Update on short-lived and cumulative fission product yields from the NPML Collaboration

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Multi-Lab multi-disciplinary collaboration

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Introduction

Motivation:

There have been few short-lived fission product yield studies since the publication of the England and Rider report in 1993 (28 years). The yields in the England and Rider report are a collection of results from experiments occurring from the 1950s through to the early 1990s.

Integral cross-section measurements in unperturbed fast fission neutron fields are rare but crucial data for a range of applied fields (criticality safety, reactor operations, etc…)

Approach:

Project is conducting the first set of self consistent irradiations of major actinides in nearly 30 years using:

- Long irradiations with fission chambers for an absolute fission basis estimate of yields
- Long irradiations of high purity mixed foil sets for cross-section measurements
- Prompt (< 60 μs) fission neutron bursts for relative fission yield measurements

High quality nuclear data essential to data evaluation and modeling;

- Improve nuclear data libraries
- Provide valuable nuclear data references for the nuclear science community

Neutron Sources

Operations conducted at the NCERC include both subcritical and critical experiments with the ability to measure a wide variety of nuclear properties.

Flattop (NCERC)

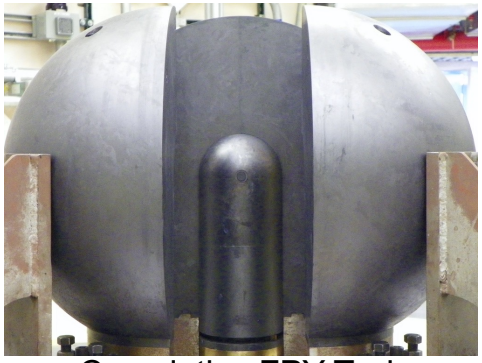
- Fast/fission Spectrum
- Horizontal ("traverse") glory hole
- 10^{13} fissions/g on samples

Godiva IV (NCERC)

- Fast/fission neutron spectrum
- Super-Prompt Critical Operations
- Vertical glory hole for samples
- 1-4 $\times$ 10^{16} Total Fissions / burst

D-T Generator (PNNL)

- Thermo D711 neutron generator
- Low scatter facility at PNNL
- Max neutron flux of 1×10^{9} n/cm^2/s
- New D-T source at LLNL Jan-2022



Cumulative FPY Task
Days to weeks post irradiation

Short-Lived FPY Task
Hours to days post irradiation

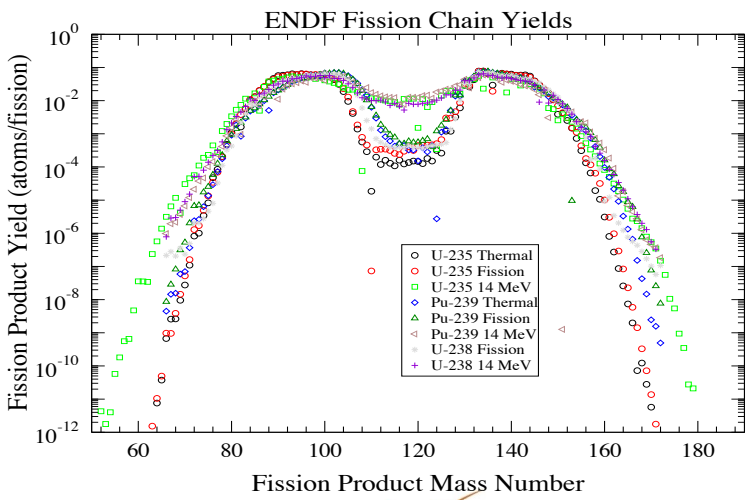
Cumulative FPY Task
Days to weeks post irradiation

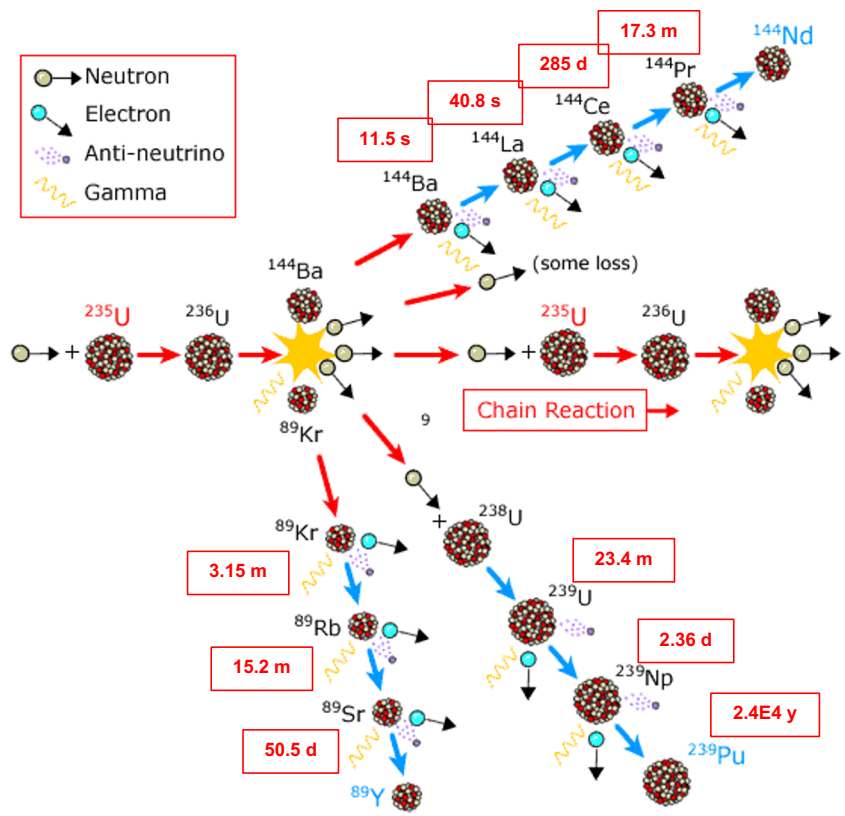
Time scales of isotopes we can investigate

Fission product isotopes cover a wide range of time scales from microsecond to day++ half-lives.

We can currently make measurements:

- At the 10s of minutes time scales by whole gamma counting
- Hours/days by performing radiochemistry and beta/gamma counting





Short-Lived Fission Product Yields (SLFPY)

Fabricate High Quality Targets → Pulse Irradiate at NCERC → Acquire γ spectral data → Symmetric Temporal Spectral Analysis (LLNL) → Asymmetric Temporal Spectral Analysis (PNNL) → Evaluate Neutron Fluence & Fissions → Publish Fission Yields in Nuclear Data Sheets for NNDC

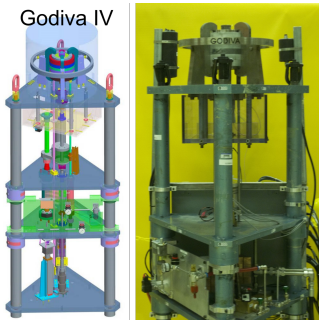
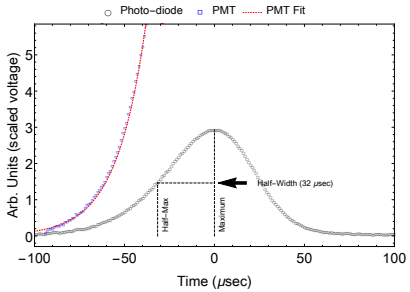
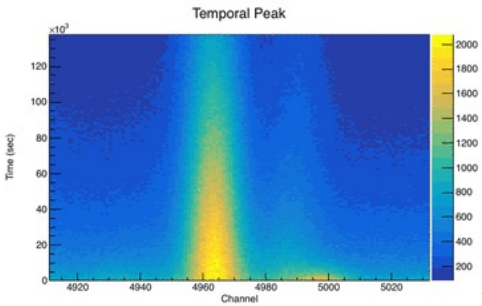
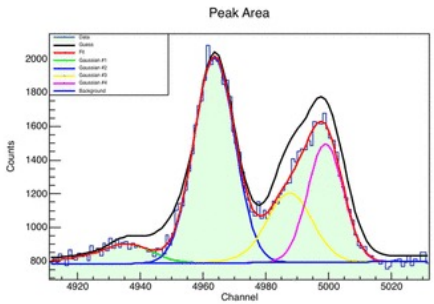


