Integral Fission Product Yields Multi-lab

Los Alamos NATIONAL LABORATORY
Lawrence Livermore National Laboratory
Pacific Northwest NATIONAL LABORATORY
NEVADA NATIONAL NNSS SECURITY SITE

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LANL / LLNL / PNNL

NCERC NATIONAL CRITICALITY EXPERIMENTS RESEARCH CENTER

WANDA 2023
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DEPARTMENT OF ENERGY · UNITED STATES OF AMERICA ·
NNSA National Nuclear Security Administration
Defense Nuclear Nonproliferation Research & Development Program

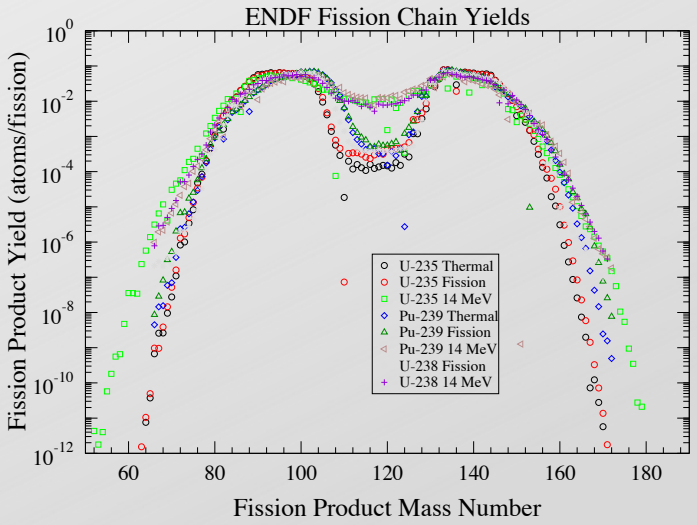
Integral Fission Product Yields

Objective: Make improved measurements of integral cumulative and short-lived fission product yields, and related cross sections, for major and minor actinides in relevant neutron fields.

Approach: Make use of burst and steady-state critical assemblies, and other neutron sources, to irradiate well characterized actinide and non-actinide samples, and use multiple techniques to extract fission product yields.

CFPY Task:
Cumulative fission product yields

SLFPY Task:
Short-lived fission product yields

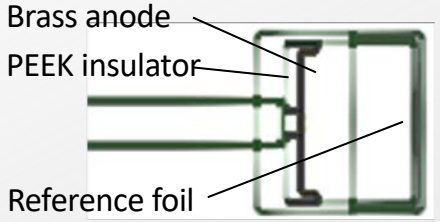


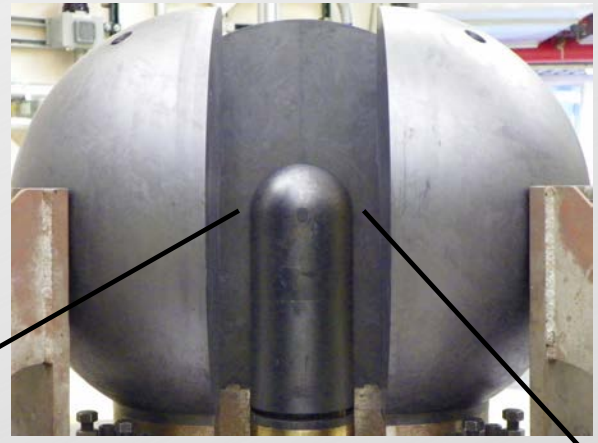
Measurements Made/Scheduled since 2012			
Task	Year(s)	Neutron Source(s)	Material(s) of Interest
CFPY*	**2012-2018**	NCERC – Planet, Comet & Flattop D-T Generator	^{233}U, ^{235}U, ^{238}U, ^{237}Np
SLFPY	**2015-2018**	NCERC – Godiva	^{238}U, ^{235}U, ^{239}Pu
CFPY	**2020**	D-T Generator	^{235}U
SLFPY	**2020**	NCERC – Godiva	^{237}Np
CFPY	**2021**	NCERC – Flattop Fission Chamber	^{235}U
SLFPY	**March 2022**	NCERC – Godiva	^{233}U
CFPY	**April 2022**	NCERC – Godiva Fission Chamber	^{239}Pu
SLFPY	**May 2022**	OSU TRIGA	^{238}U

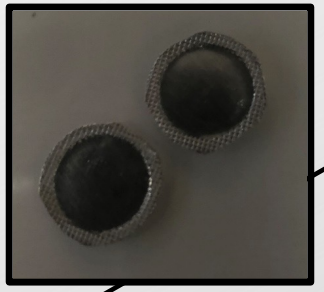
Cumulative Fission Product Yields (CFPY)

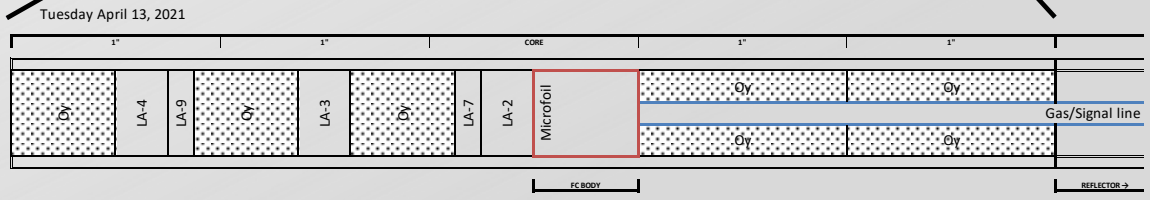
INNOVATE. COLLABORATE. DELIVER.

