Integral Measurements of Independent and Cumulative Fission Product Yields

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Integral Measurements of Independent and Cumulative Fission Product Yields Supporting Nuclear Forensics and Other Applications

Objective: Improved measurements of integral cumulative and independent fission product yields for major and minor actinides in relevant neutron fields.

Relevance: Fission product yields (FPYs) represent an important nuclear fission observable for basic science and multiple national security applications. This and related work provides experimental data to validate and improve differential nuclear data, nuclear physics models and simulations, and operational tools. The data collected under this project will directly feed into a new FPY evaluation that will be included in the release of ENDF/B VIII.

Approach: Make use of steady-state and burst critical assemblies, and other neutron sources as appropriate, to irradiate well characterized actinide samples. Samples will be analyzed by

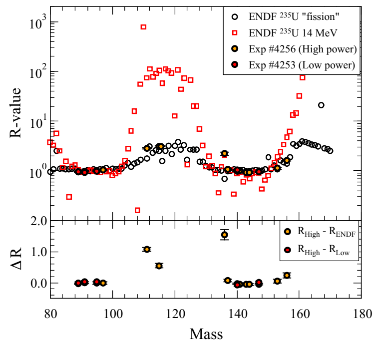
- Radiochemical analysis to determine relative ($R_i^{j,k}$) cumulative FPYs. When appropriate, fission chambers will be used to determine absolute ($Y_i^{j,k}$) fission product yields.

  and/or

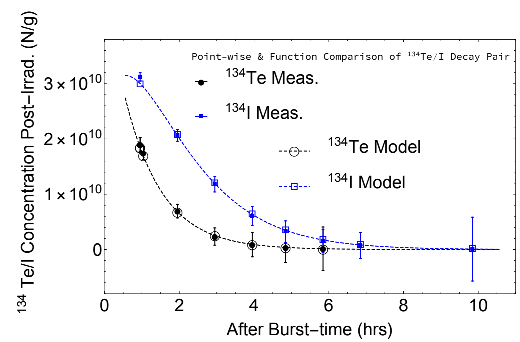
- Direct gamma-ray counting to determine independent FPYs.

Prior Work: Executed 1-2 irradiations per year since 2012. Early efforts focused on restoring operational and experimental capabilities at NCERC. Later activities resulted in new data of cumulative FPYs for ^{233}U, ^{235}U, ^{238}U and ^{237}Np using the Flattop critical assembly. New data were also collected on independent FPYs for ^{235}U and ^{238}U using burst mode with the Godiva critical assembly.

Tested and fielded prototype fission chambers with ^{235}U (FY17) and ^{237}Np (FY18) reference and macro-foils on the Flattop assembly to extract absolute fission product yields ($Y_i^{j,k}$).



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.



A plot of the measured and model estimated atom-densities of ^{134}Te and ^{134}I as a function of time following a burst irradiation of a ^{235}U sample on the Godiva IV critical assembly.

Cumulative Fission Product Yields

Research/Project Description

Objective: Measure integral cumulative fission product yields, $R_i^{j,k}$ and $Y_i^{j,k}$, in relevant neutron fields for important major and minor actinides.

Relevance: Fission product yields (FPY) represent important nuclear fission observables for basic science as well as numerous nuclear applications. This and related work provides experimental data to validate and improve differential nuclear data, nuclear physics modeling and operational tools.

Current plans in the community are to produce a new fission product yield evaluation within the next five years, to include isotopes of uranium, plutonium, americium, neptunium and others that may be deemed important by the user community.

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

Current State of Research: Executed 1-2 irradiations per year since 2012 on actinide samples including ^{233}U, ^{235}U, ^{238}U and ^{237}Np. For each actinide sample we extracted relative fission product yields, $R_i^{j,k}$, by radiochemical analysis. This exercised the Radiochemistry teams at LANL and PNNL and provided valuable data for modeling and evaluation.