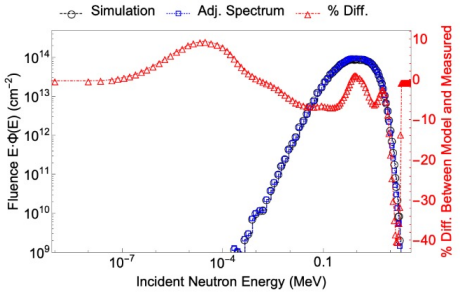
Photo & Model of Godiva

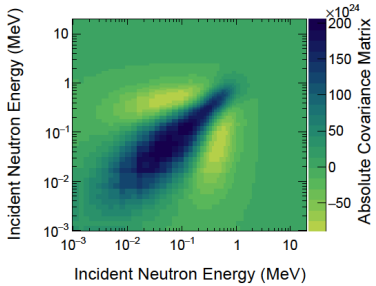


Godiva Irradiation Pulse Width

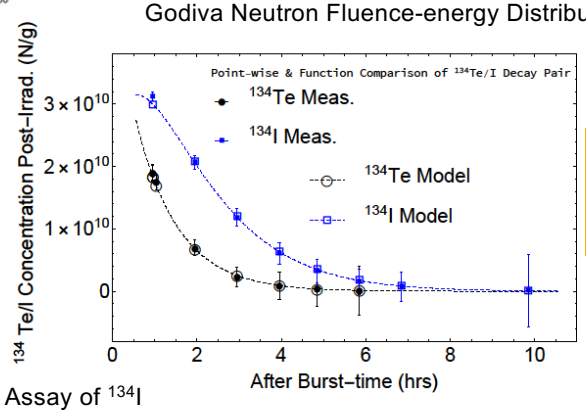








Godiva Neutron Fluence-energy Distribution & Covariance Matrix

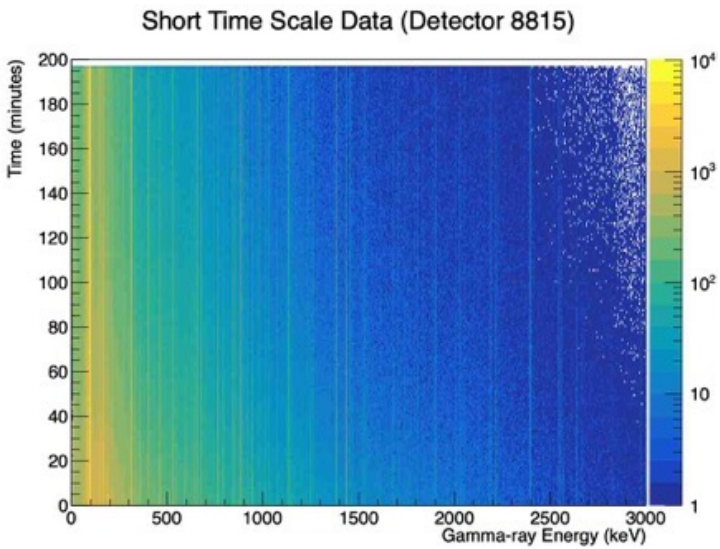


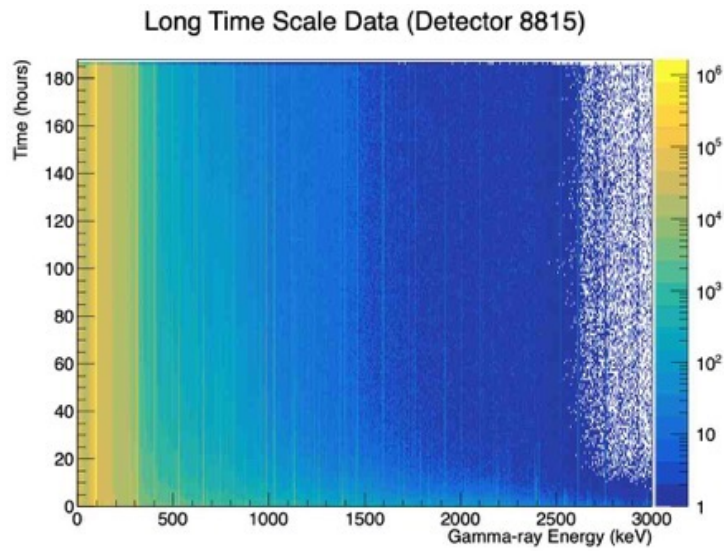
Temporal Assay of ^{134}I

Nuclear Data Sheets **155**, 86 (2019).
Nuclear Data Sheets **163**, 249 (2020).

Np237 Data Time Dependent Spectra Example

- Currently, we can retrieve the sample from Godiva ~30-40 minutes after the burst mode operation. Then begin measuring gamma-ray spectra at about 1 hour.
- 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

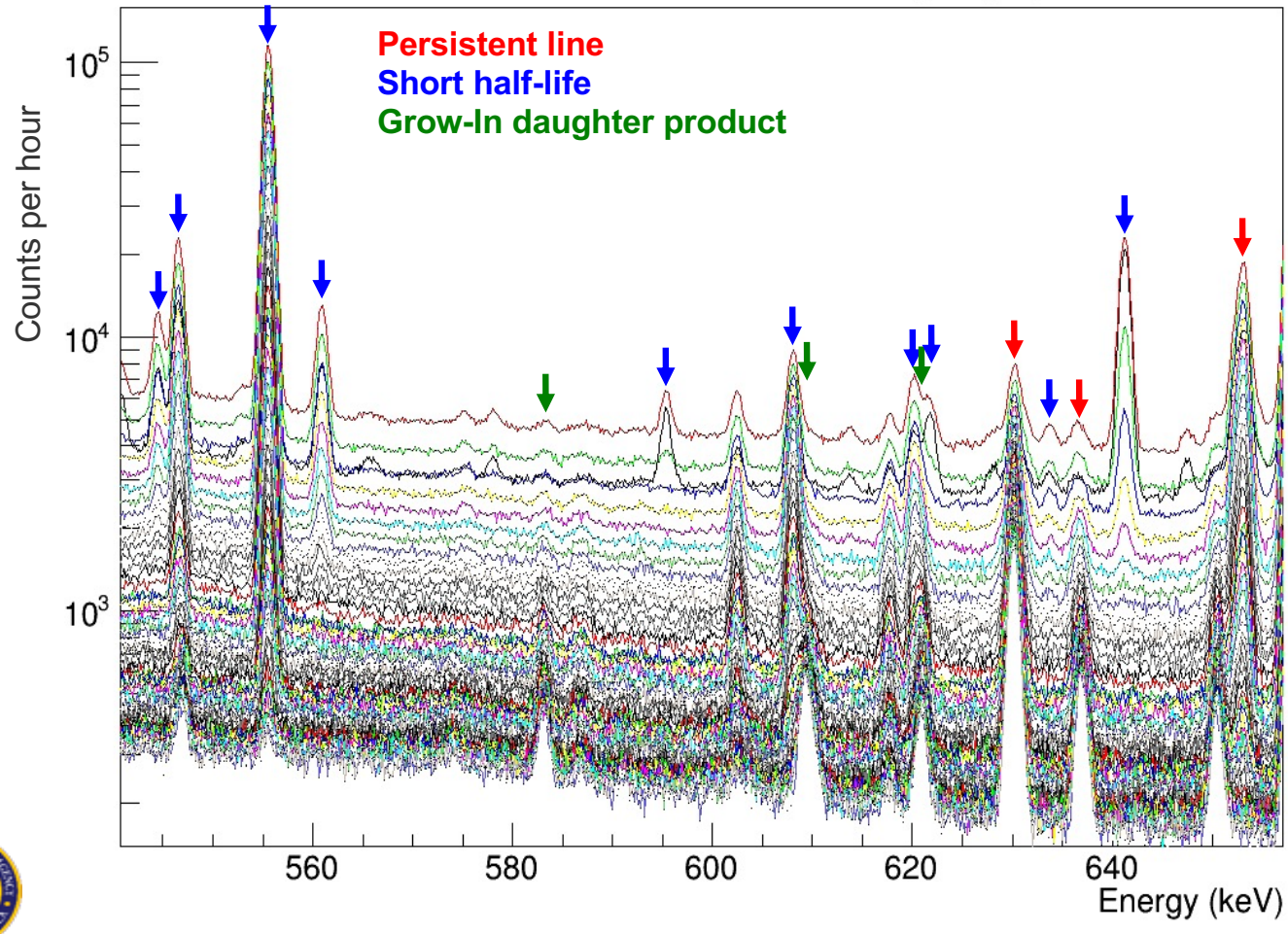




Courtesy Sean Burcher

Technical Approach: identifying gamma rays
Time dependent gamma ray spectra from 45 minutes to 7 days post-irradiation

List mode data taken with high resolution HPGe detectors to perform the gamma-ray spectroscopy is essential to unfolding the very complex spectra obtained from fission products.



