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R-Value Measurements Performed on Actinide Targets Irradiated using the GODIVA IV Critical Assembly in FY22

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Acronyms and Abbreviations

CFY	Cumulative Fission Yield
DU	Depleted uranium
HEU	Highly enriched uranium
GEA	Gamma emission analysis
ICP-MS	Inductively coupled plasma mass spectrometry
ICP-OES	Inductively coupled plasma optical emission spectroscopy
KPA	Kinetic phosphorescence analysis
LANL	Los Alamos National Laboratory
NCERC	National Criticality Experiments Research Center
PNNL	Pacific Northwest National Laboratory
TIMS	Thermal ionization mass spectrometry

1.0 Introduction

The separation and characterization of two irradiated uranium targets, a depleted uranium (DU) and a highly enriched uranium (HEU) target as well as a plutonium (Pu) target, was conducted in April of 2022. The three targets were assembled at Los Alamos National Laboratory (LANL) and irradiated using the GODIVA critical assembly at the National Criticality Experiments Research Center (NCERC). Splits of the dissolved targets were received by Pacific Northwest National Laboratory (PNNL) after which the PNNL and LANL teams chemically separated the solutions using independent separation schemes and analyzed the separated fractions for short lived actinides and fission products. Chemical separations were traced with stable or radioactive tracers to allow for the determination of chemical yields, analyzing using either inductively coupled plasma optical emission spectroscopy (ICP-OES), inductively coupled plasma mass spectrometry (ICP-MS) or gamma emission analysis (GEA) depending on the nature of the tracer. The Pu target solution was traced with stable elements at LANL to follow elemental fractionation during a Pu removal step. Many analytical techniques were used by PNNL including kinetic phosphorescence analysis (KPA), ICP-OES, ICP-MS, GEA, and thermal ionization mass spectrometry (TIMS) depending on the analyte's need.

Comparisons were made between PNNL, LANL and literature values for the R-values, number of fissions, activation products, as well as the atoms detected. There was good agreement between the two laboratories for the bulk of analytes. This experiment represents one of the first modern examinations of fission yields for Pu fission induced by fission neutrons. Included in these comparisons were the short-lived actinides ^{237}U, ^{239}Np, the fission products ^{89}Sr, ^{91}Y, $^{95/97}$Zr, ^{99}Mo, ^{111}Ag, $^{115/115m}$Cd, $^{136/137}$Cs, ^{140}Ba, $^{141/143/144}$Ce, ^{147}Nd, ^{153}Sm, ^{156}Eu, and ^{161}Tb, providing both total atoms as well as the R-values . The data presented in this report represents the first irradiation using the GODIVA critical assembly for this work. .

2.0 PNNL Sample Splitting

The targets used were assembled by LANL, using actinide metal foils. Details on the foil mass are included in **Table 1**. Each actinide target was cleaned of oxide, individually wrapped in thin high purity aluminum foil, and further sealed in aluminum using a Bronson Ultraweld® 20 ultrasonic welder. Details of target production are included in the LANL report LA_CP-22-20725. The targets were shipped to NCERC to be irradiated on the GODIVA IV critical assembly. Each of the targets were co-located during the irradiation to ensure that the neutron environment was as similar as possible with minimal attenuation. The critical assembly was operated for a total of 52 minutes, with two irradiation pulses of 31 and 21 minutes. There was a shut down due a transient temperature ramp that exceeded the safety basis, thus requiring a short shutdown and restart.

Each of the targets were disassembled the HEU and DU targets were removed from the outer Al capsule and dissolved. The transient temperature ramp caused an issue with the Pu target, requiring the dissolution of the outer Al capsule. During the irradiation the Pu heated to the point that it welded to the Al inner wrapping as well as the Al capsule. This welding caused an increase in the amount of dissolved Al nearly two orders of magnitude. Stable elements of fission products were added including Ag, Ba, Cd, Ce, Cs, Eu, Mo, Nd, Sm, Sr, Tb, Te, Y, and Zr at 100 µg.

Due to shipment requirements, the Pu was removed from the target prior to the shipment to PNNL. This was accomplished using well established ion exchange of Pu in high nitric acid

concentration. The dissolved target solution, also referred to as the A solution was split into aliquots for each lab. Table 2 contains the masses of the A solution splits, the mass of the actinide in each of the target A solutions, as well as the fissions relative to thermal ^{99}Mo. Due to the number of fissions in these samples, it was determined that it would be better if the samples were split into thirds to provide enough activity for adequate statistics on the various fission products in each PNNL A solution or separated fraction. The chemistry replicates moved on to the separations that will be discussed in a larger report, while the whole A Solution GEA was rotated among several detectors for analysis.

Table 1. Target and irradiation conditions, including irradiation time, target mass, date of irradiation, and PNNL receipt date.

	Total Irradiation Time (min)*	Mass (g)	Irradiation Date	PNNL Receipt Date
Pu (Z11135)	**52**	**0.135**	**4/26/2022**	**5/6/2022**
HEU (Z11136)		**0.251**		**5/3/2022**
DU (Z11137)		**0.624**		
* GODIVA experienced an issue so total irradiation time was two combined pulses of 31 and 21 mins.				

Table 2. Mass of PNNL A solution splits

		Mass of A solution (g)	Percent of A solution	Mass of U* (mg)	Fissions Thermal ^{99}Mo*
DU (Z11137)	Whole Solution GEA	7.5570	33.4%	102.2	5.63×10^{10}
	Chemistry Rep 1	7.5545	33.3%	102.2	5.62×10^{10}
	Chemistry Rep 2	7.5382	33.3%	101.9	5.61×10^{10}
		Mass of A solution (g)	**Percent of A solution**	**Mass of U* (mg)**	**Fissions Thermal ^{99}Mo***
HEU (Z11136)	Whole Solution GEA	7.4845	33.5%	41.2	1.53×10^{11}
	Chemistry Rep 1	7.4252	33.2%	40.8	1.51×10^{11}
	Chemistry Rep 2	7.4202	33.2%	40.8	1.51×10^{11}
		Mass of A solution (g)	**Percent of A solution**	**Mass of Pu† (µg)**	**Fissions Thermal ^{99}Mo***
Pu (Z11135)	Whole Solution GEA	6.8212	29.9%	142.3	1.04×10^{11}
	Chemistry Rep 1	7.9323	34.8%	165.5	1.21×10^{11}
	Chemistry Rep 2	7.9377	34.8%	165.6	1.21×10^{11}

*Based on PNNL GEA analysis of A solution in f/g A solution (f/g target are presented below)
†Based on ICP-MS of A solution after LANL Pu removal

There was a degree of fractionation that was found in the analysis of the Pu (Z11135) A solution by ICP-OES. Recovery in both mass and percent are included in **Table 3**. The yields were used to correct for any loss in isotope activity in the A solution or separated fraction. Tellurium analysis by ICP-OES suffers from poorly resolved spectra, the diluted mass recovered was on the lower limits of the detection limit. Due to the high recovery, no yield correction was applied for the Te analysis.