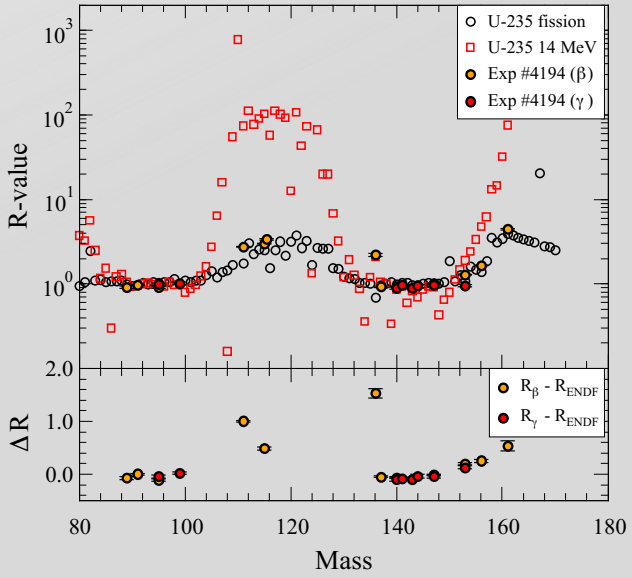
Fabricate Fission Chambers to Determine Absolute FPYs → Prepare High Quality Reference and Macro-foils → Irradiate Samples at NCERC or PNNL → Post-Irradiation Whole Foil γ–Counting → Radiochemical Dissolution and Analysis → R-values ($R_i^{j,k}$) and Absolute FP Yields ($Y_i^{j,k}$) → Publish & improve international libraries







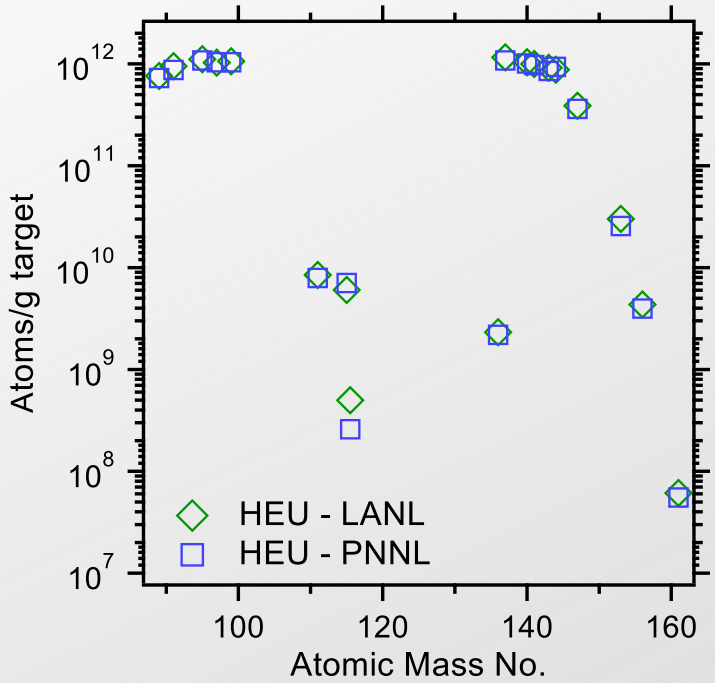


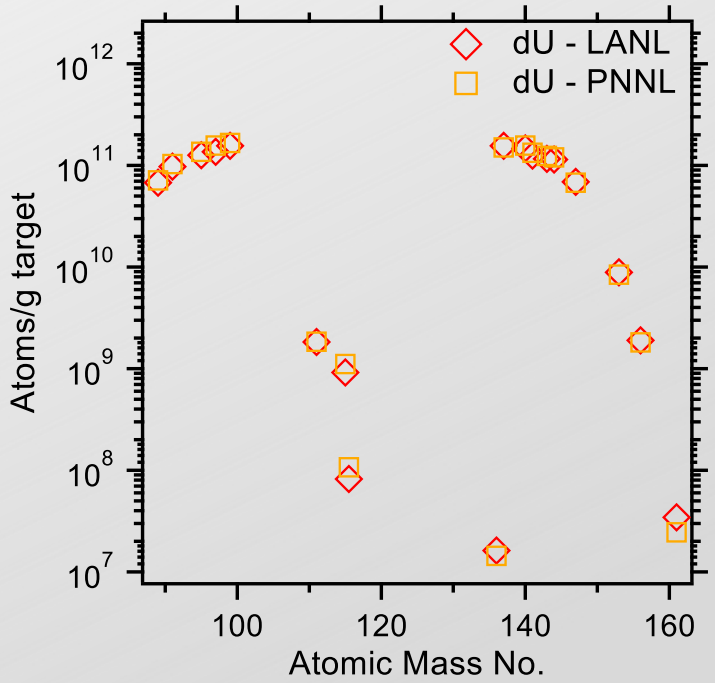


Fission Chambers | Actinide Samples | Neutrons | Radiochemistry | Relative (to Absolute) FP Yields

Defense Nuclear Nonproliferation R&D

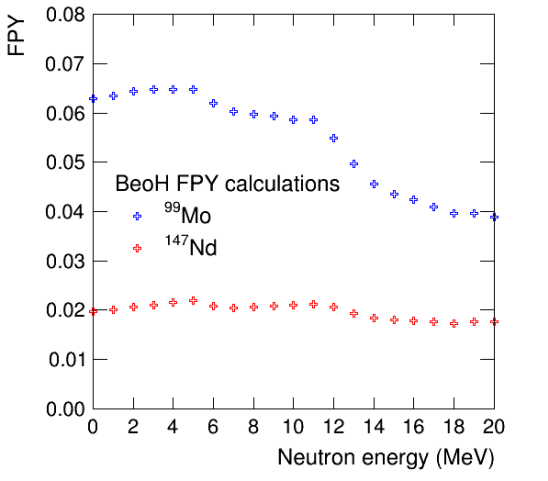
FY21 CFPY runs on Flattop – Uranium Results