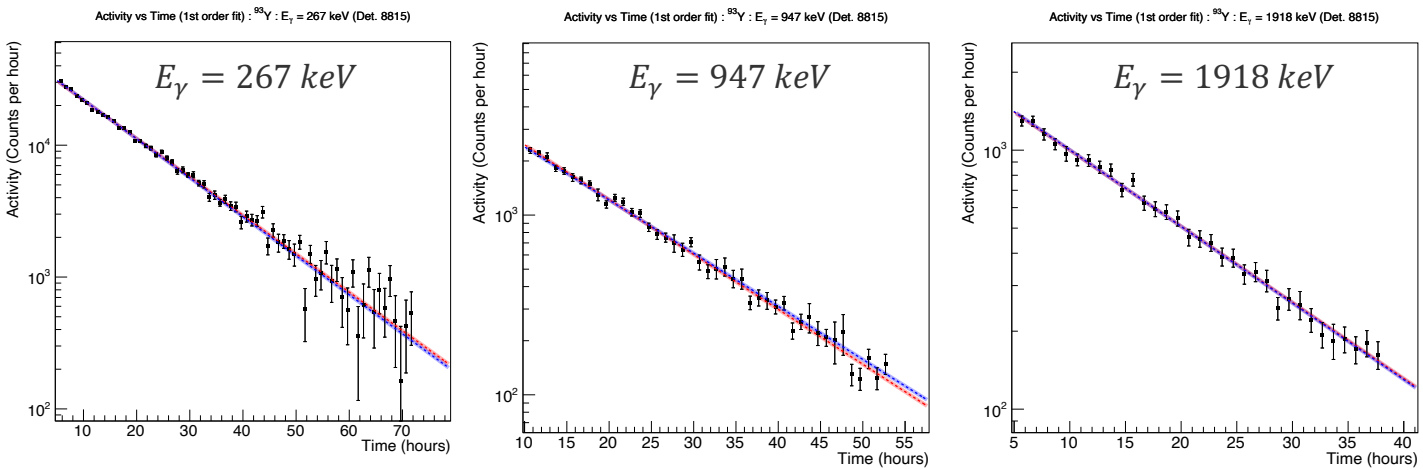
Np237 Results : Example ^{93}Y

- 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*

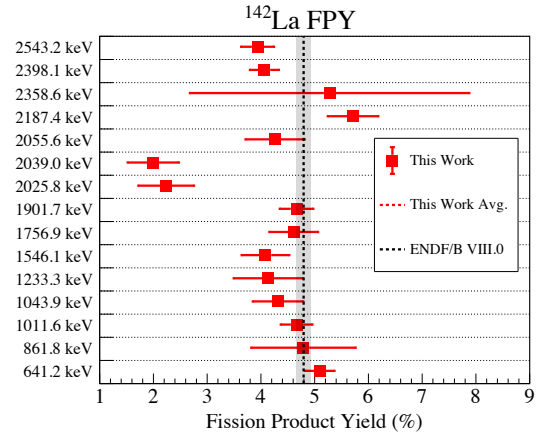
$$C(t_1, t_2) = \int_{t_1}^{t_2} A(t)dt = \frac{A_0}{\lambda} e^{-\lambda t_2}(e^{\lambda \Delta t} - 1)$$

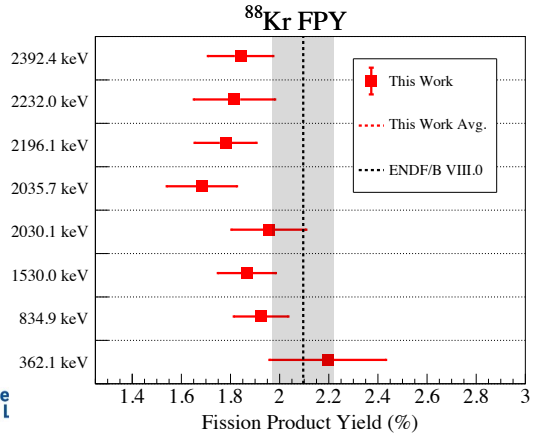


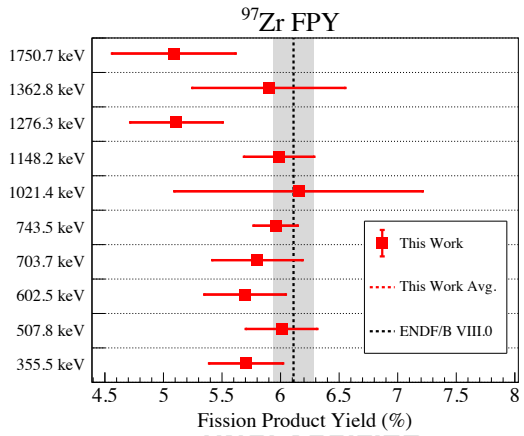
Courtesy Sean Burcher

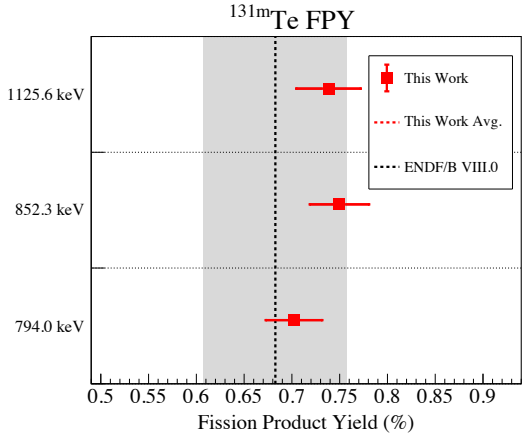
Results for ^{237}Np FPYs

- **Fission Product yields for 45 unique isotopes/isomers have been measured**
  - **Using 191 different γ-rays**
  - **Isomer Yields**
- **Half-lives ranging from 10s of minutes to a few days.**
- **Use multiple γ-rays from each decay if possible**
  - **Reduces sensitivity to systematic uncertainties introduced by potentially imprecise nuclear data**









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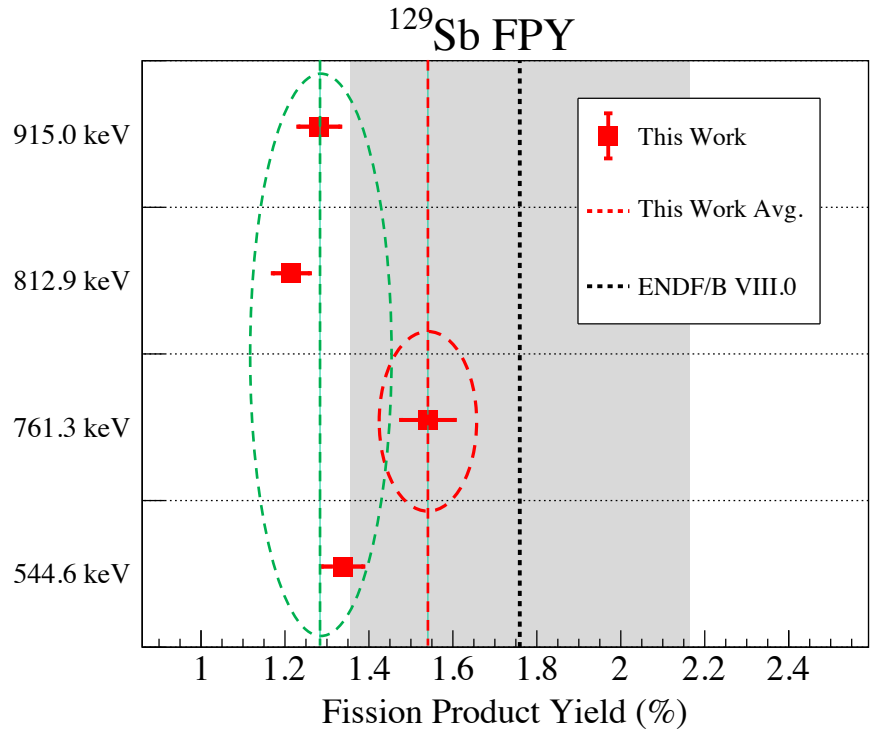
Results for ^{237}Np FPYs : Investigating Nuclear Data

