PNNL-34287

Update of the Cesium-136 Cumulative Fission Yields at Multiple Neutron Energies

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U.S. DEPARTMENT OF ENERGY

Prepared for the U.S. Department of Energy
under Contract DE-AC05-76RL01830

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Executive Summary

Nuclear data is foundational to several fields, including nuclear forensics. Nuclear forensics investigations of fission events, relies heavily on the cumulative fission yields, however several highly useful fission products suffer from very poor nuclear data including cumulative fission yields or associated uncertainties in that value. Cesium-136 is a highly relevant example of this issue, where the cumulative fission yield may be accurate but its uncertainty high enough to preclude its usefulness. In this work we evaluate and calculate an updated cumulative fission yield for ^{136}Cs for ^{235}U, ^{238}U, and ^{239}Pu at multiple neutron energies or sources using data obtained from irradiation campaigns. In all cases these newly determined cumulative fission yields provided small changes to the actual fission yields but had dramatic improvements to the uncertainty in those yields. These improvements in uncertainty will enable nuclear forensics end users to use ^{136}Cs data with more confidence.

Acknowledgments

The authors would like to acknowledge all the people that worked on the irradiation campaigns used in this work, past and present, the author list is by no means comprehensive but makes an effort to include those still at PNNL.

Acronyms and Abbreviations

CFY	Cumulative Fission Yield
ENDF	Evaluated Nuclear Data File
JEFF	Joint Evaluated Fission and Fusion File
JENDL	Japanese Evaluated Nuclear Data Library
MITR	Massachusetts Institute of Technology Reactor
HPGe	High Purity Germanium
GEA	Gamma Energy Analysis

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1.0 Introduction

Nuclear data is fundamental to predicting behavior of nuclear processes, one of the most important being the fission process. Of the nuclear data, the fission yields of a given fission product are highly useful for several activities including nuclear forensics but many of the shielded or lower yield fission products have poorly established yields or high uncertainty. An excellent example of the high uncertainty is ^{136}Cs, a blocked fission product with a particularly high uncertainty associated with its yield. A blocked fission product is a fission product that is only produced through fission with no production path from decay of fission products. This is illustrated in Table Table 1-1, where the range of cumulative fission yields of ^{136}Cs ranges from 10^{-6} to 10^{-2} depending on the actinide and neutron energy all with an associated 64% uncertainty.

Table 1-1. Cumulative fission yields of ^{136}Cs for common actinides. All yields have an associated ± 64% uncertainty. All values obtained from ENDF V.III.0 database.

	Cumulative Fission Yields (CFY)					
Neutron Energy	**^{233}U**	**^{235}U**	**^{238}U**	**^{237}Np**	**^{239}Pu**	**^{241}Am**
0.0253 eV	1.30x10^{-3}	5.54x10^{-5}		1.23 x10^{-4}	9.743x10^{-4}	2.566 x10^{-3}
500 keV	1.06x10^{-3}	1.17x10^{-4}	9.60x10^{-6}	5.85 x10^{-4}	6.182x10^{-4}	4.524x10^{-3}
2 MeV					6.315x10^{-4}	
14 MeV	8.23x10^{-3}	2.26x10^{-3}	2.12x10^{-4}	1.15x10^{-3}	2.674x10^{-3}	1.249x10^{-2}

2.0 Methodologies

The absolute fission yield can be determined using the experimentally determined atoms of a given fission product and dividing it by the number of fissions, however it should be noted that this is relative to the total number of fissions determined relative to well established fission product's fission yields. The calculation is shown below in Section 2.1, showing both the generic equation as well as the using ^{136}Cs thermal and fission neutrons as examples of the calculation.

The availability of primary references for many of the thermal calibrations are controlled due to their protected nature, however the data from these references are used. To provide examples of those reports see, PNNL-X-900-2189, or PNNL-NC-0891. A similar report exists for each of the thermal HEU irradiations, however no further thermal calibration reports will be referenced.

Due to the high variability of the ^{136}Cs fission yields with actinide fuel and neutron energy a comprehensive analysis of the neutron spectrum used for activation is shown in 5.0Appendix A 5.0A.2. The analysis of the neutron spectrum used several monitor foils with well characterized activation cross sections at many neutron energies along with modeling using STAYSL and MCNP.

2.1 Absolute Fission Yield Calculation Example

Equation 2-1 is used to calculate the absolute fission yield and is shown in Equation 2-2. Calculation of the number of fissions is determined using the literature CFY of ^{99}Mo and the atoms of ^{99}Mo determined from the dissolved irradiated target.

$$CFY_X = \frac{Atoms_X/g}{Fissions/g}$$

Equation 2-1

$$CFY_{Cs136thermal} = \frac{Atoms_{Cs136}/g}{Fissions/g} = \frac{2.04x10^8 \pm 3.7\%\ atoms/g}{3.53x10^{12} \pm 3.1\%\ fission/g} = 5.78x10^{-5} \pm 7.6\%$$

$$CFY_{Cs136fission} = \frac{Atoms_{Cs136}/g}{Fissions/g} = \frac{7.65x10^6 \pm 2.0\%\ atoms/g}{5.97x10^{10} \pm 2.5\%\ fission/g} = 1.28x10^{-4} \pm 3.2\%$$

Equation 2-2

2.2 Separation Methods and Analysis

2.2.1 Separation Methods

The separation methods used for each of the individual irradiation experiments is the topic of multiple reports included in the references, some of which are not available for wide distribution; for those reports they will be cited only. In general, the separation procedure is generally consistent between each of the irradiations, regardless of the neutron spectrum. Specific details on the separation methods are outside of the scope of this report and can be found in each individual report included in the references.

2.2.2 Analysis Methods

The analysis, particularly at earlier dates is subject to the level of practice for these measurements, as such as each individual experiment occurred the skill in the analytical measurements increased. Instrumentation changed over time as well, though each of the analyses uses similar equipment operated under the same quality control and quality assurance processes. Gamma analysis of the separated fractions of Cs was done using HPGe detectors and analyzed at least once, however multiple analyses were performed for several the irradiations.

The mathematics used to determine the R-value relies on the use of a r_{hist} that is a running average of five consecutive thermal calibration exercises. The use of the r_{hist} to determine the R-value was not used until the 2018 irradiations. Prior to that the CFY from ENDF was used, with some variation on which version of ENDF ranging from ENDF.VI to ENDF.VIII.0. There is little change to the CFY of ^{136}Cs over that time frame for any of the U isotopes examined.

3.0 Results

The calculation of the CFY of ^{136}Cs was determined from eight thermal irradiations using the MIT Reactor between 2011 and 2022, producing a value of 5.74x10^{-5} ± 3.6% compared to the value of 5.45x10^{-5} ± 64% for thermal neutrons after extensive chemical separations. Figure 3-1 shows a comparison of the Cs data obtained from the irradiation campaigns, with the average and ±1σ are plotted at dashed lines.

