

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

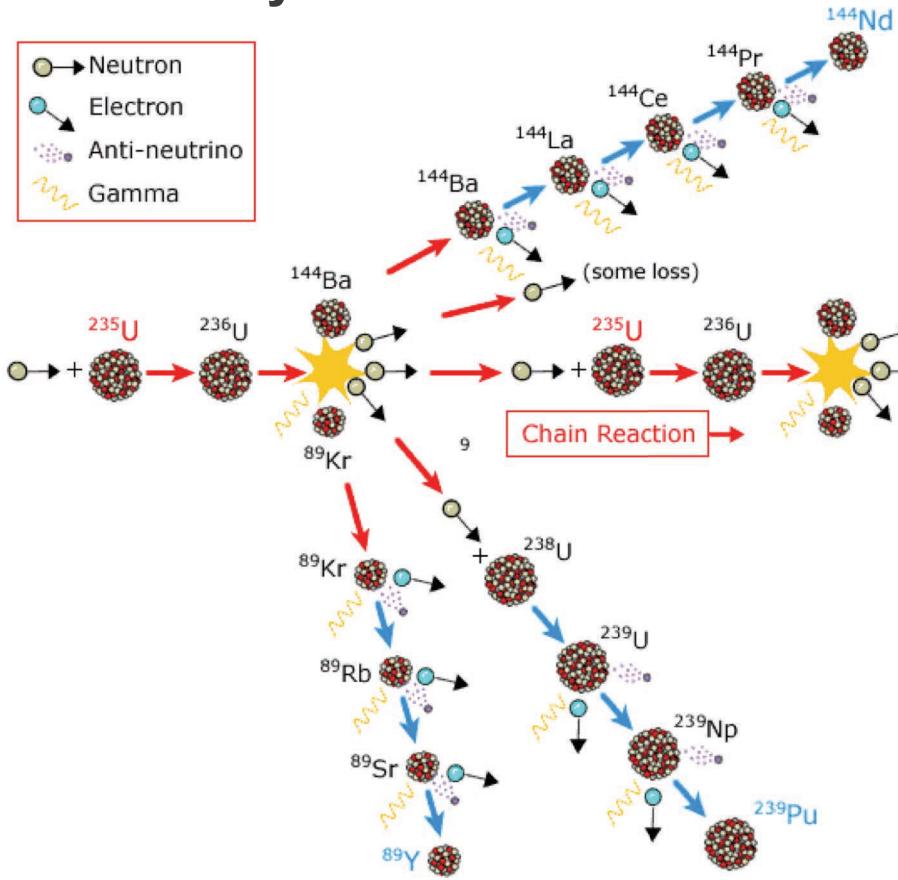
- Radiochemical analysis of irradiated samples to determine relative ($R_i^{j,k}$) cumulative FPYs and associated reaction rates.
- and/or
- Direct γ -ray counting of the irradiated samples to determine relative ($R_i^{j,k}$) cumulative FPYs and associated reaction rates.

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.

Why is nuclear physics / nuclear data important to the national security mission

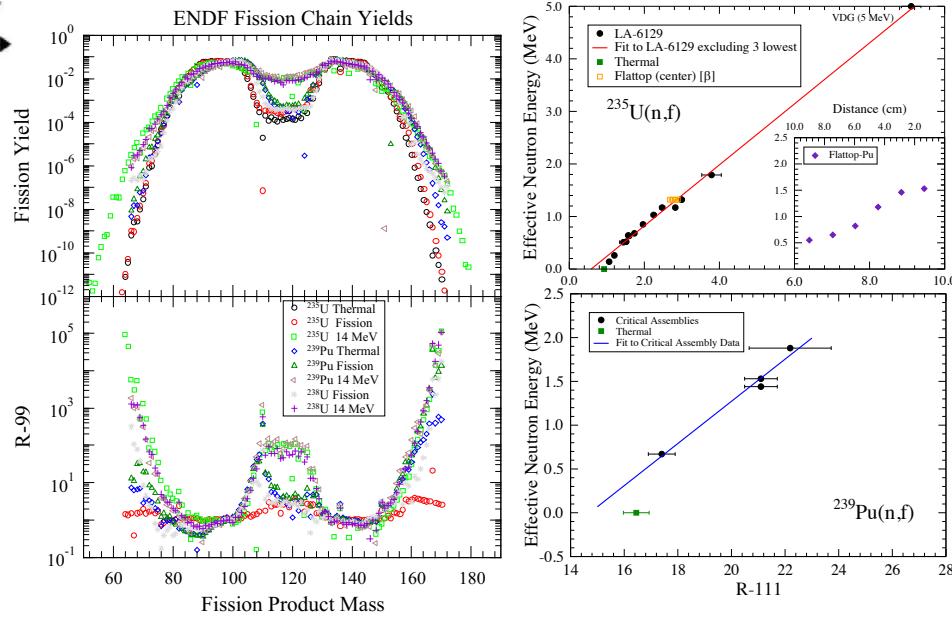


The defining signature of a nuclear detonation is the presence of nuclear reaction products in the debris. These can be from