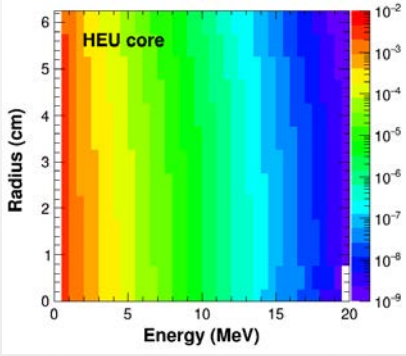


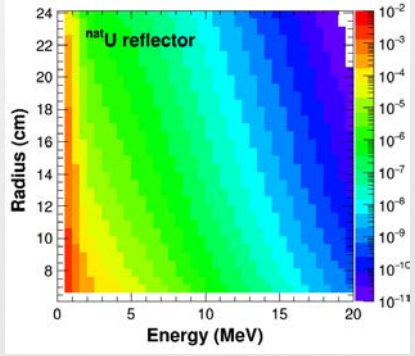


- Excellent agreement between PNNL & LANL – total fissions, actinide analysis, and across almost all fission products.
- Ultrasonic weld containment tested with irradiated depleted U targets - planned deployment April ‘22 with Pu targets.

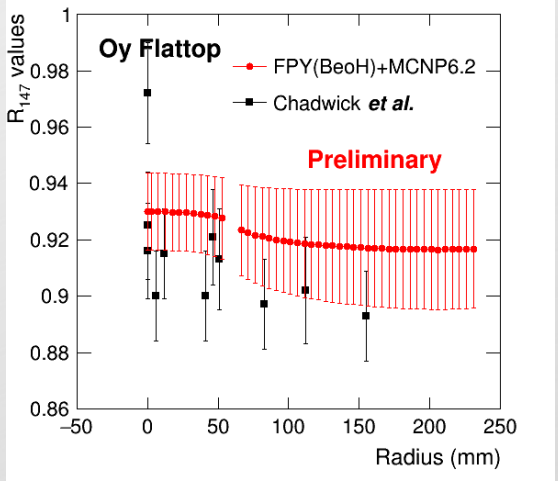
R-value calculations for NCERC assemblies

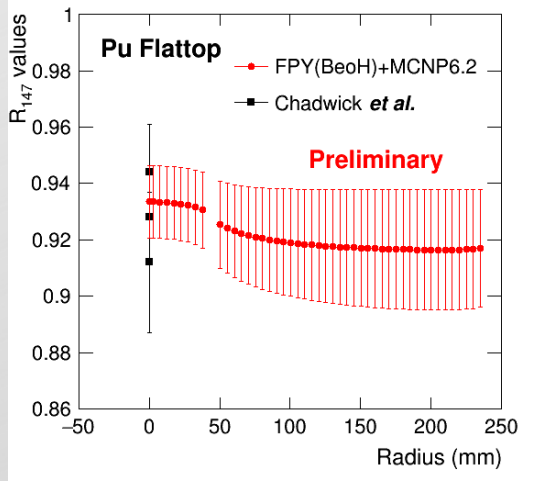


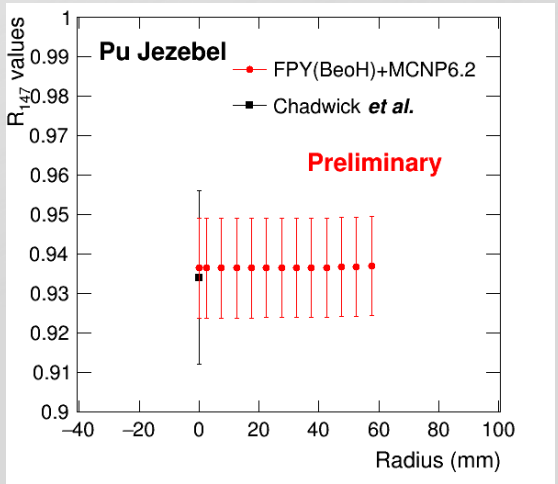


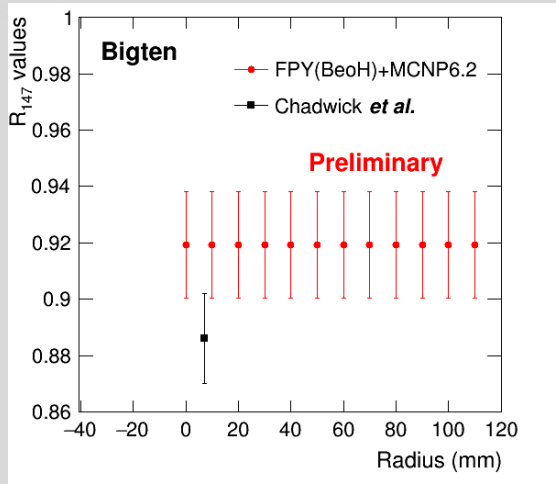


MCNP 6.2 simulations for the neutron flux of Oy Flattop.







