LA-UR- 10-06933

Approved for public release; distribution is unlimited.

Title:	Predicting Fissile Content of Spent Nuclear Fuel Assemblies with the Passive Neutron Albedo Reactivity Technique and Monte Carlo Code Emulation
Author(s):	Jeremy Lloyd Conlin Stephen J. Tobin
Intended for:	International Conference on Mathematics and Computational Methods Applied to Nuclear Science and Engineering May 8–11, 2011 Rio de Janeiro, Brazil

Form 836 (7/06)

International Conference on Mathematics and Computational Methods Applied to Nuclear Science and Engineering (M&C 2011)
Rio de Janeiro, RJ, Brazil, May 8-12, 2011, on CD-ROM, Latin American Section (LAS) / American Nuclear Society (ANS)
ISBN 978-85-63688-00-2

Predicting Fissile Content of Spent Nuclear Fuel Assemblies with the Passive Neutron Albedo Reactivity Technique and Monte Carlo Code Emulation

Jeremy Lloyd Conlin and Stephen J. Tobin

Los Alamos National Laboratory
Los Alamos, NM
jlconlin@lanl.gov; tobin@lanl.gov

ABSTRACT

There is a great need in the safeguards community to be able to nondestructively quantify the mass of plutonium of a spent nuclear fuel assembly. As part of the Next Generation of Safeguards Initiative, we are investigating several techniques, or detector systems, which, when integrated, will be capable of quantifying the plutonium mass of a spent fuel assembly without dismantling the assembly. This paper reports on the simulation of one of these techniques, the Passive Neutron Albedo Reactivity with Fission Chambers (PNAR-FC) system. The response of this system over a wide range of spent fuel assemblies with different burnup, initial enrichment, and cooling time characteristics is shown. A Monte Carlo method of using these modeled results to estimate the fissile content of a spent fuel assembly has been developed. A few numerical simulations of using this method are shown. Finally, additional developments still needed and being worked on are discussed.

Key Words: Monte Carlo, code emulator, fissile content, spent nuclear fuel, safeguards.

1. Introduction

The quantification of the plutonium mass in spent nuclear fuel assemblies is a problem of great concern to the International Atomic Energy Agency (IAEA). Their efforts to prevent the proliferation of nuclear weapons mandates close monitoring of the spent fuel assemblies after their removal from nuclear reactors around the world to ensure that no material is diverted from the assembly.

An ideal situation for quantifying the mass of plutonium in spent fuel assemblies would be a nondestructive assay (NDA) technique that can be used in a variety of surroundings (e.g., air, water, borated water), and is fast enough so as not to be a burden on the other operations in a reactor, spent fuel pool, or reprocessing facility.

Under the sponsorship of the Next Generation Safeguards Initiative (NGSI) program, an effort is underway [1] to combine a few NDA techniques into one integrated design allowing a nuclear inspector to measure the mass of plutonium in a spent nuclear fuel assembly and determine whether plutonium has been removed from the assembly.

The program to develop a combination of detectors—integrated in such a way as to quantify the mass of plutonium—is, recognizably, a grand challenge [2]. We are currently investigating fourteen different NDA techniques as part of this program. No single technique can measure the plutonium mass directly,

but each provides a measurement, which contributes a piece of the puzzle necessary to quantify the plutonium mass. Some of the quantities that can be measured from an individual measurement are the average burnup of an assembly, the ratio of the mass of the fissile isotopes to the mass of the fertile isotopes (fissile/fertile ratio); the ratio of the mass of elemental uranium to the mass of elemental plutonium (U/Pu ratio); the mass of ^{235}U to the mass of ^{239}Pu (^{235}U/^{239}Pu ratio); and a combination of the masses of the three fissile isotopes, ^{235}U, ^{239}Pu, and ^{241}Pu (fissile content).

The plutonium quantification project is nearing the end of the simulation phase. So far, we have focused on simulating each of the fourteen NDA techniques in MCNPX using a library of spent nuclear fuel models [3]. This paper will demonstrate a method of predicting the fissile content of a spent nuclear fuel assembly using a Monte Carlo estimation and emulation of existing scientific codes. It will present the Monte Carlo estimation procedure while focusing on one of the NDA techniques that measures the fissile content of a spent nuclear fuel assembly, the Passive Neutron Albedo Reactivity with Fission Chambers (PNAR-FC) technique [4].