Over the last two years we tested and fielded prototype fission chambers with ^{235}U and ^{237}Np reference and macro-foils on the Flattop assembly to extract absolute fission product yields, $Y_i^{j,k}$.

Issues: Tests of the new fission chamber show degradation of the fission product energies that was not observed the previous year. We believe this is an issue with fabrication of the reference foils, and we have developed a plan to determine the cause and correct it.

Availability of suitable materials continues to be a challenge.

Background

Fission Product Yields (FPYs) were historically analyzed by radiochemistry, and reported as ratios of activities known as R-values

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

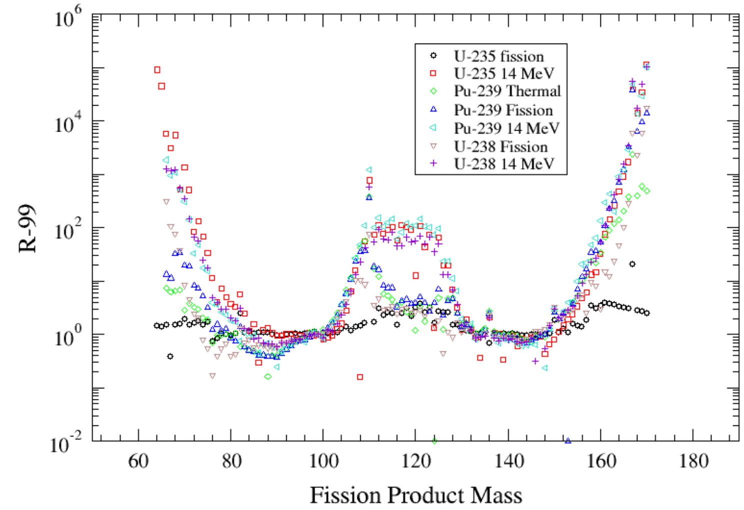
Fission chamber experiments were used to calibrate radiation detectors for a small set of FPs, e.g. ^{99}Mo, to determine # of fissions.

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

Once measured experimentally, the K-factor could be used to determine the number of fissions in a sample based on activity.

NOTE: THIS IS DETECTOR SPECIFIC!

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

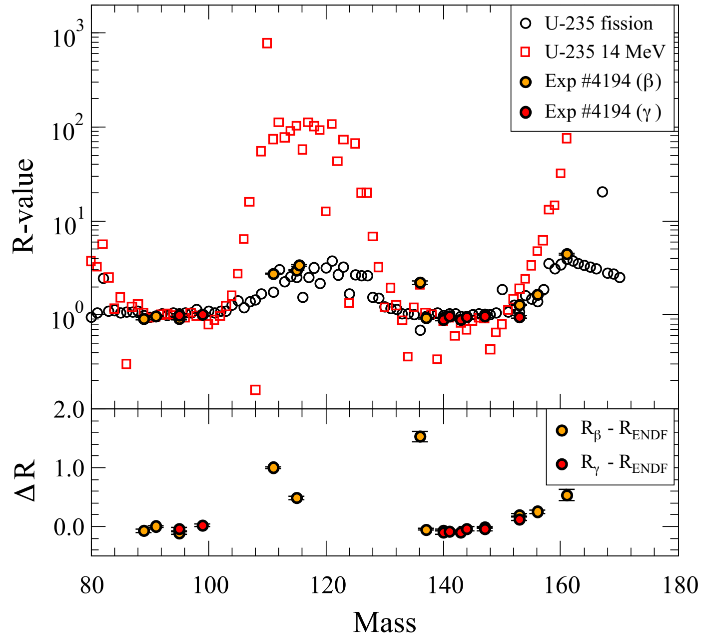


Modern tools are becoming more dependent on evaluated (usually energy-differential) nuclear data, including FPYs. These nuclear data, and models that use them, need to be validated against integral measurements such as those that have been conducted at NCERC.