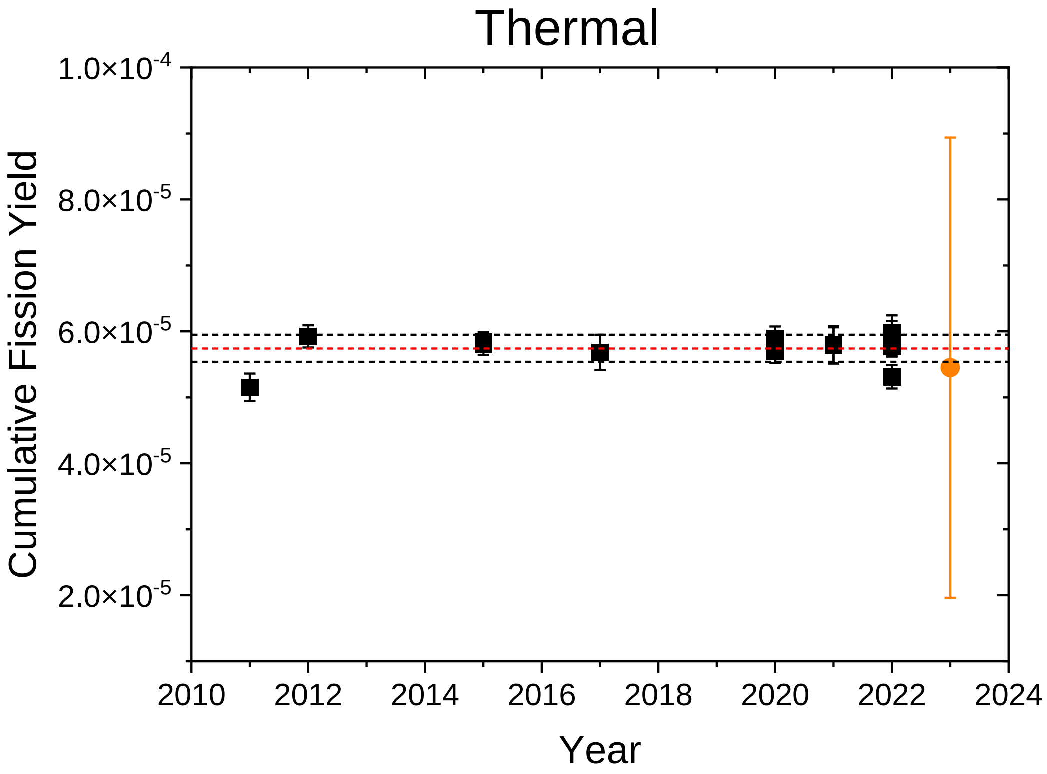

Figure 3-1. ^{136}Cs cummulative fission yields from the fission spectrum irradiation of ^{235}U using the MITR reactor at MIT. The black symbols are the calculated CFY, the orange symbol is the ENDF value. The dashed red line shows the average calculated CFY, the black dashed line show 1σ in that value.

The ^{136}Cs CFY results for fission spectrum neutrons are shown Figure 3-2 using data obtained from the Flattop and Godiva IV critical assemblies at NCERC. These experimentally determined CFYs are compared to the ENDF literature value. ENDF CFY is lower than all experimentally determined CFYs, however there is excellent agreement between the five campaigns and critical assemblies. The cores used in each campaign inside of critical assemblies are not identical, but there is a relatively narrow range of neutron energy regardless of the fuel identity i.e., ^{239}Pu vs ^{235}U.

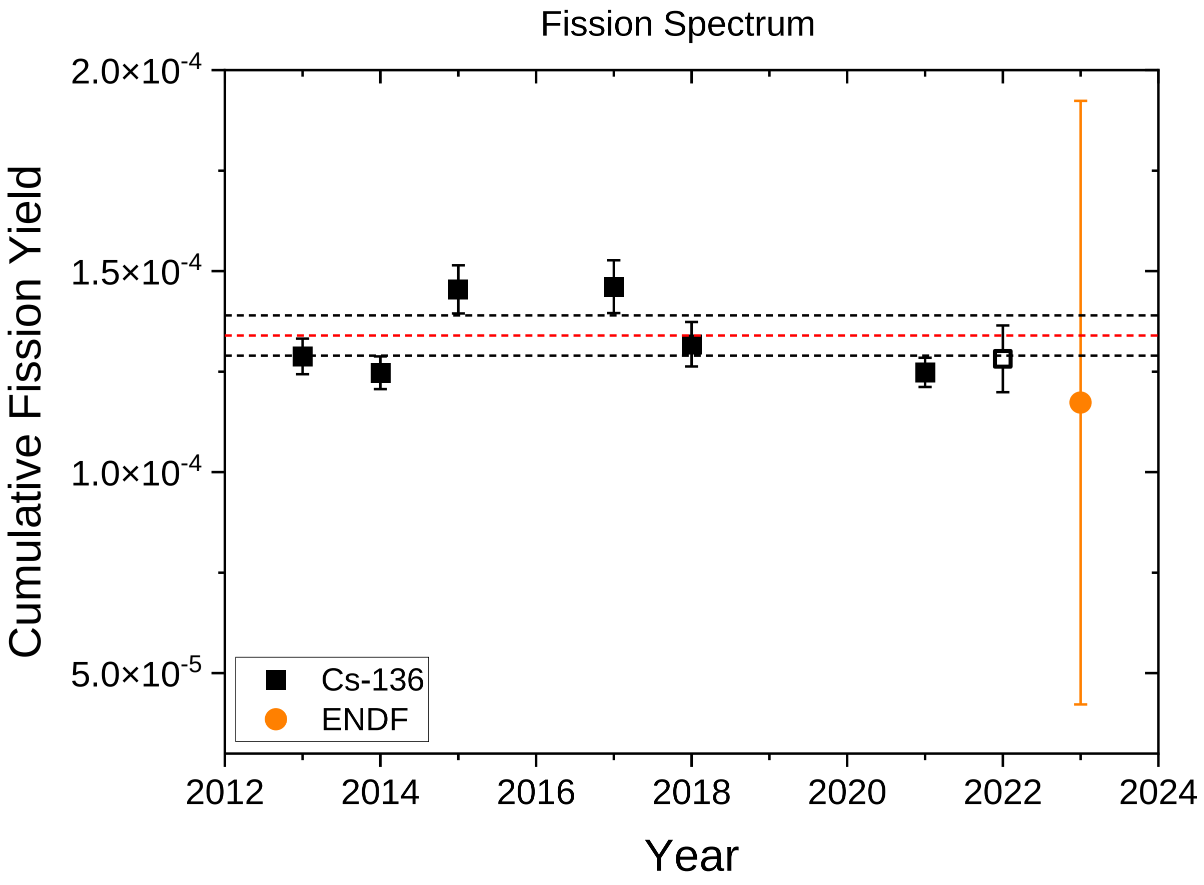

Figure 3-2. ^{136}Cs cummulative fission yields from the fission spectrum irradiation of ^{235}U using the Flattop and Godiva critical assemblies. The black symbols are the calculated CFY from the Flattop critical assembly and the open black symbol is from the Godiva critical assembly, the orange symbol is the ENDF value for 500 keV neutrons. The dashed red line shows the average calculated CFY, the black dashed line show 1σ in that value.

The ^{136}Cs CFY from ^{238}U is highly susceptible to the neutron energy, increasing with increasing neutron energy. Shown in Figure 3-3 and Figure 3-4 are the calculated CFY for each of the campaigns for 14 MeV and fission spectrum neutrons. There is high agreement between the 14 MeV campaigns with good agreement with the ENDF value, though with the associated 64% uncertainty. Determining the CFY from fission spectrum neutrons due to the low yield is difficult, chemical separations are necessary to adequately detect the isotope. However, if the separated chemical yield is low relative to other campaigns there is the potential that no Cs is detected, which is the reason the missing data from the 2014, 2015, 2017.