- Using multiple γ-rays can highlight areas where current evaluated branching ratios may be incorrect
- Example: ^{129}Sb
  - Using the 761.3 keV γ-ray to measure FPY would produce a different result
  - The other 3 observed γ-rays produce a consistent value for FPY
- Indicates that there could be an issue with the branching ratio for the 761.3 keV γ-ray
  - In this case it appears that the ENSDF/B VIII.0 FPY value was measured using the 761.3 resulting in an inaccurate FPY



Past Results from ^{235}U, ^{238}U, ^{239}Pu

^{235}U FY

Isotope	Energy (keV)	ENDF/B VII.1 FY (%)	1σ-abs.	Meas. FY (%)	1σ-abs.	% Diff.
† ^{84g}Br	881.6	1.192	0.041	1.187	0.128	-0.419
† ^{89}Rb	1032	4.37	0.48	3.99	0.36	-8.70
^{91}Sr	1384	5.73	0.12	5.27	0.36	-8.03
^{93}Y	267	6.25	4.00	6.06	0.99	-3.04
† ^{94}Y	919	6.300	0.088	5.34	0.38	-15.2
^{97}Zr	743	6.003	0.084	5.85	0.45	-2.55
^{101}Mo	192	5.24	0.20	4.95	0.39	-5.53
^{105}Ru	724	1.196	0.072	1.243	0.086	3.93
^{128}Sn	482	0.49	0.32	0.487	0.073	-0.612
^{129}Sb	813	0.70	0.18	1.073	0.077	53.29
† ^{130g}Sb	793	0.93	0.60	0.513	0.055	-44.8
^{131}Sb	943	2.92	0.18	2.52	0.259	-13.7
^{131m}Te	774	0.43	0.19	0.581	0.048	35.1
^{133}I	530	6.72	0.70	6.60	0.50	-1.79
† ^{134}Te	767	6.57	0.18	5.73	0.36	-12.8
† ^{134}I	847	7.641	0.076	6.82	0.66	-10.7
^{135}I	1132	6.30	0.25	6.02	0.54	-4.44
† ^{138}Xe	434	6.01	0.08	5.65	0.57	-5.66
^{141}Ba	304	5.95	0.24	5.34	0.51	-10.3
† ^{142}Ba	255	5.51	0.11	5.238	0.442	-4.94
† ^{142}La	641	5.542	0.078	5.241	0.430	-5.43

^{238}U FY

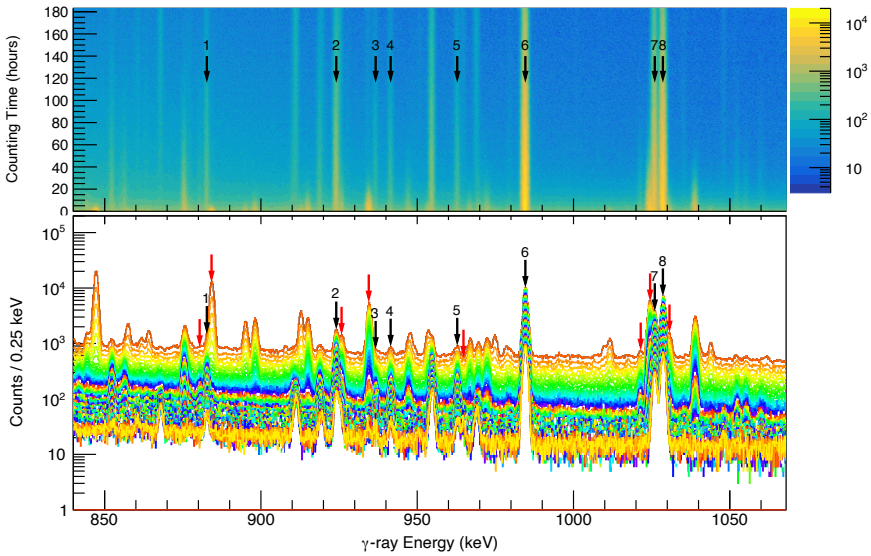
Isotope	ENDF/B VIII.0 FY ±1σ	Meas. FY ±1σ
^{84g}Br ‡	0.822(23)	0.875(89)
^{93}Y	4.910(31)	4.621(767)
^{94}Y ‡	4.610(184)	5.084(488)
^{104}Tc	5.036(101)	4.841(353)
^{128}Sn	0.284(182)	0.236(34)
^{129}Sb	1.011(162)	0.644(63)
^{130g}Sb	0.926(593)	0.487(43)
^{130}Sb	0.866(43) [28]	–
^{131m}Te	0.264(42)	0.276(20)
^{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)

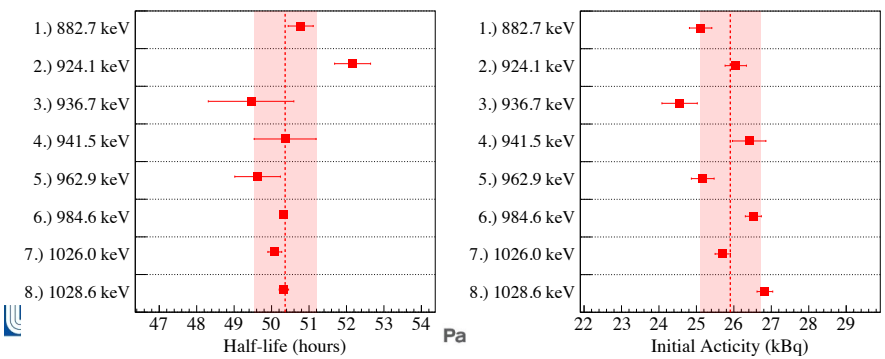
Preliminary ^{239}Pu FY

	ENDF VII.0/B		Small Target			Large Target		
Isotope	FY	±	FY	±	Ratio	FY	±	Ratio
Kr-85m	0.59	0.01	0.58	0.03	0.98	0.62	0.03	1.05
Rb-89	1.72	0.07	1.69	0.08	0.98			
Y-93	3.82	2.45	3.58	0.29	0.94	3.36	0.25	0.88
Y-94	4.22	0.06	4.13	0.19	0.98			
Mo-101	6.67	0.27	6.37	0.29	0.96			
Ru-105	5.36	0.21	5.51	0.25	1.03	5.52	0.25	1.03
Rh-107	3.22	0.52	3.19	0.16	0.99			
Sb-127	0.50	0.16				0.530	0.02	1.06
Sn-128	0.79	0.51	0.64	0.03	0.81	0.75	0.07	0.94
Sb-129	1.45	0.12	1.49	0.07	1.03	1.32	0.06	0.91
Sb-131	2.89	0.32	1.87	0.09	0.65			
Te-131	3.11	0.50	4.72	0.74	1.52			
Te-131m	0.92	0.41	1.52	0.08	1.66	1.28	0.06	1.39
I-131	3.88	0.04	3.18	0.23	0.82	4.09	0.19	1.06
Te-132	5.15	0.10	5.49	0.25	1.07			
I-133	6.91	4.42	6.28	0.30	0.91	6.83	0.32	0.99
Te-134	4.79	0.29	3.60	0.16	0.75	3.46	0.23	0.72
I-135	6.08	0.24	5.84	0.26	0.96	5.92	0.27	0.97
Xe-138	4.71	0.09	4.36	0.22	0.93			
Cs-139	5.37	0.43	4.36	1.06	0.81			
Ba-139	5.61	0.11	5.88	0.39	1.05	6.44	0.55	1.15
Ba-140	5.32	0.07	5.66	0.70	1.06	5.02	0.23	0.94
Ba-141	5.08	0.30	4.90	0.22	0.97			
Ce-141	5.15	0.14				4.57	0.21	0.89
Ba-142	4.48	0.49	3.82	0.21	0.85			
La-142	4.75	0.07	5.17	0.23	1.09	5.12	0.23	1.08
Ce-143	4.34	0.02	3.59	0.17	0.83	4.09	0.18	0.94
Ce-146	2.42	0.10	1.81	0.11	0.75			
Nd-147	1.99	0.03				1.77	0.09	0.89
Pr-147	1.99	0.06	2.01	0.14	1.01			
Nd-149	1.24	0.02	1.23	0.06	0.99	1.18	0.06	0.95
Pm-151	0.78	0.01				0.73	0.05	0.93
Sm-153	0.43	0.03				0.43	0.02	1.01