Table 3. Degree of fractionation of Pu (Z11135) stable tracers.

Element	ug added	ug recovered	1σ%	Yield (%)
Ag	99.8	88.0	3.7%	88
Ba	100.6	89.8	4.1%	89

Cd	99.2	89.8	4.7%	91
Ce	100.5	55.0	3.0%	55
Cs	99.1	94.4	0.5%	95
Eu	100.5	88.0	3.3%	88
Mo	100.6	93.5	2.3%	93
Nd	101.3	69.7	2.6%	69
Sm	99.7	84.3	2.8%	85
Sr	99.3	91.6	3.1%	92
Tb	99.8	88.0	2.6%	88
Te	99.7	106.3	2.2%	107
Y	99.3	89.8	2.9%	90
Zr	100.0	88.0	2.3%	88

3.0 R-value Calculation

The analytical results were used to calculate the R-value for each fission product; the method for calculating the R-value is shown in **Equation 1**. PNNL has a running historical r-value (r_{hist}) for each isotope based on the results from the last five thermal calibration (t-cal) exercises where available. A t-cal exercise involves the thermal irradiation of ^{235}U followed by separation and radiometric analysis. The historical r-value replaces the ENDF/B-VIII.0 Cumulative Fission Yield (CFY) in the R-value calculation. The historical r-values used in the R-value calculation are shown in **Table 4**. The applicable ENDF/B-VIII.0 CFY values are/were used for the ^{91}Sr, ^{93}Y, ^{112}Ag, and ^{156}Sm isotopes which are not measured in t-cal solutions and do not have r_{hist} values.

Equation 1: R-value calculation for measurements at PNNL

$$R = \frac{\left(\frac{N_X}{N_{Mo99}}\right)_{Measured}}{\left(\frac{N_x}{N_{Mo99}}\right)_{U235\ Thermal}} = \frac{\left(\frac{N_X}{N_{Mo99}}\right)_{Measured}}{\left(\frac{CFY_x}{CFY_{Mo99}}\right)_{U235\ Thermal}} = \frac{\left(\frac{N_X}{N_{Mo99}}\right)_{Measured}}{r_{hist}}$$

N_X – atoms of isotope X per gram of A solution
N_{Mo99} – atoms of ^{99}Mo per gram of A solution
CFY_X – cumulative fission yield for isotope X for ^{235}U thermal fission
CFY_{Mo99} – cumulative fission yield for ^{99}Mo for ^{235}U thermal fission
r_{hist} – historical r-value as determined in Equation 2

Equation 2: Historical r-value

$$r_{hist} = \left(\frac{N_X}{N_{Mo99}}\right)_{t-cal}$$

N_X – atoms of isotope X per gram of A solution in a t-cal sample
N_{Mo99} – atoms of ^{99}Mo per gram of A solution in a t-cal sample

t-cal – thermal calibration exercise sample

Table 4. PNNL historical r-values

Isotope	r_{hist}	Isotope	r_{hist}
^{89}Sr	0.793	^{136}Cs	$9.67x10^{-4}$
^{91}Sr	N/A	^{137}Cs	1.05
^{91}Y	0.939	^{140}Ba	1.05
^{93}Y	N/A	^{141}Ce	0.971
^{95}Zr	1.09	^{143}Ce	0.994
^{97}Zr	1.05	^{144}Ce	0.910
^{103}Ru	0.504	^{147}Nd	0.365
^{111}Ag	$2.80x10^{-3}$	^{153}Sm	$2.22x10^{-2}$
^{112}Ag	N/A	^{156}Sm	N/A
^{115}Cd*	$2.21x10^{-3}$	^{155}Eu	$5.37x10^{-3}$
^{115m}Cd	$7.90x10^{-5}$	^{156}Eu	$2.44x10^{-3}$
^{132}Te	0.719	^{161}Tb	$1.29x10^{-5}$

*Recent work on ^{115}Cd, for another project, revealed errors in software that have been fixed

4.0 Results from A Solution and Separated Fraction Analysis

Due to the mass differences between targets, different separation schemes were required for the Pu relative to the HEU and DU targets, Z11135, Z11136 and Z11137 respectively. The scheme used for the HEU, and DU targets is identical to that described in PNNL-31327. For the Pu target, the separation scheme is shown below in

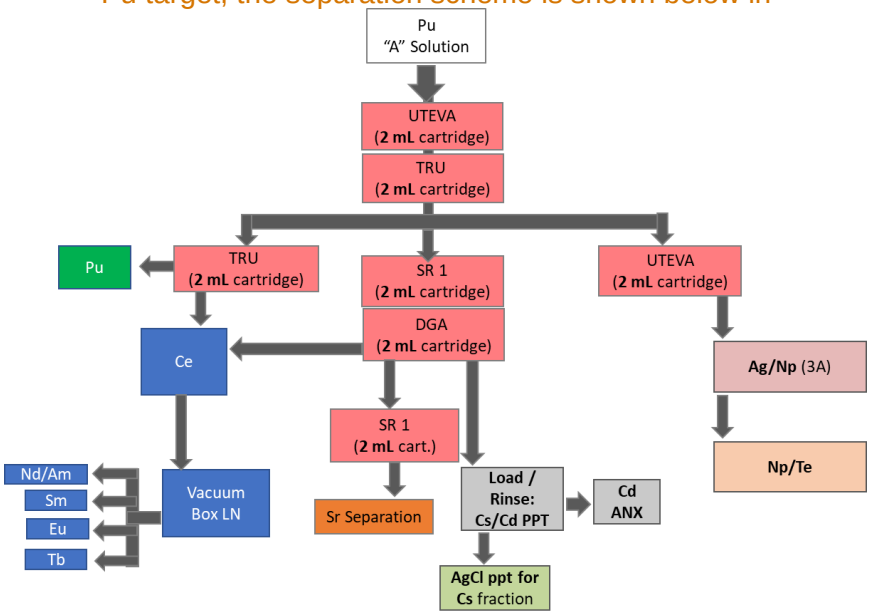

Figure 1. The stable tracers used in the initial addition prior to the Pu removal were used as the tracers for the full separation process, with the addition of radiotracers of Cd, Cs, Eu, Ag, and Np to establish chemical yields. The results for Pu (Z11135) are shown in The results for each

of the three targets are presented in Table 5, Table 6, and Table 7 for the individual targets including the fission product yields in atoms/g of target, the R-value, chemical yield, fission/g target and analysis method. A calculation of the spectral index is included for comparison between the two critical assemblies used in these campaigns (Flattop and Godiva), shown in Figure 3.