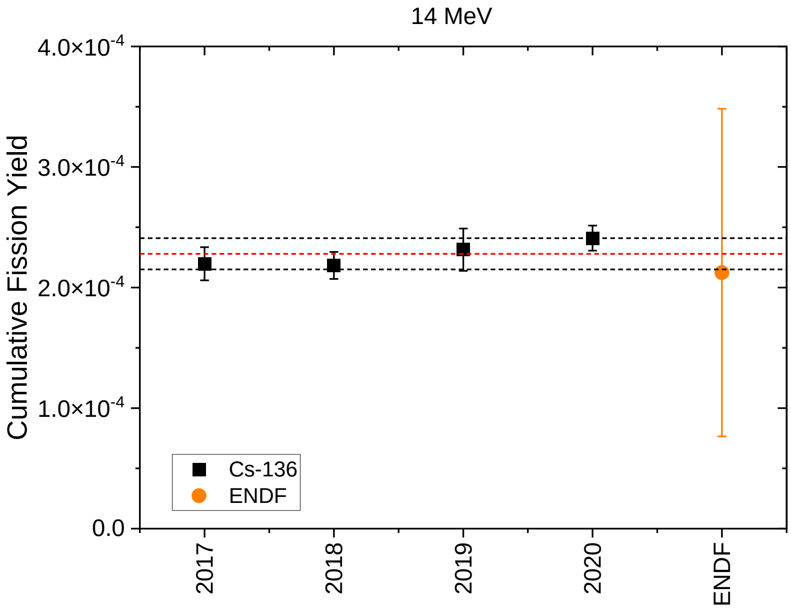

Figure 3-3. ^{136}Cs cummulative fission yields from ^{238}U fission from 14 MeV using the PNNL D711 D-T generator. The black symbols are the calculated CFY, the orange symbol is the ENDF value. The dashed red line shows the average calculated CFY, the black dashed line show 1σ in that value.

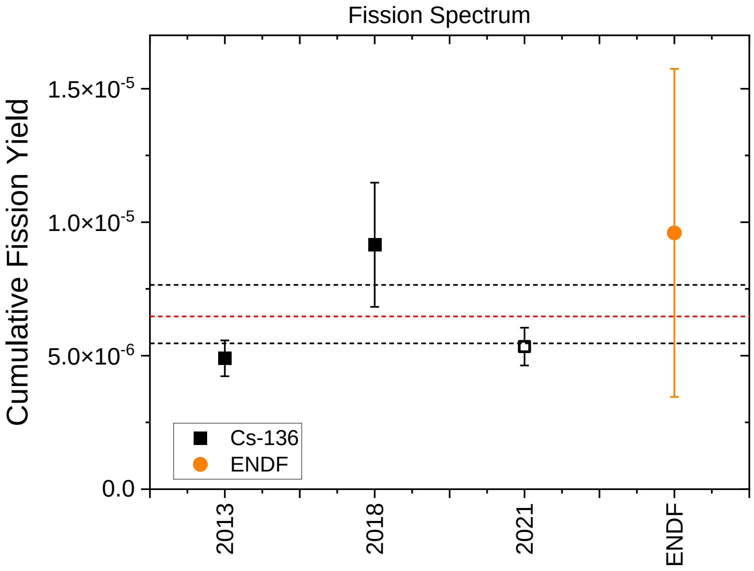

Figure 3-4. ^{136}Cs cummulative fission yields from ^{238}U fission from fission spectrum neutrons using the Flattop and Godiva IV Critical assemblies. The black symbols are the calculated CFY from Flattop, the open symbol is the calculated CFY from Godiva IV the orange symbol is the ENDF value. The dashed red line shows the average calculated CFY, the black dashed line show 1σ in that value.

Validation of the methodologies was conducted by calculating the absolute CFY of ^{137}Cs. The similarity of a peak yield fission product CFY like that of ^{137}Cs to the available literature CFY, provides a validation of the approach. The CFY for ^{136}Cs from each irradiation campaign are included in Table 3-1. Similarly, the CFY for ^{137}Cs is included in Table 3-2; the ENDF CFYs are also included for direct comparison to the neutron energy for the calculated CFY. The CFY determined for ^{238}U for fission spectrum neutrons is included for comparison only, the value found in this evaluation is dependent on highly difficult measurements to make due to low yield. Two other neutron energies have been used in the past, the Comet critical assembly and the WSU TRIGA reactor with the target in a boron carbide (B_4C) shield. Though significantly different in practice, the net neutron spectrum is very similar. Results from these investigations are included in Table 3-1. Many of the values included in the table are the result of a single irradiation experiment, these values have been denoted with an asterisk. These isotopes are both high-yield fission products with relatively long half-lives and are generally easily quantifiable which lends credence to the calculated fission yields through this method. The CFY of ^{239}Pu was also included, calculated from a single experiment for each neutron energy. It should be noted that the ^{239}Pu 14 MeV data was obtained from a single irradiation of a Pu target and measured 21 times over 110 days without dissolution or chemical processing, therefore the contribution to the gamma spectrum of ^{241}Am was significant precluding ^{137}Cs analysis, but the ^{136}Cs remained detectable. [PNNL-33060].

Table 3-1. Absolute fission yields of ^{136}Cs relative to total fissions. Values are average of multiple irradiations unless denoted by an *. The CFY of ^{136}Cs using Flattop on ^{238}U is included in italics for comparison purposes.The uncertainty is 1σ in that value.

Neutron Energy	**^{235}U**	**^{238}U**	**^{239}Pu**
0.0253 eV	5.74x10^{-5} ± 3.6%	N/A	-
Flattop (500keV)	1.34x10^{-4} ± 3.8%	*6.47x10^{-6} ± 18.3%*	-
14 MeV	2.24x10^{-3} ± 2.8%*	2.28x10^{-4} ± 5.9%	7.30x10^{-3} 4.3%*
GODIVA IV	1.28x10^{-4} ± 6.5%	ND	1.30x10^{-3} ± 3.5%*
Comet	N/A	1.17x10^{-4} ± 2.8%*	N/A
WSU B4C	N/A	1.27x10^{-4} ± 5.9%*	N/A

Table 3-2. Absolute fission yields of ^{137}Cs realtive to total fissions. Values are average of multiple irradiations unless denoted by an *. The uncertainty is 1σ in that value.

