LA-UR-19-30219

Title:	Fission Product Chain Yield Measurements
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Fission Product Chain Yield Measurements

Todd Bredeweg

2019 DNP Meeting
Crystal City, VA
October 14-17, 2019

LA-UR-19-NNNNN

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

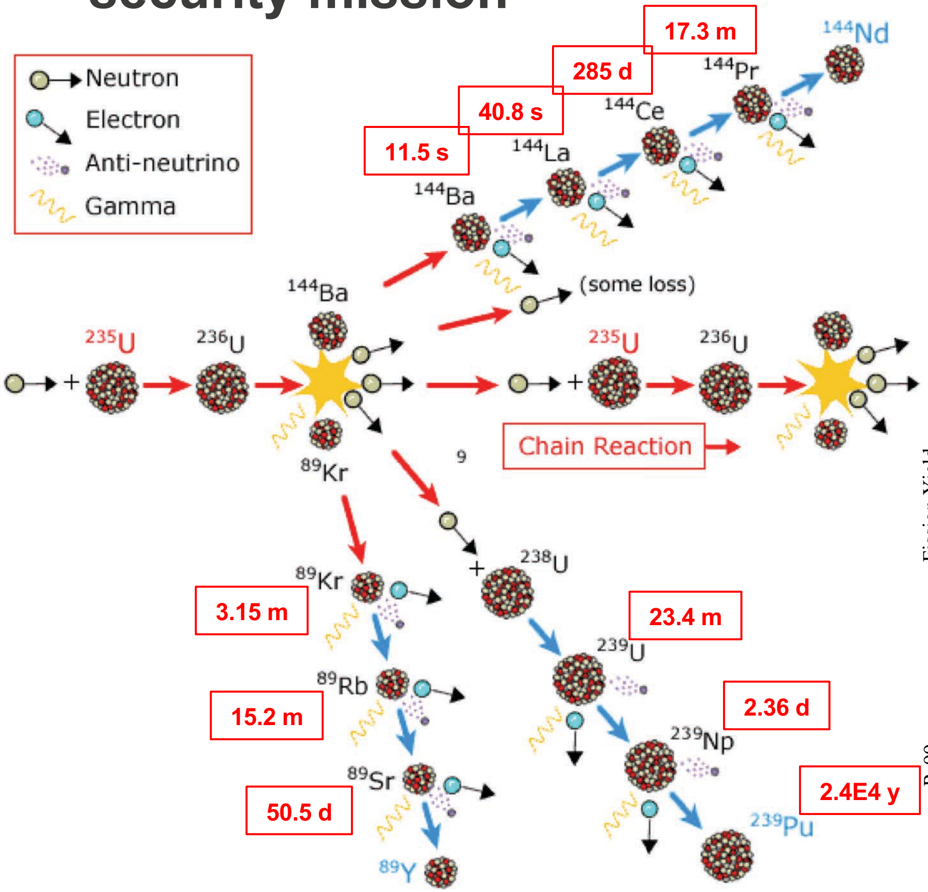
- Direct γ-ray counting of the irradiated samples to determine relative ($R_i^{j,k}$) cumulative FPYs and associated reaction rates.

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

Accomplishments/Results:

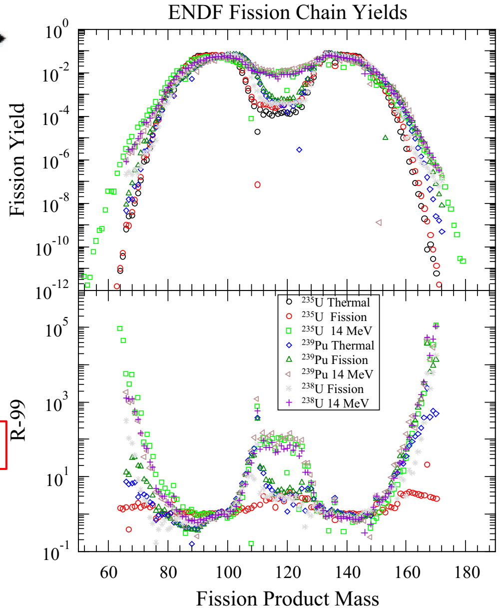
- Fission spectrum irradiations of ^{233}U, ^{235}U, ^{238}U, ^{237}Np and ^{239}Pu at NCERC. For each actinide, relative fission product yields were determined by radiochemical analysis.
- Monoenergetic irradiations of ^{235}U, ^{238}U and ^{239}Pu with collocated fission chambers at TUNL. Relative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