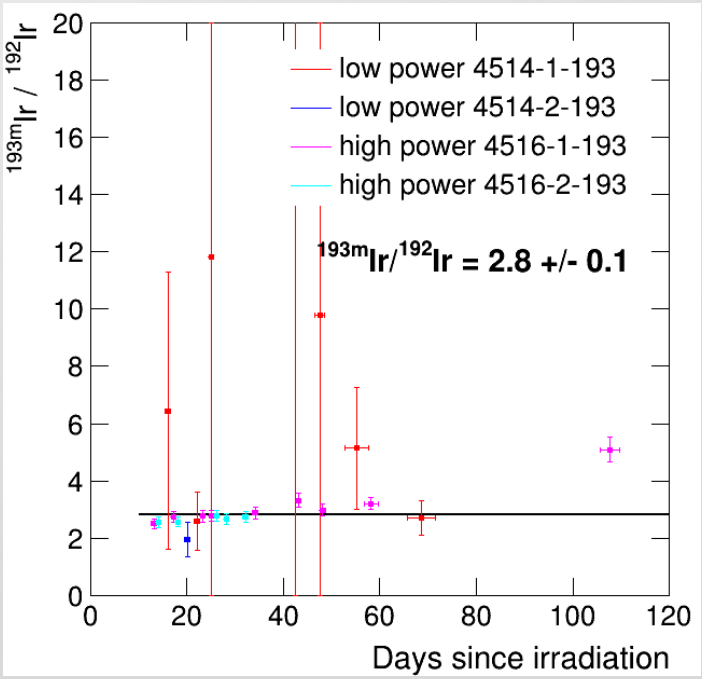


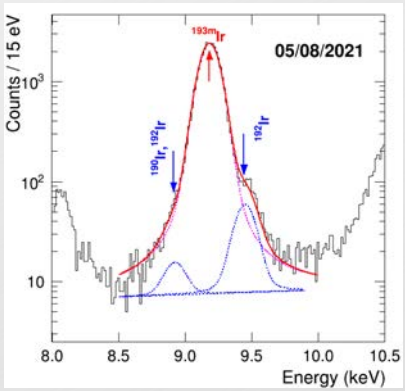
We use recent BeoH calculations for the energy dependent FPY from ^{239}Pu(n,f) and for ^{235}U(n_{th},f) to calculate the R-values for ^{147}Nd for various critical assemblies. The theory values were averaged with the neutron flux simulated with MCNP 6.2 for a given radius from the center of the critical assembly.

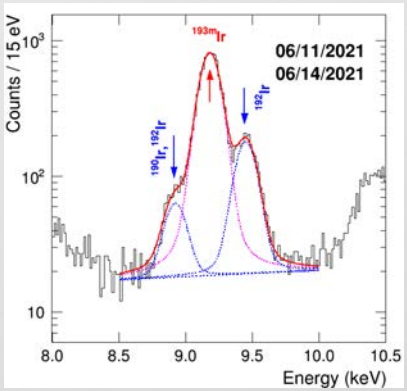
Iridium activation measurements at NCERC

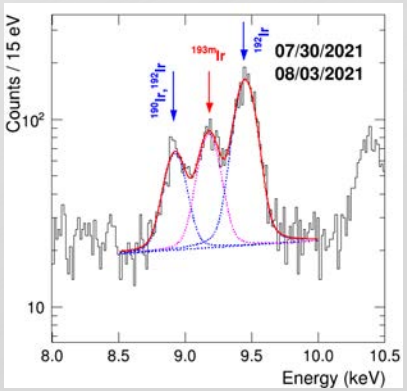
We use high-resolution silicon drift detectors to measure the X-rays from the iridium samples. We determine ^{193m}Ir/^{192}Ir ratio by fitting the three peaks with Voigt functions and correcting the peak areas for the ^{192}Ir and ^{193m}Ir decay.

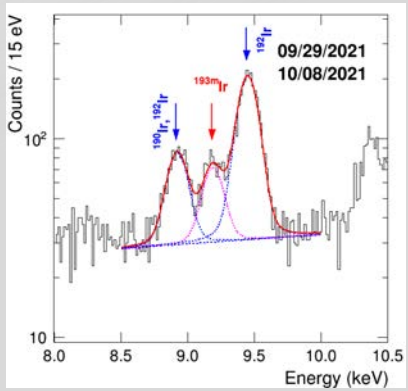
We measured the spectra at different times since the irradiation and calculated the weighted average ^{193m}Ir/^{192}Ir ratio.

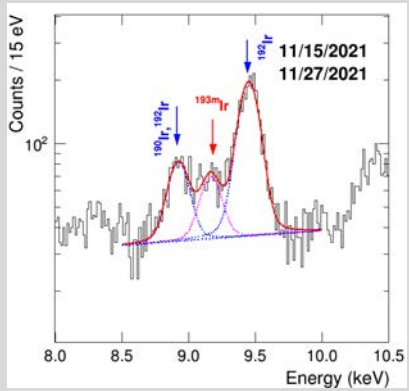












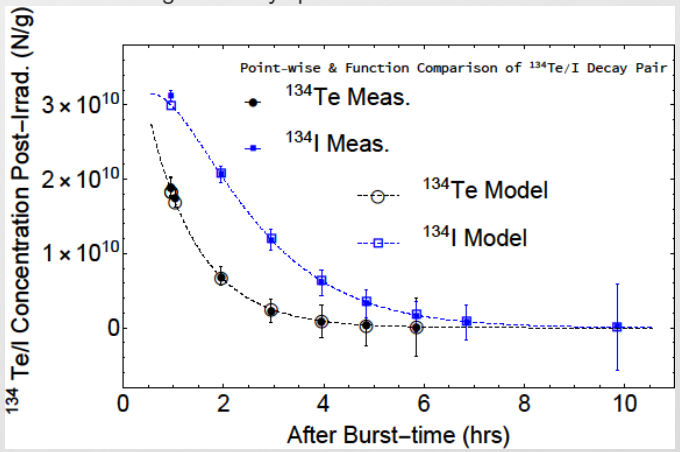
Short-Lived Fission Product Yields (SLFPY)

INNOVATE. COLLABORATE. DELIVER.

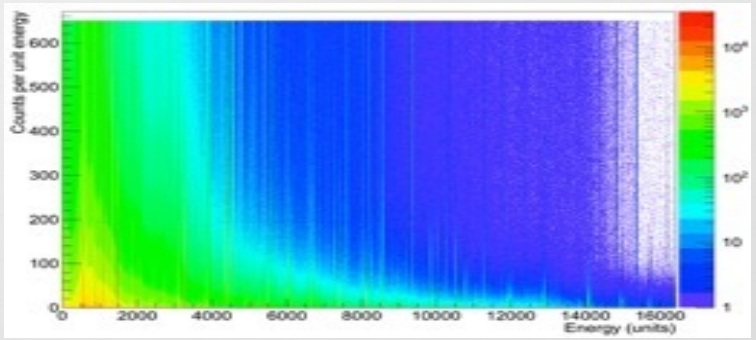
Fabricate High Quality Targets → Pulse Irradiation on the Godiva → Acquire γ spectral data → Symmetric Temporal Spectral Analysis (LLNL) → Asymmetric Temporal Spectral Analysis (PNNL) → Evaluate Neutron Fluence & Fissions → Publish Fission Yields in Nuclear Data Sheets for NNDC

Isotopically enriched actinide targets – produced at LLNL by RadChem team.