Neutron Energy	**^{235}U**	**ENDF**	**^{238}U**	**ENDF**	**^{239}Pu**	**ENDF**
0.0253 eV	6.31x10^{-2} ± 3.6%	6.18x10^{-2}	N/A		-	
Flattop (500keV)	6.24x10^{-2} ± 3.8%	6.22x10^{-2}	6.14x10^{-2} ± 4.2%	6.05x10^{-2}	-	
14 MeV	5.05x10^{-2} ± 8.2%*	4.92x10^{-2}	5.77x10^{-2} ± 7.3%	5.15x10^{2}	N/A*	
GODIVA IV	6.23x10^{-2} ± 6.1%	6.22x10^{-2}	6.12x10^{-2} ± 5.6%	6.05x10^{-2}	6.83x10^{-2} ± 5.6%*	6.58x10^{-2}
Comet	N/A		6.62x10^{-2} ± 2.0%*	6.05x10^{-2}	N/A	
WSU B4C	N/A		5.92x10^{-2} ± 5.8%*	6.05x10^{-2}	N/A	
* Interferences between the ^{137}Cs and ^{241}Am gamma emissions						

A complete list of ^{136}Cs literature CFY for ^{235}U, ^{238}U, and ^{239}Pu are included in Appendix 5.0A.1 for three nuclear databases, ENDF.V.III.0, JEFF 3.3, and JENDL 4.0. The values calculated using the methods above compare well to the literature values, ignoring the uncertainty. For example, the ENDF value for ^{136}Cs CFY is 5.54x10^{-5} ± 64%, while we determined a value of 5.74x10^{-5} ± 3.6%, a difference in yields that is within the uncertainty of our calculated value. Similarly, the other fuels or neutron energies can be compared, showing a high degree of agreement with the literature yields for most analyses. The databases represent the best current nuclear data, but as this data is updated periodically, there are CFYs that are outside of what is reported in ENDF. A stark example of this is the ^{238}U fission spectrum yields using either Flattop critical assemblies, where the CFY found in experiments is within the enormous bounds of the uncertainty of the literature CFY but is consistent between critical assemblies as shown in Figure 3-4. Without further irradiations it is difficult to draw a concrete conclusion due to the low yield of the fission product.

3.1 R-values

The R-value can be used to obtain a CFY, but it can also be used directly as an alternative to a CFY. However, to calculate the CFY using the R-values requires a well-known CFY at thermal energies and the literature CFYs have significant uncertainty associated with them. Propagating that uncertainty leads to a >90% uncertainty in the calculated value. The R-values for ^{235}U at thermal and fission spectrum are shown in Figure 3-5 and Figure 3-6, respectively. R-values for ^{238}U at fission and 14 MeV are shown in Figure 3-7 and Figure 3-8, respectively.

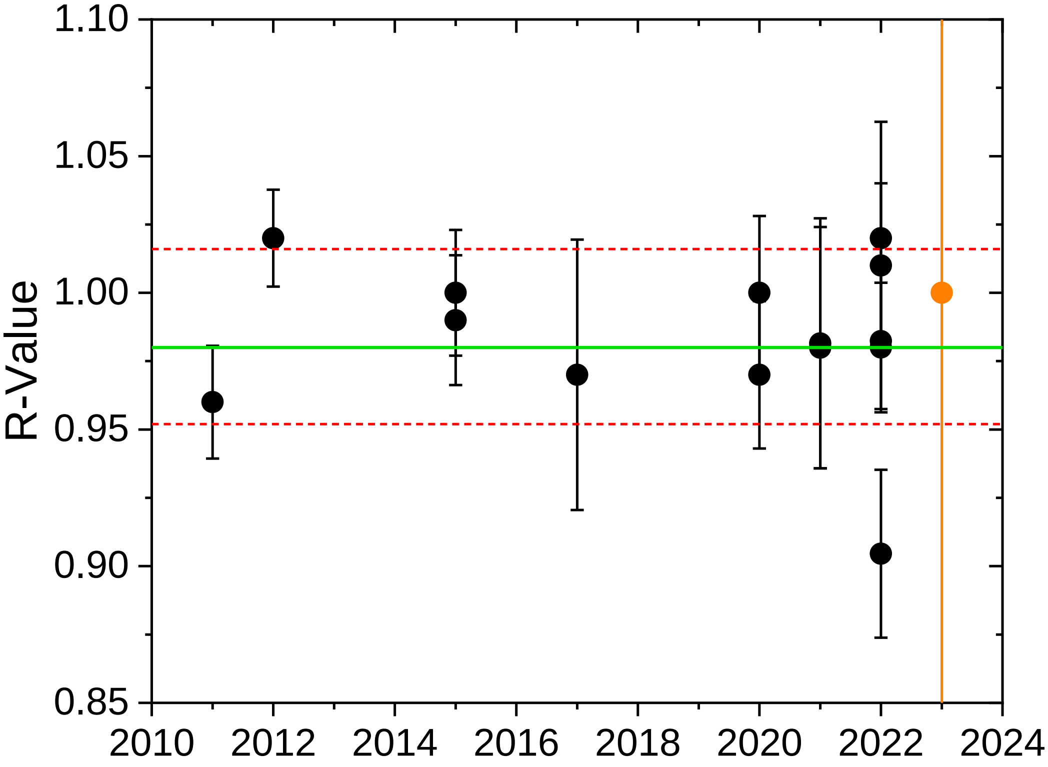

Figure 3-5. R-values of ^{136}Cs from ^{235}U at thermal energies using MITR at MIT. Black symbols show the experimental values, the orange symbols show the ENDF calculated value, the green line shows the average experimental R-value, with the red dashed lines showing the range of 1σ of the average.

The R-values shown in Figure 3-5 show a high degree of agreement, except for a single point in 2022, which brings the average down slightly. R-values are normalized to the thermal fission of ^{235}U, therefore the R-value for a thermal fission should be equal to 1. The small deviation from 1 is an indication of either a need to explore an internal bias or a need to update the fission yield.

The large uncertainty in the ENDF yield lends itself to a need for an updated fission yield, the average of the many fission analysis campaigns fills that need.

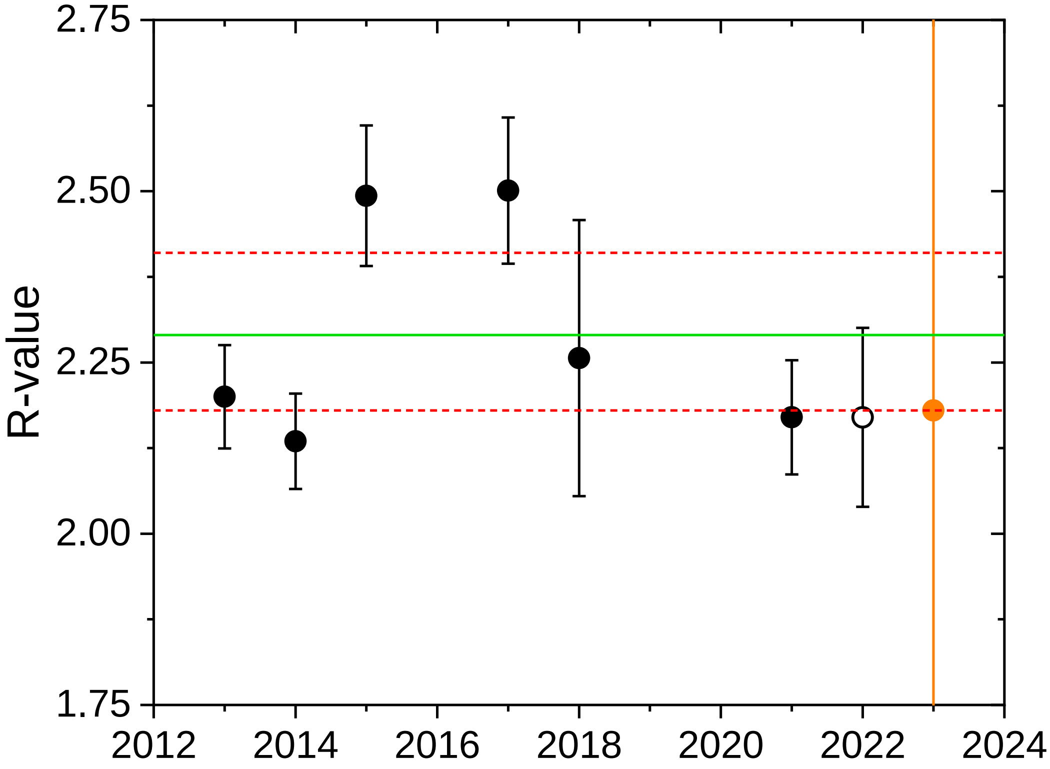

Figure 3-6. R-values of ^{136}Cs from ^{235}U at fission spectrum using the Flattop and Godiva IV critical assemblies. Solid black symbols show the experimental R-values for the Flattop assembly, the open black symbols show the experimental R-values for Godiva IV, the orange symbols show the ENDF calculated value, the green line shows the average experimental R-value, with the red dashed lines showing the range of 1σ of the average.