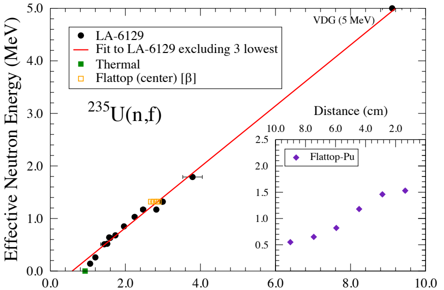
Why is nuclear physics / nuclear data important to the national security mission

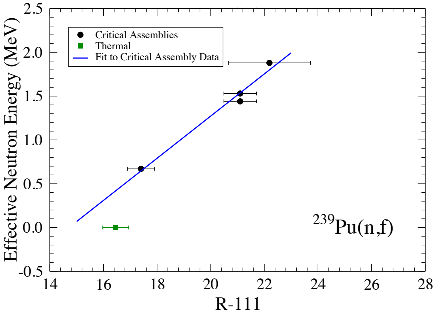


The defining signature of a nuclear detonation is the presence of nuclear reaction products in the debris. These can be from

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

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

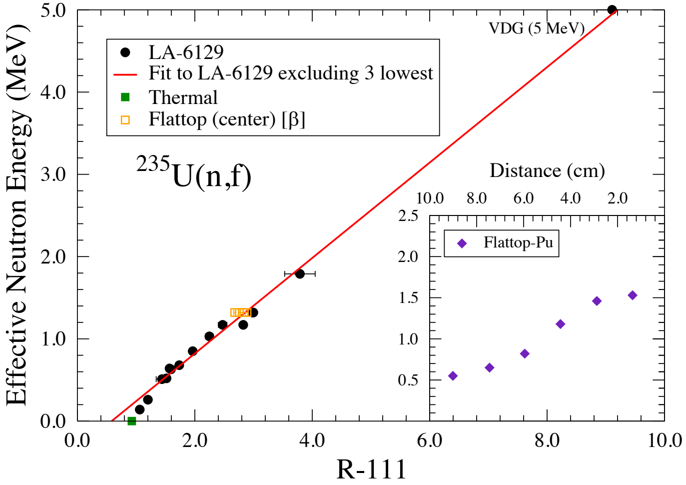
- Radiochemical (RC) analysis of the macro-foil provides $A_i^{j,k}$. The atom number and FPY is then determined by the standard equations

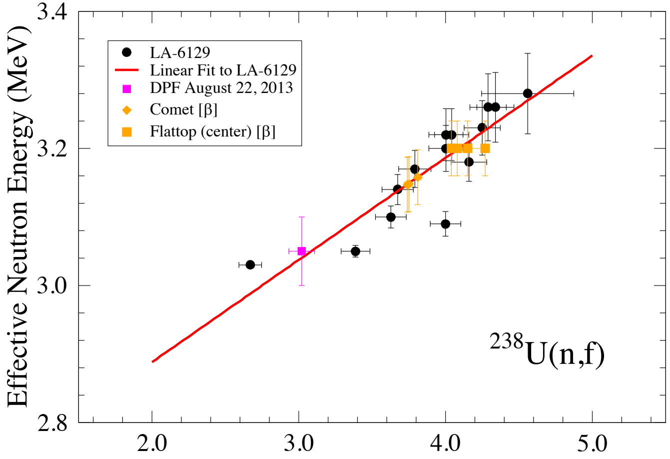
$$N_i^{j,k} = \frac{A_i^{j,k}}{\lambda_i \epsilon_i} = \frac{(1+\alpha_i)A_i^{j,k}}{\lambda_i \epsilon_i f_i}$$

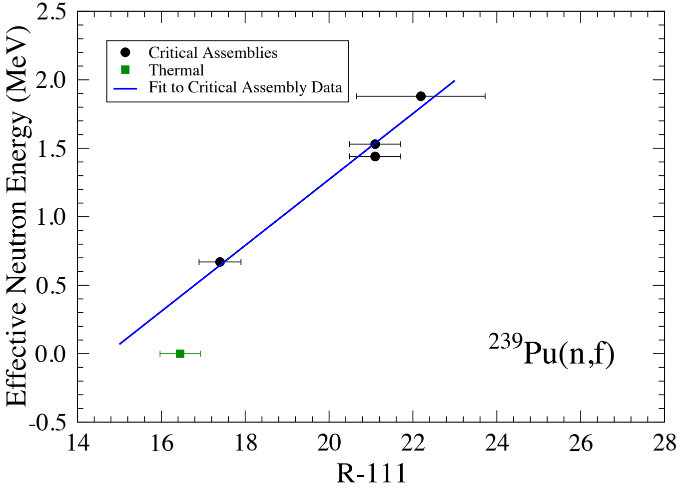
$$Y_i^{j,k} = \frac{N_i^{j,k}}{N_f^*} = \frac{(1+\alpha_i)A_i^{j,k}}{\lambda_i \epsilon_i f_i N_f^*}$$

