

Advancement of Computational Nuclear Fuel and Safeguard Tools

A Thesis Presented for the

Master of Science

Degree

The University of Tennessee, Knoxville

Nathan Shoman

May 2016

© by Nathan Shoman, 2016
All Rights Reserved.

*Dedicated to my parents, who encouraged me to always push forward no matter the
challenge*

Acknowledgments

I would like to thank my parents, S.A. & E.T. Shoman, who always encouraged me to keep fighting no matter the challenge. My high school calculus teacher, Mrs. Maggie. G, who kept pushing me and taught me that I could handle the rigors of my dreams. My advisors Dr. Steven Skutnik and Dr. Jamie Coble whose patience and wisdom instilled the skills within me to become a young engineer. Finally, I would like to thank my best friends Andrea C, who helped me to enjoy the little things despite the struggles of life, Blake B, who has shared in the many joys and sorrows of my life as my brother, and to the friends that made Knoxville home.

Abstract

This thesis work was conducted in two separate but related areas: reactor physics and nuclear fuel safeguards. Reactor physics calculations at the assembly level are important to understand the buildup of fission products and major actinides. These calculations can be used to determine the content of a spent fuel assembly prior to reprocessing or disposal at a much cheaper price. This thesis presents an extension to the SCALE (A Nuclear Systems Modeling & Simulations Suite) by developing a lattice model and cross-section library for the CANada Deuterium Uranium (CANDU) 37 and 28 element assemblies. The previous cross-section library was based on the older ENDF/B-V.2 data for which there was no pre-existing lattice template. The modernized library and creation of a new lattice are intended to provide users with an accurate means of evaluating depletion of CANDU fuel assemblies by using the lattice with TRITON/NEWT or TRITON/KENO, or by using the problem-dependent cross-section library with ORIGEN. The new problem dependent cross-section library performs similarly or better to the old cross-section library when benchmarked to radiochemical assay data. The development of the new lattice revealed discrepancies in the Europium-154 thermal capture cross-section in the ENDF/B-VII.0 evaluation compared with the ENDF/B-VI.8 evaluation, which is analyzed in detail. This thesis also presents benchmarks for SCALE's existing PWR templates. Many studies have shown the high degree of accuracy of the SCALE system, however, there is little work to show the performance of the unmodified templates. This study shows the templates perform well when reproducing the destructive assay results

of samples that are located away from features within the assembly that adversely change the spectrum, such as burnable absorbers or guide tubes. Finally, this thesis presents results of the experimental performance of the Multi-Isotope Process monitor (MIP), an online, non-destructive method for safeguards of reprocessing facilities. The system detects changes in the characteristic gamma spectra at each stage in reprocessing using multivariate techniques. The results show that the MIP monitor can distinguish between Mixed Oxide fuels (MOX) and traditional Used Nuclear Fuel (UNF). Additionally, the monitor can distinguish between different UNF materials or different MOX materials.

Table of Contents

1	Introduction	1
2	Development of modern CANDU PHWR cross-section libraries for SCALE	3
2.1	Abstract	4
2.2	Introduction	5
2.3	Motivation	6
2.4	Experimental Assay Data	7
2.4.1	Bruce-A Fuel Nuclide Inventory	7
2.4.2	Pickering-A Fuel Nuclide Inventory	8
2.5	Lattice Development	10
2.5.1	Dancoff Factors	12
2.5.2	Model parameters	14
2.6	Lattice physics validation studies	16
2.6.1	28-element bundle benchmark	17
2.6.2	37-element bundle benchmark	19
2.6.3	Europium inventory production	21
2.7	ORIGEN library performance comparison	27
2.7.1	28-element library benchmarks	27
2.7.2	37-element ORIGEN library benchmark	31
2.8	Conclusion	31

2.8.1	Effect of cross-section libraries on isotopic benchmarks as evaluated in lattice physics transport models	32
2.8.2	Performance of the new SCALE-based lattices compared to the prior WIMS-AECL library	33
2.9	Acknowledgments	35
3	Verification and Validation of New Unmodified SCALE PWR Lattice Templates	36
3.1	Abstract	37
3.2	Introduction	38
3.3	Procedure	41
3.3.1	Template Validation	41
3.3.2	Multivariate Analysis	41
3.4	Results	45
3.4.1	Westinghouse 15x15	45
3.4.2	Westinghouse 17x17	47
3.4.3	Combustion Engineering 14x14	48
3.4.4	Babcock & Wilcox 15x15	48
3.4.5	Multivariate Analysis	51
3.5	Conclusions	53
4	Experimental Performance of the Multi-Isotope Process Monitor	55
4.1	Abstract	56
4.2	Introduction	56
4.3	Experimental Data Collection	58
4.4	Methods	59
4.4.1	Background Subtraction	60
4.4.2	Spectral Alignment	61
4.5	Data Analysis	64
4.5.1	Discrimination between UNF and MOX fuel samples	64

4.5.2	Classification of UNF samples with dissimilar Uranium isotope distribution	66
4.5.3	Classification of MOX samples with dissimilar Plutonium isotope distribution	68
4.5.4	Discrimination of mixtures for detection threshold	69
4.5.5	Observation of material transients	72
4.6	Conclusions	73
4.7	Future Work	74
4.7.1	Need for global alignment	74
4.7.2	Investigation in conjunction with chemical analysis	76
4.7.3	Application of Q-stat discrimination	77
5	Conclusions and Final Remarks	78
	Bibliography	80
	Appendix	88
A.1	CANDU 28-element input for lattice validation (TRITON/NEWT)	90
A.2	CANDU 37-element input for lattice validation (TRITON/NEWT)	94
A.3	CANDU lattice and ORIGEN cross-section library E/C values	100
B.1	LWR unmodified template E/C values	104
B.1.1	Westinghouse 15x15	104
B.1.2	Westinghouse 17x17	105
B.1.3	Combustion Engineering 14x14	107
B.1.4	Babcock & Wilcox 15x15	108
	Vita	110

List of Tables

2.1	Power history used for modeling the Pickering-A CANDU 28-element bundle (with corrected final decay time) for the nominal assembly power and the calculated outer element power [1, 2].	10
2.2	Power history used for modeling the Bruce-A CANDU 37-element bundle (with corrected final decay time) [1, 2].	10
2.3	Geometry specification for CANDU 28-element bundle lattices. Phase denotes the angular position of the first element in the assembly, wherein each subsequent element is offset by angle $\Delta\theta$	11
2.4	Geometry specification for CANDU 37-element bundle lattices. Phase denotes the angular position of the first element in the assembly, wherein each subsequent element is offset by angle $\Delta\theta$	11
2.5	Calculated Dancoff Factors for CANDU 28-element and 37-element assemblies	13
2.6	Modeling parameters used for both CANDU assembly lattices	14
2.7	Reaction-based gain and loss channels for ^{154}Eu , based on the ORIGEN transition matrix for a CANDU 28-element assembly at the start of irradiation ($0 \frac{\text{MWd}}{\text{MTU}}$). (Note that only the 5 most significant transitions are given.)	23

2.8	Reaction-based gain and loss channels for ^{155}Eu , based on the ORIGEN transition matrix for a CANDU 28-element assembly at the start of irradiation ($0 \frac{\text{MWd}}{\text{MTU}}$). (Note that only the 5 most significant transitions are given.)	23
A.1	CANDU 37-element E/C values	100
A.2	CANDU 28-element nominal power NEWT E/C values	100
A.3	CANDU 28-element nominal power ORIGEN E/C values	101
A.4	CANDU 28-element outer power ORIGEN E/C values	102
B.1	H.B. Robinson (Westinghouse 15x15) calculated values	104
B.2	Turkey Point (Westinghouse 15x15) calculated values	104
B.3	Takahama (SF95) (Westinghouse 17x17) calculated values	105
B.4	Takahama (SF97) (Westinghouse 17x17) calculated values	106
B.5	Calvert Cliffs (Combustion Engineering 14x14) calculated values . . .	107
B.6	Three Mile Island ORNL assay (Babcock & Wilcox 15x15) calculated values	108
B.7	Three Mile Island ANL assay (Babcock & Wilcox 15x15) calculated values	109

List of Figures

2.1	Final holes-based geometries for CANDU assembly lattices	12
2.2	CANDU infinite-lattice multiplication values (k_∞), as a function of burnup. Error bars are too small to be shown; Δk_∞ on the order of 1 pcm for TRITON/NEWT and 100 pcm for TRITON/KENO.	15
2.3	CANDU 28-element outer-ring depletion benchmark (based on Pickering-A outer-ring data using the nominal assembly power history) for major actinoids [2, 1]	18
2.4	CANDU 28-element outer-ring depletion benchmark (based on Pickering-A outer-ring data using the nominal assembly power history) for fission products and minor actinoids [2, 1]	19
2.5	CANDU 37-element bundle depletion benchmark (based on Bruce-A assembly data) for major actinoids [2, 1].	20
2.6	Radiative capture (n, γ) cross-sections for several ENDF library evaluations (left) and capture cross-section ratios to ENDF-VI.8 data (right) for ^{153}Eu , ^{154}Eu , and ^{155}Eu . [3, 4]	24
2.7	Distribution of the 49-group collapsing flux used at the beginning-of-core for a typical PWR assembly (Westinghouse 17x17) versus a CANDU PHWR (28-element) assembly. Note the shift of the neutron population away from the resonance region and toward the thermal region for heavy-water spectra compared to a corresponding light-water reactor spectrum.	26

2.8	Radiative capture (n,γ) cross-sections for several ENDF library evaluations (left) and capture cross-section ratios to ENDF-VI.8 data (right) for ^{154}Sm . [3, 4]	27
2.9	CANDU 28-element ORIGEN outer element power history comparison; major actinoids	29
2.10	CANDU 28-element ORIGEN outer element power history comparison; minor actinoids and fission products	29
2.11	CANDU 28-element ORIGEN library comparison nominal assembly power history; major actinoids. Note that benchmark inventories are rescaled from edge element values (per [2]) to produce an assembly-averaged value.	30
2.12	CANDU 28-element ORIGEN library comparison nominal assembly power history; fission products and minor actinoids	31
2.13	CANDU 37-element ORIGEN library comparison	32
3.1	Example of a PCA model with three variables	43
3.2	Turkey Point and H.B. Robinson (Westinghouse 15x15) plutonium isotopic benchmarks	46
3.3	Takahama unit 3 (Westinghouse 17x17) plutonium isotopic benchmarks	47
3.4	Calvert Cliffs (Combustion Engineering 14x14) plutonium isotopic benchmarks	49
3.5	Three Mile Unit 1 (Babcock & Wilcox 15x15) plutonium isotopic benchmarks - prior ANL evaluation	50
3.6	Babcock & Wilcox 15x15 plutonium isotopic benchmark - new ONRL evaluation	50
3.7	PCA model to compare experimental and calculated values for major actinoids	51
3.8	PLS input and predicted burnup values (based on input isotopics), the green line is the “ideal” prediction where the red is the actual	52

3.9	Significant loading for ^{235}U and ^{239}Pu on latent variables 1 and 2 . . .	53
4.1	Background Spectra at various collection tanks in the H-canyon facility	61
4.2	A variety of background correction methods. Note that some methods do not retain all spectral features	62
4.3	Example of shifting the entire spectra to achieve proper alignment . .	63
4.4	Treatment of non-linearities with RAFFT and simple alignment methods	64
4.5	Discrimination between UNF and MOX materials using a PCA model trained on MOX fuel	65
4.6	Loadings of a PCA model trained exclusively on UNF or MOX material	66
4.7	Cluster analysis in PC space is insufficient to discriminate UNF materials of dissimilar uranium enrichment	67
4.8	Separation of Q scores for different UNF materials	68
4.9	PCA model trained on Alpha and Charlie with Delta transposed to the reduced space	69
4.10	Relative of alignment of Golf to Delta with a simple alignment based on the peak around channel 25	70
4.11	High energy peak is aligned despite not explicitly targeted	71
4.12	PCA model trained on a pure golf material with a variety of UNF mixtures	72
4.13	Changing peak ratios in gamma spectra of mixed UNF materials . . .	73
4.14	Q residuals for a PCA model trained on the first 100 channels of a pure golf material	74
4.15	Cycling of Delta, first 10 minutes are observations of a non-fuel material	75
4.16	Possible peak mismatch at channel 40	76

Chapter 1

Introduction

This thesis has three main sections: Development of modern CANDU PHWR cross-section libraries for SCALE, Verification and Validation of New Unmodified SCALE PWR Lattice Templates, and Experimental Performance of the Multi-Isotope Process Monitor. This thesis provides analysis to advance the computational tools for reactor physics and safeguards. The first analysis develops a set of CANDU lattices and cross-section libraries for the SCALE code system. The previous cross-section library was based on older cross-section evaluations (ENDF/B-V.2), whereas the lattice was not publicly distributed. These lattices and libraries can be used to accurately predict spent fuel isotopics, which are important for reprocessing or geological disposal. The CANDU assembly has several fixed parameters such as enrichment and water composition that are variable parameters for many PWR assembly types. When performing rapid depletion calculations for other PWR assembly types, such as a Westinghouse 17x17, these parameters can be interpolated. However, many assemblies will possess features such as burnable poisons which are not accounted for in the rapid depletion models. The second study considers the impact of these features on the accuracy of the calculated isotopics. Once spent fuel is discharged from the reactor and the isotopics are determined, it must ultimately reside in a geological repository or be reprocessed. The final part of the thesis covers the Multi-Isotope Process Monitor (MIP), a non-destructive monitoring program to detect diversion and tampering of nuclear material at a reprocessing facility. In this thesis, this tool is shown to accurately discriminate between multiple fuel material types. Additionally, the ability to detect system transients is demonstrated.

Chapter 2

Development of modern CANDU PHWR cross-section libraries for SCALE

This chapter is based on a pre-publication paper by N. T. Shoman and S. E. Skutnik in the journal Nuclear Engineering and Design. My primary contribution to this paper includes: (i) development of CANDU 37-element lattice, (ii) development of CANDU 28-element lattice, (iii) development of CANDU 37-element cross-section library for use with ORIGEN, (iv) development of CANDU 28-element cross-section library for use with ORIGEN, (v) benchmarking for all of the above, (vi) discovery and investigation of ^{241}Pu discrepancies (vii) benchmarking of modified ORIGEN reactor data library with ENDF/B-VI.8 ^{154}Eu cross-sections, and (viii) much of the writing of the paper. I generated the initial cross-section libraries where S. E. Skutnik modified the ^{154}Eu cross-sections which I then benchmarked. S. E. Skutnik wrote Section 2.6.3 and generated all associated plots. He also determined the transition coefficients in Table 2.7.

2.1 Abstract

A new set of SCALE fuel lattice models have been developed for the 28-element and 37-element CANDU fuel assembly designs using modern cross section data from ENDF-B/VII.0 in order to produce new reactor data libraries for SCALE/ORIGEN depletion analyses. These new libraries are intended to provide users with a convenient means of evaluating depletion of CANDU fuel assemblies using ORIGEN through pre-generated cross sections based on SCALE lattice physics calculations. The performance of the new CANDU ORIGEN libraries in depletion analysis benchmarks to radiochemical assay data were compared to the previous version of the CANDU libraries provided with SCALE (based on WIMS-AECL models).

Benchmark comparisons with available radiochemical assay data indicate that the new cross-section libraries perform well at matching major actinide species (U/Pu), which are generally within 1-4% of experimental values. The library also showed similar or better results over the WIMS-AECL library regarding fission product species and minor actinoids (Np, Am, and Cm). However, a notable exception was in

calculated inventories of ^{154}Eu and ^{155}Eu , where the new library employing modern nuclear data (ENDF/B-VII.0) performed substantially poorer than the previous WIMS-AECL library (which used ENDF-B/VI.8 cross-sections for these species). The cause for this discrepancy appears to be due to differences in the ^{154}Eu thermal capture cross-section between ENDF/B-VI.8 and ENDF/B-VII.0, an effect which is exacerbated by the highly thermalized flux of a CANDU heavy water reactor compared to that of a typical light-water reactor spectrum.

Overall, the new SCALE-based CANDU libraries appear to give very good agreement to experimental benchmarks, indicating that the new libraries should provide satisfactory depletion calculation performance generally comparable to or better than the prior WIMS-AECL libraries.

2.2 Introduction

The purpose of this project was to develop a working set of CANada Deuterium Uranium (CANDU) pressurized heavy water reactor (PHWR) fuel lattice models for cross section library production based on modern nuclear data (ENDF/B-VII.0) compatible with the latest SCALE physics modules [5, 6]. Cross section libraries for the CANDU 28-element and 37-element designs were produced for use in the ORIGEN depletion sequence [7].

In this paper, the means of developing these CANDU lattices for reactor data libraries are presented, including details of the lattice construction, validation studies to characterize the performance of the new libraries compared to both radiochemical assay data and the prior version of the library, and refinements made to the model to ensure maximum fidelity of the new lattice model. The computed isotopic inventories using these new models were then checked against previously developed benchmarks based on radiochemical assay data and found to show good agreement for most isotopes.

2.3 Motivation

SCALE includes a number of pre-generated reactor-specific data libraries based upon lattice physics calculations for use with the ORIGEN depletion module [7]. These libraries are intended to allow users to perform rapid depletion calculations without the need to re-run computationally expensive lattice physics transport calculations, namely by providing a set of pre-calculated one-group cross-sections for specific initial assembly configurations at set burnup intervals. These pre-calculated libraries can then be interpolated to problem-specific conditions (e.g., initial enrichment, cycle burnup, etc.) for use with ORIGEN [6, 7]

The reactor data libraries provided with SCALE include most major U.S. light water reactor (LWR) fuel assembly types, as well as MOX-based pressurized water reactor (PWR) and boiling water reactor (BWR) fuel assembly configurations along with several foreign reactor types, such as RBMK, VVER, Magnox, and CANDU PHWR lattice types. These libraries allow for rapid evaluation of fuel depletion problems by interpolating cross-sections calculated from prior 2-D transport calculations to problem-specific conditions (e.g., enrichment, moderator density, and burnup), thus largely preserving solution accuracy.

While current and previous versions of SCALE provide CANDU 28-element and 37-element cross-section libraries for use with ORIGEN, no current lattice templates compatible with the NEWT transport sequence currently exist. Meanwhile, the previous libraries contain a very limited burnup region, ranging from 230 MWd/MTU to 12,480 MWd/MTU [7, 8]. Because no initial zero-burnup cross-sections are provided on the included CANDU libraries, ORIGEN cannot interpolate any data in the 0 to 230 MWd/MTU region with the provided libraries.

Additionally, the previous libraries were generated using the two-dimensional transport code WIMS-AECL using the ENDF/B-V nuclear data [2, 1]. The new libraries have been evaluated using modern nuclear data (ENDF/B-VII.0) and the current SCALE 2-D deterministic transport sequence, TRITON [9]. As part of this

work, templates have been developed that are compatible for both a Monte-Carlo approach via the KENO sequence [10] or a discrete-ordinates sequence via NEWT.

2.4 Experimental Assay Data

Validation of the new CANDU reactor data libraries for ORIGEN was conducted by comparing calculated inventories from depletion to known radiochemical assay benchmark studies for CANDU fuel assemblies. Measurements were available for both Pickering-A and Bruce-A fuel assembly nuclide inventories. Available radiochemical assay data includes studies on CANDU assemblies from two reactors: Bruce-A, a 37-element assembly benchmark which was limited to uranium and plutonium isotopic ratios, and Pickering-A, which included several important fission products and minor actinoids [1, 2].

The Bruce-A assay provides measurements from each fuel ring in the assembly which can be used to construct assembly average isotopic values. However, the Pickering-A assay only includes isotopic measurements from the outer fuel ring. These results are not directly applicable to benchmarking the homogenized one group cross-sections. A number of different benchmarks are provided in an attempt to provide an adequate comparison.

2.4.1 Bruce-A Fuel Nuclide Inventory

Isotopic measurements on fuel bundle F21037C from the Bruce-A Nuclear Generating Station were obtained as part of the Thorium Fuel Reprocessing Experiment Program conducted by the AECL in 1982 [2]. The 37-element CANDU fuel design consists of a single central pin surrounded by three concentric rings of fuel pins, each with 6, 12, and 18 pins, respectively. The assay was taken from multiple pins from different rings within the bundle. These measurements were combined to determine assembly-averaged inventories. The central fuel pin was not assayed, however, it was assumed

to have the same fuel composition as the inner ring [1, 2]. The discharge burnup of the bundle was approximately 7980 MWd/kgU.

2.4.2 Pickering-A Fuel Nuclide Inventory

For the 28-element assembly benchmark, a single fuel assay was performed on an outer element of Pickering-A fuel bundle 19558C [2]. The measurements were made to provide a benchmark base for codes used to predict used fuel inventories for the Waste Management Program at AECL. The bundle consisted of 28 natural uranium oxide pellets, each surrounded by Zircaloy-4 cladding. The bundle had a discharge burnup of 9208 MWd/kgU.

The original benchmarking study reported that the relatively large discrepancies in ^{99}Tc and ^{106}Ru could be attributed to significant quantities remaining in undissolved residue after recovery, indicating this could likely be due to poor counting geometries used in analyzing the residues [1]. The report further indicated that errors in ^{129}I could similarly be attributed to measurement issues resulting from the difficulty in capturing off-gasses during the sample dissolution.

The WIMS-AECL lattice evaluations employed ENDF/B-V for fission yields whereas nuclear decay and energy release data was based on ENDF/B-VI. The original WIMS-AECL library was a hybrid containing ENDF/B-V cross sections, supplemented with ENDF/B-VI cross sections for a number of nuclides not available in ENDF/B-V. Additionally, ENDF/B-VI cross sections were used for ^{154}Eu and ^{155}Eu due to large differences in the resonance energy region [1].

The ratio of the WIMS-AECL cluster-averaged inventories to outer element inventories was used to scale the measured data in order to obtain an estimate of the cluster-averaged inventory. The scaled fission products were not reproducible using this method, so the WIMS-AECL scaled values were used instead of TRITON/NEWT scaled values from the new lattice. The ratio by which the inventories are scaled was not provided in the primary report. Caution must be used as using this method

introduces a bias. If the ratios of the outer inventory to the cluster-averaged values are incorrect for any reason it will impact the comparison. This comparison is nonetheless considered due to a lack of available cluster-averaged data. It should be noted that not all of the fission products were scaled. Nuclides with a small capture cross section and for which the dominant production route is fission yield will not be significantly impacted by error induced by assembly-averaged cross-sections and flux.

Discrepancies in Pu-241 inventories due to errors in reported decay times

Originally, the ^{241}Pu inventories for both assemblies were incorrect. The 37-element bundle ^{241}Pu inventory was under-predicted by 35-40% while the 28-element ^{241}Pu inventory was over-predicted by approximately 70%. It was eventually determined that there was an error in the radiochemical benchmark reference paper [1] used to evaluate these models. The original benchmark report [1] reported a final decay time after discharge of 5590 days for the the 37-element Bruce-A assembly (Table 20 in the original report), while the 28-element Pickering-A assembly had a decay time prior to measurement of 1162 days (Table 23 of the original report). It was not possible to reproduce their results with this data. However, a separate study referencing the radiochemical assay performed on the Pickering-A (28-element) fuel assembly [8, 2] contradicts this decay time, indicating a final decay step of 5590 days prior to measurement (rather than 1162 days), implying that the decay times for the two assemblies had inadvertently been reversed. When these changes are made in the TRITON model, the models showed substantially improved agreement with the isotopic benchmark, particularly for ^{241}Pu . The correct decay time was confirmed via private correspondence with the author of the original benchmarking study.

The corrected power histories for each assembly are presented in Tables 2.1 and 2.2. Two power histories are provided for the 28-element lattice, one for the assembly power history and an inferred power history reported for the outer element [1]. This outer element power history was used with the WIMS-AECL transport sequence to control the specific burnup of the outer element for the 28-element bundle, due to

the fact that the radiochemical assay benchmark was based upon measurements of a single outer-element pin. This history is thus provided here in addition to the nominal assembly power history for benchmarking purposes.

Table 2.1: Power history used for modeling the Pickering-A CANDU 28-element bundle (with corrected final decay time) for the nominal assembly power and the calculated outer element power [1, 2].

Irradiation (days)	Power ($\frac{\text{MW}}{\text{MTU}}$)		Cooling (days)
	Assembly nominal	Outer element	
31.33	28.41	31.4	3.25
15.38	26.26	28.9	5.61
66.67	28.15	31.1	0.00
66.67	26.71	29.5	9.00
64.55	26.07	28.7	15.0
70.78	24.18	26.7	5590.0

Table 2.2: Power history used for modeling the Bruce-A CANDU 37-element bundle (with corrected final decay time) [1, 2].

Irradiation (days)	Power ($\frac{\text{MW}}{\text{MTU}}$)	Cooling (days)
31.63	1.040	0.0
13.00	30.10	10.0
43.25	30.21	55.3
49.75	30.30	9.0
50.33	30.30	15.0
103.00	30.30	3.0
117.25	0.870	1162.0

2.5 Lattice Development

Because NEWT does not support triangular-pitch repeated-element arrays, modeling the CANDU assemblies as a true repeated-element lattice proved impossible. Alternative geometries (such as hexagonal-pitch arrays) are unable to correctly represent the non-uniform radial spacing between circular arrays of elements; i.e.,

Table 2.3: Geometry specification for CANDU 28-element bundle lattices. Phase denotes the angular position of the first element in the assembly, wherein each subsequent element is offset by angle $\Delta\theta$.

Ring	r (cm)	$\Delta\theta$	Phase
Inner	1.175	90	45.0
Middle	2.685	45	22.5
Outer	4.229	22.5	11.25

Table 2.4: Geometry specification for CANDU 37-element bundle lattices. Phase denotes the angular position of the first element in the assembly, wherein each subsequent element is offset by angle $\Delta\theta$.

Ring	r (cm)	$\Delta\theta$	Phase
Center	0	–	–
Inner	1.4885	60	0.0
Middle	2.8755	30	15.0
Outer	4.3305	20	0.0

the distance from the inner ring to the center ring is not the same as that from the middle ring to the outer ring.

Ultimately, the only available modeling solution was to insert each individual fuel element into the model as holes. Coordinates for each hole were determined by establishing the ring radius and the angular spacing ($\Delta\theta$) between each element (assuming an equal angular spacing of elements for each ring), along with an appropriate angular offset between rings (i.e., a “phase” angle). Determining the correct “phase angle” between elements of individual rings is necessary to ensure the correct line-of-sight of fuel elements between rings, which will influence the result (e.g., as indicated by the Dancoff Factors, discussed in the next section). The relationships for the ring positions, such as the ring radius, angular offset between elements ($\Delta\theta$), and the “phase” of each ring (defined as the angular position of the first element in the ring) are presented for the 28 and 37-element assemblies in Tables 2.3 and 2.4, respectively.

The final 28-element and 37-element lattice models are shown as Figures 2.1(a) and Figure 2.1(b). To maximize the accuracy of the multi-group transport (NEWT) solution, a tighter mesh was imposed inside the pressure tube and calandria along with within the fuel elements themselves. Further, to ensure a correct representation of each assembly’s relative orientation, the final models (Figures 2.1(a) and (Figure 2.1(b)) were likewise compared to available images of each assembly’s orientation [2, 1, 11].

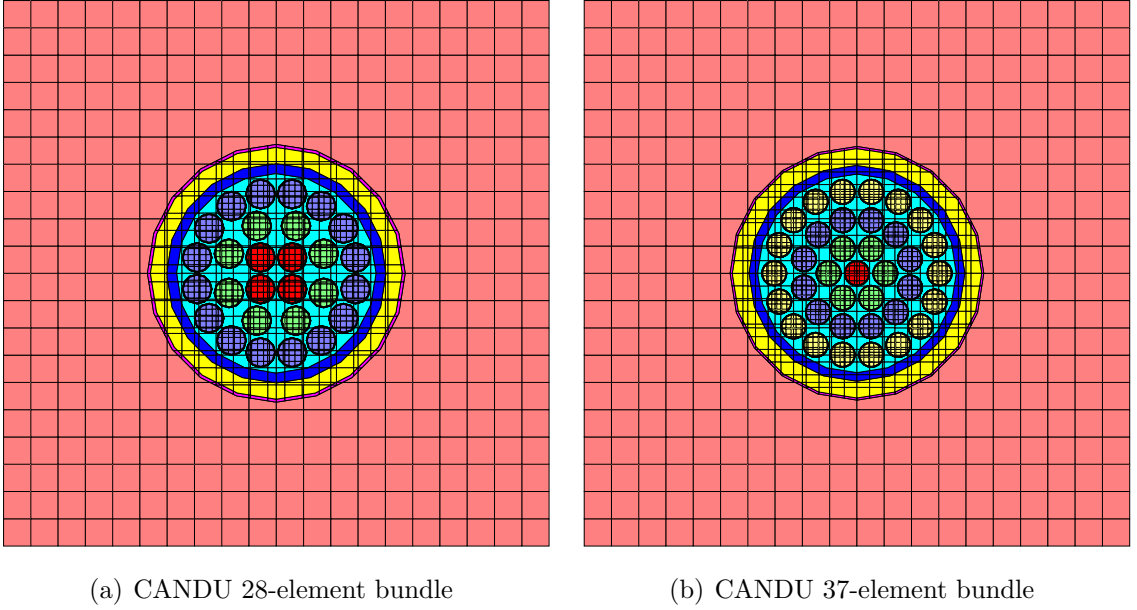


Figure 2.1: Final holes-based geometries for CANDU assembly lattices

2.5.1 Dancoff Factors

With the correct geometry model for the fuel assemblies established, it is important to determine the Dancoff factors for multi-group deterministic transport calculations (like NEWT). The Dancoff factor is the probability that a neutron emitted isotropically from the surface of one absorber lump will pass through the external media and enter a nearby absorber lump [12]. (Here it should be noted that Dancoff Factors apply exclusively to multi-group transport calculations; such a correction is not

Table 2.5: Calculated Dancoff Factors for CANDU 28-element and 37-element assemblies

Position / Ring	Dancoff Factor	
	28-element	37-element
Center pin	–	0.763
Inner ring	0.714	0.761
Middle ring	0.707	0.728
Outer ring	0.447	0.442

required for continuous-energy Monte Carlo approaches like KENO.) Dancoff factors have a significant impact on transport calculation (including for isotopic calculations), given the relatively heterogeneous environment of the CANDU fuel assembly. This finding is consistent with prior studies evaluating impacts on other heterogeneous assembly types, such as SVEA BWR assembly types [13]. The study found that differences in Dancoff factors can have large impacts in the effective cross sections, and thus, the isotopic calculations. It was also noted that at higher burnups Dancoff factors are particularly important. Deviations of 8% for ^{235}U and 3-5% in plutonium isotopics were seen when Dancoff factors were not used.