Figure 3-6 shows the R-values from fission spectrum neutron sources, the critical assemblies Flattop and Godiva IV. The R-values in 2015 and 2017 are high relative to the other fission yields, all others are nearly identical to the ENDF value. However, the average R-value is within 1σ of the ENDF value. The agreement between the Flattop (2013-2021) and Godiva IV (2022) critical assemblies is impressive, particularly because they operate fundamentally in different ways, Flattop is a large assembly that does sustained irradiation, Godiva IV on the other hand is pulsed and provides the neutrons in a burst.

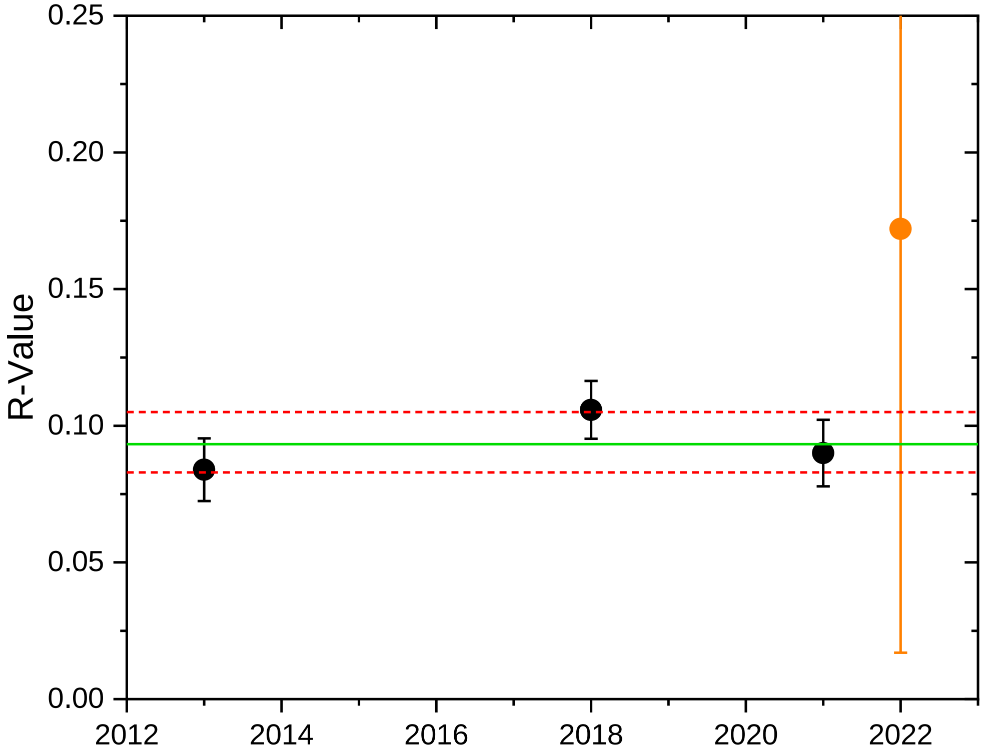

Figure 3-7. R-values of ^{136}Cs from ^{238}U at fission spectrum using the Flattop critical assembly. Black symbols show the experimental values, the orange symbols show the ENDF calculated value, the green line shows the average experimental R-value, with the red dashed lines showing the range of 1σ of the average.

The R-values shown in Figure 3-7 are well below the ENDF R-value, however the extremely low fission yield makes measurements of ^{136}Cs difficult. The measured R-values are in good agreement; however, it was not detected in the campaigns in 2014, 2015, 2017 or the Godiva IV campaign in 2022. This highlights the need for further irradiation campaigns on ^{238}U at fission spectrum energies.

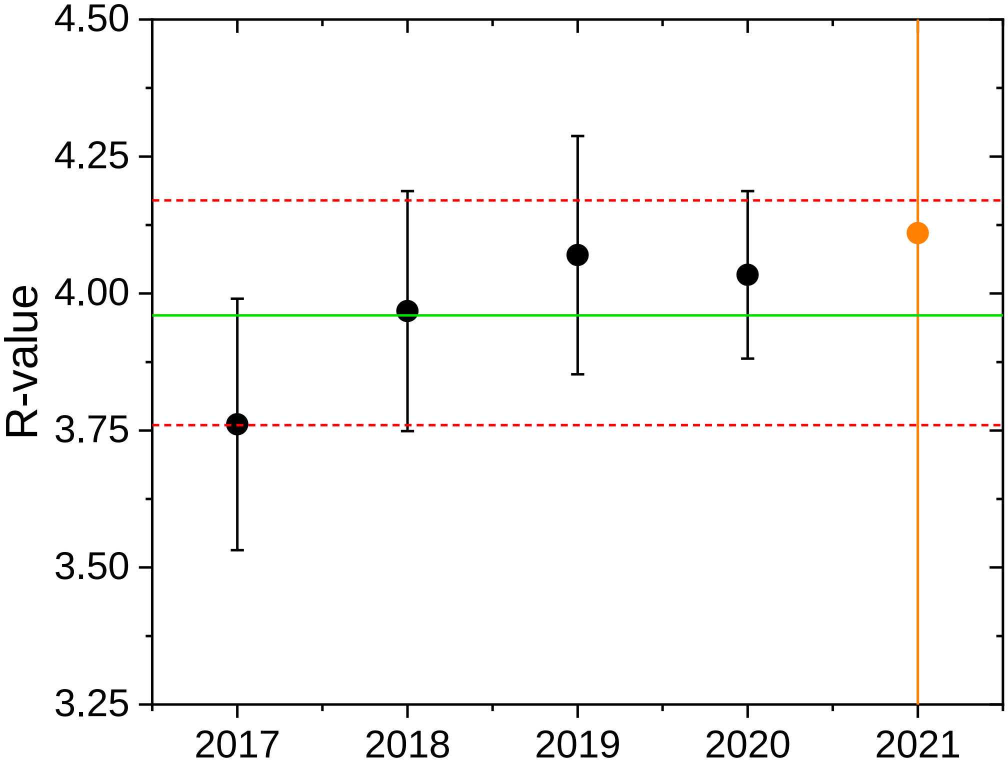

Figure 3-8. R-values of ^{136}Cs from ^{238}U at 14 MeV using a Thermo D711 D-T neutron generator. Black symbols show the experimental values, the orange symbols show the ENDF calculated value, the green line shows the average experimental R-value, with the red dashed lines showing the range of 1σ of the average.