For the Pu target (Z11135) all R-values are yield corrected for the ^{99Mo} yield after the Pu separation, as such the determined values are highly dependent on this correction. A small deviation in the ^{99Mo} analysis would have far reaching consequences to the analysis of the other elements. Because the solution was only stable traced with stable peak yield fission products there is the potential that there was deviation between the measured atoms of a given fission product and what was originally in solution prior to the Pu separation step. By in large the anion exchange process used for the Pu removal is highly selective for Pu ignoring the bulk of fission products and actinides. The R-values were in good agreement with the ENDF database calculated R-values, with a notable exception ^{136Cs,} which is a likely indication of a need for updated nuclear data. Unlike ^{136Cs} yield issues with ^{238U,} the yield from ^{239Pu} does not suffer from the high uncertainty but may also be an issue.

Table 5, including the atoms/g of target, R-values both measured and ENDF determined, chemical yields, and analytical method used.

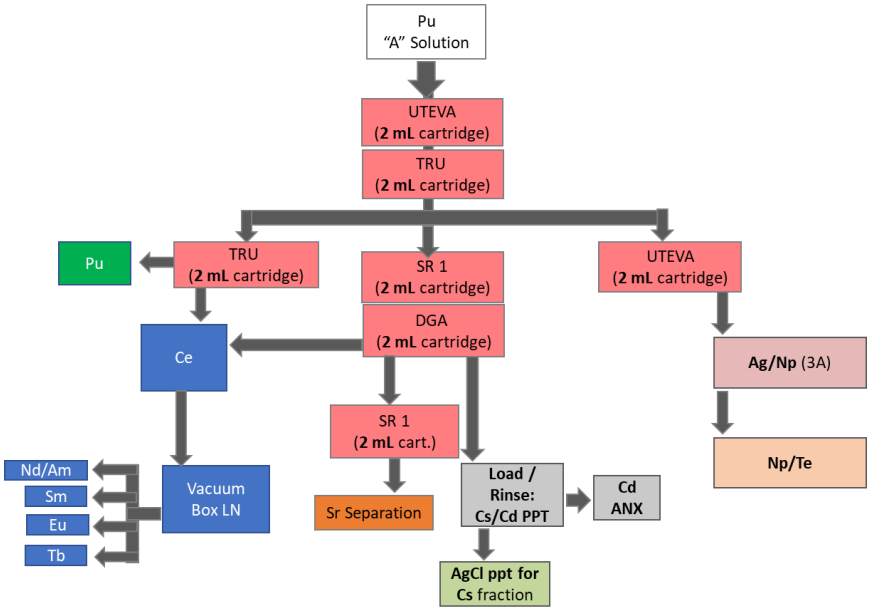

Figure 1. Separation scheme used for the Pu target solution Z11135.

There was evidence of disequilibrium between the stable tracer and the fission products from the dissolution, tracer addition and Pu separation process for the Z11135. Cerium is the most striking example of this disequilibrium, as the recovery of the isotopes of ^{141}Ce, used as an internal radiotracer, and the stable Ce tracer recoveries were different.

4.1 Separation Results

This section will only focus on separations whose results were outside of expectation. This is intended as a note for future efforts as well as a highlight of the resilience of the chemistry and chemistry team. The separation of the fission product lanthanides proceeded using a vacuum box method, favoring the expedience of the separation. Both Z11136 and Z11137 proceeded without complications, outside of issues with Ce oxidation steps, therefore only Z11135 is going to be discussed. The Ce oxidation step issues have been noted in the past most recently in PNNL-32666. An underlying issue in the preparation of the oxidant has been identified and will be rectified in future campaigns.

Aluminum is included in many of these R-value campaigns, being used as the capture layer for fission products during the irradiation process. This campaign however included a significantly higher quantity of dissolved Al, due to the welding of the Al foil and capsule to the Pu target material for Z11135. Aluminum presents a unique challenge for lanthanide separations due to a similarity in the chemistry between Al and the Ln series, i.e., +3 charge. Shown below are percent recoveries for each of the fission product lanthanides in their respective elution fraction. There is a significant difference from what is expected.

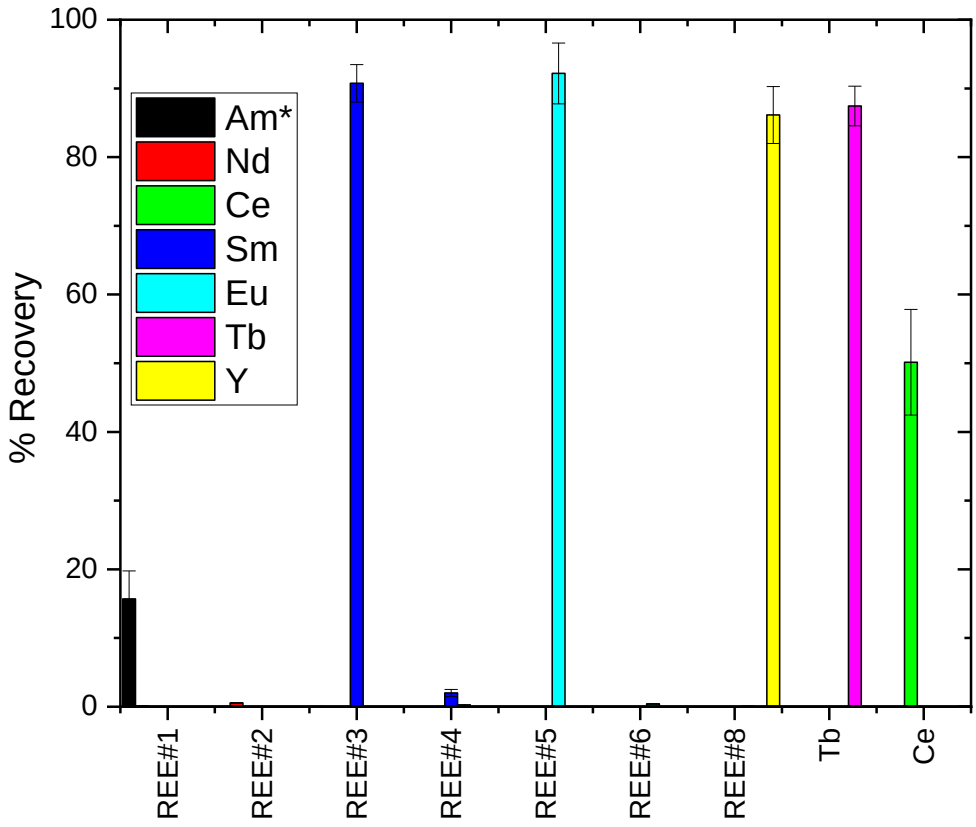

Figure 2. Recovery of rare earth elements from LN separation procedure.

The results shown in Figure 2 show a slight alteration from the previously reported results from FY21 in PNNL-32666. The data from ^{140}La was not reported as it was not stable traced, therefore it was not followed through the Pu removal step . In PNNL-32666, Nd was effectively all eluted in REE#2, a result that is shared in both Z11136 and Z11137 from this campaign. Based on the distribution of Am compared to Nd, Am is expected to elute in the same fraction as Nd in fraction REE#2. The early lanthanides (La, Nd) and Am are more favorably held on to the TRU resin step prior to the LN separation method because of the Al concentration, this trend had been seen in literature reports. (Horowitz 1993) In future campaign large Al concentrations will need to be mitigated, either through alternative target containment or through Al removal chemistry to ensure that the separation chemistries work as designed. Though this does cause issues with the separation of several the lanthanide elements, it does not have a deleterious effect on the reported results for the lanthanide fission products that require separation for better detection such as ^{153}Sm and ^{161}Tb.

