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The Use of Non Destructive Assay Measurements to Estimate Burnup and Plutonium Inventory in Pressurized Water Reactor Spent Fuel Assemblies

By H. R. Trellue, D. I. Poston, and S. J. Tobin (LANL), and Y. Ham (LLNL)

1.0 Introduction

One of the overarching goals of the Next Generation of Safeguards Initiative (NGSI) Spent Fuel (SF) Non-Destructive Assay (NDA) Project is to determine the Pu mass in spent fuel assemblies with NDA instruments.¹ Other goals include verifying initial enrichment, burn-up and cooling time of declaration, detecting diversion or replacement of pins, and/or determining heat emitted from an assembly (not a safeguards goal but important nonetheless). In pursuit of these goals, various NDA instruments of interest need to be built, tested, and then calibrated. Ideally simple calibration sources can be used such as isotopic sources or fresh assemblies. However, given the fact that many of the measured signals are influenced by the complex, energy-dependent interaction of neutrons with a range of isotopes, the connection between the measured signals and the Pu mass is difficult to make.

Thus, the concept of Working Standards was developed as follows.

- Quantify, with as great an accuracy as possible, the isotopes of relevance (or groups of isotopes: fissile isotopes, neutron absorbers) to the relevant NDA signal. This exceptional accuracy is to be obtained through the combined effort of measurement made of the assembly as well as additional simulations that have been informed by the new measurements. Example of these additional measurements include: Destructive Analysis (DA) of a few pellets, absolutely calibrated NDA signals, and relative NDA signals. The extent to which these measurements can improve the characterization of the isotopic content of the assembly is a research objective for this task. (Ideally, no NDA signals being used here for Working Standard development will come from the NDA instruments being calibrated in the next step.)
- Once the “Working Standard” exists, the “Working Standard” assembly is measured with the NDA system of instruments being calibrated (for discussion’s sake, assume the system to be Differential Die Away (DDA) + Passive Gamma (PG) + Total Neutron (TN)).

- Next an unknown assembly is measured with the “calibrated” NDA system of instruments (DDA + PG + TN).
- The content of the unknown assembly is determined by comparing it to the “Working Standard” assembly. Under the simplest case, imagine the unknown assembly turns out to have all the same measured signatures as the “Working Standard” assembly. Then we could conclude it had the same content. The more realistic case is that it will be a little different. The measured signature may show that the BU was greater, cooling time less, and fissile content lower. All of these *relative changes* from the “Working Standard” are quantified from the measured values. How much the Pu mass changes because of these differences then needs to be quantified. The fact that simulations are used to quantify a “relative change” from a known quantity is important since the uncertainty in the final answer is reduced.

In particular, development of a realistic “base case” simulation of assemblies would involve the following steps:

- Compare isotopic estimations in base fuel to DA.
- Change (“tweak”) operating conditions and rerun the simulation.
- Compare the declared value to a post-iteration-with-measurements burnup calculation to assess how much the inclusion of measurement data changes the Pu mass differences.

In preparation for fully developing Working Standard assemblies, there were several pieces of research performed during Fiscal Year 2013 that will be discussed in upcoming sections.

- 1) We performed simulations of “well-characterized” Republic of Korea (ROK) assemblies given extensive operating data and pin-by-pin burnup maps for seven assemblies (8 are available at the PIEF, but we were only given pin-by-pin burnup maps for 7). Then, we changed the pin-by-pin burnup distribution using NDA from Partial Defect Tester (PDET) measurements and regenerated isotopics for the assemblies. Finally, we performed simulations for Delayed Neutron (DN) and Californium Interrogation Prompt Neutron (CIPN) instruments surrounding the assemblies with new isotopics and analyzed the results to determine the impact to the count rates and changes in plutonium content.
- 2) We determined effects of including neighboring assemblies and modeling uncertainties on the simulated isotopics, guiding how operating conditions can be changed in further Working Standard calculations. For this calculation, we simulated

irradiation of the Takahama Pressurized Water Reactor (PWR) spent fuel assembly NT3G23 from Japan.

Future work will expand on that presented in this document and focus on the actual creation of “Working Standards” to make the connection between measured signals and Pu mass through the following.

- Quantify, with estimated uncertainties, assembly concentrations of the isotopes of relevance (or groups of isotopes: fissile, neutron absorbers, etc.) to advanced NDA signals. The level of accuracy is defined in part by the availability of assembly design information and reactor operator data. A critical component of a Working Standard is the uncertainty associated with the simulated assembly contents to quantify the impact of these uncertainties on the calibration of advanced NDA instruments.
- Apply experimental measurements of the assembly in an attempt to confirm declared assembly parameters and improve the data used in the assembly simulations, thereby reducing uncertainties in the Working Standard. Examples of these additional measurements include: DA of selected pellets or rods, absolutely calibrated NDA signals, and relative NDA signals.
- Once calibrated assemblies are obtained, NDA instruments will measure signatures for an unknown assembly and compare it to the Working Standards. Determination of input parameters (initial enrichment (IE), burnup (BU), cooling time (CT)) may require comparison to calibration curves, which represent expected results for a range of inputs and/or operating parameters.

Efforts have been made to quantify uncertainty in plutonium isotopics through irradiation simulations of spent fuel assemblies compared to destructive analysis.¹ Uncertainties are typically less than 5% in ^{239}Pu , ^{240}Pu , and ^{241}Pu and 2-3% in total elemental plutonium when detailed operating data is known. However, uncertainties are greater and working standards harder to implement when irradiation information such as location in core and composition of neighboring assemblies is unknown. Thus, the use of NDA to help guide development of Working Standards is more helpful when little is known about the assembly (such as starting with an infinitely-reflected simulation as will be discussed). This document describes efforts to quantify uncertainty in plutonium isotopics from irradiation simulations of Republic of Korea and Takahama spent fuel assemblies.

2.0. Background

In general, the best and easiest way to estimate the transuranic (TRU) inventory in a spent-fuel bundle is to utilize the detailed fabrication and operation history of the bundle, and the core/reactor it was placed in. Assembly, and in some cases full-core modeling techniques have

improved substantially over the past decade, and in particular, Monte Carlo calculations are becoming more practical with advances in computing power. The problem with relying on a solely computational-based technique is not the modeling tools, but the integrity of the input data (i.e. the design, fabrication and operational history). Even in the best case scenarios, some of this data is not available or worse, it can be reported/recorded incorrectly. If the owner of the reactor is not-cooperative, or has poor record-keeping, then there may be little fabrication or operation information available. If the owner of the reactor is deceptive, then there might be data provided that is intentionally misleading. Additionally, detailed information about where the assembly was located within a reactor and estimated pin-by-pin burnups are often not released or difficult to obtain. The highest integrity data is any that can be collected by physical inspection, either by destructive or non-destructive techniques. Destructive assays are difficult and expensive to perform, which leaves NDA to estimate and/or verify the contents of a fuel bundle.

Simulation of irradiation in nuclear reactor assemblies can take place on many different levels. Irradiation of a single fuel pin can be simulated, or a fuel assembly or even core can be modeled in entirety. Individual fuel pins can be modeled separately but often get smeared together for simplicity. Pin-level simulation within assemblies is often necessary because small burnup changes can produce significant differences in nuclide inventories across the assembly. Fuel burnup is not uniform; it is subject to axial and pin-by-pin variations. Pin-by-pin (radial) variations are strongly influenced by neighboring assemblies and exposure to burnable poisons. Pu production and ^{239}Pu fraction are sensitive to burnup and can vary radially and axially, especially in BWRs.

Additionally, many NDA parameters scale non-linearly with burnup (e.g. neutron production from $^{244}\text{Cm} \sim \text{BU}^4$). In terms of importance to NDA instrument response:

- Techniques that use neutron multiplication are relatively insensitive to distribution (< 1% variation),
- Photon detection techniques preferentially measure only the outer rows of fuel pins and are very sensitive to pin-by-pin burnup, and
- Interpreting measurement data requires knowledge of detailed pin-by-pin level spatial information.

NDA techniques can be used to provide estimated isotopic detail to the Working Standards process as in the examples given below:

- Passive Gamma: ^{134}Cs , ^{137}Cs , and ^{154}Eu , ^{106}Ru , ^{144}Ce , $^{110\text{m}}\text{Ag}$, ^{125}Sb , and others,
- Other photon detection: ^{241}Am , elemental Pu (XRF),
- Multiplication techniques: ^{244}Cm , mixture of $^{235}\text{U} + ^{239}\text{Pu} + ^{241}\text{Pu}$, and

- Self-Integration Neutron Resonance Densitometry (SINRD): ^{239}Pu (for fully burned spent fuel).

Passive neutron and gamma measurements are routinely made of spent fuel assemblies using Fork detectors or other instruments, and results can help guide the creation of spent fuel assemblies as Working Standards. Irradiation of several spent fuel assemblies were simulated at LANL in FY13 using NDA measurements to show the improvement of the simulations from using NDA measurements: two from the Republic of Korea (ROK), and one from Takahama in Japan.