The correlation of the R-values determined in all the irradiations, demonstrates the repeatability and reliability of the processes from the separation to the analysis methods. Averages of the experimental R-values generally compare well to the ENDF R-value, with ^{238}U fission spectrum being the exception, being lower by roughly a factor of 2 with significantly decreased uncertainty. Improving these uncertainties would supply end users with data that provides better diagnostic information, with ^{136}Cs being of some interest.

4.0 Conclusion

Calculations of new CFY for ^{136}Cs using multiple irradiations at several neutron energies and fuel materials. Though the calculated CFY values do not deviate significantly from the literature value there is a significant improvement in the uncertainty relative to the literature values, particularly from ENDF or JENDL databases. Future irradiations will be included in further refinement of these values, with particular interest in the non-thermal and non-^{235}U targets. The thermal ^{235}U ^{136}Cs CFY is $5.74\text{x}10^{-5}$ ± 3.6% should provide the foundation in the future irradiations to provide relative cumulative fission yield calculated using the R-value.

5.0 References

Uhnak, NE; Haney, MM; Pierson, BD; Greenwood, LR; Herman, SM; Arnold, ES; Lawler, B; Risenhuber, M; Irwin, L; Warzecha, E; Trang-Le, T; Byram, D; Liezers, M; Thomas, M; Gajos, N; Springer, K; Spitler, G; Barnett, DS; Friese, JI; Metz, LA; Boswell, M; Dembowski, M; Dry, DE; Gaunt, AJ; Hanson, SK; Hudston, LA; James, MR; Kinman, WS; Lance, CA; Lee, G; Marigiotta, C; May, I; Meininger, D; Miller, JL; Rielly SD; Rendon, RJ; Romero, JR; Smythe, NC; White, JM; Williams, JM; Wren, MS. R-value Measurements Performed on Actinide Targets Irradiated using the GODIVA IV Critical Assembly in FY22. **PNNL-33660** (2022).

Uhnak, NE; Haney, MM; Greenwood, LR; Pierson, BD; Friese, JI; Metz, LA. R-Value Measurements Performed on Uranium Targets Irradiated with Fission Spectrum Neutrons in FY 2021 for F2019 Project. **PNNL-32666** (2022).

Uhnak, NE; Haney, MM; Greenwood, LR; Pierson, BD; Trang-Le, T; Byram, DW; McNamara, BK; Bowen, JM; Munley, WO; Hilton, CD; Friese, JI; Metz, LA. 14 MeV Irradiation and Analysis of a 93% ^{239}Pu Target. **PNNL-33060** (2022).

Gartman, BN; Pierson, BD; Estrada, JH. *PNNL After Action Report for the Fission Products Thermal Calibration Exercise (QA-PTFP-2022-3).* **PNNL-NC-0891** (2022).

Uhnak, NE; Haney, MM; Arrigo, LM; Greenwood, LR; Pierson, BD; Metz, LA; Friese, JI; Boswell, M; Dembowski, M; Dry, DE; Gaunt, AJ; Hanson, SK; Hudston, LA; James, MR; Kinman, WS; Lance, CA; Lee, G; Margiotta, C; May, I; Meininger, D; Miller, JL; Reilly, SD; Rendon, RJ; Romero, JR; Smythe, NC; White, JM; Williams, JM; Wren, MS. FY20 R-value Measurement Results for 14 MeV Neutron Irradiation of DU and HEU Targets. **PNNL-31327** (2021).

Seiner, DR; Gartman, BN; Haney, MM; Pierson, BD; Archambault, B; Estrada, JH; Friese, JI. *PNNL After Action Report for the Fission Products Thermal Calibration (QA-PTFP-2021-02).* **PNNL-X-900-2189** (2021).

Uhnak, NE; Haney, M; Pierson, B; Greenwood, LR; Friese, JI; Metz, LA; Boswell, M; Dembowski, M; Dry, DE; Gaunt, AJ; Hanson, SK; Hudston, LA; James, MR; Kinman, WS; Lance, CA; Lee, G; Margiotta, C; May, I; Meininger, D; Miller, JL; Reilly, SD; Rendon, RJ; Romero, JR; Smythe, NC; White, JM; Williams, JM; Wren, MS. R-value Measurements Performed in FY21 on Uranium Targets Irradiated by Fission Spectrum Neutrons-Short Report. **PNNL-31718** (2021).

Uhnak, NE; Haney, MM; Arrigo, LM; Greenwood, LR; Pierson, BD; Metz, LA; Friese, JI; Boswell, M; Dembowski, M; Dry, DE; Gaunt, AJ; Hanson, SK; Hudston, LA; James, MR; Kinman, WS; Lance, CA; Lee, G; Margiotta, C; May, I; Meininger, D; Miller, JL; Reilly, SD; Rendon, RJ; Romero, JR; Smythe, NC; White, JM; Williams, JM; Wren, MS. FY20 R-value Measurement Results for 14 MeV Neutron Irradiation of DU and HEU Targets. **PNNL-31327** (2020)

Arrigo, LM; Greenwood, LR; Pierson, BD; Metz, LA; Friese, JI; Berger, J; Boggs, M; Boswell, M; Dry, D; Gaunt, A; Hanson, S; Hudston, L; James, M; Kinman, W; Lance, C; Lee, G; Margiotta, C; May, I; Meininger, D; Miller, J; Oldham, W; Reilly, S; Rendon, R;

Romero, J; Smythe, N; Williams, J; Wren, M. FY19 R-value Measurement Results for 14 MeV Neutron Irradiation of DU and HEU. **PNNL-29545** (2019).

Arrigo, LM; Friese, JI; Greenwood, LR; Metz, LA. R-value Measurements performed in FY18 at PNNL under the NCNS F2016 Nuclear Physics Project. **PNNL-28368** (2018).

Arrigo, LM; Friese, JI; Greenwood, LR; Metz, LA. R-value Measurements performed in FY2017 under the NCNS F2016 Nuclear Physics Project. **PNNL-27046** (2017).

Friese, JI; Metz, LA; Arrigo, LM; Estrada, JH. F2012: Comparison of LANL and PNNL Radiochemical Results for HEU and DU Flattop Irradiations. **PNNL-26166** (2016)

Friese, JI; Morley, SM; Finn, EC; Seiner, BN; Snow, L; Arrigo, LM; Beacham, TA; Lucas, DD; Morrison, SS; Smith, SC; Metz, LA; Bowen, J; Gregory, SJ; Beck, CL; Greenwood, LR; Haney, MM; Noyes, KL. R-Value Measurements Performed in FY 2015 under the NCNS F2012 Nuclear Physics Project. **PNNL-25141** (2016).

Friese, JI; Morley, SM; Seiner, BN; Snow, L; Arrigo, LM; Beacham, TA; Lucas, DD; Smith, SC; Metz, LA; Gregory, SJ; Beck, CL; Greenwood, LR; Haney, MM; Noyes, KL. R-Value Measurements Performed in FY 2014 under the NCNS F2012 Nuclear Physics Project. **PNNL-23945** (2015).

Friese, JI; Metz, LA; Seiner, BN; Douglas, M. R-Value Measurements Performed in Fiscal Year 2012 under the NCNS Nuclear Physics Project. **PNNL-23846** (2014).

Metz, LA; Morley, SM; Doll, SR; Friese, JI; Arrigo, LM; Beacham, TA; Lucas, DD; Smith, SC; Seiner, BN; Gregory, SJ; Beck, CL; Greenwood, LR; Haney, MM; Noyes, KL. R-Value Measurements Performed in FY 2013 under the NCNS F2012 Nuclear Physics Project. **PNNL-23191** (2014).

