INNOVATE. COLLABORATE. DELIVER.

Integral Fission Product Yield Multi-lab

T.A. Bredeweg / J.T. Harke / J. Friese

LANL / LLNL / PNNL

WANDA 2022
February 28 - March 4, 2022

Defense Nuclear Nonproliferation Research & Development Program

Integral Fission Product Yields

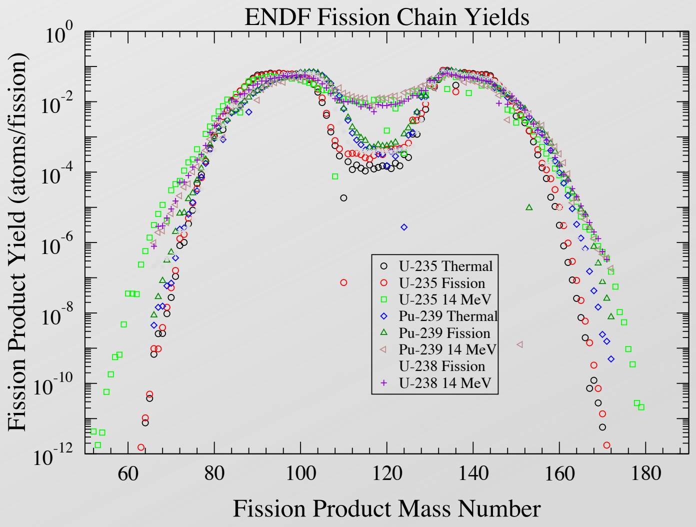
INNOVATE. COLLABORATE. DELIVER.

Objective: 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.

CFPY Task:
Cumulative fission product yields

SLFPY Task:
Short-lived fission product yields



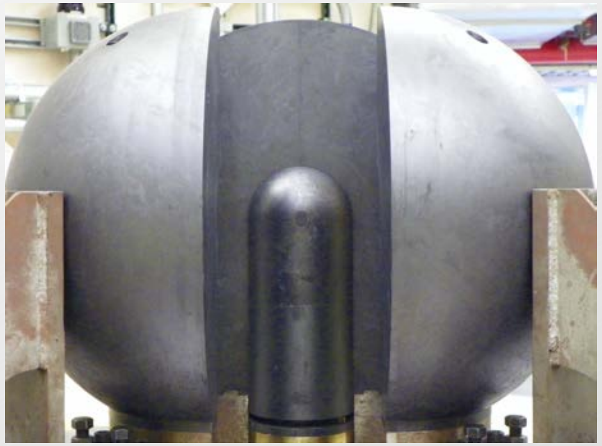
Measurements Made/Scheduled since 2012			
Task	Year(s)	Neutron Source(s)	Material(s) of Interest
CFPY*	**2012-2018**	NCERC – Planet, Comet & Flattop D-T Generator	^{233}U, ^{235}U, ^{238}U, ^{237}Np
SLFPY	**2015-2018**	NCERC – Godiva	^{238}U, ^{235}U, ^{239}Pu
CFPY	**2019**	D-T Generator	^{235}U
SLFPY	**2020**	NCERC – Godiva	^{237}Np
CFPY	**2021**	NCERC – Flattop Fission Chamber	^{235}U
SLFPY	**Scheduled March 2022**	NCERC – Godiva	^{233}U
CFPY	**Scheduled April 2022**	NCERC – Godiva Fission Chamber	^{239}Pu
SLFPY	**Scheduled April 2022**	OSU TRIGA	^{238}U

Neutron Sources

INNOVATE. COLLABORATE. DELIVER.

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



Cumulative FPY Task
Days to weeks post irradiation

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 $\times$ 10^{16} Total Fissions / burst

Short-Lived FPY Task
Hours to days post irradiation

D-T Generator (PNNL)

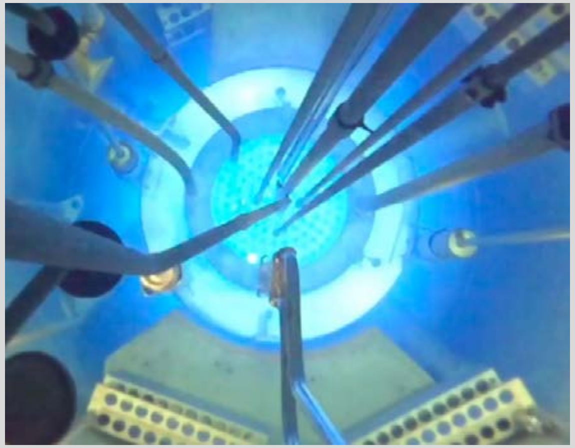
- Thermo D711 neutron generator
- Low scatter facility at PNNL
- Max neutron flux of 1×10^9 n/cm²/s

Cumulative FPY Task
Days to weeks post irradiation

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)