3.0 Results of Working Standards Calculations from ROK

The first assemblies chosen to create “Working Standards” were two that are being measured by the NDA instruments ^{252}Cf Interrogation with Prompt Neutron (CIPN) and/or Self-integration Neutron Resonance Densitometry (SINRD) at a Post Irradiation Examination Facility (PIEF) in the Republic of Korea. Thus, for representative simulations to occur, accurate models of the spent fuel assemblies had to be created. Detailed operating histories and pin-by-pin burnups according to the simulation code TOTE were provided by ROK.² Additionally, axial gamma scans of 10 rods within two of the 14x14 assemblies were given as well as destructive analysis for a sample within one rod. However, at the beginning of FY13, Young Ham from LLNL provided us NDA results from the Partial Defect Tester (PDET) instrument that measured six of the spent fuel assemblies at the PIEF facility in ROK. The PDET instrument uses multiple tiny neutron and gamma detectors in the form of a gravity-driven cluster to obtain underwater radiation measurements inside the guide tubes of a Pressurized Water Reactor (PWR) spent fuel assembly. Results from the PDET measurements were thus used to create Working Standard assemblies, and the changes to the plutonium content and resulting two NDA instrument measurements (Delayed Neutron and ^{252}Cf Interrogation with Prompt Neutron (CIPN)) from the assemblies generated with the TOTE operator estimate of burnup were compared.

Data Sets

Both neutron and gamma measurements were performed using PDET, and two different techniques for both neutron and gamma data collection were attempted. The first measurement of gamma contributions were from an ion chamber. For the second gamma technique, some channels were integrated below a threshold to obtain gamma contribution from the spectra obtained with a fission chamber and an analog preamplifier. Usually such a threshold is applied to this data set to obtain neutron signals. However, knowing that lower channels are mainly from alphas and gammas, some channels were selected to estimate

gamma flux (at least at a relative scale). Because the second technique was rather unconventional, the ion chamber results were used here instead.

For neutrons, again there were two sets of data. One was from the same spectra mentioned above, and these results were used in this analysis. The other was using a fission chamber with a preamplifier to simply obtain counts (pulse signals) at a given measurement time. Once neutron and gamma data were obtained, it was assumed that the counts were directly proportional to the burnup of the pins directly surrounding a guide tube as described in the next paragraph.

Modified Burnup Maps

The PDET measurements were used to obtain new pin-by-pin burnup estimates of each fuel rod in two of the ROK assemblies (Note that for each of these assemblies, rods had been removed for previous measurement purposes). Because PDET measurements occur in guide tubes (see Fig. 1), mathematical techniques have to be applied to derive estimated burnups in the fuel rods surrounding the guide tubes.

The general idea of the calculations was to assume that the 4-8 pins directly surrounding a guide tube influence the burnup estimated for that tube. Several steps were taken to obtain a new pin-by-pin burnup map. Five different methods for determining pin-by-pin burnup estimates were used, but only two were carried through to the analysis of the impact of the changes on a NDA instrument, methods 1 and 4.

		34.1			40.5			37.6			32.8		
				45.9					42.0				
		38.9									38.1		
		38.1									37.2		
				39.3					41.2				
		31.7			37.7			38.0			32.4		

Fig. 1. Example of average representative burnups in guide tubes surrounding fuel rods

Method 1

- First, the average count rate over all 16 guide tubes was obtained. Then the counts in each individual guide tube were divided by the average for a “relative” burnup fraction in each guide tube. This “relative” fraction was then multiplied by the average declared burnup in an assembly to give a representative burnup for the guide tube.
- An estimated burnup in each of the 4 (1b) or 8 (1) fuel pins surrounding the guide tube was then obtained by assuming they all had an average of the central value (see Fig. 2).

	37.5		X = Fuel pin X = Guide Tube
37	38	38.5	
	39		

Fig. 2. Example of estimating burnup for fuel pins surrounding a guide tube

- Burnups in the remaining cells were estimated simply by a weighted-by-inverse-distance (Euclidean methodology) averaging of all fuel rods from the previous step.

Method 2

- Same first three steps as Method 1
- Operator declared pin-by-pin burnups filled up the remaining rods

Method 3

- Same first two steps as Method 1.
- Operator-declared BU was used at all available locations to define the gradient instead of the distance-based interpolation used in Method 1.

Method 4

- Same first three steps as Method 1
- Additionally used operator burnup at 4 corners (4b) or at 4 corners plus 4 outer rods located approximately in the middle of the outer row of the assembly (4) to determine pin burnups away from the guide tubes.

Method 5

- Same first three steps as Method 1
- Uses operator burnup rescaled slightly, by multiplication by $\text{mean(PDET)}/\text{mean(BU)}$, using available burnups from PDET measurements.

Using these techniques, the average burnup of the assembly was usually different than the initial declared value, but such differences were expected. Additionally, rods that have been removed from the assembly can complicate calculations for determining pin-by-pin burnup maps (i.e. only 3 instead of the 4 fuel rods surrounding a guide tube may be valid). If weighted-by-inverse-distance techniques were used (all but Method 3), then the effect of missing rods was small. For that reason, two ROK assemblies with the fewest rods removed were chosen for this analysis. Unfortunately, DA was not available for either of these assemblies. Note that DA was available for one ROK assembly, but it had a significant number of rods removed and a strong gradient from one corner to another, making it too difficult to calculate pin-by-pin burnup distributions using PDET.

Once new pin-by-pin burnup maps were obtained, Oak Ridge National Laboratory then used ORIGEN within the SCALE code to generate material compositions indicative of burnup in all pin locations. We then input these material cards back into MCNP models of instruments and simulated count rates corresponding to the new burnup maps.

Impact on NDA Instruments

Delayed Neutron (DN) counting is an active assay technique that consists of turning on and off an interrogating source, in this case a deuterium-tritium (DT) generator, on one side of the assembly and counting the number of neutrons detected on the other side when the source is off (see Fig. 3). First a passive neutron measurement is performed to determine the background count rate (singles), mainly originating from the spontaneous fission of ^{244}Cm in spent fuel and amplified by multiplication. An active assay is then undertaken by switching on the DT generator. Spectrum

Fig. 3. Geometry used for DN where the numbers represent “unique” fuel pins.

tailoring is implemented to lower the energy of the ~14.1 MeV neutrons created before they enter the fuel [Ref. 3]. DN uses a relatively long neutron burst (0.9 s), followed by a short pause (~0.1s) and then one second counting periods repeated over 300 seconds.

Table 1 contains results from Delayed Neutron calculations using a variety of data and methods for determining pin-by-pin burnups in spent fuel assemblies derived from PDET measurements and corresponding changes in total plutonium mass. In general, the delayed neutron count rates were mainly affected by changes in the estimated average burnup of the assembly because they were sensitive to how much ^{235}U was present (i.e., DN count rates were higher at low burnups when more ^{235}U was present). The percent difference from DN counts between the new cases and the operator-declared pin-by-pin burnup maps (the “base” case) was also larger using Method 4 than Method 1. Note that Method 4 for assembly 1 significantly underestimated the burnup and both the delayed neutron count rate and plutonium content were much different than the base case as a result.

Differences of less than one percent were seen from similar simulations of Californium Interrogation Prompt Neutron (CIPN) measurements using different pin-by-pin burnup maps as presented in the ORNL end-of-the-year report.

4.0 Results of Working Standards Calculations from ROK

The next assembly examined as a “Working Standard” was a Takahama 3 spent fuel assembly from Japan. In the last section, we discussed the use of NDA measurements to enhance a calculated pin-by-pin burnup map. For Takahama, representative of many other spent fuel assemblies, no pin-by-pin or core map was given, so we must start with an infinitely-reflected model instead. The infinitely-reflected (base case) model assumes that the assembly is symmetric on all four sides, so the radial burnup gradient is unknown. Input and modeling parameters (temperature, power, cycle length, shutdown, neighboring assembly effects) for the burnup simulation were then modified to provide better agreement with NDA, which helps provide the burnup gradient.

Methodology

Burnup and isotopic generation and depletion calculations were again performed using the code MONTEBURNS, which links the Monte Carlo transport code MCNP6 and the isotope generation and depletion code CINDER90. The initial design of the bundle was known, while the burnup conditions of the assembly were initially based on “generic” PWR conditions. The NDA measurements used in the study were the gamma scans named NT3G23, which were obtained at 9 different locations as shown in Fig. 3. The majority of pins had an IE of 4.1wt%; these pins are referred to as SF95 pins. The pins denoted with an “x”, including D11, B12 and B10 are 2.6wt% enriched pins that contain 6 w/o Gd; these pins are referred to as SF96 pins.

Table 1. Changes to DN count rate from Variations in Pin-by-Pin Burnup Maps

Data	Method	DN counts	average BU	% diff. - DN	% diff. - Pu
Assembly 1		/per source neutron			
Operator-declared burnups		5.67E-06	17.20		
Neutron - PDT preamp.	1	5.81E-06	16.95	2.38	-0.89
Gamma - ion chamber	1	5.72E-06	16.97	0.87	-0.63
Neutron - PDT preamp.	4	6.52E-06	15.39	14.97	-5.49
Gamma - ion chamber	4	6.56E-06	15.39	15.67	-5.49
Assembly 2					
Operator-declared burnups		3.08E-06	37.84		
Gamma - ion chamber	4	3.18E-06	37.85	3.32	-0.84
Gamma - ion chamber	4b	3.21E-06	36.57	4.11	-0.97
Gamma - ion chamber	1b	3.04E-06	37.68	-1.29	0.40
Gamma - ion chamber	1	3.03E-06	37.66	-1.59	0.39
Gamma - integrated channels	1	3.04E-06	37.71	-1.26	0.44
Neutron	1	3.13E-06	37.35	1.49	-0.20

The destructive analysis data used in this study were for pins A4 (SF95) and B10 (SF96). The geometric details of the bundle were assumed to be known. The bundles were a 17x17 bundle in a core with a bundle pitch of 21.4 cm.³

The MCNP model of the Takahama 3 assembly assumed the following:

- All pins in the 17x17 bundle were individually modeled, both in MCNP and MONTEBURNS.
- Up to 6 radial rings per pin were modeled, but only one axial zone.
- Upper and Lower Plena were modeled above/below active stack.
- Reflective or open water axial boundary conditions were used beyond the plena.
- Option to model water perimeter between lattice and model x-y boundaries existed.
- X-Y boundaries were modeled as reflective boundary conditions.