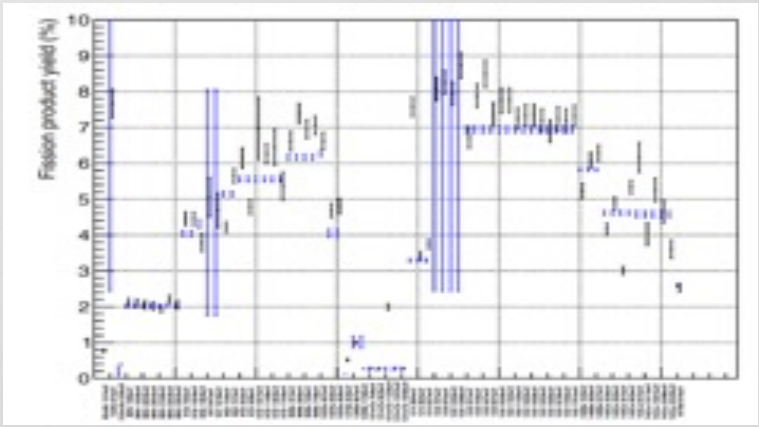
Fit gamma-ray spectra and obtain FPY



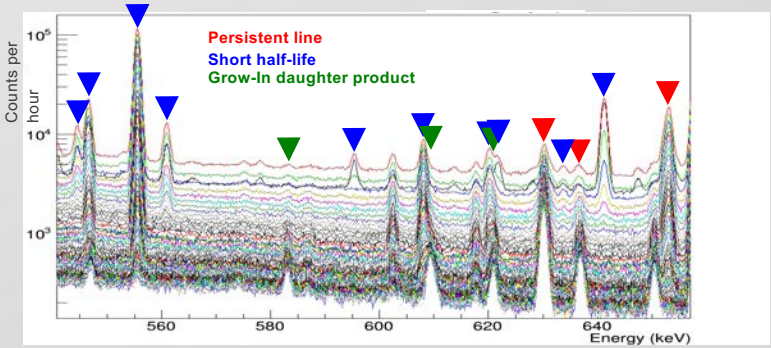
Time dependent gamma-ray spectra from seconds-days time scale



Compare multiple FPYs to old database



Very rich high-resolution spectroscopy allows us to unfold the time dependent behavior of FPs



We are seeing good improvement in FPYs and issues with branching ratios effecting FPY results. This has implications for user community. Results are published in NDS.

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

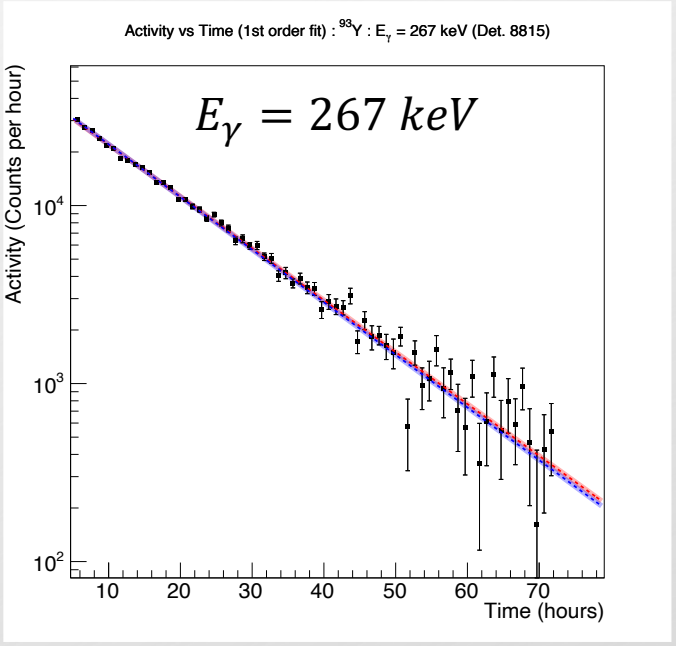
^{237}Np Results : Example ^{93}Y

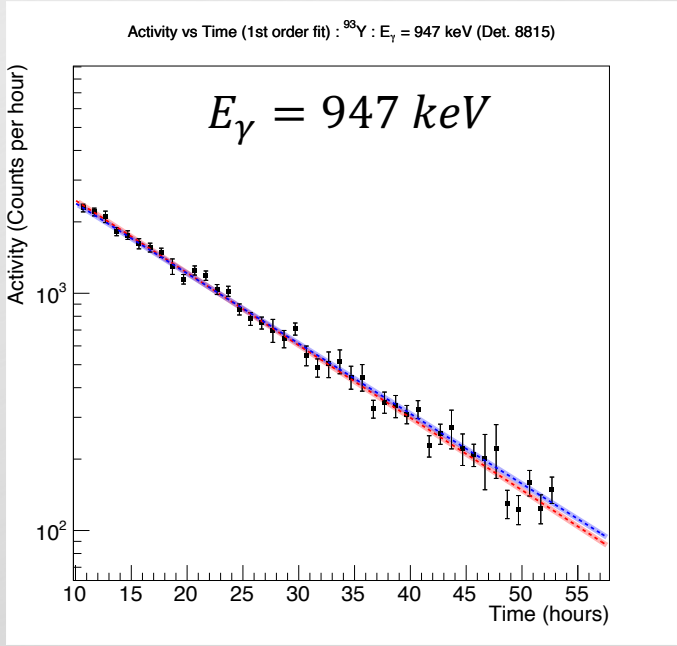
- Observed 3 'clean' γ-rays from the decay of ^{93}Y
- Extrapolate Decay Curve fits back to irradiation time : A_0
  - Correct for DAQ live-time, detection efficiency, and self-attenuation

