

# **High-Energy, Beta-Delayed Gamma-Ray Spectroscopy Experiments**

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Workshop on Nuclear Data Needs and Capabilities for Applications

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# Delayed Gamma-Ray Spectroscopy for Non-Destructive Assay of Nuclear Materials

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Vladimir Mozin



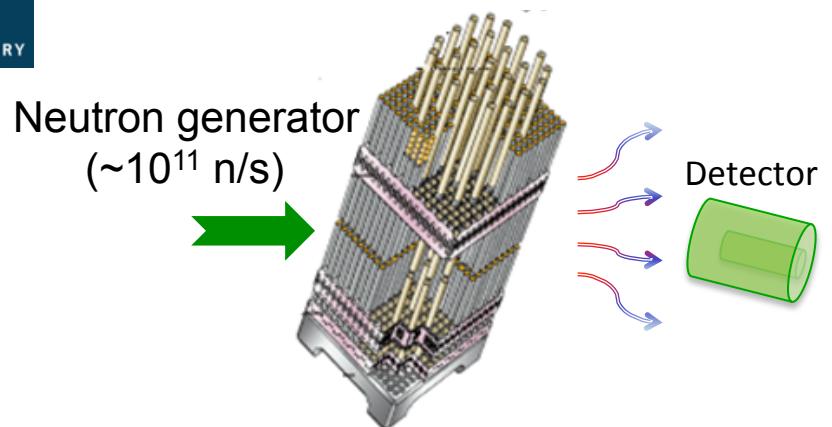
Alan Hunt  
Edward Reedy  
Heather Seipel



Luke Campbell  
Rusty Williford, OSU



Andrea Favalli, LANL



Delayed gamma-ray spectra are analyzed as superposition of U-235, Pu-239, Pu-241, U-238 contributions.



# Delayed Gamma-Ray Experiments

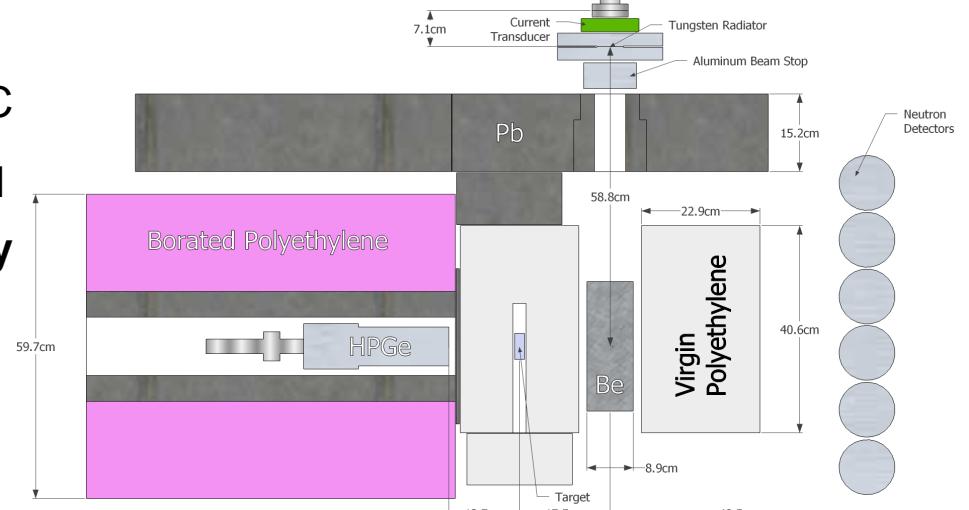
## At IAC

**Photo-neutron source, 15 MeV Electron LINAC**

**Targets:** U-235, Pu-239, Pu-241, DU, Combined

**Data sets for several irradiation/spectroscopy cycles:**

- 15 min/30 min (multiple passes to increase statistics)
- Many cycles: 5 min/5 min, 90 sec/90 sec, 60 sec/60 sec, 10 sec/10 sec

