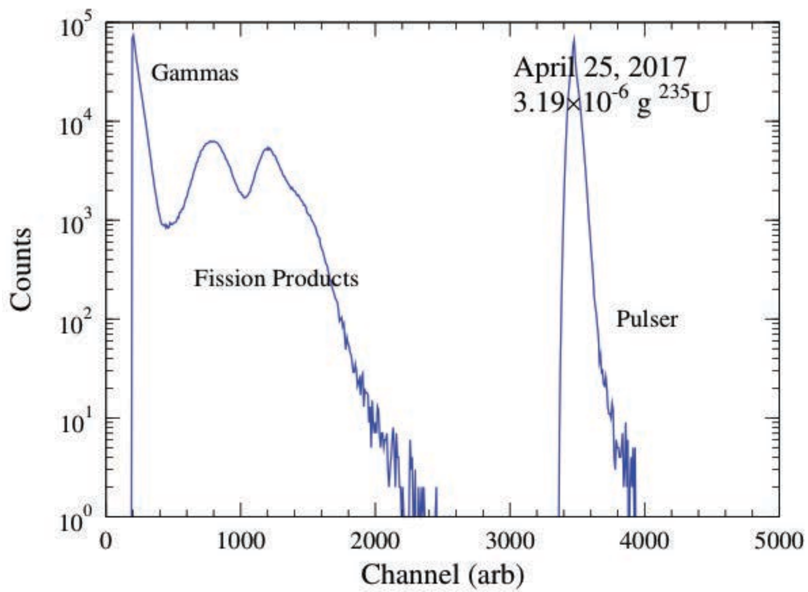
The lower panel shows the absolute difference between the values determined by β counting (black circles) and γ counting (red circles) and ENDF/B-VII.1.

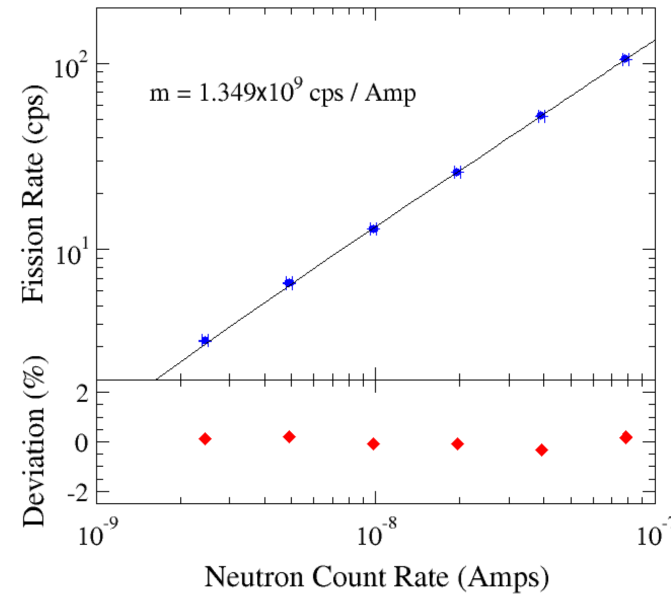


Outlier	ENDF/B	LANL	PNNL
^{111}Ag (β)	1.76	2.76	n/a
^{111}Ag (γ)	1.76	n/a	3.30
^{136}Cs (β)	0.69	2.22	n/a
^{136}Cs (γ)	0.69	n/a	2.47
^{153}Sm (β)	1.08	1.28	n/a
^{153}Sm (γ)	1.08	0.95	1.20