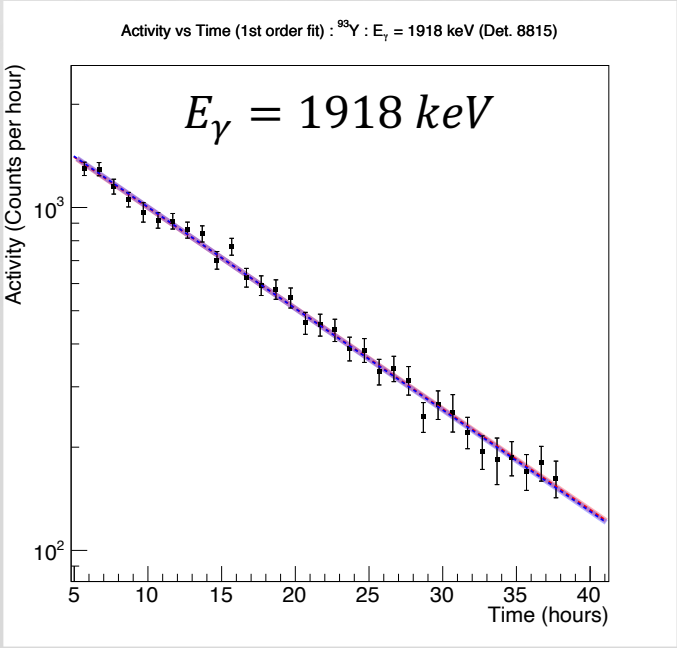
$$Y = \frac{A_0\, t_{1/2}}{\ln(2)\, \Gamma\, N_f}$$

Γ : *Branching Ratio*
N_f : *Number of Fissions*

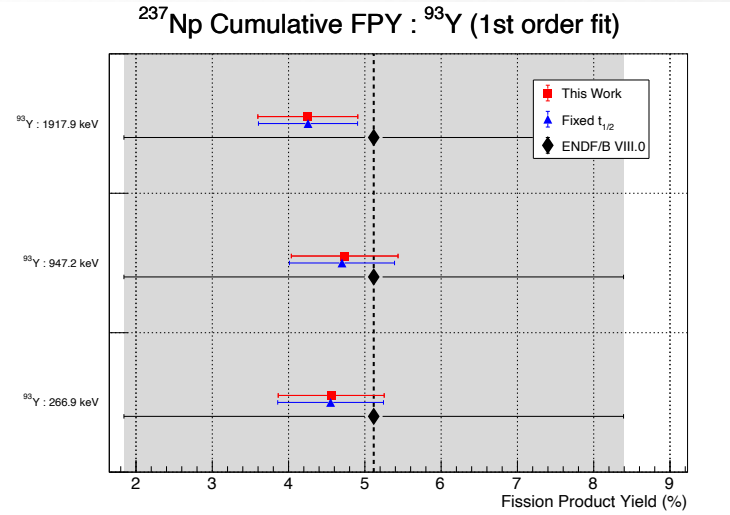
$$C(t_1, t_2) = \int_{t_1}^{t_2} A(t)dt = \frac{A_0}{\lambda} e^{-\lambda t_2}(e^{\lambda \Delta t} - 1)$$