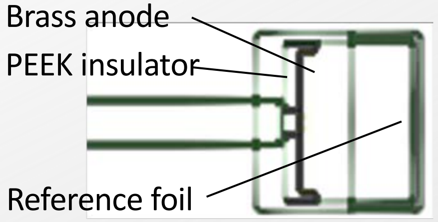


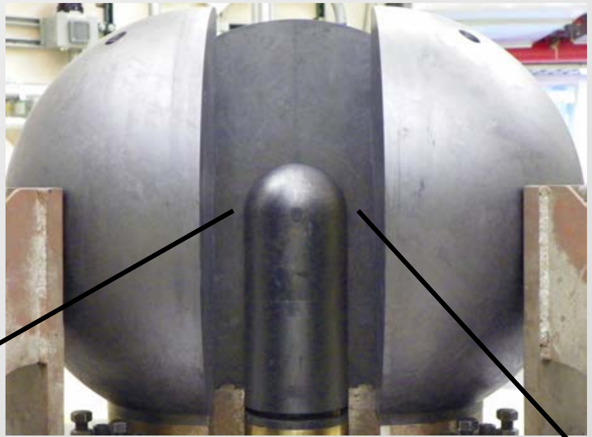
Short-Lived FPY Task
Seconds to days post irradiation

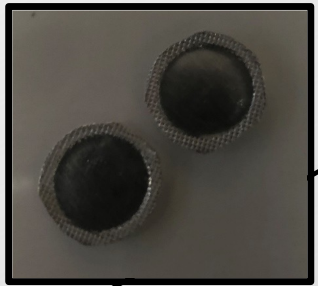
NCERC = National Criticality Experiments Research Center - at Nevada National Security Site (NNSS)

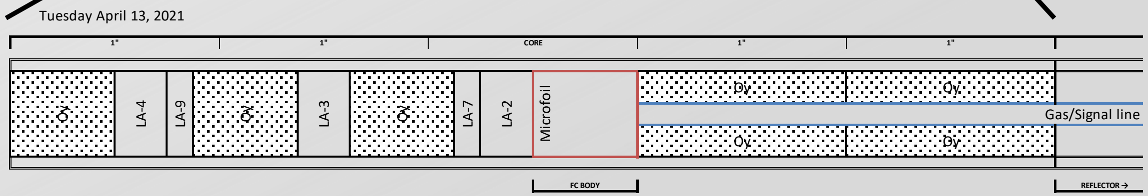
Cumulative Fission Product Yields (CFPY)

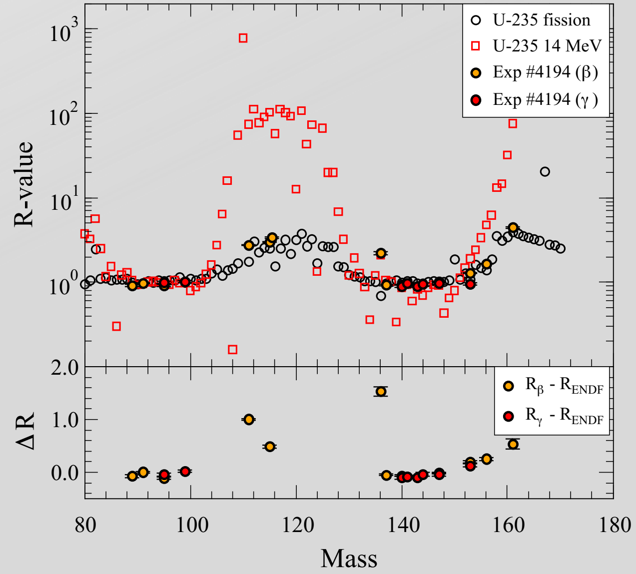
1. Fabricate Fission Chambers to Determine Absolute FPYs
2. Prepare High Quality Reference and Macro-foils
3. Irradiate Samples at NCERC or PNNL
4. Post-Irradiation Whole Foil γ–Counting
5. Radiochemical Dissolution and Analysis
6. R-values ($R_i^{j,k}$) and Absolute FP Yields ($Y_i^{j,k}$)
7. Publish Results and Post in XFOR