- 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



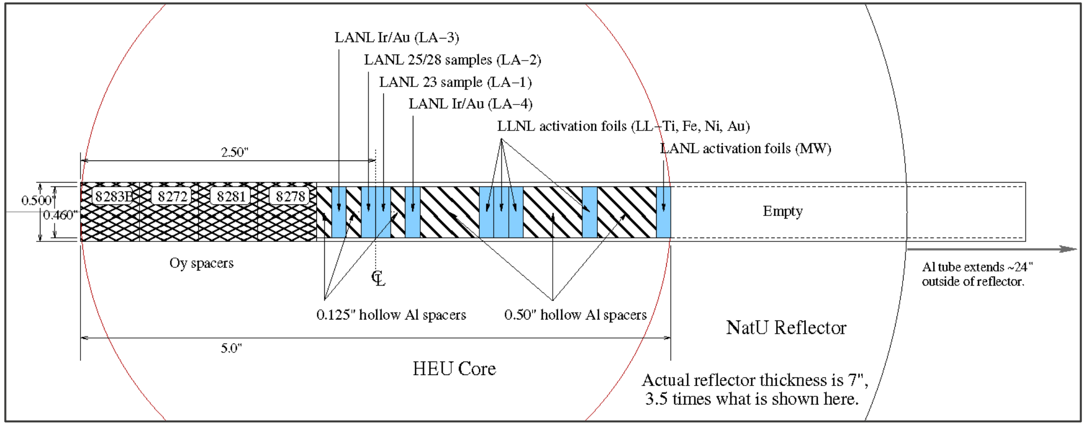




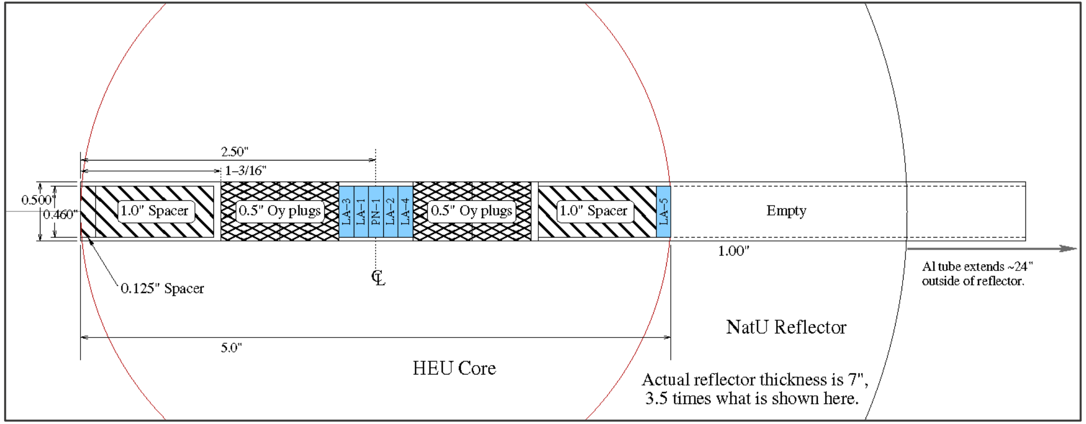
- G.P. Ford and A.E. Norris, LA-6129 (1976)
- H.D. Selby, et al., Nucl Data Sheets 111, 2891 (2010)
- M.B. Chadwick, et al., Nucl Data Sheets 111, 2923 (2010)
- J. Laurec, et al., Nucl Data Sheets 111, 2965 (2010)

Flat-Top Configurations

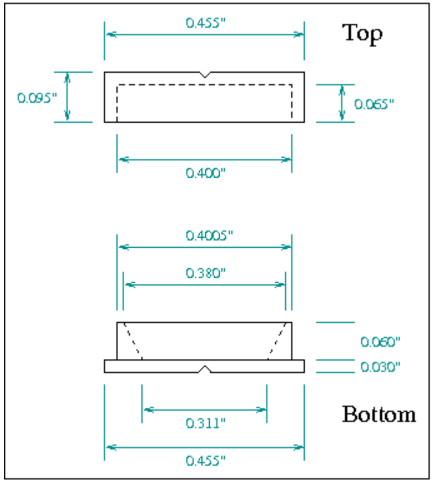
2013 / 2014



2015 (Experiment #4194)