Dancoff factors were analyzed to using the MCDancoff module to determine a ring-averaged Dancoff factor. Included in Table 2.5 are the calculated ring-averaged Dancoff Factors for both the 28-element and 37-element CANDU assembly types. From Table 2.5, one apparent feature of both lattices is the fact that the outer-ring Dancoff factor is substantially lower than those of the inner bundle rings (indicating a relatively higher probability of absorption). Here, this can be understood with respect to the average moderation of neutrons in the outer-ring elements compared to the inner rings. Neutrons created in other bundles will be highly thermalized; likewise, given the solid angle subtended by the outer ring elements, they have a comparatively high rate of absorption in the outer elements. This higher rate of absorption will thus manifest as a comparatively higher power in the outer ring elements compared to the bundle average.

2.5.2 Model parameters

The modeling parameters used for transport and depletion calculations are presented in Table 2.6. In order to achieve maximum accuracy in isotopic predictions, four depletion subintervals were used per depletion cycle (**nlib=4**). Trace levels of 230 nuclides were added to the fuel mixtures (**addnux=3**) in order to accurately account for the effect of key isotopes on depletion calculations (especially at higher burnups). Parameters specific to multi-group transport included the convergence criteria (ϵ), P_N scattering order, and coarse-mesh finite differencing (CMFD, used to accelerate source convergence).

Table 2.6: Modeling parameters used for both CANDU assembly lattices

Parameter	Value	Description
addnux	3	Add trace levels of 230 nuclides to the fuel mixture composition at the start of depletion (to ensure inclusion in the first cross-section update)
ϵ	8.0E-4	Source convergence criteria for multi-group deterministic transport
CMFD	yes	Coarse-mesh finite differencing acceleration
xycmfd	4×4	Four fine mesh cells per coarse mesh cell (for CMFD acceleration)
P_N	2 / 1	P_N scattering order; set to 2 for moderator and coolant, 1 for all other materials
nlib	4	Number of depletion sub-intervals per irradiation cycle

In order to accurately validate the model, results from both deterministic and Monte Carlo methods were compared to known assay data. Monte Carlo results were obtained by using the KENO transport model and the discrete ordinates results were obtained using TRITON/NEWT. Additionally, the effect of employing ENDF/B-V versus ENDF/B-VII.0 cross-section data for the models was compared. One benchmark used to compare the agreement of the KENO and NEWT models was to compare their k_∞ value as a function of the assembly irradiation time, shown in

Figures 2.2(a) and 2.2(b). This helped to rule out the transport solution as the cause of any discrepancies in isotopic benchmarks.

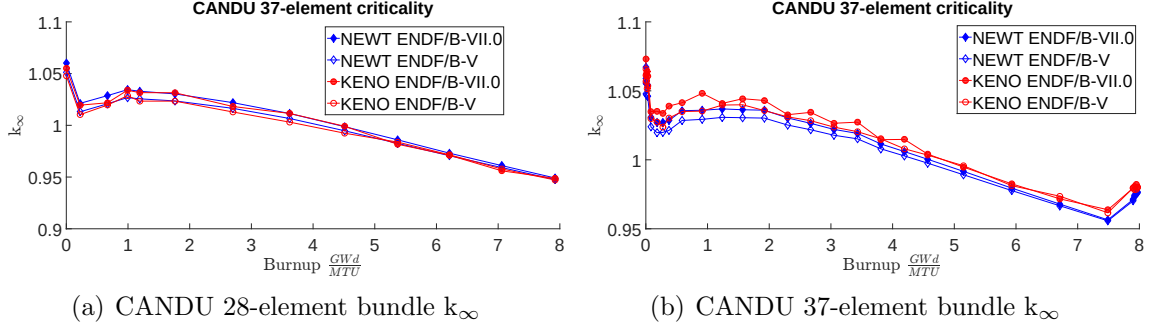


Figure 2.2: CANDU infinite-lattice multiplication values (k_∞), as a function of burnup. Error bars are too small to be shown; Δk_∞ on the order of 1 pcm for TRITON/NEWT and 100 pcm for TRITON/KENO.

Effect of cross-section processing options and modeling assumptions on isotopic benchmark agreement

Because the problem in the reported power histories from the original benchmark studies [1] was not immediately apparent, several avenues were also explored to explain the discrepancies in the isotopic benchmark, particularly with respect to ^{241}Pu . This included evaluating alternative cross-section processing options for their effect on discharge isotopics [14] along with minor modeling adjustments in materials.

Prior work evaluating cross-section processing options with ENDF/B-VII.0 libraries in SCALE considered their effect on calculated discharge isotopics [14]. The study found that of several cross section processing options, only `PMC_OMIT` (which determines whether non-fuel materials are included in pointwise resonance self-shielding calculations) had any significant impact on the results. Other processing parameters, such as `xs_dilute` (i.e., infinitely-dilute treatment of fuel materials), while important for criticality benchmarks, had impacts on calculated discharge isotopics of $< 0.2\%$. The largest contribution to a change in the inventories arose from using the NITAWL (Nordheim Integral Treatment and Working Library

Production) cross-section processing approach (limited to ENDF/B-V-based libraries) as opposed to the default CENTRM (**C**ontinuous **E**nergy **T**ransport **M**odule) approach, consistent with prior investigations on the effects of cross-section processing on isotopic inventories [15].

Further, additional modeling details were evaluated in an attempt to root out the source of discrepancy, including changing the gas in the air gap from standard atmosphere to helium, decreasing the mesh size, and increasing the number of sides used to approximate cylindrical surfaces.

All of the above-listed approaches had minimal impact, with none of these adjustments providing significant change in the inventories. Ultimately the error in the reported decay times was discovered in the original reference paper [1] and determined to be the primary source of discrepancies between the model and the isotopic benchmark.

2.6 Lattice physics validation studies

The primary method of validation for the model was to compare the computational results from the developed model to measured isotopic data from radiochemical assay experiments. Two such studies were available [1, 8], and as such, these studies were used to benchmark the performance of the new fuel bundle lattice models. Further, the new lattice models were similarly benchmarked against the performance of the prior WIMS-AECL model (which served as the basis for the prior CANDU-based cross-section libraries distributed with SCALE) by comparing their isotopic calculations the WIMS model [2]. The WIMS-AECL lattice was unavailable for use so reported values were used in lieu of calculated values. Additional E/C tables and input files can be found in appendix A.

2.6.1 28-element bundle benchmark

A single benchmark based on transport calculations are conducted for the 28-element lattice. The aim of the transport benchmark is to test the accuracy of the lattice parameters such as geometry, materials, and Dancoff factors. The library benchmarks objective is to determine the performance of the ORIGEN library produced from the 28-element lattice.

For the transport benchmark, the outer assembly element isotopics were calculated through TRITON/NEWT and TRITON/KENO by depleting each ring of fuel-elements individually, which was necessary in the multi-group transport solution due to ring-specific Dancoff factors. NEWT and KENO both provide ring-specific isotopic measurements from which the outer ring was compared. This benchmark will indicate how well the lattice geometry performs and determine how well the transport method distributes the power among the rings. For the first benchmark (referred to hereout as the “nominal” benchmark), the nominal assembly power history in Table 2.1 was used for the total assembly power, where power in the individual depletion materials was allowed to float (conserving total assembly power). Here, the isotopic benchmark was calculated by comparing the outer ring inventory to the measured outer ring inventory from the radiochemical assay studies.

Nominal assembly power benchmark

For the nominal assembly power benchmark, the assembly was depleted using the average assembly power, where powers of individual assembly elements were allowed to float, preserving total assembly power (i.e., the assembly power was normalized to the nominal power history expressed in Table 2.1). Validation was performed by comparing the calculated outer ring isotopic inventory to the measured outer ring inventory. The benchmark was run with Monte Carlo (KENO) and Discrete Ordinate (NEWT) transport using ENDF/B-V and ENDF/B-VII.0 cross-sections. An ENDF/B-VI.8 benchmark was also conducted with the new lattice to evaluate

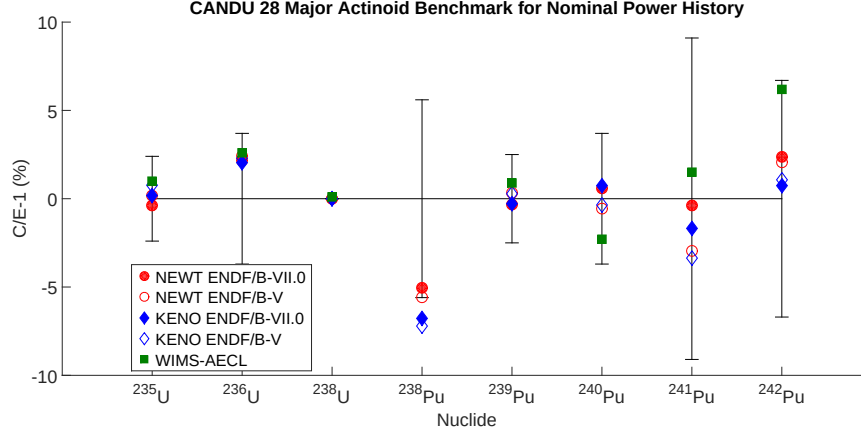


Figure 2.3: CANDU 28-element outer-ring depletion benchmark (based on Pickering-A outer-ring data using the nominal assembly power history) for major actinoids [2, 1]

the effect of cross-section libraries on the production of the europium isotopes ^{154}Eu and ^{155}Eu , but only using the NEWT transport sequence.

Both ENDF/B-V.2 and ENDF/B-VII.0 produced similar results for the major actinoids. However, ENDF/B-VII.0 showed slightly improved agreement to the benchmark values, which is consistent with prior comparisons for PWR assemblies [15]. Although many of the minor actinoids and fission products showed comparatively high error, several of the errors can be explained by issues with the experimental procedure described in Section 2.4.2. However, there remains good agreement with key burnup indicators, such as ^{137}Cs and ^{134}Cs (Figure 2.4), thus indicating that the transport-calculated flux is likely accurate. It is important to note that unlike both ENDF/B-V.2 and ENDF/B-VII.0, ENDF/B-VI.8 did produce accurate values for ^{154}Eu .

Many of the minor actinoids and fission products were not available for comparison with the WIMS-AECL code, due to missing nuclides in the WIMS-AECL library. With respect to the measured isotopes, the new 28-element lattice appears to produce accurate inventories for the major actinides (Figure 2.3) regardless of the cross-section library used. There was a significant improvement in ^{237}Np accuracy over WIMS-AECL in the new lattice, as well as ^{238}Pu .

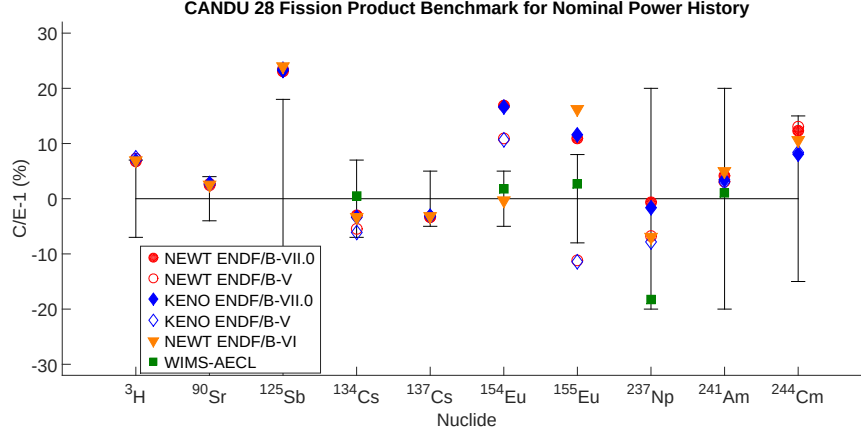


Figure 2.4: CANDU 28-element outer-ring depletion benchmark (based on Pickering-A outer-ring data using the nominal assembly power history) for fission products and minor actinoids [2, 1]

Here, accurate assessment of many of the transuranic species is especially important for used nuclear fuel disposition studies; ^{237}Np is a nuclide of particular concern from a modeling standpoint for geologic disposal evaluations; under certain geochemical conditions, ^{237}Np can act as a significant dose contributor, given its relatively high solubility and mobility in groundwater, while ^{244}Cm acts as a significant neutron source for used fuel storage.

2.6.2 37-element bundle benchmark

Here, the most accurate experimental data were for the uranium and plutonium atom ratios. Like the Pickering-A bundle, the uranium and plutonium ratios were selected for comparison. The (C/E)-1 values are likewise presented in Figure 2.5. Error bars are presented as one sigma uncertainties of the experimental data. However, unlike the 28-element bundle, a detailed assay of the fission products was not performed on the 37-element bundle, thus an extended isotopic comparison cannot be made.

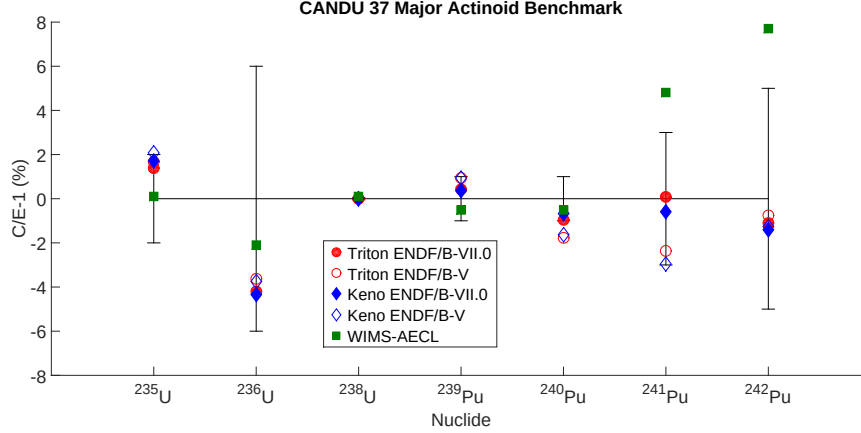


Figure 2.5: CANDU 37-element bundle depletion benchmark (based on Bruce-A assembly data) for major actinoids [2, 1].

Nominal assembly power benchmark

NEWT and KENO produced very similar results for both ENDF/B-VII.0 and ENDF/B-V.2 with the 37-element lattice. The ENDF/B-VII.0 library produced more accurate results for many of the major actinoids, with the exception of ^{236}U . The largest discrepancy between the libraries is less than 3%, which is consistent with prior literature [15]. It should be noted that the 37-element isotopic assay was determined from several samples from each ring and combined into an assembly-average value which enabled an assembly wide comparison.

The most accurate 37-element lattice model involved using the NEWT transport sequence and ENDF/B-VII.0 library. The resulting calculated inventory lies within the experimental uncertainty, with the results for the ENDF/B-V model residing just outside the experimental uncertainty for the plutonium isotopes. The new lattice produced accurate values that were inside experimental uncertainty for all assayed major actinides. This is an improvement over the WIMS-AECL based lattice where ^{241}Pu and ^{242}Pu were calculated outside the experimental uncertainty.

2.6.3 Europium inventory production

The previous WIMS-based 28-element library was able to achieve reasonable prediction of ^{154}Eu and ^{155}Eu by using ENDF/B-VI cross-sections for these isotopes. The ENDF/B-VI.8 library did produce accurate inventory predictions for ^{154}Eu (with a difference of about 1.34%, well within experimental uncertainty), however ^{155}Eu was still predicted higher than the experimental value by about 15%. One plausible explanation for this deviation is due to the evaluated radiative capture cross-sections for Eu species in the thermal region, and in particular the differences in evaluated capture cross-section values between ENDF/B-V.2, VI.8, and VII.0. The ENDF/B-VI.8 cross-sections for ^{154}Eu produced good agreement for ^{154}Eu , however, ^{155}Eu was not produced within the measurement uncertainty bounds for any cross-section library.

To understand this issue, it is first important to understand the production and loss channels for Eu species, which can be represented by the general expression in Equation 2.1.

$$\frac{dN_i}{dt} = \underbrace{\sum_{m=1}^M Y_m \lambda_m N_m}_{\text{(A)}} + \underbrace{\phi \sigma_{i-1}^a N_{i-1}}_{\text{(B)}} - \underbrace{\lambda_i N_i}_{\text{(C)}} - \underbrace{\phi \sigma_i^a N_i}_{\text{(D)}} + \underbrace{Y_i \Sigma_f \phi}_{\text{(E)}} + \underbrace{\sum_{m \neq i} N_m \sigma_{m \rightarrow i} \phi}_{\text{(F)}} \quad (2.1)$$

where

- Term A = Production of nuclide i from M parent isotope decays
- Term B = Production of nuclide i from neutron absorption
- Term C = Loss of nuclide i to decay
- Term D = Loss of nuclide i to neutron absorption

- Term E = Direct production of nuclide i from fission
- Term F = Other neutron-induced reactions (e.g., (n,p), (n,2n), etc.)

The dominant terms controlling the ^{154}Eu and ^{155}Eu inventories are (A), (B), and (D). Given that the direct fission yields of ^{154}Eu and ^{155}Eu from ^{235}U and ^{239}Pu are both vanishingly small, the predominant production channels for both species are therefore through parent isotope decays (e.g., for ^{155}Eu from the decay of ^{155}Sm) as well as through successive neutron capture reactions, i.e. $^{153}\text{Eu} (n, \gamma) ^{154}\text{Eu}$ and $^{154}\text{Eu} (n, \gamma) ^{155}\text{Eu}$. ^{153}Eu , ^{154}Eu , and ^{155}Eu all have relatively high thermal-region neutron capture cross-sections, ranging from about 335 barns for ^{153}Eu [16] to several thousand barns for ^{154}Eu and ^{155}Eu (with even higher capture cross-sections in the resonance energy regions), making capture a predominant production and loss channel for these species.

To understand the relative contributions to nuclide production and loss, it is helpful to refer to the ORIGEN transition matrix $\overline{\overline{A}}$, which describes the total transition rate constant for each set of coupled differential equations describing the system, i.e. [7]:

$$\frac{d\overline{N}}{dt} = \overline{\overline{A}} \cdot \overline{N}(t) \quad (2.2)$$

The transition matrix $\overline{\overline{A}}$ describes the transition rate coefficients for each isotopic transition from neutron reactions and decay. Thus, each individual element of $\overline{\overline{A}}$ (denoted a_{ij}) describes the relative transition rate from nuclide i to j [7]:

$$a_{ij} = -\ell_{ij}\lambda_i - f_{ij}\overline{\phi}\sigma_i \quad (2.3)$$

Where ℓ_{ij} is the decay yield from nuclide i to nuclide j and f_{ij} is the reaction yield, $\overline{\phi}$ is the collapsed one-group flux, and σ_i is the one-group flux-weighted reaction cross-section. Thus, individual transition coefficients can be thought of as relative indicators of the rates of production and loss into individual nuclides. Evaluated this way, it is

evident in Tables 2.7 and 2.8 (which list the most significant gain and loss transition coefficients for ^{154}Eu and ^{155}Eu) that neutron capture is by far the dominant gain and loss channel for both species.

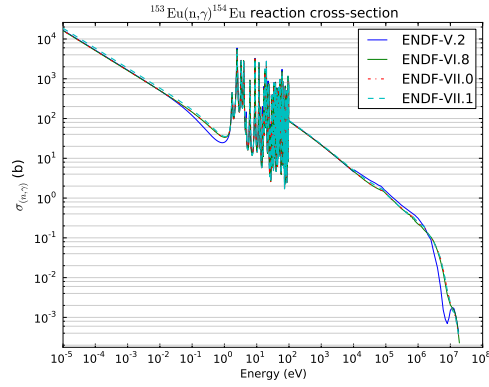
Table 2.7: Reaction-based gain and loss channels for ^{154}Eu , based on the ORIGEN transition matrix for a CANDU 28-element assembly at the start of irradiation ($0 \frac{\text{MWd}}{\text{MTU}}$). (Note that only the 5 most significant transitions are given.)

^{154}Eu gains		^{154}Eu losses	
Reaction	a_{ij}	Reaction	a_{ij}
$^{153}\text{Eu} (n, \gamma) ^{154}\text{Eu}$	1.1038E+02	$^{154}\text{Eu} (n, \gamma) ^{155}\text{Eu}$	6.6322E+02
$^{249}\text{Cf} (n, f) ^{154}\text{Eu}$	3.5425E-03	$^{154}\text{Eu} (n, n) ^{154\text{m}}\text{Eu}$	5.5964E-02
$^{243}\text{Cm} (n, f) ^{154}\text{Eu}$	7.0951E-04	$^{154}\text{Eu} (n, 2n) ^{153}\text{Eu}$	2.1322E-03
$^{242}\text{Am} (n, f) ^{154}\text{Eu}$	6.3834E-04	$^{154}\text{Eu} (n, p) ^{154}\text{Sm}$	6.1756E-06
$^{251}\text{Cf} (n, f) ^{154}\text{Eu}$	3.6207E-04	$^{154}\text{Eu} (n, \alpha) ^{150}\text{Pm}$	4.3990E-08

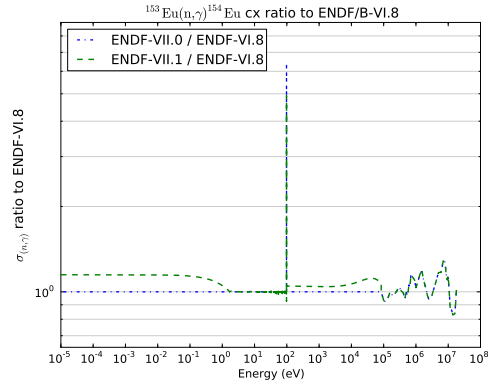
Table 2.8: Reaction-based gain and loss channels for ^{155}Eu , based on the ORIGEN transition matrix for a CANDU 28-element assembly at the start of irradiation ($0 \frac{\text{MWd}}{\text{MTU}}$). (Note that only the 5 most significant transitions are given.)

^{155}Eu gains		^{155}Eu losses	
Reaction	a_{ij}	Reaction	a_{ij}
$^{154}\text{Eu} (n, \gamma) ^{155}\text{Eu}$	6.6322E+02	$^{155}\text{Eu} (n, \gamma) ^{156}\text{Eu}$	1.4438E+03
$^{249}\text{Cf} (n, f) ^{155}\text{Eu}$	4.2343E-02	$^{155}\text{Eu} (n, 2n) ^{154}\text{Eu}$	3.0927E-04
$^{242}\text{Am} (n, f) ^{155}\text{Eu}$	1.0235E-02	$^{155}\text{Eu} (n, 2n) ^{154\text{m}}\text{Eu}$	1.2106E-04
$^{243}\text{Cm} (n, f) ^{155}\text{Eu}$	9.5522E-03	$^{155}\text{Eu} (n, p) ^{155}\text{Sm}$	3.5362E-06
$^{251}\text{Cf} (n, f) ^{155}\text{Eu}$	7.1689E-03	$^{155}\text{Eu} (n, \alpha) ^{152\text{m}}\text{Pm}$	1.6364E-06

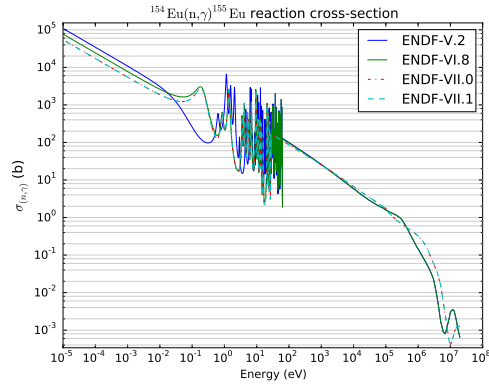
Particularly useful in understanding the miscalculations of Eu species is a comparison of the thermal-region capture cross-sections for ^{153}Eu and ^{154}Eu , shown in Figure 2.6. Here, the $^{153}\text{Eu}(n, \gamma)^{154}\text{Eu}$ cross-section is consistent between ENDF/B-VI.8 and ENDF/B-VII.0 [3, 4]. However the ENDF/B-VII.1 evaluation reports a discrepancy of approximately 11% found with experimental values [17]; thus the 2200 m/s thermal cross-section was corrected from 312 bn to 358 bn in the newest evaluation, consistent with what is seen in Figure 2.6(b). However, given the



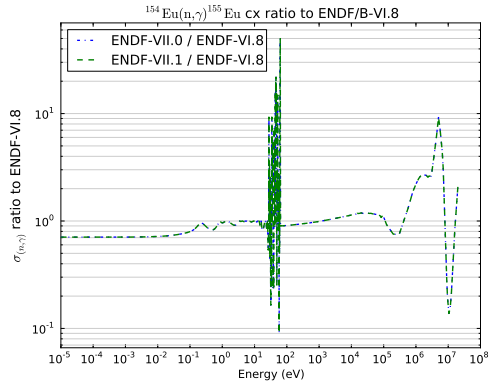
(a) $^{153}\text{Eu}(n,\gamma)^{154}\text{Eu}$ cross-section



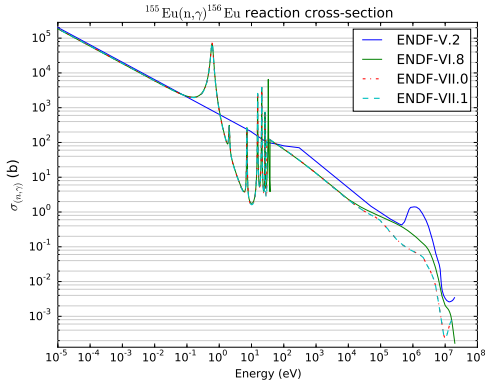
(b) $^{153}\text{Eu}(n,\gamma)^{154}\text{Eu}$ cross-section ratios



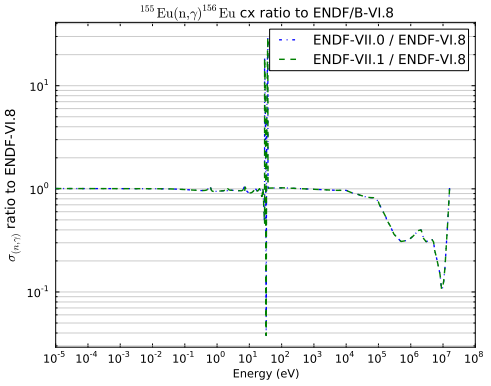
(c) $^{154}\text{Eu}(n,\gamma)^{155}\text{Eu}$ cross-section



(d) $^{154}\text{Eu}(n,\gamma)^{155}\text{Eu}$ cross-section ratios



(e) $^{155}\text{Eu}(n,\gamma)^{156}\text{Eu}$ cross-section



(f) $^{155}\text{Eu}(n,\gamma)^{156}\text{Eu}$ cross-section ratios

Figure 2.6: Radiative capture (n,γ) cross-sections for several ENDF library evaluations (left) and capture cross-section ratios to ENDF-VI.8 data (right) for ^{153}Eu , ^{154}Eu , and ^{155}Eu . [3, 4]

consistency between ENDF-VI.8 and ENDF-VII.0, this can be ruled out as the source of the discrepancy in ^{154}Eu inventories.

Likely at cause for the over-prediction is the $^{154}\text{Eu}(n, \gamma)^{155}\text{Eu}$ capture cross-section, which is approximately 40% lower for the 2200 m/s evaluation for ENDF-VII.0 compared to ENDF-VI.8. The 0.0254 eV cross-section in ENDF/B-VI.8 is based on evaluations by Sekine et. al., indicating a value of 1840 barns for the radiative capture cross-section [18]. The evaluation used in ENDF/B-VII.0 cites Mughabghab for an evaluated thermal capture cross-section of 1348.5 barns [19], however no mention is made of the prior evaluation by Sekine et. al. [18]. Further, File 2 (MF=2) of the ENDF/B-VII.0 library gives a calculated thermal capture cross-section of 1841.6 barns [3], which is 36.6% higher than the evaluated value and consistent with Sekine [18]. Thus, given this and the better observed agreement seen for ^{154}Eu when using the ENDF/B-VI.8 library, it appears likely that the ENDF/B-VII.0 and VII.1 libraries are in error for the ^{154}Eu thermal capture cross-section, which would thus explain the observed discrepancy.