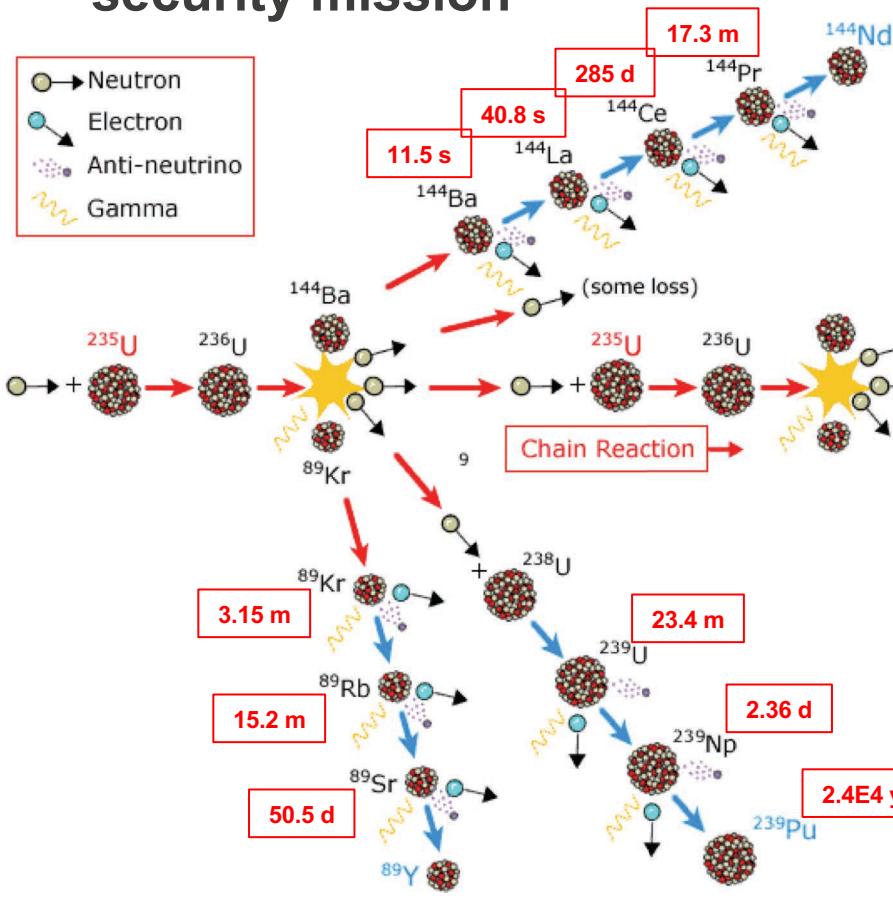
- Fission products (FP) from the fuel

$$^{An}(n,f)FP_{1,2}, {}^A FP_{1,2}(\text{decay}) {}^A FP'_{1,2}$$
 - Activation products (AP) from the fuel or nearby materials

$${}^AZ(n,2n){}^{A-1}Z, {}^AZ(n,\gamma){}^{A+1}Z, {}^AZ(n,p){}^A(Z-1), {}^AZ(\text{decay}) {}^A Z, \text{etc.}$$

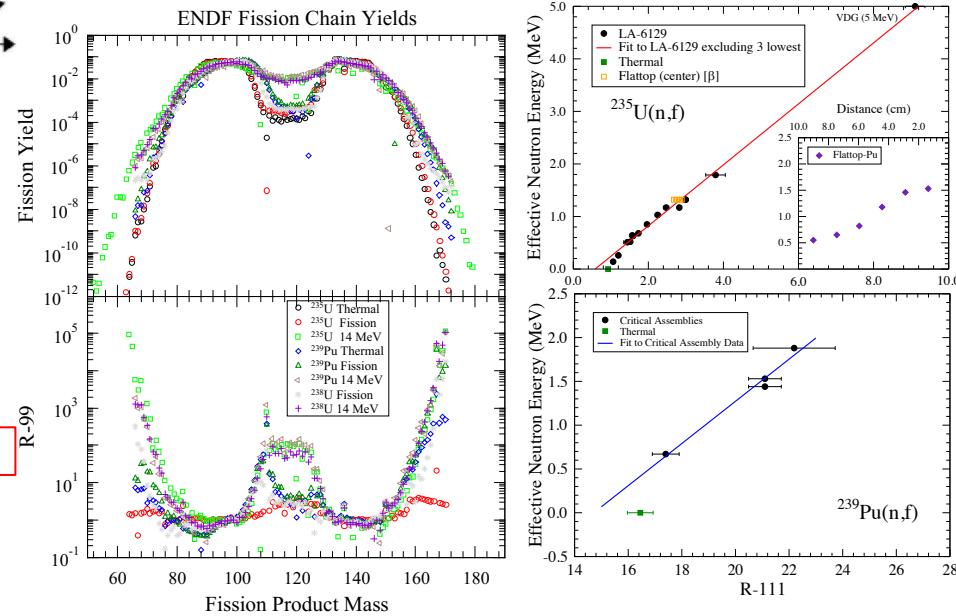


Why is nuclear physics / nuclear data important to the national security mission



The defining signature of a nuclear detonation is the presence of nuclear reaction products in the debris. These can be from

- Fission products (FP) from the fuel
 $\text{An}(\text{n},\text{f})\text{FP}_{1,2}$, $^{\text{A}}\text{FP}_{1,2}(\text{decay})^{\text{A}'}\text{FP}'_{1,2}$
- Activation products (AP) from the fuel or nearby materials
 $^{\text{A}}\text{Z}(\text{n},2\text{n})^{\text{A}-1}\text{Z}$, $^{\text{A}}\text{Z}(\text{n},\gamma)^{\text{A}+1}\text{Z}$, $^{\text{A}}\text{Z}(\text{n},\text{p})^{\text{A}}(\text{Z}-1)$, $^{\text{A}}\text{Z}(\text{decay})^{\text{A}'}\text{Z}$, etc.



Background

Fission Product Yields (FPYs) were historically determined by radiochemical analysis of the irradiated material, and reported as ratios of activities known as R-values*

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

Fission chamber experiments were used to calibrate radiation detectors for a small set of FPs, e.g. ^{99}Mo , to quickly determine # of fissions that occurred in a sample

$$K_i^{j,k} = \frac{N_f^*}{A_i^{j,k}} \quad \Rightarrow \quad N_f^* = K_i^{j,k} A_i^{j,k}$$

The K-factor was then used to determine the number of fissions that occurred in other samples directly from the measured activity of the chosen fission product.

NOTE: THE K-FACTOR IS DETECTOR SPECIFIC!

* G.P. Ford and A.E. Norris, LA-6129 (1976)

- The fission chamber (FC) is used to determine the number of fissions, N_f , in a thin deposit of fissile material, called the reference foil.
- The number of fissions, N_f^* , occurring in a “macro-foil” is scaled from the reference foil by mass.

$$N_f^* = \frac{m_M}{m_R} N_f$$

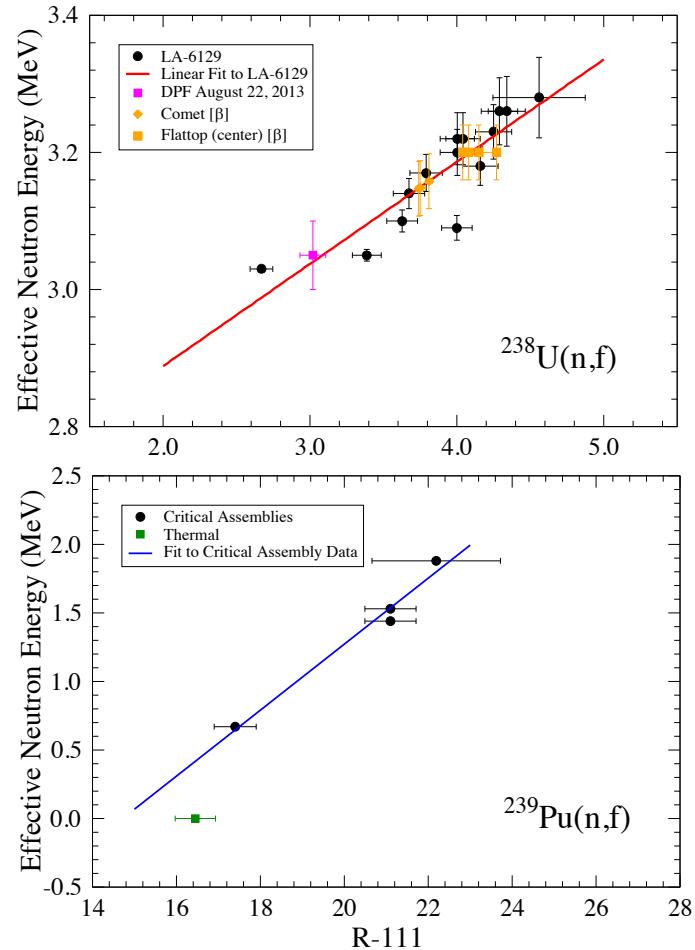
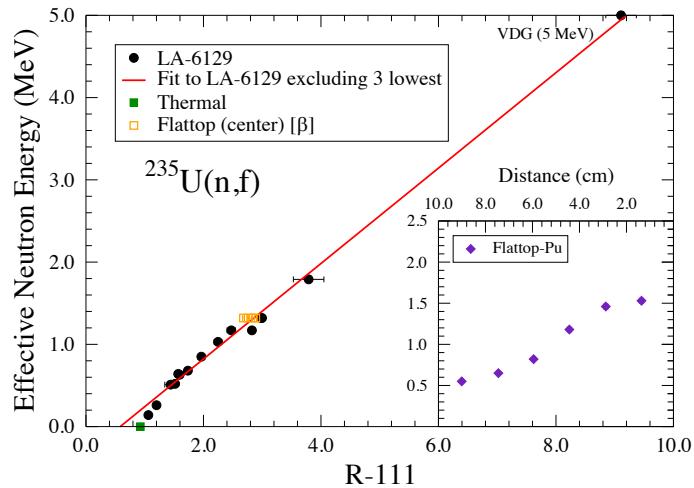
- Radiochemical (RC) analysis of the macro-foil provides $A_i^{j,k}$. The atom number and FPY is then determined by the standard equations