The new MCNP capability to model albedo boundary conditions by changing the amount and/or energy spectrum of reflection at the edges of an assembly was also used in an attempt to simulate whether the bundle may have been next to fresh, burnt or core-periphery regions.⁴ Details on run time and uncertainty analyses can be found in Appendix A.

C3																A6
				x	w			w			w	xD11				
			w					x					w			
		x												xB12		
		w			w			w			w			w		
						x				x						
A8		w			w			w			w			w		
						x				x						
		w			w			w			w			w		
		xB10												x		
			w					x					w			
				x	w			w			w	x				
A4								C7								C5

Fig. 3. Takahama Fuel Assembly

Assumptions

If one is attempting to determine the plutonium inventory of a PWR bundle using NDA gamma measurements, then at a minimum one has physical access to the bundle. Many parameters can be estimated based on declared operating data and standard PWR protocol and experience. The dimensions of the bundle will give a decent indication of the approximate power density during operation (at least within a factor of 2). Reactivity requirements usually dictate PWR cycle lengths of ~12 to 18 months and refueling/outage times of ~3 months. Operating temperatures are in the neighborhood of 300 C, pressures around 15 MPa, and expected boron concentration, gaps between bundles, etc. can be derived. If NDA measurements indicate significant differences, then possible neutronic reasons could be that they have a different enrichment or Gadolinium (Gd) poisoning. Other reasons could be partial defect of fuel pins or misinformation from the operator. Knowing the IE, BU, and CT provides an advantage for this study.

Parameters Modified

The parameters that were varied to match the model with the measurements were:

- Operating temperature (bundle average): 290 C to 310 C
- Operating Pressure: 13 to 17 MPa
- Gap between assemblies: 0 to 2 mm
- Boron concentration (average over cycle) 500 to 1000 ppm
- Number of operating cycles: 1 2 or 3
- Cycle length: 12 to 16 months
- Shutdown length: 3 months

Results

Tables 2 and 3 list the parameters for each of the examined cases. The results of the calculations for each case, and the measured values are listed in Appendix B.

For each case, the percentage differences between the calculated and measured activity ratios were “plotted” in an Excel spreadsheet. Figures 4 and 5 below show this plot for 2 cases of the most interest (taki2c and takj2b). Note that the numbers represent the percent error times 10 (e.g. a value of 13 represents 1.3%), which was a better format for displaying the data.

Table 2. Parameters Used for the First Set of Cases

Case	Base	takf2a	takf2b	tagk2a	takh2a	takh2b	takh2c	takh2d	takh2e	takh2f
Power	20.09	19.2	19.2	18.8	19.2	19.2	18.75	18.75	18.75	18.65
h2oden	0.7373	0.721	0.721	0.721	0.71	0.7373	0.7373	0.7463	0.7265	0.7265
Ppm	652.5	800	800	800	800	500	500	500	500	500
Boundary	Inf. Refl.	1.00	1.00	1.25	1.00	1.00	1.00	1.00	1.50	1.50
Days	875	787	787	890	787	787	890	890	890	890
Nstep	24	16	16	29	16	13	29	29	29	29
Nring	1	4	4	4	1	1	1	1	1	1
Fimp	0.000001	0.00001	0.000001	0.000001	0.0001	0.0001	0.00001	0.000001	0.00001	0.00001
Kcode	250000	250000	250000	250000	125000	100000	125000	125000	125000	125000
axialBC	Reflected	reflected	reflected	reflected	nominal	nominal	nominal	nominal	nominal	nominal

Table 3. Parameters Used for the Second Set of Cases

[illegible]

Fig. 4. 10 times the percent-error between the calculated and measured activity ratios (and burnup) at each pin location for case taki2c.

Fig. 5. 10 times the percent-error between the calculated and measured activity ratios (and burnup) at each pin location for case takf2b.

Table 4 lists the percentage difference between the calculated activity ratios and the ratios based on the NDA data. The rows labeled Ave(Abs(%)) are the average value of the absolute value of the errors for each isotope in that pin. It was felt that this would be a better comparison metric than standard deviation (e.g. 2 measurements 5% off were just as good/bad as 1 measurement 10% off and the other exactly matching).

Table 4. Difference Between Calculated and Measured Activity Ratios (curies/curies Cs-137)

All cases below are 4-ring cases – see previous table for differences between cases								
Pin A8	takf2a	takf2b	tag2a	taki2a	taki2b	taki2c	taki3c	takk2a
Cs134	4.5%	5.7%	3.9%	0.8%	0.3%	-0.1%	1.4%	1.6%
Eu154	8.4%	9.4%	10.2%	5.7%	5.1%	4.5%	7.3%	8.4%
Ce144	3.1%	2.9%	-3.3%	-3.1%	-2.9%	-3.0%	-3.0%	-3.0%
Ru106	-2.8%	-2.4%	-3.0%	-4.6%	-5.0%	-5.3%	-4.7%	-4.5%
Sb125	7.3%	7.6%	5.5%	4.8%	4.6%	4.6%	4.8%	4.9%
<i>Ave(Abs(%))</i>	5.2%	5.6%	5.2%	3.8%	3.6%	3.5%	4.2%	4.5%
Pin B12	takf2a	takf2b	tag2a	taki2a	taki2b	taki2c	taki3c	takk2a
Cs134	3.0%	4.6%	3.4%	0.6%	0.2%	-1.1%	-1.7%	0.5%
Eu154	10.6%	19.9%	22.3%	16.8%	16.6%	16.2%	17.2%	20.2%
Ce144	7.4%	7.3%	1.7%	2.6%	2.5%	2.6%	2.6%	2.3%
Ru106	-0.9%	-0.5%	-1.5%	-2.9%	-3.0%	-3.7%	-3.0%	-2.6%
Sb125	2.7%	3.0%	1.0%	0.4%	0.3%	0.0%	0.3%	0.6%
<i>Ave(Abs(%))</i>	4.9%	7.0%	6.0%	4.7%	4.5%	4.7%	5.0%	5.2%
Pin D11	takf2a	takf2b	tag2a	taki2a	taki2b	taki2c	taki3c	takk2a
Cs134	2.5%	3.7%	2.1%	-1.8%	-1.3%	-1.8%	-2.0%	-0.6%
Eu154	10.7%	18.5%	20.5%	15.6%	14.8%	14.8%	16.6%	18.8%
Ce144	8.1%	8.1%	2.5%	3.1%	3.3%	3.6%	3.3%	3.0%
Ru106	-1.1%	-0.5%	-1.5%	-3.1%	-2.7%	-3.2%	-2.8%	-2.4%
Sb125	5.6%	6.2%	4.1%	3.4%	3.4%	3.2%	3.5%	3.7%
<i>Ave(Abs(%))</i>	5.6%	7.4%	6.1%	5.4%	5.1%	5.3%	5.7%	5.7%
Pin B10	takf2a	takf2b	tag2a	taki2a	taki2b	taki2c	taki3c	takk2a
Cs134	1.0%	2.3%	1.4%	-2.2%	-1.2%	-2.8%	-2.6%	-0.9%
Eu154	9.2%	16.9%	20.6%	14.6%	13.7%	13.7%	16.0%	17.3%
Ce144	10.3%	10.0%	4.2%	4.9%	4.8%	5.0%	5.1%	4.7%
Ru106	-1.8%	-1.3%	-2.5%	-4.3%	-4.1%	-4.7%	-4.0%	-3.7%
Sb125	6.0%	6.3%	4.1%	3.4%	3.4%	3.2%	3.4%	3.7%
<i>Ave(Abs(%))</i>	5.6%	7.3%	6.6%	5.9%	5.4%	5.9%	6.2%	6.0%

Table 4 (cont). Difference Between Calculated and Measured Activity Ratios (curies/curies Cs-137)