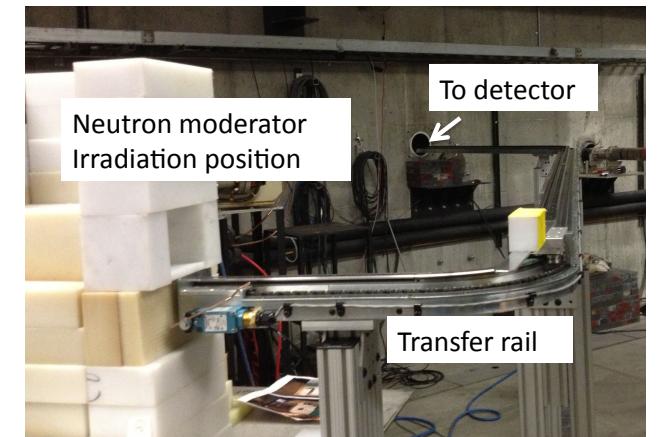
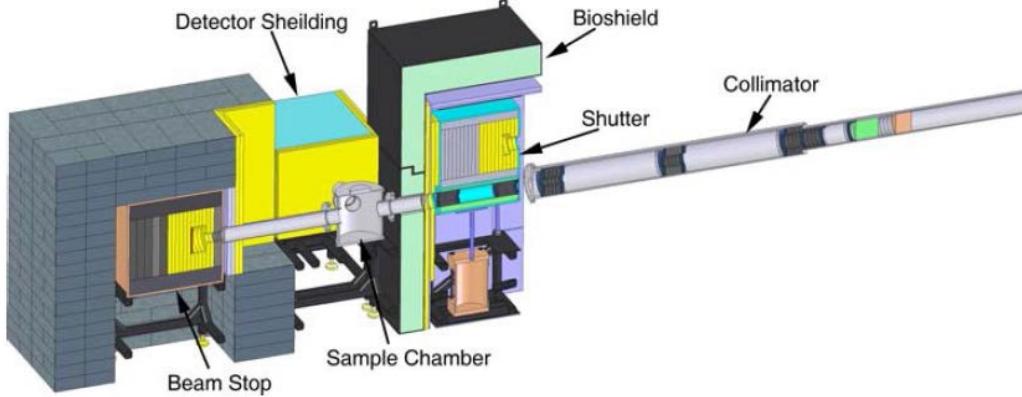