Further compounding this issue is the fact that the highly-thermalized neutron energy spectrum for heavy-water moderators heightens the sensitivity to these cross-section differences compared with light-water spectra, as seen by the representative beginning-of-core assembly-average flux profile shown in Figure 2.7. Here, the average thermal-region neutron population for a HWR is nearly double that of a typical LWR (calculated for a Westinghouse 17x17 PWR assembly). Thus, the heavy-water lattice models are arguably more sensitive to the observed differences in the thermal-region absorption cross-sections (while being subsequently less sensitive to changes in the resonance region).

Meanwhile, ^{155}Eu is not accurately predicted by any of the libraries. Here, the production is too high for ENDF-B/V and too low for ENDF/B-VII.0. Looking at the production of ^{155}Eu via the neutron absorption from the $^{154}\text{Eu}(n, \gamma)^{155}\text{Eu}$ reaction does not appear to immediately explain the discrepancy. ENDF/B-V.2 contains no resonance information; the cross-section is interpolated through the resonance

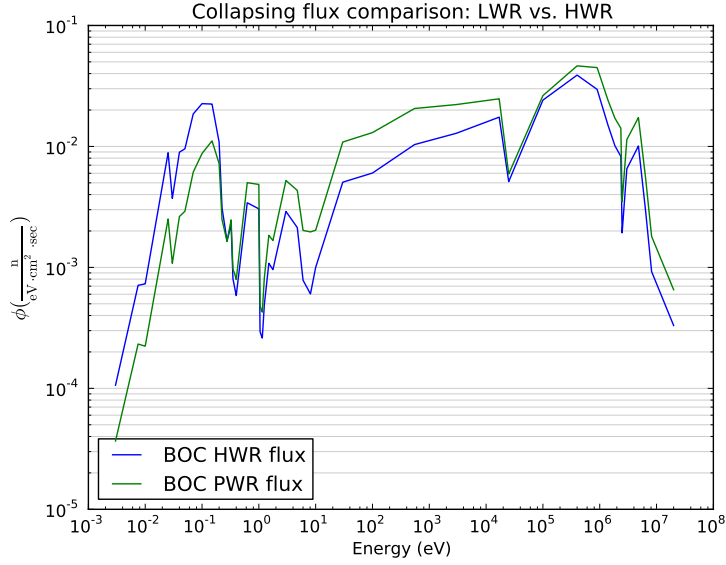


Figure 2.7: Distribution of the 49-group collapsing flux used at the beginning-of-core for a typical PWR assembly (Westinghouse 17x17) versus a CANDU PHWR (28-element) assembly. Note the shift of the neutron population away from the resonance region and toward the thermal region for heavy-water spectra compared to a corresponding light-water reactor spectrum.

region, likely explaining the over-prediction for ENDF-V libraries. Meanwhile, the ENDF/B-VII.0 evaluation reports a thermal capture cross-section of 3,758.4 barns, with a resonance integral of 15,527.5 barns [3]; identical values are reported for ENDF/B-VII.1 [17]. The ENDF/B-VI.8 evaluation reports a thermal capture cross-section of 3,760.22 barns with a resonance integral of 15,288 barns [4]. Given the similarities between the ENDF-VI.8 and VII.0 evaluations, it appears unlikely that the $^{155}\text{Eu}(n,\gamma)^{156}\text{Eu}$ cross-section is at issue, but rather that there may be other production channels at issue (i.e., neutron capture by ^{154}Sm , the dominant loss channel with a transition coefficient of 3.3521 s^{-1} , drives β^- decay by ^{155}Sm into ^{155}Eu). This hypothesis is further supported by the differences in the evaluation of the $^{154}\text{Sm}(n,\gamma)^{155}\text{Sm}$ reaction between ENDF/B-VI.2 and ENDF/B-VII.0 and VII.1, wherein the thermal capture cross-section for the later evaluations are approximately

10% higher than the ENDF/B-VI.2 evaluation, thus driving a larger decay source term by ^{155}Sm into ^{155}Eu .

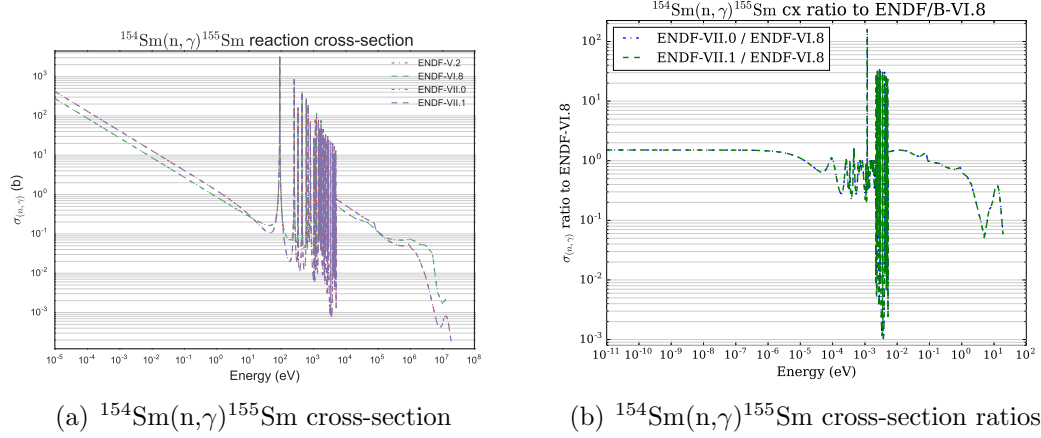


Figure 2.8: Radiative capture (n,γ) cross-sections for several ENDF library evaluations (left) and capture cross-section ratios to ENDF-VI.8 data (right) for ^{154}Sm . [3, 4]

2.7 ORIGEN library performance comparison

With the general validation of the new lattice used for lattice physics transport, the performance of the new ORIGEN libraries generated from these calculations can now be benchmarked in comparison to the previous version of the CANDU libraries distributed with SCALE (based on WIMS transport calculations). Since the older, WIMS-AECL based libraries were available, they were used to directly calculate the isotopic inventories.

2.7.1 28-element library benchmarks

Two different 28-element ORIGEN libraries are benchmarked. The first (referred to as the “unmodified” library) was created using the default 238-group AMPX master multi-group library using TRITON/NEWT with ENDF/VII.0 cross-sections. The second library (referred to as the “modified” library) uses a modified AMPX

master multi-group library for ^{154}Eu , employing modified cross-sections for the thermal-region neutron capture reaction (discussed previously in Section 2.6.3). Two benchmarks are conducted using both libraries, corresponding to the two power histories provided: an inferred outer element power history, and the nominal (assembly-average) power history.

The outer element power history benchmark aims to test the accuracy of the new ORIGEN library by evaluating the library using the same power history as the fuel element used for the destructive analysis benchmark. This benchmark is not directly appropriate because the assembly is using a power history from a particular zone applied across an assembly-averaged spectrum. As a result, nuclides that are strongly influenced by local variations in the flux spectrum (i.e., nuclides whose primary or only production channel is through neutron capture reactions) will suffer in terms of accuracy, while less impact will be seen in isotopes where the dominant production channel is through fission yield (be it either direct yield or accumulated yield through decay). The nominal power history benchmark takes the opposite approach in that it applies the known, average power of the assembly across an assembly-averaged flux spectrum.

Outer element power history benchmark

This benchmark was conducted using both modified and unmodified ORIGEN libraries produced by the new lattice design and compared to the old CANDU 28-element library. The new CANDU 28-element library was generated by irradiating the 28-element lattice at a constant specific power of $25 \text{ MW}/\text{MTU}$ applied over 461 days to reach an exit burnup of $11,520 \text{ MWd}/\text{MTU}$, the same as the previous library. The previous library consisted of 8 library positions, however, the new library used 21 library positions. The results of this benchmark are shown in Figures 2.9 and 2.10.

The new library produced similar major actinide inventories as the old library, with most of the results residing inside the experimental uncertainty. It should be noted that neither the old or new ORIGEN library produced an ^{235}U value that

resided within the experimental uncertainty. Compared to the experimental value the new library produced better correspondence for ^{236}U and ^{241}Pu . There was a large decrease in accuracy for ^{244}Cm , compared to the nominal case, indicating the incorrect flux shape and distribution. Key burnup indicators ^{134}Cs , ^{137}Cs and key fission products ^{237}Np and ^{241}Am were within the experimental uncertainty bounds for both libraries.

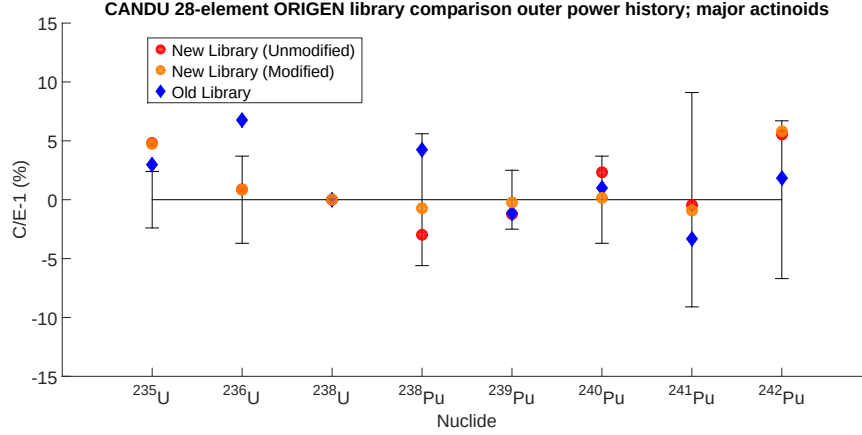


Figure 2.9: CANDU 28-element ORIGEN outer element power history comparison; major actinoids

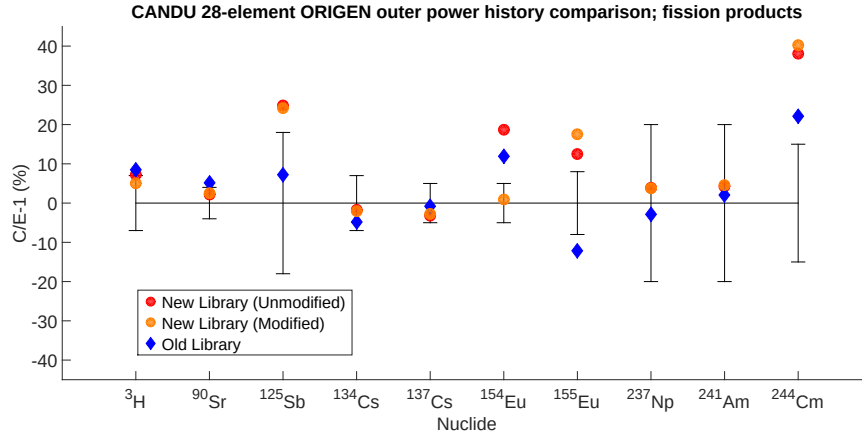


Figure 2.10: CANDU 28-element ORIGEN outer element power history comparison; minor actinoids and fission products

Nominal power history benchmark

This benchmark was conducted using the new ORIGEN library with the nominal assembly power. This benchmark compares the cluster-averaged values from ORIGEN to the scaled experimental data, as described in 2.4.2. It must be reiterated that this will likely bias the experimental data as a result, but nonetheless this benchmark is considered in an effort to test the accuracy of the ORIGEN library. The results of this benchmark are given in Figures 2.11 and 2.12.

All of the major actinoids for the nominal power history case saw agreement and were within experimental uncertainty with exception of ^{238}Pu . The new library exhibited better agreement for all of the uranium isotopes. The plutonium isotopes were similar to the old library. Additionally, the new modified library produced accurate values within uncertainty for both ^{244}Cm and ^{154}Eu which were outside experimental uncertainty for the old library. The improvement in ^{154}Eu is due to the replacement of ENDF/B-VII.0 ^{154}Eu cross-section with that of ENDF/B-VI.8.

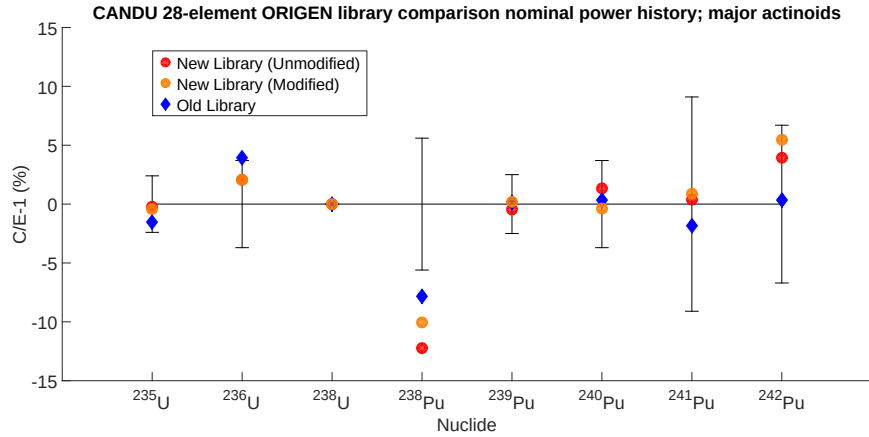


Figure 2.11: CANDU 28-element ORIGEN library comparison nominal assembly power history; major actinoids. Note that benchmark inventories are rescaled from edge element values (per [2]) to produce an assembly-averaged value.

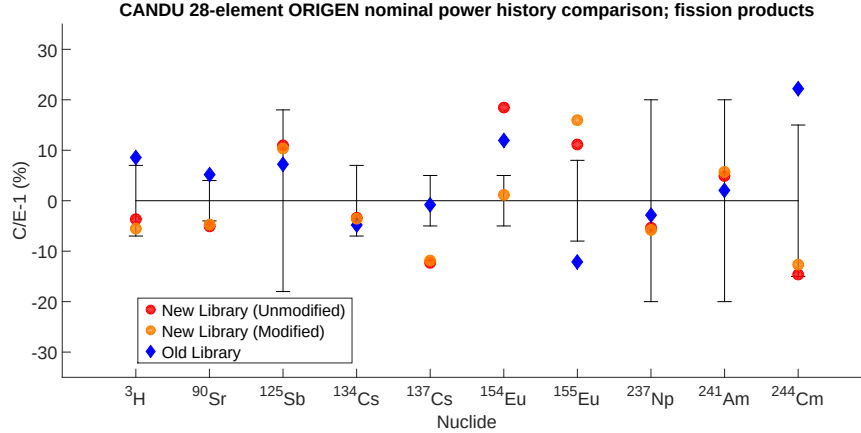


Figure 2.12: CANDU 28-element ORIGEN library comparison nominal assembly power history; fission products and minor actinoids

2.7.2 37-element ORIGEN library benchmark

Figure 2.13 gives a benchmark comparison for major actinide species calculated in ORIGEN using the prior WIMS-based library distributed with SCALE and the newly-developed ENDF/B-VII.0-based library. All values for both libraries lie within the experimental uncertainty. However, the new library produced results that were closer to the experimental value for all of the major actinoids with the exception of ^{241}Pu . Here, assembly-averaged cross-sections are used by ORIGEN to produce assembly-averaged isotopic inventories, which can then be directly compared to the 37-element radiochemical assay benchmark.

2.8 Conclusion

Overall, data from the available benchmarks thus suggest that the new CANDU lattices should provide satisfactory accuracy in line with the levels of accuracy of other provided SCALE fuel lattice templates. While all of the evaluated major actinoids for each CANDU fuel assembly lattice models presented here fit within the experimental uncertainty, some models performed better than others.

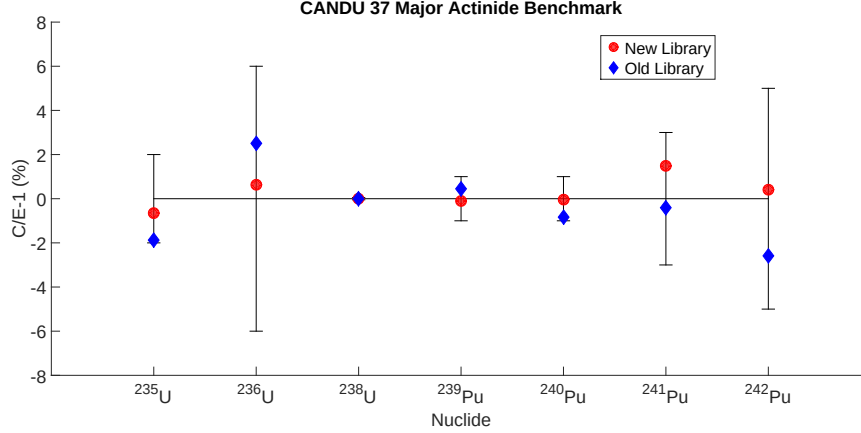


Figure 2.13: CANDU 37-element ORIGEN library comparison

2.8.1 Effect of cross-section libraries on isotopic benchmarks as evaluated in lattice physics transport models

For the 37-element bundle, ENDF/B-VII.0 libraries outperform the ENDF/V libraries with respect to predictions of plutonium isotopes. For the 28-element bundle, all libraries produced similar results for the uranium isotopes. ENDF/B-VII.0 and ENDF/B-VI produced better results for all the plutonium isotopes except for ^{242}Pu . ENDF/B-VI produced the best results for ^{238}Pu and ^{240}Pu . All libraries produced results inside the experimental uncertainty for the major actinoids. Additionally, the ENDF/B-VII.0 library performed comparably or better than the prior WIMS-AECL model with significant improvement in correspondence for ^{238}Pu and ^{242}Pu inventories.

For fission product comparisons, the ENDF/B-V and ENDF/B-VII.0-based libraries generally performed similarly, with ENDF/B-VII.0 showing slightly worse correspondence to experimental assay data for ^{154}Eu but slightly better for ^{155}Eu . For both of these isotopes, the calculated values are outside of the experimental uncertainties for all cross-section libraries evaluated and show substantially poorer agreement compared with the WIMS-AECL model. This is likely due to the cross-section disagreements outlined previously. Prior studies on the effect of cross-section

data and cross-section processing options (e.g., NITAWL versus CENTRM) found effects on the order of 2-3% for ^{154}Eu in LWR lattices. In this study however, the use ENDF/B-VII.0 (from ENDF/B-VI.2) showed up to a 20% difference in predicted inventories for ^{155}Eu , one of the more significantly changed nuclides between the cross-section libraries [15]. ENDF/B-VI also had similar performance to ENDF/B-VII.0 for many of the fission products. While the ENDF/B-VI library did produce accurate results for ^{154}Eu , it performed far worse for ^{155}Eu , over-predicting this nuclide by 15%, a significantly poorer agreement than both ENDF/B-VII.0 and ENDF/B-V.2-based calculations.

For minor actinoids, while ^{237}Np showed increased correspondence from ENDF/B-V to ENDF/B-VII.0, other minor actinoids such as ^{241}Am and ^{244}Cm showed slightly poorer correspondence. For all of the minor actinoids, the move to the ENDF/B-VII.0 libraries appears to create a bias in favor of higher actinide production (i.e., more of the transuranic species are produced for ENDF/B-VII.0 libraries), likely due to the improved resonance-region resolution for these libraries [15]. This trend is likewise supported in the behavior of the plutonium isotopes, where a slight decline in ^{239}Pu is observed in favor of the higher plutonium isotopes in inventories generated using ENDF/B-VII.0. ENDF/B-VI produced poorer results for minor actinoids ^{241}Am and ^{237}Np , however, it did see an improvement in ^{244}Cm over ENDF/B-VII.0.

2.8.2 Performance of the new SCALE-based lattices compared to the prior WIMS-AECL library

In general, the new ENDF/B-VII.0-based SCALE lattices showed improvements in major actinide predictions for both lattices compared to the prior WIMS-AECL model, as well as in predictions of ^{237}Np . In general, ^{154}Eu and ^{237}Np showed the most marked differences in the two models, likely due to improvements in cross-section treatments inherent to the ENDF/B-VII.0 cross-section libraries [15]. For most measured isotopes, the calculated inventories were well within experimental

uncertainties. Predictions for fission products fared slightly worse in the new lattice models, particularly for ^{155}Eu , which is likely due to the adjustment of the ^{154}Eu cross-section.

CANDU 28-element lattice

It is likely that the WIMS-AECL inventories were derived by normalizing the outer element power to a specific pin. However, the power history for a particular element (i.e., an individual fuel pin) is not usually available. In practice, the nominal power case (Section 2.6.1) will be used.

For the major actinoids, the new lattice using the nominal power history performed similarly to the WIMS-AECL model. Improvements in accuracy with respect to the experimental value were observed for uranium isotopics, with decreases noted for the plutonium isotopics. While the differences between the model performance was small for most isotopes, the performance of the new SCALE-based lattice showed a decrease in agreement for ^{238}Pu which was the only isotopic measurement outside experimental uncertainty. However, the previous WIMS-AECL library also produced ^{238}Pu outside the measurement uncertainty.

The accuracy of the minor actinoids and fission products of the new SCALE library were similar to the WIMS-AECL model, with the exception of ^{154}Eu . The specific use of ENDF/B-VI.8 cross-sections for ^{154}Eu saw a significant increase in the reported accuracy of the ORIGEN library (by about 10%). Additionally, ^{237}Np saw a increase in accuracy of 4.5% in the new ENDF/B-VII.0-based SCALE model. ^{155}Eu was not accurately reflected by any library iteration; the inclusion of the more accurate ^{154}Eu capture cross-section lead to substantial over-predictions in the ^{155}Eu inventory (outside experimental uncertainty bounds), while the WIMS-AECL-based ORIGEN library significantly under-predicted ^{155}Eu inventories (again, outside of experimental uncertainty).

CANDU 37-element lattice

The new ENDF/B-VII.0-based lattice showed at least a slight improvement in all calculated inventories compared to the WIMS-AECL model except for ^{241}Pu . A notable improvement was observed for ^{242}Pu , which was outside the uncertainty for the prior WIMS-AECL-based library. All calculated ENDF/B-VII.0 results lie within the experimental uncertainty.

2.9 Acknowledgments

This work was supported by an Nuclear Energy University Programs (NEUP) grant sponsored by the U.S. Department of Energy, Office of Nuclear Energy, award number DE-NE0000737.

The authors wish to thank Ian Gauld of Oak Ridge National Laboratory, who in addition to furnishing them with the original CANDU validation studies, provided much-needed assistance and insights in determining the source of validation issues with the lattices as well as thoughtful comments on drafts of this paper. Finally, the authors would like to acknowledge Brian Ade of Oak Ridge National Laboratory, who likewise provided guidance and troubleshooting assistance in the lattice validation studies.

Chapter 3

Verification and Validation of New Unmodified SCALE PWR Lattice Templates

This chapter is based on a conference article by N .T. Shoman and S. E. Skutnik that will appear in PHYSOR 2016. My primary contributions to this paper include (i) modification of templates to match experimental conditions, (ii) benchmarking the templates, (iii) applying and interpreting results of PLS and PCA methods, and (iv) writing much of the paper.

3.1 Abstract

The goal of this analysis was to determine the effectiveness of using pre-developed, unmodified templates to create cross-section libraries using a full reactor physics simulation. Those cross-section libraries can then be interpolated to problem-specific conditions (e.g., the burnup, enrichment, and void fraction specific to the problem) in order to perform rapid depletion calculations without the need to re-run time-intensive transports calculations. However, unlike in typical benchmark studies the templates were modified to a minimal degree, much as what would be seen in practical application. The templates provide certain parameters that are available for modification such as fuel enrichment and burnup. These parameters were the only parameters modified except for some certain cases in which minor geometry modifications were required, such as fuel pellet diameter. The assembly-level lattice arrangement of pins remained the same as the template in all cases. A more accurate approach would be to use full assembly models and to specify certain pins with known burnup histories for depletion. However, another focus of this report is to examine the effect of applying a burnup of a specific pin to the entire assembly, i.e. in evaluating how well an assembly-average model can capture specific pin features measured in prior radiochemical assay studies. The goal of this study is not to test the full capabilities of the SCALE system. This study is a parallel to an ORIGIN depletion calculation, where the depletion is calculated by solving the Bateman equations using problem dependent cross-sections and fluxes. Lattice physics calculations with problem specific conditions can take several hours whereas ORIGIN calculations can

be computed on the order of seconds. This study aims to determine what is lost when using average or generic properties.

Additionally, multivariate analysis techniques such as principal component analysis (PCA) and partial least squares (PLS) regression is applied to existing radiochemical assay data to determine relationships between nuclides and burnup. Using PLS it is possible to determine the linear variation in the assay data that is due to the linear variation in burnup. In effect, this will help determine the impact of using an unmodified lattice versus a detailed and modified lattice. The use of unmodified, low fidelity calculations works well for simple modeling conditions. However, detailed modeling is required for situations where flux shape changes rapidly or frequently. Multivariate techniques indicate that 85% of the linear variation in the isotopics is due to the variation in burnup, however, more research is required to test the robustness of the models.

3.2 Introduction

One of the main features of the ORIGEN depletion package for SCALE is its suite of pre-generated reactor data libraries (covering a wide variety of fuel lattice types) useful for rapid calculation of assembly-averaged properties, such as isotopic content, decay heat, and radiation source terms [6]. By providing a series of libraries with collapsed one-group cross-sections at fixed burnups, enrichments, and moderator densities, users are able to easily interpolate to problem-specific conditions to directly solve for quantities of interest without the need to repeat computationally-expensive lattice physics transport calculations.

As part of an overall modernization effort for SCALE, the ORIGEN reactor data libraries included with SCALE are being expanded to cover most domestic fuel types as well as being updated to employ the latest nuclear data. A key component to this effort includes validation of these libraries in their isotopic predictions through benchmarks against known destructive assay data from well-characterized

fuel assemblies. While the isotopic prediction capabilities of SCALE (and ORIGEN in particular) have been extensively validated [20], the goal of this report is to characterize the performance of lattice templates “as-is;” in other words, evaluating the performance of the lattice templates used for ORIGEN reactor data libraries using only the modifications that would be expected from interpolation. As a result, the templates were modified to a minimal degree, much as what would be seen in practical application, exclusively modifying parameters such as the power history and enrichment.

This analysis will present the results of a validation campaign conducted on the new pressurized water reactor (PWR) fuel lattice templates to be included with the upcoming SCALE 6.2 release. Here, the goal of this analysis is not to repeat the validation of SCALE’s depletion capabilities but rather to present the relative degree of accuracy that can be expected from pre-generated reactor data libraries, such as those distributed with ORIGEN. This analysis thus relies on generic templates without reactor-specific features, such as placement of burnable poisons, soluble boron history, etc., instead relying solely on the typical user-specified interpolation parameters such as the power history and initial fuel enrichment [7]. This analysis thus closely approximates the interpolated cross-sections employed when using the ORIGEN reactor data libraries interpolated to problem-specific conditions. Note that the goal of this analysis is not to validate the capabilities of SCALE.

An additional aim of this analysis is to evaluate the accuracy of using assembly-averaged models to estimate the isotopic content of specific fuel pins. Because ORIGEN cross-section libraries use the assembly-averaged collapsed one-group spectrum, these data libraries therefore inherently represent an assembly-averaged flux. While a more accurate approach to evaluating pin-specific properties would be to model the full assembly and deplete pins of interest separately, here the goal is to convey how well an assembly-averaged model (such as those represented by the ORIGEN reactor data libraries) can capture specific pin features measured in prior radiochemical assay studies.

For the purposes of this study, SCALE 6.1 was used to carry out depletion calculations which were compared against experimental assays. This report examines fuel samples from a variety of programs with a wide range of burnups. Except where noted, the geometry of the templates was held constant; only the interpolatable template parameters (e.g., enrichment, cycle power, cycle length, and inter-cycle decay time) were modified.

Researchers at ORNL conducted an extensive validation study of the depletion capabilities of SCALE 5.1, benchmarking SCALE isotopic predictions to radiochemical assay studies [20]. Much of the benchmarking data for this study rely on the same radiochemical benchmarks as prior study. For both studies, the benchmark data originated from multiple spent fuel measurement campaigns; this study uses an aggregate of many of the PWR assays conducted to date.

In addition to benchmarking several unmodified PWR lattices, this study examines the use of multivariate analysis techniques to analyze trends in the isotopics, including principal component analysis (PCA) and partial least squares regressions (PLS). Principal component analysis is used to compare the linear relationships in large sets of data. Currently, PCA is being used to determine how well experimental and calculation results agree with each other in a linear sense.

PLS is used to determine which parameters are important for lattice physics calculation. In specific, how much of the isotopic variance can be explained by particular parameters such as burnup? This study primary focuses on the lattice performance without modeling more advanced parameters such as the boron letdown curve or proximity to burnable absorbers. It is important to note that PLS can only capture linear variance; thus, non-linear variances cannot be accounted for. Initial results show that 85% of the variance in the measured major actinoids can be explained by the variance in the burnup. PLS and PCA analysis was conducted using the PLS-Toolbox[21] for MATLAB[22].