Prototype Fission Chamber Design and Testing



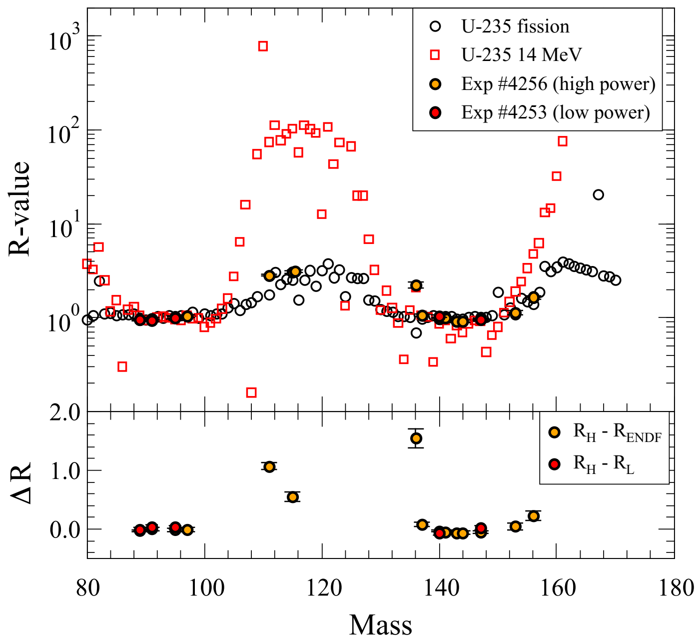


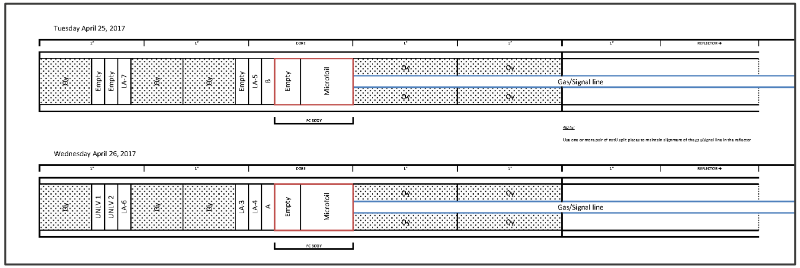


Observations:

- At low power levels the prototype FC provided excellent energy resolution given its small size.
- The gamma rate from the assembly fuel presaged issues with running the fission chamber at the power level required for full radiochemical analysis.

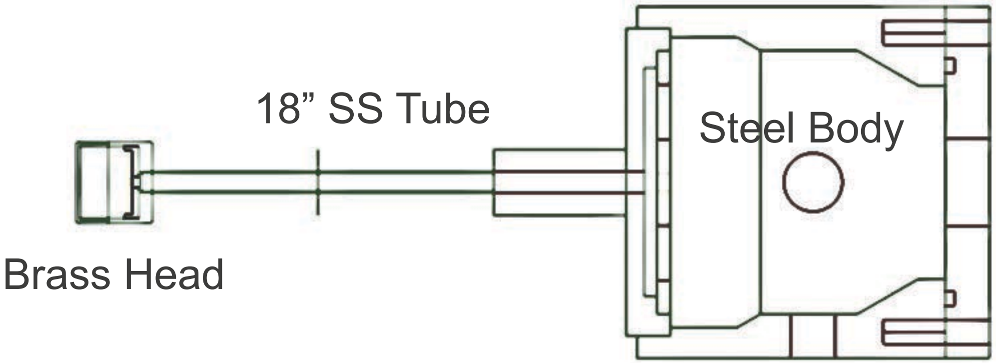
Results For Experiments #4253 and 4256 (HEU)