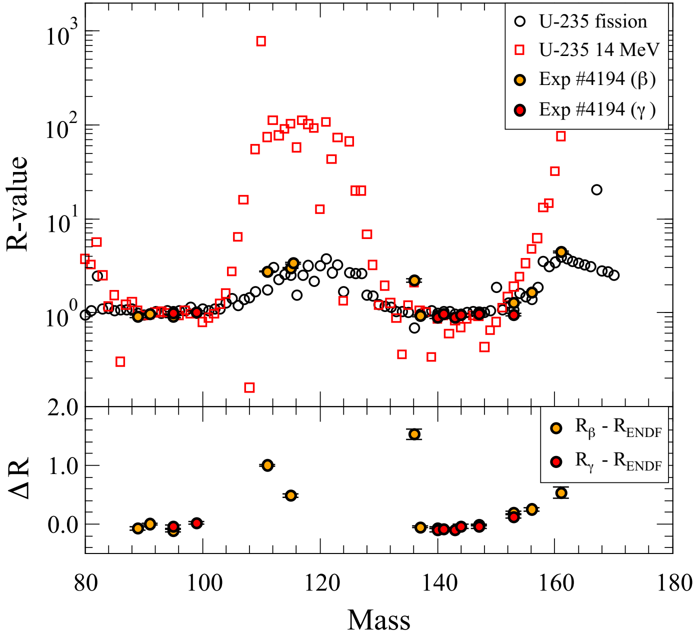
Sample Capsules



Results For Experiment #4194 (HEU)

	Flattop Run	4/27/15				Flattop Run	4/27/15				Flattop Ru	4/27/15
	Separated Beta Exp # 4194	FINAL				Separated Gamma Exp # 4194	FINAL				Separated Gamma PNNL	U-235
	Atoms/g of "A" Sol'n	Uncert. [%]	R_β	Uncert. [%]		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%	[a]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	--	--	--	--		--	--

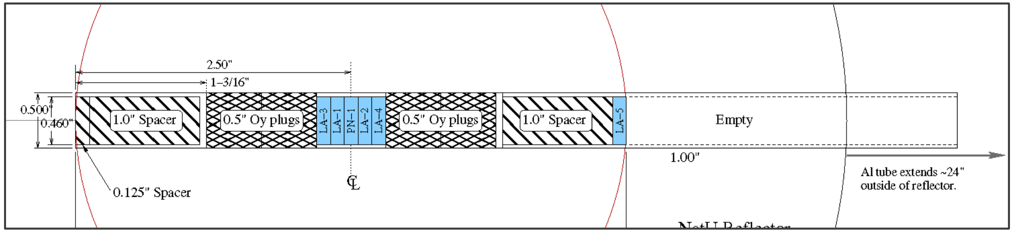
Results For Experiment #4194 (HEU)



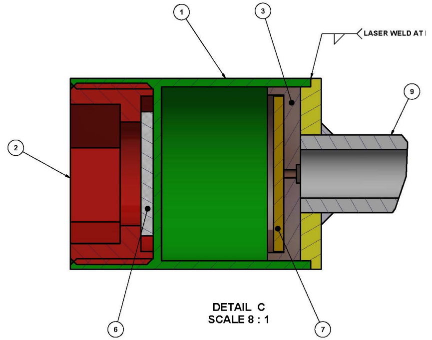
Experimentally determined R-values for ^{235}U in the center position of Flattop compared to R-values calculated from ENDF/B-VII.1 fission product yields for 0.5 and 14 MeV incident neutrons.

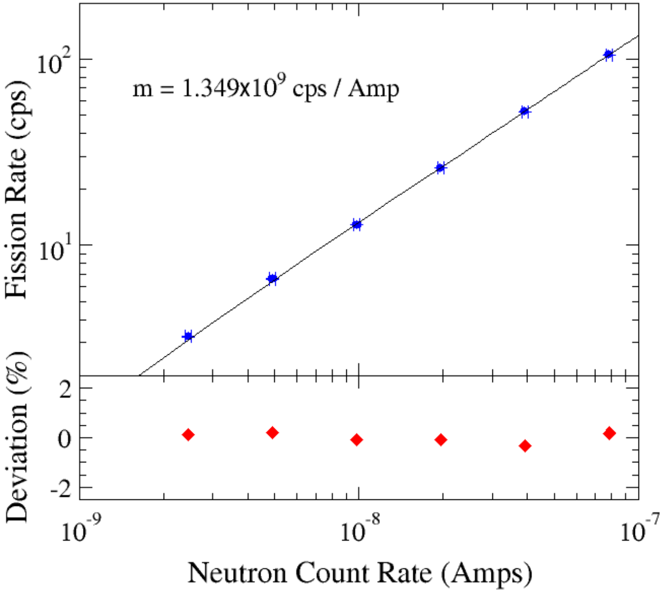
The lower panel shows the absolute difference between the values determined by β counting (black circles) and γ counting (red circles) and ENDF/B-VII.1.