3.3 Procedure

3.3.1 Template Validation

This study considers the results of four different PWR lattices: Babcock and Wilcox 15x15, Siemens 14x14 Westinghouse 15x15, and Westinghouse 17x17. Each template was run only changing the default parameters for enrichment and assembly power. The templates were run through the TRITON transport sequence using the 238-group ENDF/B-VII.0 cross-section library. The results were then compared to experimental assay data.

The TRITON/NEWT 2-D transport sequence was used with models that were developed for each of the assemblies. The transport and depletion sequence (T-DEPL) was run using the 238-group ENDF/B-VII.0 cross-section library using SCALE 6.1. The T-DEPL sequence uses BONAMI and WORKER/CENTRM/PMC for resonance cross section processing, NEWT for 2-D discrete ordinate transport calculations, COUPLE for calculating collapsed one-group cross-sections for each burnup point, and ORIGEN for depletion calculations and updating isotopic compositions.

In order to more effectively analyze trends in evaluated assembly isotopic values compared to benchmark data, plots are given for select major and minor actinoids. This report utilizes assay data from a variety of different studies. The selected actinoids were the most common collected data cross the different assay studies. The plots aim to analyze trends in these isotopes across burnup for a particular assembly.

3.3.2 Multivariate Analysis

Principal Component Analysis

Principal component analysis is classified as a statistical method that utilizes an orthogonal transformation to convert observations of possibly correlated variables into a set of values that are linearly uncorrelated called principal components. The

number of components corresponds to the dimensionality of the reduced space with the maximum number equaling the number of original variables. Each principal component is such that the first component has the most variance, the second the next highest, and so on. Each new principal component must be orthogonal to all the components before it [23, 24].

Essentially the linear transformation changes the coordinate space x of n variables and m observations to principal component space. The new variable (t_1) is a linear combination of the original variables so that t_1 has maximum variance. The loadings can be defined as $w_{(k)} = (w_1, \dots, w_p)^T_k$ that map each row vector $x_{(i)}$ of x to a vector of principal component scores $t_i = (t_1, \dots, t_p)$ such that $t_{k(i)} = x_{(i)} \cdot w_{(k)}$. The first loading has to satisfy

$$\mathbf{w}_{(1)} = \arg \max_{\|\mathbf{w}\|=1} \left\{ \sum_i (t_1)_{(i)}^2 \right\} \quad (3.1)$$

where further components can be found by subtracting the first $k - 1$ principal components from x

$$\hat{\mathbf{X}}_k = \mathbf{X} - \sum_{s=1}^{k-1} \mathbf{X} \mathbf{w}_{(s)} \mathbf{w}_{(s)}^T \quad (3.2)$$

so that the final decomposition of x can be represented as $T = xW$.

An example of PCA for three variables is provided in Figure 3.1. For the example data set, two principal components capture most of the variance and are likely sufficient. PCA can also be considered as a dimensionality reduction technique. Data can be reduced to fewer principal components without significant data loss.

PCA can be applied by the following steps:

1. Place all data into a matrix x of dimension $N \times M$ where N is the number of observations and M is the number of features.

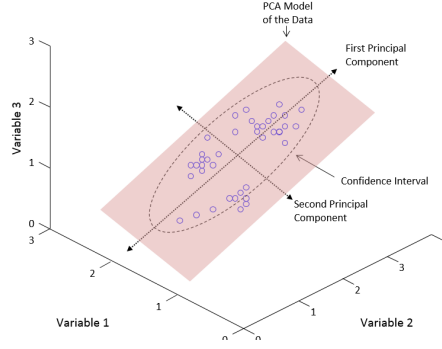


Figure 3.1: Example of a PCA model with three variables

2. Mean center the matrix x
3. Calculate the covariance matrix of the data matrix, x
4. Compute the eigenvectors and eigenvalues of the covariance matrix
5. Sort the eigenvectors by decreasing eigenvalues and choose the D number of eigenvectors by decreasing eigenvalues. Use these eigenvectors to form the transform matrix W which is size $D \times N$. D is the number of reduced dimension subspace.
6. Determine the transformed data Y by multiplying the transpose of the transform matrix by the original data, x , such that $Y = W^T x$.

Partial Least Squares

Partial Least Squares Regression (PLSR)[25, 26] is a method that finds a linear regression model by projecting predicted and observed variables to a new space. A PLS model will attempt to find the multidimensional direction in the X space that explains the maximum multidimensional variance direction in the Y space. When more than two variables are present the data will be represented by a hyperplane in multidimensional space. Under particular settings PLS will provide identical results to that of a linear discriminate analysis or euclidean distance to centroid method. The components of the model's components can be calculated using the non-linear

iterative partial least squares (NIPALS)[27, 28] algorithm. The method can calculate the components without direct calculation of the covariance matrix which results in a reduction of computational time.

The NIPALS algorithm is as follows: First, decompose the X and Y matrices in the form:

$$\begin{aligned} X &= TP^T + E \\ Y &= UQ^T + F \end{aligned} \tag{3.3}$$

where T and U are the $(n \times p)$ matrices of the p extracted score vectors (components, latent vectors), the $(N \times p)$ matrix P and the $(M \times p)$ matrix Q represent matrices of loadings and the $(n \times N)$ matrix E and the $(n \times M)$ matrix F are the matrices of the residuals. The method finds the weight vectors w, c such that

$$[\text{Cov}(t, u)]^2 = [\text{Cov}(Xw, Yc)]^2 = \max_{|r|=|s|=1} [\text{Cov}(Xr, Ys)]^2 \tag{3.4}$$

where $\text{Cov}(t, u) = \frac{t^T u}{n}$ is the sample covariance between the score vectors t and u . A variety of different algorithms can be used to solve the problem. The components of the model can be calculated using the non-linear iterative partial least squares (NIPALS)[27, 28] algorithm. The method can calculate the components without direct calculation of the covariance matrix which results in a reduction of computational time.

The algorithm then proceeds over several steps as follows: First, project the matrix X onto u to determine the loading vector w (Equation 3.5).

$$w = \frac{X^T u}{u^T u} \tag{3.5}$$

Next, the loading vector w is normalized to a length of one by dividing by its norm (Equation 3.6).

$$w = \frac{w}{|w|} \quad (3.6)$$

Calculate the corresponding score from the loading vector (Equation 3.7)

$$t = Xw \quad (3.7)$$

The weights and scores of Y are computed in a similar fashion. It is important to note that PLS can be prone to over-fitting. If the number of features is greater than the number of observations then the model will perfectly fit the given observations but fail to accurately predict new data. In this application 39 observations (different PWR fuel assays) with 8 features (the major actinide assay values) are used.

3.4 Results

The results of the TRITON/NEWT calculations are given below. Additional tables of all benchmarked nuclides are provided in appendix B.

3.4.1 Westinghouse 15x15

The Westinghouse 15x15 benchmark data consisted of experimental data[20] from Turkey Point Unit 3 and H.B. Robinson Unit 2. Samples from Turkey Point were collected from near the center of the fuel rod. Four samples from H.B. Robinson were obtained from various positions of a single rod while the other two were measured from a second pin.

The results from the Turkey Point benchmark averaged an error of 2.03% over all plutonium isotopes (Figure 3.2) for all runs. Most plutonium isotopes had $\leq 10\%$ error. However, ^{238}Pu was over-predicted by as much as 16%. ^{148}Nd was also benchmarked and had an error $\leq 5\%$ (appendix B.1.1). The accuracy of ^{148}Nd

indicates that the power was calculated and distributed correctly across the assembly. The use of ^{148}Nd to determine burnup is outlined in ASTM-E321[29]. ^{235}U was found on average to agree with experimental results within 3%. The experimental uncertainty was not recorded in the primary report.

The results for the H.B. Robinson benchmark were less accurate than Turkey Point. This is likely a result of the specific assembly history; the H.B. Robinson assembly was irradiated for two cycles in two different locations in the core. During the first cycle only, the fuel pin was adjacent to a burnable poison rod, which will significantly impact the shape of the local neutron spectrum; this burnable poison rod was not modeled (the rod was removed following the first cycle). Additionally, reflective infinite lattice boundary conditions were assumed, which may be violated by changing the assembly position in the reactor core. Nonetheless, ^{235}U was typically within 4-5% of the measured value. Many of the plutonium isotopes resided within 10% of the experimental value. ^{238}Pu values were particularly inaccurate seeing error as high as 90%. ^{238}Pu is difficult to accurately predict with even a high-fidelity model, due to the sensitivity to the spectral shape. The production of ^{238}Pu arises from ^{237}Np irradiation, ^{242}Cm alpha decay, or (n,2n) reactions.

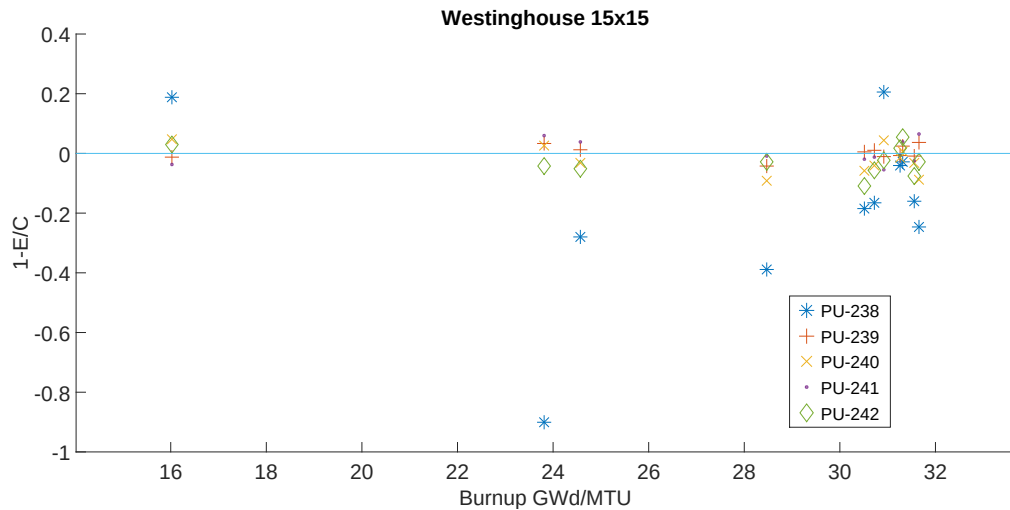


Figure 3.2: Turkey Point and H.B. Robinson (Westinghouse 15x15) plutonium isotopic benchmarks

3.4.2 Westinghouse 17x17

The experimental benchmark data for the Westinghouse 17x17 came from Takahama Unit 3[20]. The assay collected 16 different samples, however, 5 of these are $\text{UO}_2\text{-Gd}_2\text{O}_3$ which were not considered. The eleven axial samples used originated from two different pins near the edge of the assembly. There was an extended isotopic inventory conducted on the samples. Most of the calculated plutonium inventories were within 10% of the measured value (Figure 3.3), with many residing inside 5%. One sample, SF97-1 had a larger error than other samples from the same pin. However, this result is consistent with a prior validation study[20]. Additionally, gadolinia fuel pins were present, but not accounted for in the model.

Several different neodymium isotopes were benchmarked (Appendix B.1.2). Most of the isotopes showed good agreement and were within 5% of the experimental value for rod SF95, indicating good agreement with measured burnup. ^{142}Nd and ^{144}Nd had higher errors than the other isotopes, although ^{144}Nd was within 10%. ^{142}Nd data was not available for all samples. Cesium isotopes were also benchmarked and saw good agreement within 3-4%. Edge effects, and the assumption of an infinite lattice, had little impact on the isotopic calculations.

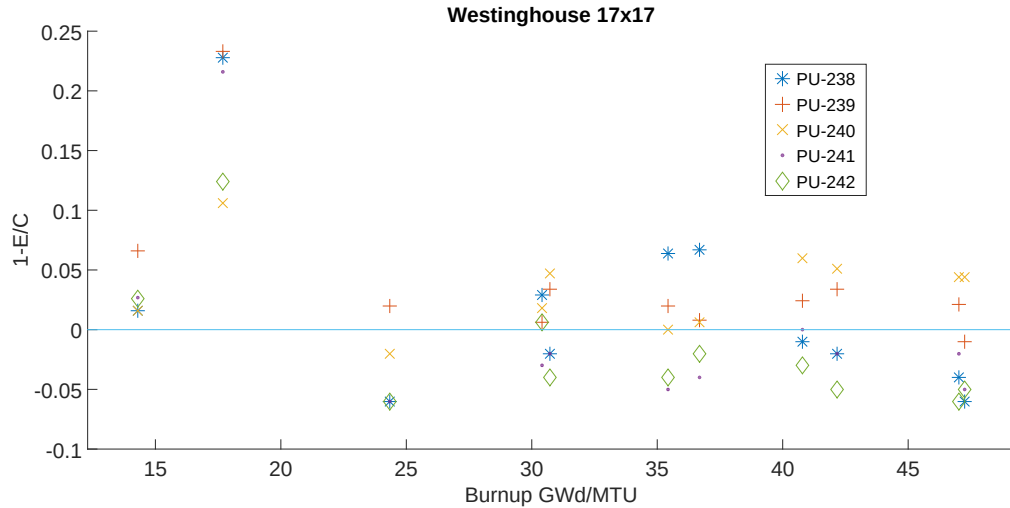


Figure 3.3: Takahama unit 3 (Westinghouse 17x17) plutonium isotopic benchmarks

3.4.3 Combustion Engineering 14x14

The Combustion Engineering benchmark was derived from data from Calvert Cliffs Unit 1[20]. There were three samples from two different assemblies for a total of six samples. The concentration of soluble boron was known, but was not modeled. ^{148}Nd content and linear heat generation rates were available for each sample. At each axial location the linear burnup of time intervals was calculated as the product of the interval size and linear heat generation rate were summed for each cycle. The linear burnups were then normalized to the total burnups which were used in the simulation.

Major actinoids for assembly D101 saw reasonable agreement (Figure 3.4), however, ^{238}Pu and ^{241}Pu were overpredicted with respect to the experimental value (Appendix B.1.3). This is likely due to the assayed pin which resided at the edge of the assembly. Previous research has suggested that edge pins require special modeling.

The major actinide values for assembly D047 had good correspondence with the measured value except for ^{238}Pu and ^{235}U , both of which were overpredicted. Additionally, all Nd isotopes for the three samples were within 5% of recorded values indicating good burnup agreement. The previous benchmark study indicated that the irradiation history was adjusted to achieve good results[20].

3.4.4 Babcock & Wilcox 15x15

Eleven samples from Three Mile Island Unit 1[20] (TMI-1) were used to benchmark the unmodified performance of the Babcock & Wilcox 15x15 lattice. All of the samples originated from different axial positions of the same fuel rod. Uranium isotopic predictions were generally in good agreement with experimental values and similar to prior validation studies. Plutonium isotopics were similar to previous studies, however, were quite inaccurate in some cases. Burnable poison rods were present for the first irradiation cycle, but were removed for the second cycle which were not modeled. In order to achieve good results and an accurate local environment, the

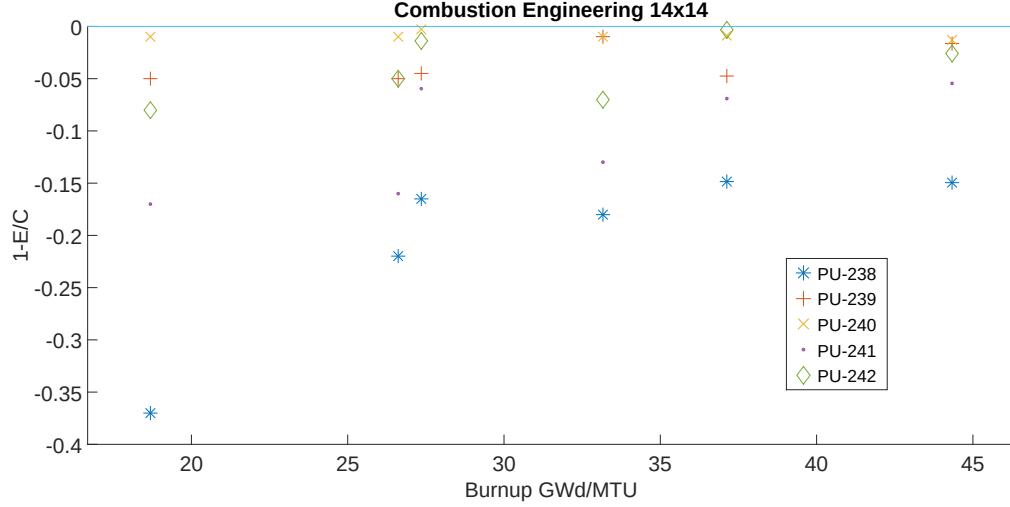


Figure 3.4: Calvert Cliffs (Combustion Engineering 14x14) plutonium isotopic benchmarks

previous benchmark study depleted nearest neighbor pins individually which was not performed for this study. The previous study achieved accuracy around 10-20% error for the plutonium isotopics (Figure 3.5). Neodymium saw excellent correspondence with experimental values typically having an error of only a few percent, indicating good agreement between the modeled and actual fuel burnup (Appendix B.1.4). The TMI-1 assay provided an extended isotopic comparison. Fission products such as Sm saw reasonable agreement. However, other fission products such as ^{155}Eu saw poorer correspondence.

Some studies[30, 31] have reported large discrepancies between measured and predicted plutonium concentrations, even when a high fidelity model was used. The differences are larger than what can be explained on the basis of laboratory-estimated bias and uncertainty, or uncertainty associated with reactor operating conditions. The assayed fuel samples were operated in cycle 10 when significant operating anomalies occurred causing fuel failure. These issues prompted a recent re-evaluation of the Three Mile Island assay data conducted by Oak Ridge National Laboratory on five samples[32].

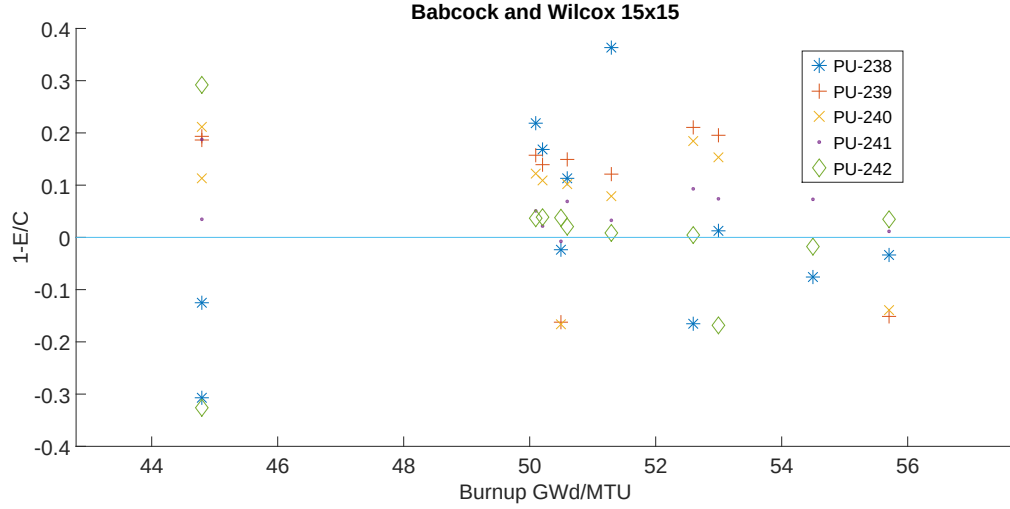


Figure 3.5: Three Mile Unit 1 (Babcock & Wilcox 15x15) plutonium isotopic benchmarks - prior ANL evaluation

The unmodified templates produced a much more consistent range of values when compared against the newer assay data. Plutonium isotopics were calculated within 10% of the experimental value (Figure 3.6). The error is likely due to the proximity to guide tubes which were not modeled.

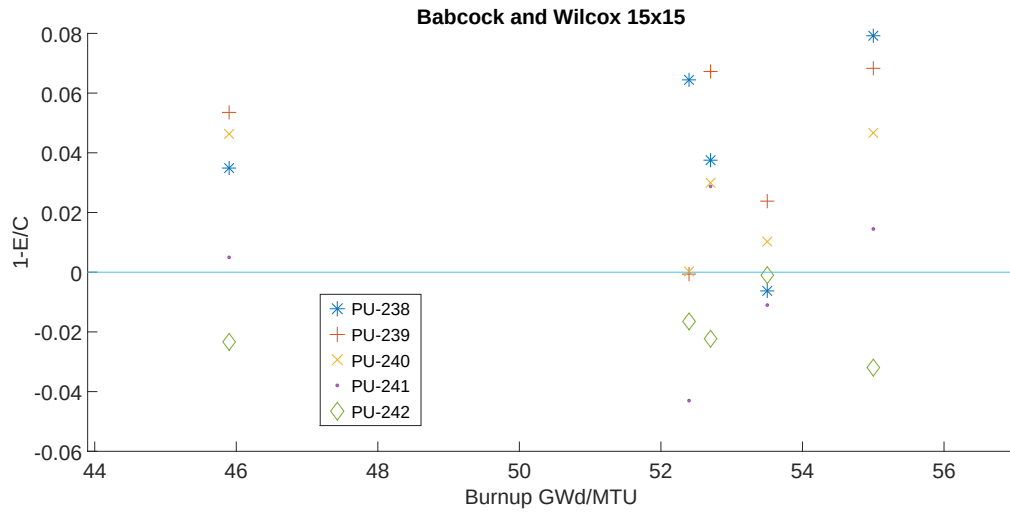


Figure 3.6: Babcock & Wilcox 15x15 plutonium isotopic benchmark - new ONRL evaluation

3.4.5 Multivariate Analysis

This study also considers the use of PCA to analyze trends in large fuel assay sets. Previous work has been conducted that indicated that similar techniques can be used to determine reactor origin, either pressurized or boiling water reactor, and burnup of the fuel using nuclide concentrations as input data[33]. Initial work has been generating a PCA model of the spent fuel assay as seen in Figure 3.7.

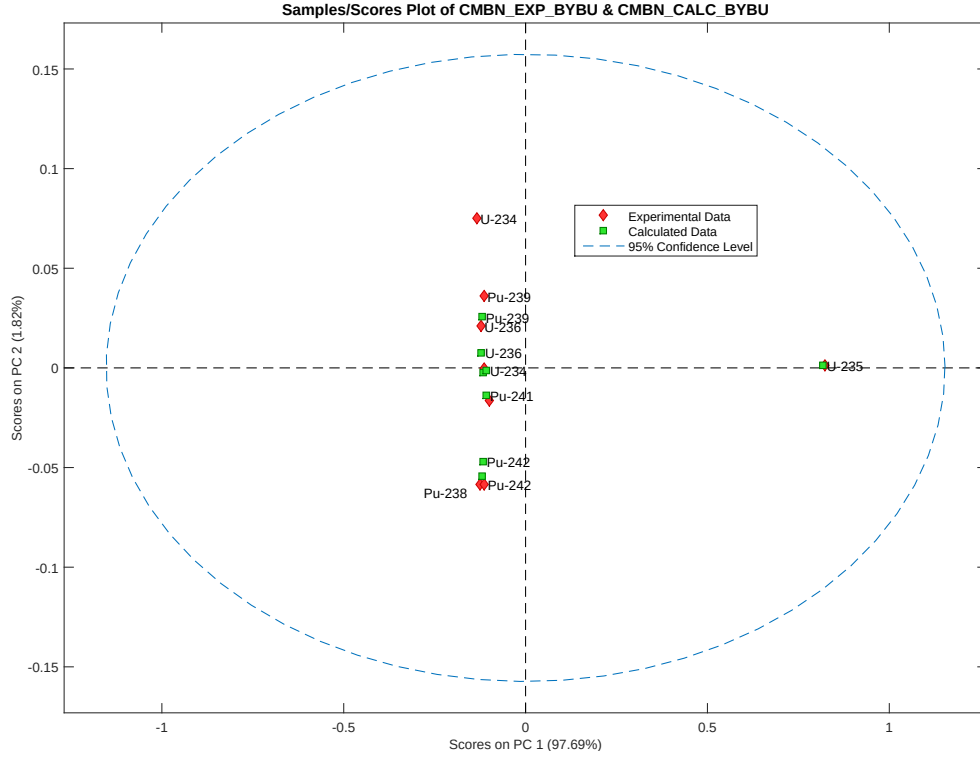


Figure 3.7: PCA model to compare experimental and calculated values for major actinoids

Plutonium and uranium inventories were selected as the input data for the model due to their commonality across all experimental assays. The entire experimental data set of inventories across all assemblies was used to calibrate a PCA model. The calculated data resulting from the lattice physics simulations were used as validation data. This technique is in its early stages and was primarily used as a heuristic tool. The calculated major actinoids fit within the uncertainty for the generated model.

Additionally, clustering occurred for individual nuclides indicating good agreement between calculated and experimental values in PC space. It is currently unknown why ^{235}U loads differently on the first principal component. Since the first principal component captures 97% of the variance it, is expected for most of the nuclides to have similar values for PC1. This behavior is shown in Figure 3.7 which shows that the major actinoids have similar linear variation.

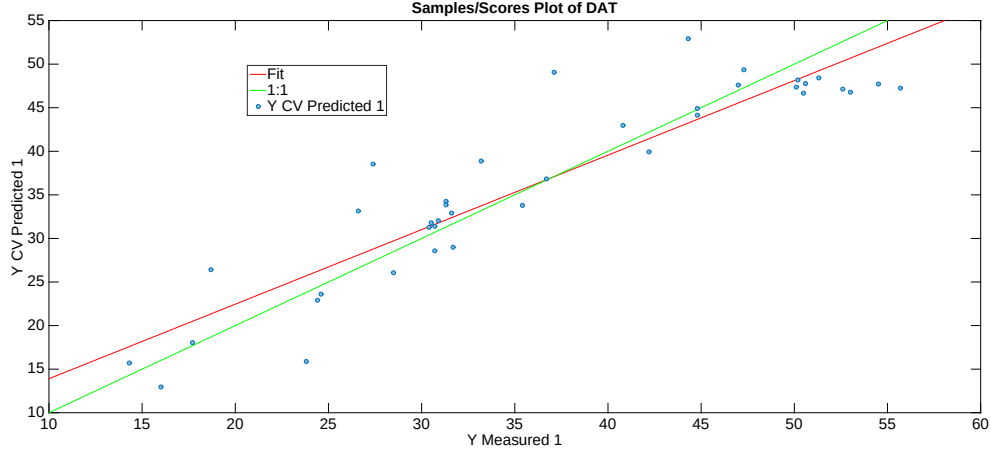
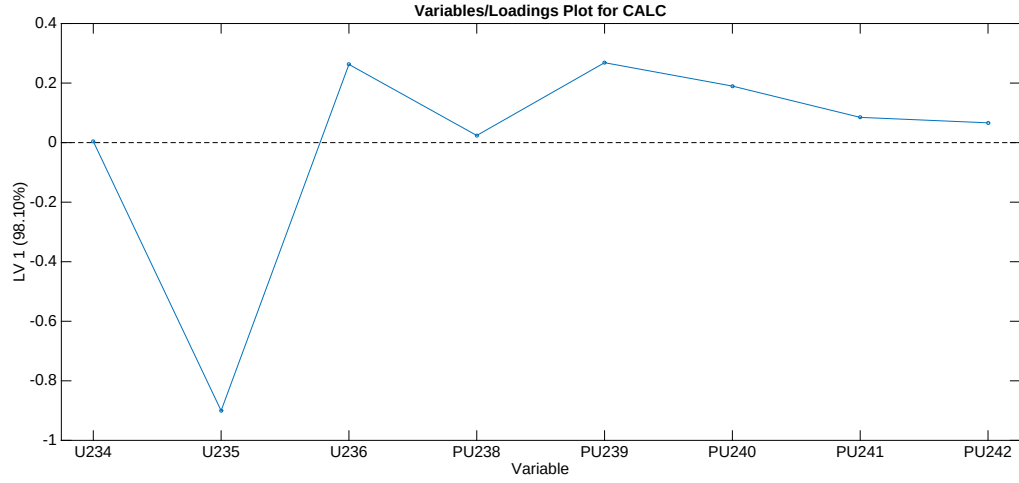


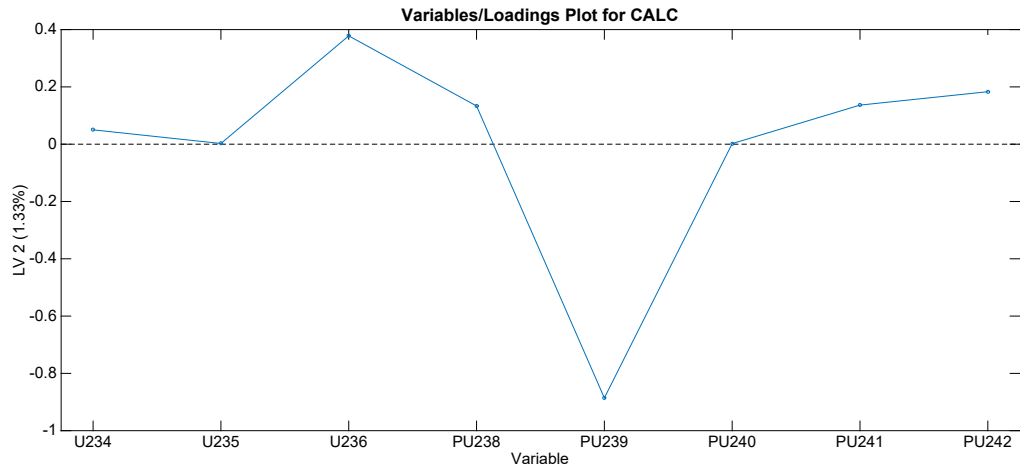
Figure 3.8: PLS input and predicted burnup values (based on input isotopics), the green line is the “ideal” prediction where the red is the actual

A PLS (Partial Least Squares) study was generated using all of the experimental assay data, again with the uranium and plutonium isotopics. Additionally, the burnup for each assay was then loaded into the model. Results show that 85% of the linear variation in the burnup can be explained by using 99.43% of the information in the isotopics. This is an important finding in determining which lattice parameters are of importance for accurate major actinide predictions. Figure 3.8 shows the actual and predicted results of the model. The green line is a 1:1 fit of the input data to the predicted, which would be the ideal fit. The loadings of latent variable 1 and 2 are provided in Figure 3.9. Note that ^{235}U has a strong correlation with burnup as indicated by the large weight for the feature. Another particular interesting analysis would be to determine the inverse problem, that is, how much of the variation in the isotopics is due to the burnup. However, the resulting model would be essentially a

linear regression model with a single input and output, which would not provide any meaningful insight.