## At OSU

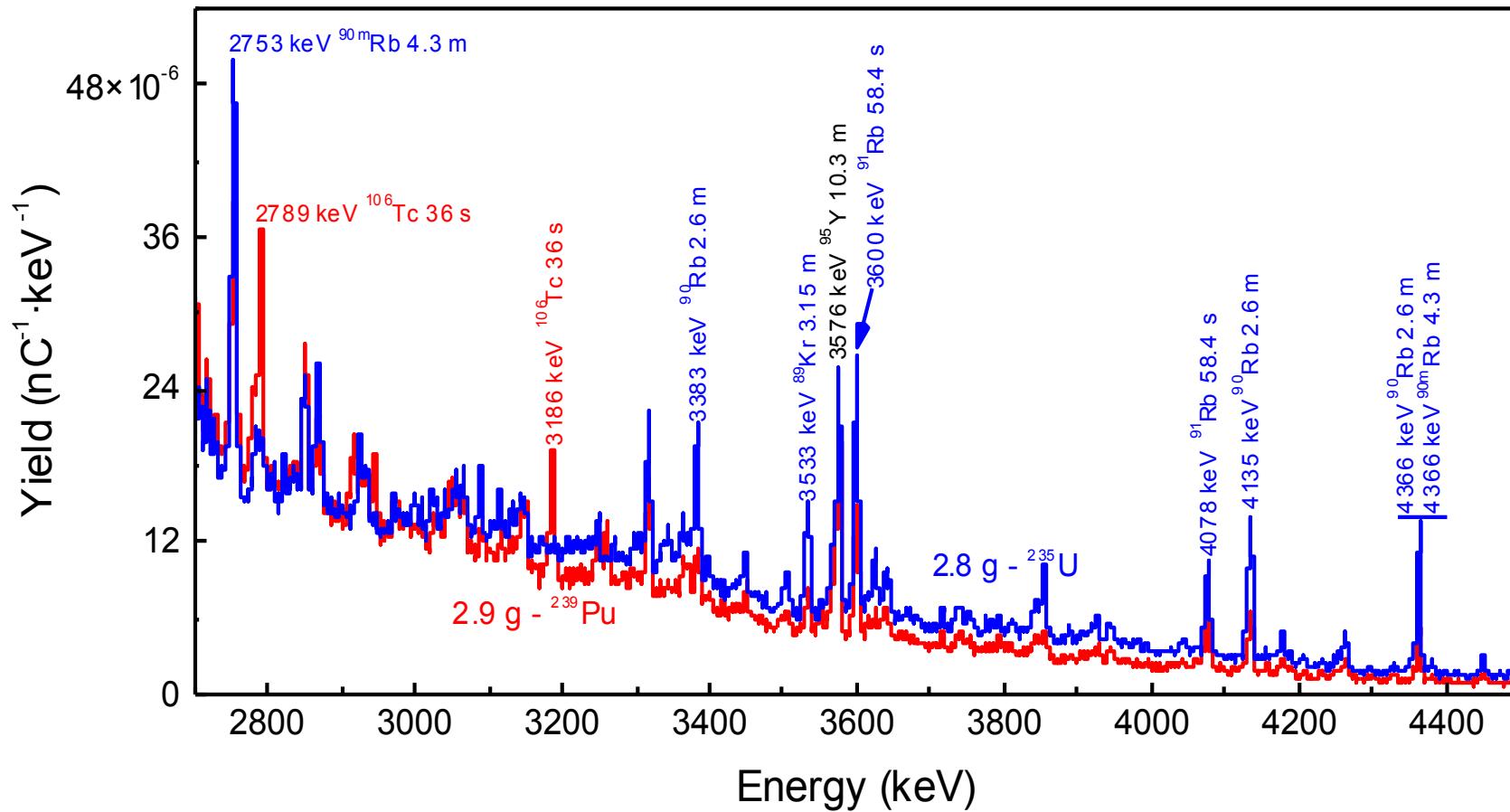
TRIGA reactor, pure thermal neutron beam