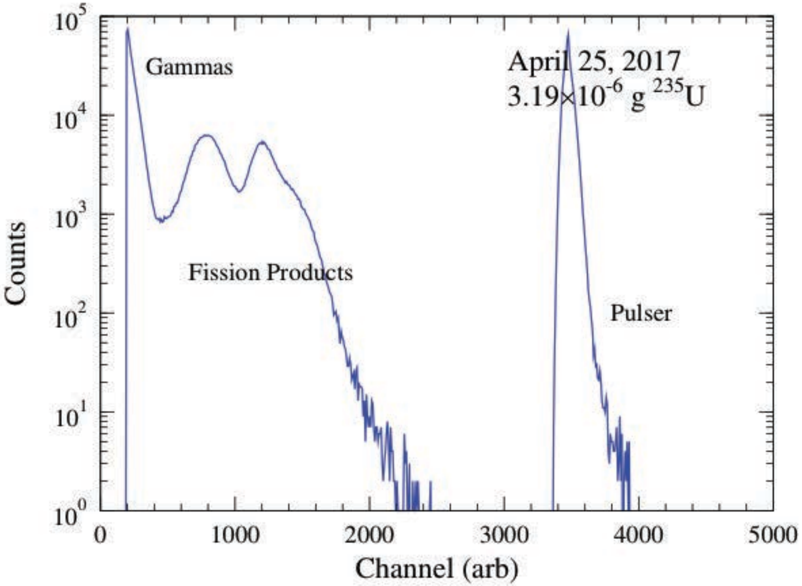
Outlier	ENDF/B	LANL	PNNL
^{111}Ag (β)	1.76	2.76	n/a
^{111}Ag (γ)	1.76	n/a	3.30
^{136}Cs (β)	0.69	2.22	n/a
^{136}Cs (γ)	0.69	n/a	2.47
^{153}Sm (β)	1.08	1.28	n/a
^{153}Sm (γ)	1.08	0.95	1.20



Prototype Fission Chamber Design and Testing



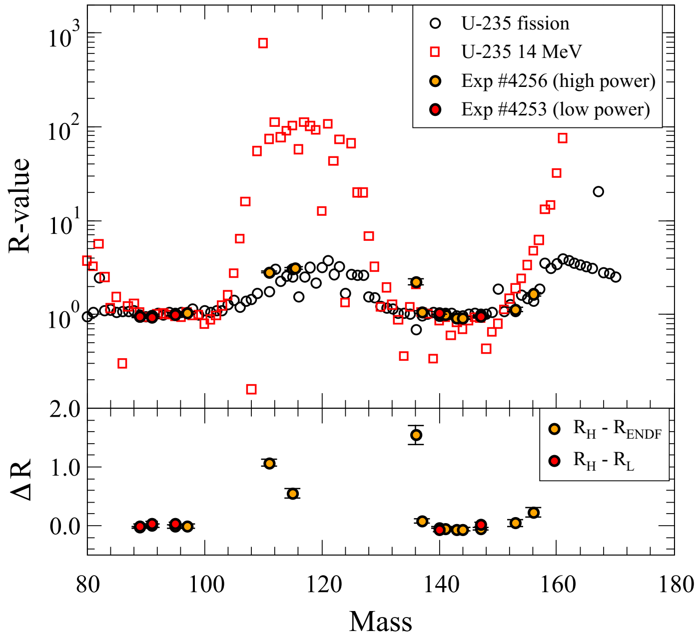


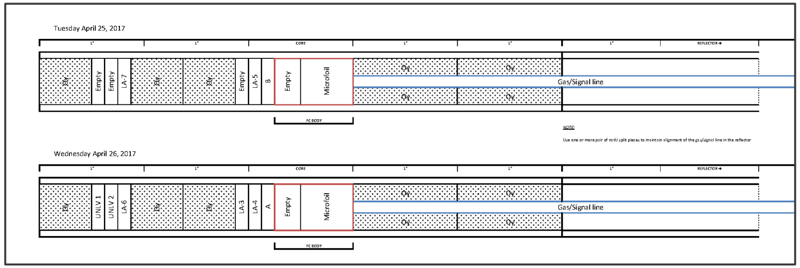


Observations:

- 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.