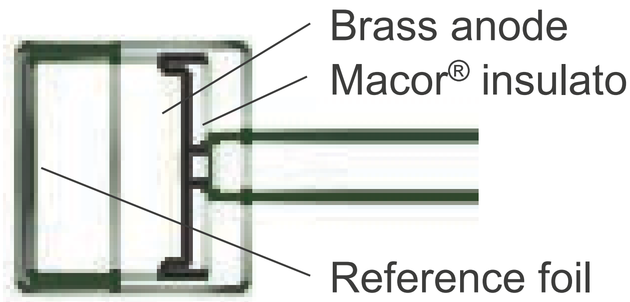


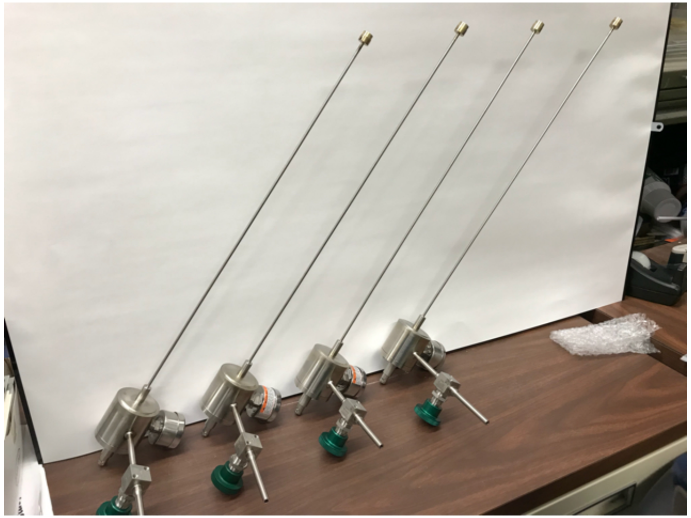


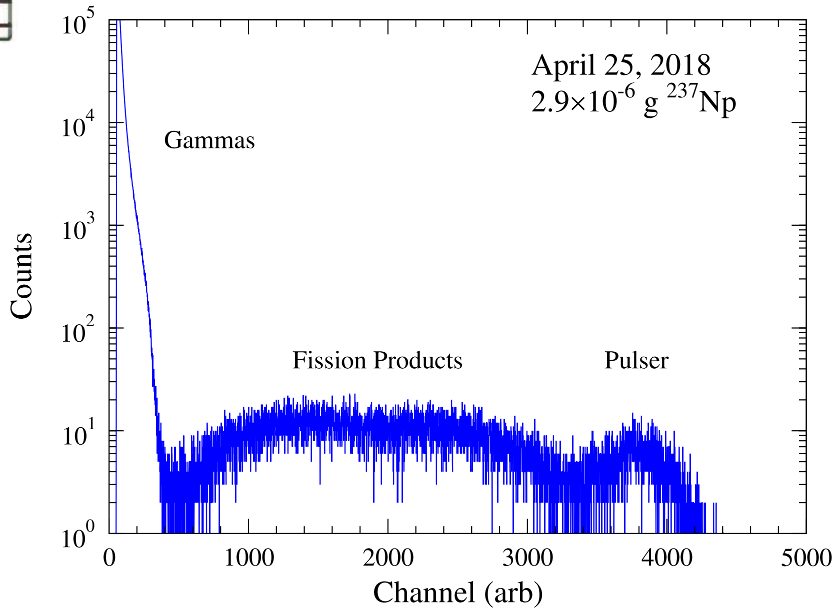
- Comparison of current R-value results with R-values calculated from ENDF/B-VII.0 cumulative fission product yields for fission of ^{235}U from 0.5 (fission) and 14 MeV neutrons.
- Difference, $\Delta R = R_H - R_n$, between R-values determined from the high-power run (H) and those calculated for ENDF fission (n = ENDF) and from the low-power run (n = L).
- There is very good agreement between both data sets and ENDF for peak yield fission products, where we expect to see little fluctuation with incident neutron energy. However, differences can be seen in the valley and high mass wing.

