Methods to Collect, Compile, and Analyze Observed Short-lived Fission Product Gamma Data

EC Finn   RF Payne   LR Greenwood   BD Pierson
LA Metz   JI Friese   JD Kephart   TA Ellis

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Pacific Northwest
NATIONAL LABORATORY

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Summary

A unique set of fission product gamma spectra was collected at short times (4 minutes to 1 week) on various fissionable materials. Gamma spectra were collected from the neutron-induced fission of uranium, neptunium, and plutonium isotopes at thermal, epithermal, fission spectrum, and 14-MeV neutron energies. This report describes the experimental methods used to produce and collect the gamma data, defines the experimental parameters for each method, and demonstrates the consistency of the measurements.

Acronyms

DSM	direct simultaneous measurement
HPGe	high purity germanium
IBI	International Bio-Analytical Industries
IPL	Isotope Products Laboratories
MCNP	Monte Carlo N-Particle (transport code)
NBL	New Brunswick Laboratory
ORNL	Oak Ridge National Laboratory
PNNL	Pacific Northwest National Laboratory
WSU	Washington State University

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1.0   Introduction

Experiments involving the neutron irradiation of various actinides and subsequent fission product measurements have traditionally been conducted at times greater than 5 days, resulting in the analysis of fission products with half-lives in the range of 1 day to 30 years following chemical separations. Interpreting gamma spectra on samples of unseparated shorter-lived fission products (radionuclides with half-lives ranging from 75 s to 3 days) is not as well understood because of the complexity of these spectra and the limited availability of experimental spectra containing the full suite of fission products at these half-lives. Modeling of gamma spectra is thus frequently used to deduce information about short-lived nuclear data. A goal of this work was to compile a set of fission-product gamma data at short times on various fissionable materials.

This report describes the methods used to collect, compile, and analyze the set of short-lived fission-product gamma data. The experimental procedure used to collect fission-product data from the neutron-induced fission of uranium, neptunium, and plutonium at four different energy bins (Table 1.1) is outlined in this report. Experiments with thermal, epithermal, and fission energy neutrons were completed at the Washington State University (WSU) TRIGA reactor by irradiating the samples bare, cadmium shielded, and boron carbide shielded, respectively. High neutron energy (14-MeV) experiments were completed at Pacific Northwest National Laboratory (PNNL) with a D-T 14-MeV neutron generator. Samples were counted on a standard high purity germanium (HPGe) detection system as well as an advanced coincidence system, starting at 4 minutes post irradiation and continuing for up to 1 week.

It is important to preserve these data in such a way that they are available for public use for various nuclear physics or chemistry applications. This report describes the experimental parameters and methods used to verify the consistency of the data. This knowledge facilitates understanding and use of the data set. Detailed information about neutron energy ranges and fluences used for each experiment is provided. A brief description of how short-lived isotopes were measured in the complicated gamma spectra is also provided in this report.

Table 1.1. Short-Lived Fission Product Experiments Completed

Samples	Thermal	Epithermal	Fission Spectrum	14 MeV
^{233}U	✓	✓	✓	
^{235}U	✓	✓	✓	
^{238}U		✓	✓	✓
^{237}Np		✓	✓	
^{239}Pu	✓	✓	✓	

2.0 Experimental

2.1 Sample Preparation

2.1.1 Thermal, Epithermal, and Fission Energy Neutron Samples

Working solutions containing known concentrations of the radionuclides ^{233}U, ^{235}U, ^{238}U, ^{237}Np, and ^{239}Pu individually at >97% isotopic purity were prepared. The stock materials came from isotopic standards listed in Table 2.1, except for the materials for the 14 MeV experiments that are described below. The sample matrix was less than or equal to 2 M nitric acid. Nitric acid was used to minimize formation of activation products during irradiation. Other matrices such as hydrochloric acid can result in significant activation that interferes with the gamma spectra collected. The mass of sample used in each case is given in Table 2.1, where the mass of sample was calculated to produce approximately 1 x 10^8 fissions from a pulse of a 1 MW TRIGA reactor.

In order to avoid cross-contamination, supplies used for sample preparation were leached using an 8 M NH$_4$OH/8 M HNO$_3$ procedure prior to use. In addition, sample preparation and packaging for irradiation were done in a clean environment. Samples of each isotope were prepared in triplicate. A gravimetric aliquot of each working solution was placed into a vial and transposed to a few microliters of 2 M HNO$_3$.Whatman ashless filters were cut to 8 mm using a hole boring tool. The transposed solution was added to the filter in 0.5 μL aliquots and dried under a heat lamp after each addition to keep the deposited material as concentrated as possible. Complete transfer of the material was ensured by rinsing the vial with a few microliters of <2 M HNO$_3$ and adding to the filter in the same fashion as the sample. It is important to use a moderate concentration of nitric acid to be sure that quantitative transfer is complete while avoiding the fragility of the filter that occurs when higher concentrations are used. When sample loading was complete, each filter was heat-sealed into a Tedlar® sleeve. Tedlar® was used in an effort to retain gaseous fission products, however losses were still observed (discussed below). In Figure 2.1, the sample is shown drying onto the filter.

2.1.2 14 MeV Neutron Samples

A single sample was prepared from thin foils of 99.998% pure ^{238}U metal. These foils are circles 0.5 in. in diameter and 0.005 in. thick. Each foil weighs approximately 0.3 g. The foil was laid on a piece of packing tape and a second piece of packing tape was carefully laid over the top and pressed to seal. The excess tape was cut away to leave approximately 3 mm of tape around the edges of the foil. Multiple foils were stacked to create a larger sample of 1.57 g. Due to the low neutron flux of the D-T generator, the largest sample mass resulted in only ~1.5×10^7 fissions.