4.2 Results

The results for each of the three targets are presented in Table 5, Table 6, and Table 7 for the individual targets including the fission product yields in atoms/g of target, the R-value, chemical yield, fission/g target and analysis method. A calculation of the spectral index is included for comparison between the two critical assemblies used in these campaigns (Flattop and Godiva), shown in Figure 3.

For the Pu target (Z11135) all R-values are yield corrected for the ^{99}Mo yield after the Pu separation, as such the determined values are highly dependent on this correction. A small deviation in the ^{99}Mo analysis would have far reaching consequences to the analysis of the other elements. Because the solution was only stable traced with stable peak yield fission products there is the potential that there was deviation between the measured atoms of a given fission product and what was originally in solution prior to the Pu separation step. By in large the anion exchange process used for the Pu removal is highly selective for Pu ignoring the bulk of fission products and actinides. The R-values were in good agreement with the ENDF database calculated R-values, with a notable exception ^{136}Cs, which is a likely indication of a need for updated nuclear data. Unlike ^{136}Cs yield issues with ^{238}U, the yield from ^{239}Pu does not suffer from the high uncertainty but may also be an issue.

Table 5. Results for analysis of a Pu (Z11135) A solution and separated fractions from the FY22 fission spectrum irradiation using GODIVA IV (R ± 1σ%) compared to ENDF values. Results include atoms/g, R values, ENDF R, Chemical yields, and analysis methods. Values in italics are the atoms/g for the mass of the initial target prepared by LANL.

Isotope	Atoms/g A (*Atoms/g target*)	R Pu	R ENDF – ^{239}Pu "Fission" Spectrum	Chemical Yield (%)	Method
^{89}Sr	9.60x10^{10} ± 3.6%	0.382 ± 5.6%	0.357 ± 4.1%	46.4%	Separated Fraction (TIMS)
	9.30x10^{10} ± 5.0%	0.387 ± 5.6%			Separated Fraction (LSC)
^{91}Y†	N/A	N/A	0.423 ± 90.5%	N/A	A Solution (GEA)
	1.04x10^{10}	0.421		78.8%	Separated Fraction(MS)
^{95}Zr	2.43x10^{11} ± 2.0%	0.704 ± 3.2%	0.718 ± 2.2%	N/A	A Solution (GEA)
^{99}Mo	2.75x10^{11} ± 2.5%	1.00 ± 2.8%	1.00 ± 2.2%	N/A	A Solution (GEA)
^{103}Ru	1.08x10^{11} ± 2.0%	0.678 ± 3.2%	2.21 ± 4.7%	N/A	A Solution (GEA)
^{111}Ag	2.04x10^{10} ± 11.1%	23.0 ± 11.4%	20.0 ± 4.7%	N/A	A Solution (GEA)
	2.09x10^{10} ± 2.0%	23.6 ± 3.2%		85.5%	Separated Fraction
^{115}Cd	1.05x10^{10} ± 9.4%	15.0 ± 9.7%*	6.53 ± 7.4%	N/A	A Solution (GEA)
	3.43x10^{9} ± 3.6%	5.71 ± 2.7%		84.6%	Separated Fraction (GEA)
^{115m}Cd	1.98x10^{8} ± 51.7%	71.1 ± 64.7%	4.09 ± 6.8%	84.6%	Separated Fraction (GEA)
^{132}Te	2.21x10^{11} ± 2.7%	0.971 ± 3.7%	1.17 ± 2.9%	N/A	A Solution (GEA)
^{136}Cs	7.26x10^{9} ± 2.0%	23.7 ± 3.2%	10.95 ± 90.5	N/A	A Solution (GEA)
	7.14x10^{9} ± 3.3%	23.5 ± 4.1%		99.8%	Separated Fraction (GEA)
^{137}Cs	7.38x10^{11} ± 3.3%*	2.21 ± 4.1%	1.04 ± 1.8%	N/A	A Solution (GEA)

Isotope	Atoms/g A (*Atoms/g target*)	R Pu	R ENDF – ^{239}Pu "Fission" Spectrum	Chemical Yield (%)	Method
	$3.76x10^{11}$ ± 4.6%	1.14 ± 5.2%		99.8%	Separated Fraction (GEA)
^{140}Ba	$2.81x10^{11}$ ± 2.0%	0.844 ± 3.2%	0.836 ± 2.0%	N/A	A Solution (GEA)
^{141}Ce[a]	$9.85x10^{10}$ ± 4.1%	0.320 ± 4.7%	0.862 ± 3.4%	N/A	A Solution (GEA)
^{143}Ce[a]	$7.83x10^{10}$ ± 9.6%	0.248 ± 9.9%	0.713 ± 2.2%	N/A	A Solution (GEA)
^{144}Ce[a]	$4.24x10^{10}$ ± 29.1%	0.147 ± 29.1%	0.658 ± 1.9%	N/A	A Solution (GEA)
	$1.42x10^{11}$ ± 6.8%	0.492± 6.8%		50.1%[a]	Separated Fraction (GEA)
^{147}Nd	$1.22x10^{11}$ ± 2.0%	1.05 ± 3.2%	0.877 ± 2.2%	N/A	A Solution (GEA)
^{153}Sm	$2.26x10^{10}$ ± 3.4%	3.23 ± 6.9%	2.63 ± 9.1%	90.7%	Separated Fraction (GEA)
^{155}Eu	$1.03x10^{10}$ ± 41.3%	6.15 ± 36.3%	6.36 ± 23.4%	98.7%	Separated Fraction (GEA)
	$1.03x10^{10}$ ± 35.5%	6.10 ± 35.6%		92.2%	Separated Fraction (OES)
^{156}Eu	$9.22x10^{9}$ ± 4.7%	12.0 ± 5.3%	10.2 ± 7.4%	N/A	A Solution (GEA)
	$9.74x10^{9}$ ± 7.9%	12.6 ± 8.3%		98.7%	Separated Fraction (GEA)
	$9.75x10^{9}$ ± 5.2%	12.7 ± 5.8%		92.2%	Separated Fraction (OES)
^{161}Tb	$3.50x10^{8}$ ± 4.5%	85.5 ± 5.2%	99.2 ± 5.9%	87.4%	Separated Fraction (GEA)
Isotope	**Atoms/g A (*Atoms/g target*)**		**Chemical Yield (%)**		**Method**
^{235}U	$1.76x10^{16}$ ± 57.1% *($5.99x10^{18}$ ± 57.1%)*		N/A		A Solution (GEA of ^{235}U)
^{237}U	$4.28x10^{7}$ ± 37.5% *($1.46x10^{10}$ ± 37.5%)*		N/A		A Solution (GEA of ^{235}U)
Np[b]	N/A[b]		74.8%		Separated Fraction (GEA)
Total Fissions	$5.19x10^{12}$ ± 2.83%				A Solution (GEA)