Mark II Fission Chamber

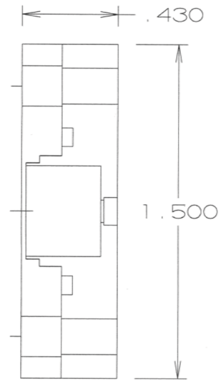


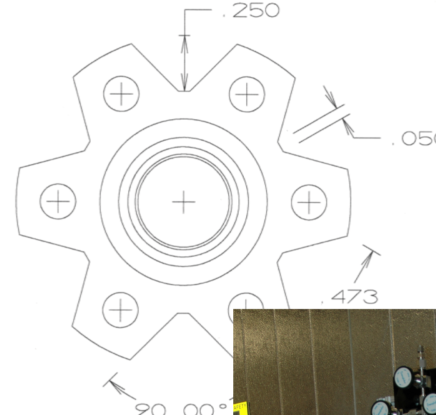


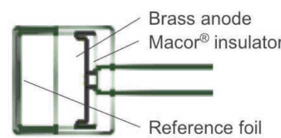




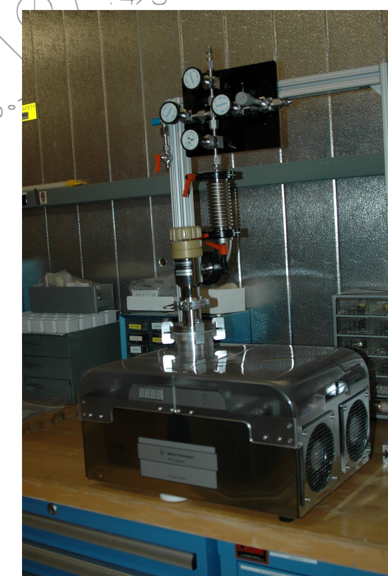
Tests at McClellan Nuclear Research Center (UC Davis)







Testing U(93) reference foils at MNRC in September



Work To Date

NCERC (LANL):

- Energy integral foil irradiations on the Comet, Flattop, Godiva and Planet assemblies.

  ^{233}U, ^{235}U, ^{238}U and ^{237}Np (^{239}Pu)
- Direct foil counting and radiochemical analyses to determine relative CFPYs.
- Developing fission chambers to determine absolute CFPYs.

CEA/Valduc (CEA-DAM):

- Energy integral foil irradiations on the CALIBAN reactor.

  ^{235}U and ^{239}Pu
- Collocated fission chamber and direct foil counting to determine absolute FPYs

Path Forward

NCERC (LANL/CEA-DAM):

- Integrate LANL fission chamber with CEA fast pre-amplifier.
- Energy integral foil irradiations on the Flattop, and Godiva assemblies.

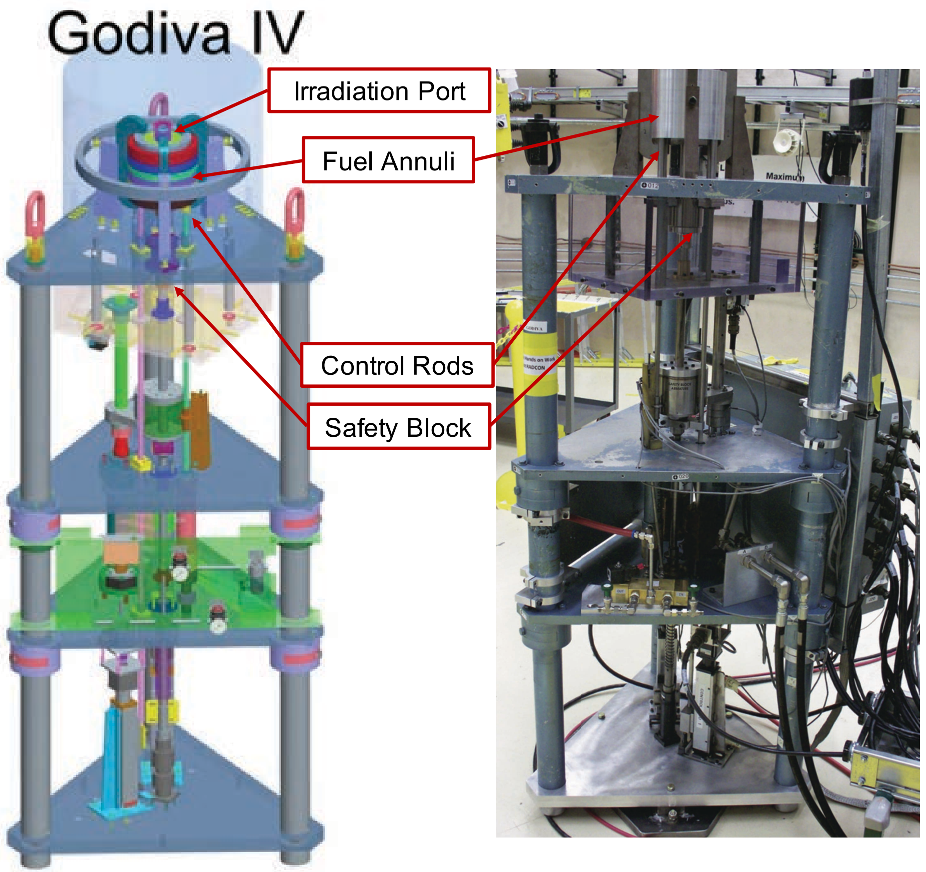
  ^{235}U, ^{238}U and ^{239}Pu
- Direct foil counting and radiochemical analyses to determine relative CFPYs.
- Collocated fission chambers to determine absolute CFPYs.