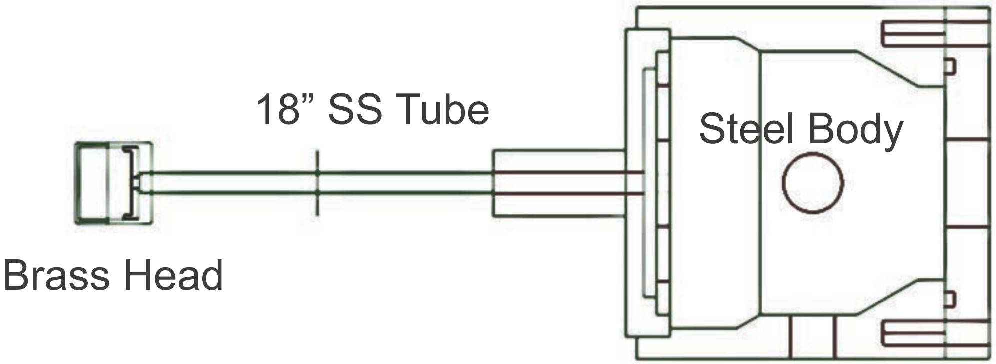
Results For Experiments #4253 and 4256 (HEU)

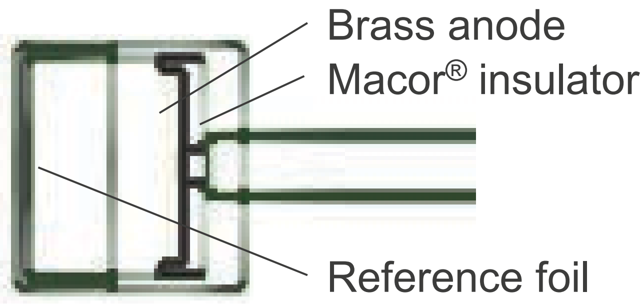


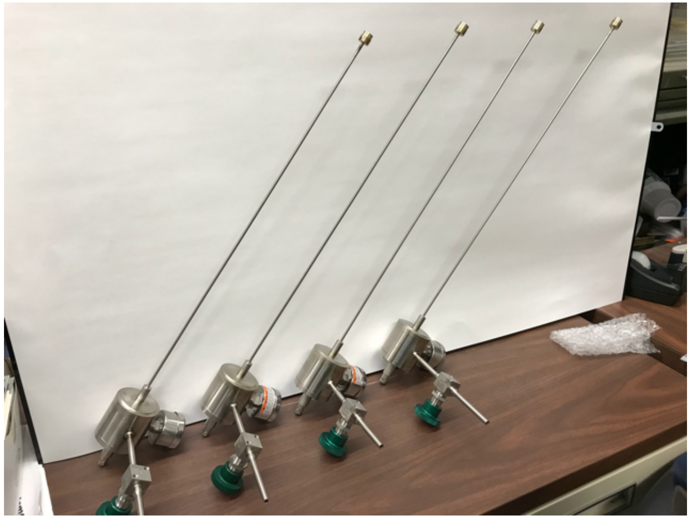


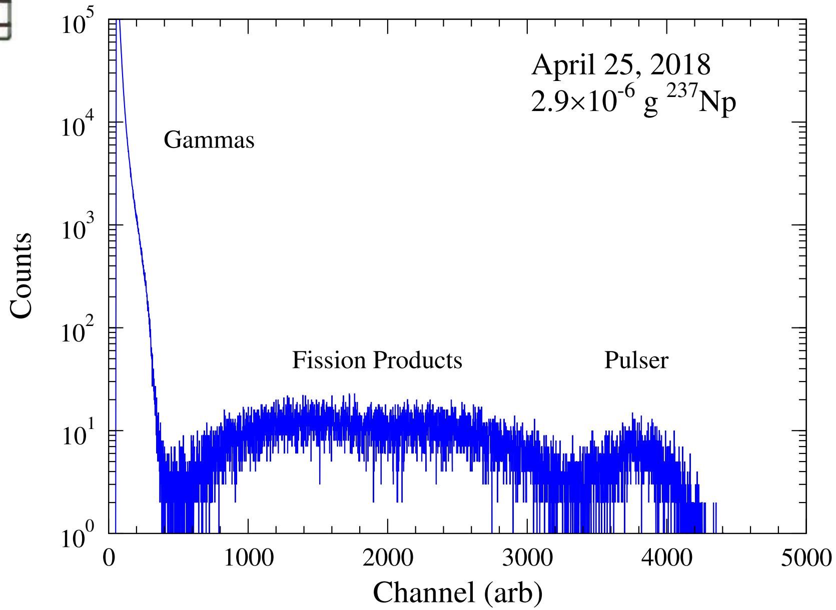
- Comparison of current R-value results with R-values calculated from ENDF/B-VII.0 cumulative fission product yields for fission of ^{235}U from 0.5 (fission) and 14 MeV neutrons.
- Difference, $\Delta R = R_H - R_n$, between R-values determined from the high-power run (H) and those calculated for ENDF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