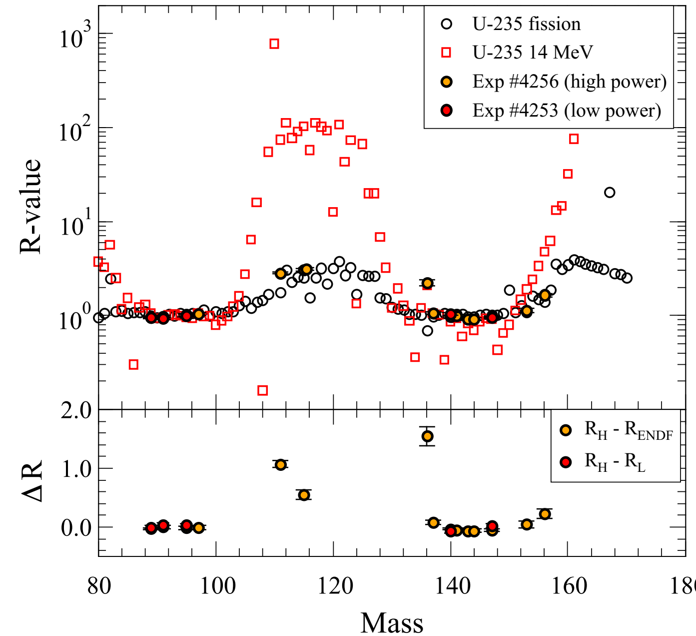
For FPYs this is best accomplished using fission chambers in conjunction with radiochemical analyses.

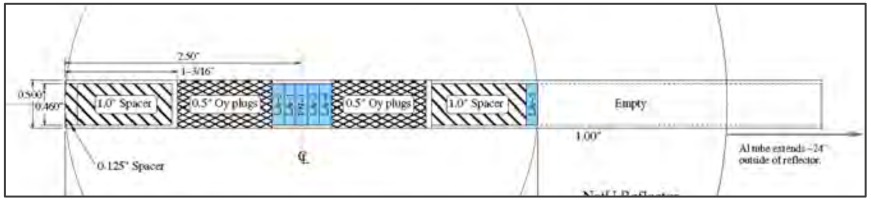
Example Results

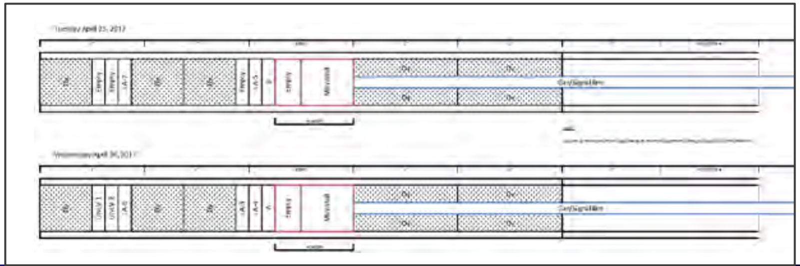
2015: No fission chamber



2017: ^{235}U fission chamber







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Independent Fission Product Yields

Overview

There has been no serious scrutiny of the short lived fission product yields since the publication of the England and Rider report in 1993 (24 years). The results in the England and Rider report are a collection of results from experiments occurring from the 1950s through to the early 1990s.

We are conducting the first set of self consistent irradiations of major actinides in nearly 30 years with a prompt (< 100 μs) fission neutron spectrum.