(a) Loadings on LV1



(b) Loadings on LV2

Figure 3.9: Significant loading for ^{235}U and ^{239}Pu on latent variables 1 and 2

3.5 Conclusions

This analysis has shown that low fidelity, “out-of-the-box” templates work well for simplistic modeling conditions. However, detailed modeling is required for areas with significant local flux and spectral variations, such as in the presence of burnable

poisons. More research is needed to determine if competing effects in a simplistic model are canceling out and providing more reasonable results than expected. Additionally, multivariate techniques have provided a unique insight into the trends in the isotopic data set. However, more research is required to test the robustness of the models to ensure they are not over-fitted. It is unlikely that the model is over-fit, since there are more observations than features however, further analysis of the residuals is required.

Chapter 4

Experimental Performance of the Multi-Isotope Process Monitor

This chapter is based on a journal article in preparation for Nuclear Instruments and Methods by N. T. Shoman, J. B. Coble and D. Meier. Parts of this report are based on readiness reports of the MIP project for PNNL. I am responsible for writing the manuscript and was responsible for the analysis and determining associated pre-processing methods. Dr. J. B. Coble provided assistance in interpreting and troubleshooting implemented methods. D. Meyer and his team at Pacific Northwest National Laboratory carried out the experimental measurements at Savannah River National Laboratory.

4.1 Abstract

The goal of this analysis is to test the experimental performance of the Multi Isotope Process monitor. Previous research has demonstrated the sensitivity of the MIP monitor to acid concentrations and burnup of the fuel[34, 35]. New analysis presented shows the ability for the MIP monitor to discriminate between UNF and MOX materials. The monitor can discriminate between materials of the same type (either UNF or MOX) within a threshold of about 10%. It is possible that with additional analytical techniques this threshold can be further reduced. Finally, the MIP monitor has been demonstrated to capture transients. Since simulating a diversion scenario is not feasible, data is presented on the introduction of a fuel material into a sample box, which, represents a change in the observed spectra. Some results suggest that further pre-processing (such as global alignment) is necessary to produce accurate results. Overall, the MIP monitor is effective in discriminating materials in a number of situations during a real world deployment.

4.2 Introduction

The recent growth in the nuclear sector has renewed interest in used nuclear fuel reprocessing. The once through cycle will not be sustainable long term, so the

alternatives must be considered. One alternative is extending the fuel cycle by using fast reactors and reprocessing spent nuclear fuel. This could potentially extend the fuel supply for several thousand years.

One concern for fuel reprocessing is the diversion of materials for weapons use. Commonly, fuel is recycled through the Plutonium Uranium Extraction (PUREX) process. The process is a safeguards concern because the process extracts pure plutonium which can be diverted. The current International Atomic Energy Agency (IAEA) standard is that a reprocessing plant must account for extracted plutonium within one significant quantity per month (8 kg for Plutonium)[36]. This is typically checked through the use of destructive analysis which can be costly and time consuming.

The Multi-Isotope Process (MIP) Monitor is a non-destructive monitoring program to detect diversion and tampering of nuclear material at a reprocessing facility[34, 35]. The MIP is not a material accountancy system, rather, a system to flag potential diversion or off-normal conditions. While the MIP monitor is used as a safeguards monitor it can be deployed as a process monitor. The system measures several gamma-emitting isotopes in different stages of the reprocessing cycle. The isotopic distributions are measured in near-real time by using gamma spectrometry and comparing measured distributions to “normal” process distributions processed by multivariate statistics, either principal component analysis (PCA)[24] or partial least squares regression (PLS)[37]. The MIP monitor is paired with gamma detectors that can be easily deployed into an existing facility. The MIP monitor is currently deployed at Savannah River National Laboratory (SRNL) H-canyon reprocessing facility. This paper discusses recent results from the data collected at SRNL. This chapter demonstrates the capability of MIP to discriminate between Used Nuclear Fuel (UNF) and Mixed Oxide Fuels (MOX), between different UNF samples, and between different MOX samples. Additionally, an initial analysis to quantify the MIP discrimination capability is included.

4.3 Experimental Data Collection

The MIP monitor is deployed at H-Canyon to capture the changing radiological conditions within a nuclear separations plant in near-real-time (NRT) while the facility conducts normal processing, cycling, and transfer operations. The deployment locations were metal sampling boxes inside the hot sampling aisle in the separation facility that provided direct access to the dissolution and mixing tanks that are located deep inside the lower levels of H-Canyon. Each sampling box is roughly 1 m³ in dimension with a sidewall thickness of approximately 25 mm. Attached to each of these boxes is an air pneumatic system that allows for the sample transfer of material from the large storage tanks located elsewhere in the facility into the hot sampling aisle. The tank solution is circulated through the sampling box during the cycling of the tanks providing a constant stream of solution into the sampling box. Inside each of these boxes is a 10 mL collection vial that was used by facility personnel as a means of capturing grab samples of the tank contents for chemical and radiological analysis. The project deployment team used the same grab vials as the primary radiological source for measuring gamma-ray spectra from the material in the tanks. Given appropriate training at the H-Canyon and SRNL facilities, deployment team members could be positioned a few feet away from the sampling box while it was opened and cycling eliminating the need for collimation and expensive stand-off facility modifications to the sampling boxes for this effort.

All of the materials analyzed have been characterized using both a NaI(Tl) and LaBr₃ detectors. Data were collected using Maestro software using List Mode. This mode puts a time stamp on each count collected by the software. Currently, five-second spectra are analyzed.

4.4 Methods

The MIP monitor involves several steps of pre-processing before the PCA model can be calculated. First, the observed gamma spectra are aligned together. Next, the background is subtracted from each of the samples. These two steps are important because the PCA model maximizes variation to separate classes. If the spectra are not aligned properly, the PCA model will incorrectly flag the sample as off-normal. Additionally, the background is subtracted in order to obtain the true signal. Since reprocessing facilities may have multiple operations occurring at the same time this can cause the model to incorrectly classify the material. The final pre-processing steps are to perform normalization and mean centering. Currently, infinite normalization is used. That is, the sample is normalized to the maximum value observed for all variables. Normalization is used to ensure all spectra have the same weight in the model. It is important to note that normalization occurs after background correction. The presence of the baseline signal could produce an incorrect normalization. Mean centering is performed in order to redefine the 0, or center of the data.

Once the data has been pre-processed, the data can then be used in a PCA model. Typically models are trained on a set of data while other samples are placed in the PC space for comparison (Section 3.3.2). The PCA model treats each channel as a feature. The original 1024 channel spectra are converted to a two-dimensional reduced PC space. In the following section each data point in the PCA models are reduced spectra observations. Note that many of the models include a confidence interval. Currently, these ellipses represent a 95% Gaussian confidence interval and do not account for counting statistics. Future work is needed to develop a more rigorous confidence interval.

4.4.1 Background Subtraction

One of the important methods introduced into the MIP data processing is background subtraction. In facilities where there may be a high level of background due ongoing processes it is important to remove the unwanted background. Additionally, the background signal can vary at different locations in a reprocessing facility, as in Figure 4.1. If the true gamma spectra is not compared to the “normal” spectra it will produce false alarms.

A variety of background correction methods were considered, including a simple background subtraction, weighted least squares (WLS)[38] and weighted least squared with a Whittaker filter[39]. The simple background correction consists of calculating an average background spectra from multiple observations that have been collected. The average spectra is then subtracted from each observed spectra of interest. This method can result in a negative number of counts in certain channels, which is a non-physical answer. This could be solved by simply setting the negative areas to zero, however, important information may be lost, as seen in Figure 4.2.

The next method is WLS[38], which uses an iterative asymmetric least squares algorithm. Since background observations were recorded, they serve as the baseline. The background signal is removed by subtracting some of the collected background signal which results in a low background signal. Since only some of the background is removed, the resulting signal is positive. The algorithm iteratively fits a baseline to each spectra to determine which channels are clearly above or below the baseline. The points above the baseline are more important to the signal, whereas points below the baseline are more important in fitting the baseline to the spectrum.

The WLS-WHIT[39, 40] method is similar to the WLS method. The method uses a Whittaker filter and aims to create a smooth baseline approximation. The algorithm has two conflicting goals, that is, the fitted baseline must remain close to the spectral baseline and it must be smooth. These goals can be formulated as a

sum of squares problem with penalization where more smoothing occurs the larger the deterioration of the data (which is the penalization).

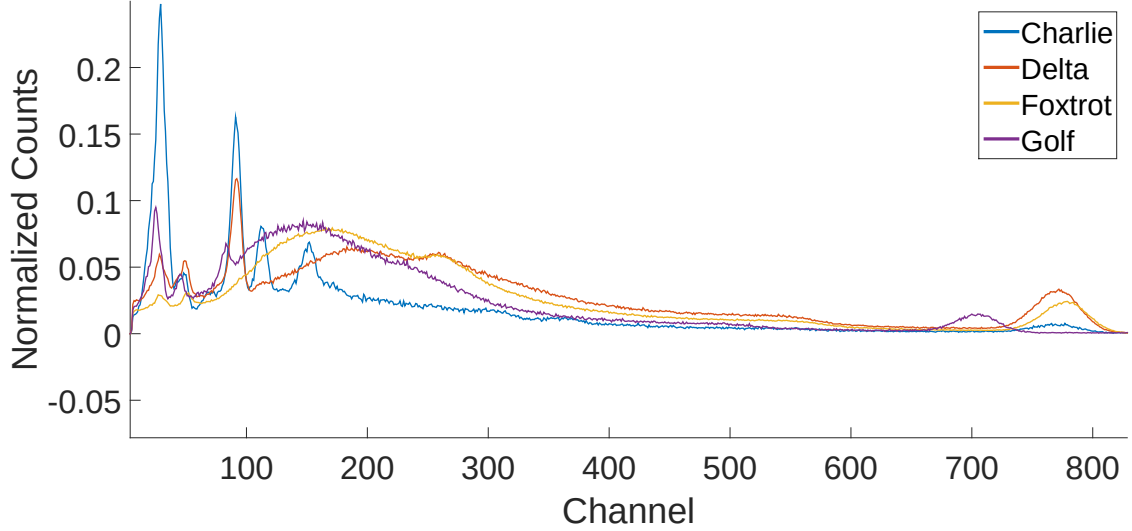


Figure 4.1: Background Spectra at various collection tanks in the H-canyon facility

4.4.2 Spectral Alignment

Spectral alignment is an essential operation that corrects for detector drift or channel drift due to small changes in geometry. Alignment results in population clustering in the transformed space which allows for more precise comparisons. Early data collection indicated that simply shifting the spectra to the designated reference spectrum would be sufficient to align the spectra, as seen in Figure 4.3. However, newly collected data show that non-linear gain shifts are affecting the spectra. In order to account for this non-linear gain shift a Rapid Alignment via Fast Fourier Transform (RAFFT)[41] is used. A comparison between RAFFT and simple alignment can be seen in Figure 4.4.

A fast Fourier transform is used to compute a cross-correlation function that enables alignment between samples to be optimized. The method is based on spectral segmentation, that is, the spectrum is divided into an arbitrary number of segments

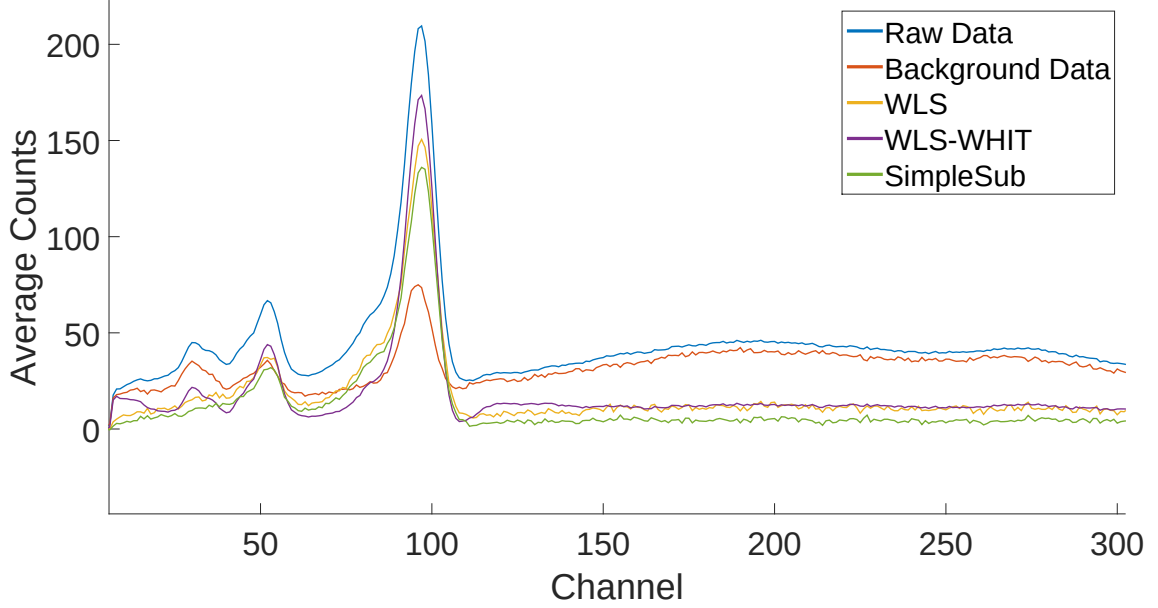


Figure 4.2: A variety of background correction methods. Note that some methods do not retain all spectral features

and the correlation coefficient is used to determine the optimal shift for each segment (in this case, the optimal location is determined via FFT). The minimum segment size for alignment is determined automatically by using a recursive alignment. The spectra is first aligned on a global scale then on a local scale. The advantage is that the algorithm can automatically locate and align local areas that require particular attention. A constraint that the segment size cannot fall below 10 points is implemented as to prevent aligning small segments of noise.

The shift between the reference data set and the observed data set can be calculated as follows. The cross-correlation function, $\text{Corr}(r, s)_u$ of two functions, $r(x)$ and $s(x)$, at any shift position u can be defined by Equation 4.1:

$$\text{Corr}(r, s)_u = \int_{-\infty}^{\infty} r(x)s(x+u)dx \quad (4.1)$$

And the Fourier transform for some function $h(x)$ is given by Equation 4.2

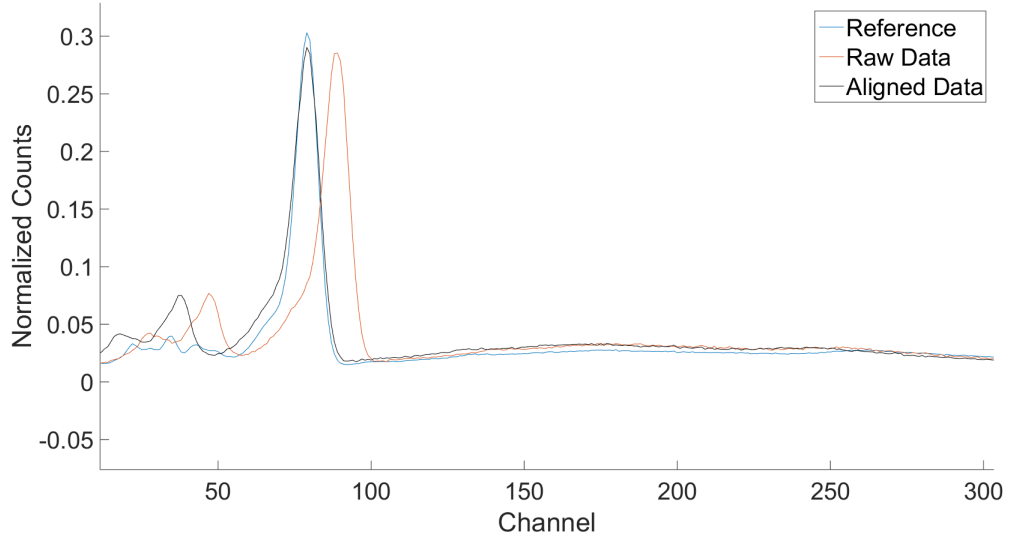


Figure 4.3: Example of shifting the entire spectra to achieve proper alignment

$$\begin{aligned}
 H(\chi) &= \int_{-\infty}^{\infty} h(x) e^{2\pi i \chi x} dx \\
 h(x) &= \int_{-\infty}^{\infty} H(\chi) e^{-2\pi i \chi x} d\chi \\
 h(x) &\Longleftrightarrow H(\chi)
 \end{aligned} \tag{4.2}$$

The discrete correlation theorem states (Equation 4.3)

$$\text{Corr}(r, x)_u \Longleftrightarrow R(\chi) S^*(\chi) \tag{4.3}$$

Note that $H(\chi)$ is the Fourier transformed function in the inverse χ domain. Applying the forward Fourier transform to $r(x)$ and $s(x)$, multiplying by transformed functions $R(\chi)$ and $S^*(\chi)$ and performing a reverse Fourier transform on the product will generate a cross-correlation function between $r(x)$ and $s(x)$. The optimal shift, u_{op} , between the functions $r(x)$ and $s(x)$ is found at the maximums of $\text{Corr}(r, s)_u$. Recursive alignment is performed such that alignment proceeds from larger global

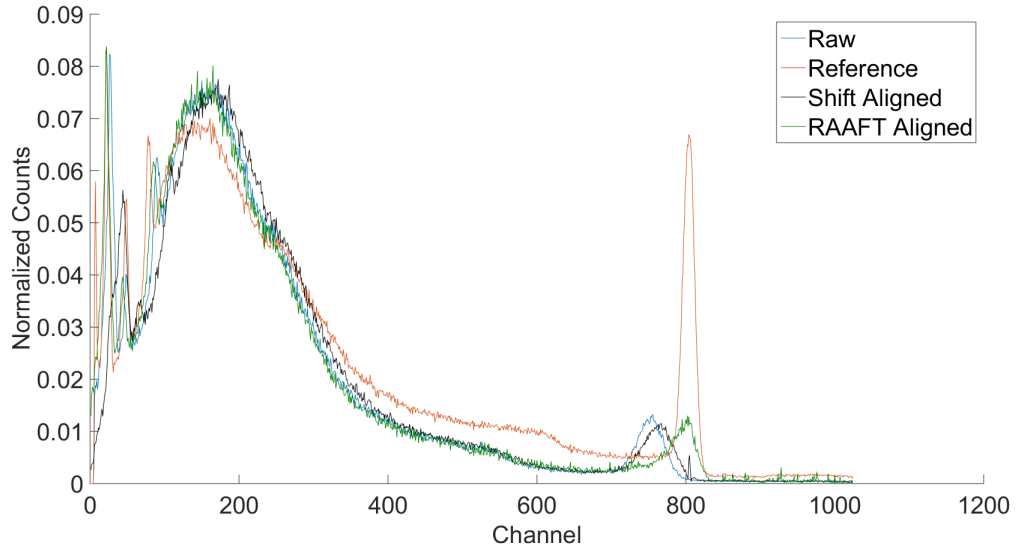


Figure 4.4: Treatment of non-linearities with RAAFT and simple alignment methods segments to smaller, more local segments. The algorithm for this procedure is detailed in [41].

4.5 Data Analysis

The following sections outline the benchmarks conducted to evaluate the performance of the MIP monitor. The benchmarks are conducted on a multiple materials under steady state and transient conditions.

4.5.1 Discrimination between UNF and MOX fuel samples

This analysis aims to discriminate between UNF samples (designated as Echo, Foxtrot, Golf, and Hotel) and MOX samples (designated as Alpha, Charlie and Delta). Discrimination of two different materials is relatively easy for the MIP system due to the significant differences between the materials. This would represent a scenario in which the operator does not correctly disclose the material being reprocessed. Since UNF and MOX have different discharge plutonium content this is of significant

importance. This model was trained on the MOX materials and the UNF materials were transformed into the reduced space using the loadings from the training data. As seen in Figure 4.5 the MIP monitor can distinguish between dissolved UNF and MOX fuel samples. Note the large degree of separation between UNF and MOX samples along PC 1, which carries the majority of the variance. Golf does not lie with the other UNF samples due to dissimilar uranium enrichment. Separation between materials on PC2 is less significant because it only carries 17.43% of the variation compared to PC1 which carries 52.91%.

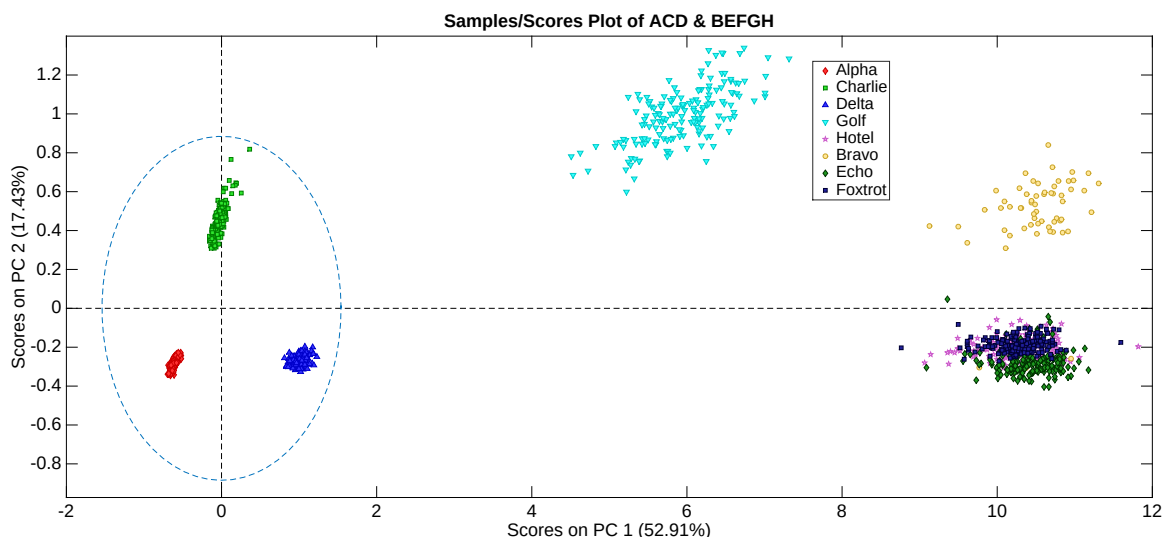


Figure 4.5: Discrimination between UNF and MOX materials using a PCA model trained on MOX fuel

The first model separates the UNF and MOX materials by using MOX as training data, therefore using the MOX loadings. The intensity of the loadings correspond to areas that contain significant information. In Figure 4.6 the loadings for a model trained on only MOX fuel and a model trained only on UNF material is shown. The low energy region shows a notable difference in the loadings indicating that the region may be more significant in discriminating the materials.

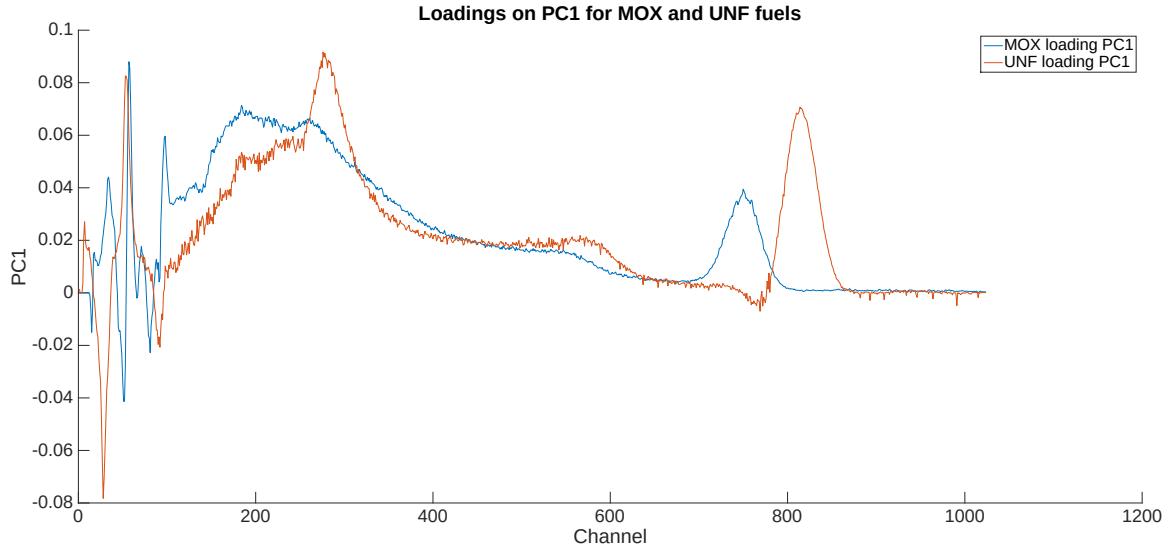


Figure 4.6: Loadings of a PCA model trained exclusively on UNF or MOX material

4.5.2 Classification of UNF samples with dissimilar Uranium isotope distribution

The goal of this analysis was to classify materials Golf and Hotel in a model that was trained on Bravo, Echo, and Foxtrot. Golf and Hotel are translated to PC space by multiplying the calibrated loadings by the input data (Section 3.3.2). Bravo, Echo, Foxtrot, and Hotel have similar enrichment. However, the Golf fuel sample has a much higher enrichment than the others. The goal is to train a model on three of the materials and to classify the other two. This is a parallel to a possible diversion scenario where HEU is hidden in different reprocessing stages. It is expected that Hotel will be classified with the other similar enrichment fuels while Golf will not. In the PC space, Golf clusters near Foxtrot inside the model (Figure 4.7), which was trained on Bravo, Echo, and Foxtrot. This is contradictory to the expected behavior, that Hotel would reside inside the model and Golf would not.

Unlike many previous cases, cluster analysis alone is not enough to correctly discriminate between the fuel materials. However, discrimination can be achieved

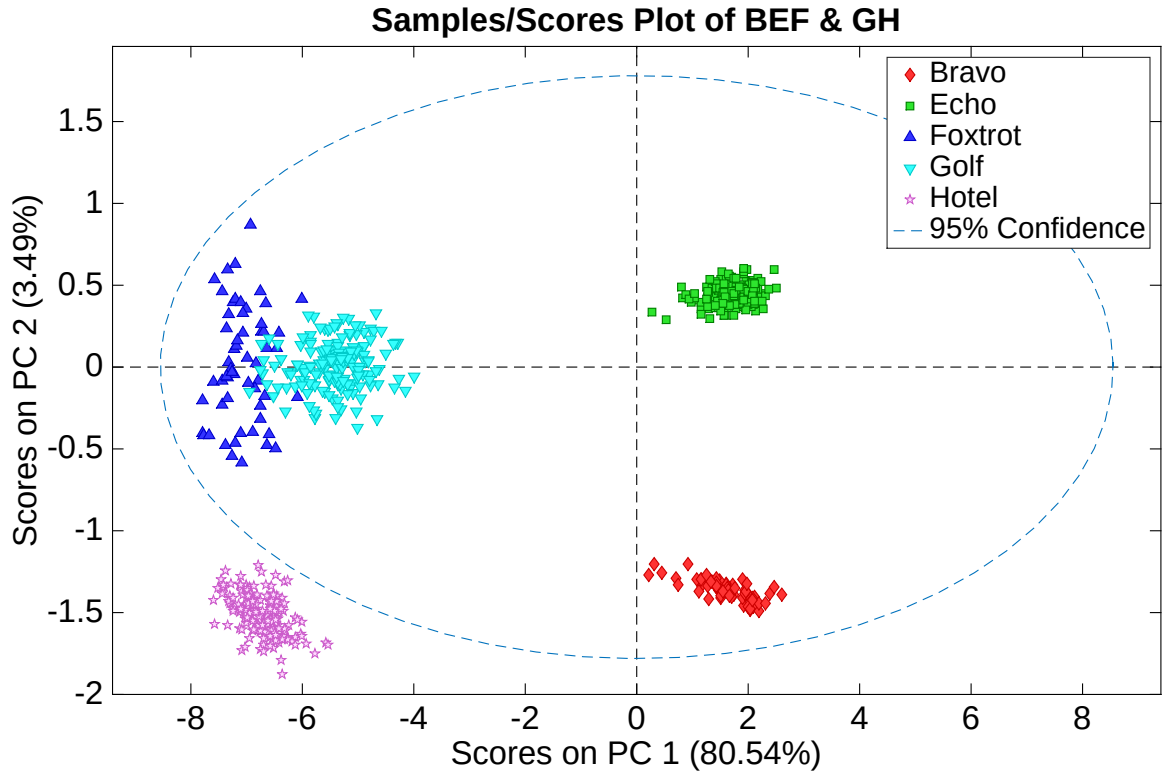


Figure 4.7: Cluster analysis in PC space is insufficient to discriminate UNF materials of dissimilar uranium enrichment

by utilizing the Q-residual. The Q-residual is a measure of how much of the sample is not captured by the model. As seen in Figure 4.8, Golf has a much higher Q-residual than the other samples. This implies that the Golf spectra is quite different from the other samples because a significant portion of the spectra is not captured by the model. It can be concluded that Golf's composition is different from the other samples in some way, in this case, the uranium enrichment.