2K v3.4.1 manual, Canberra Industries, Inc. www.canberra.com

England, TR; Rider, BF. “Evaluation and Compilation of Fission Product Yields. ENDF-349, LA-UR-94-3106, Los Alamos National Laboratory (1994), https://t2.lanl.gov/nis/publications/endf349.pdf

N. Soppera, M. Bossant, E. Dupont, "JANIS 4: An Improved Version of the NEA Java-based Nuclear Data

Appendix A – Title

A.1 Literature CFY for ^{136}Cs

Table A.1-5-1. CFY values from the ENDF.V.III.0, JEFF3.3, and JENDL4.0 databases. [Soppera]

Fuel	**Databases**	**Neutron Energy**		
		0.0253 eV	**500 keV**	**14 MeV**
235U	ENDF.V.III.0	5.54x10-5 ± 64%	1.17 x10-4 ± 64%	2.26x10-3 ± 64%
	JEFF3.3	2.91 x10-5 ± 64%	3.40 x10-5 ± 64%	9.87x10-3 ± 64%
	JENDL4.0	5.53 x10-5 ± 64%	1.17 x10-4 ± 64%	2.26x10-3 ± 64%
238U	ENDF.V.III.0	N/A	9.60 x10-6 ± 64%	2.12 x10-4 ± 64%
	JEFF3.3	N/A	6.96 x10-7 ± 29%	2.76 x10-4 ± 25%
	JENDL4.0	N/A	9.60 x10-6 ± 64%	2.13 x10-5 ± 60%
239Pu	ENDF.V.III.0	9.74 x10-4 ± 64%	6.18 x10-4 ± 64%	2.67 x10-3 ± 64%
	JEFF3.3	7.96 x10-4 ± 32%	8.35 x10-4 ± 31%	N/A
	JENDL4.0	9.74 x10-4 ± 64%	1.23 x10-3 ± 64%	7.537 x10-3 ± 60%

A.2 Neutron Source Analysis

Neutron spectrum measurements are determined using a series of metal foils or wires, each metal is highly pure and well characterized nuclear reactions, referred to as fluence monitors. The activated fluence monitors are then analyzed by gamma energy analysis, these results are combined with modeling by MCNP and STAYSL. Many of the irradiations included these fluence monitors, for many of the irradiations the neutron spectrum analysis is included in the figures and tables below. Both the flux and fluence are included below, however regardless of flux or fluence the relative difference between the different neutron energy ranges is the most important. There is no neutron spectral analysis for MITR irradiations, or the Godiva IV irradiation.

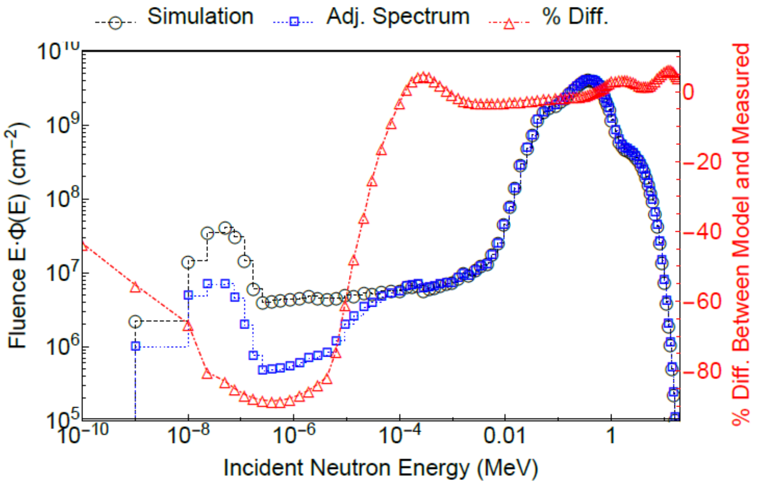

Figure A.2-1.STAYSL analysis of 2013 Flattop irradiation.

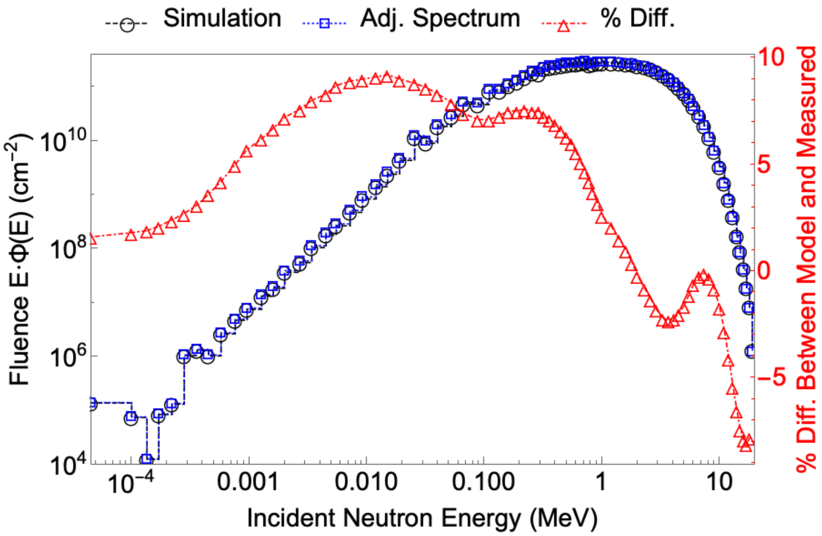

Figure A.2-2. STAYSL analysis of 2014 Flattop

Table A.2-2. STAYSL analysis of 2014 Flattop irradiation.

STAYSL PNNL Results		
ENERGY	**FLUX**	**STDEV %**
<0.5 eV	N/A	
(0.5 eV -100 keV)	3.99E+14	14
>0.1 M eV	3.99E+15	8
>1 MeV	1.84E+15	9
Total	4.21E+15	3

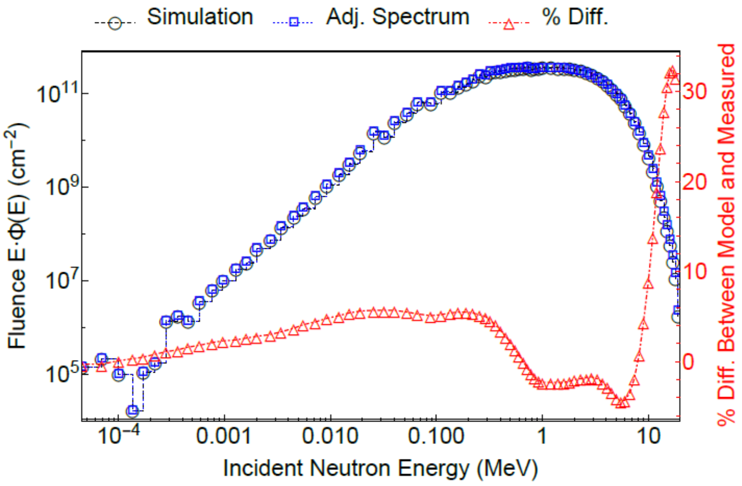

Figure A.2-3. STAYSL analysis of 2015 Flattop

Table A.2-3. STAYSL analysis of 2015 Flattop irradiation.