**Targets:** U-235, Pu-239, DU

**Irradiation/spectroscopy cycles:** 10/10 sec, 1/1 min, 10/10 min



# Comparison of $^{239}\text{Pu}$ , $^{235}\text{U}$ spectra collected for 90s/90s irradiation/spectroscopy cycle.



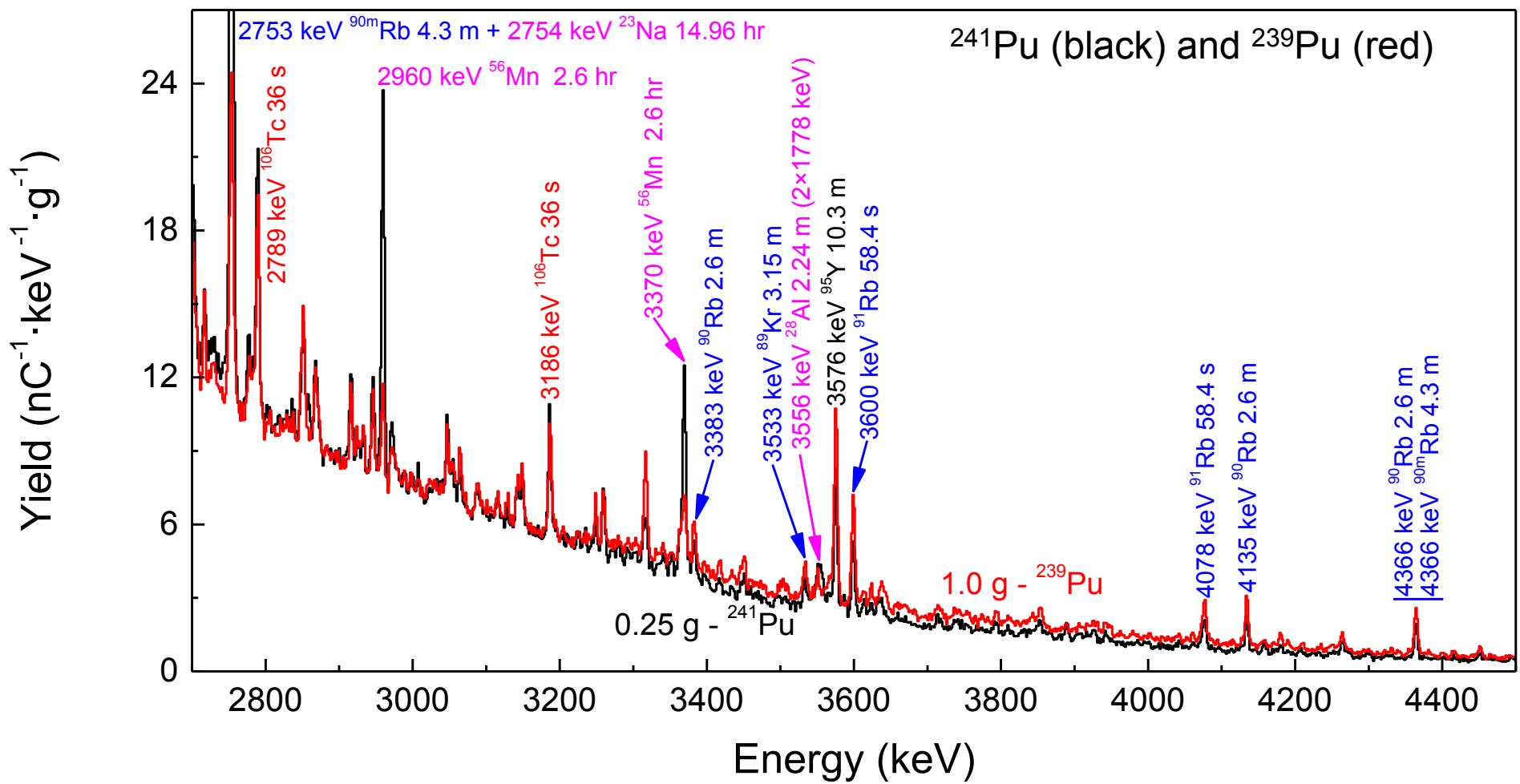
Significant differences between  $^{235}\text{U}$  and  $^{239}\text{Pu}$  for a number of lines:

Strong  $^{235}\text{U}$  indicators include  $^{90}\text{Rb}$  +  $^{90\text{m}}\text{Rb}$  with half-lives of 2.6 and 4.3 min,  $^{91}\text{Rb}$ ,  $t_{1/2} = 58.4$  s.

Strong  $^{239}\text{Pu}$  indicator:  $^{106}\text{Tc}$ ,  $t_{1/2} = 36$  s

$^{95}\text{Y}$ ,  $t_{1/2} = 10.3$  min, is strong in both spectra and important.

# Measured Delayed Gammas from $^{241}\text{Pu}$



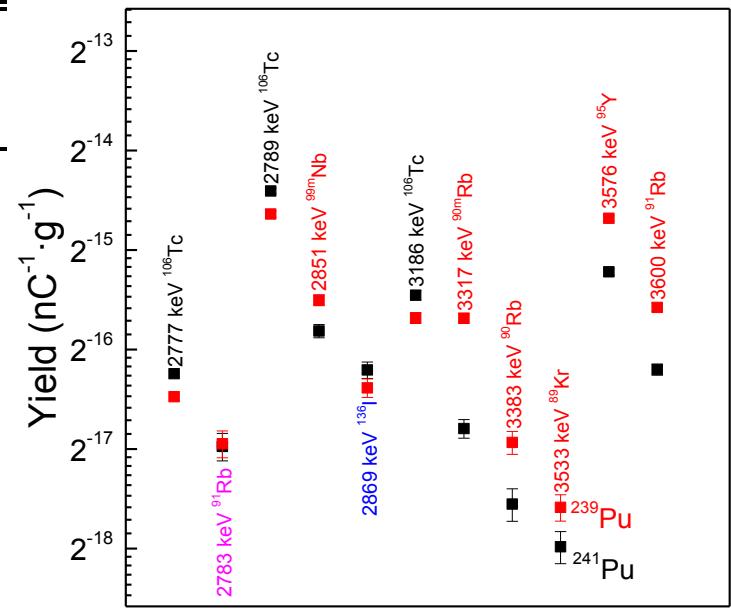
The  $^{241}\text{Pu}/^{239}\text{Pu}$  ratios of prominent peaks vary between 1.2 and 0.5.