Table 2.1. Sample Information for Each Fission Experiment

Experiment	Isotopic Standard	Initial Chemical Form	Isotopic Purity (%)	Average Sample Mass (g)
^{233}U Thermal	NBL CRM 111A	$UO_2(NO_3)_2$ (aq)	99.4911	6.80E-07
^{233}U Epithermal	NBL CRM 111A	$UO_2(NO_3)_2$ (aq)	99.4911	2.77E-05
^{233}U Fission	NBL CRM 111A	$UO_2(NO_3)_2$ (aq)	99.4911	5.02E-04
^{235}U Thermal	NBL CRM U970	U_3O_8	97.663	3.51E-07
^{235}U Epithermal	NBL CRM U970	U_3O_8	97.663	5.21E-05
^{235}U Fission	ORNL U235-165-AR	U_3O_8	99.71	2.05E-03
^{238}U Epithermal	IBI	UO_2	99.80	2.60E-03
^{238}U Fission	IBI	UO_2	99.80	2.62E-03
^{238}U 14 MeV	PNNL	Metal foil	99.998	1.57
^{237}Np Epithermal	PNNL	NpO_2	99.99	9.98E-04
^{237}Np Fission	PNNL	NpO_2	99.99	1.01E-03
^{239}Pu Thermal	IPL (Lot# 1195.15)	$Pu(NO_3)_4$	97.937	3.55E-07
^{239}Pu Epithermal	PNNL	Metal foil	99.11	4.29E-05
^{239}Pu Fission	PNNL	Metal foil	99.11	7.68E-04

NBL—New Brunswick Laboratory
ORNL—Oak Ridge National Laboratory
IBI—International Bio-Analytical Industries
IPL—Isotope Products Laboratories

Figure 2.1. Sample Preparation. Sample is drying onto filter.

2.2 Sample Packaging

2.2.1 Thermal Neutron Spectrum Data

Thermal neutron spectrum data were collected with unshielded samples. The three individual filter papers enclosed in Tedlar® were stacked and pushed into the bottom of a 2/5 dram polyethylene vial. This vial was heat sealed to make sure that the sample and fission products were contained. The neutron fluence varies between reactor pulses and cannot be predicted accurately (Payne et al. 2009); therefore, a series of neutron fluence monitors were irradiated with each sample to quantitatively analyze the neutron

spectrum. The fluence monitors were composed of high-purity elements and alloys, described in Table 2.2. The wires were placed in the bottom of a second 2/5 dram vial, which was also heat sealed. To ensure that the neutron spectrum obtained from analyzing the fluence monitors was representative of that incident on the samples, the samples and fluence monitors were placed as closely together as possible. This was accomplished by placing the vial containing the samples upside-down into a 2-dram vial. The vial containing the fluence monitors was then placed on top of this, and the 2-dram vial was heat-sealed. The 2-dram vial was then fixed to a rotator assembly using zip ties.

Table 2.2. Approximate Masses of Fluence Wires Used in Each Set of Experiments

Material (purity), manufacturer	Approximate Mass Used for Thermal (unshielded) Experiments (mg)	Approximate Mass Used for Epithermal (Cd-shielded) Experiments (mg)	Approximate Mass Used for Fission Spectrum (B_4C-shielded) Experiments (mg)
Cu (99.999%), Cominco	3.6	7.7	8.7
Fe (99.999%), MARZ	121.8	125.8	156.7
Ti (99.917%), Reactor Experiments, Inc.	11.2	10.9	12.5
1.0% Co-V alloy, Mol (Fabry)	2.0	15.3	7.0
0.135% Au-Al alloy (99.916%), Reactor Experiments, Inc.	2.2	7.5	8.6

2.2.2   Epithermal Neutron Spectrum Data

Epithermal neutron spectrum data were collected using cadmium shielded samples. Cadmium shields were 99.95% pure and were purchased from Reactor Experiments, Inc. The shields measured 1 in. in outer diameter and 0.020 in. thick. The fluence monitors were heat-sealed into a plastic bag and laid in the bottom of the cadmium shield. The fluence monitors are the same materials as were used in the thermal experiments (Table 2.2). The filter papers enclosed in Tedlar® were laid on top of the fluence monitor package. The cadmium shield was closed by lightly crimping the edges by hand and using two narrow strips of Cd metal wrapped completely around the shield perpendicular to each other. This assembly was then placed in the bottom of an 8-dram vial with a loosely crumpled KimWipe® on top to prevent excessive movement. The vial was heat sealed and fixed to a rotator assembly with zip ties.

2.2.3   Fission Energy Neutron Data

Boron is known (McLane et al. 1988; Galy et al. 2000; Turner and Haynes 2009) to absorb thermal and epithermal neutrons, allowing only fission energy neutrons to interact with the sample. Therefore, PNNL designed a boron carbide shield to be used to collect fission energy neutron data. The shield was manufactured by Ceradyne Inc. A cross-sectional schematic of this shield is shown in Figure 2.2. The shield is 5 cm in diameter and 12 cm high with a total mass of 577.6 g. It has a male and female side that fit snugly together to form a 1 cm by 8 cm cylindrical interior cavity.