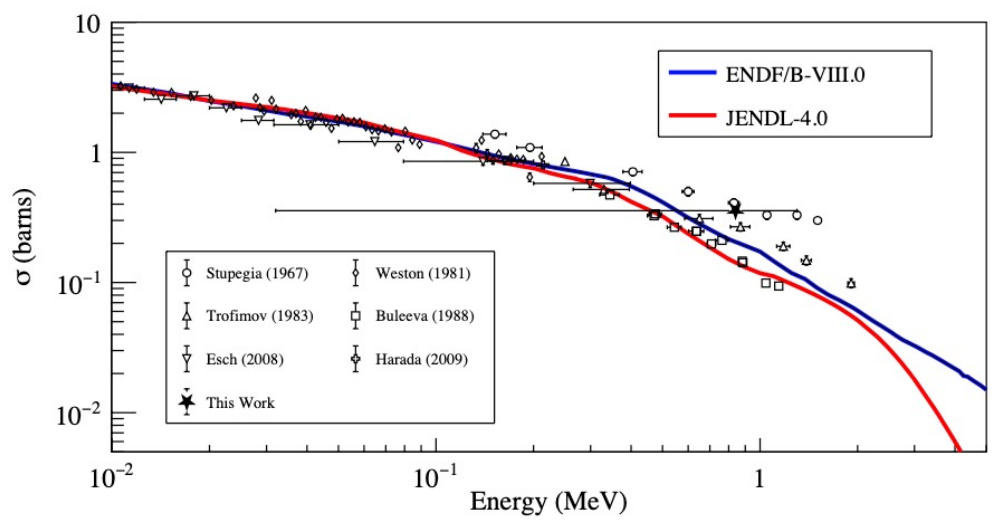
Highlighted in GREEN are improved measurements

Integral Measurement $^{237}Np(n,\gamma)^{238}Np$ Cross Section



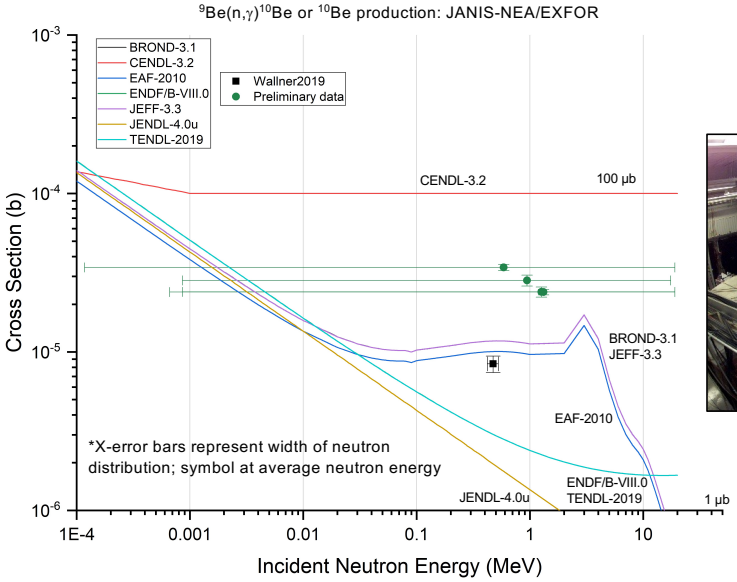


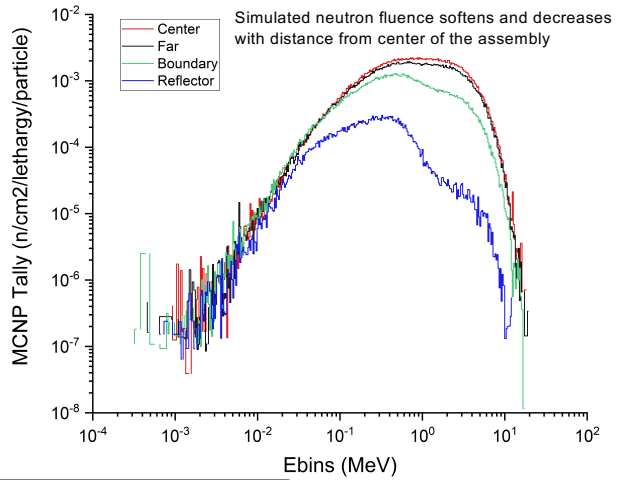
- γ-rays from the decay of ^{238}Np were observed in the time dependent spectrum
- Activity of ^{238}Np was used to determine the amount of ^{238}Np produced during the irradiation

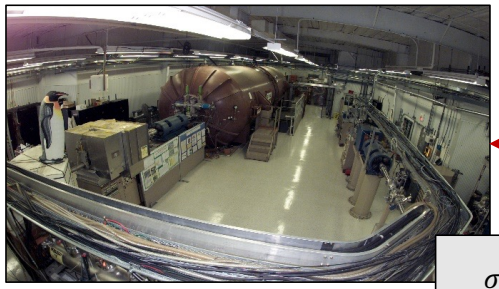


NCERC Flattop assembly irradiation and modeling to evaluate the $^{9}Be(n,\gamma)^{10}Be$ cross section at fission energies
Courtesy of Jack Goodell, Jennifer Church and Bryan Bandong

- High-fidelity model of the Flattop assembly and experimental configuration simulated using MCNP6.2
- $^{9}Be(n,\gamma)^{10}Be$ integral cross section determined by total neutron fluence and the $^{10}Be/^{9}Be$ ratio
- Neutron fluence estimates determined by $^{197}Au(n,\gamma)^{198}Au$ analysis
- $^{10}Be/^{9}Be$ ratio measured by accelerator mass spectrometry
- Preliminary $^{9}Be(n,\gamma)^{10}Be$ integral cross section results do not agree with current evaluated libraries







LLNL Center for Accelerator Mass Spectrometry (CAMS)

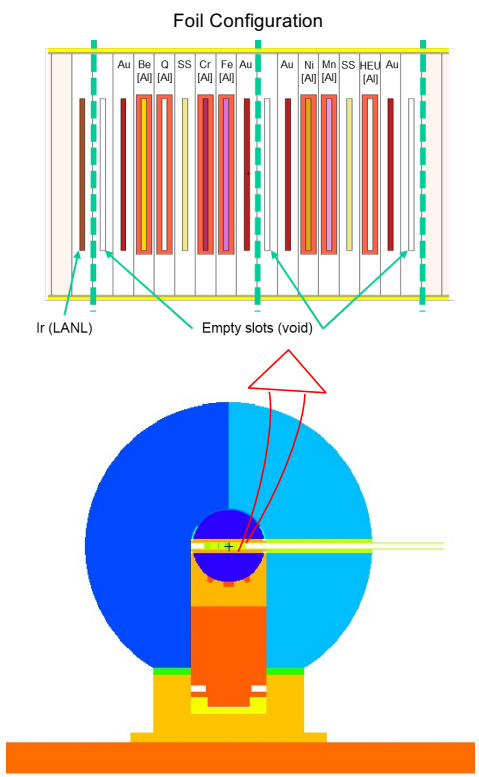
CAMS assay

$$\sigma_{exp} = \frac{1}{\Phi_{total}}\left(\frac{^{10}Be}{^{9}Be}\right) \quad [1]$$

Φ_{total} = Total fluence (n/cm^2) from ^{198}Au fluence estimates

[1] Wallner *et al.* Phys Rev C 99, 015804 (20129)

MCNP Visualization of the Flattop Model



*From Corey Keith - LANL

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We are aiming to return to Oregon State University Jan-2022 to perform the short time scale U238 irradiations

Rabbit shuttle injection station

OSU TRIGA reactor pulsed

Reports with compendia of results for SLFPY

- "Interim Report on Fission Product Yields from the Irradiation of Np237 using the Godiva Critical Assembly", S. Burcher et al. Sept. 2020 LLNL-TR-1023214
- "Report on Short-Lived Fission Product Yields from Pu239", S. Padgett et al. LLNL-TR-799578 Dec 2019
- "Interim Report on Short-Lived Fission Product Yields from U238", J.T. Burke et al. LLNL-TR-738508 Sept 2017
- "Interim Report on Short-Lived Fission Product Yields from U235", J.T. Burke et al. LLNL-TR-704065 Sept 2016
- "Improved Cumulative Fission Yield Measurements with Fission Spectrum Neutrons on ^{235}U," *Nuclear Data Sheets*, 155, 86–97, Jan. 2019, doi: 10.1016/j.nds.2019.01.005
- "Improved Cumulative Fission Yield Measurements with Fission Spectrum Neutrons on 238U," *Nuclear Data Sheets*, 163, 249–260, 2020, doi: 10.1016/j.nds.2019.12.006.