The data from Hotel lies outside the confidence interval for the model, however, it lies within the threshold for the Q-statistics. This indicates that Hotel shares similar features to the training data (Bravo, Echo, and Foxtrot), but that those features are different. That is, Hotel has significant peaks in the same location as the training data, however, their relative ratios are different causing the PCA model to place Hotel outside the confidence interval.

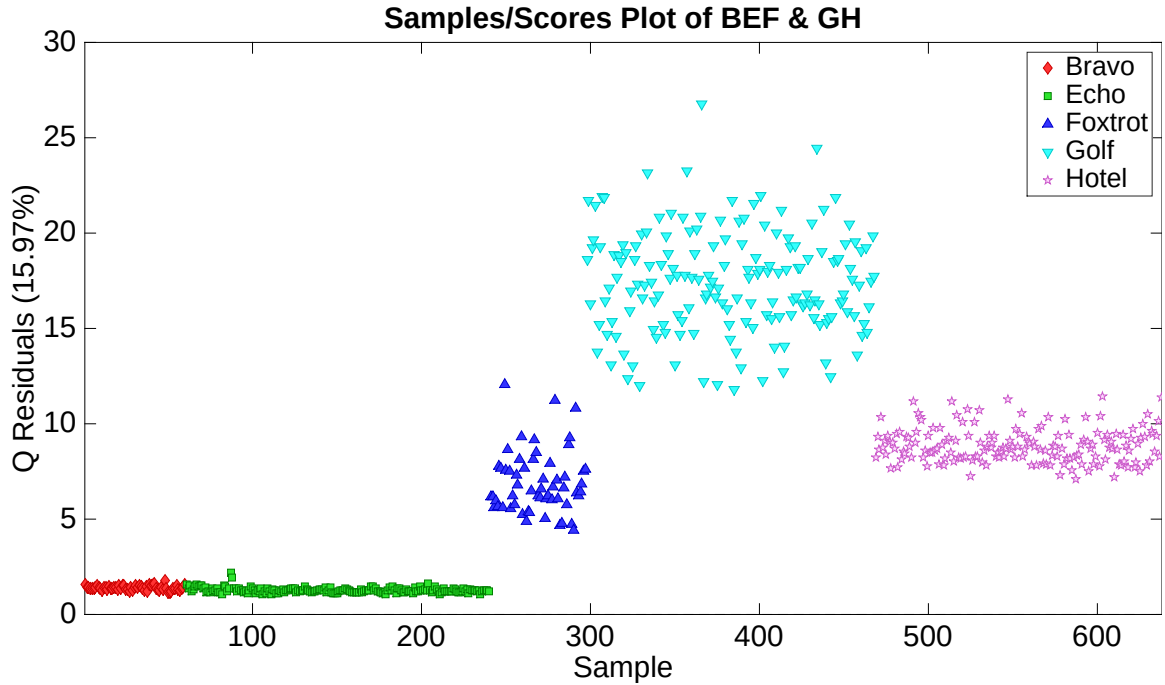


Figure 4.8: Separation of Q scores for different UNF materials

4.5.3 Classification of MOX samples with dissimilar Plutonium isotope distribution

Three Pu-samples have currently been collected: Alpha, Charlie, and Delta. Note that Delta is a composite of Alpha and Charlie material. Gamma spectra from these materials were analyzed for classification between samples. Data was collected during the mixing of Alpha and Charlie to form Delta. A PCA model was trained on Alpha and Charlie while Delta was transformed to the PC space. Because Delta combines the two Pu samples, it is expected to lie between the two in PC-space. Figure 4.9 shows the spectra of well-mixed Delta material projected onto the PC-space identified for Alpha and Charlie data. As expected, the Delta data (indicated with blue triangles) is classified inside the model with Alpha and Charlie. However, if the model is trained on a single MOX material it is able to correctly discriminate the three materials.

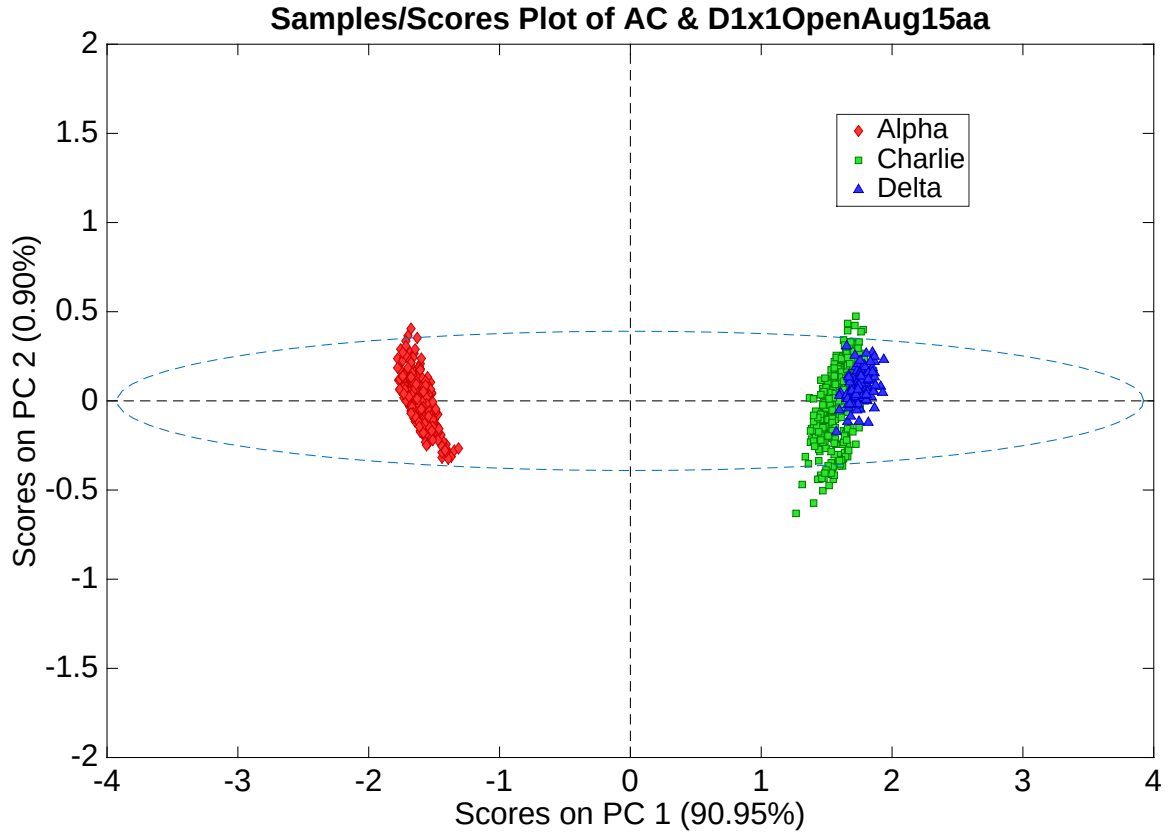


Figure 4.9: PCA model trained on Alpha and Charlie with Delta transposed to the reduced space

4.5.4 Discrimination of mixtures for detection threshold

The purpose of this analysis is to determine an approximate threshold for detection. The model was trained on the UNF material Golf. Various percentages of Delta, a MOX fuel sample, was added to the Golf spectra in order to simulate an off-normal process. The spectra were added in a linear sense that is various percentages of the average Delta spectrum was added to each Golf sample. This ensures that each individual observation is representative of an off-normal condition. In order to properly add Delta to Golf, it was necessary to ensure that the samples were aligned properly. Due to the alignment used it was ensured that samples of the same material would be aligned, (i.e., all Golf samples would be aligned to each other), however,

this does not imply Delta and Golf would be aligned to each other. There was a small shift between Golf and Delta spectra which was removed by using a simple shift targeted at the Golf peak around channel 25, seen in Figure 4.10. Note that while only one peak was targeted for alignment, another high energy peak was also aligned correctly (Figure 4.11) indicating that the RAFFT alignment resolved any non-linear shifts.

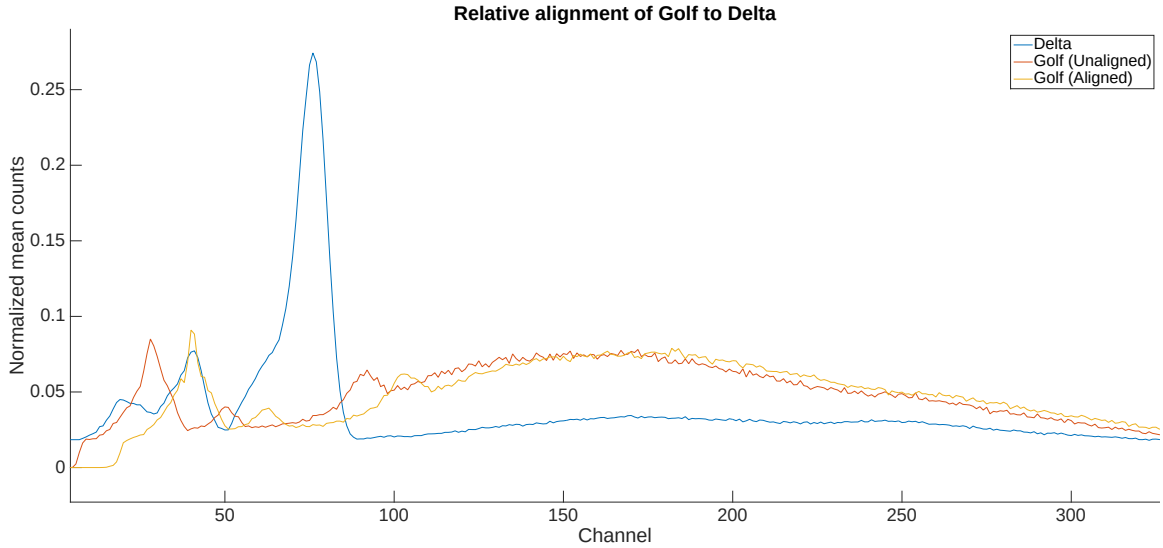


Figure 4.10: Relative of alignment of Golf to Delta with a simple alignment based on the peak around channel 25

When the model is trained on the expected fuel sample, discrimination via cluster analysis can be detected to as low as 15% added spectra (Figure 4.12), that is, discrimination of a pure spectra and a pure spectra with 15% spectral intensity added from another sample. Note that this model captures much less data (6.71%) than the model that was created in the previous section to classify Golf and Hotel. This is expected, because this model was trained on a single material rather than multiple, the variation that is being captured is among samples. The variance among samples is due to counting statistics and harder to capture than variation among different materials. However, despite the low percent of data captured, it is still sufficient to classify the data mixtures.

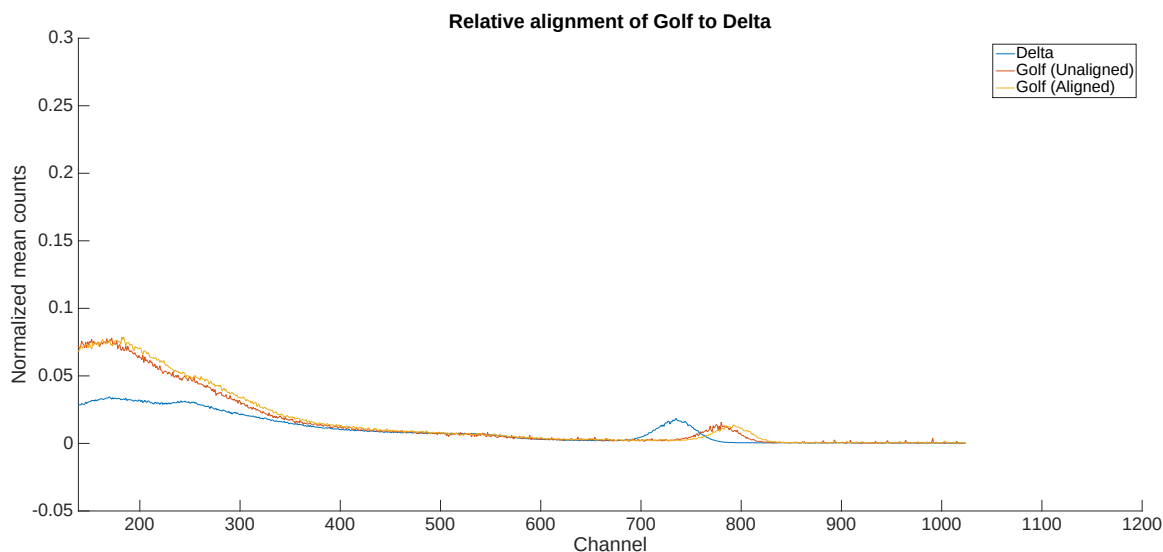


Figure 4.11: High energy peak is aligned despite not explicitly targeted

For security purposes, it is currently unknown what the corresponding physical detection threshold is. In the event that further resolution is needed, the PCA analysis can be targeted at a specific segment of interest in the spectra. In this case, the variation between the pure Golf and a mixture of Golf appears to be related to the ratio of the two smaller peaks in Figure 4.13. For the pure Golf sample the left peak (channel 20) is quite large compared to the right peak (channel 35). As more Delta is added, it is apparent that the ratio of the peaks is inversely proportional. That is, as the intensity of the peak at channel 20 decreases, the intensity of the peak at channel 75 increases. Since the primary changes in the spectra occur below channel 100, the PCA model is focused at the first 100 channels.

Cluster analysis is insufficient to discriminate at added ratios below 15%. However, the Q residuals do exhibit some degree of separation as seen in Figure 4.14. The degree of separation between the pure sample and the 10% mixture is likely sufficient to discriminate based on a simple threshold. That is, if the majority of the spectra are above the threshold the material is likely not pure golf. Below 10% added spectra the population begins to overlap with the pure golf sample. It is possible that by

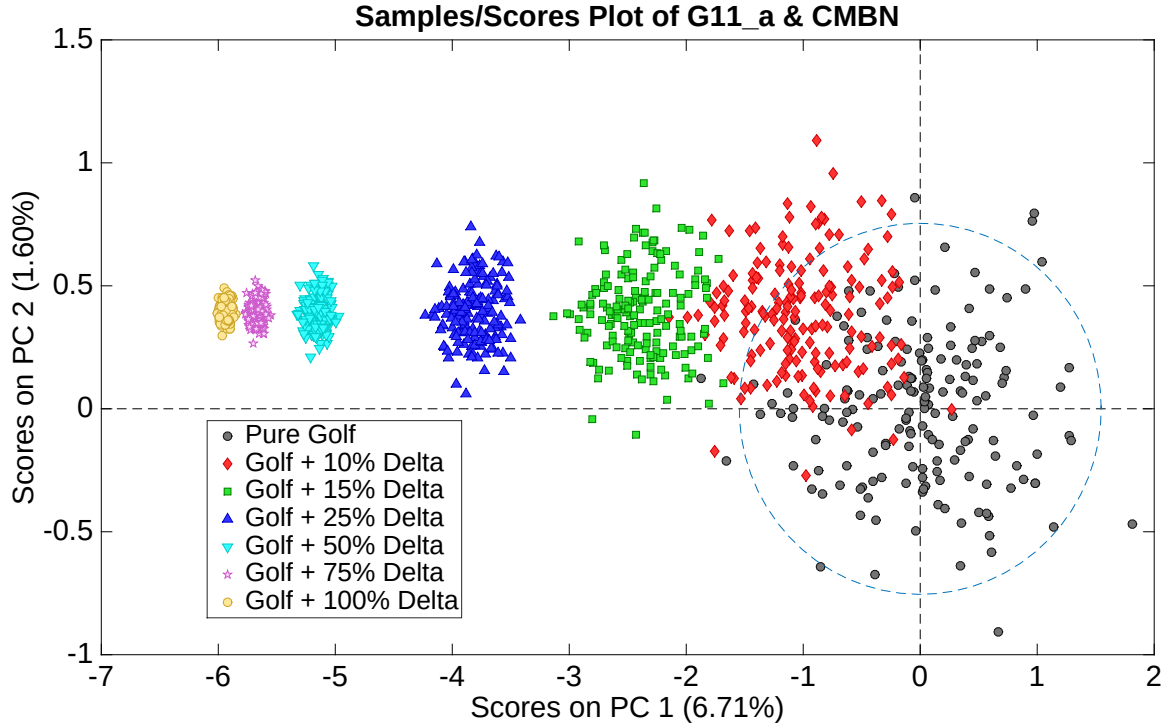


Figure 4.12: PCA model trained on a pure golf material with a variety of UNF mixtures

employing a sequential probability ratio test, which is discussed in Section 4.7.3, it would be possible to further enhance discrimination capability.

4.5.5 Observation of material transients

In a diversion scenario it is important to detect transients in the tank chemistry. Simulation of a diversion scenario is not feasible, however, transient sensitivity can be evaluated by cycling the fuel material. Past analysis has consisted of constant five second observations of a sample box cycling a fuel material at a steady state. In this test the sample box is cycled with cold, non fuel chemicals for the first ten minutes of observation. Once ten minutes have elapsed, the fuel material is cycled through the box. In order to observe this transient, a PCA model was trained on steady state Delta data. The Delta cycle data was then translated to the reduced PC space. As

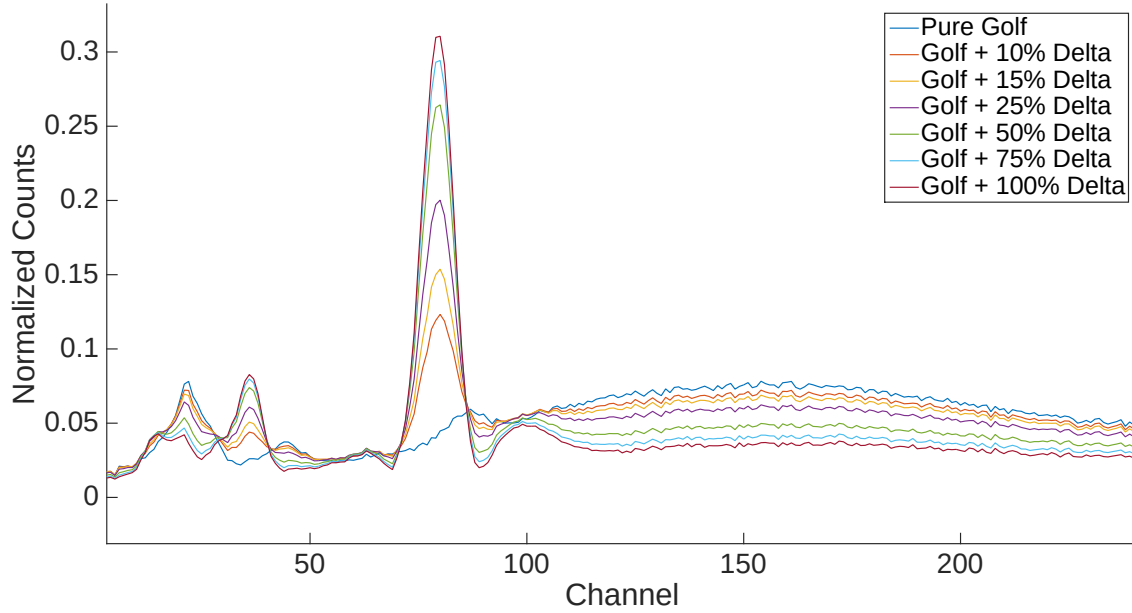


Figure 4.13: Changing peak ratios in gamma spectra of mixed UNF materials

seen in Figure 4.15, the cycle Delta material originally lies outside the confidence interval of PC1. However, when the material begins to cycle through the sample box, the observations transition inside the confidence bounds in PC space. This shows that the MIP monitor is likely capable of detecting transients as they occur.

4.6 Conclusions

The MIP monitor can sufficiently discriminate between different types of fuel, MOX and UNF. The distinctions between the populations are large and will enable accurate detection. The MIP monitor can also distinguish between different samples of the same fuel type. Depending on how the PCA model is trained, additional analysis may be required. Analyzing the Q statistics, as discussed in section 4.7.3 would be required in cases where discrimination based on uranium enrichment is desired. However, discrimination of MOX materials with dissimilar plutonium isotopic distributions requires only cluster analysis through the PCA model. (These additional analysis

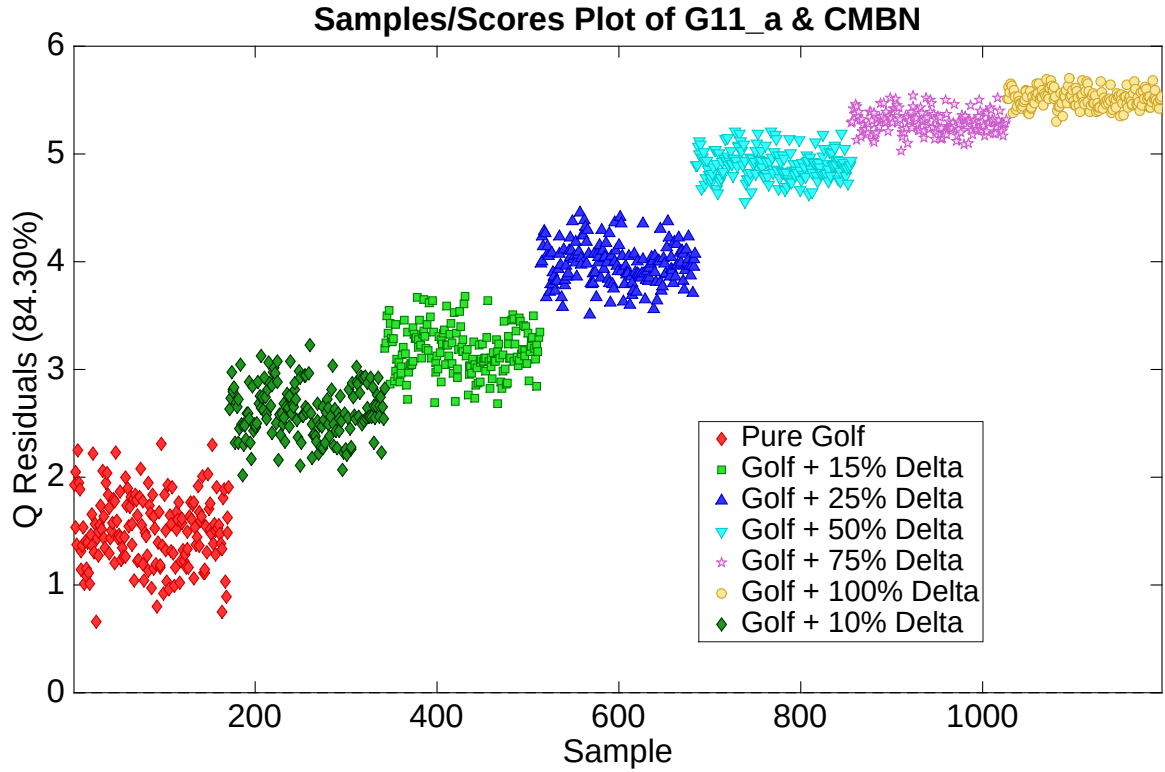


Figure 4.14: Q residuals for a PCA model trained on the first 100 channels of a pure golf material

requirements of the UNF materials could be explained with future chemical analysis.) It is possible that the uranium enrichment of the UNF fuel has a lesser effect on the gamma spectra than the plutonium isotopics of the MOX fuel. Note that chemical analysis would not be required for operations when the MIP monitor is deployed, rather, it would provide information to further improve data analysis.

4.7 Future Work

4.7.1 Need for global alignment

Presently, spectral alignment is done on a local scale. That is samples are aligned within material; Delta is aligned to a Delta reference, Charlie is aligned to a Charlie

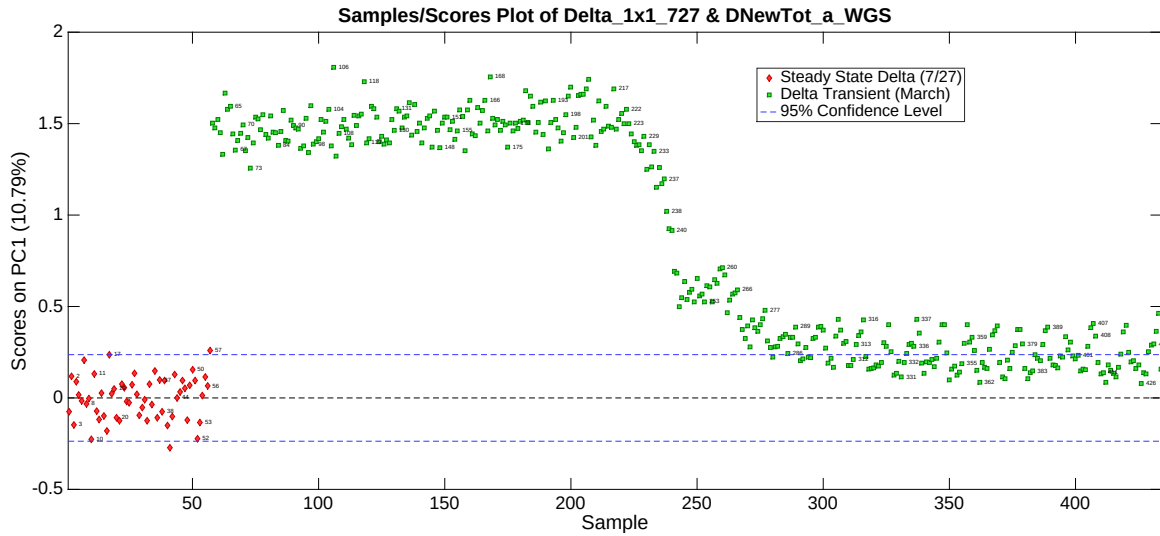


Figure 4.15: Cycling of Delta, first 10 minutes are observations of a non-fuel material reference, and so on. However, this does not guarantee peaks common to two different materials will be aligned to each other. In Figure 4.16 it is possible that the Golf peak at channel 40 should actually be aligned to channel 60. One possible solution would be to identify all of the peaks. This would require development of energy calibration curves for every material and subsequent peak identification. Peak identification would be challenging due to the multitude of gammas emitted from fission products and actinides present in the fuel. An easier solution would require the presence of a calibration source during operation of the MIP monitor. A known source would allow alignment to easily identified peaks. The MIP monitor does not require that peaks reside at a particular channel, but only that common peaks reside at the same channel. Ideally, a source with at least three easily identifiable photopeaks would be used, however, a correct alignment can be produced with only two peaks that span the energy range of interest. This method would also ensure that later material observations would align properly without any knowledge of the isotopic distribution. MIP does not require the identification of isotopes present in the tank solutions, only that a “normal” spectra be identified.

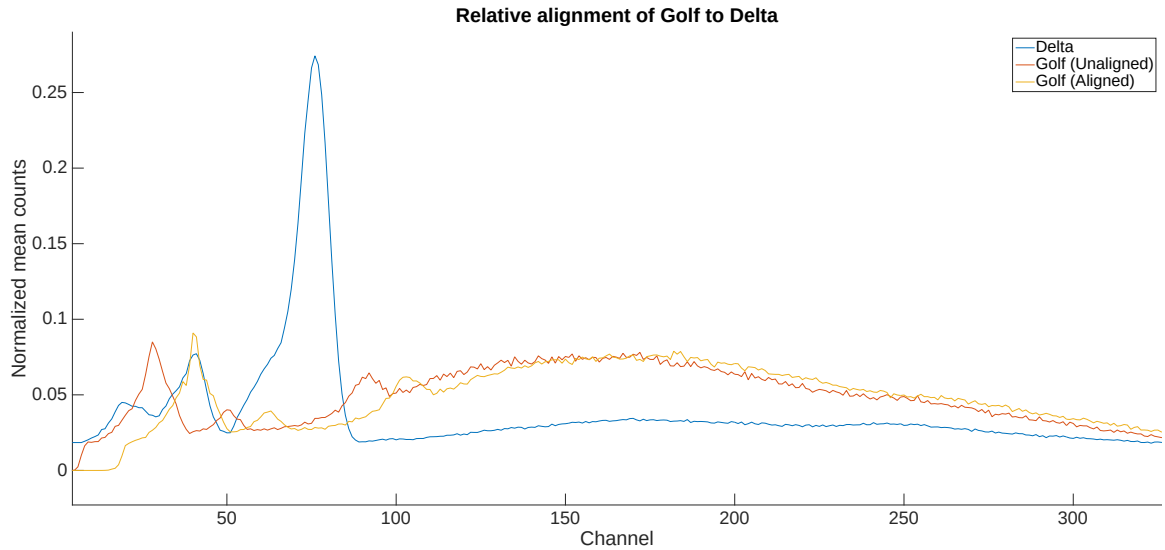


Figure 4.16: Possible peak mismatch at channel 40

4.7.2 Investigation in conjunction with chemical analysis

As mentioned previously, the MIP monitor does not require explicit knowledge of an isotopic distribution to correctly flag off-normal processes. However, it is important that the PCA model reflects a correct comparison. The PCA model would incorrectly judge that the variation between two materials is larger than the true value if not aligned correctly. That would be an example of a process that does not rely on any outside knowledge to resolve. However, there are multiple questions that could be answered if isotopic distributions and densities were obtained. A rough estimate of the detection threshold could be quantified. In section 4.5.3 the Delta mixing data was found to be inconsistent with the steady-state observation. Information about the fuel composition could provide insight into this discrepancy. Further, more specific information on the materials could validate current MIP findings, such as the classification of materials in Section 4.5.2.