[a] Disequilibrium of fission product and stable tracer

[b] Traced with ^{237}Np

* Gamma spectral interferences

† Result of a single measurement

The HEU (Z11136) and DU (Z11137) target's chemical yields for isotopes of Cd, Cs, Eu, Ag and Np were established using radiotracers analyzed using GEA. Chemical yielding for Sr, Y, Sm, Tb, and Eu were conducted using stable tracer analyzed by ICP-OES of the separated fractions after GEA. Confirmation of the Y chemical yield was conducted using ICP-MS. Uranium analysis of both the A solution and the separated fraction were conducted to obtain yields using both GEA and KPA, while isotopic information was established through GEA analysis. Chemical yields were equal or better than previous irradiations, a result of improvements in chemistry or analysis methods. Results from the HEU (Z11136) and DU (Z11137) are presented in Table 6 and Table 7 respectively. As a note the uncertainty for R-values are higher than reported in the past, this was due to a transient increase in the background of the measurement's laboratory. This is particularly important considering the uncertainty stemming directly from the ^{99}Mo atoms,

all R-values are determined relative to the ^{99}Mo data. This was a facility issue that has been rectified and will not happen in future campaigns. It did not affect the Pu data, as the issue had been alleviated between the initial analysis of the A solution of the HEU and DU targets and the Pu target A solution.

Table 6. Results for the analysis of an HEU (Z11136) A solution, from the FY22 fission spectrum irradiation (R ± 1σ%) compared to ENDF values. Results include atoms/g, R values, ENDF R, Chemical yields, and analysis methods. Values in italics are the atoms/g for the mass of the initial target prepared by LANL.

Isotope	Atoms/g A (*Atoms/g target*)	R HEU	R ENDF - ^{235}U Fission Spectrum	Chemical Yield (%)	Method
^{89}Sr	1.80x10^{11} ± 5.1%	1.03 ± 5.1%	0.950 ± 2.62%	48.6%	Separated Fraction (TIMS)
	1.93x10^{11} ± 4.1%	1.10 ± 6.1%			Separated Fraction (LSC)
^{91}Y	2.17x10^{11} ± 16.9%	0.960 ± 17.4%	1.01 ± 90.5%	N/A	A Solution (GEA)
	1.91x10^{11} ± 6.0%	0.870 ± 6.3%		90.3%	Separated Fraction(OES)
	1.85x10^{11} ± 4.6%	0.897 ± 7.4%		93.1%	Separated Fraction(MS)
^{95}Zr	2.33x10^{11} ± 2.0%	0.943 ± 4.7%	1.02 ± 2.62%	N/A	A Solution (GEA)
^{97}Zr	2.35x10^{11} ± 8.1%	0.988 ± 9.1%	1.03 ± 3.14%	N/A	A Solution (GEA)
^{99}Mo	2.26x10^{11} ± 4.2%	N/A	N/A	N/A	A Solution (GEA)
^{103}Ru	1.19x10^{11} ± 2.0%	1.05 ± 4.7%	1.10 ± 2.80%	N/A	A Solution (GEA)
^{111}Ag	1.66x10^{9} ± 5.1%	2.62 ± 7.0%	2.51 ± 5.27%	89.7%	Separated Fraction (GEA)
^{115}Cd	7.22x10^{8} ± 11.5%	1.43 ± 12.3%	2.76 ± 6.91%	N/A	A Solution (GEA)
	1.31x10^{9} ± 2.2%	2.51 ± 5.3%		88.5%	Separated Fraction (GEA)
^{115m}Cd	N/A	N/A	2.86 ± 23.85%‡	88.5%	Separated Fraction (GEA)
^{132}Te	1.70x10^{11} ± 2.7%	1.04 ± 5.0%	1.12 ± 3.14%	N/A	A Solution (GEA)
^{136}Cs	4.31x10^{8} ± 7.3%	1.97 ± 8.4%	2.18 ± 90.5%	N/A	A Solution (GEA)
	4.75x10^{8} ± 3.3%	2.17 ± 6.0%		94.6%	Separated Fraction
^{137}Cs	2.36x10^{11} ± 6.0%	0.991 ± 7.4%	1.03 ± 2.10%	N/A	A Solution (GEA)
	2.31x10^{11} ± 3.0%	0.954 ± 5.5%		94.6%	Separated Fraction (GEA)
^{140}Ba	2.16x10^{11} ± 2.0%	0.910 ± 4.7%	0.989 ± 2.43%	N/A	A Solution (GEA)
^{141}Ce	2.07x10^{11} ± 2.0%	0.941 ± 4.7%	1.05 ± 3.57%	N/A	A Solution (GEA)
^{143}Ce	1.95x10^{11} ± 2.0%	0.869 ± 4.7%	0.989 ± 2.80%	N/A	A Solution (GEA)
^{144}Ce	1.75x10^{11} ± 4.3%	0.850 ± 6.0%	0.985 ± 2.52%	N/A	A Solution (GEA)
	1.81x10^{11} ± 3.6%	0.880 ± 6.0%		92.0%	Separated Fraction
^{147}Nd	7.74x10^{10} ± 2.3%	0.936 ± 4.8%	1.14 ± 2.80%	N/A	A Solution (GEA)
^{153}Sm	5.93x10^{9} ± 4.1%	1.19 ± 7.2%	1.27 ± 6.47%	70.7%	Separated Fraction
^{156}Eu	N/A	N/A	1.40 ± 4.89%	N/A	A Solution (GEA)

Isotope	Atoms/g A (*Atoms/g target*)	R HEU	R ENDF - ^{235}U Fission Spectrum	Chemical Yield (%)	Method
	8.61x10^{8} ± 9.5%	1.57 ± 10.8%		95.5%	Separated Fraction (GEA)
	9.12x10^{8} ± 9.9%	1.66 ± 10.7%		90.2%	Separated Fraction (OES)
^{161}Tb	1.25x10^{7} ± 9.0%	4.28 ± 10.0%	4.01 ± 7.48%	92.7%	Separated Fraction
Isotope	**Atoms/g A (*Atoms/g target*)**		**Chemical Yield (%)**		**Method**
^{237}U	3.19x10^{9} ± 20.3%		N/A		A Solution (GEA)
^{237}U	2.69x10^{9} ± 2.8%		82.0%		Separated Fraction (GEA)
^{239}Np	2.21x10^{10} ± 15.1%		N/A		A Solution (GEA)
^{239}Np	1.97x10^{10} ± 3.2%		72.3%		Separated Fraction (GEA)
Fissions/g target	3.71x10^{12} ± 4.47%				A Solution (GEA)
* Chemical yielding issues present					
‡ Likely requires updates to nuclear data					

Table 7. Results for the analysis of a DU (Z11137) A solution, from the FY22 fission spectrum irradiation (R ± 1σ%) compared to ENDF values. Results include atoms/g, R values, ENDF R, Chemical yields, and analysis methods. Values in italics are the atoms/g for the mass of the initial target prepared by LANL.