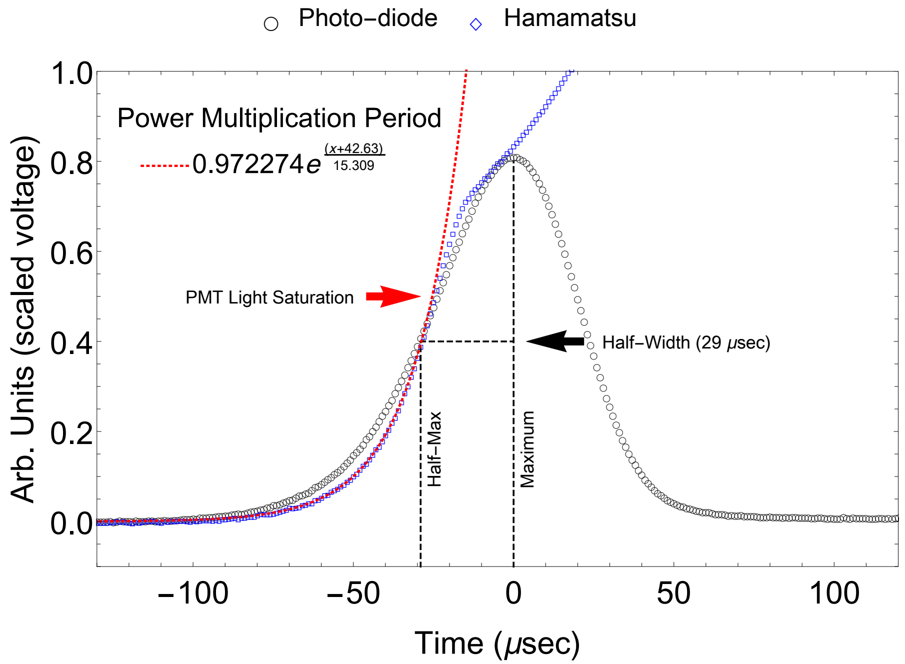
Farther forward:

- Move on to other actinides.

Irradiation

1. Sample Loaded
2. Reactor Pulse
3. Optical Response Recorded:
   - PMT
   - 2x Photo-diodes
4. Radiation Decay Period
   - ~20-30 min.