In packaging the samples for irradiation, it is important to ensure that the filters and fluence monitors are in close proximity to one another for the most reliable fluence measurements. Vertical alignment is especially important as the neutron fluence is very sensitive to this parameter. The fluence monitors were heat-sealed into a small plastic bag. To keep the filters and fluence monitors together, the filters in Tedlar® were stacked with the fluence monitor package and slipped into a plastic sleeve. The plastic sleeve containing the filters and fluence monitors is rolled and slipped into the female side of the boron carbide shield. The female side is used to facilitate easier removal of the samples following irradiation.
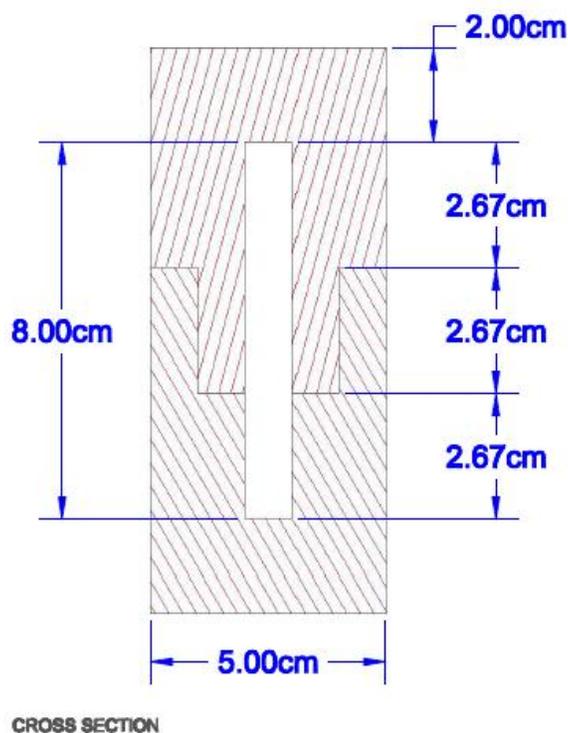
CROSS SECTION

Figure 2.2. Cross-Sectional Schematic of the B$_4$C Capsule

The assembled boron carbide shield was heat sealed into a plastic bag (Figure 2.3). The boron carbide was then placed into a polyethylene container custom designed and fabricated at WSU (Figure 2.4), which was sealed with o-rings and zip ties.

Figure 2.3. B$_4$C Shield Sealed in Plastic Before Irradiation

Figure 2.4. Custom-Fabricated Polyethylene Containment Used for B$_4$C Irradiations

2.2.4   14 MeV Neutron Spectrum

High-purity fluence foils 0.5 in. in diameter were used to measure the neutron spectrum. The materials used are described in Table 2.3. The aluminum foil discs were cut from a metal strip. The foils were stacked in the order Fe, Au, In, Cu, and Al and heat-sealed into a plastic bag. The Al was placed directly behind the sample foils inside a Cd shield (as used for the epithermal experiments) to eliminate thermal fissions due to environmental down-scattering of the neutrons. The Cd shield holding the fluence foils and sample was sealed in a plastic bag.

Table 2.3. Approximate Masses of Fluence Wires Used in 14 MeV Neutron Experiments

Material (purity), manufacturer	Thickness (in.)	Approximate Mass Used for Experiment (mg)
Fe (99.9%), Reactor Experiments, Inc.	0.005	100
Au (99.992%), Reactor Experiments, Inc.	0.002	100
In (99.99%), Reactor Experiments, Inc.	0.005	200
Cu (99.9928%), Reactor Experiments, Inc.	0.005	100
Al (99.99%), Materials Research Corporation	0.010	100

2.3 Sample Irradiation

2.3.1 1 MW TRIGA Irradiations

Thermal, epithermal, and fission neutron spectrum irradiations were performed at the 1-MW TRIGA reactor at WSU in Pullman, Washington. For the thermal and epithermal neutron spectrum experiments, the reactor was stabilized at 80 W before inserting the samples. Samples were placed into position D8 and irradiated for approximately 50 ms following a 2-dollar reactivity insertion. Samples were immediately removed from the reactor without cooling, resulting in contact dose rates of 50 to 100 mrem/hr (bare irradiations) and 18 to 39 mrem/hr (Cd-shielded) upon removal.

Adjustments to the irradiation conditions were required for the fission neutron spectrum experiments. The B_4C shield was carefully designed and fabricated to ensure reactor safety and correct experimental conditions during use. Prior to fabrication, a piece of the proposed B_4C material from Ceradyne, Inc. was irradiated at WSU. No interfering impurities were present and the material from Ceradyne, Inc. was deemed suitable for fabricating the shield. It was anticipated that the large mass of the boron carbide could influence the stabilization of the reactor, so the shield was briefly irradiated empty. It was found that reactor power dropped significantly following boron carbide insertion; re-stabilization was required before pulsing the reactor. Based on these results, the reactor was stabilized at ~100 W prior to insertion of actinide samples. After the sample in the shield was inserted the reactor was re-stabilized within 3-5 minutes to a power of 80 W. Once stabilized, the reactor was pulsed as was done to achieve thermal and epithermal neutron spectra. Samples were removed without cooling and the observed dose rate was 20-34 mrem/hr on contact. The neutron fluence to the samples during reactor stabilization was negligible relative to the fluence during the pulse.

2.3.2 14 MeV Irradiations

Irradiations were conducted at PNNL with a Thermo Scientific P 385 neutron generator. The generator was positioned on an aluminum support in the middle of the laboratory to reduce the down scattering of neutrons (Figure 2.5A). The location of maximum neutron fluence was determined by making neutron fluence measurements at four different locations on the outside of the neutron generator head. The location directly adjacent to the target plane resulted in the highest measured neutron fluence (9.11×10^{11} n/cm^2 ± 12%). For each experiment, the plastic bag holding the sample and fluence foils was taped to a stainless steel band clamp loosely placed around the target plane of the neutron generator head

4.5 in. from the front of the generator tube (Figure 2.5B). Each irradiation lasted for 2 hours. Following irradiation, the sample was repackaged and transported as quickly as possible for gamma analysis.