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

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

- Use MCNP and other tools to model as-run configuration and compare results with measured values to test nuclear data.

Integral CFPY Measurements and Re-evaluations



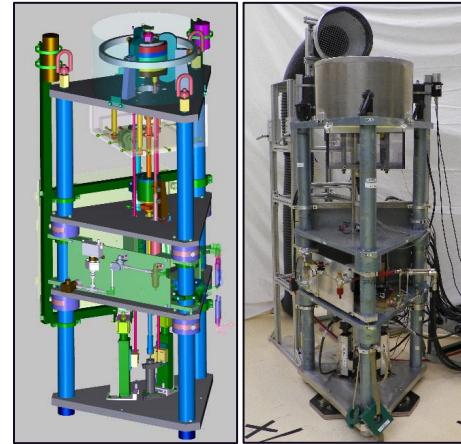
- G.P. Ford and A.E. Norris, LA-6129 (1976)
- H.D. Selby, et al., Nucl Data Sheets 111, 2891 (2010)
- M.B. Chadwick, et al., Nucl Data Sheets 111, 2923 (2010)
- J. Laurec, et al., Nucl Data Sheets 111, 2965 (2010)

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
- $1-4 \times 10^{16}$ Total Fissions, Hard Spectrum



Comet

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



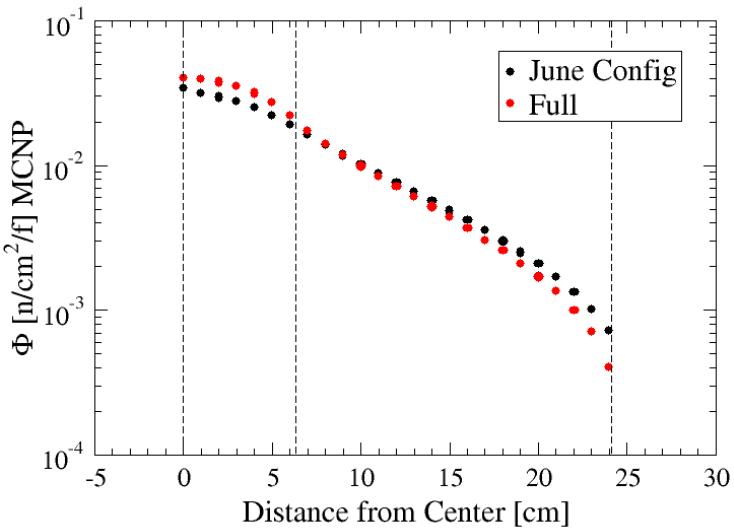
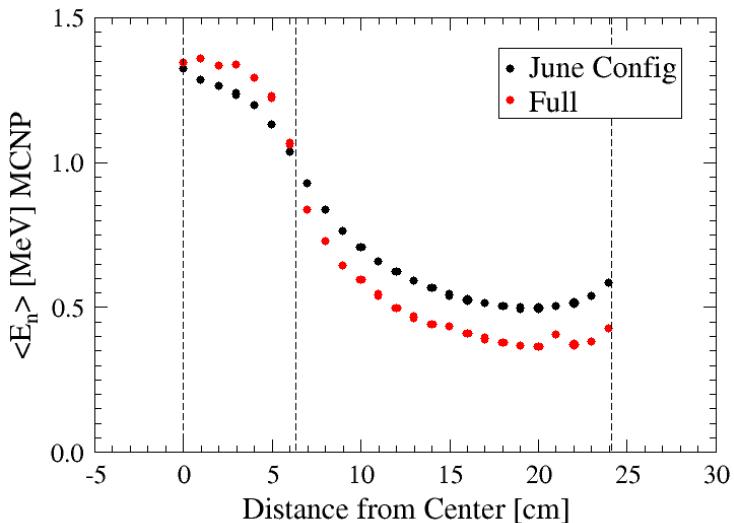
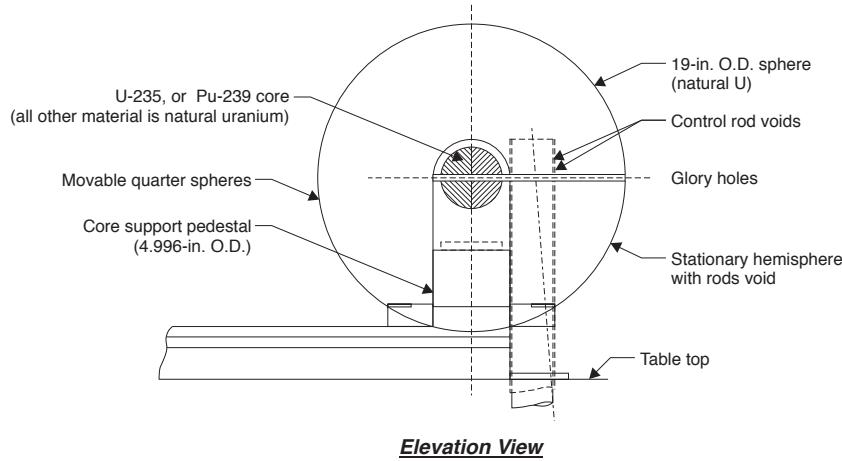
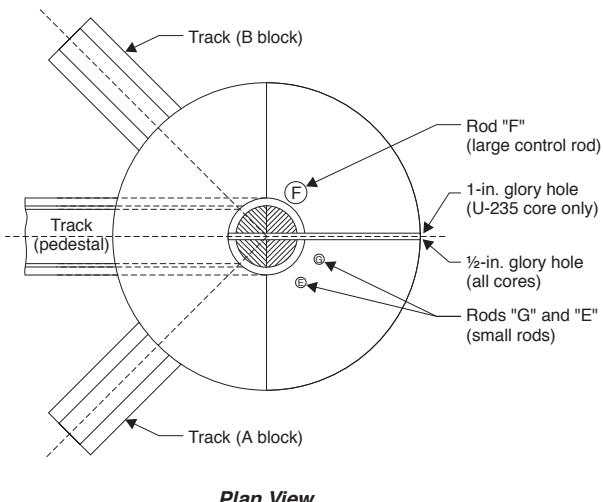
Flat-Top