1.1 Quantifying Fissile Content for Spent Nuclear Fuel

Before we begin discussion of the PNAR-FC system, it is important to define what is meant by the fissile content of a nuclear fuel assembly. The fissile content of fresh nuclear fuel is simply the mass of ^{235}U or the uranium enrichment used in the assembly. In spent nuclear fuel assemblies, determining the fissile content is more ambiguous than simply the uranium enrichment. Several issues make this definition challenging: what fissile isotopes should be included in the definition, how much of those isotopes is in the spent fuel assembly, and should each isotope be weighted equivalently.

In order to quantify the fissile content in a spent nuclear fuel assembly, we have borrowed a concept from neutron coincidence counting [5, Chapter 16], *effective mass*. In our calculations we have defined the ^{239}Pu effective mass as

$$^{239}\mathrm{Pu}_{\mathrm{e}} = C_1 M_{U235} + M_{Pu239} + C_2 M_{Pu241}, \tag{1}$$

where M_i is the mass in grams of isotope i in the assembly and C_1 and C_2 are coefficients defined as

$$C_1 = \frac{\text{Net neutron contribution from induced fission in } ^{235}\text{U per gram of } ^{235}\text{U}}{\text{Net neutron contribution from induced fission in } ^{239}\text{Pu per gram of } ^{239}\text{Pu}}, \tag{2a}$$

$$C_2 = \frac{\text{Net neutron contribution from induced fission in } ^{241}\text{Pu per gram of } ^{241}\text{Pu}}{\text{Net neutron contribution from induced fission in } ^{239}\text{Pu per gram of } ^{239}\text{Pu}}. \tag{2b}$$

Defined in this manner, the units of ^{239}Pu$_{\mathrm{e}}$ are in grams of ^{239}Pu. Alternatively C_1 and C_2 can be written as

$$C_i = \frac{\left(\nu^{(i)} - 1\right)\sigma_f^{(i)} - \sigma_a^{(i)}}{\left(\nu^{(\mathrm{Pu239})} - 1\right)\sigma_f^{(\mathrm{Pu239})} - \sigma_a^{(\mathrm{Pu239})}}, \tag{3}$$

where ν, σ_f, and σ_a are the number of neutrons emitted from fission, fission cross section, and absorption cross section, respectively; ν, σ_f, and σ_a are all energy dependent. Using the definition of ^{239}Pu effective mass in Equation (1), we have a method for quantifying the fissile content of our spent fuel assemblies.

In our simulations of spent nuclear fuel assemblies, a number of tallies in MCNPX are necessary to estimate the C_1 and C_2 coefficients. In each fuel pin of an assembly, for each of the three fissile isotopes, we must tally:

1. Number of fissions (σ_f);
2. Number of fission neutrons ($\nu\sigma_f$); and

3. Number of neutron captures by the isotope (σ_a).

Using these quantities for each of the fissile isotopes and knowing the masses of each of the fissile isotopes, we can calculate the coefficients C_1 and C_2 and the ^{239}Pu effective mass.

2. Passive Neutron Albedo Reactivity with Fission Chambers

As described above, the PNAR-FC [6, 7] technique is one of the methods being investigated as part of the effort to quantify plutonium mass in spent nuclear fuel. The PNAR-FC measures the fissile content of an assembly. As its name suggests, the PNAR technique uses the reflected neutrons (albedo) from the intrinsic neutron emission of a sample of fissile material to self-interrogate and induce fission.

A PNAR detector system surrounds the fissile material with some hydrogenous material (i.e., polyethylene) so that neutrons emitted by the assembly may be thermalized and reflected back into the assembly to potentially induce fission. Along the inside edge of the hydrogenous material is a removable cadmium liner. Neutron detectors are embedded in the hydrogenous material to measure the emission rate of neutrons from the fissile material.

Two measurements are made with the PNAR detector system, one with and one without the cadmium liner present. The ratio of the neutron count *without* the cadmium liner (ω_o) to the neutron count *with* the cadmium liner (ω) can be measured and calculated

$$R_{Cd} = \frac{\omega_o}{\omega}, \tag{4}$$

and is called the cadmium ratio.