Figure 2.5. A) Photograph of the 14 MeV Generator Used for Experiments. B) Sample Positioned on the 14 MeV Generator Perpendicular to the Target Plane (4.5 inches from the front of the tube).

2.4   Sample Counting

The triplicate samples of each isotope were irradiated together for each experiment performed at WSU. One of the replicates was counted beginning 4 minutes after irradiation at WSU using a single-crystal HPGe detector. The remaining two samples and the fluence monitors were returned to PNNL for counting. From 6 hours to 1 week following the pulse, the second sample was counted at PNNL with a single crystal HPGe detector. The fluence monitors were also analyzed at PNNL with HPGe detection. The third sample was counted on a direct simultaneous measurement (DSM) detector system at PNNL from ~6 hours to 1 week post-pulse.

Only one sample was irradiated during the 14-MeV neutron spectrum experiments. Gamma acquisition was performed with a single-crystal HPGe detector at PNNL 70 min after completing the irradiation and continuing for up to 1 week. The following sections provide further details about each detection system and methods used for their operation.

2.4.1   Gamma Acquisition by High Purity Germanium Detector at WSU

In the first 5 ½ - 6 hours following the pulse, 14 total spectra were collected. The count time was always based on live time. Counting began 4 minutes after the pulse. The following gamma spectra were collected: three 5-minute spectra, three 10-minute spectra, three 15-minute spectra, two 30-minute spectra, and three 60-minute spectra. The durations of the counts were targeted to approximate the half-lives of the isotopes being analyzed in that time slice. The detector used was a 68% relative efficiency HPGe detector, previously calibrated using standard methods. It was operated by the Genie 2000 software package from Canberra Industries. The first counts were on Shelf 10, 4.5 inches from the detector face. The sample was moved to Shelf 5, 2 inches from the detector face, as the dead time dropped to ~5%. As the dead time again reached 5% the sample was moved to Shelf 3, 1 inch from the detector face.

2.4.2   Gamma Acquisition by High Purity Germanium Detector at PNNL

Gamma acquisition was performed at PNNL with several single-crystal HPGe detectors with relative efficiencies ranging from 40% to 105%. These detectors were operated by either APEX Gamma or Genie VMS (Canberra Industries). The calibrated geometries were on the detector face, or 5 or 10 cm distant from the face. Upon arrival at PNNL, fluence wires had to be first unpackaged to allow separation and placement in individual plastic vials to match previously calibrated detector geometry. Filters and fluence wires were counted in various calibrated geometries for times ranging from 10 minutes to 10 hours, depending on the characteristics of the particular sample. Counting of the samples continued for 1 week after each pulse. Specific information on count geometry, duration, and calibration files are provided with each spectrum.

2.4.3   Gamma Acquisition by Direct Simultaneous Measurement at PNNL

The DSM detector system was developed at PNNL and is an array of three HPGe crystals connected in coincidence. The DSM system has three HPGe detectors, two on top and one on bottom. The detectors on top are a planar p-type detector and a semi-coaxial N-type detector. The planar detector (relative efficiency 20%) has a diameter of 70 mm and a thickness of 20 mm; its face is 6 mm from the Be window. The semi-coaxial detector (relative efficiency 140%) has a diameter of 80 mm and a thickness of 80 mm. The semi-coaxial detector is located 10 mm from the back of the planar detector, or 36 mm from the Be window. The bottom detector (relative efficiency 90%) is a p-type semi-coaxial crystal. Detection efficiency and dead time can be optimized by adjusting the relative spacing of the detectors. The sample filter inside Tedlar® is mounted in a card geometry and placed 1 cm from the front face of both detector cryostat faces. This system uses an XIA dgf4c electronics package, which allows list mode data of the energy and synchronized time of each event. The data are parsed into files to create time slice histograms and analyze peaks and coincidence using a PNNL-developed code. The detector system was calibrated for energy and efficiency using standard methods.

3.0   Data Analysis

The purpose of data analysis for this work was to verify the quality of the collected data. Sample and fluence monitor data were all analyzed using Genie 2000 v. 3.2 software (Canberra Industries). Below are descriptions of data analysis methods used to determine the neutron spectrum and verify data quality.

3.1   Neutron Fluence Monitors and Observed Spectrum

The neutron spectrum may be quantified by use of fluence monitors that have activation reactions sensitive to different neutron energies. The reactions observed during reactor pulses are described in Table 3.1. The reaction rate is calculated for each reaction in Table 3.1, and an average fluence was calculated for each pulse. The fluence values calculated from each reaction indicated good agreement, with a standard deviation of less than 10% for each reactor pulse. Table 3.2 shows the range of values observed in thermal, epithermal, and fission neutron spectrum experiments. The fluence between reactor pulses cannot be directly compared as this value may be highly variable (Payne et. al. 2009). The reactions monitored and fluences observed in the 14-MeV experiments are presented in Table 3.3. The standard deviation between fluence values obtained from different reactions in 14-MeV neutron irradiations was 12%. This is slightly higher than in the pulsed experiments, but nonetheless indicates good agreement between the fluences determined.