STAYSL PNNL Results		
ENERGY	**FLUX**	**STDEV %**
<0.5 eV	N/A	
(0.5 eV -100 keV)	2.25E+14	10
>0.1 M eV	4.00E+15	3
>1 MeV	1.86E+15	4
Total	4.22E+15	3

Table A.2-4. STAYSL analysis of 2017 14 MeV neutron irradiation using PNNL D711 DT generator.

STAYSL PNNL Results		
ENERGY	**FLUENCE**	**STDEV %**
<0.50 eV	5.69E+9	464
(0.50 eV–100.0 keV)	3.00E+11	39
>0.1 MeV	8.89E+13	2
>1 MeV	8.75E+13	2

Table A.2-5. STAYSL analysis of 2018 14 MeV neutron irradiation using PNNL D711 DT generator.

STAYSL PNNL Results		
ENERGY	**FLUENCE**	**STDEV %**
<0.50 eV	5.E+10	
(0.50 eV–100.0 keV)	3.10E+11	43
>0.1 MeV	9.74E+13	2
>1 MeV	9.59E+13	2

Table A.2-6. STAYSL analysis of 2019 14 MeV neutron irradiation using PNNL D711 DT generator.

STAYSL PNNL Results		
ENERGY	**FLUX**	**STDEV %**
(0.100 meV–0.550 eV)	5.02E4	464
(0.550 eV–110.0 keV)	2.44E+06	40.3
(110.0 keV–16.5 MeV)	7.27E+08	2.6
(1.0 MeV–16.5 MeV)	7.15E+08	2.6

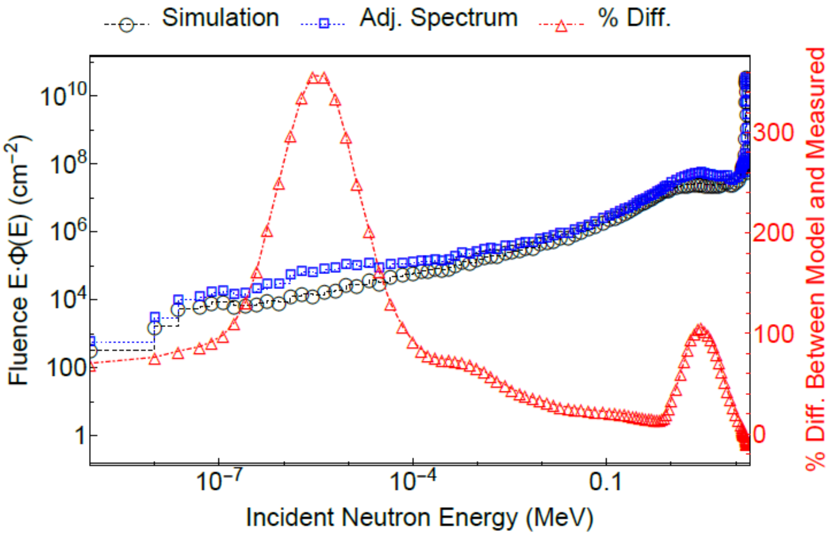

Figure A.2-4. STAYSL analysis of the 2020 14 MeV neutron spectrum from PNNL D711 DT generator.

Table A.2-7. Table of the neutron energy of the PNNL D711 DT generator irradiation in 2020.

STAYSL PNNL Results		
ENERGY	**FLUX**	**STDEV %**
(0.100 meV–0.550 eV)	7.60E+04	166.69
(0.550 eV–110.0 keV)	3.74E+06	25.23
(110.0 keV–16.5 MeV)	1.05E+09	1.28
(1.0 MeV–16.5 MeV)	1.03E+09	1.2

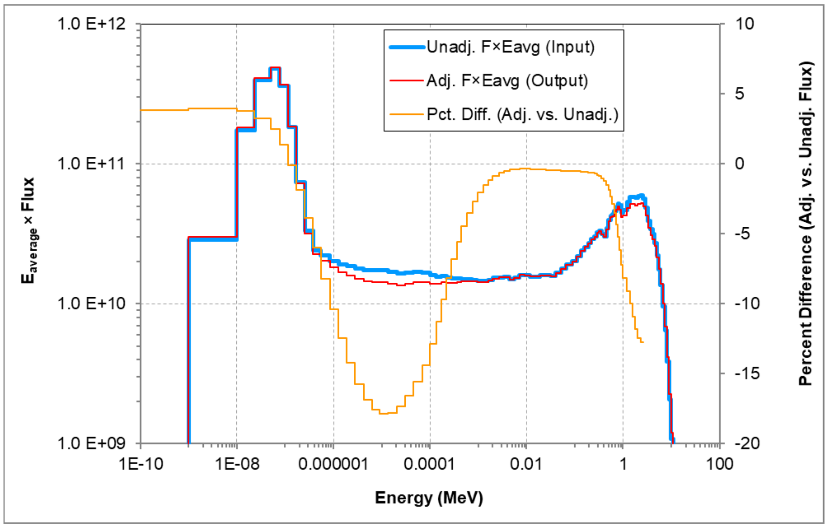

Figure A.2-5. STAYSL analysis of WSU TRIGA reactor at port E9 for the 2022 thermal irradiations.

Table A.2-8. STAYSL analysis of 2022 irradiation at the WSU TRIGA reactor.

STAYSL PNNL Results		
ENERGY	**FLUX**	**STDEV %**
(0.100 meV–0.550 eV)	7.60E+04	166.69
(0.550 eV–110.0 keV)	3.74E+06	25.23
(110.0 keV–16.5 MeV)	1.05E+09	1.28
(1.0 MeV–16.5 MeV)	1.03E+09	1.2

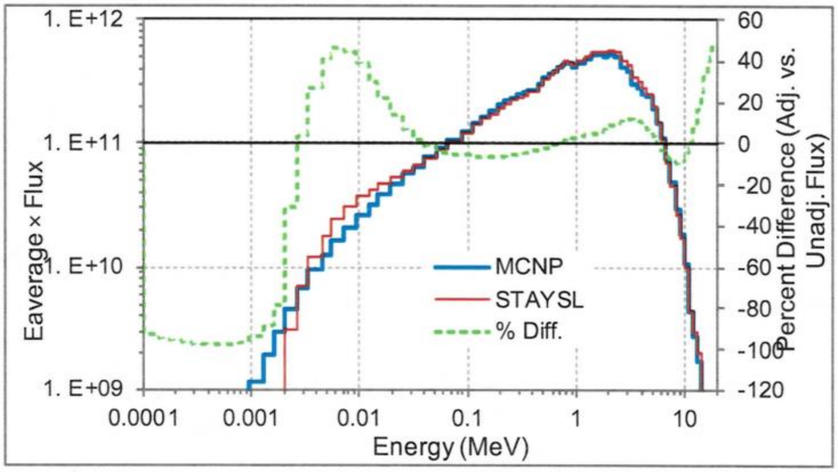

Figure A.2-6. STAYSL analysis of WSU TRIGA reactor for a representative B4C shielded irradiation.

Table A.2-9. STAYSL analysis of the 2014 B4C shielded irradiation in the WSU TRIGA reaction.

STAYSL PNNL Results		
ENERGY	**FLUX**	**STDEV %**
<0.5 eV	N/A	
(0.5 eV -100 keV)	9.65E+15	14
>0.1 M eV	7.23E+16	8
>1 MeV	4.09E+16	9
Total	8.20E+16	7

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