# Relative Delayed Gamma-Ray Yields from $^{241}\text{Pu}$ , $^{239}\text{Pu}$

Integrated discrete delayed  $\gamma$ -ray yields from the  $^{241}\text{Pu}$  and  $^{239}\text{Pu}$  targets.

(Prel. Data)

Discrete $\gamma$ -ray energy (keV)	$^{241}\text{Pu}$ Discrete $\gamma$ -ray yield ( $\text{nC}^{-1} \cdot \text{g}^{-1}$ )	$^{239}\text{Pu}$ Discrete $\gamma$ -ray yield ( $\text{nC}^{-1} \cdot \text{g}^{-1}$ )	$^{241}\text{Pu}/^{239}\text{Pu}$
2783 - $^{91}\text{Rb}$	$(7.8 \pm 0.7) \times 10^{-6}$	$(7.9 \pm 0.7) \times 10^{-6}$	1.0
2789 - $^{106}\text{Tc}$	$(4.60 \pm 0.09) \times 10^{-5}$	$(3.92 \pm 0.09) \times 10^{-5}$	1.2
2851 - $^{99m}\text{Nb}$	$(1.74 \pm 0.08) \times 10^{-5}$	$(2.15 \pm 0.08) \times 10^{-5}$	0.8
2869 - $^{136}\text{I}$	$(1.32 \pm 0.07) \times 10^{-5}$	$(1.17 \pm 0.07) \times 10^{-5}$	1.1
3186 - $^{106}\text{Tc}$	$(2.23 \pm 0.07) \times 10^{-5}$	$(1.90 \pm 0.07) \times 10^{-5}$	1.2
3317 - $^{90m}\text{Rb}$	$(8.8 \pm 0.6) \times 10^{-6}$	$(1.90 \pm 0.07) \times 10^{-5}$	0.5
3383 - $^{90}\text{Rb}$	$(5.2 \pm 0.6) \times 10^{-6}$	$(8.0 \pm 0.6) \times 10^{-6}$	0.65
3533 - $^{89}\text{Kr}$	$(3.9 \pm 0.4) \times 10^{-6}$	$(5.1 \pm 0.5) \times 10^{-6}$	0.76
3576 - $^{95}\text{Y}$	$(2.62 \pm 0.06) \times 10^{-5}$	$(3.81 \pm 0.07) \times 10^{-5}$	0.69
3600 - $^{91}\text{Rb}$	$(1.33 \pm 0.05) \times 10^{-5}$	$(2.05 \pm 0.06) \times 10^{-5}$	0.65