Table 3.1. Activation Reactions Observed for Calculation of Neutron Spectrum

Reaction	Energy Range (MeV)	
$^{63}Cu(n, \gamma)^{64}Cu$	1.0×10^{-8}	1.7×10^{-7}
$^{58}Fe(n, \gamma)^{59}Fe$	1.0×10^{-8}	1.7×10^{-7}
$^{59}Co(n, \gamma)^{60}Co$	1.0×10^{-8}	1.0×10^{-4}
$^{197}Au(n, \gamma)^{198}Au$	1.0×10^{-8}	4.3×10^{-6}
$^{54}Fe(n, p)^{54}Mn$	2.3	7.4
$^{56}Fe(n,p)^{56}Mn$	6.0	11.0
$^{27}Al(n, \alpha)^{24}Na$	6.7	12.0
$^{47}Ti(n, p)^{47}Sc$	6.7	12.0
$^{48}Ti(n,p)^{48}Sc$	5.5	12.0

Table 3.2. Ranges of Fluence Values Observed in Each Neutron Spectrum

	Thermal Neutron Spectrum		Epithermal Neutron Spectrum		Fission Neutron Spectrum	
	Low	High	Low	High	Low	High
Thermal fluence (n/cm^2)	1.96×10^{14} $\pm 3\%$	2.28×10^{14} $\pm 3\%$	$<1 \times 10^{11}$			
Epithermal fluence (n/cm^2)	4.92×10^{13} $\pm 10\%$	6.17×10^{13} $\pm 10\%$	6.02×10^{13} $\pm 5\%$	9.91×10^{13} $\pm 5\%$		
Fission fluence (n/cm^2)	2.05×10^{13} $\pm 4\%$	2.75×10^{13} $\pm 3\%$	2.04×10^{13} $\pm 3\%$	3.93×10^{13} $\pm 2\%$	1.96×10^{13} $\pm 3\%$	4.11×10^{13} $\pm 5\%$

Table 3.3. Example of Observed Neutron Fluence for Cd-Shielded 14-MeV Experiments

Reaction	Observed fluence (n/cm^2)	Error (%)
^{56}Fe(n, p)^{56}Mn	1.06×10^{10}	6
^{65}Cu(n, 2n)^{64}Cu	8.84×10^9	2
^{197}Au(n, 2n)^{196}Au	7.96×10^9	3
^{27}Al(n, α)^{24}Na	9.05×10^9	2
Average	9.11×10^9	12

A Monte Carlo N-Particle (MCNP) model (not shown) was created from the core deck provided by WSU for position D8. The model represents the neutron spectrum anticipated inside each of the shielding materials used. The model confirms that low-energy neutrons are absorbed to an increasing extent when comparing thermal, Cd-shielded, and boron carbide-shielded neutron spectra.

The MCNP models are purely theoretical and therefore cannot be directly compared to experimental fluence data. In order to make this comparison, the MCNP models were modified by use of the STAY'SL computer code (Perey 1977). The format of STAY'SL input and output files was modified at PNNL and standard libraries were established. Input to the STAY'SL code included the reaction rates for each of the nuclear reactions described in Table 3.1 and Table 3.3 and the MCNP calculations. The STAY'SL adjusted neutron spectra for the WSU core are shown in Figure 3.1. The same corrections were made for the MCNP model of the 14 MeV neutron generator (Figure 3.2).

The initial MCNP for thermal, epithermal, and 14-MeV calculations showed some differences from the measurements that varied considerably for each neutron environment. Typically, the problem was that MCNP did not predict the correct ratio of thermal neutrons to the rest of the spectrum with differences typically on the order of 20 to 30%. However, after spectral adjustment with STAY'SL, all the measured reaction rates showed very good agreement, indicating that the measurements are internally consistent, and they provide confidence in the adjusted neutron spectra.