Pin A4	takf2a	takf2b	tag2a	taki2a	taki2b	taki2c	taki3c	takk2a
Cs134	2.5%	3.2%	2.2%	0.1%	-1.5%	-1.1%	-0.4%	-0.5%
Eu154	9.6%	10.6%	10.9%	4.9%	6.4%	4.9%	6.1%	9.1%
Ce144	7.6%	7.4%	1.2%	1.2%	1.7%	1.1%	1.1%	1.2%
Ru106	-3.8%	-3.5%	-3.9%	-5.3%	-6.2%	-5.8%	-5.4%	-5.3%
Sb125	14.8%	15.2%	13.0%	12.3%	12.1%	12.1%	12.4%	12.5%
<i>Ave(Abs(%))</i>	7.7%	8.0%	6.3%	4.7%	5.6%	5.0%	5.1%	5.7%
Pin C7	takf2a	takf2b	tag2a	taki2a	taki2b	taki2c	taki3c	takk2a
Cs134	3.9%	6.5%	4.0%	1.2%	-0.1%	-0.1%	1.0%	1.6%
Eu154	10.5%	12.8%	13.4%	8.0%	8.3%	7.2%	9.1%	10.4%
Ce144	6.3%	6.3%	-0.1%	0.1%	0.2%	0.2%	0.2%	0.2%
Ru106	-2.8%	-2.2%	-3.0%	-4.5%	-5.2%	-5.3%	-4.7%	-4.3%
Sb125	9.1%	9.7%	7.3%	6.7%	6.4%	6.4%	6.7%	6.9%
<i>Ave(Abs(%))</i>	6.5%	7.5%	5.6%	4.1%	4.0%	3.8%	4.3%	4.7%
Pin C5	takf2a	takf2b	tag2a	taki2a	taki2b	taki2c	taki3c	takk2a
Cs134	4.0%	4.7%	3.6%	1.3%	-0.7%	0.3%	1.9%	0.8%
Eu154	13.3%	14.2%	14.9%	8.7%	9.9%	7.5%	10.0%	12.6%
Ce144	5.6%	5.6%	-0.6%	-0.8%	-0.1%	-0.6%	-0.7%	-0.4%
Ru106	-4.3%	-4.1%	-4.3%	-5.9%	-6.7%	-6.3%	-5.9%	-5.9%
Sb125	7.7%	8.1%	6.1%	5.3%	5.1%	5.3%	5.5%	5.4%
<i>Ave(Abs(%))</i>	7.0%	7.3%	5.9%	4.4%	4.5%	4.0%	4.8%	5.0%
Pin C3	takf2a	takf2b	tag2a	taki2a	taki2b	taki2c	taki3c	takk2a
Cs134	4.3%	6.0%	3.5%	1.1%	-0.7%	1.3%	1.6%	1.2%
Eu154	8.1%	9.1%	9.9%	4.5%	5.4%	4.1%	5.6%	8.9%
Ce144	5.1%	5.0%	-1.3%	-1.5%	-0.7%	-1.3%	-1.4%	-1.1%
Ru106	-3.1%	-2.6%	-3.2%	-4.5%	-5.1%	-5.1%	-4.5%	-4.3%
Sb125	7.8%	8.3%	6.3%	5.4%	5.4%	5.3%	5.6%	5.7%
<i>Ave(Abs(%))</i>	5.7%	6.2%	4.8%	3.4%	3.5%	3.4%	3.8%	4.2%
Pin A6	takf2a	takf2b	tag2a	taki2a	taki2b	taki2c	taki3c	takk2a
Cs134	4.0%	4.8%	4.0%	1.5%	-0.1%	0.8%	1.7%	1.7%
Eu154	12.3%	13.3%	14.4%	8.8%	9.0%	7.6%	9.4%	12.5%
Ce144	1.4%	1.3%	-4.7%	-4.8%	-4.2%	-4.6%	-4.8%	-4.4%
Ru106	-4.5%	-3.9%	-4.9%	-6.2%	-6.8%	-6.9%	-6.3%	-6.1%
Sb125	3.0%	3.4%	1.4%	0.7%	0.4%	0.4%	0.6%	1.0%
<i>Ave(Abs(%))</i>	5.0%	5.4%	5.9%	4.4%	4.1%	4.1%	4.6%	5.1%

Table 5. Difference Between Calculated and Measured Activity Ratios (curies/curies Cs-137)

<i>Average of difference in all pins</i>								
All Pins	takf2a	takf2b	takg2a	taki2a	taki2b	taki2c	taki3c	takk2a
Cs134	3.3%	4.6%	3.1%	0.3%	-0.6%	-0.5%	0.1%	0.6%
Eu154	10.3%	13.9%	15.2%	9.7%	9.9%	8.9%	10.8%	13.1%
Ce144	6.1%	6.0%	0.0%	0.2%	0.5%	0.3%	0.3%	0.3%
Ru106	-2.8%	-2.3%	-3.1%	-4.6%	-5.0%	-5.1%	-4.6%	-4.3%
Sb125	7.1%	7.5%	5.4%	4.7%	4.6%	4.5%	4.8%	4.9%
<i>Ave(Abs(%))</i>	5.9%	6.9%	5.4%	3.9%	4.1%	3.9%	4.1%	4.7%

Table 5 lists the percentage error of each activity ratio averaged over all 9 pins (as opposed to the above table which averages the ratios in each pin). The bottom row again provides the average of the absolute value of error in the 5 activity ratios.

For most cases, the percent error between the calculated and measured activity ratios was relatively small (<5%), except for Eu154 which was significantly over predicted (from 8.9% to 15.2%). The best case was taki2c, which matched Cs134 and Ce144 very well, Ru106 and Sb125 reasonably well, and had the lowest error (8.9%) for Eu154. The cases that performed best at predicting fission products were generally the more highly moderated (higher h2o density and lower boron concentration), with even more boundary moderation (i.e. had a thicker water layer between assemblies).

The purpose of the study was to determine a) if a reasonable match of NDA measurements would reproduce a reasonable match of plutonium inventory and b) whether the model that best matched NDA measurements would best reproduce the plutonium inventory.

Table 6 contains the difference between the calculated and measured inventory of plutonium and americium isotopes. The differences are displayed in grams per ton-initial-heavy-metal (g/TIHM), where TIHM is based on the initial actinide loading. This is more informative than percent differences because trace isotopes can often have large percentage errors and can detract from the bigger picture. The “Net Pu Diff” is simply the sum of the Pu isotopes, thus providing the grams/TIHM difference of Pu overall. The “Abs Pu Diff” is the absolute mass of the Pu isotopes, which gives a better indication of plutonium isotopic differences. The “Pct Pu Diff” divides the “Net Pu Diff” by the measured g/TIHM of the Pu, thus providing percent difference between the calculated amount of Pu and the measured amount of Pu.

Table 6. Difference Between Calculated and Measured Pu and Am Isotopics (g/TIHM)

SF95	Base	takf2a	takf2b	takg2a	taki2a	taki2b	taki2c	taki3c	takk2a
Pu-238	-21.54	-18.61	-15.61	-14.01	-18.61	-23.81	-21.91	-20.31	-21.01
Pu-239	151.62	-59.98	22.02	-35.98	-393.98	-264.98	-405.98	-311.98	-53.98
Pu-240	12.35	60.49	48.49	71.49	52.49	5.49	26.49	28.49	12.49
Pu-241	-70.51	-58.85	-28.85	-46.85	-90.85	-112.85	-100.85	-76.85	-69.85
Pu-242	-68.91	-33.09	-32.49	-30.79	-17.89	-51.89	-24.29	-26.09	-50.09
Am-241	6.67	6.43	6.90	8.72	7.13	6.90	7.07	7.60	8.19
Am-242m	-0.22	-0.23	-0.22	-0.20	-0.24	-0.23	-0.24	-0.23	-0.21
Am-243	-7.96	-0.93	-1.16	1.54	0.66	-6.27	-1.40	-1.28	-3.98
Net Pu Diff	3.01	-110.05	-6.45	-56.15	-468.85	-448.05	-526.55	-406.75	-182.45
Abs Pu Diff	324.9	231.03	147.46	199.13	573.83	459.03	579.53	463.73	207.43
Pct Pu Diff	-0.03%	-1.1%	-0.1%	-0.6%	-4.8%	-4.6%	-5.3%	-4.1%	-1.9%
SF96	Base	takf2a	takf2b	takg2a	taki2a	taki2b	taki2c	taki3c	takk2a
Pu-238	-29.21	-20.23	-17.89	-15.80	-23.16	-22.18	-24.59	-24.01	-21.34
Pu-239	213.53	-134.62	-55.62	-88.62	-342.62	-374.62	-348.62	-264.62	-147.62
Pu-240	-67.82	107.51	56.51	80.51	21.51	34.51	14.51	6.51	35.51
Pu-241	13.04	-15.94	41.06	16.06	-58.94	-54.94	-74.94	-48.94	-26.94
Pu-242	0.71	-18.90	-9.10	-12.40	-29.30	-24.70	-42.30	-39.70	-33.20
Am-241	-5.41	3.34	4.44	6.20	4.30	4.23	4.06	4.72	5.50
Am-242m	0.18	-0.21	-0.19	-0.16	-0.20	-0.21	-0.21	-0.20	-0.18
Am-243	-10.50	2.09	2.80	3.45	-4.03	-2.28	-6.64	-4.95	-3.19
Net Pu Diff	130.26	-82.17	14.97	-20.24	-432.50	-441.92	-475.93	-370.75	-193.58
Abs Pu Diff	340.4	297.20	180.18	213.39	475.53	510.95	504.96	383.78	264.61
Pct Pu Diff	-1.4%	-0.8%	0.2%	-0.2%	-4.4%	-4.5%	-4.8%	-3.8%	-2.0%

For percentage error between the calculated and measured plutonium inventories was relatively small (<5%) for all cases for both pins (SF95 and SF96). The best case was takf2b, which was extremely close in calculating overall Pu inventory, and did fairly well for all Pu and Am isotopes. The cases that performed best were generally the less moderated (lower h₂O density and higher boron concentration), with less boundary moderation (i.e. had a thinner water layer between assemblies), opposite of that seen for the activity ratios. Note that the overall plutonium elemental calculation for the basecase for SF95 was fairly close, which as coincidence because the individual plutonium isotopics were not. Changes to input parameters for the simulations did help decrease plutonium isotopic errors.