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Uncertainty Analysis

Rough Uncertainty Budget:

- Fissions—5-7%
  - Could be 2-3% with track etch counting or fission chambers
- Branching Ratios
  - 1-15%, isotope dependent
- Half-lives
  - Generally <2%
- Efficiency—0.6-3%
- Self-Attenuation~2%
- Counting statistics—1-10%

$$\sqrt{0.05^2 + 0.05^2 + 0.02^2 + 0.015^2 + 0.02^2} = 0.078 \approx 8\%\ \text{error}$$

$$\sqrt{0.03^2 + 0.05^2 + 0.02^2 + 0.015^2 + 0.02^2} = 0.066 \approx 7\%\ \text{error}$$

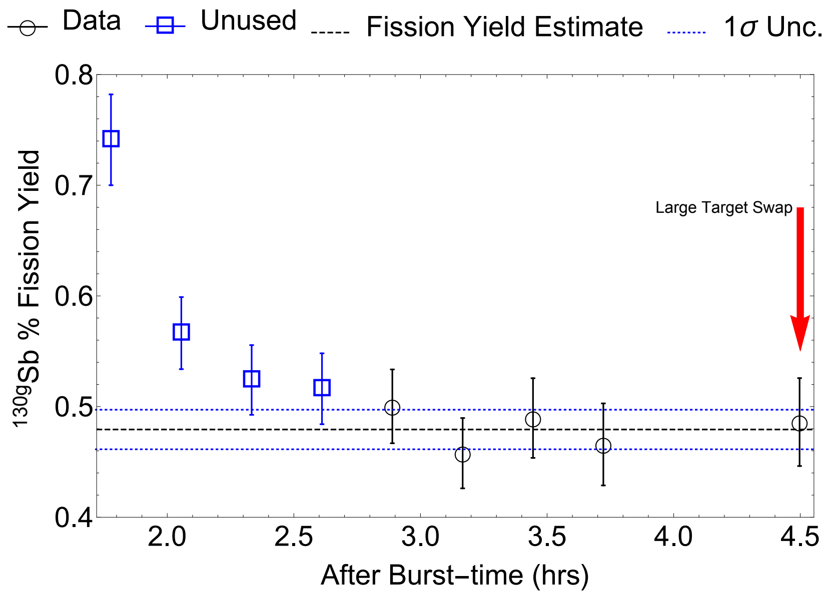
Isotope	Gamma-line (keV)	Γ	$\%1\sigma$	$T_{1/2}$ (sec)	$\%\ 1\sigma$	ε	$\%1\sigma$	F_{ss}	$\%1\sigma$
^{84g}Br	882	0.416	7.45	1908	0.2516	4.342E-3	0.635	0.985	0.222
^{93}Y	267	0.073	15.1	36648	0.7859	1.111E-2	1.003	0.894	1.76
^{94}Y	919	0.56	0.91	1122	0.9107	4.202E-3	0.610	0.985	0.212
^{104}Tc	358	0.89	3.37	1098	1.63	8.704E-3	2.061	0.914	0.914
^{128}Sn	482	0.59	12.	3544	0.2370	7.058E-3	0.782	0.965	0.522
^{129}Sb	813	0.43	2.3	15840	0.2272	4.635E-3	0.673	0.983	0.244
^{130g}Sb	793	1.	5.	2370	2.025	4.728E-3	0.681	0.983	0.252
^{131m}Te	774	0.368	1.90	119700	0.7519	4.821E-3	0.688	0.982	0.260
^{133}I	530	0.87	3.5	74880	0.4808	6.540E-3	0.753	0.970	0.446
^{134}I	847	0.96	3.1	3150	0.3810	4.485E-3	0.656	0.984	0.233
^{135}I	1132	0.226	3.10	23652	0.3044	3.562E-3	0.520	0.988	0.171
^{138}Xe	846	0.315	4.13	844.8	0.5682	7.675E-3	0.795	0.958	0.630
^{139}Ba	166	0.2376	1.05	4984	0.3371	1.668E-2	3.149	0.505	6.90
^{141}Ba	190	0.254	7.09	1096.2	0.3831	1.011E-2	0.701	0.919	1.29
^{142}Ba	255	0.205	3.90	636	1.887	1.147E-2	1.180	0.883	1.97
^{142}La	641	0.474	1.05	5466	0.5488	5.611E-3	0.711	0.977	0.335
^{146}Ce	218	0.208	5.77	811.2	0.9612	1.437E-2	3.462	0.681	3.35
^{149}Nd	211	0.259	5.56	6221	0.0578	1.478E-2	3.506	0.666	3.11