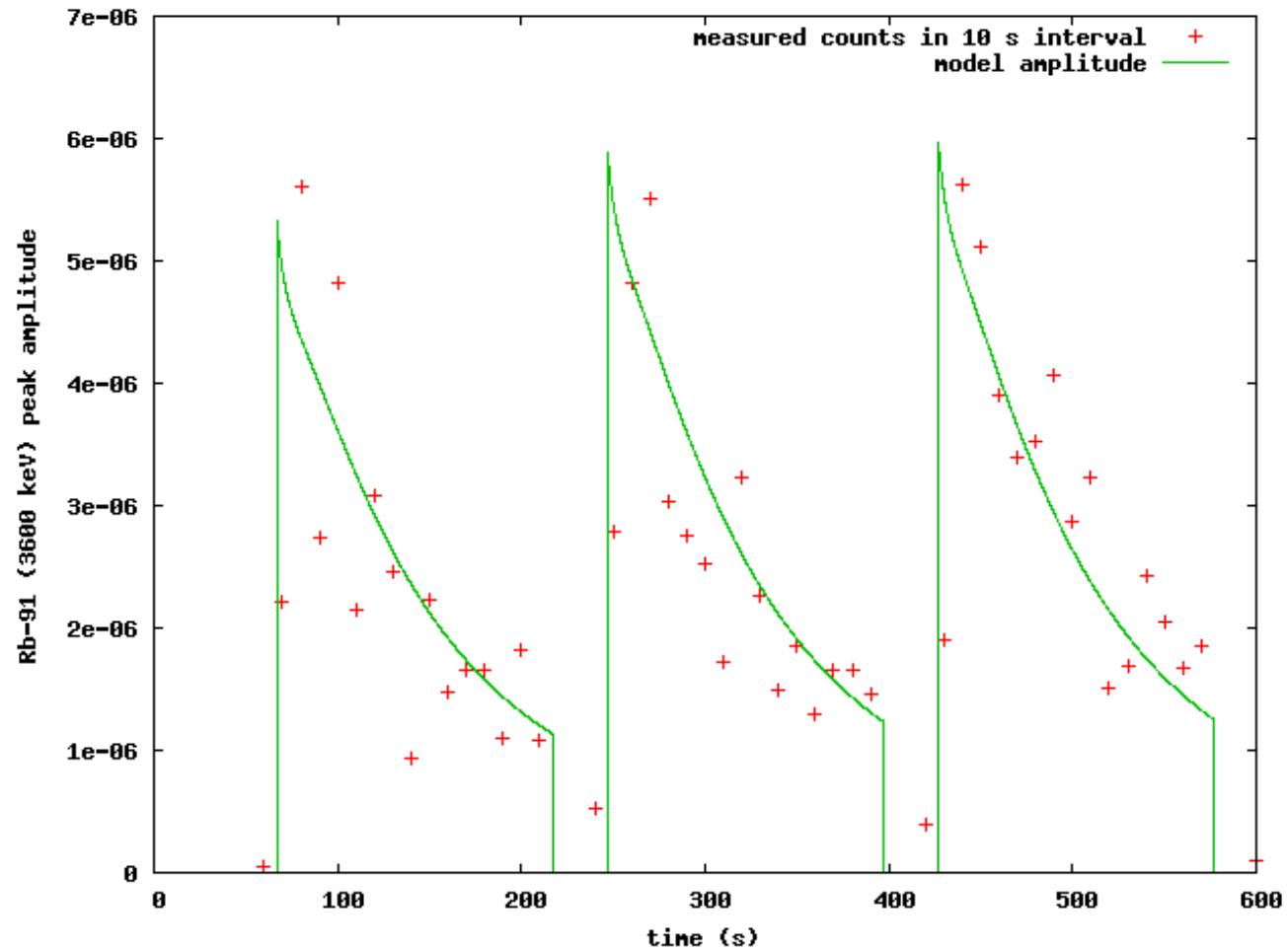


# Determination of Fission Yields from Time-dependent Delayed Gamma-ray Data

**Physical model:** Systems of decaying isotopes produce time-varying spectral peaks & background

Independent fission yields, parameters in the model, are found by fitting the experimental time-energy data using a maximum-likelihood estimation method.