- 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)

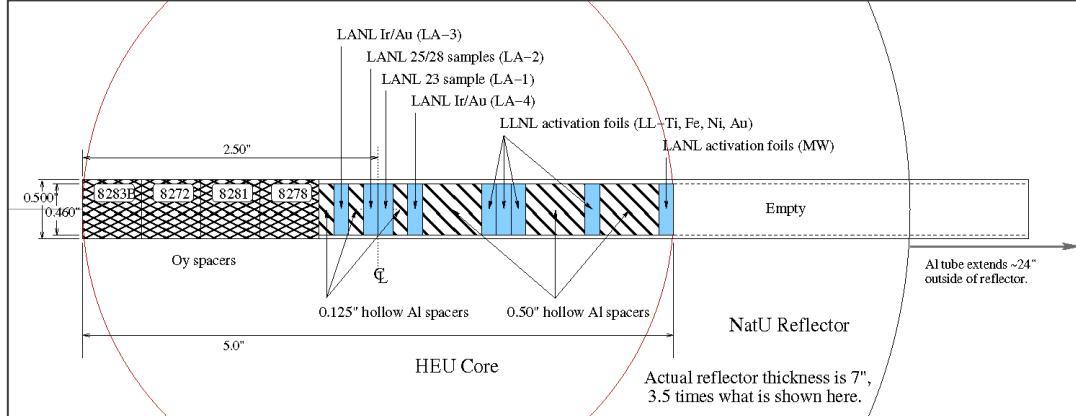
Assembly	Date	233U	235U	238U	237Np	239Pu	R-values	Actinides	FC	PNNL	LLNL	Sponsor(s)
Comet/ZEUS	Sep-11	✓	✓		✓		✓	✓	✓			NA-10/80
Comet/ZEUS	Sep-12	✓	✓				✓	✓		✓		NA-22
Flattop (half)	Jun-13	✓	✓	✓			✓	✓		✓		NA-22
Flattop (half)	Aug-14	✓	✓	✓			✓	✓		✓		NA-22
Flattop (full)	Apr-15	✓	✓		✓	✓	✓	✓		✓		NA-22
Planet (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

Flattop

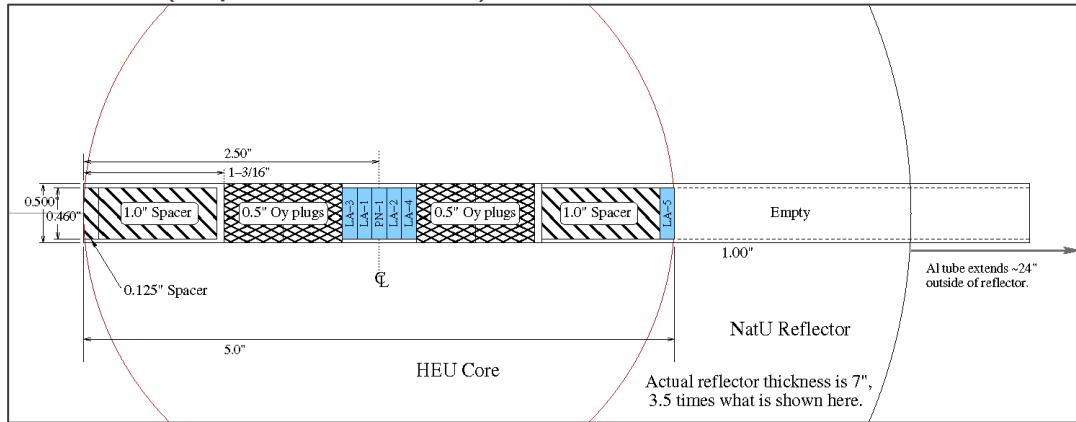


Flat-Top Configurations

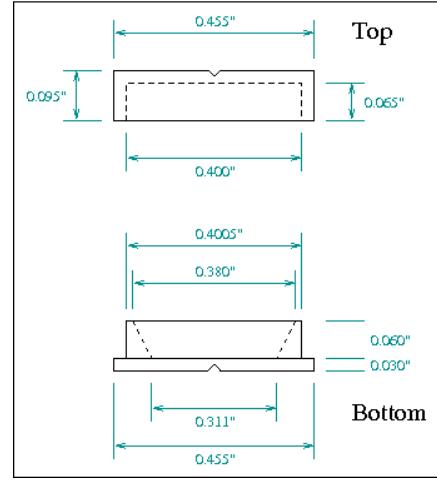
2013 / 2014



2015 (Experiment #4194)