4.7.3 Application of Q-stat discrimination

As mentioned in Sections 4.5.1 and 4.5.4 further analysis may require discrimination on the basis of the Q-residual. Many of the populations are well separated in the PCA models. However, the Q scores are often similar and not well discriminated. One possible method to enhance Q score discrimination would be the sequential probability ratio test (SPRT)[42]. Primarily, the test can compare a hypothesis and an alternative hypothesis. The test is able to determine which hypothesis should be confirmed. The advantage of the SPRT method is that a confirmation or rejection of the hypothesis can be made without testing the entire population. The test proceeds until a given quantity, typically the probability of false alarm, is reached. In the application of the MIP monitor the hypothesis would be developed to classify a set of observations into a population. That is, if we have an observation should it reside in the “normal” distribution or the off-normal distribution. This would require fitting a distribution to the “normal” data set. The most obvious choice would be to fit a Gaussian distribution to the “normal” data set and determine its associated mean and standard variation. Then, calculated scores would be tested to see if they fit in the given distribution. However, there is no guarantee that the Q-residuals or PC scores fit a normal distribution. Further work is required to test the viability of this approach. Currently, PCA is the primary method used to discriminate between the fuel materials. It is possible employing PLS could provide a better discrimination than PCA. This would eliminate the need for discrimination on Q-statistics.

Chapter 5

Conclusions and Final Remarks

The new CANDU lattice and cross-section libraries produce results that are in good agreement with experimental assay values. These validated templates and libraries will provide SCALE users an excellent starting point for CANDU depletion studies. Destructive assays are expensive and time consuming. Accurate depletion calculations are essential for proper disposal or reprocessing of assemblies. Many users will use the base templates to do these calculations with minimal modification. While the CANDU assembly does not have significant variation (there are no burnable rods, no guide tubes, constant enrichment, etc), PWR systems have a variety of parameters that vary from plant to plant. The second study provided in this thesis indicates that the unmodified templates, which are similar to ORIGEN depletion cases, work well for systems without significant flux and spectral variations. However, in the presence of features that effect flux shape, a more detailed model is required for accurate calculations. Once the isotopic content of assemblies have been calculated, they must be either buried or reprocessed. Analysis presented in this thesis demonstrates the ability of the MIP monitor to discriminate between a number of steady-state materials or transients. Additionally, the MIP monitor has a good detection threshold of 15% of a material diverted using a NaI(Tl) detector with five-second counting intervals.

Bibliography

- [1] I. C. Gauld, K. A. Litwin, [Verification and Validation of the ORIGEN-S Code and Nuclear Data Libraries](#), Tech. Rep. RC-1429 / COG-I-95-150, Atomic Energy of Canada Limited (AECL) (August 1995).
URL <http://web.ornl.gov/sci/scale/pubs/rc1429.pdf> x, xii, 6, 7, 8, 9, 10, 12, 15, 16, 18, 19, 20
- [2] I. Gauld, P. Carlson, K. Litwin, Production and Validation of ORIGEN-S Cross-Section Libraries for CANDU Reactor Fuel Studies, Tech. Rep. RC-1442; COG-I-95-200, Whiteshell Laboratories (October 1995). x, xii, xiii, 6, 7, 8, 9, 10, 12, 16, 18, 19, 20, 30
- [3] M. Chadwick, P. Obložinský, M. Herman, N. Greene, R. McKnight, D. Smith, P. Young, R. MacFarlane, G. Hale, S. Frankle, A. Kahler, T. Kawano, R. Little, D. Madland, P. Moller, R. Mosteller, P. Page, P. Talou, H. Trellue, M. White, W. Wilson, R. Arcilla, C. Dunford, S. Mughabghab, B. Pritychenko, D. Rochman, A. Sonzogni, C. Lubitz, T. Trumbull, J. Weinman, D. Brown, D. Cullen, D. Heinrichs, D. McNabb, H. Derrien, M. Dunn, N. Larson, L. Leal, A. Carlson, R. Block, J. Briggs, E. Cheng, H. Huria, M. Zerkle, K. Kozier, A. Courcelle, V. Pronyaev, S. van der Marck, [ENDF/B-VII.0: Next Generation Evaluated Nuclear Data Library for Nuclear Science and Technology](#), Nuclear Data Sheets 107 (12) (2006) 2931–3060, Evaluated Nuclear Data File ENDF/B-VII.0. doi:<http://dx.doi.org/10.1016/j.nds.2006.11.001>.
URL <http://www.sciencedirect.com/science/article/pii/S0090375206000871> xii, xiii, 23, 24, 25, 26, 27

- [4] Cross-Section Evaluation Working Group, ENDF/B-VI Summary Documentation, Tech. Rep. BNL-NCS-17541 (ENDF-201), Data Library ENDF/B-VI, update 1998, by the U.S. National Nuclear Data Center on behalf of the Cross-Section Evaluation Working Group (1998). [xii](#), [xiii](#), [23](#), [24](#), [26](#), [27](#)
- [5] Oak Ridge National Laboratory, SCALE: A Comprehensive Modeling and Simulation Suite for Nuclear Safety Analysis and Design, Version 6.1 (June 2011). [5](#)
- [6] S. Bowman, SCALE 6: Comprehensive Nuclear Safety Analysis Code System, Nuclear Technology 174 (2) (2011) 126–148. [5](#), [6](#), [38](#)
- [7] I. C. Gauld, G. Radulescu, G. Ilas, B. D. Murphy, M. L. Williams, D. Wiarda, Isotopic Depletion and Decay Methods and Analysis Capabilities in SCALE, Nuclear Technology 174 (2) (2011) 169–195. [5](#), [6](#), [22](#), [39](#)
- [8] J. Tait, I. Gauld, A. H. Kerr, [Validation of the ORIGEN-S Code for Predicting Radionuclide Inventories in Used CANDU Fuel](#), Journal of Nuclear Materials 223 (2) (1995) 109–121.
URL <http://www.sciencedirect.com/science/article/pii/S0022311594006830> [6](#), [9](#), [16](#)
- [9] M. D. DeHart, S. M. Bowman, Reactor Physics Methods and Analysis Capabilities in SCALE, Nuclear Technology 174 (2) (2011) 196–213. [6](#)
- [10] S. Goluoglu, L. M. Petrie, M. E. Dunn, D. F. Hollenbach, B. T. Rearden, Monte Carlo Criticality Methods and Analysis Capabilities in SCALE, Nuclear Technology 174 (2) (2011) 214–235. [7](#)
- [11] C. H. Chung, K. B. Oh, C. H. Kim, Y. H. Chang, S. Y. Kim, J. S. Kim, [Nuclear Design Analysis of Wolsung-1 CANDU-PHW Nuclear Generating Station](#), Journal of the Korean Nuclear Society 10 (4) (1978) 203–213.

- URL <http://www.kns.org/jknsfile/v10/A04803288321.pdf?PHPSESSID=a078b088435d92a883ed144a4b241782> 12
- [12] S. Goluoglu, D. F. Hollenbach, N. F. Landers, L. M. Petrie, J. A. Bucholz, C. F. Weber, C. M. Hopper, The Material Information Processor for SCALE, Oak Ridge National Laboratory (June 2011). 12
- [13] H. Smith, Modeling depletion simulations for a high-burnup, highly heterogeneous bwr fuel assembly with scale, in: PHYSOR - Advances in Reactor Physics, American Nuclear Society, 2012. 13
- [14] C. McGraw, G. Ilas, [Pressurized Water Reactor ENDF/B-VII Cross-Section Libraries for ORIGEN-ARP](#), Nuclear Technology 183 (3) (2013) 436–445.
URL http://www.ans.org/pubs/journals/nt/a_19431 15
- [15] G. Ilas, I. C. Gauld, G. Radulescu, [Validation of new depletion capabilities and ENDF/B-VII data libraries in SCALE](#), Annals of Nuclear Energy 46 (2012) 43–55.
URL <http://dx.doi.org/10.1016/j.anucene.2012.03.012> 16, 18, 20, 33
- [16] M. Basunia, R. Firestone, Z. Révay, H. Choi, T. Belgia, J. Escher, A. Hurt, M. Kr̕t̕čka, L. Szentmiklósi, B. Sleaford, N. Summers, Determination of the $^{151}\text{Eu}(n,\gamma)^{152m1,g}\text{Eu}$ and $^{153}\text{Eu}(n,\gamma)^{154}\text{Eu}$ Reaction Cross Sections at Thermal Neutron Energy, Nuclear Data Sheets 119 (2014) 88–90. 22
- [17] M. Chadwick, M. Herman, P. Obložinský, M. Dunn, Y. Danon, A. Kahler, D. Smith, B. Pritychenko, G. Arbanas, R. Arcilla, R. Brewer, D. Brown, R. Capote, A. Carlson, Y. Cho, H. Derrien, K. Guber, G. Hale, S. Hoblit, S. Holloway, T. Johnson, T. Kawano, B. Kiedrowski, H. Kim, S. Kunieda, N. Larson, L. Leal, J. Lestone, R. Little, E. McCutchan, R. MacFarlane, M. MacInnes, C. Mattoon, R. McKnight, S. Mughabghab, G. Nobre, G. Palmiotti, A. Palumbo, M. Pigni, V. Pronyaev, R. Sayer,

- A. Sonzogni, N. Summers, P. Talou, I. Thompson, A. Trkov, R. Vogt, S. van der Marck, A. Wallner, M. White, D. Wiarda, P. Young, [ENDF/B-VII.1 Nuclear Data for Science and Technology: Cross Sections, Covariances, Fission Product Yields and Decay Data](#), Nuclear Data Sheets 112 (12) (2011) 2887–2996, special Issue on ENDF/B-VII.1 Library. doi:<http://dx.doi.org/10.1016/j.nds.2011.11.002>.
URL <http://www.sciencedirect.com/science/article/pii/S009037521100113X> 23, 26
- [18] T. Sekine, S.-I. Ichikawa, S. Baba, Triple Neutron Capture of ^{153}Eu in a Reactor: The Cross Sections of ^{154}Eu and ^{155}Eu , International Journal of Radiation Applications and Instrumentation. Part A. Applied Radiation and Isotopes 38 (7) (1987) 513–516. 25
- [19] S. F. Mughabghab, Atlas of Neutron Resonances, Fifth Edition: Resonance Parameters and Thermal Cross Sections. Z=1-100. 25
- [20] G. Radulescu, I. C. Gauld, G. Ilas@TechReportenrich, Title = Technical Support for a Proposed Decay Heat Guide Using SAS2H/ORIGEN-S Data, Author = O.W. Hermann and C. V. Parks and J. P. Renier, Institution = Oak Ridge National Laboratory, Year = 1994, Owner = Nathan, Timestamp = 2015.06.04 , Scale 5.1 predictions of pwr spent nuclear fuel isotopic compositions, Tech. Rep. ORNL/TM-2010/44, Oak Ridge National Laboratory (2010). 39, 40, 45, 47, 48
- [21] PLSToolbox, 7.9.4(19353), Eigenvector Research Inc, 2015. 40
- [22] MATLAB, 2015a (8.0.5.197613), The Mathworks Inc, Natick, Massachusetts, 2015. 40
- [23] K. R. Bebbe, R. J. Pell, M. B. Seasholtz, Chemometrics: A Practical Guide, John Wiley & Sons, Inc, 1998. 42

- [24] E. Malinowski, Factor Analysis in Chemistry, John Wiley & Sons, Inc, 2008. 42, 57
- [25] H. Abdi, L. J. Williams, Partial least squares methods: Partial least squares correlation and partial least square regression, Tech. rep., University of Texas - Dallas (2013).
URL http://www.utd.edu/~herve/abdi-PLSC_and_PLSR2012.pdf 43
- [26] P. Geladi, B. R. Kowalski, Partial least-squares regression: a tutorial, *Analytica Chimica Acta* 185 (1986) 1–17. 43
- [27] H. Blalock, Quantitative Sociology: International Perspectives on Mathematical and Statistical Modeling, Academic Press, Inc., 1975. 44
- [28] B. G. M. Vandeginste, C. Sielhorst, M. Gerritsen, The nipals algorithm for the calculation of the principal components of a matrix, *Trends in analytical chemistry* 7 (8) (1988) 286–287. 44
- [29] Standard test method for atom percent fission in uranium and plutonium fuel (neodymium-148 method). 46
- [30] J. M. Scaglione, Three mile island unit 1 radiochemical assay comparisons to sas2h calculations, Tech. rep., WPNCS Expert Group on Assay Data of Spent Nuclear Fuel (2002). 49
- [31] F. C. D. G. Ilas, I. C. Gauld, M. B. Emmett, Analysis of experimental data for high burnup pwr spent fuel isotopic validation - calvert cliffs, takahama, and three mile island reactors, Tech. rep., U.S. NRC (2010). 49
- [32] I. Gauld, J. Giaquinto, J. Delashmitt, J. Hu, G. Ilas, T. Haverlock, C. Romano, Re-evaluation of spent nuclear fuel assay data for the three mile island unit 1 reactor and application to code validation, *Annals of Nuclear Energy* 87 (2016) 267–281. doi:10.1016/j.anucene.2015.08.026. 49

- [33] K. J. Dayman, Multivariate analysis applied to the characterization of spent nuclear fuel, Master's thesis, The University of Texas at Austin (2012). [51](#)
- [34] C. R. Orton, C. G. Fraga, R. N. Christensen, J. M. Schwantes, Proof of concept experiments of the multi-isotope process monitor: An online, nondestructive, near real-time monitor for spent nuclear fuel reprocessing facilities, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment (672) (2012) 38–45. [doi:doi:10.1016/j.nima.2011.12.083](#). [56](#), [57](#)
- [35] C. R. Orton, C. G. Fraga, R. N. Christensen, J. M. Schwantes, Proof of concept simulations of the multi-isotope process monitor: An online, nondestructive, near-real-time safeguards monitor for nuclear fuel reprocessing facilities, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 629 (1) (2011) 209–219. [doi:doi:10.1016/j.nima.2010.10.024](#). [56](#), [57](#)
- [36] IAEA, Iaea safeguards glossary, Tech. rep., International Atomic Energy Agency (2001). [57](#)
- [37] P. Geladi, B. R. Kowalski, Partial least-squares regression - a tutorial, Analytica Chimica Acta 185 (1986) 1–171. [doi:10.1016/0003-2670\(86\)80028-9](#). [57](#)
- [38] M. Balcilar, A. C. Sonmez, Background estimation method with incremental iterative re-weighted least squares, Signal, Image and Video Processing 10 (1). [60](#)
- [39] P. H. C. Eilers, H. F. M. Boelens, Baseline correction with asymmetric least squares smoothing, Tech. rep., Swammerdam Institute for Life Sciences (SILS) - University of Amsterdam (2005). [60](#)
- [40] P. H. C. Eilers, A perfect smoother, Analytical Chemistry 75 (14) (2003) 3631–3636. [doi:10.1021/ac034173t](#). [60](#)

- [41] J. W. H. Wong, C. Durante, H. M. Cartwright, Application of fast fourier transform cross-correlation for the alignment of large chromatographic and spectral datasets, *Analytical Chemistry* 77 (2005) 5655–5661. [61](#), [64](#)
- [42] A. Wald, Sequential test of statistical hypotheses, *Annals of Mathematical Statistics* 16 (2) (1945) 117–186. [77](#)

Appendix

Appendix A

A.1 CANDU 28-element input for lattice validation (TRITON/NEWT)

```
=t-depl parm=(centrm, addnux=3)
candu 28-element assembly
v7-238
'
' -----
'   template to generate libraries for ORIGEN-S
'   parameters are:
'u235wt%      - wt% U235
'u234wt%      - wt% U234
'u236wt%      - wt% U236
'u238wt%      - wt% U238
'ddd          - coolant density (g/cc)
'Dancoff1     - Dancoff factor 1
'Dancoff2     - Dancoff factor 2
'Dancoff3     - Dancoff factor 3
'namelibrary  - name of generated ORIGEN library
'specpow      - average specific power
'daystoburn   - depletion interval in days
' options are:  name          - candu 28 element
'enrichment   - 0.711 (natural)
'cool. density - 0.8445
'Dancoff1     - 0.6770
'Dancoff2     - 0.6650
'Dancoff3     - 0.4260
'spec. power  - 29.203
'burnups      -
' -----
'   Documentation and Notes:
'   - Information from "Verification and Validation of ORIGEN-S Code
'     and Nuclear Data Libraries" ORNL RC-1429, COG-I-95-150
'
'   - Validation of the ORIGEN-S code for predicting radionuclide
'     inventories in used CANDU fuel DOI:10.10.59/0022-3115(94)00683-0
'
'   - Nuclear Design Analysis of Wolsung-1 CANDU-PHW Nuclear
'     Generating Station IAEA RN:11522694
'
'   - Air gap was assumed to be air -- was not specified in documents
'     as helium or another inert gas. Effects of using helium explored
'     in report.
'
'   - Note that d2o can be called directly if desired, calling d2o
'     as a composition of deuterium and oxygen not necessarily required
'   - by N. T. Shoman
' -----
' -----
'   Mixture data
' -----
read comp
' fuel (natural uranium) -- Inner Ring
uo2      1 den=10.59 1 1003 92234 u234wt% 92235 u235wt% 92236 u236wt% 92238 u238wt% end
' clad (Zircaloy)
```

```

    zirc4 2 1 569 end
,
' pressure tube (Zr-Nb 2.5%)
  wtptclad 3 8.573 2 40000 97.5 41093 2.5 1 559 end
,
' air gap (air fill gas -- not specified)
  he 4 den=0.001205 1.0 559 end
,
' d2o coolant (99.75% purity d2o)
  d2o 5 den=ddd 0.9975 545 end
  h2o 5 den=ddd 0.0025 545 end
,
' calandria tube
  zirc4 6 1 360 end
,
' d2o moderator (99.91% purity d2o -- Outside calandria)
  d2o 7 den=ddd2 0.9991 336 end
  h2o 7 den=ddd2 0.0009 336 end
,
' -----
' Additional materials for self-shielding
' -----
' clad
  zirc4 21 1 569 end
,
' clad
  zirc4 22 1 569 end
,
' d2o coolant (99.75% purity d2o -- Inside of pressure tube)
  d2o 51 den=ddd 0.9975 545 end
  h2o 51 den=ddd 0.0025 545 end
,
' d2o coolant (99.75% purity d2o -- Inside of pressure tube)
  d2o 52 den=ddd 0.9975 545 end
  h2o 52 den=ddd 0.0025 545 end
,
' fuel -- Middle Ring
  uo2 10 den=10.59 1 1003 92234 u234wt% 92235 u235wt% 92236 u236wt% 92238 u238wt% end
' fuel -- Outer Ring
  uo2 20 den=10.59 1 1003 92234 u234wt% 92235 u235wt% 92236 u236wt% 92238 u238wt% end
,
end comp
' -----
' Cell data
' -----
read celldata
  latticecell triangpitch fuelr=0.7120 1 cladr=0.7605 2 pitch=1.9125 5 end
  centrmdata dan2pitch(1)=0.7145 end centrmdata
  latticecell triangpitch fuelr=0.7120 10 cladr=0.7605 21 pitch=1.9283 51 end
  centrmdata dan2pitch(10)=0.7074 end centrmdata
  latticecell triangpitch fuelr=0.7120 20 cladr=0.7605 22 pitch=2.6433 52 end
  centrmdata dan2pitch(20)=0.4480 end centrmdata
end celldata
' -----
' Depletion data
' -----
read depletion
  1 10 20 end
end depletion

```

```

' -----
' Burn data
' -----

read burndata
  power=specpower burn=daystoburn down=0 end
end burndata

read opus
  units=gatoms
  typarms=nucl
  symnuc=u-235 u-236 u-238 pu-238 pu-239 pu-240 pu-241 pu-242 end
  matl=20 end

newcase
  units=becq
  typarms=nucl
  symnuc=h-3 sr-90 tc-99 ru-106 sb-125 i-129 cs-134 cs-137
    eu-154 eu-155 np-237 am-241 cm-244 end
end opus

' -----
' NEWT model data
' -----

read model
candu 28-element

read parm
  echo=yes
  drawit=no
  epsinner=-1.5e-4
  cmfd=yes
  xycmfd=4
  epsilon=8.0e-04
end parm

read materials
  mix=1 pn=1 com='inner fuel ring' end
  mix=2 pn=1 com='clad' end
  mix=3 pn=1 com='pressure tube' end
  mix=4 pn=1 com='air gap' end
  mix=5 pn=2 com='coolant' end
  mix=6 pn=1 com='calandria' end
  mix=7 pn=2 com='moderator' end
  mix=10 pn=1 com='middle fuel ring' end
  mix=20 pn=1 com='outer fuel ring' end
end materials

read geom
  unit 1
    com='fuel element (inner)'
    cylinder 1 0.7120
    cylinder 4 0.7605
    media 1 1 1
    media 2 1 -1 4
    boundary 4 5 5
  unit 10
    com='fuel element (middle)'
    cylinder 1 0.7120
    cylinder 4 0.7605
    media 10 1 1
    media 2 1 -1 4
    boundary 4 5 5
  unit 20
    com='fuel element (outer)'
    cylinder 1 0.7120

```

```

cylinder 4 0.7605
media 20 1 1
media 2 1 -1 4
boundary 4 5 5
unit 300
com='pressure tube'
cylinder 101 5.1815 sides=20
cylinder 102 5.6965 sides=20
media 5 1 101
media 3 1 -101 102
com='air gap and calandria'
cylinder 103 6.5405 sides=20
cylinder 104 6.6955 sides=20
media 4 1 -102 103
media 6 1 -103 104
com='fuel elements ring 1'
com='1' hole 1 origin x=0.83085 y=0.83085
com='2' hole 1 origin x=-0.83085 y=0.83085
com='3' hole 1 origin x=-0.83085 y=-0.83085
com='4' hole 1 origin x=0.83085 y=-0.83085
com='fuel elements ring 2'
com='5' hole 10 origin x=2.480617 y=1.027505
com='6' hole 10 origin x=1.027505 y=2.480617
com='7' hole 10 origin x=-1.02751 y=2.480617
com='8' hole 10 origin x=-2.48062 y=1.027505
com='9' hole 10 origin x=-2.48062 y=-1.027505
com='10' hole 10 origin x=-1.02751 y=-2.480616
com='11' hole 10 origin x=1.027505 y=-2.48062
com='12' hole 10 origin x=2.480617 y=-1.02751
com='fuel elements ring 3'
com='13' hole 20 origin x=4.147741 y=0.825037
com='14' hole 20 origin x=3.516285 y=2.349507
com='15' hole 20 origin x=2.349507 y=3.516285
com='16' hole 20 origin x=0.825037 y=4.147741
com='17' hole 20 origin x=-0.82504 y=4.147741
com='18' hole 20 origin x=-2.34951 y=3.516258
com='19' hole 20 origin x=-3.51628 y=2.349507
com='20' hole 20 origin x=-4.14774 y=0.825037
com='21' hole 20 origin x=-4.14774 y=-0.825037
com='22' hole 20 origin x=-3.51628 y=-2.34951
com='23' hole 20 origin x=-2.34951 y=-3.51628
com='24' hole 20 origin x=-0.82504 y=-4.14774
com='25' hole 20 origin x=0.82504 y=-4.14774
com='26' hole 20 origin x=2.349507 y=-3.51628
com='27' hole 20 origin x=3.516258 y=-2.34951
com='28' hole 20 origin x=4.147741 y=-0.82504
boundary 104 15 15
global unit 400
cuboid 1 4p14.2875
media 7 1 1
hole 300
boundary 1 25 25
end geom
read bounds
all=refl
end bounds
end model
end
~

```

A.2 CANDU 37-element input for lattice validation (TRITON/NEWT)

```
=t-depl parm=(centrm, addnux=3)
CANDU 37-element assembly
v7-238
'
' -----
'   template to generate libraries for ORIGEN-S
'   parameters are:
'   u235wt%      - wt% U235
'   u234wt%      - wt% U234
'   u236wt%      - wt% U236
'   u238wt%      - wt% U238
'   ddd          - coolant density (g/cc)
'   Dancoff1     - Dancoff factor 1
'   Dancoff2     - Dancoff factor 2
'   Dancoff3     - Dancoff factor 3
'   Dancoff4     - Dancoff factor 4
'   namelibrary  - name of generated ORIGEN library
'   specpow      - average specific power
'   daystoburn   - depletion interval in days
'   options are:
'               name      - candu 37 element
'               enrichment - 0.711 (Natural)
'               cool. density - 0.8360
'               Dancoff1    - 0.7630
'               Dancoff2    - 0.7610
'                       Dancoff3      - 0.7280
'                       Dancoff4      - 0.4200
'               spec. power - 19.564
'               burnups     -
'
'
' -----
'   Documentation and Notes:
'   - Information from "Verification and Validation of ORIGEN-S Code
'     and Nuclear Data Libraries" ORNL RC-1429, COG-I-95-150
'
'   - Validation of the ORIGEN-S code for predicting radionuclide
'     inventories in used CANDU fuel DOI:10.1016/0022-3115(94)00683-0
'
'   - Nuclear Design Analysis of Wolsung-1 CANDU-PHW Nuclear
'     Generating Station IAEA RN:11522694
'
'   - Air gap was assumed to be air -- was not specified in documents
'     as helium or another inert gas. Effects of using helium explored
'     in report.
'
'   - Note that d2o can be called directly if desired, calling d2o
'     as a composition of deuterium and oxygen not necessarily required
'   - by N. T. Shoman
'
' -----
'   Mixture data
```

```

' -----
read comp
'fuel (natural uranium) --- Center Pin
uo2      1 den=10.6 1 960 92234 u234wt%
          92235 u235wt%
          92236 u236wt%
          92238 u238wt% end

' clad (Zircaloy)
zirc4    2 1 569 end

' pressure tube (Zr-Nb 2.5%)
wtptclad    3 8.573 2
             40000 97.5
             41093 2.5
             1 559 end

' air gap (gas not specified, air)
c          4 den=0.001205 0.0124 559 end
n          4 den=0.001205 76.83538 559 end
o          4 den=0.001205 23.1781 559 end

'Coolant (99.75% pure d2o --- Inside pressure tube)
d2o 5 den=ddd 0.9975 561 end
h2o 5 den=ddd 0.0025 561 end
'calandria tube
zirc4      6 1 360 end

'd2o moderator (99.91% pure d2o --- Outside Calandria)
d2o 7 den=ddd2 0.9991 346 end
h2o 7 den=ddd2 0.0009 346 end

' -----
' Additional mixtures for self-shielding calculations
' -----
' clad
zirc4    21 1 569 end

' clad
zirc4    22 1 569 end

' clad
zirc4    23 1 569 end

'd2o coolant --- Inside Pressure Tube
d2o 51 den=ddd 0.9975 561 end
h2o 51 den=ddd 0.0025 561 end
,
d2o 52 den=ddd 0.9975 561 end
h2o 52 den=ddd 0.0025 561 end
,
d2o 53 den=ddd 0.9975 561 end
h2o 53 den=ddd 0.0025 561 end
,
'fuel --- Inner Ring
uo2      10 den=10.6 1 960 92234 u234wt%
          92235 u235wt%
          92236 u236wt%

```

```

          92238  u238wt% end
'fuel -- Middle Ring
uo2      20 den=10.6 1 960 92234 u234wt%
          92235 u235wt%
          92236 u236wt%
          92238 u238wt% end
'-fuel -- Outer Ring
uo2      30 den=10.6 1 960 92234 u234wt%
          92235 u235wt%
          92236 u236wt%
          92238 u238wt% end

end comp
',
' Cell data
',

read celldata
latticecell triangpitch fuelr=0.6075 1 cladr=0.6540 2 pitch=1.6058 5 end
centrm data
dan2pitch(1)=0.7630
end centrm
latticecell triangpitch fuelr=0.6075 10 cladr=0.6540 21 pitch=1.6100 51 end
centrm data
dan2pitch(10)=0.7610
end centrm
latticecell triangpitch fuelr=0.6075 20 cladr=0.6540 22 pitch=1.6807 52 end
centrm data
dan2pitch(20)=0.7280
end centrm
latticecell triangpitch fuelr=0.6075 30 cladr=0.6540 23 pitch=2.5191 53 end
centrm data
dan2pitch(30)=0.4200
end centrm
end celldata
',
' Depletion data
',

read depletion
1 10 20 30 end
end depletion
',
' Burn data
',

read burndata
power=specpower burn=daystoburn down=0 end
end burndata

read opus
units=gatoms
symnuc=u-235 u-236 u-238 pu-238 pu-239 pu-240 pu-241 pu-242 end
matl=1 10 20 30 end
end opus

read keep
newt opus couple
end keep
',
' NEWT model data
',

read model
candu 37 element
',