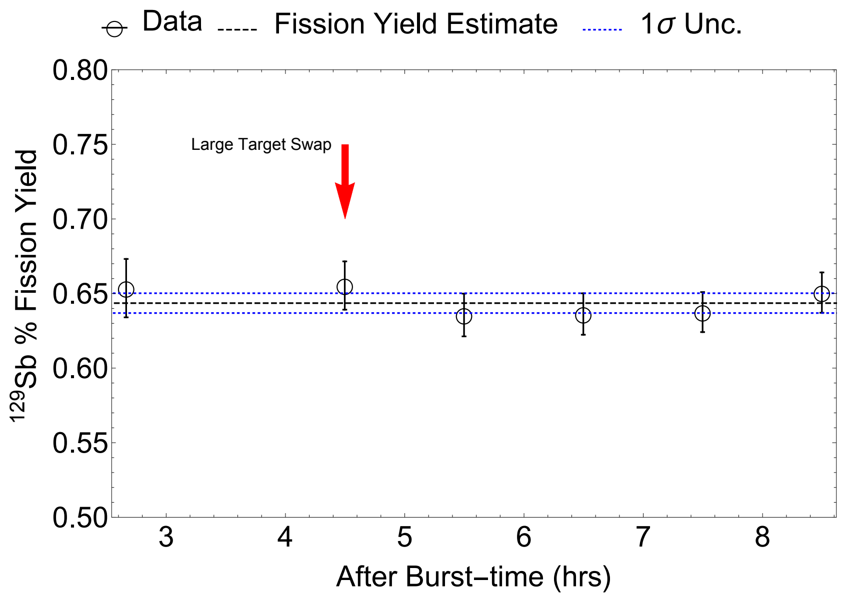
**Highlights need for improved photon branching ratio

Results & Discussion

Isotope	ENDF/B VIII.0 FY $\pm 1\sigma$	Meas. FY $\pm 1\sigma$
^{84g}Br ‡	0.822(23)	0.875(89)
^{93}Y	4.910(31)	4.621(767)
^{94}Y ‡	4.610(184)	5.084(488)
^{104}Tc	5.036(101)	4.841(353)
^{128}Sn	0.284(182)	0.236(34)
^{129}Sb	1.011(162)	0.644(63)
^{130g}Sb	0.926(593)	0.487(43)
^{130}Sb	0.866(43) [28]	–
^{131m}Te	0.264(42)	0.276(20)
^{133}I	6.760(4326)	7.296(547)
^{134}I ‡	7.60(456)	8.076(606)
^{135}I	6.941(97)	7.334(557)
^{138}Xe ‡	5.702(160)	5.694(547)
^{139}Ba	5.670(113)	6.338(602)
^{141}Ba	5.336(320)	5.028(513)
^{142}Ba ‡	4.581(183)	3.927(401)
^{142}La ‡	4.586(92)	4.991(359)
^{146}Ce ‡	3.445(96)	3.251(348)
^{149}Nd	1.625(65)	1.664(150)

[28] H. Naik, S. P. Dange, and R. J. Singh, Phys. Rev. C **71**, 014304 (2005).





Our measurements agree with other works but highlight some disagreement between the ENDF evaluation near the proton shell closure @ Z=50

Objectives & Future Work

- This data is made available for evaluators through publication and EXFOR
- The experimental method, data reduction & analysis techniques, uncertainty quantification, and reporting can be refined/improved to make the data more useful
- Incorporate GEANT4 full spectral simulation of expected and measured inventory for comparison to raw data to refine the analysis and improve completeness in data utilization
- Collaborate and interact more regularly with evaluators to improve data utilization

Thank you

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