As the B$_4$C shield was fabricated for these experiments, special attention was given to verifying that the shield was performing in a consistent and predictable manner. In the fission energy spectrum experiments with B$_4$C shielding, there was excellent agreement between the rates of the various activation reactions, four of which are sensitive to thermal neutrons and five of which are threshold reactions. Results calculated from each of the reactions agree within 10% and the deviation between theoretical and experimental reaction rates is 4.7%. The total neutron fluence is 6.18 x 10^{13} n/cm^2, with 2.67 x 10^{13} n/cm^2 above 1 MeV. Less than one percent of the neutrons are below 0.1 keV. These data and their consistency indicate that the boron carbide shield is performing as expected and that the thermal contribution to the overall fluence is minimal. See Figure 3.6 below for further evidence of the quality and consistency of the performance of the boron carbide shield.
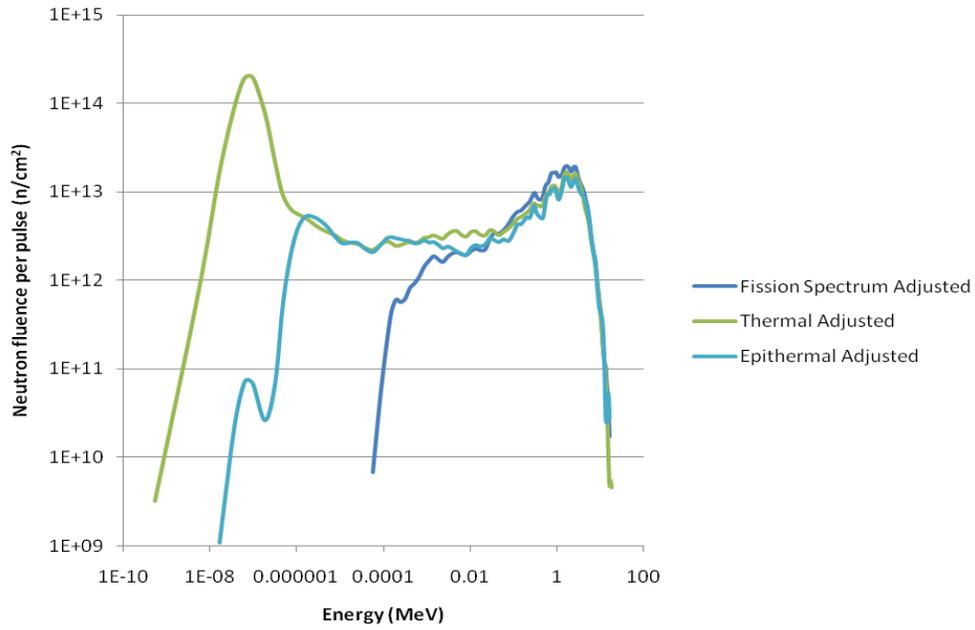
**Figure 3.1.** Adjusted STAY'SL Model of Neutron Spectrum Within the WSU Core
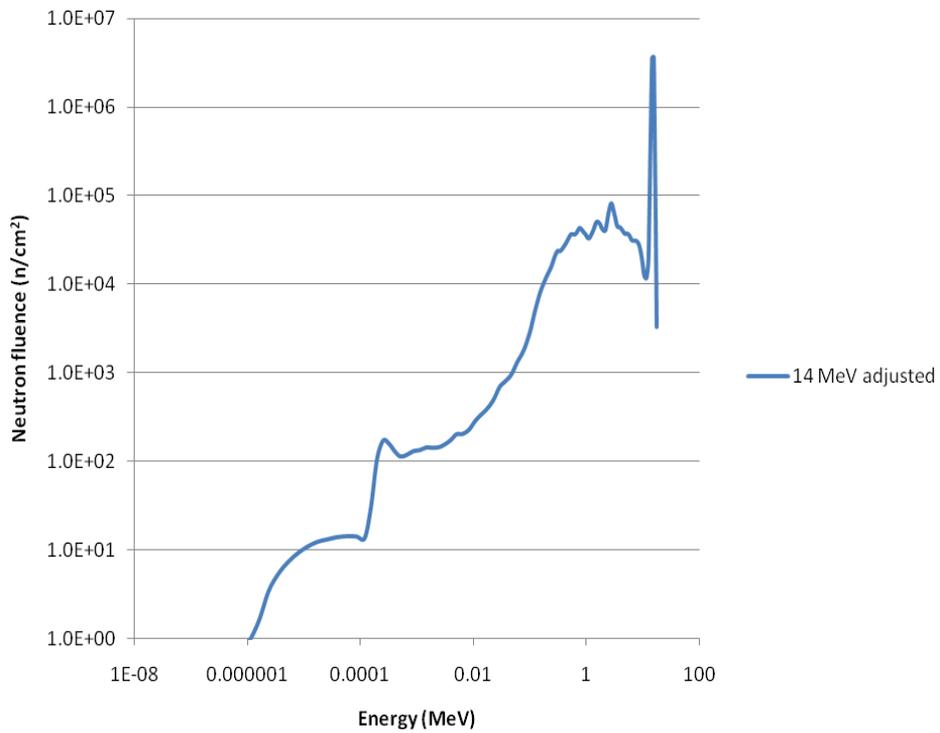
**Figure 3.2.** Adjusted STAY'SL Model for 14 MeV Neutron Generator

3.2 Verification of Data Quality

Sample analysis for both PNNL's and WSU's single-crystal HPGe detectors was performed using Genie 2000 v. 3.2 software (Canberra Industries). Short-lived fission and activation products were identified and quantified. Novel data analysis methods were developed for the time-stamped DSM data. Following data conversion, time slices from the DSM detector system were analyzed using Genie 2000. Data from single-crystal HPGe detectors and the converted DSM data are available on this website. The "How to Access the Data" document provides information on obtaining additional time slices of DSM data. To improve analysis of short-lived isotopes in Genie 2000, a traditional fission product library containing longer-lived isotopes was expanded to include short-lived isotopes with half-lives ranging from 75 seconds to 3 days.