```

```

read parm
    echo=yes
    drawit=yes
    epsinner=-1.5e-4
    cmfd=yes
    xycmfd=4
    epsilon=8.0e-04
    cell_tol=1.0e-8
    grid_tol=1.0e-5
end parm
,

read materials
1 1 'center fuel pin' end
2 1 'clad' end
3 1 'pressure tube' end
4 1 'air gap' end
5 2 'coolant' end
6 1 'calandria' end
7 2 'moderator' end
10 1 'inner fuel ring' end
20 1 'middle fuel ring' end
30 1 'outer fuel ring' end
end materials
,

read geom
unit 1
com='fuel element (center)'
    cylinder 1 0.6075 sides=20
    cylinder 2 0.6540 sides=20
    cylinder 3 0.6545 sides=20
    media 1 1 1
    media 2 1 -1 2
    media 5 1 3 -2
    boundary 3 4 4
unit 10
com='fuel element(inner ring)'
    cylinder 1 0.6075 sides=20
    cylinder 2 0.6540 sides=20
    cylinder 3 0.6545 sides=20
    media 10 1 1
    media 2 1 -1 2
    media 5 1 3 -2
    boundary 3 4 4
unit 20
com='fuel element(middle ring)'
    cylinder 1 0.6075 sides=20
    cylinder 2 0.6540 sides=20
    cylinder 3 0.6545 sides=20
    media 20 1 1
    media 2 1 -1 2
    media 5 1 3 -2
    boundary 3 4 4
unit 30
com='fuel element(outer ring)'
    cylinder 1 0.6075 sides=20
    cylinder 2 0.6540 sides=20
    cylinder 3 0.6545 sides=20
    media 30 1 1
    media 2 1 -1 2

```

```

media 5 1 3 -2
boundary 3 4 4
unit 2
com='pressure tube'
cylinder 1 5.1589 sides=25
cylinder 2 5.6032 sides=25
media 5 1 1
media 3 1 -1 2
com='air gap and calandria'
cylinder 3 6.4778 sides=25
cylinder 4 6.5875 sides=25
media 4 1 -2 3
media 6 1 -3 4
com='fuel elements ring 1'
com='1' hole 1 origin x=0 y=0
com='2' hole 10 origin x=1.4885 y=0
com='3' hole 10 origin x=0.74425 y=1.289079
com='4' hole 10 origin x=-0.74425 y=1.289079
com='5' hole 10 origin x=-1.4885 y=0
com='6' hole 10 origin x=-0.74425 y=-1.28908
com='7' hole 10 origin x=0.74425 y=-1.28908
com='fuel elements ring 2'
com='8' hole 20 origin x=2.77752 y=0.744234
com='9' hole 20 origin x=2.033286 y=2.033286
com='10' hole 20 origin x=0.744234 y=2.77752
com='11' hole 20 origin x=-0.74423 y=2.77752
com='12' hole 20 origin x=-2.03329 y=2.03329
com='13' hole 20 origin x=-2.77752 y=0.744234
com='14' hole 20 origin x=-2.77752 y=-0.74423
com='15' hole 20 origin x=-2.03329 y=-2.03329
com='16' hole 20 origin x=-0.74423 y=-2.77752
com='17' hole 20 origin x=0.744234 y=-2.7752
com='18' hole 20 origin x=2.033286 y=-2.03329
com='19' hole 20 origin x=2.77752 y=-0.74423
com='fuel elements ring 3'
com='20' hole 30 origin x=4.3305 y=0
com='21' hole 30 origin x=4.069339 y=1.481118
com='22' hole 30 origin x=3.317355 y=2.783592
com='23' hole 30 origin x=2.16525 y=3.750323
com='24' hole 30 origin x=0.751983 y=4.26471
com='25' hole 30 origin x=-0.75198 y=4.26471
com='26' hole 30 origin x=-2.16525 y=3.750323
com='27' hole 30 origin x=-3.31736 y=2.783592
com='28' hole 30 origin x=-4.06934 y=1.481118
com='29' hole 30 origin x=-4.3305 y=0
com='30' hole 30 origin x=-4.06934 y=-1.48112
com='31' hole 30 origin x=-3.31736 y=-2.78359
com='32' hole 30 origin x=-2.16525 y=-3.75032
com='33' hole 30 origin x=-0.75198 y=-4.26471
com='34' hole 30 origin x=0.751982 y=-4.26471
com='35' hole 30 origin x=2.16525 y=-3.75032
com='36' hole 30 origin x=3.317355 y=-2.78359
com='37' hole 30 origin x=4.069339 y=-1.48112
boundary 4 15 15
global unit 3
' com='moderator rings (improved processing) and outer moderator cell'
cuboid 1 4p14.2875
' cylinder 2 8 sides=20
' cylinder 3 8.50 sides=20

```

```

'   cylinder 4 11.00 sides=20
'   cylinder 5 11.50 sides=20
'   media 7 1 2
'   media 7 1 -2 3
'   media 7 1 -3 4
'   media 7 1 -4 5
'   media 7 1 -5 1
      media 7 1 1
      hole 2
      boundary 1 20 20
end geom
'

read bounds
all=refl
end bounds
'

end model
end

```

A.3 CANDU lattice and ORIGEN cross-section library E/C values

Table A.1: CANDU 37-element E/C values

Isotope	Atom %	$\pm\sigma$ (%)	NEWT (ENDF/B-VII.0)	ORIGEN (ENDF/B-VII.0)
²³⁵ U	0.213	2	1.39	1.37
²³⁶ U	0.08	6	-4.19	-4.23
²³⁸ U	99.707	0.05	0.0004	0.0005
²³⁹ Pu	65.82	1	0.42	0.30
²⁴⁰ Pu	27.46	1	-0.95	-1.73
²⁴¹ Pu	4.96	3	-0.07	-0.05
²⁴² Pu	1.76	5	-1.74	-1.25

Table A.2: CANDU 28-element nominal power NEWT E/C values

Isotope	Atom %	$\pm\sigma$ (%)	NEWT (ENDF/B-VII.0)	NEWT (ENDF/B-VII.0) [†]
²³⁵ U	0.168	2.4	0.013	-0.26
²³⁶ U	0.082	6	1.81	2.12
²³⁸ U	99.747	0.05	0.002	0.002
²³⁸ Pu	0.14	5.6	-4.846	-8.0
²³⁹ Pu	64.91	1	-0.21	0.35
²⁴⁰ Pu	29.45	1	0.41	-0.57
²⁴¹ Pu	3.24	3	-0.70	-2.00
²⁴² Pu	2.27	5	2.31	0.28
³ H [‡]	2.07E+9	7.0	6.44	5.08
⁹⁰ Sr [‡]	4.86E+11	4.0	2.77	3.13
¹²⁵ Sb [‡]	2.2E+9	18	22.26	23.06
¹³⁴ Cs [‡]	4.16E+9	7.0	-3.46	-3.20
¹³⁷ Cs [‡]	8.05E+11	5.0	-3.20	-2.70
¹⁵⁴ Eu [‡]	8.14E+9	5.0	16.22	-0.25
¹⁵⁵ Eu [‡]	3.35E+9	8.0	10.33	16.44
²³⁷ Np [‡]	9.99E+5	20.0	0.97	-2.11
²⁴¹ Am [‡]	1.86E+10	20.0	3.68	4.34
²⁴⁴ Cm [‡]	7.12E+8	15.0	10.68	11.46

[†] Modified AMPX library with ENDF/B-VI.8 ¹⁵⁴Eu capture cross sections

[‡] Bq/KgU

Table A.3: CANDU 28-element nominal power ORIGEN E/C values

Isotope	Atom %	$\pm\sigma$ (%)	ORIGEN (ENDF/B-VII.0)	ORIGEN (ENDF/B-VII.0) [†]
²³⁵ U	0.202	2.4	-0.22	-0.43
²³⁶ U	0.077	6	2.08	2.08
²³⁸ U	99.717	0.05	0.002	0.003
²³⁸ Pu	0.135	5.6	-12.23	-10.04
²³⁹ Pu	66.65	1	-0.47	0.20
²⁴⁰ Pu	28.21	1	1.35	-0.40
²⁴¹ Pu	2.98	3	0.38	0.88
²⁴² Pu	1.89	5	3.93	5.47
³ H [‡]	2.07E+9	7.0	-3.62	-5.56
⁹⁰ Sr [‡]	4.86E+11	4.0	-5.10	-4.71
¹²⁵ Sb [‡]	2.2E+9	18	11.0	10.32
¹³⁴ Cs [‡]	3.5E+9	7.0	-3.37	-3.60
¹³⁷ Cs [‡]	8.05E+11	5.0	-12.32	-11.88
¹⁵⁴ Eu [‡]	6.86E+9	5.0	18.43	1.12
¹⁵⁵ Eu [‡]	2.97E+9	8.0	11.11	15.99
²³⁷ Np [‡]	9.81E+5	20.0	-5.39	-5.79
²⁴¹ Am [‡]	1.862+10	20.0	4.94	5.74
²⁴⁴ Cm [‡]	7.12E+8	15.0	-14.62	-12.67

[†] Modified AMPX library with ENDF/B-VI.8 ¹⁵⁴Eu capture cross sections

[‡] Bq/KgU

Table A.4: CANDU 28-element outer power ORIGEN E/C values

Isotope	Atom %	$\pm\sigma$ (%)	ORIGEN (ENDF/B-VII.0)	ORIGEN (ENDF/B-VII.0) [†]
²³⁵ U	0.168	2.4	4.82	4.73
²³⁶ U	0.082	6	0.88	0.85
²³⁸ U	99.747	0.05	-0.01	-0.01
²³⁸ Pu	0.140	5.6	-2.97	-0.72
²³⁹ Pu	64.91	1	-1.24	-0.23
²⁴⁰ Pu	29.45	1	2.33	0.14
²⁴¹ Pu	3.24	3	-0.44	-0.91
²⁴² Pu	2.27	5	5.54	5.79
³ H [‡]	2.07E+9	7.0	7.20	5.02
⁹⁰ Sr [‡]	4.86E+11	4.0	2.16	2.55
¹²⁵ Sb [‡]	2.2E+9	18	24.91	24.23
¹³⁴ Cs [‡]	4.16E+9	7.0	-1.75	-2.07
¹³⁷ Cs [‡]	8.05E+11	5.0	-3.20	-2.75
¹⁵⁴ Eu [‡]	8.14E+9	5.0	18.67	0.90
¹⁵⁵ Eu [‡]	3.35E+9	8.0	12.54	17.52
²³⁷ Np [‡]	9.99E+5	20.0	3.90	3.80
²⁴¹ Am [‡]	1.86E+10	20.0	4.25	4.62
²⁴⁴ Cm [‡]	7.12E+8	15.0	38.06	40.28

[†] Modified AMPX library with ENDF/B-VI.8 ¹⁵⁴Eu capture cross sections

[‡] Bq/KgU

Appendix B

B.1 LWR unmodified template E/C values

B.1.1 Westinghouse 15x15

Table B.1: H.B. Robinson (Westinghouse 15x15) calculated values

Sample ID	BO5-N9-1	BO5-N9-2	BO5-N9-3	BO5-N9-4	BO5-P8-5	BO5-P8-6
Burnup (GWd/MTU)	16.02	23.81	28.47	31.66	24.57	30.92
Nuclide ID	E/C	E/C	E/C	E/C	E/C	E/C
²³⁴ U	0.2463	3.7113	3.3048	3.0601	4.1715	0.2489
²³⁵ U	0.9866	1.0006	1.0808	1.0256	0.9691	0.9600
²³⁶ U	0.9156	1.0764	1.0438	1.0453	1.0853	0.9583
²³⁸ U	1.0002	0.9995	1.0384	1.0001	0.9995	1.0007
²³⁸ Pu	0.8118	1.9005	1.3893	1.2467	1.2806	0.7946
²³⁹ Pu	1.0127	0.9661	1.0428	0.9628	0.9884	1.0104
²⁴⁰ Pu	0.9529	0.9740	1.0909	1.0882	1.0322	0.9557
²⁴¹ Pu	1.0377	0.9406	1.0091	0.9343	0.9618	1.0551
²⁴² Pu	0.9696	1.0418	1.0291	1.0285	1.0509	1.0228
¹³⁷ Cs	1.0096	0.9978	0.9500	0.9628	-	-
²³⁷ Np	0.8909	1.1242	1.0822	1.0311	-	-

Table B.2: Turkey Point (Westinghouse 15x15) calculated values

Sample ID	D01-G9	D01-G10	D01-H9	D04-G9	D04-G10
Burnup (GWd/MTU)	30.720	30.510	31.560	31.260	31.310
Nuclide ID	E/C	E/C	E/C	E/C	E/C
²³⁴ U	0.9285	0.9252	0.8738	0.8244	0.9622
²³⁵ U	0.9531	0.9123	0.9486	1.0002	1.0308
²³⁶ U	0.9844	0.9872	0.9508	0.9330	0.9610
²³⁸ U	0.9999	1.0002	1.0000	1.0015	1.0015
²³⁸ Pu	1.1647	1.1851	1.1599	1.0404	1.0290
²³⁹ Pu	0.9902	0.9941	1.0085	1.0069	0.9755
²⁴⁰ Pu	1.0410	1.0582	1.0317	1.0139	0.9939
²⁴¹ Pu	1.0122	1.0189	1.0239	1.0080	0.9609
²⁴² Pu	1.0563	1.1096	1.0765	0.9831	0.9450
¹⁴⁸ Nd	1.0180	1.0178	1.0178	0.9695	0.9693

B.1.2 Westinghouse 17x17

Table B.3: Takahama (SF95) (Westinghouse 17x17) calculated values

Sample ID	SF95-1	SF95-2	SF95-3	SF95-4	SF95-5
Burnup (GWd/MTU)	14.3	24.35	35.42	36.69	30.40
Nuclide ID	E/C	E/C	E/C	E/C	E/C
²³⁴ U	0.9046	0.9986	0.7775	0.7857	1.0712
²³⁵ U	0.9966	0.9882	1.0015	0.9943	0.9987
²³⁶ U	1.0117	1.0415	1.0253	1.0265	1.0309
²³⁸ U	1.0008	1.0005	1.0003	1.0003	1.0004
²³⁸ Pu	0.9840	1.0578	0.9356	0.9330	0.9714
²³⁹ Pu	0.9341	0.9799	0.9804	0.9916	0.9937
²⁴⁰ Pu	0.9836	1.0199	1.0032	0.9941	0.9817
²⁴¹ Pu	0.9726	1.0561	1.0472	1.0375	1.0267
²⁴² Pu	0.9738	1.0592	1.0386	1.0215	0.9937
²⁴¹ Am	0.9132	0.6722	0.6566	0.4759	0.6878
²⁴² Am	113161	115706	116214	113057	108842
²⁴³ Am	0.8739	0.9230	0.8932	0.8666	0.8722
²⁴² Cm	1.1716	1.7763	1.9217	2.1818	1.4540
²⁴³ Cm	1.4194	1.5742	1.3449	1.3659	1.5155
²⁴⁴ Cm	0.9381	1.1400	1.0016	1.0064	0.9543
²⁴⁵ Cm	0.7621	1.0240	0.8830	0.9264	0.8816
¹⁴² Nd	1.2757	1.1092	1.2036	1.1701	1.0795
¹⁴³ Nd	0.9972	0.9994	1.0033	0.9949	0.9944
¹⁴⁴ Nd	0.9202	0.9394	0.9352	0.9725	0.9383
¹⁴⁵ Nd	0.9985	1.0052	1.0100	1.0057	1.0022
¹⁴⁶ Nd	0.9753	0.9842	0.9833	0.9834	0.9776
¹⁴⁸ Nd	0.9900	0.9989	0.9972	0.9976	0.9933
¹⁵⁰ Nd	1.0152	1.0088	1.0155	1.0135	0.9994
¹³⁴ Cs	1.1000	1.1456	1.1073	1.1068	1.1206
¹³⁷ Cs	1.0323	1.0500	1.0386	1.0417	1.0305
¹⁴⁴ Ce	1.1854	1.1835	1.2199	1.1122	1.1793
¹⁵⁴ Eu	0.9177	0.9827	0.9689	1.0030	0.9753
¹⁰⁶ Ru	1.1313	0.9592	0.8871	0.8756	1.0177
¹²⁵ Sb	0.4623	0.5008	0.4001	0.3285	0.4269

Table B.4: Takahama (SF97) (Westinghouse 17x17) calculated values

Sample ID	SF97-1	SF97-2	SF97-3	SF97-4	SF97-5	SF97-6
Burnup (GWd/MTU)	17.69	30.73	42.16	47.03	47.25	40.79
Nuclide ID	E/C	E/C	E/C	E/C	E/C	E/C
²³⁴ U	0.9336	0.9080	0.9318	0.9346	0.9288	0.9179
²³⁵ U	0.9834	1.0045	1.0000	0.9944	1.0037	0.9855
²³⁶ U	1.0026	1.0217	1.0329	1.0383	1.0365	1.0351
²³⁸ U	1.0026	1.0007	1.0003	1.0002	0.9998	1.0006
²³⁸ Pu	0.7720	1.0163	1.0173	1.0366	1.0609	1.0079
²³⁹ Pu	0.7665	0.9659	0.9657	0.9789	1.0132	0.9763
²⁴⁰ Pu	0.8936	0.9526	0.9493	0.9556	0.9556	0.9398
²⁴¹ Pu	0.7838	1.0249	1.0212	1.0214	1.0453	1.0047
²⁴² Pu	0.8762	1.0358	1.0535	1.0570	1.0537	1.0312
²⁴¹ Am	0.5916	0.7861	0.7905	0.8774	0.9199	0.7675
²⁴² Am	10.9082	11.1282	8.3380	7.5830	7.4953	7.6458
²⁴³ Am	0.6307	0.9316	0.9413	0.9515	0.9589	0.9056
²⁴² Cm	0.8008	0.9659	0.9015	0.8513	0.8033	0.8845
²⁴³ Cm	0.9554	1.4257	1.3743	1.3534	1.3856	1.3507
²⁴⁴ Cm	0.5836	1.0469	1.0609	1.0773	1.0964	1.0036
²⁴⁵ Cm	0.4090	0.9473	0.9567	0.9750	1.0109	0.8788
¹⁴³ Nd	0.9994	1.0017	1.0070	1.0019	1.0106	1.0037
¹⁴⁴ Nd	0.9658	0.9811	1.0168	1.0364	1.0417	1.0218
¹⁴⁵ Nd	1.0010	0.9923	1.0016	1.0009	1.0016	1.0030
¹⁴⁶ Nd	0.9771	0.9815	0.9888	0.9882	0.9893	0.9866
¹⁴⁸ Nd	0.9818	0.9843	0.9904	0.9905	0.9896	0.9879
¹⁵⁰ Nd	0.9666	0.9863	0.9916	0.9929	0.9909	0.9852
¹³⁴ Cs	0.9657	1.1115	1.0778	1.0394	1.0474	1.0632
¹³⁷ Cs	1.0323	1.0304	1.0360	1.0264	1.0286	1.0352
¹⁴⁴ Ce	1.2161	1.1095	1.0285	0.9435	0.9366	1.0506
¹⁵⁴ Eu	0.7588	0.9568	0.9528	0.9604	0.9789	0.9414
¹⁰⁶ Ru	1.0835	1.0487	1.0241	0.9228	0.5531	1.1745
¹²⁵ Sb	0.6595	0.7058	0.4698	0.5083	0.6272	0.4551
¹⁴⁷ Sm	2.2433	2.2160	2.3225	2.3731	2.3520	2.2798
¹⁴⁸ Sm	0.8796	1.0144	1.0313	1.0484	1.0612	1.0220
¹⁴⁹ Sm	1.1689	1.5345	1.6527	1.6354	1.6655	1.6811
¹⁵⁰ Sm	0.9477	0.9527	0.9567	0.9586	0.9653	0.9459
¹⁵¹ Sm	0.8281	1.0181	1.0204	1.0191	1.0427	0.9773
¹⁵² Sm	1.0157	0.9572	0.9691	0.8915	0.9706	0.9710
¹⁵⁴ Sm	0.9687	0.9835	0.6387	0.5504	0.5501	0.6798

B.1.3 Combustion Engineering 14x14

Table B.5: Calvert Cliffs (Combustion Engineering 14x14) calculated values

Sample ID	MKP109-LL	MKP109-P	MKP109-CC	MLA098-P	MLA098-BB	MLA098-JJ
Burnup (GWd/MTU)	27.35	44.34	37.12	33.17	26.62	18.68
Nuclide ID	E/C	E/C	E/C	E/C	E/C	E/C
²³⁴ U	1.0011	0.9728	1.0166	0.9367	0.8576	0.8793
²³⁵ U	1.0532	1.1193	1.0987	1.0351	1.0329	1.0104
²³⁶ U	0.9775	0.9783	0.9726	1.0190	1.0180	1.0287
²³⁸ U	1.0060	1.0010	1.0030	1.0089	1.0167	1.0113
²³⁸ Pu	1.1652	1.1497	1.1487	1.1804	1.2204	1.3714
²³⁹ Pu	1.0452	1.0162	1.0475	1.0132	1.0492	1.0482
²⁴⁰ Pu	1.0026	1.0130	1.0087	1.0063	1.0086	1.0130
²⁴¹ Pu	1.0597	1.0547	1.0690	1.1337	1.1556	1.1747
²⁴² Pu	1.0139	1.0259	1.0034	1.0657	1.0543	1.0816
¹³³ Cs	0.9892	0.9784	0.9849	-	-	-
¹⁴³ Nd	1.0004	1.0113	1.0115	-	-	-
¹⁴⁴ Nd	0.9898	0.9920	0.9883	-	-	-
¹⁴⁵ Nd	1.0050	1.0191	1.0145	-	-	-
¹⁴⁶ Nd	0.9827	0.9785	0.9807	-	-	-
¹⁴⁸ Nd	0.9865	0.9850	0.9862	-	-	-
¹⁵⁰ Nd	0.9784	0.9637	0.9706	-	-	-
¹⁴⁷ Sm	0.9747	1.0008	0.9857	-	-	-
¹⁴⁸ Sm	1.2426	1.1889	1.1661	-	-	-
¹⁴⁹ Sm	1.7261	2.3406	1.6408	-	-	-
¹⁵⁰ Sm	1.0548	1.0796	0.9865	-	-	-
¹⁵¹ Sm	1.1474	0.9942	1.0472	-	-	-
¹⁵² Sm	1.0081	1.0254	0.9760	-	-	-
¹⁵¹ Eu	2.1864	2.1303	2.0377	-	-	-
¹⁵³ Eu	0.9844	1.0326	0.9266	-	-	-
¹⁵⁵ Eu	0.8955	0.9210	0.8685	-	-	-
²³⁷ Np	-	-	-	1.0175	1.1536	1.0627
²⁴¹ Am	-	-	-	0.9546	0.9452	0.9604
¹³⁵ Cs	-	-	-	0.9835	0.9728	0.9649
¹³⁷ Cs	-	-	-	1.0488	1.0588	1.0605

B.1.4 Babcock & Wilcox 15x15

Table B.6: Three Mile Island ORNL assay (Babcock & Wilcox 15x15) calculated values

Sample ID	A1	B1	C2D1	C2D2	B2
Burnup (GWd/MTU)	45.9	55.0	53.5	52.7	52.4
Nuclide ID	E/C	E/C	E/C	E/C	E/C
^{234}U	0.9384	0.9436	0.9344	0.9541	0.9400
^{235}U	1.1270	1.1621	1.1921	1.0987	1.2035
^{236}U	1.0433	1.0604	1.0611	1.0591	1.0607
^{238}U	1.0038	1.0112	1.0163	1.0124	1.0108
^{238}Pu	0.9652	0.9208	1.0062	0.9625	0.9355
^{239}Pu	0.9464	0.9317	0.9762	0.9328	1.0006
^{240}Pu	0.9537	0.9533	0.9898	0.9700	0.9997
^{241}Pu	0.9946	0.9856	1.0110	0.9714	1.0431
^{242}Pu	1.0233	1.0320	1.0010	1.0222	1.0164
^{237}Np	1.0168	1.0720	1.0971	0.0011	1.0790
^{143}Nd	1.0344	1.0348	1.0324	1.0343	1.0499
^{145}Nd	1.0315	1.0375	1.0264	1.0447	1.0357
^{148}Nd	0.9867	0.9881	0.9773	1.0005	0.9908
^{137}Cs	0.9298	0.9132	0.8627	0.9857	0.9040

Table B.7: Three Mile Island ANL assay (Babcock & Wilcox 15x15) calculated values

Sample ID	A1B	C1	B2	C3	B1B	C2B	B3J	D2	D1A2	D1A4
Burnup (GWd/MTU)	45.9	55.0	53.5	52.7	52.4	45.9	55.0	53.5	52.7	52.4
Nuclide ID	E/C	E/C	E/C	E/C	E/C	E/C	E/C	E/C	E/C	E/C
²³⁴ U	0.96	1.00	0.95	0.93	0.95	0.92	0.93	0.89	1.03	1.01
²³⁵ U	1.05	1.03	0.99	0.97	0.97	0.94	0.94	0.82	0.92	0.89
²³⁶ U	0.99	1.03	1.01	1.00	1.01	0.97	1.02	1.04	0.97	0.99
²³⁸ Pu	1.31	0.83	0.78	0.64	1.08	1.17	0.99	1.13	1.03	1.02
²³⁹ Pu	0.81	0.86	0.84	0.88	0.81	0.79	0.80	0.81	1.15	1.16
²⁴⁰ Pu	0.79	0.89	0.88	0.92	0.84	0.82	0.85	0.89	1.14	1.17
²⁴¹ Pu	0.81	0.98	0.95	0.97	0.93	0.91	0.93	0.97	0.99	1.01
²⁴² Pu	0.71	0.96	0.96	0.99	1.02	1.00	1.17	1.33	0.97	0.96
²³⁷ Np	0.81	0.82	0.81	0.80	0.82	0.80	0.82	0.88	1.20	1.22
²⁴¹ Am	0.86	0.87	0.78	0.69	0.65	1.15	1.15	1.20	1.30	0.82
²⁴³ Am	0.66	0.00	0.94	0.94	0.77	0.74	0.79	0.94	1.28	1.37
¹⁴³ Nd	0.94	0.89	0.91	0.87	0.98	0.94	0.96	0.99	0.99	1.01
¹⁴⁵ Nd	0.96	0.93	0.93	0.93	1.02	0.98	1.01	1.06	0.96	0.99
¹⁴⁸ Nd	0.96	0.96	0.94	0.97	1.03	1.00	1.00	1.07	0.95	1.03
¹³⁷ Cs	1.14	1.08	1.04	1.02	1.05	1.06	1.03	6.84	1.09	0.99
¹⁴⁷ Sm	0.90	0.74	0.73	0.71	1.00	0.89	0.97	0.98	1.02	1.08
¹⁴⁹ Sm	0.81	0.80	0.86	0.76	0.90	0.89	0.82	0.79	0.99	1.08
¹⁵⁰ Sm	0.89	0.83	0.81	0.79	1.01	0.91	0.98	1.05	1.02	1.10
¹⁵¹ Sm	0.91	0.84	0.90	0.85	1.01	0.89	0.99	1.01	0.95	1.04
¹⁵² Sm	0.94	0.91	0.93	0.91	1.03	0.93	1.02	1.06	0.97	1.03
¹⁵¹ Eu	1.23	1.22	1.41	1.51	1.01	1.25	1.32	1.36	0.85	0.84
¹⁵³ Eu	0.91	0.95	0.91	0.88	1.02	0.95	1.00	1.07	0.96	1.03
¹⁵⁵ Eu	1.72	2.08	1.89	1.86	2.24	1.45	1.49	1.62	0.70	0.53
¹⁵⁵ Gd	1.41	0.93	0.94	0.98	1.46	1.37	1.50	1.63	0.67	0.48
⁹⁵ Mo	1.02	0.98	1.00	0.90	1.02	0.98	1.00	1.05	1.01	1.02
⁹⁹ Tc	1.34	0.93	0.93	0.89	1.13	1.17	1.07	1.12	1.02	0.97
¹⁰¹ Ru	1.05	0.98	1.00	0.86	1.00	0.99	0.98	1.04	1.05	1.56
¹⁰³ Rh	0.96	0.92	0.93	0.81	0.92	0.91	0.91	0.95	1.09	1.11
¹⁰⁹ Ag	0.49	0.43	0.42	0.75	0.35	0.53	0.62	0.68	2.68	1.43

Vita

Nathan Shoman was born in Macon, GA to Sheryl and Tyler Shoman. He is the only child. He attended Houston County High School in Warner Robins, GA. After graduation he spent two years at Middle Georgia State University on scholarship before transferring to the University of Tennessee - Knoxville. There he finished his undergraduate degree in Nuclear Engineering in December of 2014. He accepted a graduate research assistantship at the University of Tennessee - Knoxville in the Nuclear Engineering program under his advisors Dr. Steve Skutnik and Dr. Jamie Coble. He graduates with his Master of Science degree in Nuclear Engineering with a focus in reactor physics and safeguards in May 2016.