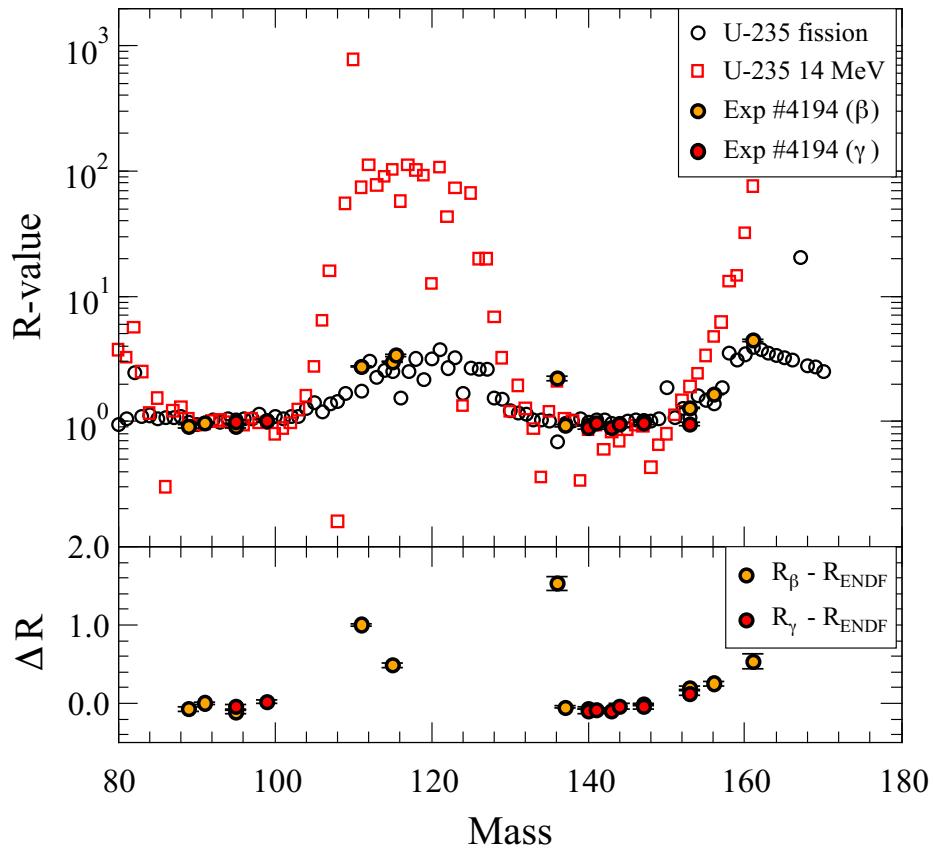
Sample Capsules



Results For Experiment #4194 (HEU)

	Flattop Run	4/27/15				Flattop Run	4/27/15				Flattop Run	4/27/15	
	Separated Beta	FINAL				Separated Gamma	FINAL				Separated Gamma	PNNL	U-235
	Exp #4194	Atoms/g of "A" Sol'n	Uncert. [%]	R _B	Uncert. [%]	Exp #4194	Atoms/g of "A" Sol'n	Uncert. [%]	R _γ	Uncert. [%]	R _γ	Uncert. [%]	
⁸⁹ Sr		1.421E+09	3.60%	0.92	3.40%	OK	--	--	--	--	0.98	5.10%	
⁹⁰ Sr	--	--	--	--	--		--	--	--	--	--	--	
⁹¹ Y		1.840E+09	1.70%	0.97	0.90%	OK	--	--	--	--	--	--	
⁹⁵ Zr		1.956E+09	3.30%	0.92	2.90%	OK	2.099E+09	2.60%	0.99	3.00%	OK	0.97	2.70%
⁹⁷ Zr	--	--	--	--	--		--	--	--	--	0.98	5.60%	
⁹⁹ Mo		1.996E+09	1.40%	3.267E+10	1.40%	³ OK	2.029E+09	2.20%	1.02	2.60%	OK	--	--
¹⁰³ Ru	--	--	--	--	--		--	--	--	--	1.10	3.20%	
¹⁰⁵ Rh	--	--	--	--	--		--	--	--	--	--	--	
¹¹¹ Ag		1.531E+07	1.50%	2.69	0.60%	OK	--	--	--	--	3.30	7.30%	
¹¹⁵ Cd		1.142E+07	1.70%	3.01	0.90%	OK	--	--	--	--	3.14	7.80%	
^{115m} Cd		1.105E+06	2.10%	3.38	1.50%	OK	--	--	--	--	--	--	
¹³² Te	--	--	--	--	--		--	--	--	--	1.11	3.50%	
¹³⁶ Cs		4.311E+06	4.20%	2.22	4.00%	OK	--	--	--	--	2.47	3.50%	
¹³⁷ Cs		1.873E+09	2.40%	0.93	2.00%	OK	--	--	--	--	0.93	5.40%	
¹⁴⁰ Ba		1.904E+09	1.80%	0.94	1.10%	OK	1.822E+09	3.10%	0.90	3.40%	OK	0.97	3.60%
¹⁴¹ Ce		2.051E+09	11.50%	1.07	11.40%	OK	1.854E+09	2.15%	0.97	2.57%	OK	0.97	3.70%
¹⁴³ Ce		1.882E+09	11.80%	0.97	11.70%	OK	1.741E+09	2.28%	0.89	2.68%	OK	0.96	2.60%
¹⁴⁴ Ce		1.528E+09	14.40%	0.89	14.40%	OK	1.714E+09	3.66%	0.95	3.93%	OK	0.87	3.80%
¹⁴⁷ Nd		7.398E+08	1.90%	1.01	1.30%	OK	7.183E+08	2.90%	0.98	3.20%	OK	0.99	4.90%
¹⁵³ Sm		6.595E+07	1.90%	1.28	1.30%	OK	4.914E+07	2.70%	0.95	3.00%	OK	1.20	9.90%
¹⁵⁶ Eu		8.062E+06	2.00%	1.66	1.40%	OK	--	--	--	--	1.70	6.50%	
¹⁶¹ Tb		1.249E+05	2.50%	4.48	2.10%	OK	--	--	--	--	--	--	

Results For Experiment #4194 (HEU)

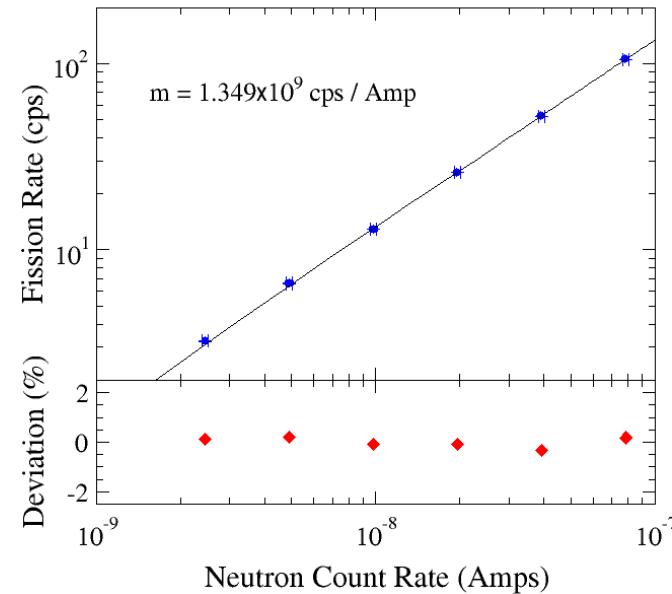
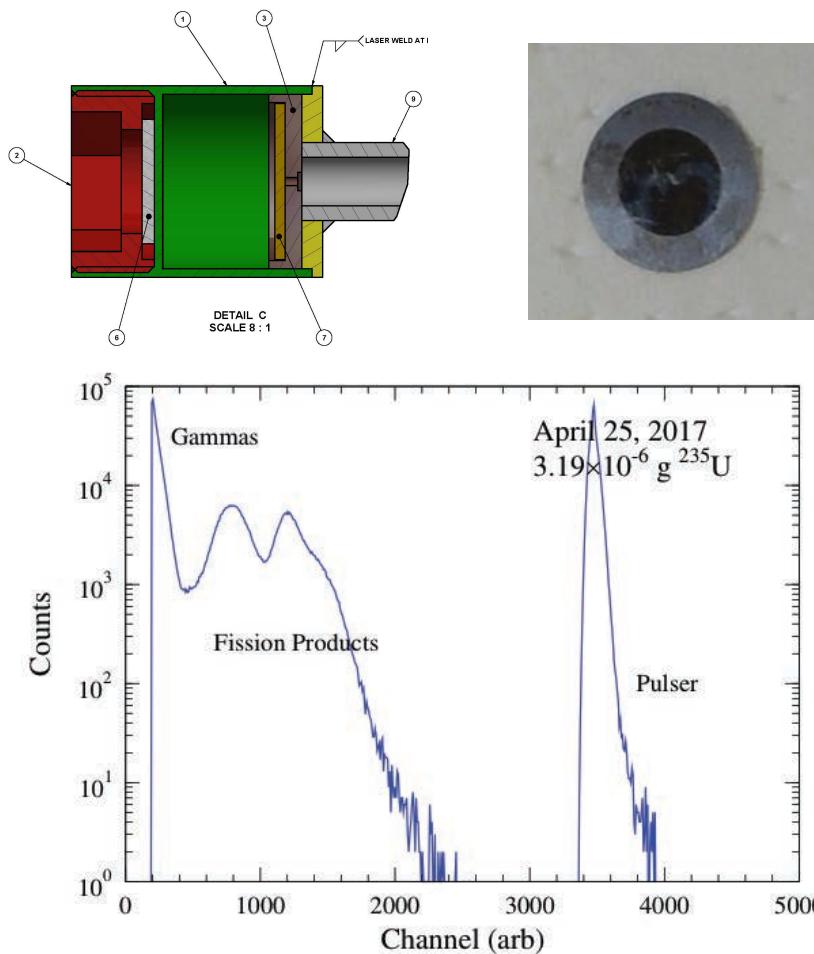