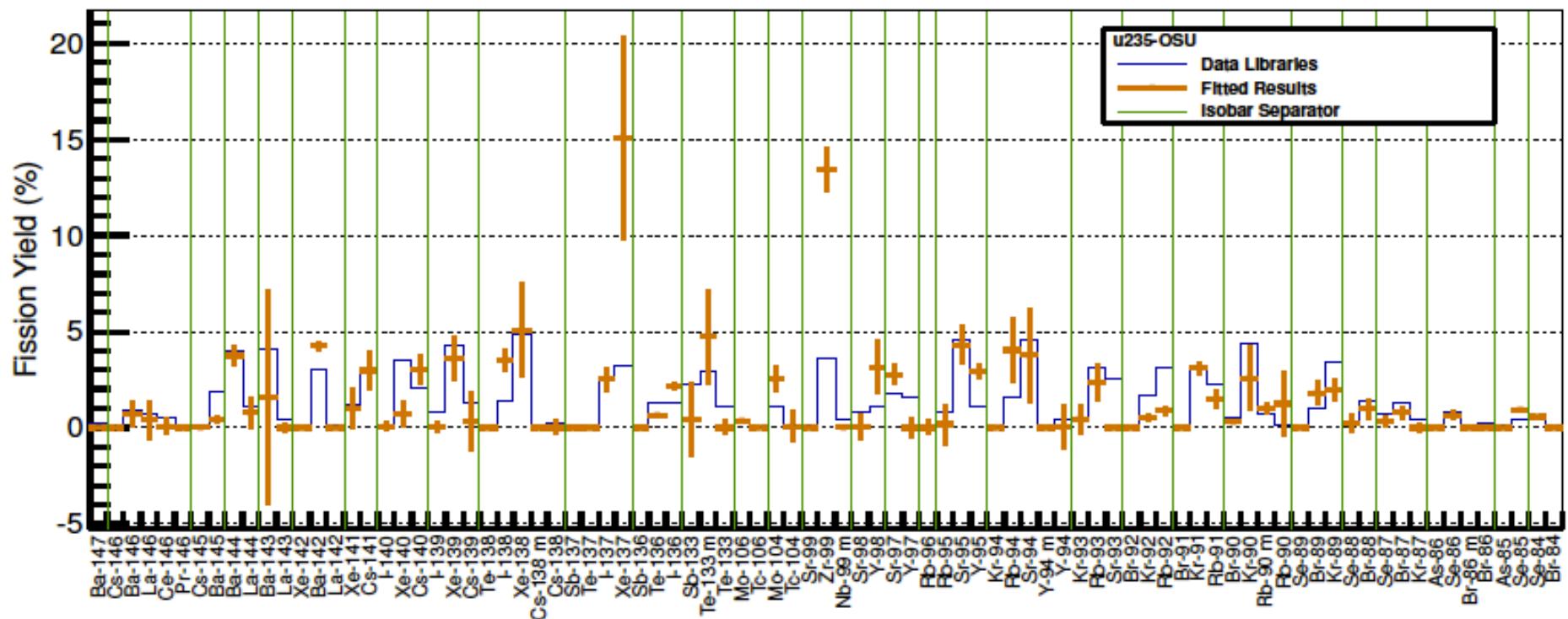
LW Campbell,  
PNNL-SA-99655



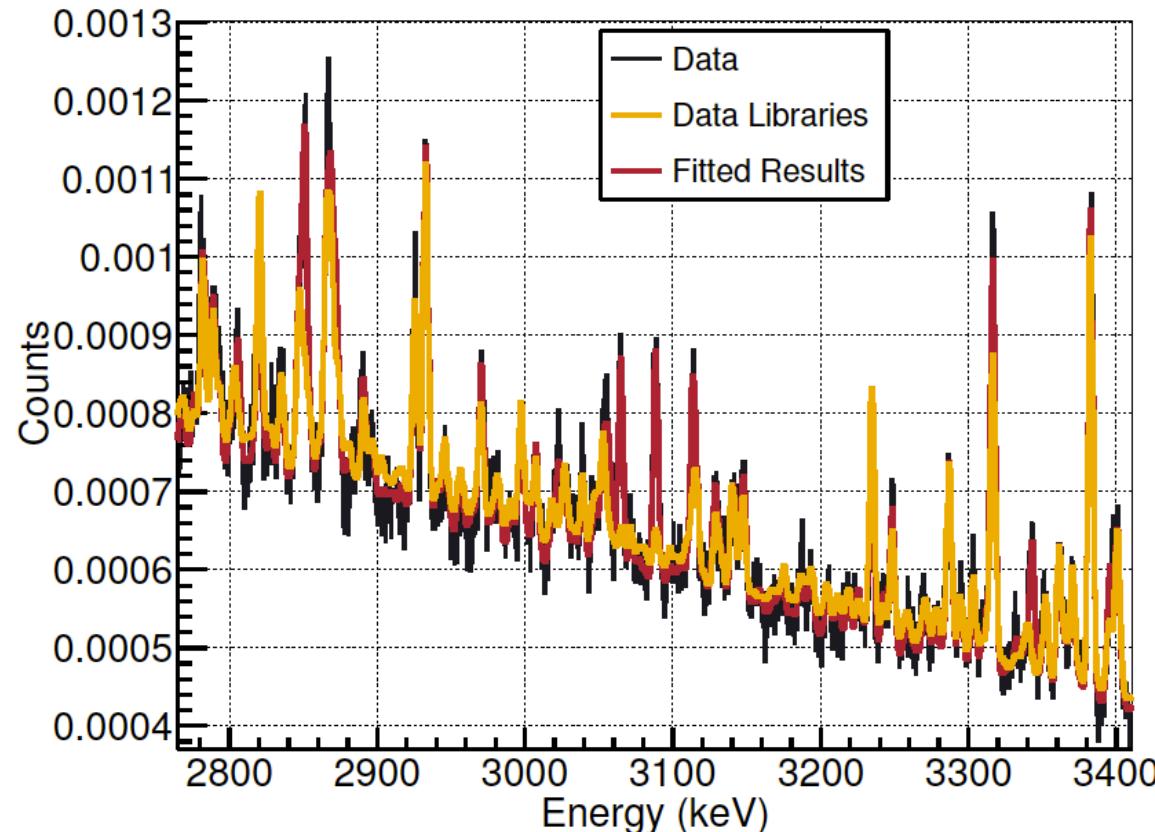
Fit of the time-dependence of the number of counts measured in 10 s intervals in the  $^{91}\text{Rb}$  peak at 3600 keV.

# Fitted $^{235}\text{U}$ Fission Yields

Fitted  $^{235}\text{U}$  fission yields (orange) extracted from delayed gamma-ray measurements between 2.8 and 3.4 MeV normalized to the cumulative  $^{95}\text{Y}$  yield and ENDF/B-VII data library values (blue line).



# $^{235}\text{U}$ Delayed Gamma-ray Spectrum Calculated with Fitted Fission Yields



- Black: measured.
- Yellow: calculated based on ENDF/B-VII data libraries.
- Red: calculated based on the fitted fission yields.

- More data sets needed to be fitted to improve the accuracy of the extracted fission yields.
- Also  $^{239}\text{Pu}$  and  $^{241}\text{Pu}$ .
- Experimental data available – Prof. Alan Hunt, ISU

## Physical model

- Systems of decaying isotopes produce time-varying spectral peaks & background