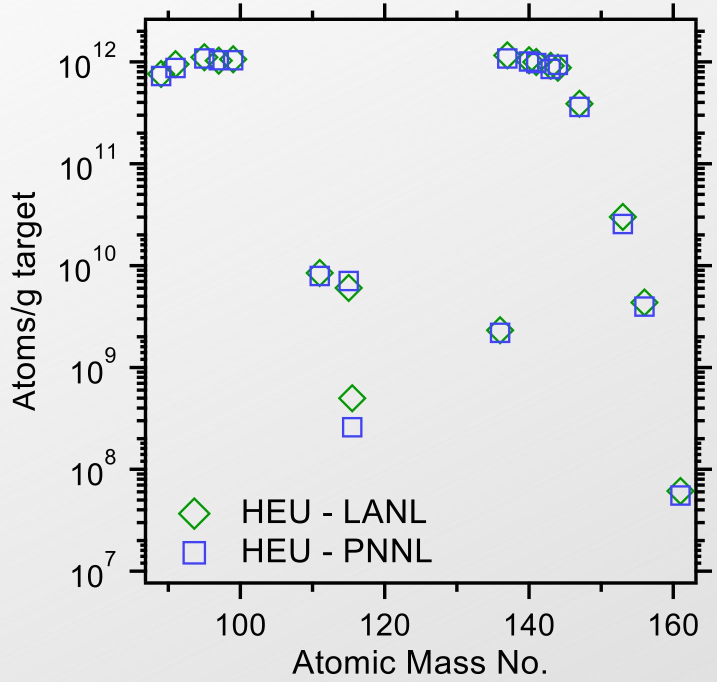


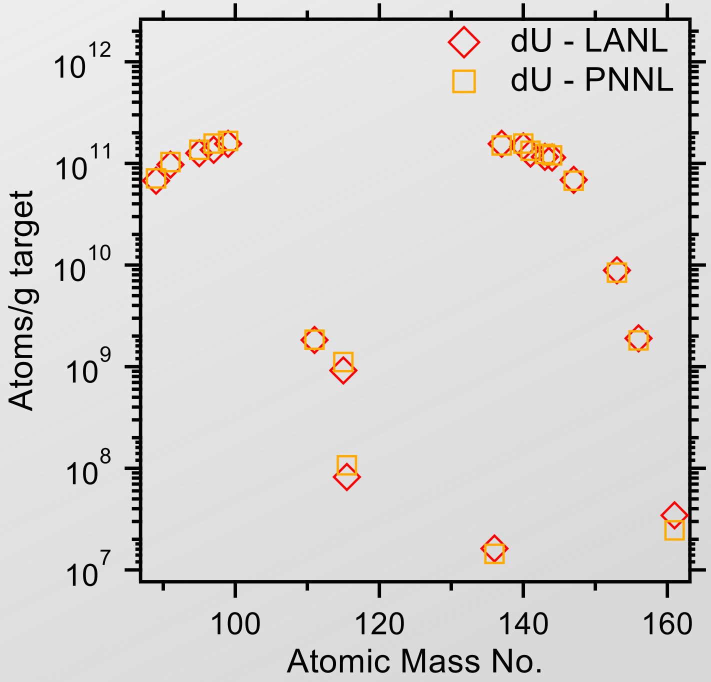




Fission Chambers | Actinide Samples | Neutrons | Radiochemistry | Relative (to Absolute) FP Yields

^{235}U Radiochemistry 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.

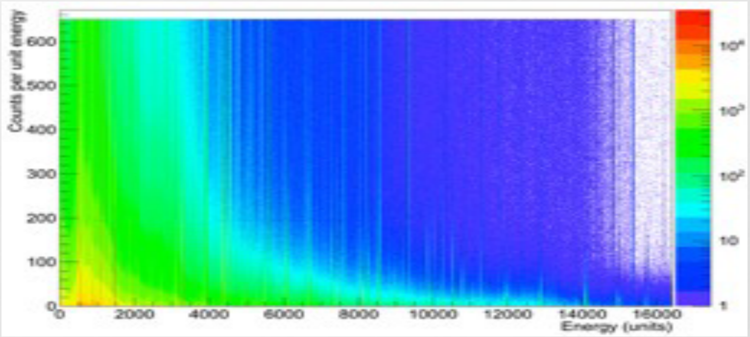
Short-Lived Fission Product Yields (SLFPY)

INNOVATE. COLLABORATE. DELIVER.

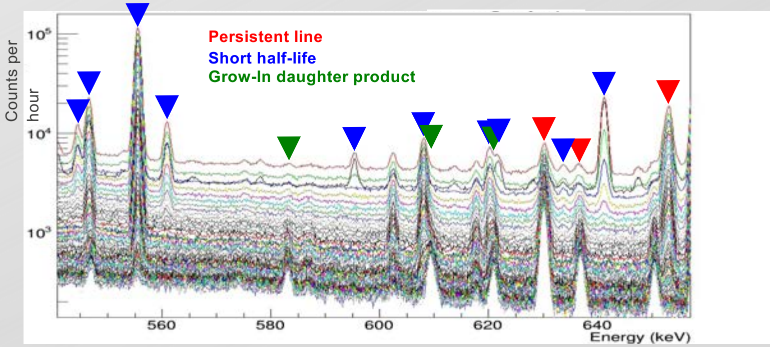
Fabricate High Quality Targets → Pulse Irradiate at NCERC → Acquire γ spectral data → Symmetric Temporal Spectral Analysis (LLNL) → Asymmetric Temporal Spectral Analysis (PNNL) → Evaluate Neutron Fluence & Fissions → Publish Fission Yields in Nuclear Data Sheets for NNDC

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

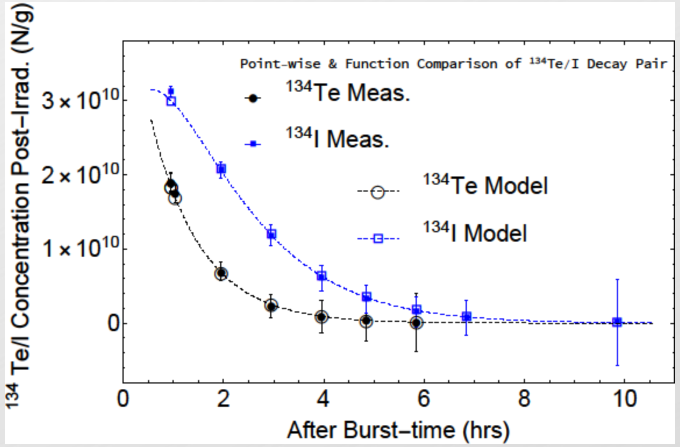
Time dependent gamma-ray spectra from seconds-days time scale