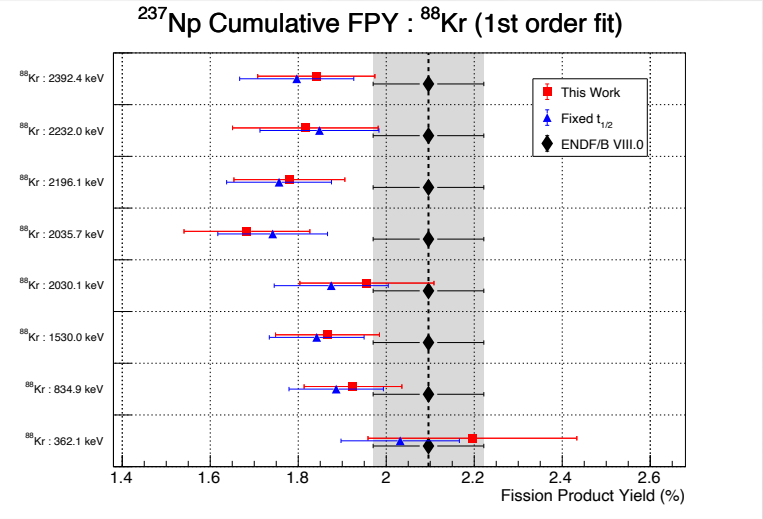


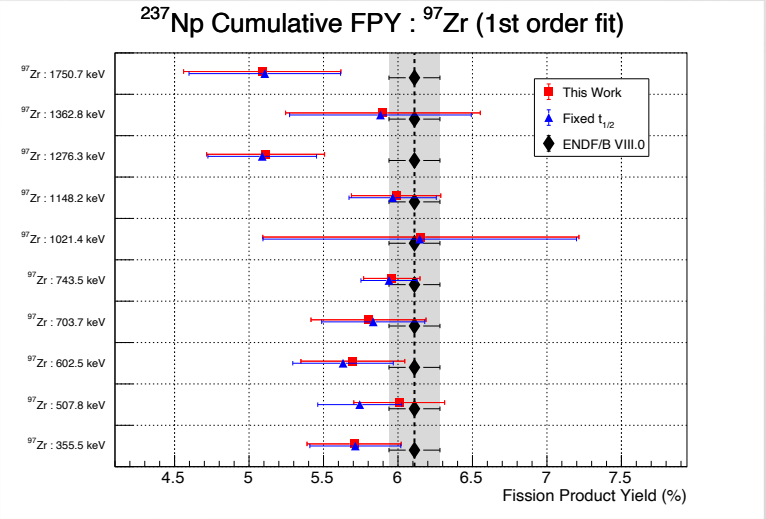


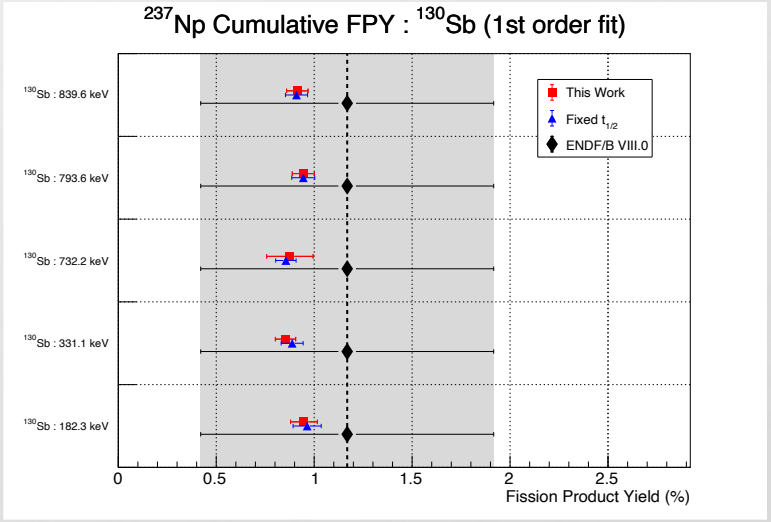


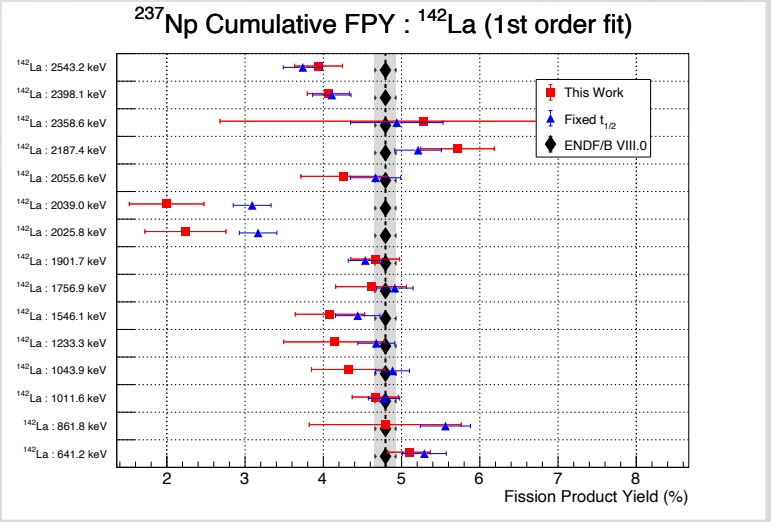
^{237}Np Results : 45 Isotopes/Isomers : 191 γ-rays











Summary: FY21 Experimental Activities

Dates	Task	Activity
Oct 27-29, 2020	CFPY	^{235}U fission chamber testing at the MIT NRL
Apr 7-9, 2021	CFPY	^{235}U fission chamber testing on Flattop
Apr 12-14, 2021	**CFPY**	**^{235}U production irradiation on Flattop**
Apr 26-29, 2021	API	Flattop core swap (Oy → Pu)
May 3-5, 2021	**API**	**Production irradiation on Flattop-Pu**
Jun 14-15, 2021	**SLFPY**	**^{233}U production irradiation on Godiva – POSTPONED (detector failure)**
Jul 13-15, 2021	CFPY	^{235}U fission chamber testing on Godiva
Jul 19-21, 2021	**CFPY**	**Supported CSoM on ^{238}U production irradiation on Flattop**
Sep 15-16, 2021	CFPY	^{235}U fission chamber testing at the MIT NRL
Sep 2021	**CFPY**	**^{239}Pu test irradiation on 14 MeV D-T source. Analysis in FY22.**

We completed 3 experimental campaigns at NCERC and 1 at PNNL in FY21.

Summary: FY22 Experimental Activities

Dates	Task	Activity
Feb 1-3, 2022	CFPY	^{235}U fission chamber testing at the MIT NRL
Mar 7-10, 2022	CFPY	^{235}U fission chamber testing on Godiva
Mar 22, 2022	**SLFPY**	**^{233}U production irradiation on Godiva**
Apr 18-21, 2022	CFPY	Final setup and testing for the ^{239}Pu irradiation on Godiva
Apr 25-28, 2022	**CFPY**	**^{239}Pu production irradiation on Godiva**
Apr 2022	**SLFPY**	**^{238}U 14 MeV D-T generator – proof in principle measurement**
Apr 2022	**SLFPY**	**^{238}U Oregon State University TRIGA Nuclear Reactor**
May 2-5, 2022	API	Flattop core swap (Oy → Pu)
May 9-12, 2022	**API**	**API production irradiation on Flattop-Pu**
March 2023	**CFPY**	**^{239}Pu production irradiation on 14 MeV D-T source**

We completed 3 experimental campaigns at NCERC, 1 at LLNL, and 1 at OSU in FY22.

We have 1 experimental campaign planned at PNNL in FY23.