Effectiveness of Methodology

The burnup of a PWR bundle was modeled with various assumptions and some of the results were compared with measured values. The goal was to tweak the model to match measured activity ratios (based on gamma scans) and then determine how well the model matched the measured inventory of plutonium (and other isotopes). There are many possible metrics for comparison (individual isotope inventories, variations across the bundle, etc.), but the overall net difference between calculated and measured activity ratios and plutonium inventory gives the broadest comparison – Table 7 shows these differences for 8 of the cases that were simulated.

Overall, the method used in the study of predicting plutonium inventory based on NDA gamma measurements could be labeled as adequate. If the operational parameters of the bundle were set such that the average error of a particular isotope activity was <5%, then the total Pu inventory was calculated within 5%.

The disappointing aspect of this method is that the cases that best matched activity ratios were the worst cases with respect to matching Pu inventory. Recall that the activity difference is an absolute value, so there is no direct consequence for the activity ratios being positive, while the Pu inventories are mostly negative. However, there is some significance that in general the activity ratios calculated were mostly over-predicted, and that some isotopes were much harder to match than others. Such effects are almost certainly a consequence of gamma absorption within the fuel and possible structural or probe materials between the source and the detector. Most of the decay gammas for the isotopes measured are on the order of 1 MeV, so the attenuation coefficients are in the several 1/mm range (up to 1/cm range for energies < 1 MeV). This is significant because the dimensions of the fuel pellet and clad are in the mm range, so a good fraction of gammas will be absorbed between emission and detection.

It should be expected that for all isotopes the measured gamma flux will be lower than what would exist within the fuel; thus, isotopes with lower energy gammas will be impacted more by absorption effects. Therefore, to produce accurate calculational results, the gamma source must be transported from the fuel to the detector, as opposed to simply calculating the activity of the isotopes within the fuel. Also, the fuel would be best modeled with nodes that are not

Table 7. Summary of Differences Between Calculated and Measured Cases

<i>Average of difference in all pins</i>								
All Pins	takf2a	takf2b	tag2a	taki2a	taki2b	taki2c	taki3c	takk2a
<i>Activity Diff</i>	5.9%	6.9%	5.4%	3.9%	4.1%	3.9%	4.1%	4.7%
<i>Pu Diff</i>	-0.8%	0.2%	-0.2%	-4.4%	-4.5%	-4.8%	-3.8%	-2.0%

radially symmetric, so that the fuel on the outside of the pin (closest to the measurement point) is best represented. The transport of gammas would be the most important aspect of a follow on study, and there are other possible improvements as well – listed in the following Possible Improvements to Methodology section.

Future Work

There are several ways to improve the technique of estimating Pu inventory based on NDA gamma measurements. Some of them are listed below, in relative order of importance.

Including Radial Nodes in Fuel/Clad is important for 2 reasons. 1) The spectrum of the perimeter pins (which are most of the pins of interest) should be significantly different on the outer (boundary) edge of the pin than the inner, thus they will burn differently. 2) Of the gammas that are to be transported, it is important to include the spatial dependence of the gamma emissions within the fuel (since some gammas will have to pass through more fuel than others to reach the outer (boundary) surface). This would be easy to implement, but will require longer computation times; although this added fidelity could only be added on the 9 pins for which there is NDA data.

Change Boron Concentration with Burnup: In the current cases, the boron concentration was held steady throughout the burn. The change in spectrum over the burn will probably have a significant impact on the isotopics at the end of the burn. Boron changes would be easy to implement because the MONTEBURNS feed file allows materials to be added or removed step-by-step.

Modeling Axial Nodes would be valuable for 2 reasons. 1) The spectrum is significantly different on the axial ends than the center, so the calculation of the overall pin burnup would be more accurate. 2) The actual NDA and da data used in this study is presented as a function of axial position (average to a total pin average for this study), so the axial results could be compared directly to the axial measurements, thus adding much more fidelity. However, getting accurate geometries axially is difficult.

Simulate Bundle Shuffling with Changes in Albedo: In the current case, the albedo at each x-y surface is kept constant throughout the burn; some sides see more incoming neutrons than departing neutrons and vice-versa. There would be more flexibility, and thus potential accuracy by changing the albedo between burn cycles; in effect simulating the arrival of a fresh bundle on one side and a twice-burned bundle on the other. The changes in albedo (magnitude and energy spectrum) could be estimated by tallying the fluxes leaving bundle as it moves from a fresh to a twice-burned assembly. This would be easy to implement, but may require several modeling simulations to arrive at the “correct” answer (in terms of how the bundles may have been shuffled).

Modeling Surrounding Bundles and Including Shuffle would be a more accurate representation than the above method of simply changing albedo. It would take longer to run and require more work to set up the models.

5.0. Applications of Working Standards in Sweden

Although Working Standards could be useful in conjunction with NDA in many countries including the United States, the most promising opportunity at the present time for NDA measurements of spent fuel assemblies is in Sweden as will be discussed here. In the past 4 years the Next Generation Safeguards Initiative Spent Fuel (NGSI-SF) Project has spent around \$10 million researching the Nondestructive assay (NDA) of spent fuel. The final step in this project, *the final step that is essential to any technology being deployed by the safeguards profession*, is testing NDA technologies in the field. The opportunity in Sweden is far superior to other options for infield testing in term of the following: (1) quality and quantity of fuel, (2) potential to lead to long term deployment, (3) seriousness of our collaborator to invest in research effort and to allow us to be actively involved, (4) cost effectiveness, (5) quality of NDA signals being measured, (6) potential to aid safeguards over the coming decades, and (7) potential to help the U.S.A in fulfilling our international safeguards and eventual repository needs. Addressing each of these points individually:

- 1. Quality and quantity of fuel:** No facility exists in the U.S.A. like the Swedish interim fuel storage facility called Clab (*Centralt mellanlager för använt kärnbränsle*). The Swedes have operated 10 commercial nuclear power plants over 40 years, both PWR and BWR. The fuel from all these reactors and some experimental fuel are all at Clab and available to be measured since the Swedes minimize on-site storage at reactors. In selecting fuel for our collaborative research, we looked at the full breadth of the fuel. As a minimum, we will measure 25 BWR and 25 PWR assemblies that vary significantly in initial enrichment, burnup and cooling time. The Swedish opportunity is far superior to the other field deployment opportunities in process under the NGSI-SF Project: ROK (8 PWR assemblies) and Japan (7 MOX research reactor assemblies).
- 2. Likely long term deployment:** Sweden and Finland are leading the world regarding spent fuel repositories. They are no longer delaying the technical challenges involved; they are developing solutions. NDA can help them satisfy the relevant safeguards and safety needs as required by the relevant regulatory bodies (IAEA, Euratom and domestic Swedish inspectorate). The solutions they will determine will be used to measure more than 20,000 spent fuel assemblies.

3. **Seriousness of our collaborator to invest in this research effort and to allow us to be actively involved:** The Swedish encapsulation/repository will cost over a billion U.S. dollars to design, build and operate. The company responsible for the encapsulation/repository facilities (Svensk Kärnbränslehantering, *SKB*) is very dedicated to intelligently designing the facility. Optimal selection of the NDA system is recognized as an important part of the design. Steve Tobin will have an office at SKB headquarters in Stockholm as well as at the primary University (Uppsala) involved in NDA research in Sweden.
4. **Cost effectiveness:** In 2010, representatives from LANL and INL meet at INL to discuss the possibility of measuring commercial spent fuel at INL. The preliminary cost estimate to move fuel from storage casks at INL to a pool at INL was ~\$2 million. Note this cost did not include the cost of the measurement campaign; also, the quality, quantity and likelihood of a long term deployment, as outlined above, are noticeably inferior at INL compared to Sweden given the encapsulation/repository needs that are driving the research in Sweden. In collaborating with Sweden we pay our own costs only, the Swedes cover 100% of the operational costs of Clab.
5. **Quality of research (NDA signals and burnup code) being conducted is unprecedented:** Because of the seriousness of the encapsulation/repository needs, SKB has been willing to host/provide more facility demanding NDA techniques than our other NGSF-SF collaborators; additionally SKB is investing in burnup code research. From an NDA perspective the following are planned:
  1. Differential Die-Away (DDA) – neutron generator based technique
    - a. Note the Californium Interrogation Prompt Neutron (CIPN) technique can be tested out with the same hardware by running the generator in steady-state.
  2. Differential Die-Away Self-interrogation (DDSI) – correlated neutron detection
  3. Full length (~4 meters) calorimeter (*already functioning at Clab*, over 100 assemblies measured)
  4. Passive gamma from four sides of assembly (collimated pool side view *built into Clab*)
    - a. HPGe and LaBr3 are certain to be used
    - b. Others under discussion (CZT, NaI,
  5. Total Neutron (TN) and Gross Gamma with ion chambersIn the context of burnup code research, SKB is combining research done by ORNL (SCALE) with the industrial codes used by the reactor operator (codes developed by Stutsvik).