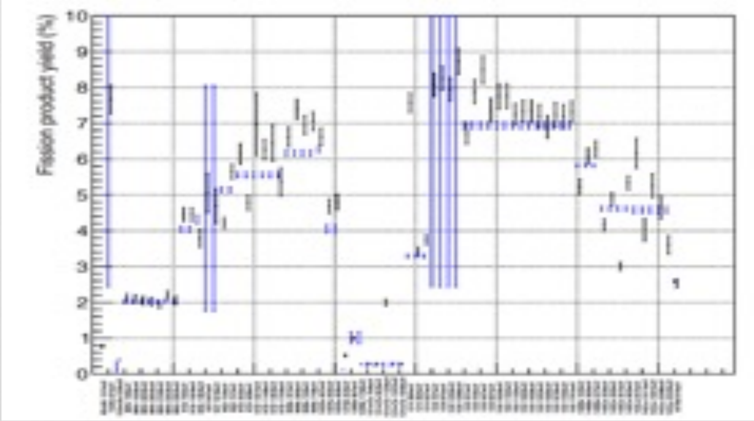
Very rich high-resolution spectroscopy allows us to unfold the time dependent behavior of FPs



Fit gamma-ray spectra and obtain FPY



Compare multiple FPYs to old database



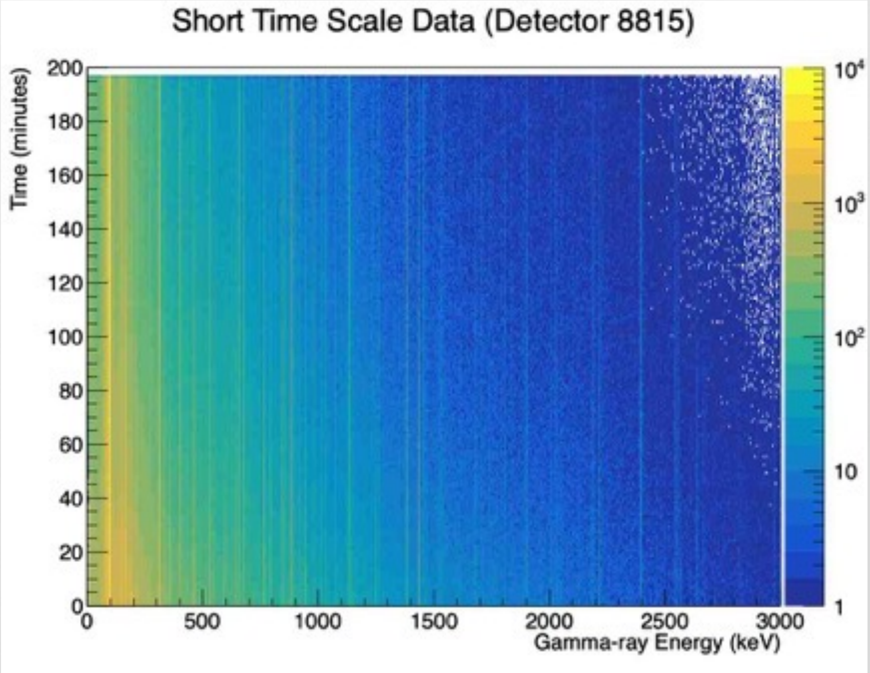
We are seeing good improvement in FPYs and issues with branching ratios effecting FPY results. This has implications for user community. Results are published in NDS.

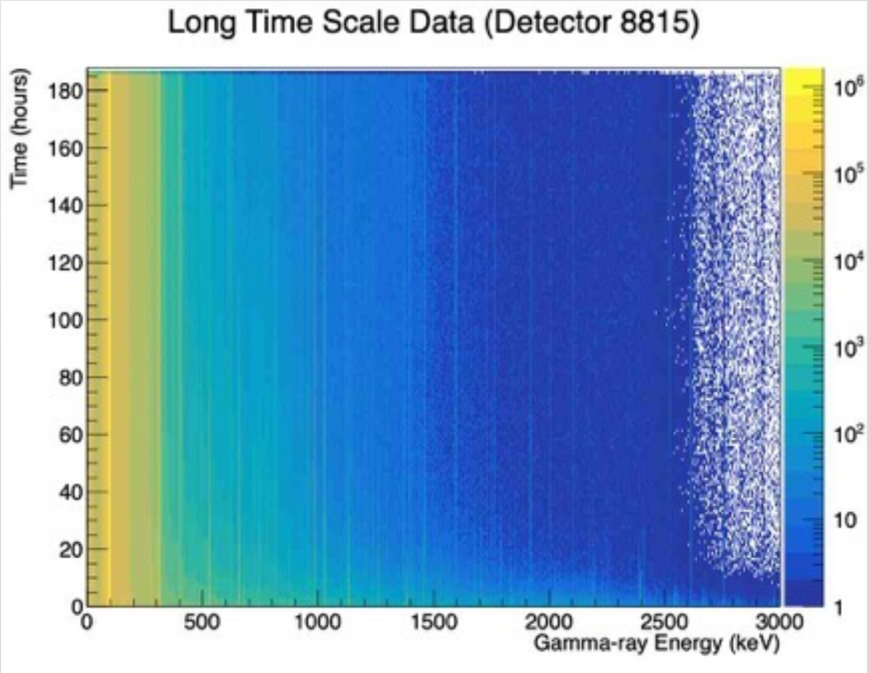
Nuclear Data Sheets **155**, 86 (2019).
Nuclear Data Sheets **163**, 249 (2020).