Figure 3.3 provides an example of data from a fission spectrum neutron pulse plotted as fission yield to make it easier to visualize multiple sample nuclides. The number of fissions is calculated by comparing the experimental data with published fission yields from England and Rider (1993). For thermal, epithermal, and fission energy neutron experiments, well-defined double-humped fission product curves were generated. A somewhat smaller number of radionuclides were observed in the 14-MeV experiments (Figure 3.4) because of a lower neutron fluence and fewer resulting fissions, but clusters of radionuclides centered on two masses are still visible.
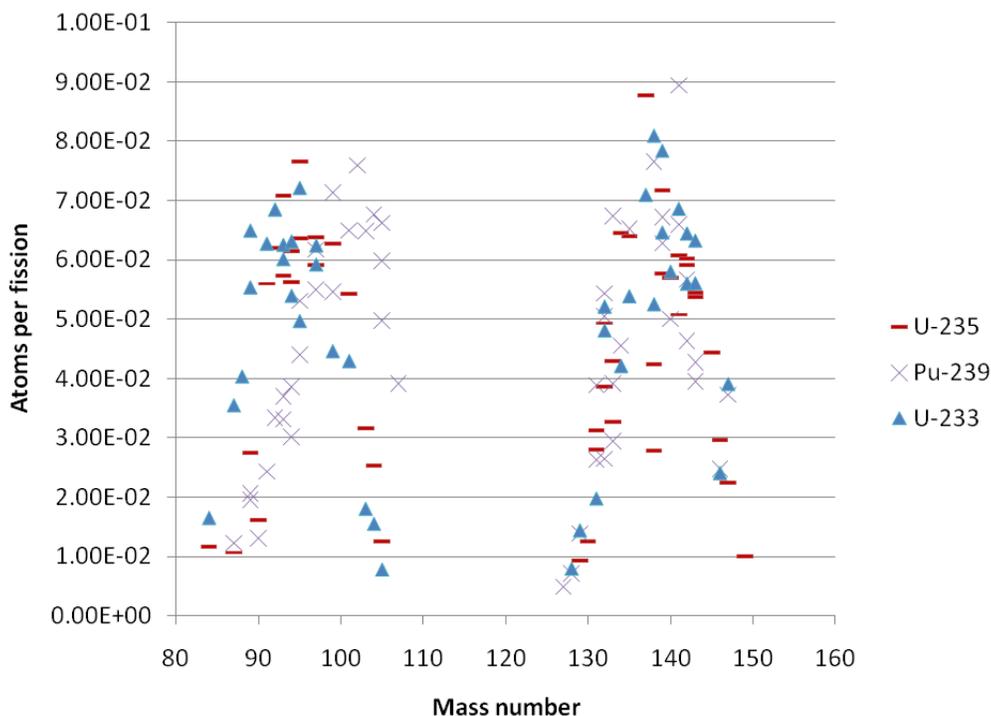
Figure 3.3. Measured Fission Yields in Fission Energy Neutron Experiments
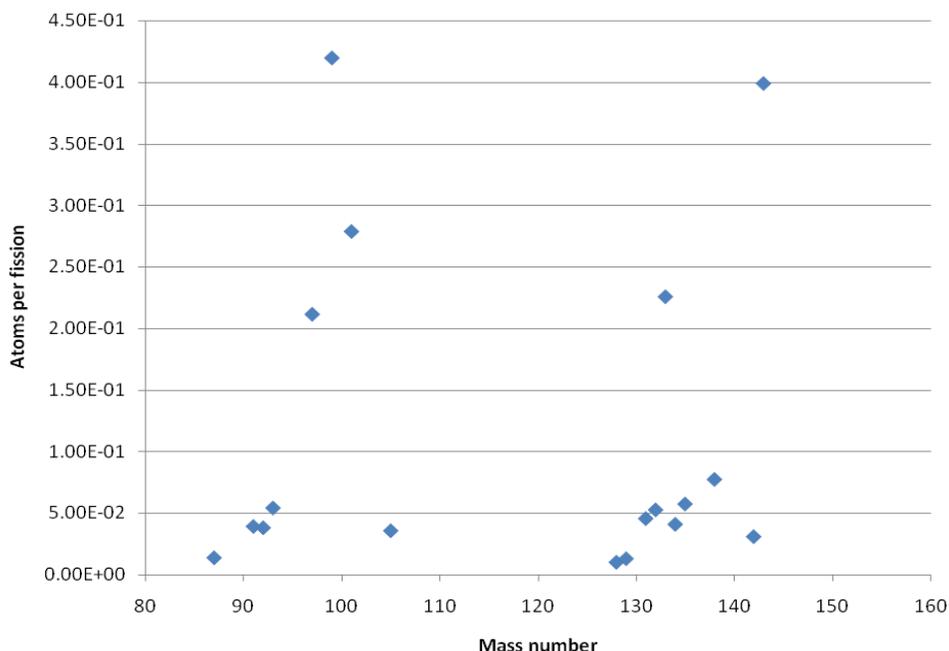
Figure 3.4. Measured Fission Yields in 14-MeV Experiments with ^{238}U

There are several methods available to analyze the gamma data for internal consistency, which provides a measure of its quality and reliability. These methods assure that the correct radionuclides have been identified and that the quantification of these radionuclides is accurate. A critical component in quality control is the collection of multiple spectra for each sample.

Verification of radionuclide identification is challenging because of the complexity of the spectra that contain a broad array of fission products at short times. However, by analyzing multiple spectra, the nuclide identification can be determined with greater confidence (Metz et. al. 2009). The activity of a radionuclide identified in multiple spectra can be decay-corrected back to the time of irradiation. When the decay-corrected activity is constant across multiple spectra, the radionuclide has been correctly identified. Figure 3.5 is an example of this type of data analysis for a boron carbide shielded pulse of ^{235}U. The radioisotopes ^{91}Sr and ^{92}Sr, which have 9.5 h and 2.7 h half-lives, respectively, were observed at various times. The data in Figure 3.5 form horizontal lines, which indicate that the correct isotopes have been identified and that the peaks used are free from gamma interferences.

Slow leakage of fission product gases from the Tedlar® was detected by quantifying the gaseous fission products and their daughters over time. This explains the abnormally low values at mass 138 in Figure 3.3. These two data points correspond to ^{138}Xe and its daughter ^{138}Cs. Since the containment of the gaseous Xe was incomplete, the resulting production of its daughter was affected. To enable quantitative analysis of gaseous fission products and their daughters, alternative containment designs must be considered.
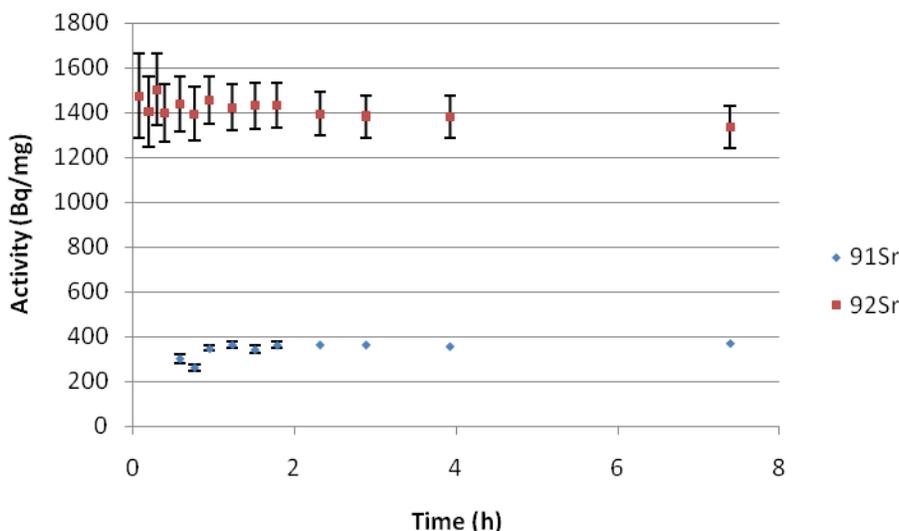
Figure 3.5. Decay-Corrected Counts to Time Zero of ^{91}Sr ($t_{1/2}$ = 9.5 h) and ^{92}Sr ($t_{1/2}$ = 2.7 h) at Various Times After Irradiation of ^{235}U Under Boron Carbide.