Cumulative Fissions Product Yields and the R-Value

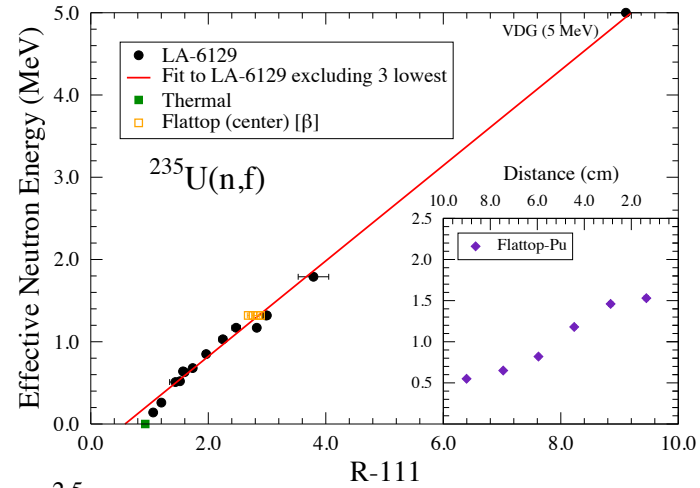
An R-value is the ratio of the yield of a FP (i) to the yield of ^{99}Mo ($i = 99$) in an unknown, relative to the same FP ratio from thermal fission ($k = th$) of ^{235}U ($j = 25$).

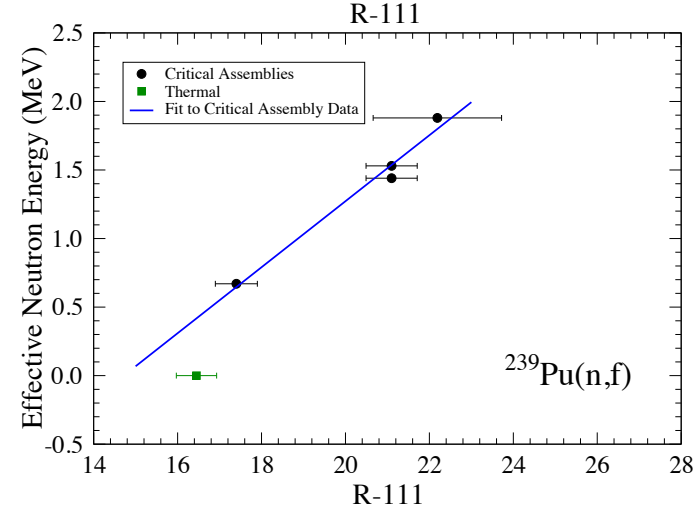
Developed in the late 40's such that a number of calculational inputs cancel out, such as $\Phi_n(E_n, t)$.

$$R_i^{j,k} = \left(\frac{A_i^{j,k} / A_{99}^{j,k}}{A_i^{25,th} / A_{99}^{25,th}} \right) = \left(\frac{Y_i^{j,k} / Y_{99}^{j,k}}{Y_i^{25,th} / Y_{99}^{25,th}} \right)$$

$$Y_i^{j,k} = \frac{(1 + \alpha_i) A_i^{j,k}}{\lambda_i \epsilon_i f_i N_f^*}$$

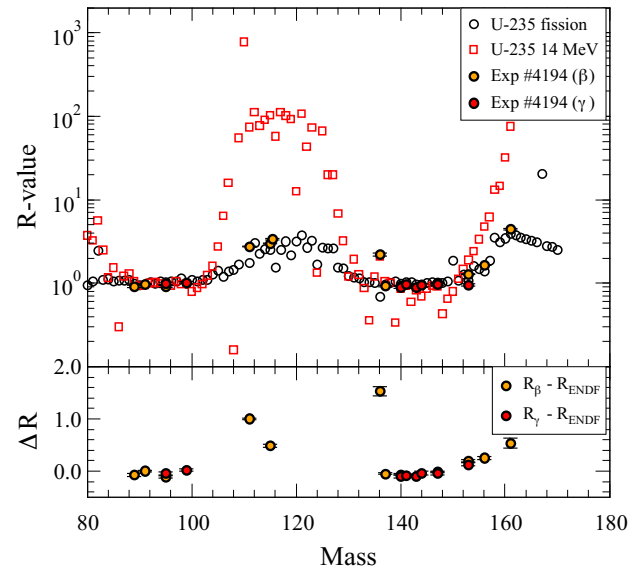
- G.P. Ford and A.E. Norris, LA-6129 (1976)
- H.D. Selby, et al., Nucl Data Sheets 111, 2891 (2010)
- M.B. Chadwick, et al., Nucl Data Sheets 111, 2923 (2010)
- J. Laurec, et al., Nucl Data Sheets 111, 2965 (2010)





Execution

- **Target preparation**
  - **Actinides for FPY measurements by:**
    - **Direct counting**
    - **Radiochemistry**
  - **Stacked foils for integral cross-section measurements in- and ex-core**
- **Irradiation at NCERC & Other (NNSS/DAF)**
- **Sample assay @ NCERC**
  - **LANL/LLNL counting facilities (HPGe coincidence)**
- **Sample shipping (from NNSS/DAF to labs)**
  - **Assay by gamma & beta**
  - **Post assay of list-mode data**



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

$$R_i^{j,k} = \left(\frac{A_i^{j,k} / A_{99}^{j,k}}{A_i^{25,th} / A_{99}^{25,th}} \right) = \left(\frac{Y_i^{j,k} / Y_{99}^{j,k}}{Y_i^{25,th} / Y_{99}^{25,th}} \right)$$

Cumulative Fission Product Yield Sample Preparation

- Sample preparation (can involve more than one lab)
  - Select stock material and determine/verify chemical and isotopic composition
  - Actinides, e.g. ^{233}U, ^{235}U, ^{238}U, ^{237}Np, ^{239}Pu, in 100s of milligram quantities
  - Geometric characterization of all target foils (mass and dimensions)
  - Sample containment prior to shipping (containment of Pu samples is part of the NPML)
- Sample shipping NCERC
  - Current experimental activities involve multiple actinide (SNM) samples

Cumulative Fission Product Yields: Previous work

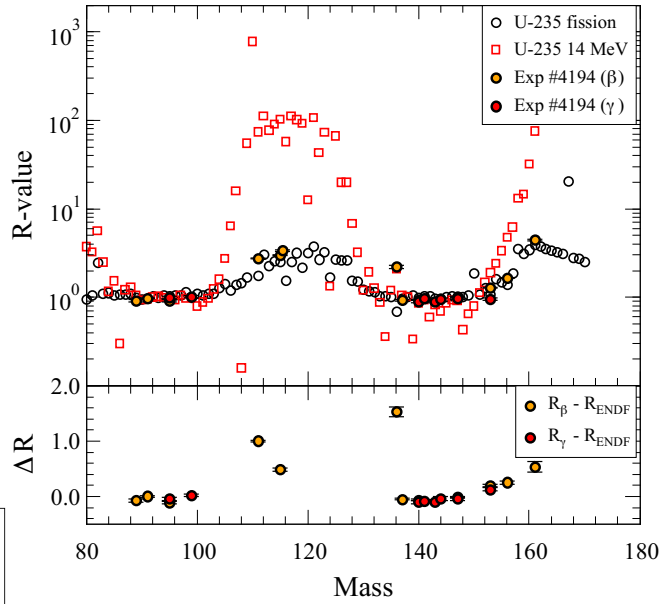
Experimentally determined R-values for ^{235}U in the center position of Flattop compared to R-values calculated from ENDF/B-VII.1 fission product yields for 0.5 and 14 MeV incident neutrons.