Short-Lived Fission Product Yields (SLFPY)

- Currently, we can retrieve the sample from Godiva ~30-40 minutes after the burst mode operation. Then begin measuring gamma-ray spectra at about 1 hour.
- Fit a peak in each in time bin and plot the intensity versus time (decay curve). Use decay curve to extract the activity of the isotope immediately after irradiation.
- Gamma-ray spectra have 1 keV one sigma resolution. Able to resolve individual isotopes.

Np237 FPY data from prompt burst operation at Godiva.



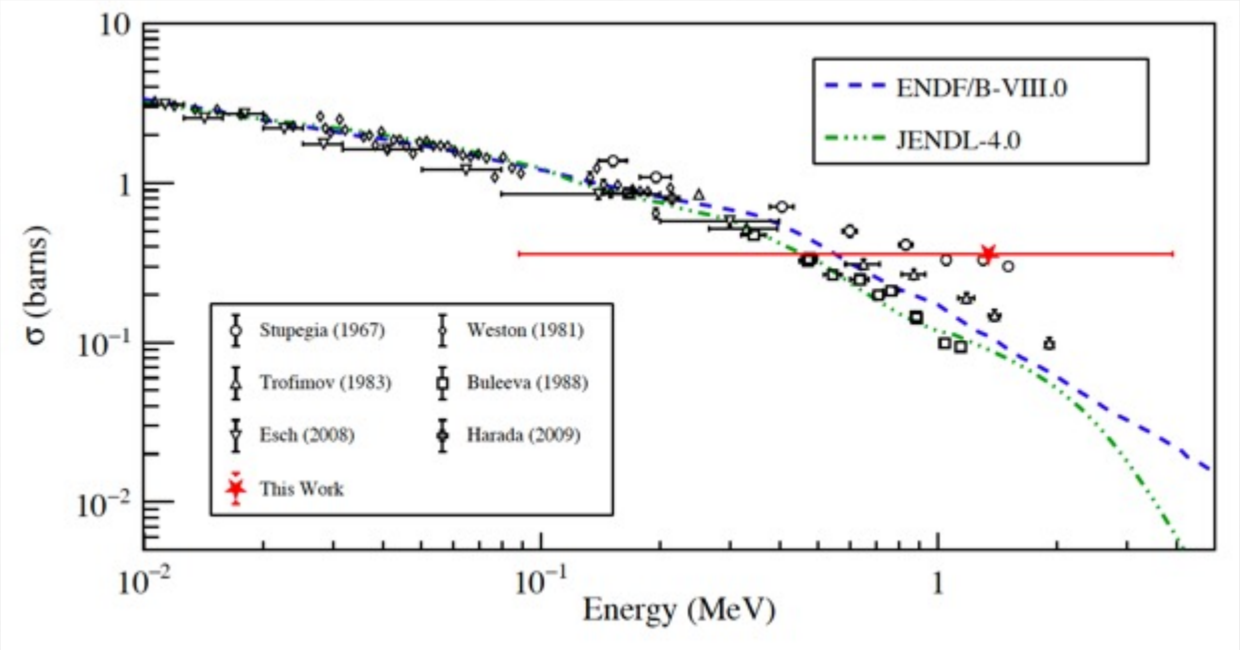


Short-Lived Fission Product Yields (SLFPY)