Once the various radionuclides have been identified and quantified according to the procedures above, data can be plotted as fissions/µg vs. mass. Plotting the data in this way provides an additional mechanism to verify data quality. To generate the plots, experimental data is compared to published fission yields (England and Rider 1993) and the number of fissions generated is calculated from each radionuclide quantified. The number of fissions generated should be constant and independent of the nuclide used in the calculations. Figure 3.6 is an example of a fissions/µg plot for an irradiation of ^{239}Pu under B_4C. Individual nuclides are represented by points, with blue diamonds showing data collected at WSU and red triangles representing data collected at PNNL. The corresponding colored lines are the average of the respective data set. Data collected at both locations is clustered around a central value with errors in the range of 10 to 15%. Comparison of the number of fissions calculated from each data source (PNNL vs. WSU) and MCNP modeling provides further confidence of data validity since all numbers agree within a few percent. These statistics are typical of all the experiments performed. The data in Figure 3.6 show a high degree of internal consistency, indicating the high quality of the data and the fact that the boron carbide shield is performing according to expectations.
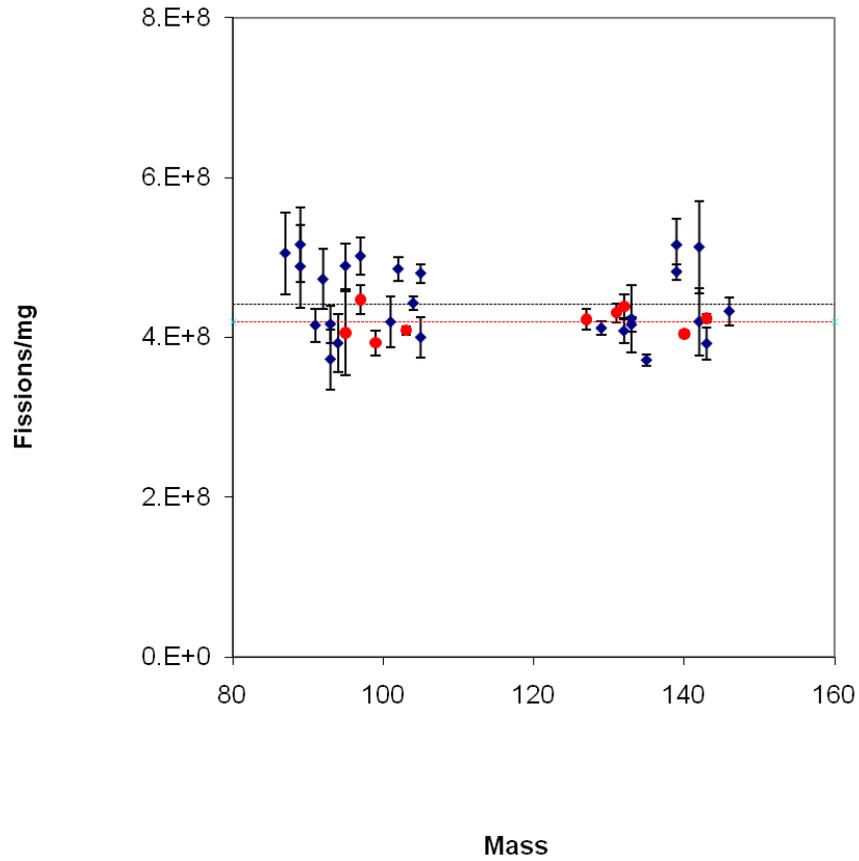
Figure 3.6. Example of Data Consistency for [239]Pu Fission Spectrum Irradiation. Blue diamonds are data collected at WSU immediately after the pulse, with the blue line being the average. Red triangles are data collected after transport to PNNL, with the red line being the average.

4.0   Conclusions

Gamma spectra of the fission products of $^{233, 235, 238}$U, ^{237}Np, and ^{239}Pu were collected beginning four minutes after neutron pulses at thermal, epithermal, and fission energy spectra. A wide array of fission products were observed, and the data were shown to be consistent and of high quality. The fission products were in agreement with published values. Fission product gases were lost from the heat-sealed Tedlar® containment. For complete capture of fission product gases for quantitative analysis, alternate containment must be designed. Gamma spectra were also collected following neutron irradiation of ^{238}U using a 14-MeV neutron generator. The sample size and irradiation time were increased for the 14 MeV experiments, but unfortunately only resulted in samples with ~1.5×10^7 fissions or less due to the low neutron fluence of the generator.

5.0 References

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