- 6. Potential to aid safeguards over the coming decades:** Given the combined NDA signatures and burnup code research on a wide range of PWR and BWR fuel, SKB is creating the best researched spent fuel library in the world. These may function as a resource for inspectorates worldwide. A key issue underlining all spent fuel NDA research is the absence of any form of standards; these Swedish assemblies may function as working standards. In previous conversations, SKB is interested in separating the assemblies we will research from the others at Clab and in keeping these assemblies out of the repository for the foreseeable future.
- 7. Potential to help the U.S.A in fulfilling our international safeguards and eventual repository needs:** Given that the Swedes are advance in designing, building and operating their repository, particularly in the context of international safeguards, the U.S. A. has much to learn by being actively involved with them.

6.0. Conclusions

In conclusion, the benefit of using NDA to inform Working Standards calculations has been demonstrated in this document as researched in Tasks 6.1, 6.3, and 6.4 of the NGSF-SF project in FY13. PDET measurements were used to estimate pin-by-pin burnup maps in spent fuel assemblies at a Republic of Korea PIEF. The resulting impact of these calculations in terms of plutonium content and contribution to NDA instruments compared to operator-declared values were relatively small and mainly a function of changes in the average assembly burnup. The ROK assemblies are relatively unique in that they are well-characterized in terms of the high level of operating history information available as provided by the Korean nuclear operators. This information, combined with available DA measurements, makes the ROK assemblies ideal for initial NDA instrument testing and calibration. However, as the application of advanced NDA moves beyond the research and development environment to commercial power plants, the availability of detailed operator data may be more limited than ROK experience, and there will be increased reliance on independent methods to establish Working Standard fuel assemblies for the purpose of instrument calibration.

At a typical commercial facility, the amount of data provided is questionable (in particular, pin-by-pin burnup maps may not be provided), so performing Working Standards simulations may prove more challenging. An example was given in this document on how changes in input data affect infinitely-reflected assembly simulations by trying to match NDA gamma scans in rods spread throughout a PWR assembly. Unfortunately, the ability to obtain similar results for fission product peaks did always correspond to more accurate plutonium predictions.

However, improvements were found by using NDA data compared to a base case without such information.

A focus of work in FY13 was to show how the use of DA and/or NDA, and the completeness and the accuracy of declared assembly information change simulated predictions of plutonium content and other nuclides important to advanced NDA instrument calibration and performance. Such changes were documented and did show improvement compared to simulations that did not use NDA. How NDA measurements are currently being included for spent fuel assemblies at a Swedish facility was also discussed. The resulting NDA measurements will be processed similarly to those included in this report to help develop Working Standards of some of the Swedish assemblies in the near future.

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Appendix A. Details on Run Time and Uncertainty for Takahama Assembly

Minimizing Monteburns Execution Time and Computational Uncertainty

There are 4 primary variables that impact run time – number of material zones, number of burn steps, number of MCNP source neutrons, and number of isotopes tracked per material. It was anticipated that there would be several (dozens) scoping/sensitivity calculations performed for this study, and initial MONTEBURNS cases were taking many days. Thus, the first task was to reduce run time as much as possible while still producing meaningful results for scoping calculations. It was found that a maximum burn step lengths (impacting number of burn steps) of ~60 days were acceptable for scoping, along with ~8 million source neutrons and a MONTEBURNS fractional importance (impacting isotopes tracked per material) of 0.00001. The number of materials could effectively go no lower than the number of pins, because symmetry could not be utilized. The question was if more nodes would be needed. In this regard, it was assumed that axial noding would not substantially change the bulk pin results, whereas radial noding might (especially for pins that contain Gd). The impact of using 4 radial zones (rings) in the pins was found to be significant, but not large enough such that scoping studies wouldn't be effective with one ring. As a result, the scoping Monteburns cases had 17 burn steps, 8 megahistory MCNP runs, a fractional importance of 0.00001, and one material zone for every fuel pin (258 zones). These cases took between 8 to 12 hours using about 40 processors (80 threads), with an error of <1% for each individual isotope result. For the final results of the study, a more accurate calculation was performed, with 15 megahistories, a fractional importance of 0.000001, 4 rings per fuel pin (>1000 burn materials), and <40 day burn steps, resulting in 29 total steps.

Computational Uncertainties

In a Monte Carlo burnup solution, there is always a question of how to propagate statistical error from the individual Monte Carlo solutions (cross sections and fluxes) from each burn step. In an attempt to answer this question, a sample burn case was run 6 times with different random seed numbers for each MCNP run, and the actual standard deviation of the data was calculated. Each of the calculations for this statistical test used the reference scoping parameters mentioned above (e.g. 8 million source neutrons, etc.).

On average, the relative statistical error on the burnup and NDA isotopes was <<1%. Some of the relative error values are zero because there were only 3 significant digits on the results, and the output was identical for all 6 cases. The only isotope with an average relative error >0.34% was Cs134, which saw an average relative error of 0.91%. This suggests that the Cs133 and Cs134 have cross sections that are more variable with energy than the other isotopes, thus they would require more energies in the sample to arrive at a statistically accurate cross section.

Appendix B. Calculated Data and Measurements for All Cases.

Measured and Calculated Isotopics (g/TIHM) for pin SF95 -- First Set of Cases										
g/TIHM	measured	takf2a	takf2b	takg2a	takh2a	takh2b	takh2c	takh2d	takh2e	takh2f
U-235	1.40E+04	1.41E+04	1.42E+04	1.40E+04	1.41E+04	1.39E+04	1.41E+04	1.41E+04	1.37E+04	1.38E+04
U-236	4.76E+03	4.76E+03	4.75E+03	4.77E+03	4.78E+03	4.78E+03	4.75E+03	4.75E+03	4.80E+03	4.78E+03
U-238	9.36E+05	9.35E+05	9.35E+05	9.35E+05	9.35E+05	9.41E+05	9.41E+05	9.41E+05	9.41E+05	9.41E+05
Pu-238	1.39E+02	1.20E+02	1.23E+02	1.25E+02	1.22E+02	1.17E+02	1.18E+02	1.17E+02	1.22E+02	1.19E+02
Pu-239	5.89E+03	5.83E+03	5.92E+03	5.86E+03	5.88E+03	5.59E+03	5.62E+03	5.66E+03	5.67E+03	5.67E+03
Pu-240	2.05E+03	2.11E+03	2.10E+03	2.12E+03	2.12E+03	2.09E+03	2.08E+03	2.07E+03	2.12E+03	2.10E+03
Pu-241	1.35E+03	1.29E+03	1.32E+03	1.30E+03	1.29E+03	1.24E+03	1.24E+03	1.25E+03	1.27E+03	1.26E+03
Pu-242	4.13E+02	3.80E+02	3.81E+02	3.83E+02	3.72E+02	3.69E+02	3.64E+02	3.64E+02	3.83E+02	3.77E+02
Am-241	2.58E+01	3.23E+01	3.28E+01	3.46E+01	3.21E+01	3.05E+01	3.28E+01	3.29E+01	3.36E+01	3.32E+01
Am-242m	6.84E-01	4.50E-01	4.60E-01	4.85E-01	4.44E-01	4.06E-01	4.49E-01	4.51E-01	4.57E-01	4.47E-01
Am-243	7.13E+01	7.04E+01	7.02E+01	7.29E+01	7.12E+01	6.65E+01	6.59E+01	6.59E+01	7.12E+01	6.71E+01
Cs-137	1.29E+03	1.22E+03	1.22E+03	1.23E+03	1.22E+03	1.21E+03	1.21E+03	1.21E+03	1.24E+03	1.23E+03
Cs-134	1.30E+02	1.27E+02	1.28E+02	1.27E+02	1.28E+02	1.24E+02	1.22E+02	1.20E+02	1.25E+02	1.24E+02
Eu-154	2.34E+01	2.42E+01	2.44E+01	2.47E+01	2.25E+01	2.12E+01	2.34E+01	2.33E+01	2.42E+01	2.38E+01
Ce-144	4.13E+02	4.33E+02	4.31E+02	4.10E+02	4.32E+02	4.27E+02	4.06E+02	4.06E+02	4.13E+02	4.09E+02
Sb-125	3.21E+00	7.62E+00	7.64E+00	7.56E+00	7.65E+00	7.47E+00	7.41E+00	7.41E+00	7.59E+00	7.47E+00
Ru-106	1.29E+02	1.50E+02	1.50E+02	1.51E+02	1.50E+02	1.42E+02	1.46E+02	1.46E+02	1.51E+02	1.48E+02