Experimentally determined R-values for ^{235}U in the center position of Flattop compared to R-values calculated from ENDF/B-VII.1 fission product yields for 0.5 and 14 MeV incident neutrons.

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
$^{111}\text{Ag} (\beta)$	1.76	2.76	n/a
$^{111}\text{Ag} (\gamma)$	1.76	n/a	3.30
$^{136}\text{Cs} (\beta)$	0.69	2.22	n/a
$^{136}\text{Cs} (\gamma)$	0.69	n/a	2.47
$^{153}\text{Sm} (\beta)$	1.08	1.28	n/a
$^{153}\text{Sm} (\gamma)$	1.08	0.95	1.20

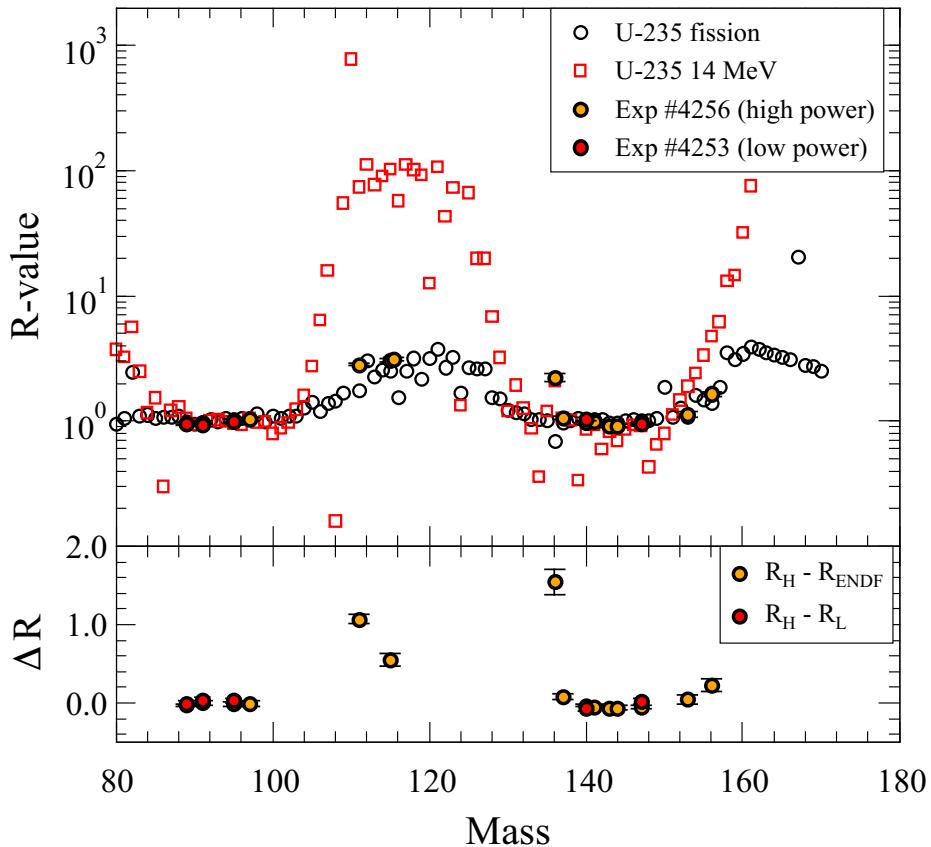
Prototype Fission Chamber Design and Testing



Observations:

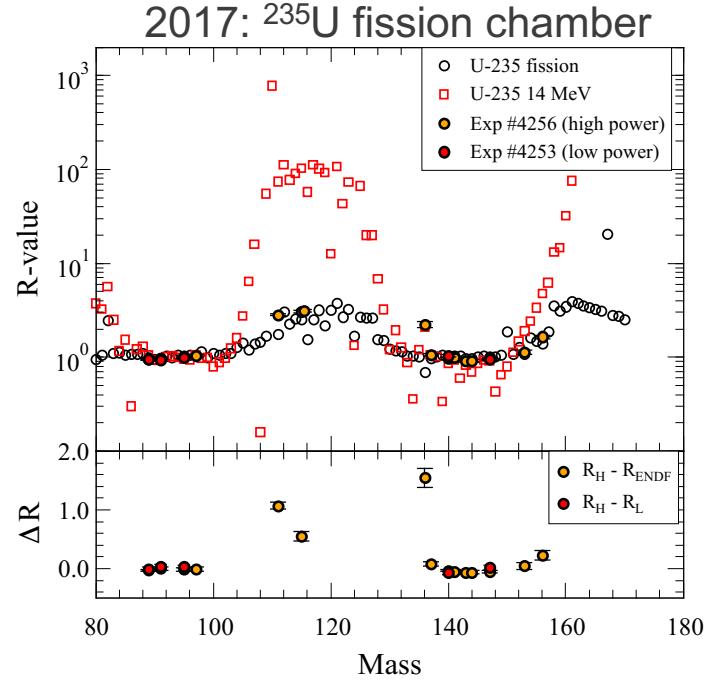
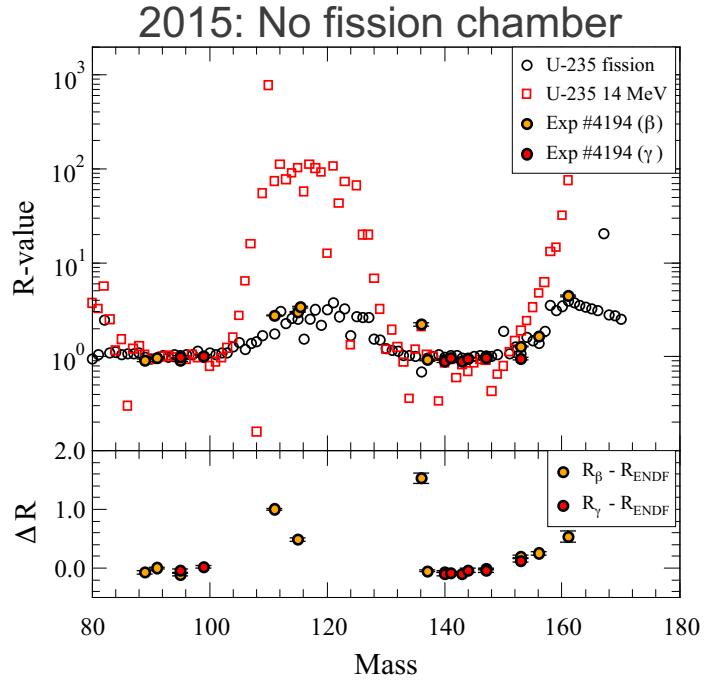
- At low power levels the prototype FC provided excellent energy resolution given its small size.
- The gamma rate from the assembly fuel presaged issues with running the fission chamber at the power level required for full radiochemical analysis.

Results For Experiments #4253 and 4256 (HEU)

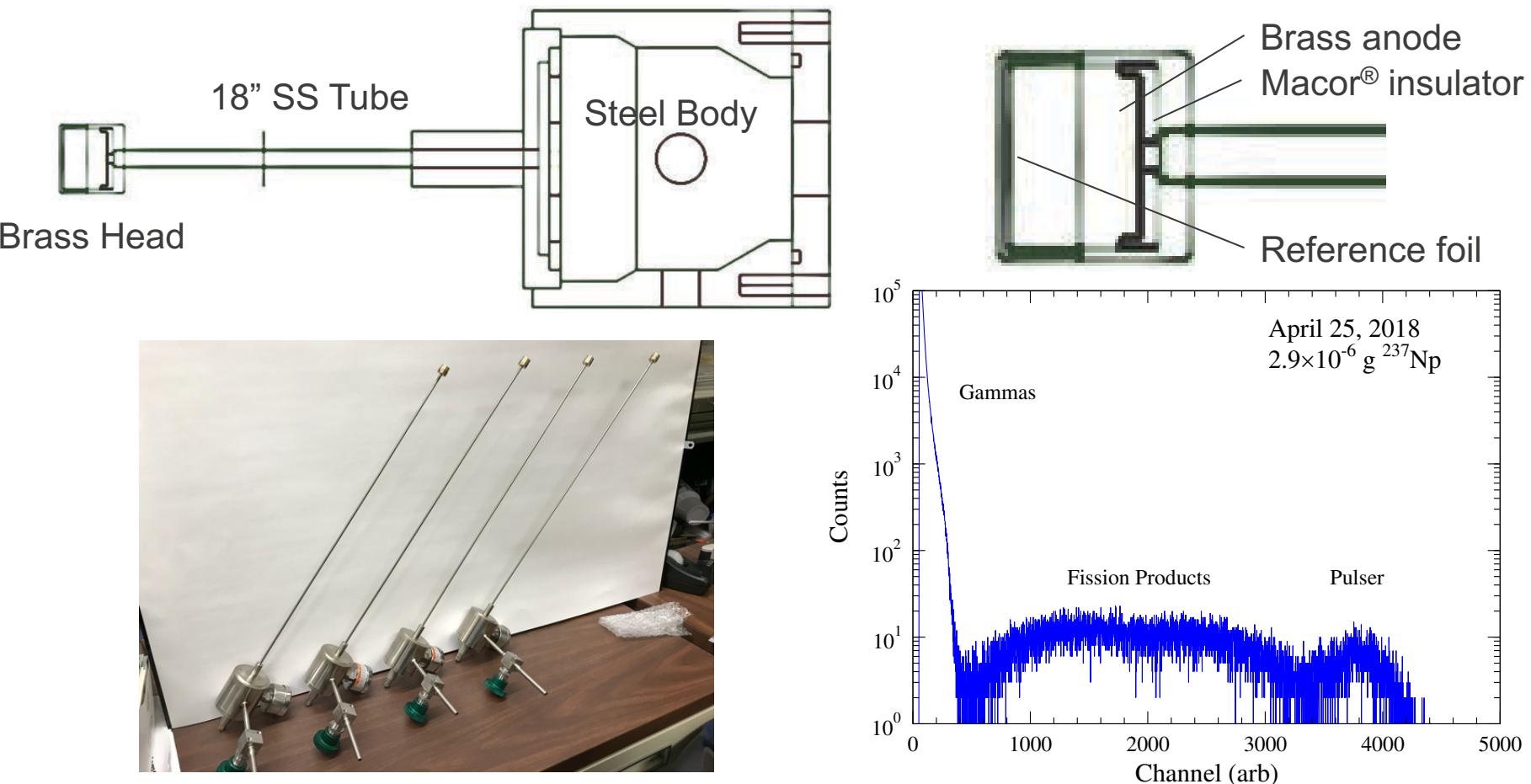


- Comparison of current R-value results with R-values calculated from ENDF/B-VII.0 cumulative fission product yields for fission of ²³⁵U from 0.5 (fission) and 14 MeV neutrons.
- Difference, $\Delta R = R_H - R_n$, between R-values determined from the high-power run (H) and those calculated for ENDF fission ($n = \text{ENDF}$) 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.

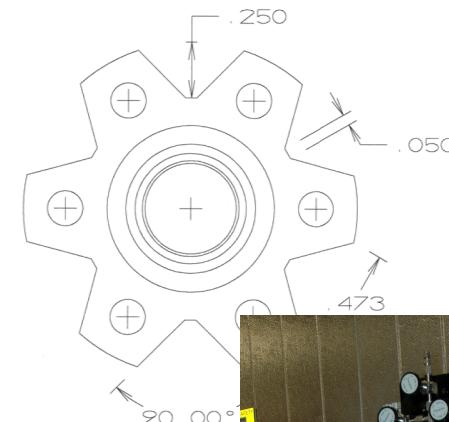
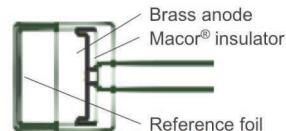
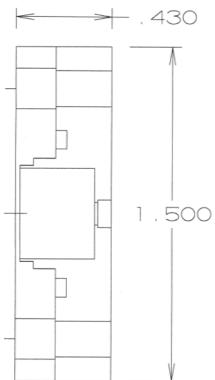
Comparison



Mark II Fission Chamber



Tests at McClellan Nuclear Research Center (UC Davis)



Testing U(93) reference
foils at MNRC in July
OK, 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.