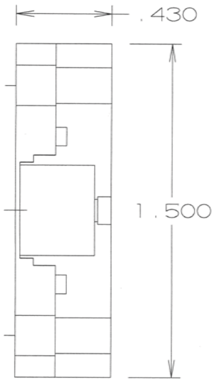


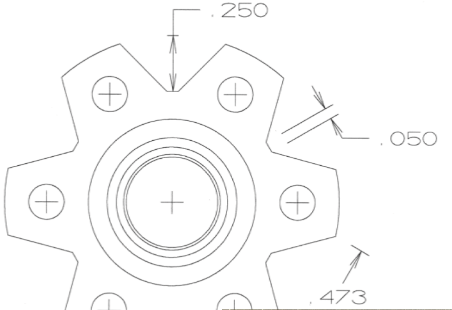


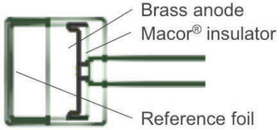




Tests at McClellan Nuclear Research Center (UC Davis)







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.

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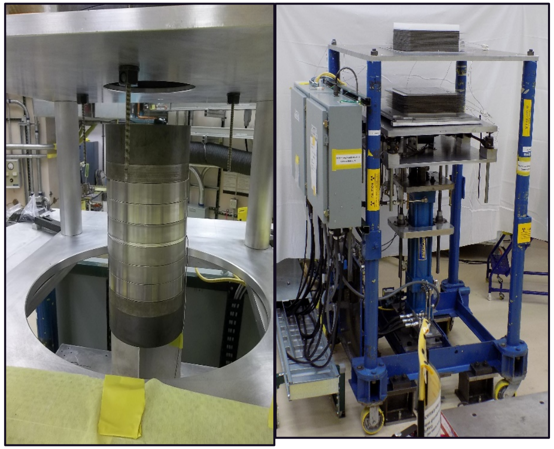
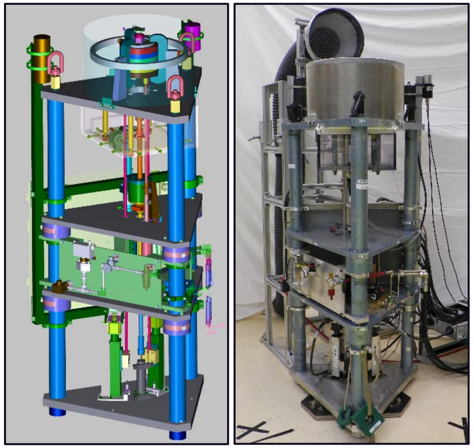
Spares

Critical Assemblies at NCERC



Planet

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

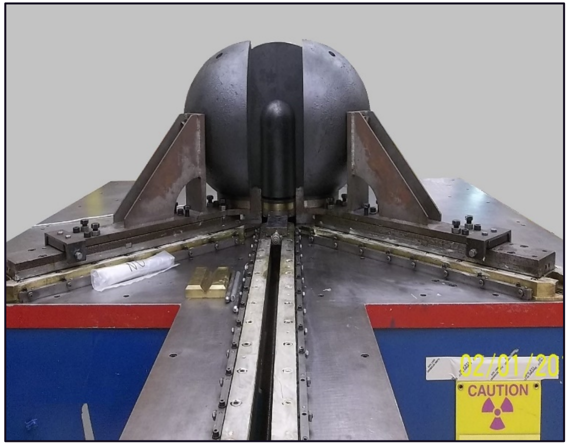


Godiva-IV

- 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

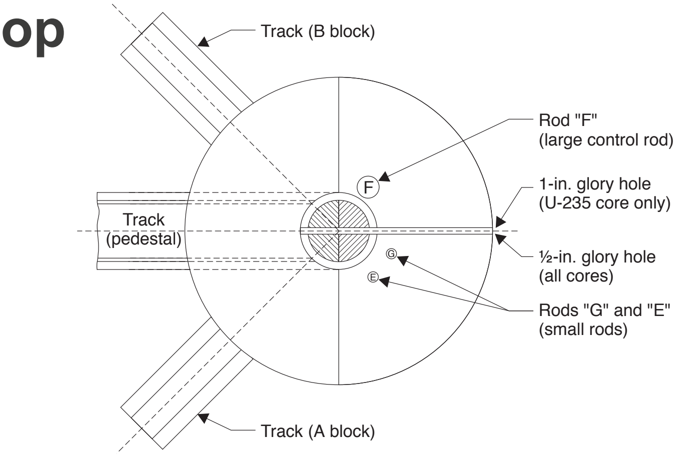
- High Capacity Vertical Lift Assembly
- Supports Large Experiments
- 10^{13} fissions/g on samples, Variable Spectrum