## Determine set of fission yields that reproduces observed data

- Have preliminary fits for certain spectral regions.
- Single and double escape peaks included in analysis.

Preliminary best-fit relative fission yields for U-235 thermal fission

(normalized to cumulative yield of Y-95 = 0.064)

Se 85	0.007691	+/-	0.00049
Se 87	0.008732	+/-	0.002671
Br 87	0.006921	+/-	0.003191
Br 88	0.015212	+/-	0.001169
Kr 89	0.023941	+/-	0.001884
Rb 89	0.005172	+/-	0.002859
Rb 90 M	0.012803	+/-	0.001086
Rb 90	0.036539	+/-	0.000603
Rb 91	0.045594	+/-	0.000441
Rb 92	0.005367	+/-	0.007878
Rb 93	0.026013	+/-	0.000965
Sr 95	0.040862	+/-	0.001605
Y 95	0.022825	+/-	0.001804
Y 97	0.020542	+/-	0.002252
Tc 106	0.009858	+/-	0.002155
Te 135	2.85E-08	+/-	2.19E-05
I 136	1.84E-07	+/-	5.33E-05
I 137	0.019238	+/-	0.002389
I 138	0.028651	+/-	0.005515
Xe 139	0.086401	+/-	0.051444
Cs 139	0.051121	+/-	0.054131
Cs 140	0.059973	+/-	0.003645
La 142	0.050324	+/-	0.002785
La 144	1.31E-10	+/-	7.99E-07
Ba 145	1.29E-06	+/-	0.000161