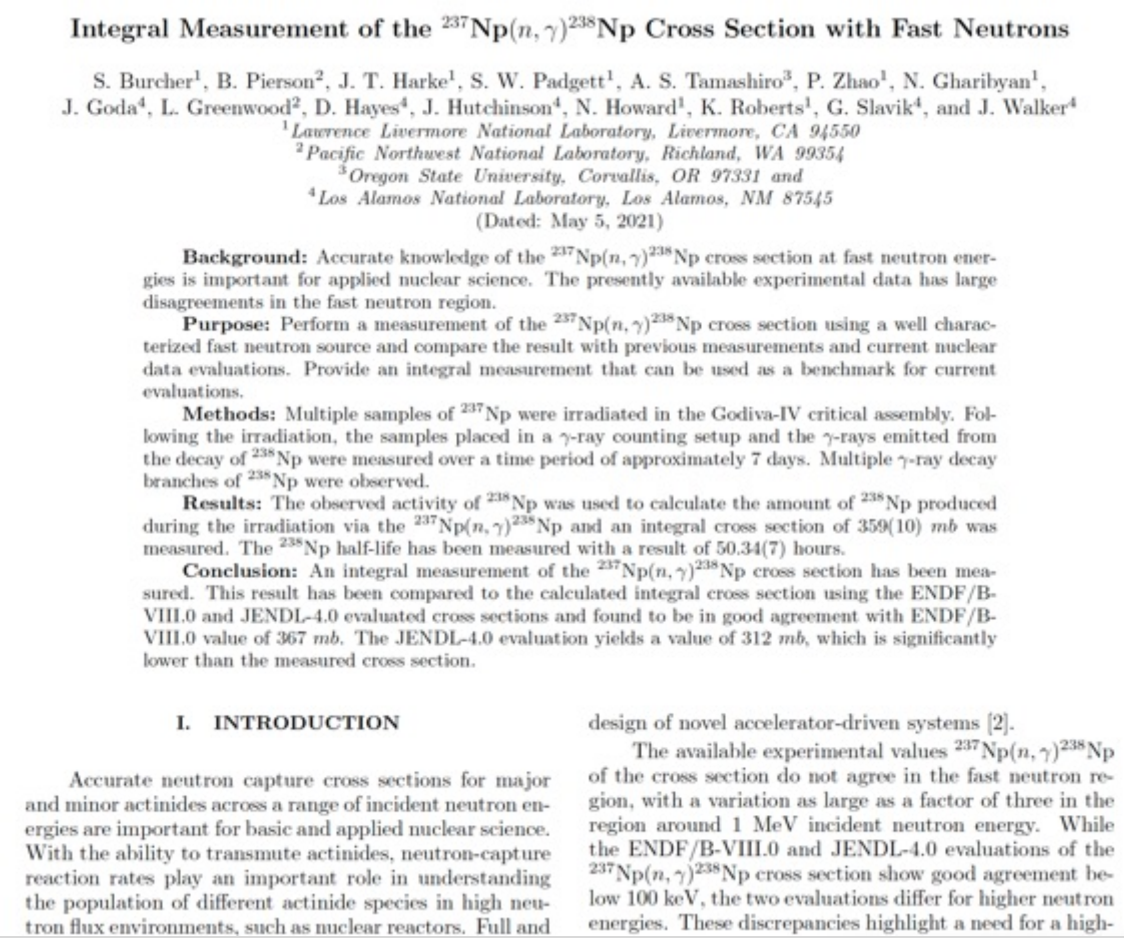
INNOVATE. COLLABORATE. DELIVER.

- Using Godiva, we are also able to make integral cross section measurements of neutron capture, (n,g), on actinide targets.

- Results from the Np237 shot produced a Np237(n,g)Np238 cross section which helped resolve a discrepancy in the national nuclear database.



Paper being submitted to Physical Review C journal



Integral Measurement of the ^{237}Np$(n,\gamma)^{238}$Np Cross Section with Fast Neutrons

S. Burcher[1], B. Pierson[2], J. T. Harke[1], S. W. Padgett[1], A. S. Tamashiro[3], P. Zhao[1], N. Gharibyan[1], J. Goda[4], L. Greenwood[2], D. Hayes[4], J. Hutchinson[4], N. Howard[1], K. Roberts[1], G. Slavik[4], and J. Walker[4]

[1]*Lawrence Livermore National Laboratory, Livermore, CA 94550*
[2]*Pacific Northwest National Laboratory, Richland, WA 99354*
[3]*Oregon State University, Corvallis, OR 97331 and*
[4]*Los Alamos National Laboratory, Los Alamos, NM 87545*

(Dated: May 5, 2021)

Background: Accurate knowledge of the ^{237}Np$(n,\gamma)^{238}$Np cross section at fast neutron energies is important for applied nuclear science. The presently available experimental data has large disagreements in the fast neutron region.
Purpose: Perform a measurement of the ^{237}Np$(n,\gamma)^{238}$Np cross section using a well characterized fast neutron source and compare the result with previous measurements and current nuclear data evaluations. Provide an integral measurement that can be used as a benchmark for current evaluations.
Methods: Multiple samples of ^{237}Np were irradiated in the Godiva-IV critical assembly. Following the irradiation, the samples placed in a γ-ray counting setup and the γ-rays emitted from the decay of ^{238}Np were measured over a time period of approximately 7 days. Multiple γ-ray decay branches of ^{238}Np were observed.
Results: The observed activity of ^{238}Np was used to calculate the amount of ^{238}Np produced during the irradiation via the ^{237}Np$(n,\gamma)^{238}$Np and an integral cross section of 359(10) *mb* was measured. The ^{238}Np half-life has been measured with a result of 50.34(7) hours.
Conclusion: An integral measurement of the ^{237}Np$(n,\gamma)^{238}$Np cross section has been measured. This result has been compared to the calculated integral cross section using the ENDF/B-VIII.0 and JENDL-4.0 evaluated cross sections and found to be in good agreement with ENDF/B-VIII.0 value of 367 *mb*. The JENDL-4.0 evaluation yields a value of 312 *mb*, which is significantly lower than the measured cross section.