Outlook: FY23 Experimental Activities

Dates	Task	Activity
Mar 20-23, 2023	CFPY	^{252}Cf fission chamber testing at LANL with CEA collaborators
Apr 10-12, 2023	**SLFPY**	**^{235}U production irradiation on Godiva w/ comparative radiochemistry**
May 1, 2023	**SLFPY**	**^{239}Pu production irradiation on 14 MeV D-T source at PNNL (w/ a fission chamber?)**
May 15-18, 2023	**CFPY**	**^{235}U fission chamber testing on Flattop with CEA collaborators**

The Full Team

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Collaborations with Colorado School of Mines and Oregon State University

Collaborative project with CEA-DAM

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Extras

Neutron Sources

Flattop (NCERC)

- Fast/fission Spectrum
- U(93) (17.7 Kg) & WG Pu (6 kg) cores / ^{Nat}U Reflector (~1000 kg)
- Horizontal (“traverse”) glory hole
- 10^{13} fissions/g on samples

Godiva IV (NCERC)

- Fast/fission neutron spectrum
- U(93) (65.5 kg, 1.5% Mo by wt)
- Super-Prompt Critical Operations
- Vertical glory hole for samples
- 1-4 × 10^{16} Total Fissions / burst

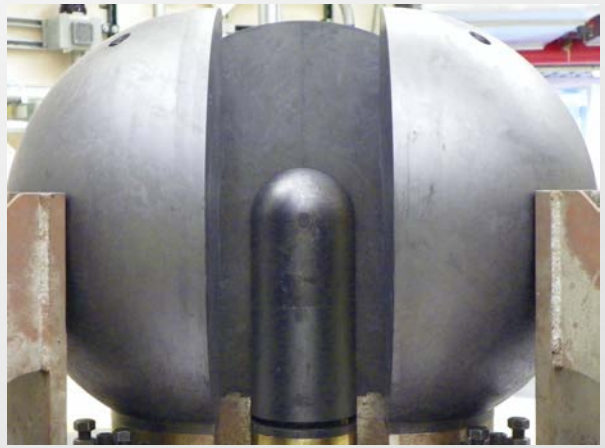
D-T Generator (PNNL)

- Thermo D711 neutron generator
- Low scatter facility at PNNL
- Max neutron flux of 1×10^{9} n/cm^2/s

Oregon State TRIGA Reactor

1.1 MW Mark II Pulsing Research Reactor

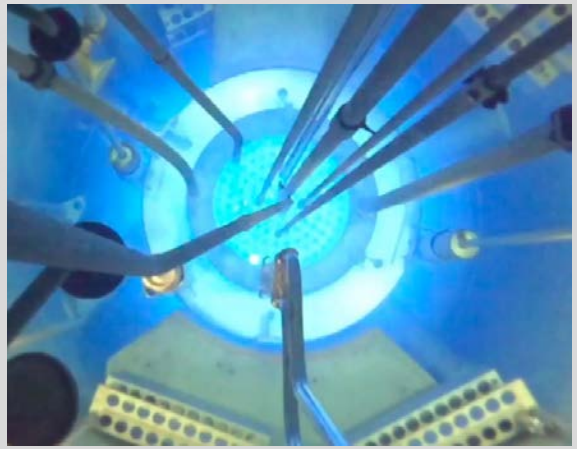
- Neutron flux of the Rabbit irradiation port
- 1.73×10^{13} n/(cm^2 s) (Thermal)
- 5.91×10^{12} n/(cm^2 s) (Epithermal)
- 5.37×10^{12} n/(cm^2 s) (Fast)



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



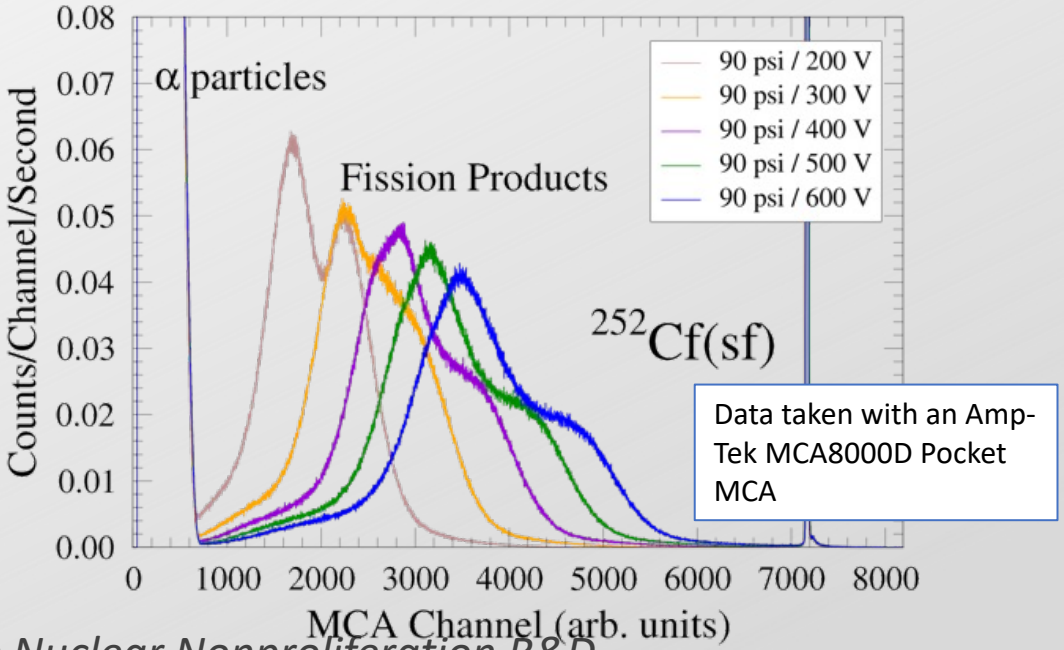
Short-Lived FPY Task
Seconds to days post irradiation

NCERC = National Criticality Experiments Research Center at Nevada National Security Site (NNSS)

Fission Chamber Performance

Pulse height spectra from the Mark II fission chamber

- Testing 0.17 cm gap with ^{252}Cf
- Testing 0.34 cm gap with ^{235}U
- Both using P-10 fill gas



MIT Nuclear Reactor Laboratory