Isotope	Atoms/g A (*Atoms/g target*)	R DU	R ENDF - ^{238}U Fission Spectrum	Chemical Yield (%)	Method
^{89}Sr	1.72x10^{10} ± 3.9%	0.645 ± 5.8%	0.578 ± 2.62%	47.7%	Separated Fraction (TIMS)
	1.63x10^{10} ± 4.0%	0.620 ± 5.9%			Separated Fraction (LSC)
^{91}Sr	N/A	N/A	0.686± 2.99%	N/A	N/A
^{91}Y	1.77x10^{10} ± 22.3%	0.525 ± 22.7	0.686 ± 90.5%	N/A	A Solution (GEA)
	1.89x10^{10} ± 8.3%	0.587 ± 8.5%		91.2%	Separated Fraction(OES)*
	1.85x10^{10} ± 7.4%	0.587 ± 8.5%		92.7%	Separated Fraction(MS)*
^{95}Zr	2.74x10^{10} ± 2.0%	0.748 ± 4.7%	0.783 ± 2.80%	N/A	A Solution (GEA)
^{97}Zr	3.15x10^{10} ± 2.0%	0.892 ± 6.3%	0.921 ± 3.14%	N/A	A Solution (GEA)
^{99}Mo	3.36x10^{10} ± 4.3%	N/A	N/A	N/A	A Solution (GEA)
^{103}Ru	3.24x10^{10} ± 2.7%	1.91 ± 5.0%	2.05 ± 2.80%	N/A	A Solution (GEA)

Isotope	Atoms/g A (*Atoms/g target*)	R DU	R ENDF - ^{238}U Fission Spectrum	Chemical Yield (%)	Method
^{111}Ag	3.98x10^{8} ± 15%	4.13 ± 15.7%	4.05 ± 4.89%	81.8%	Separated Fraction (GEA)
^{115}Cd	1.65x10^{8} ± 11.2%	2.22 ± 12.0%	2.95 ± 6.63%	N/A	A Solution (GEA)
	1.86x10^{8} ± 3.5%	2.51 ± 6.0%		88.8%	Separated Fraction (GEA)
^{132}Te	3.28x10^{10} ± 4.7%	1.36 ± 6.4%	1.18 ± 2.80%	N/A	A Solution (GEA)
^{137}Cs	3.39x10^{10} ± 9.7%	0.960 ± 10.6%	0.969 ± 2.27%	N/A	A Solution (GEA)
	3.37x10^{10} ± 2.3%	0.954 ± 5.3%		97.7%	Separated Fraction (GEA)
^{140}Ba	3.16x10^{10} ± 2.0%	0.896 ± 4.7%	0.927 ± 2.33%	N/A	A Solution (GEA)
^{141}Ce	2.68x10^{10} ± 2.0%	0.820 ± 5.2%	0.904 ± 3.57%	N/A	A Solution (GEA)
^{143}Ce	2.41x10^{10} ± 3.6%	0.721 ± 5.6%	0.769 ± 2.80%	N/A	A Solution (GEA)
^{144}Ce	2.33x10^{10} ± 3.4%	0.761 ± 5.5%	0.819 ± 2.52%	N/A	A Solution (GEA)
	2.39x10^{10} ± 4.4%	0.781 ± 6.6%		90.6%	Separated Fraction
^{147}Nd	1.41x10^{10} ± 2.9%	1.15 ± 5.2%	1.14 ± 2.80%	N/A	A Solution (GEA)
^{153}Sm[a]	2.54x10^{9} ± 4.4%	3.42 ± 6.1%	3.04 ± 5.5%	49.2%	Separated Fraction
^{156}Eu	3.68x10^{8} ± 11.8%	4.85 ± 11.8%	4.36 ± 16.6%	N/A	A Solution (GEA)
	3.18x10^{8} ± 7.9%	3.89 ± 9.0%		95.7%	Separated Fraction (GEA)
	3.33x10^{8} ± 8.5%	4.08 ± 9.5%		91.2%	Separated Fraction (OES)
^{161}Tb	6.34x10^{6} ± 7.4%	14.6 ± 8.5%	14.1 ± 5.3%	89.6%	Separated Fraction
Isotope	**Atoms/g A (*Atoms/g target*)**		**Chemical Yield (%)**		**Method**
^{237}U	2.27x10^{10} ± 4.3%		N/A		A Solution (GEA)
	2.00x10^{10} ± 3.8%		44.2%		Separated Fraction (GEA)
^{239}Np	4.31x10^{11} ± 2.1%		N/A		A Solution (GEA)
	3.71x10^{11} ± 3.7%		64.2%		Separated Fraction (GEA)
Fissions/g target	5.50x10^{11} ± 4.50%				A Solution (GEA)

[a] Uses updated ^{153}Sm yield from Jackson et. al.

* Chemical yielding issues present

‡ Likely requires updates to nuclear data

Table 8 contains short-lived actinides ^{237}U and ^{239}Np information, this includes the atoms per fission and ratio of the two short-lived actinides for the two uranium targets. Valuable information on the neutron spectrum can be gleaned from the atoms per fission and ratio of the two actinides, because of the sensitivity to neutron energy of the path of production of ^{237}U. Information on HEU (Z11136) short lived actinide was included due to the use of HEU with residual ^{238}U, thus allowing a more favorable pathway to produce the short-lived actinides ^{237}U and ^{239}Np associated with ^{235}U. Double capture on ^{235}U is less favorable than the n,2n reaction on ^{238}U, therefore a low production rate is expected from the ^{235}U activation path. The inclusion of HEU in the examination of ^{237}U and ^{239}Np production is for comparison only to stress the neutron energy and production path differences. Due to the production path, the production rate is such for the DU (Z11137) target that the activity in the A solution of both ^{237}U and ^{239}Np are easily quantifiable.

Table 8. Atoms per fission of ^{237}U and ^{239}Np in HEU (Z11136) Separated Fraction and DU (Z11137) A solution for the FY21 irradiation (N/f ± 1σ%).

	Isotope	FY22	Measurement Method
HEU (Z11136)	^{237}U	7.19×10^{-4} ± 5.3%	Separated Fraction (GEA)
	^{239}Np	5.31×10^{-3} ± 5.5%	Separated Fraction (GEA)
	$^{237}U/^{239}Np$	0.135 ± 7.6%	
DU (Z11137)	^{237}U	0.0413 ± 6.2%	A Solution (GEA)
	^{239}Np	0.571 ± 5.0%	A Solution (GEA)
	$^{237}U/^{239}Np$	0.0721 ± 8.0%	

Shown in Figure 3 is a direct comparison of the neutron spectrum as the $^{237}U/^{239}Np$ over the course of the NCERC campaigns. The difference between FY21 and FY22 is expected due to the differences between the critical assemblies and the differences in their respective neutron spectrum.