I. INTRODUCTION

Accurate neutron capture cross sections for major and minor actinides across a range of incident neutron energies are important for basic and applied nuclear science. With the ability to transmute actinides, neutron-capture reaction rates play an important role in understanding the population of different actinide species in high neutron flux environments, such as nuclear reactors. Full and design of novel accelerator-driven systems [2].

The available experimental values ^{237}Np$(n,\gamma)^{238}$Np of the cross section do not agree in the fast neutron region, with a variation as large as a factor of three in the region around 1 MeV incident neutron energy. While the ENDF/B-VIII.0 and JENDL-4.0 evaluations of the ^{237}Np$(n,\gamma)^{238}$Np cross section show good agreement below 100 keV, the two evaluations differ for higher neutron energies. These discrepancies highlight a need for a high-

Summary: FY21 Experimental Activities

INNOVATE. COLLABORATE. DELIVER.

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. Sample analysis in FY22.**

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

Outlook: FY22 Experimental Activities

INNOVATE. COLLABORATE. DELIVER.

Dates	Task	Activity
Feb 1-3, 2022	CFPY	^{239}Pu fission chamber testing at the MIT NRL
Mar 7-10, 2022	CFPY	^{239}Pu 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**
Feb 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**
TBD	**CFPY**	**^{239}Pu production irradiation on 14 MeV D-T source**

We are planning 3 experimental campaigns at NCERC, 1 at PNNL, 1 at LLNL, and 1 at OSU in FY22.

The Full Team

INNOVATE. COLLABORATE. DELIVER.

- J. Berger
- M.A. Boggs
- M. Boswell
- E.M. Bond
- S.M. Bowen
- T.A. Bredeweg
- J.A. Bounds
- G.H. Brooks, Jr.
- M.R. Cisneros
- D.L. Cox III
- T.E. Cutler
- D.E. Dry
- J.A. Favorite
- M.J. Gallegos
- A.J. Gaunt
- R.R. Gibson
- J.M. Goda
- M.E. Gooden
- S.K. Hanson
- D.K. Hayes
- L.A. Hudston
- J.D. Hutchinson
- K.R. Jackman
- M.R. James
- C.C. Keith
- W.S. Kinman
- C.A. Lance
- G. Lee
- R.C. Little
- M.R. MacInnes
- C. Margiotta
- A.M. Marenco
- I. May
- J.L. McGovern
- G.E. McKenzie IV
- D. Meininger
- D.K. Melton
- J.L. Miller
- A.D. Montoya
- W.L. Myers
- W.J. Oldham
- A.C. Olson
- S.D. Pacheco
- S.D. Reilly
- R.J. Rendon
- A.R. Roman
- J.R. Romero
- R.S. Rundberg
- R.G. Sanchez
- A.R. Schake
- N.C. Smythe
- J.L. Walker
- M.C. White
- C.W. Wilkerson, Jr.
- J.M. Williams
- M.S. Wren
- E. Arnold
- L. Arrigo
- C. Beck
- J. Bowen
- M. Cantaloub
- J. Friese
- B. Gartman
- L. Greenwood
- M. Haney
- S. Herman
- D. Lucas
- L. Metz
- B.D. Pierson
- S. Tedrow
- T. Trang-le
- N. Uhnak
- B. Bandong
- S. Burcher
- J.A. Church
- N. Gharibyan
- J.J. Goodell
- J.T. Harke
- N. Harward
- S.W. Padgett
- P. Zhao
- K. Roberts
- G. Slavik
- S. Menn
- L. Minc
- C. Palmer
- S. Reese
- A.S. Tamashiro

This work was funded by the Office of Defense Nuclear Nonproliferation Research and Development of the U.S. Department of Energy’s National Nuclear Security Administration.

This work utilized the National Criticality Experiments Research Center supported by the Nuclear Criticality Safety Program of the U.S. Department of Energy’s National Nuclear Security Administration

Backup Slides

FY21-22 Experimental Plan

INNOVATE. COLLABORATE. DELIVER.

^{235}U Cumulative FPYs at NCERC/Flattop:

- ^{235}U FC testing at the MIT Nuclear Reactor Lab
  Oct 27 - 29, 2020 – COMPLETED
- ^{235}U FC testing on Flattop at NCERC
  *Nov 17 - 19, 2020 – POSTPONED**
- ^{235}U FC testing at the MIT Nuclear Reactor Lab
  *March 2 - 4, 2021 – CANCELED**
- ^{235}U FC testing on Flattop at NCERC
  April 7 - 9, 2021 – COMPLETED
- ^{235}U foil irradiations on the Flattop critical assembly
  April 12 - 14, 2021 – COMPLETED
  - Direct foil counting <u>and</u> radiochemical analyses to determine relative CFPYs.
  - Parallel radiochemistry separations (days) and counting (weeks) at LANL and PNNL.
  - Collocated fission chambers to determine absolute CFPYs.