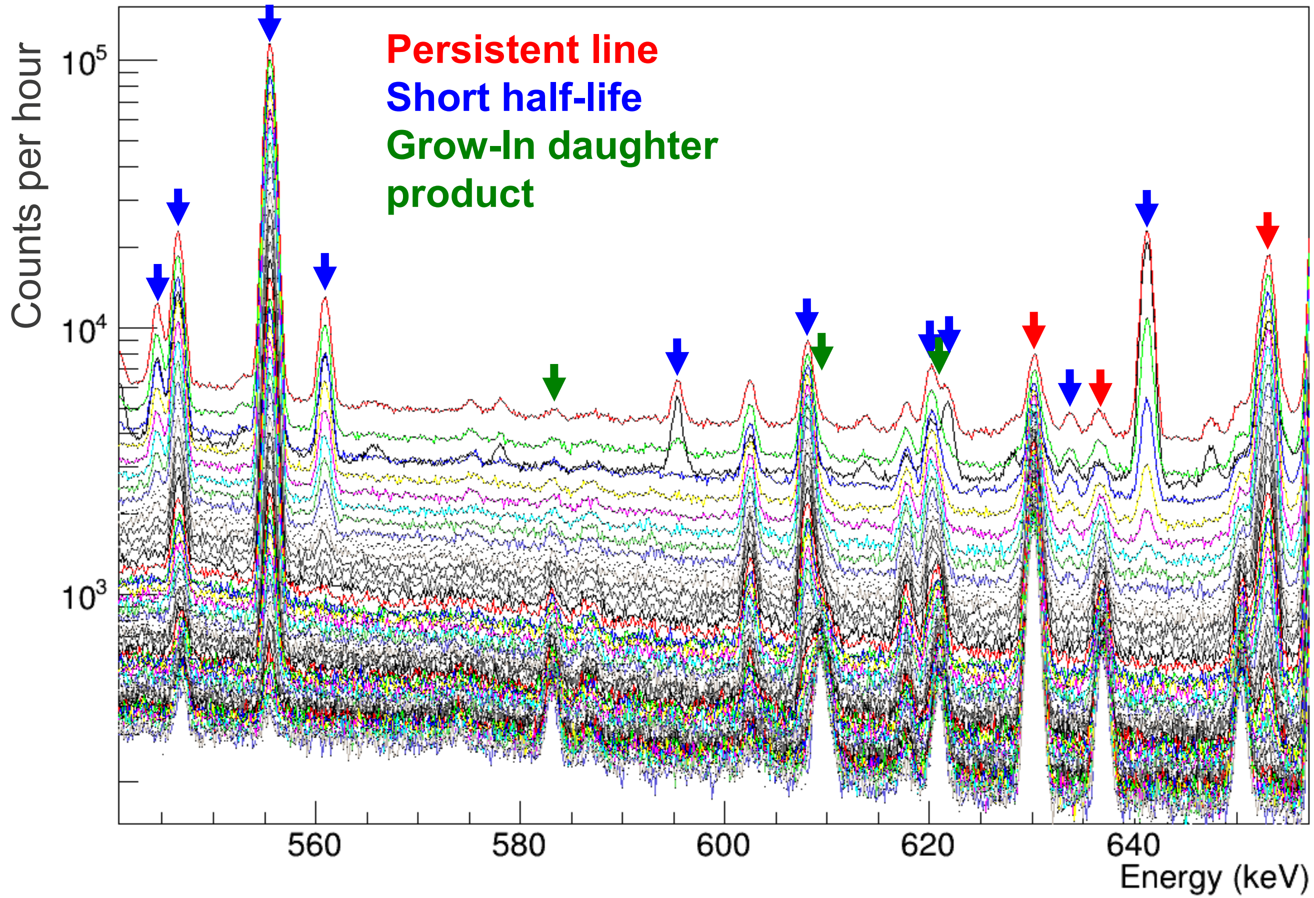
- These measurements make use of the unique capability of pulsed critical assemblies which are not available elsewhere. (assemblies at Valduc have been decommissioned)
- Counting whole samples allows us to identify clear decay signals and interferences
- Rigorous treatment and documentation of all sources of uncertainty performed to provide as accurate as possible results

High quality nuclear data are essential to data evaluation and modeling;

- Improve nuclear data libraries
- Generate time dependent gamma ray libraries from actual measurements and provide it to the user community

Technical Approach: identifying gamma rays

Time dependent gamma ray spectra from 45 minutes to 7 days post-irradiation



Results to date

- Established a reliable and repeatable gamma ray measurement system near the critical assembly area.
  - can retrieve and start counting samples in 60 minutes or less post irradiation
- have reported values on **20 unique isotopes** and **47 unique gamma rays** for ^{235}U fission product yields.
- have reported values on **23 unique isotopes** and **74 unique gamma rays** for ^{238}U fission product yields.
- There are **~230 gamma ray lines** that have been identified. More sophisticated gamma ray fitting routines are being developed to extract as much information as possible from the data sets.