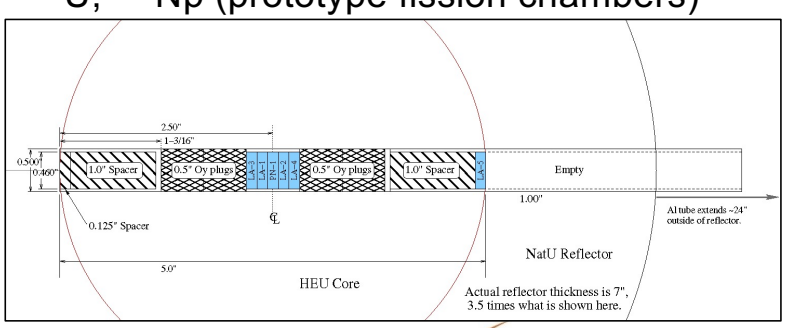
The lower panel shows the absolute difference between the experimental values determined by β counting (black circles) and γ counting (red circles) and ENDF/B-VII.1.

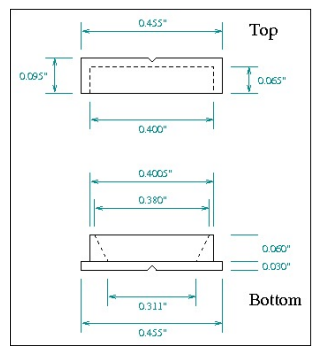
Previous measurements on the Flattop assembly

^{235}U, ^{238}U, ^{233}U, ^{237}Np and ^{239}Pu (no fission chambers)

^{235}U, ^{237}Np (prototype fission chambers)







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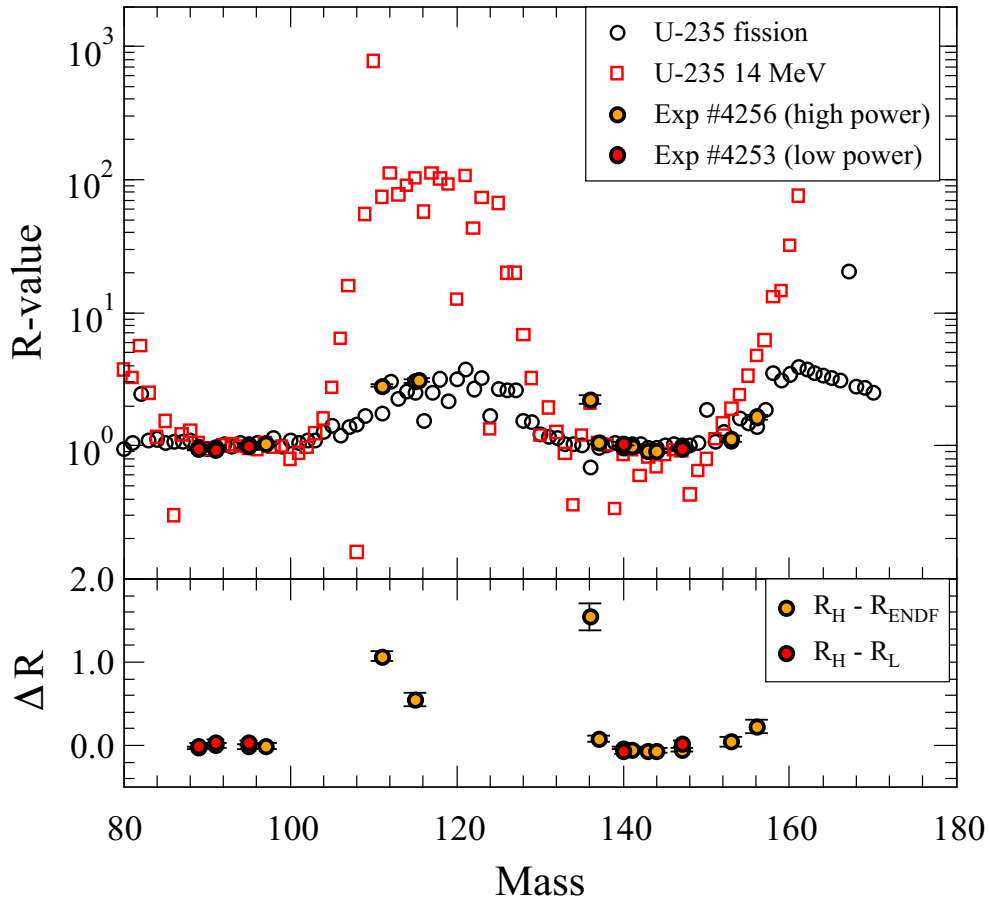
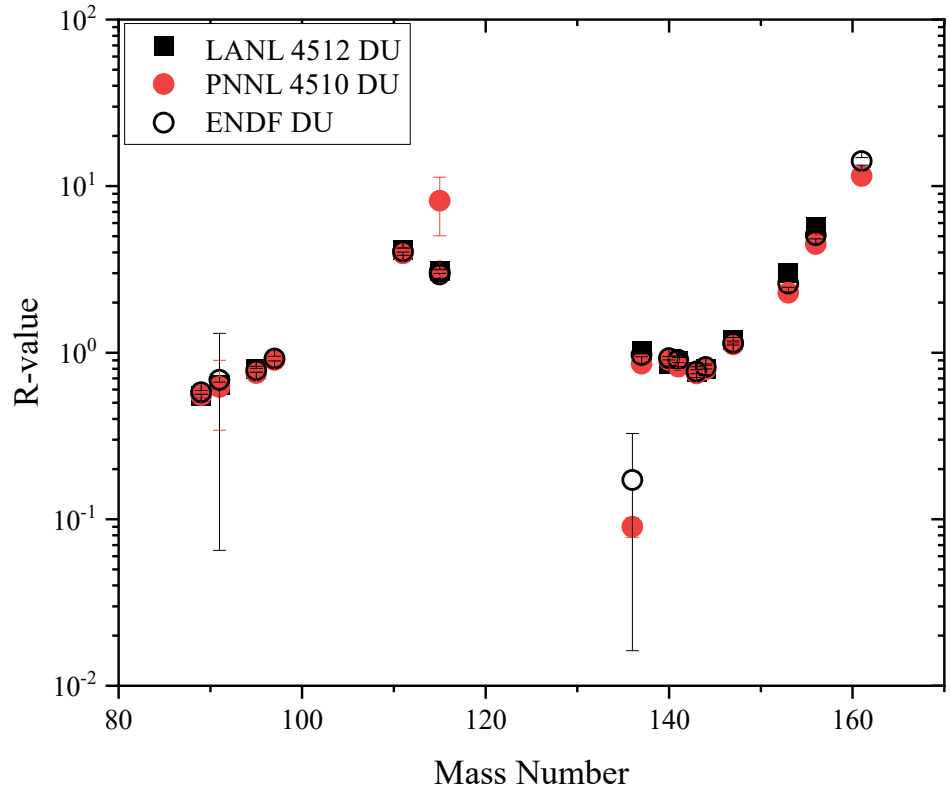
Results For Experiments #4253 and 4256 (HEU)

- 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.