*COVID-19 related issues

^{239}Pu Cumulative FPYs at NCERC/Godiva:

- ^{239}Pu FC testing at the MIT Nuclear Reactor Lab
  To be Scheduled (Jan/Feb 2022)
- ^{239}Pu FC testing on Godiva at NCERC
  Scheduled Feb/March 2022
- ^{239}Pu foil irradiations on the Godiva critical assembly
  Scheduled April 25-28, 2022
  - Direct foil counting <u>and</u> radiochemical analyses to determine relative CFPYs.
  - Parallel radiochemistry separations (days) and counting (weeks) at LANL and PNNL.
  - Collocated fission chambers to determine absolute CFPYs.

^{239}Pu Cumulative FPYs at 14 MeV:

- ^{239}Pu foil irradiations on the PNNL D-T Generator
  To be Scheduled
  - Same as above

Why is nuclear physics / nuclear data important

INNOVATE. COLLABORATE. DELIVER.

The defining signature of a nuclear detonation is the presence of nuclear reaction products in the debris.

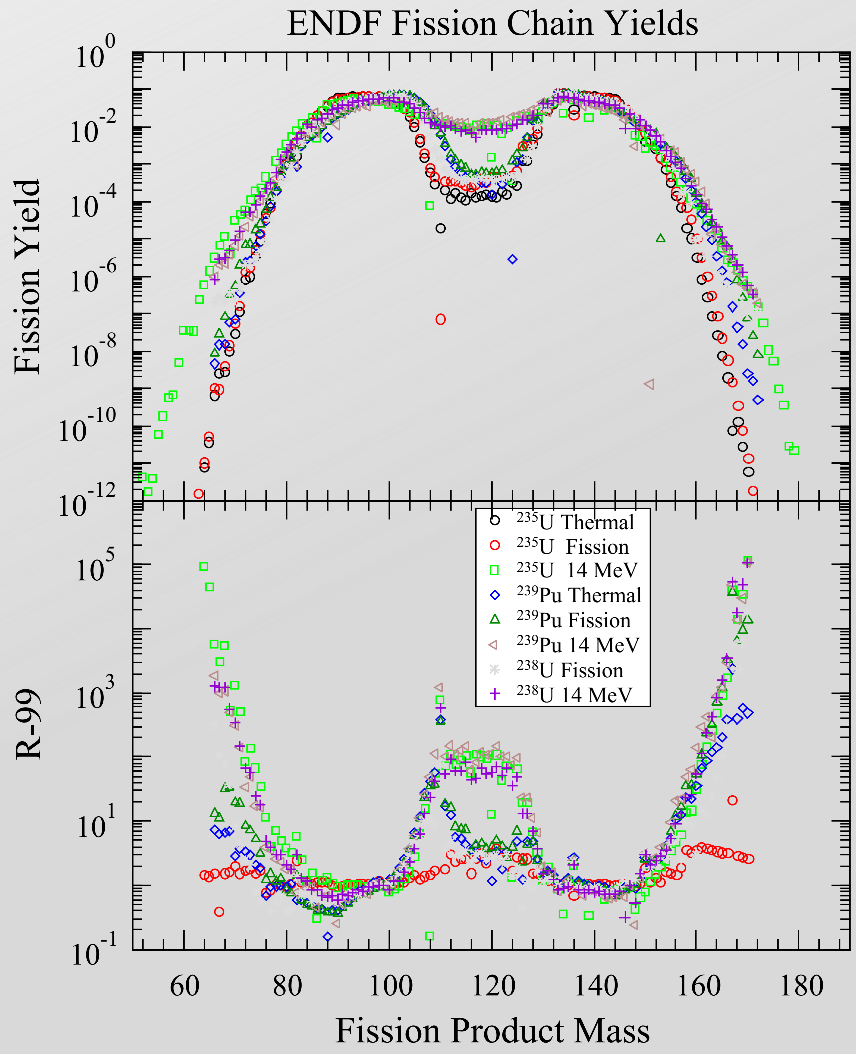
These can be from

- Fission products (FP) from the fuel.
- Activation products (AP) from the fuel or nearby materials.

Production/Destruction depends on fuel type and neutron spectrum.

$$RR = \int n\sigma(E_n)\phi(E_n)dE_n$$

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



MCNP neutron-flux simulations

Motivation: With the progress in FPY measurements and theoretical models, we want to update our estimates for the R-values using new energy dependent FPY data.

Technical approach:

1) Reproducing the average E_n as a function of position as published in M.B. Chadwick, et al., Nucl. Data Sheets 111, 2923 (2010) for Jezebel, Big-10, Godiva, Flattop-U, Flattop-Pu (done).
2) Reproducing the average E_n and R-values for ^{239}Pu+n as published in M.B. Chadwick, et al., Nucl. Data Sheets 111, 2923 (2010) (in progress).
3) Folding the simulated neutron flux with new FPY calculations performed by T-2 to obtain calculated R-values and compare to experimental results (to be performed).

Personnel: T. A. Bredeweg, A. E. Lovell, G. Rusev, P. Talou.

Leverage of other work: FPY theory development at T-2.