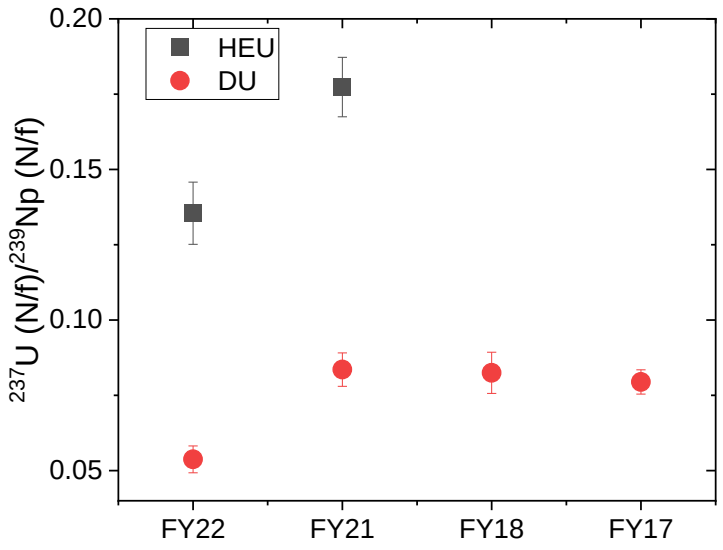

Figure 3. Comparison of the short-lived actinide production between the GODIVA and Flattop critical assemblies examining the FY21 and FY22 results for HEU and FY17, FY18, FY21 and FY22 for DU targets.

5.0 LANL Results

The results from LANL are shown for reference and are discussed with direct reference to their unique situation, further information can be found in LA-CP-22-20725. Rather than the three targets that PNNL received, LANL received and processed a high and low power sample for each of the actinide materials i.e., high, and low power HEU, DU and Pu. This report will only discuss the LANL high power results, as they are directly comparable to the PNNL results.

Table 9. LANL Pu processing results, including atoms/target, R-values, and analysis method.

Isotope	Atoms/g A (*Atoms/g target*)	R Pu	R ENDF – ^{239}Pu Fission Spectrum	Method
^{89}Sr	8.68x10^{10} ± 2.03%	0.37 ± 2.02%	0.357 ± 4.1%	Separated Beta
^{95}Zr	1.85x10^{11} ± 10.0%	0.60 ± 11.2%	0.718 ± 2.2%	A sol. Gamma
	2.31x10^{11} ± 2.96%	0.71 ± 3.56%		Separated Beta (Gamma)
^{97}Zr	2.47x10^{11} ± 2.2%	0.87 ± 5.6%	0.861 ± 2.9%	A sol. Gamma
	2.49x10^{11} ± 5.92%	0.83 ± 6.24%		Separated Beta (Gamma)
^{99}Mo	2.91x10^{11} ± 5.1%	4.76x10^{12} ± 5.1%	1.00 ± 2.2%	A sol. Gamma
	N/A	5.0qx10^{12} ± 1.96%		Separated Beta (Gamma)
^{103}Ru	2.08x10^{11} ± 10.0%	3.00x10^{11} ± 10.0%	2.21 ± 4.7%	A sol. Gamma
^{111}Ag	1.75x10^{10} ± 2.22%	20.06 ± 2.21%	20.0 ± 4.7%	Separated Beta
^{115}Cd	3.40x10^{9} ± 2.44%	5.85 ± 2.43%	6.53 ± 7.4%	Separated Beta
^{115m}Cd	2.84x10^{8} ± 2.44%	5.68 ± 2.43%	4.09 ± 6.8%	Separated Beta
^{136}Cs	7.15x10^{9} ± 4.17%	23.99 ± 4.25%	10.95 ± 90.5	Separated Gamma (Beta)
^{137}Cs	3.10x10^{11} ± 2.54%	1.00 ± 2.33%	1.04 ± 1.8%	Separated Beta (Gamma)
^{140}Ba	2.34x10^{11} ± 3.6%	0.79 ± 6.3%	0.836 ± 2.0%	A sol. Gamma
	2..53x10^{11} ± 2.01	0.81 ± 1.99%		Separated Beta (Gamma)
^{141}Ce	2.80x10^{11} ± 2.2%	1.01 ± 5.6%	0.862 ± 3.4%	A sol. Gamma
	2.74x10^{11} ± 4.24%	0.94 ± 4.68%		Separated Gamma (Beta)
^{143}Ce	1.96x10^{11} ± 3.2%	0.69 ± 6.0%	0.713 ± 2.2%	A sol. Gamma
	2.22x10^{11} ± 4.78%	0.74 ± 5.17%		Separated Gamma

Isotope	Atoms/g A (*Atoms/g target*)	R Pu	R ENDF – ^{239}Pu Fission Spectrum	Method
^{144}Ce	1.91x10^{11} ± 4.33%	0.69 ± 4.76%	0.658 ± 1.9%	Separated Gamma
^{147}Nd	8.77x10^{10} ± 15.9%	0.82 ± 16.7%	0.877 ± 2.2%	A sol. Gamma
^{153}Sm	2.10x10^{10} ± 3.04%	3.13 ± 3.03%	2.63 ± 9.1%	Separated Beta (Gamma)
^{155}Eu	1.03x10^{10} ± 3.68%	6.70 ± 3.67%	6.36 ± 23.4%	Separated Gamma
^{156}Eu	7.85x10^{9} ± 2.79%	10.51 ± 2.79%	10.2 ± 7.4%	Separated Beta
^{161}Tb	3.70x10^{8} ± 2.25%	86.69 ± 2.24%	99.2 ± 5.9%	Separated Beta

Isotope	Atoms/g A (Atoms/g target)	Method
^{235}U	1.76x10^{16} ± 57.1% (5.99x10^{18} ± 57.1%)	A Solution (GEA of ^{235}U)
^{237}U	4.28x10^{7} ± 37.5% (1.46x10^{10} ± 37.5%)	A Solution (GEA of ^{235}U)
^{239}Np	**N/A[b]**	**Separated Fraction (GEA)**

Table 10. LANL HEU processing results, including atoms/target, R-values, and analysis method. Methods within parentheses indicate a second technique within 1σ of the quoted value.