Results For Recent Experiments #4510 (PNNL DU), #4512 (LANL DU)

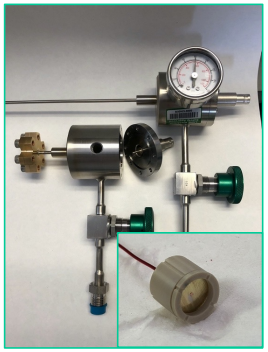
- Irradiation of DU at NCERC on Flattop in April 2021, samples split between LANL and PNNL.
- Comparison of R-values determined through whole A-solution or radiochemical separations performed by LANL and PNNL on a DU compared ENDF.V.III.0
- There is excellent agreement between the two laboratories and calculated ENDF values with some notable improvements to R-value uncertainties
  - ^{91}Y, ^{136}Cs

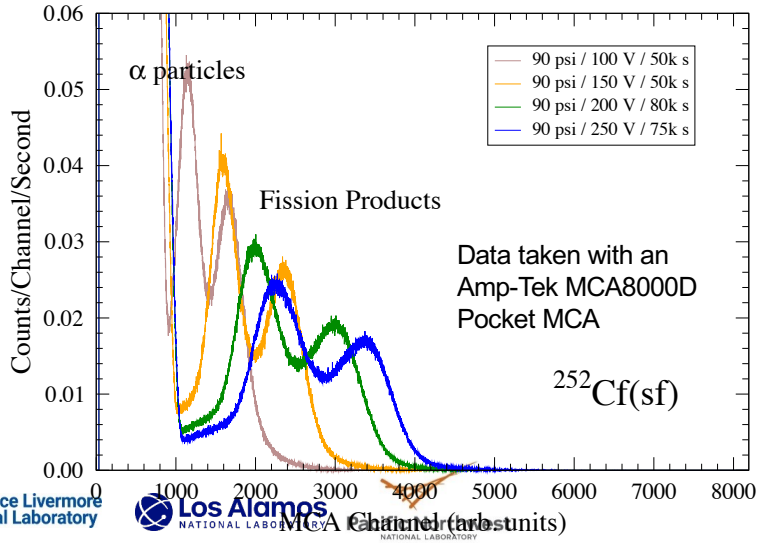


Fission Chamber Performance

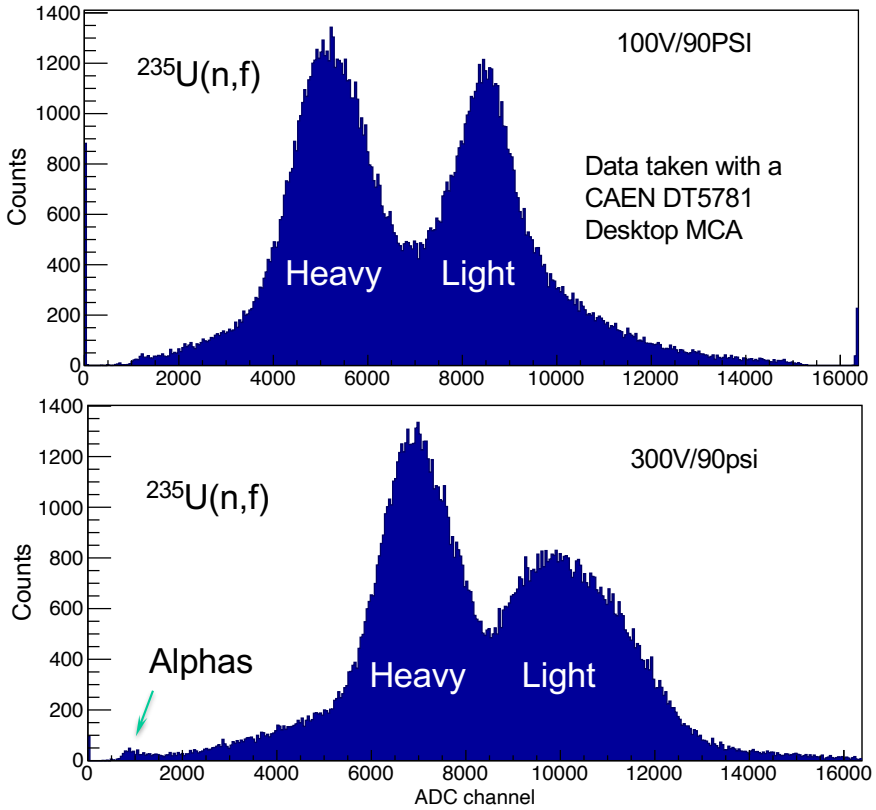
Pulse height spectra from the Mark II fission chamber

- Benchtop testing with ^{252}Cf
- Testing at MIT with ^{235}U
- Both using P-10 fill gas



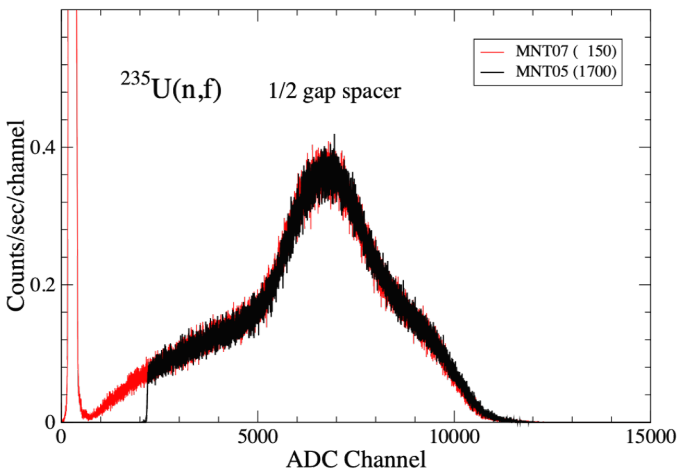


MIT Nuclear Reactor Laboratory



Results

MIT FC Tests 9-13-21 (½ gap)



The red curve is "zero" threshold. The black curve is the same settings used during the Flattop irradiation in April.

Need to confirm with ^{252}Cf, then correct detector efficiency for the high threshold.

+

Radiochemistry Results (R-values)

Isotope	Beta or gamma	R	±%	Atoms/g target	±%	Notes
^{89}Sr	B	0.921	2.4	3.15E+10	2.2	
^{91}Y	B	0.888	4.6	3.75E+10	4.5	
^{95}Zr	G	0.973	3.8	4.58E+10	3.0	Beta analysis - within one s
^{97}Zr	G	0.986	3.8	4.27E+10	3.1	Beta analysis - within one s
^{99}Mo	B	7.24E+11	2.2	4.43E+10	2.2	R- value column used to report fissions/g target
^{111}Ag	B	2.79	2.9	3.52E+08	2.8	
^{115}Cd	B	3.15	3.8	2.65E+08	3.7	
^{115m}Cd	B	-	-	-	-	Too few counts to fit beta data with confidence
^{136}Cs	G	2.28	5.7	9.81E+07	5.2	
^{137}Cs	G	1.07	3.6	4.81E+10	2.8	
^{140}Ba	B	0.904	2.7	4.07E+10	2.5	Beta analysis - within one s
^{141}Ce	-	-	-	-	-	Incomplete analysis
^{143}Ce	-	-	-	-	-	Incomplete analysis
^{144}Ce	-	-	-	-	-	Incomplete analysis
^{147}Nd	B	0.927	6.9	1.51E+10	6.8	A beta 'no absorber' measurement, within 1 s of the beta 'with absorber and gamma measurements
^{153}Sm	B	1.33	10.7	1.29E+09	10.6	High uncertainty driven by low gravimetric yield
^{156}Eu	B	1.59	6.3	1.72E+08	6.2	
^{161}Tb	-	-	-	-	-	Analysis not attempted

HEU Low-power Irradiation

Fission Product Yield Evaluations

- Data from this and other projects will feed into a new FPY evaluation that will happen over the next few years.
- Regular interactions with another NA-22 funded multi-lab project “Evaluation of Energy Dependent Fission Product Yields”
  - LANL (leading lab) develops FPY models and produce the final FPY data files
  - BNL compiles experimental FPY data and produces a set of recommended FPY values, performs FPY data validation calculations
  - LBNL performs measurements of energy-integrated and differential CNAA (Cyclical Neutron Activation Analysis) using the intense neutron source by the LBNL cyclotron, and data interpretation by the FIER code
  - PNNL develops a new Bragg curve-based fission TPC (Time Projection Chamber) analysis in collaboration with LANL and LLNL
  - LLNL develops theories and methods to calculate primary fission fragment yields, and performs FREYA calculations for prompt decay
- The theory effort draws on the experimental results of several experimental activities funded by federal agencies through the Nuclear Data Interagency Working Group

Thanks for your time and attention.

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church4@llnl.gov – AP

tonchev2@llnl.gov – SLFPY – mono-energetic TUNL