Measured and Calculated Isotopics (g/TIHM) for pin SF96 -- First Set of Cases										
g/TIHM	measured	takf2a	takf2b	takg2a	takh2a	takh2b	takh2c	takh2d	takh2e	takh2f
U-235	9.31E+03	9.42E+03	9.50E+03	9.31E+03	9.35E+03	9.28E+03	9.09E+03	9.09E+03	9.15E+03	9.22E+03
U-236	3.11E+03	3.10E+03	3.10E+03	3.10E+03	3.14E+03	3.13E+03	3.14E+03	3.14E+03	3.13E+03	3.12E+03
U-238	9.50E+05	9.49E+05	9.49E+05	9.49E+05	9.54E+05	9.54E+05	9.54E+05	9.54E+05	9.54E+05	9.54E+05
Pu-238	1.08E+02	8.79E+01	9.02E+01	9.23E+01	9.09E+01	8.43E+01	8.63E+01	8.76E+01	8.89E+01	8.69E+01
Pu-239	5.70E+03	5.57E+03	5.65E+03	5.61E+03	5.60E+03	5.31E+03	5.38E+03	5.38E+03	5.45E+03	5.43E+03
Pu-240	2.15E+03	2.26E+03	2.21E+03	2.23E+03	2.25E+03	2.26E+03	2.22E+03	2.23E+03	2.22E+03	2.22E+03
Pu-241	1.36E+03	1.34E+03	1.40E+03	1.37E+03	1.37E+03	1.28E+03	1.31E+03	1.31E+03	1.33E+03	1.30E+03
Pu-242	4.66E+02	4.47E+02	4.56E+02	4.53E+02	4.52E+02	4.44E+02	4.48E+02	4.43E+02	4.45E+02	4.39E+02
Am-241	2.72E+01	3.06E+01	3.17E+01	3.34E+01	3.11E+01	2.94E+01	3.18E+01	3.16E+01	3.22E+01	3.20E+01
Am-242m	6.17E-01	4.06E-01	4.25E-01	4.52E-01	4.13E-01	3.76E-01	4.11E-01	4.12E-01	4.22E-01	4.22E-01
Am-243	7.99E+01	8.20E+01	8.27E+01	8.33E+01	8.11E+01	7.84E+01	7.84E+01	8.04E+01	7.97E+01	7.91E+01
Cs-137	9.59E+02	9.63E+02	9.65E+02	9.77E+02	9.74E+02	9.61E+02	9.80E+02	9.80E+02	9.80E+02	9.74E+02
Cs-134	9.13E+01	9.25E+01	9.39E+01	9.41E+01	9.41E+01	9.02E+01	9.15E+01	9.22E+01	9.28E+01	9.02E+01
Eu-154	1.77E+01	1.97E+01	2.12E+01	2.21E+01	1.83E+01	1.70E+01	1.98E+01	2.08E+01	1.99E+01	1.98E+01
Ce-144	3.22E+02	3.69E+02	3.70E+02	3.54E+02	3.71E+02	3.65E+02	3.56E+02	3.56E+02	3.56E+02	3.53E+02
Sb-125	4.04E+00	7.08E+00	7.13E+00	7.08E+00	7.12E+00	6.99E+00	6.99E+00	6.99E+00	6.99E+00	6.99E+00
Ru-106	1.24E+02	1.60E+02	1.61E+02	1.62E+02	1.61E+02	1.52E+02	1.59E+02	1.59E+02	1.60E+02	1.58E+02

Activity Ratios (curies/curies Cs-137) -- Second Set of Cases										
Pin C7	measured	takf2a	takf2b	takg2a	takh2a	takh2b	takh2c	takh2d	takh2e	takh2f
BU	3.32E+01	33.63	33.662	33.535	33.66	33.7	33.34	33.32	33.55	33.37
Cs134	1.47E+00	1.53E+00	1.57E+00	1.53E+00	1.56E+00	1.54E+00	1.49E+00	1.48E+00	1.49E+00	1.49E+00
Eu154	5.42E-02	5.99E-02	6.11E-02	6.14E-02	5.58E-02	5.33E-02	5.83E-02	5.83E-02	5.97E-02	5.94E-02
Ce144	1.23E+01	1.31E+01	1.31E+01	1.23E+01	1.31E+01	1.31E+01	1.24E+01	1.24E+01	1.23E+01	1.24E+01
Ru106	4.79E+00	4.66E+00	4.69E+00	4.65E+00	4.66E+00	4.46E+00	4.55E+00	4.56E+00	4.59E+00	4.57E+00
Sb125	6.84E-02	7.46E-02	7.51E-02	7.34E-02	7.46E-02	7.39E-02	7.28E-02	7.28E-02	7.29E-02	7.28E-02
Pin C5	measured	takf2a	takf2b	takg2a	takh2a	takh2b	takh2c	takh2d	takh2e	takh2f
BU	3.16E+01	31.163	31.16	31.292	31	31.18	30.81	30.87	31.41	31.27
Cs134	1.41E+00	1.47E+00	1.48E+00	1.46E+00	1.47E+00	1.43E+00	1.40E+00	1.40E+00	1.43E+00	1.41E+00
Eu154	5.28E-02	5.98E-02	6.03E-02	6.07E-02	5.53E-02	5.27E-02	5.81E-02	5.79E-02	5.87E-02	5.82E-02
Ce144	1.25E+01	1.32E+01	1.32E+01	1.24E+01	1.32E+01	1.31E+01	1.25E+01	1.25E+01	1.24E+01	1.24E+01
Ru106	4.77E+00	4.56E+00	4.58E+00	4.56E+00	4.55E+00	4.37E+00	4.45E+00	4.47E+00	4.49E+00	4.48E+00
Sb125	6.89E-02	7.42E-02	7.45E-02	7.31E-02	7.43E-02	7.37E-02	7.23E-02	7.25E-02	7.24E-02	7.22E-02
Pin C3	measured	takf2a	takf2b	takg2a	takh2a	takh2b	takh2c	takh2d	takh2e	takh2f
BU	3.29E+01	32.983	33.095	33.12	32.99	32.86	32.68	32.7	33.27	33.09
Cs134	1.48E+00	1.54E+00	1.57E+00	1.53E+00	1.55E+00	1.52E+00	1.47E+00	1.48E+00	1.49E+00	1.48E+00
Eu154	5.66E-02	6.12E-02	6.18E-02	6.22E-02	5.71E-02	5.46E-02	5.97E-02	5.93E-02	6.00E-02	6.03E-02
Ce144	1.24E+01	1.31E+01	1.31E+01	1.23E+01	1.31E+01	1.30E+01	1.24E+01	1.23E+01	1.23E+01	1.23E+01
Ru106	4.81E+00	4.66E+00	4.68E+00	4.66E+00	4.67E+00	4.44E+00	4.56E+00	4.56E+00	4.58E+00	4.58E+00
Sb125	6.93E-02	7.47E-02	7.51E-02	7.37E-02	7.46E-02	7.39E-02	7.27E-02	7.29E-02	7.28E-02	7.26E-02
Pin A6	measured	takf2a	takf2b	takg2a	takh2a	takh2b	takh2c	takh2d	takh2e	takh2f
BU	3.10E+01	30.78	30.775	30.95	30.74	30.73	30.43	30.51	31.09	30.91
Cs134	1.39E+00	1.45E+00	1.46E+00	1.44E+00	1.47E+00	1.42E+00	1.39E+00	1.37E+00	1.40E+00	1.40E+00
Eu154	5.26E-02	5.91E-02	5.97E-02	6.02E-02	5.52E-02	5.26E-02	5.76E-02	5.73E-02	5.83E-02	5.76E-02
Ce144	1.31E+01	1.32E+01	1.32E+01	1.24E+01	1.33E+01	1.32E+01	1.26E+01	1.25E+01	1.24E+01	1.25E+01
Ru106	4.76E+00	4.55E+00	4.57E+00	4.52E+00	4.55E+00	4.35E+00	4.44E+00	4.44E+00	4.46E+00	4.46E+00
Sb125	7.20E-02	7.41E-02	7.44E-02	7.30E-02	7.39E-02	7.32E-02	7.26E-02	7.21E-02	7.20E-02	7.25E-02

Measured and Calculated Isotopics (g/TIHM) for pin SF95 -- Second Set of Cases										
g/TIHM	measured	taki2a	taki2b	taki2c	taki3c	takj2a	takj2b	takj3a	takj3b	takk2a
U-235	1.40E+04	1.34E+04	1.42E+04	1.35E+04	1.37E+04	1.42E+04	1.42E+04	1.44E+04	1.41E+04	1.44E+04
U-236	4.76E+03	4.81E+03	4.72E+03	4.78E+03	4.79E+03	4.73E+03	4.77E+03	4.74E+03	4.77E+03	4.72E+03
U-238	9.36E+05	9.37E+05	9.37E+05	9.37E+05	9.37E+05	9.41E+05	9.35E+05	9.41E+05	9.41E+05	9.37E+05
Pu-238	1.39E+02	1.20E+02	1.15E+02	1.17E+02	1.19E+02	1.15E+02	1.20E+02	1.18E+02	1.18E+02	1.18E+02
Pu-239	5.89E+03	5.50E+03	5.63E+03	5.49E+03	5.58E+03	5.65E+03	5.85E+03	5.82E+03	5.81E+03	5.84E+03
Pu-240	2.05E+03	2.10E+03	2.05E+03	2.07E+03	2.08E+03	2.08E+03	2.10E+03	2.09E+03	2.08E+03	2.06E+03
Pu-241	1.35E+03	1.26E+03	1.24E+03	1.25E+03	1.27E+03	1.24E+03	1.30E+03	1.27E+03	1.27E+03	1.28E+03
Pu-242	4.13E+02	3.96E+02	3.62E+02	3.89E+02	3.87E+02	3.59E+02	3.71E+02	3.59E+02	3.67E+02	3.63E+02
Am-241	2.58E+01	3.30E+01	3.28E+01	3.29E+01	3.35E+01	3.29E+01	3.45E+01	3.34E+01	3.35E+01	3.40E+01
Am-242m	6.84E-01	4.40E-01	4.50E-01	4.43E-01	4.55E-01	4.47E-01	4.82E-01	4.63E-01	4.59E-01	4.75E-01
Am-243	7.13E+01	7.20E+01	6.51E+01	7.00E+01	7.01E+01	6.47E+01	7.00E+01	6.35E+01	6.65E+01	6.74E+01
Cs-137	1.29E+03	1.25E+03	1.20E+03	1.24E+03	1.23E+03	1.21E+03	1.22E+03	1.21E+03	1.22E+03	1.20E+03
Cs-134	1.30E+02	1.27E+02	1.20E+02	1.24E+02	1.25E+02	1.21E+02	1.24E+02	1.22E+02	1.22E+02	1.21E+02
Eu-154	2.34E+01	2.37E+01	2.32E+01	2.35E+01	2.37E+01	2.32E+01	2.44E+01	2.36E+01	2.38E+01	2.38E+01
Ce-144	4.13E+02	4.15E+02	4.03E+02	4.12E+02	4.10E+02	4.04E+02	4.07E+02	4.02E+02	4.07E+02	4.01E+02
Sb-125	3.21E+00	7.63E+00	7.34E+00	7.54E+00	7.54E+00	7.35E+00	7.47E+00	7.35E+00	7.41E+00	7.37E+00
Ru-106	1.29E+02	1.51E+02	1.44E+02	1.49E+02	1.49E+02	1.45E+02	1.48E+02	1.45E+02	1.47E+02	1.46E+02