The absorption cross section of ^{113}Cd (the important absorbing isotope of cadmium) is shown in Fig. 1 along with the fission cross sections of the three main fissile isotopes; ^{235}U, ^{239}Pu, and ^{241}Pu. It is clear that ^{113}Cd has a very large absorption cross section for thermal neutrons; 99.9% of thermal neutrons impinging on a 1 mm thick sheet of cadmium are absorbed.

The presence of the cadmium liner hardens the neutron spectrum incident on the fissile material; without the cadmium liner, the assembly sees an additional thermal neutron spectrum. This extra thermal spectrum will induce fissions in the fissile material, proportional to the multiplication of the fissile material, and a change in the neutron count will be measured as compared with the measurement with the cadmium liner. Thus the cadmium ratio is a measure of the multiplication induced by thermal neutrons in the fissile material and it scales with fissile content.

We have adapted the PNAR technique for use with a typical, 17 by 17 PWR spent fuel assembly. The geometry is shown in Fig. 2. There are four fission chambers oriented orthogonally to the long axis of the spent fuel assembly, embedded in 5 cm thick polyethylene. The polyethylene is 60 cm long. A removable, 1 mm thick cadmium liner is on the inside of the polyethylene block and the cadmium liner is just 0.5 cm from the edge of the spent fuel assembly. This version of the PNAR technique, using fission chambers, is referred to as PNAR-FC.

We have designed the PNAR-FC detector system using MCNPX [8]. Previously [4], we have reported on the response of the PNAR-FC detector system for assemblies loaded with fresh fuel. In order to simulate the response of the PNAR-FC system with realistic spent fuel assemblies, a library of spent fuel assembly models suitable for use in MCNPX was created [3]. The models have an initial ^{235}U enrichment of 2–5 %, burnup values of 15–60 GWd/t, and time spent outside the of reactor 1–80 years. The combination of all these parameters yields 64 assembly models and represents a range of spent fuel assemblies we expect to see when making measurements of actual spent nuclear fuel assemblies. We have the capability of simulating each of these assemblies surrounded by either water, borated water, or air.