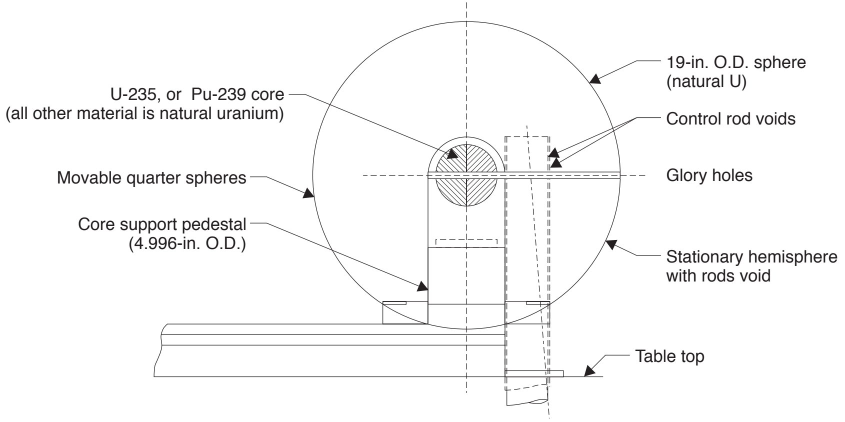
Flat-Top

- 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

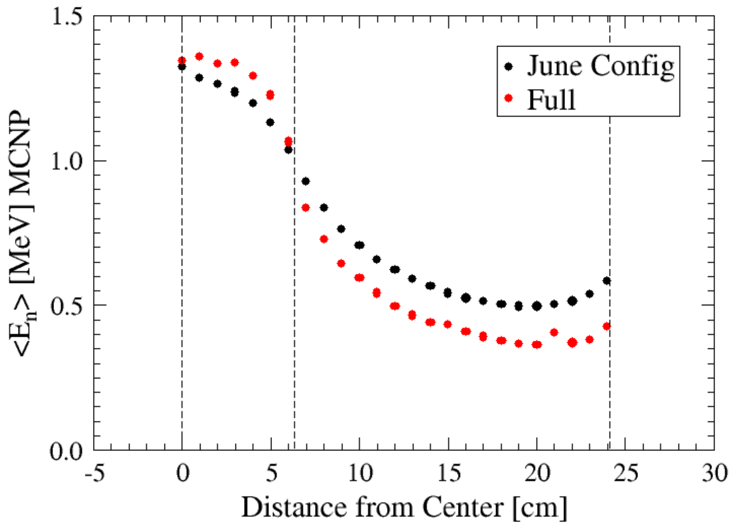
Flattop

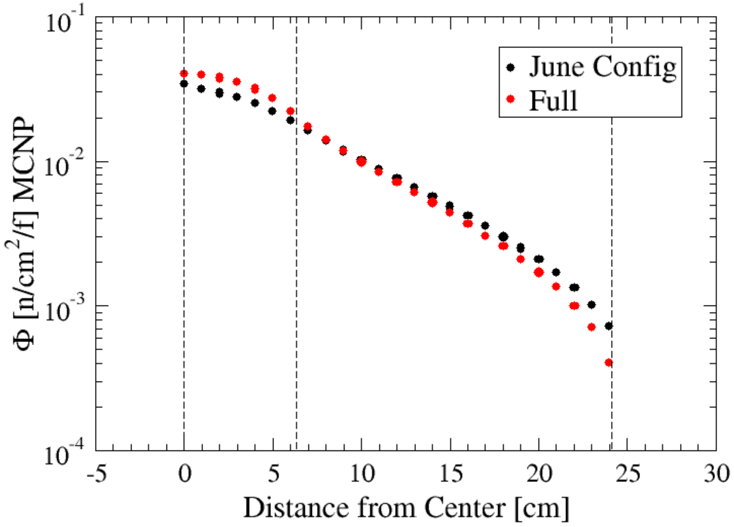


Plan View



Elevation View





LANL’s Approach Uses R-Values, Q-Values:

Ratios Allow Us to Minimize Systematic Errors

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

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 $^{233}U_3O_8$
  - 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