Measured and Calculated Isotopics (g/TIHM) for pin SF96 -- Second Set of Cases										
g/TIHM	measured	taki2a	taki2b	taki2c	taki3c	takj2a	takj2b	takj3a	takj3b	takk2a
U-235	9.31E+03	9.26E+03	9.19E+03	9.41E+03	9.49E+03	9.22E+03	9.41E+03	9.28E+03	9.28E+03	9.53E+03
U-236	3.11E+03	3.09E+03	3.10E+03	3.06E+03	3.06E+03	3.12E+03	3.11E+03	3.12E+03	3.12E+03	3.07E+03
U-238	9.50E+05	9.50E+05	9.50E+05	9.51E+05	9.51E+05	9.54E+05	9.54E+05	9.54E+05	9.54E+05	9.50E+05
Pu-238	1.08E+02	8.50E+01	8.59E+01	8.35E+01	8.41E+01	8.63E+01	8.95E+01	8.76E+01	8.76E+01	8.68E+01
Pu-239	5.70E+03	5.36E+03	5.33E+03	5.35E+03	5.44E+03	5.37E+03	5.59E+03	5.50E+03	5.48E+03	5.55E+03
Pu-240	2.15E+03	2.17E+03	2.19E+03	2.17E+03	2.16E+03	2.20E+03	2.23E+03	2.24E+03	2.22E+03	2.19E+03
Pu-241	1.36E+03	1.30E+03	1.30E+03	1.28E+03	1.31E+03	1.31E+03	1.34E+03	1.33E+03	1.33E+03	1.33E+03
Pu-242	4.66E+02	4.36E+02	4.41E+02	4.23E+02	4.26E+02	4.37E+02	4.41E+02	4.35E+02	4.43E+02	4.32E+02
Am-241	2.72E+01	3.15E+01	3.15E+01	3.13E+01	3.19E+01	3.16E+01	3.31E+01	3.21E+01	3.26E+01	3.27E+01
Am-242m	6.17E-01	4.12E-01	4.11E-01	4.08E-01	4.21E-01	4.14E-01	4.37E-01	4.28E-01	4.31E-01	4.38E-01
Am-243	7.99E+01	7.59E+01	7.76E+01	7.32E+01	7.49E+01	7.71E+01	7.91E+01	7.84E+01	7.71E+01	7.67E+01
Cs-137	9.59E+02	9.60E+02	9.65E+02	9.47E+02	9.48E+02	9.67E+02	9.74E+02	9.74E+02	9.74E+02	9.52E+02
Cs-134	9.13E+01	8.92E+01	9.06E+01	8.76E+01	8.78E+01	9.15E+01	9.22E+01	9.28E+01	9.22E+01	8.98E+01
Eu-154	1.77E+01	2.06E+01	2.06E+01	2.02E+01	2.06E+01	2.07E+01	2.14E+01	2.12E+01	2.11E+01	2.09E+01
Ce-144	3.22E+02	3.50E+02	3.52E+02	3.46E+02	3.46E+02	3.52E+02	3.52E+02	3.52E+02	3.53E+02	3.47E+02
Sb-125	4.04E+00	6.89E+00	6.93E+00	6.80E+00	6.81E+00	6.93E+00	6.99E+00	6.99E+00	6.99E+00	6.87E+00
Ru-106	1.24E+02	1.56E+02	1.57E+02	1.53E+02	1.54E+02	1.57E+02	1.59E+02	1.58E+02	1.58E+02	1.56E+02

Activity Ratios (curies/curies Cs-137) -- Second Set of Cases										
Pin C7	measured	taki2a	taki2b	taki2c	taki3c	takj2a	takj2b	takj3a	takj3b	takk2a
BU	3.32E+01	33.48	33.1	33.095	33.105	33.11	33.14	33.11	33.15	32.817
Cs134	1.47E+00	1.49E+00	1.47E+00	1.47E+00	1.48E+00	1.47E+00	1.51E+00	1.49E+00	1.51E+00	1.49E+00
Eu154	5.42E-02	5.85E-02	5.87E-02	5.81E-02	5.91E-02	5.75E-02	6.03E-02	5.98E-02	5.92E-02	5.98E-02
Ce144	1.23E+01	1.24E+01	1.24E+01	1.24E+01	1.24E+01	1.24E+01	1.24E+01	1.24E+01	1.24E+01	1.24E+01
Ru106	4.79E+00	4.57E+00	4.54E+00	4.54E+00	4.56E+00	4.54E+00	4.60E+00	4.57E+00	4.56E+00	4.58E+00
Sb125	6.84E-02	7.30E-02	7.28E-02	7.28E-02	7.30E-02	7.26E-02	7.32E-02	7.32E-02	7.32E-02	7.32E-02
Pin C5	measured	taki2a	taki2b	taki2c	taki3c	takj2a	takj2b	takj3a	takj3b	takk2a
BU	3.16E+01	31.71	30.65	31.408	31.442	30.59	30.93	30.54	30.88	30.587
Cs134	1.41E+00	1.43E+00	1.40E+00	1.42E+00	1.44E+00	1.39E+00	1.45E+00	1.41E+00	1.42E+00	1.42E+00
Eu154	5.28E-02	5.74E-02	5.80E-02	5.68E-02	5.81E-02	5.76E-02	5.93E-02	5.85E-02	5.94E-02	5.95E-02
Ce144	1.25E+01	1.24E+01	1.25E+01	1.24E+01	1.24E+01	1.26E+01	1.25E+01	1.25E+01	1.25E+01	1.25E+01
Ru106	4.77E+00	4.49E+00	4.45E+00	4.47E+00	4.49E+00	4.45E+00	4.51E+00	4.46E+00	4.48E+00	4.49E+00
Sb125	6.89E-02	7.26E-02	7.24E-02	7.25E-02	7.27E-02	7.27E-02	7.25E-02	7.27E-02	7.25E-02	7.26E-02
Pin C3	measured	taki2a	taki2b	taki2c	taki3c	takj2a	takj2b	takj3a	takj3b	takk2a
BU	3.29E+01	33.68	32.438	33.342	33.34	32.44	32.79	32.48	32.77	32.43
Cs134	1.48E+00	1.50E+00	1.47E+00	1.50E+00	1.50E+00	1.47E+00	1.51E+00	1.49E+00	1.49E+00	1.50E+00
Eu154	5.66E-02	5.92E-02	5.97E-02	5.89E-02	5.98E-02	6.00E-02	6.22E-02	6.11E-02	6.10E-02	6.16E-02
Ce144	1.24E+01	1.23E+01	1.24E+01	1.23E+01	1.23E+01	1.24E+01	1.23E+01	1.23E+01	1.23E+01	1.23E+01
Ru106	4.81E+00	4.59E+00	4.56E+00	4.56E+00	4.59E+00	4.57E+00	4.61E+00	4.58E+00	4.59E+00	4.60E+00
Sb125	6.93E-02	7.31E-02	7.30E-02	7.30E-02	7.32E-02	7.31E-02	7.35E-02	7.31E-02	7.35E-02	7.33E-02
Pin A6	measured	taki2a	taki2b	taki2c	taki3c	takj2a	takj2b	takj3a	takj3b	takk2a
BU	3.10E+01	31.403	30.248	31.06	31.038	30.22	30.59	30.19	30.52	30.225
Cs134	1.39E+00	1.41E+00	1.39E+00	1.40E+00	1.41E+00	1.37E+00	1.44E+00	1.40E+00	1.41E+00	1.41E+00
Eu154	5.26E-02	5.73E-02	5.74E-02	5.67E-02	5.76E-02	5.70E-02	5.97E-02	5.89E-02	5.87E-02	5.92E-02
Ce144	1.31E+01	1.24E+01	1.25E+01	1.25E+01	1.24E+01	1.25E+01	1.25E+01	1.25E+01	1.25E+01	1.25E+01
Ru106	4.76E+00	4.47E+00	4.44E+00	4.43E+00	4.46E+00	4.41E+00	4.50E+00	4.45E+00	4.46E+00	4.47E+00
Sb125	7.20E-02	7.25E-02	7.23E-02	7.23E-02	7.24E-02	7.24E-02	7.27E-02	7.24E-02	7.27E-02	7.27E-02