TUNL (LANL/LLNL/Duke):

- Energy differential foil irradiations using monoenergetic neutrons.
 ^{235}U , ^{238}U and ^{239}Pu
- Collocated fission chamber and direct foil counting to determine absolute CFPYs.
- Currently developing capability to measure short-lived FPYs.

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.

MIT/NRL (LANL/LLNL/Duke):

- Thermal neutron foil irradiations.
 ^{235}U (^{239}Pu)
- Collocated fission chamber and direct foil counting to determine absolute FPYs.
- Planning to add radiochemical analyses.

ILL/Grenoble (CEA-DAM):

- Thermal energy solution irradiations.
 ^{235}U
- Collocated fission tag (LSC) and direct sample 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.

TUNL (LANL/LLNL/Duke/CEA-DAM):

- Energy differential actinide foil irradiations using monoenergetic neutrons.
 ^{235}U , ^{238}U and ^{239}Pu
- Collocated fission chambers and direct foil counting to determine absolute CFPYs and short-lived FPYs.

MIT/NRL (LANL/LLNL/Duke/CEA-DAM):

- Measure detection efficiency for CEA fission chambers.
- Thermal neutron foil irradiations.
 ^{235}U and ^{239}Pu
- Direct foil counting and radiochemical analyses to determine relative FPYs.
- Collocated fission chambers to determine absolute FPYs.

Farther forward:

- Move on to other actinides.

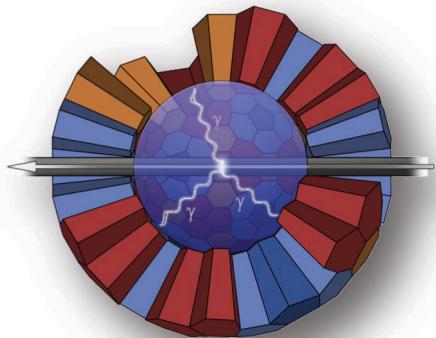
Acknowledgements

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Spares

Instruments at LANSCE



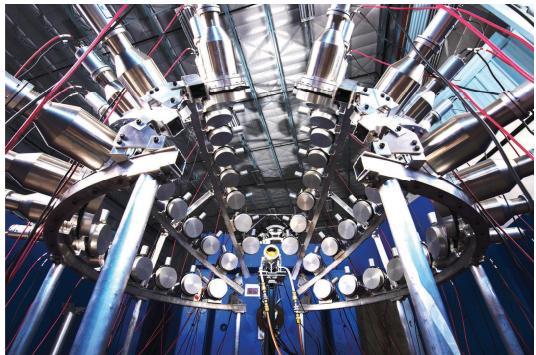
DANCE

- Studies of neutron capture and gamma decay
- Can be combined with PPAC for capture measurements on fissile isotopes



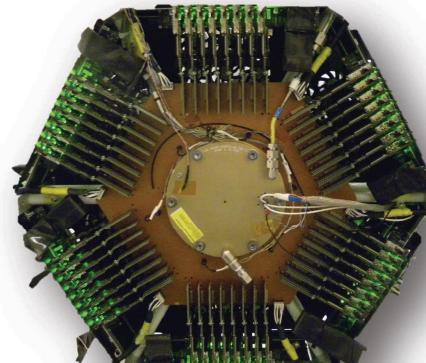
SPIDER

- High-resolution measurements of fission fragment yields as a function of A, Z, incident En , and total kinetic energy
- Uses “2E-2V” method with 8 arm pairs— MCPs for fast timing/velocity component and axial ionization chambers for fission fragment energy



Chi-Nu

- Completing measurements of the PFNS on ^{239}Pu and ^{235}U
- Future efforts focused on minor actinides and neutron scattering measurements for transport
- Uses double time-of-flight technique for incident and emitted neutron energies



TPC

- Designed for precision (n,f) cross-section measurements
- Time Projection Chamber, uses MICROMEGAS amplification and 6000 readout pads for tracking and background suppression

LANL's Approach Uses R-Values, Q-Values: Ratios Allow Us to Minimize Systematic Errors

$$Y_{147}^{Pu,fast} = Y_{147}^{25,th*} \cdot \frac{R_{147}^{Pu,fast}}{\frac{[Y_{147}^{Pu,fast} / Y_{99}^{Pu,fast}]}{[Y_{147}^{25,th} / Y_{99}^{25,th}]}} \cdot \frac{Q_{99}^{Pu,fast}}{\frac{Y_{99}^{Pu,fast}}{Y_{99}^{25,th}}}$$

Often accurately measured
(many thermal reactor
experiments)

R: Precise “Lab independent”
Measurements – ratio of ratios.

Cancellation of systematic errors
in counting FPs (99 and 147)

Cancellation of # of fissions for
A given neutron environment

Q: “lab dependent” and difficult
absolute measurement (K-factors
from fission chamber experiments)
needed for both numerator and
denominator,

BUT, cancellation of systematic errors
in counting FPs (99). Errors in the # of
fissions remain.

Experiments to Date

- Comet/ZEUS – September 2011
 - Irradiated U(93), DU and WG-Pu metal foils
 - Chemistry → R-values + actinide isotopics
- Comet/ZEUS – September 2012
 - Irradiated U(93) and DU metal foils (NO Pu)
 - Chemistry → R-values + actinide activations/isotopics
- Flattop (center) – June 2013
 - Irradiated U(93) and DU metal, and $^{233}\text{U}_3\text{O}_8$
 - Chemistry → R-values + actinide activations/isotopics.
- Flattop (center) – August 2014
 - Irradiated U(93) and DU and ^{237}Np metal
 - Chemistry → R-values + actinide activations/isotopics
- Flattop (center) – April 2015
 - Irradiated U(93) and DU and ^{239}Pu metal foils
 - Chemistry → R-values + actinide activations/isotopics
- Flattop (center) – March 2017
 - Irradiated U(93), DU and ^{237}Np metal foils
 - Chemistry → R-values + actinide activations/isotopics
- Flattop (center) – April 2017
 - Irradiated U(93) and DU metal foils + U(93) FC
 - Chemistry → R-values + actinide activations/isotopics
- Flattop (center) – April 2018
 - Irradiated U(93), DU and ^{237}Np metal + ^{237}Np FC
 - Chemistry → R-values + actinide activations/isotopics
- Two Custom Assemblies on Planet 2016-2018
 - Irradiated U(93) and DU metal foils
 - Chemistry → R-values + actinide activations/isotopics