Isotope	Atoms/g target	R HEU	R ENDF - ^{235}U Fission Spectrum	Method
^{89}Sr	1.69x10^{11} ± 2.02	0.96 ± 1.98	0.950 ± 2.62%	Separated Beta
^{91}Y	2.01x10^{11} ± 2.20	0.92 ± 2.16	1.01 ± 90.5%	Separated Beta
^{95}Zr	2.27x10^{11} ± 3.3	0.96 ± 3.08	1.02 ± 2.62%	A sol. Gamma Separated Beta
^{97}Zr	2.19x10^{11}	0.95 ± 3.94	1.03 ± 3.14%	A sol. Gamma
^{99}Mo	2.22x10^{11}± 3.3	3.64x10^{12}	N/A	A sol. Gamma
	2.28x10^{11} ± 1.95	3.73x10^{12} ± 1.95		Separated Beta
^{103}Ru	1.14x10^{11}	1.04 ± 5.2	1.10 ± 2.80%	A sol. Gamma
^{111}Ag	1.82x10^{9} ± 2.35	2.80 ± 2.32	2.51 ± 5.27%	Separated Beta
^{115}Cd	1.36x10^{9} ± 2.82	3.14 ± 2.79	2.76 ± 6.91%	Separated Beta
^{115m}Cd	1.15x10^{8} ± 2.89	3.08 ± 2.86	2.86 ± 23.85%‡	Separated Beta
^{136}Cs	5.39x10^{8} ± 5.36	2.43 ± 5.34	2.18 ± 90.5%	Separated Gamma (Beta)
^{137}Cs	2.37x10^{11} ± 2.10	1.03 ± 2.06	1.03 ± 2.10%	Separated Beta (Gamma)
^{140}Ba	2.21x10^{11} ± 1.98	0.95 ± 1.95	0.989 ± 2.43%	Separated Beta (Gamma)

Isotope	Atoms/g target	R HEU	R ENDF - ^{235}U Fission Spectrum	Method
^{141}Ce	2.12x10^{11} ± 2.63	0.97 ± 3.27	1.05 ± 3.57%	Separated Gamma (Beta)
^{143}Ce	1.94x10^{11} ± 3.1	0.90 ± 4.5	0.989 ± 2.80%	A sol. Gamma
	2.13x10^{11} ± 2.64	0.89 ± 2.61		Separated Beta (Gamma)
^{144}Ce	2.04x10^{11} ± 15.6	1.02 ± 16.0	0.985 ± 2.52%	A sol. Gamma
	1.86x10^{11} ± 2.70	0.91 ± 3.33		Separated Gamma
^{147}Nd	7.57x10^{10} ± 4.5	0.93 ± 5.6	1.14 ± 2.80%	A sol. Gamma
^{153}Sm	**6.42x10^{9} ± 3.17**	**1.28 ± 3.15**	**1.27 ± 6.47%**	**Separated Beta (Gamma)**

Table 11. LANL DU processing results, including atoms/target, R-values, and analysis method.

Isotope	Atoms/g A (*Atoms/g target*)	R DU	R ENDF - ^{238}U Fission Spectrum	Method
^{89}Sr	1.51x10^{10} ± 2.02	0.56 ± 1.99	0.578 ± 2.62%	Separated Beta
^{91}Y	2.15x10^{10} ± 2.32	0.64 ± 2.30	0.686 ± 90.5%	Separated Beta
^{95}Zr	2.63x10^{10} ± 5.4	0.70 ± 6.6	0.783 ± 2.80%	A sol. Gamma
	2.78x10^{10} ± 2.38	0.74 ± 3.08		Separated Gamma
^{97}Zr	2.92x10^{10} ± 8.1	0.85 ± 9.0	0.921 ± 3.14%	A sol. Gamma
	2.91x10^{10} ± 4.00	0.85 ± 4.45		Separated Gamma
^{99}Mo	3.53x10^{10} ± 3.9	5.77x10^{11*}	N/A	A sol. Gamma
	3.52x10^{10} ± 1.96	5.76x10^{11} ± 1.96		Separated Beta (Gamma)
^{103}Ru	3.16x10^{10} ± 5.4	1.80 ± 6.6	2.05 ± 2.80%	A sol. Gamma
^{111}Ag	3.91x10^{8} ± 2.27	3.90 ± 2.25	4.05 ± 4.89%	Separated Beta
^{115}Cd	1.99x10^{8} ± 3.16	2.98 ± 3.15	2.95 ± 6.63%	Separated Beta
^{115m}Cd	1.84x10^{7} ± 3.45	3.19 ± 3.44	3.07 ± 8.71%‡	Separated Beta
^{136}Cs	3.05X10^{6} ± 10.02	0.09 ± 10.01	0.172 ± 90.5%	Separated Gamma
^{137}Cs	3.89x10^{10} ± 4.1	1.09 ± 5.7	0.969 ± 2.27%	A sol. Gamma
	3.50x10^{10} ± 2.10	0.98 ± 2.08		Separated Beta (Gamma)
^{140}Ba	3.55x10^{10} ± 5.2	0.99 ± 6.5	0.927 ± 2.33%	A sol. Gamma
	3.27x10^{10} ± 1.98	0.92 ± 1.96		Separated Beta (Gamma)
^{141}Ce	2.86x10^{10} ± 3.0	0.85 ± 4.9	0.904 ± 3.57%	A sol. Gamma
	2.87x10^{10} ± 2.66	0.85 ± 3.30		Separated Gamma (Beta)
^{143}Ce	2.48x10^{10} ± 2.7	0.72 ± 4.8	0.769 ± 2.80%	A sol. Gamma
	2.92x10^{10} ± 2.83	0.74 ± 2.82		Separated Gamma (Beta)
^{144}Ce	3.11x10^{10} ± 13.0	0.98 ± 13.6	0.819 ± 2.52%	A sol. Gamma
	2.59x10^{10} ± 2.82	0.82 ± 3.43		Separated Gamma
^{147}Nd	1.36x10^{10} ± 5.6	1.09 ± 5.7	1.14 ± 2.80%	A sol. Gamma
^{153}Sm[a]	2.03x10^{9} ± 3.40	2.63 ± 3.39	3.04 ± 5.5%[a]	Separated Beta
Isotope	**Atoms/fission**		**Atoms/g A**	**Method**

Isotope	Atoms/g A (*Atoms/g target*)	R DU	R ENDF - ^{238}U Fission Spectrum	Method
			(*Atoms/g target*)	
^{237}U	3.98x10^{-2} ± 3.20		2.29x10^{10} ± 3.75	Separated Gamma
^{239}Np	0.558 ± 3.1		3.21x10^{11} ± 2.3	Separated Gamma

6.0 Conclusions

Three targets were assembled at LANL, irradiated at NCERC, dissolved, and split between two national laboratories: LANL and PNNL. The target solutions were chemically separated and analyzed in parallel at LANL and PNNL using different methods. Results from PNNL of the analysis of most of the fission products, agreed with literature as well as LANL for all three targets.

The ^{237}U/^{239}Np was consistent with the expected neutron spectrum for the irradiation of DU, though is slightly different than what was found using Flattop. Modifications to chemistry were made, which improved timelines and yields for several analytes. The results from this work represents significant improvements to the uncertainties associated with several fission product's R-values. A repeated analysis using the Godiva critical assembly should conducted with the same core material to provide a second data point for comparison.

It cannot be understated that these campaigns are a team effort between the two national laboratory's teams, but the FY22 campaign was an exemplary demonstration of the teamwork between the two labs, whether that was hosting visiting scientists or assisting with shipping issues.

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