We have performed 4 successful irradiations.

- ^{235}U irradiation and count June 2014 on Godiva – burst mode
- ^{235}U irradiation and count July 2015 on Flattop – short high flux
- ^{238}U irradiation and count January 2016 on Godiva – burst mode
- ^{239}Pu irradiation and count November 2018 on Godiva – burst mode

Reporting:

- Created a ^{235}U gamma ray library and interference reference - tech report
- Reported on the ^{235}U fission product yields - tech report
- Preparing a report on the ^{238}U fission product yields (Sept 2017) – tech report to be submitted
- Multiple presentations at Venture Quarterly Reviews to update on progress

Future Plans

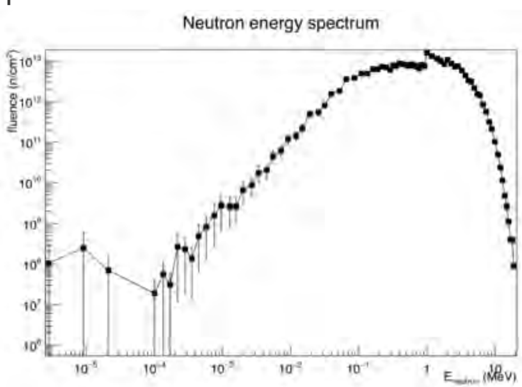
1) Complete analysis of the ^{239}Pu data collected in November and prepare a report.
2) Perform inter-comparisons between ^{235}U, ^{238}U and ^{239}Pu FPYs to ensure consistency of results and fully understand discrepancies with current database
3) Publish the results in a journal so that it is available to the nuclear physics community and the data libraries at the National Nuclear Data Center at Brookhaven National Laboratory which is funded by the US Nuclear Data Program
4) Continue program investigating minor actinides and FPY time dependent behavior exposed to a prompt fission neutron spectrum. We know there are issues here as well.
5) Release gamma ray libraries to the user community
6) Perform measurements at 14 MeV to validate that energy region and ensure we understand potential interferences.

NCERC Team essential: nuclear engineers, operations and support staff
LLNL Team: Jason Burke, Stephen Padgett, Gary Slavik, Doug McAvoy, Becca Hudson, Kevin Roberts, Sherry Faye, Jennifer Shusterman, Kip Harward, Bob Haslett, Todd Woody, Roger Henderson and Phil Toretto
PNNL Team: Bruce Pierson, Larry Greenwood, Leah Arrigo, Sean Stave, Amanda Prinke, Lori Metz and Judah Friese

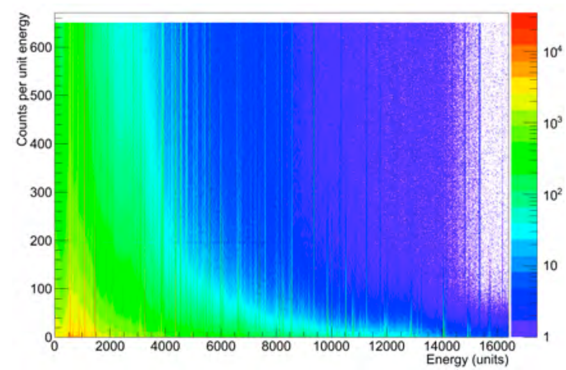
Technical Approach: the experiment

Actinide samples are prepared at LLNL and shipped to NCERC along with a witness foil pack from PNNL.

Prior to the irradiation, two BeGe gamma ray detectors are cooled, tested and calibrated in preparation for the irradiated samples.



Irradiate with prompt fission neutron spectrum



Collect list mode data of gamma rays emitted over a 7 day period