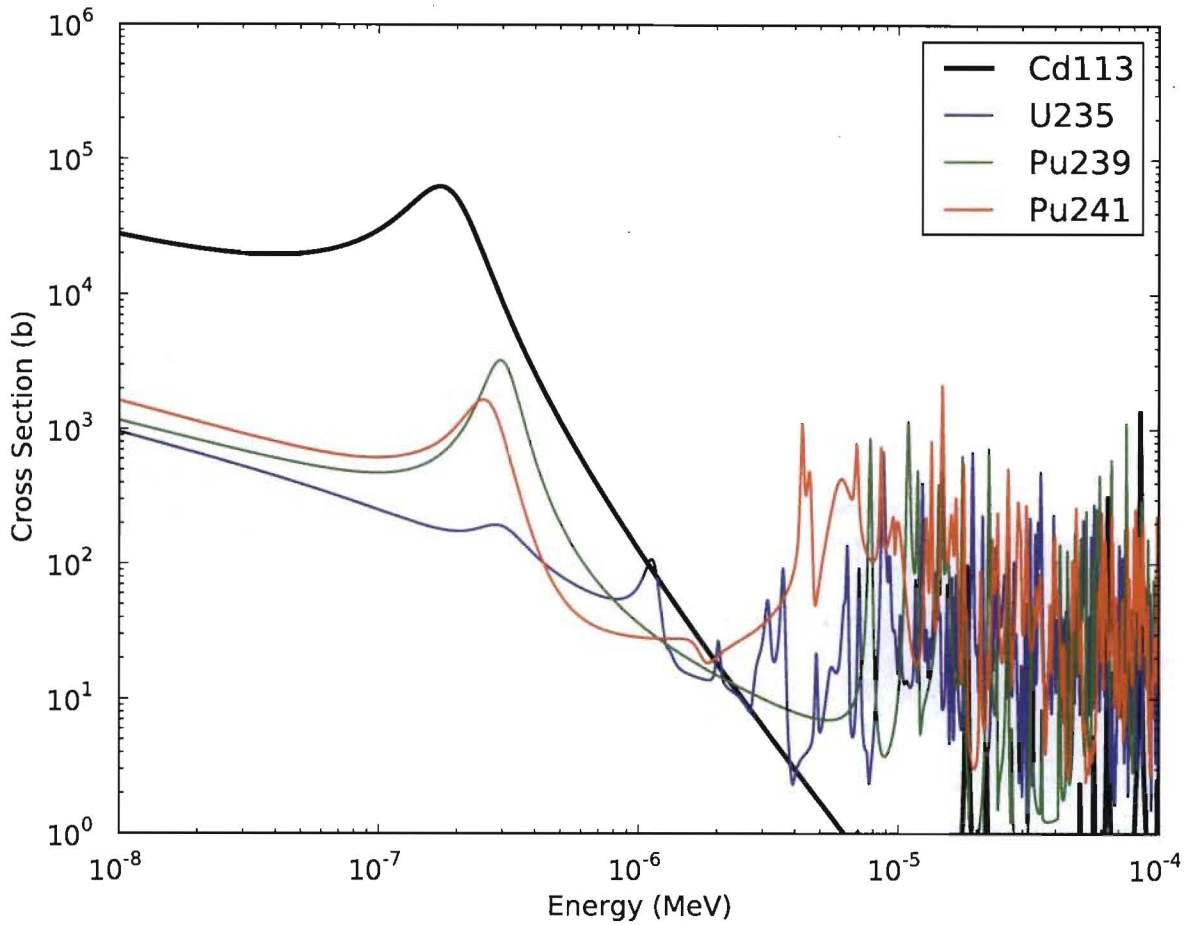
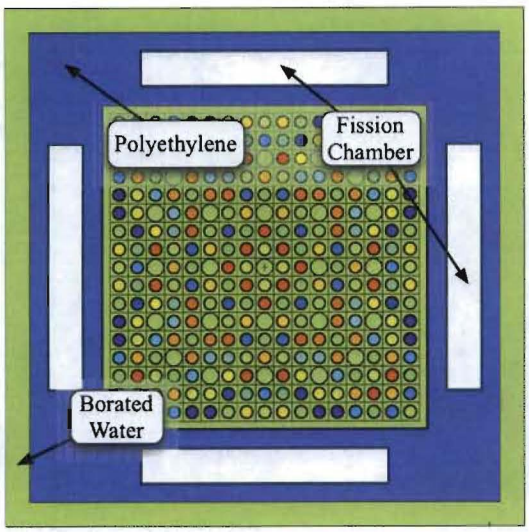


Figure 1: Capture cross section of ^{113}Cd plotted with induced fission cross sections for ^{235}U, ^{239}Pu, and ^{241}Pu.

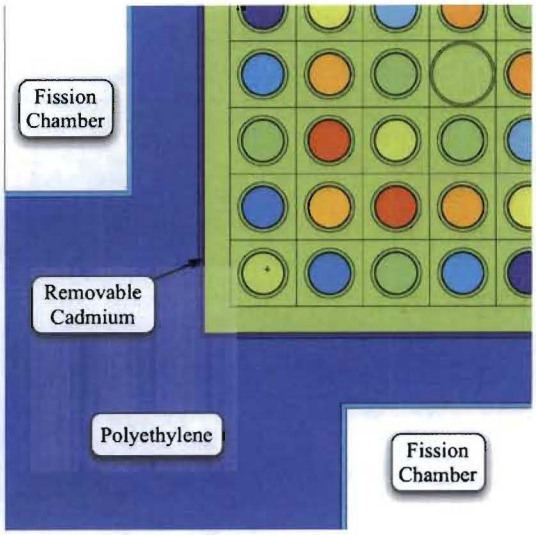
2.1 Estimating ^{239}Pu Effective Mass from Cadmium Ratio

We have simulated the PNAR-FC detector system using the library of spent fuel assembly models (introduced above) using MCNPX. We tallied the induced fission rate in the fission chambers to simulate the counts made in the detector. We also made tallies in each of the fuel pins to give us the data we need to calculate C_1 and C_2 for use in Eq. (1). Fig. 3 shows the response of the PNAR-FC system as a function of the ^{239}Pu effective mass. The assembly parameters of burnup and initial enrichment are delineated by the color and symbol of the data point. The cooling time dependence—while not differentiated by a difference in appearance of the plot marker—can still be identified by recognizing that the ^{239}Pu effective mass decreases with an increase in the cooling time.

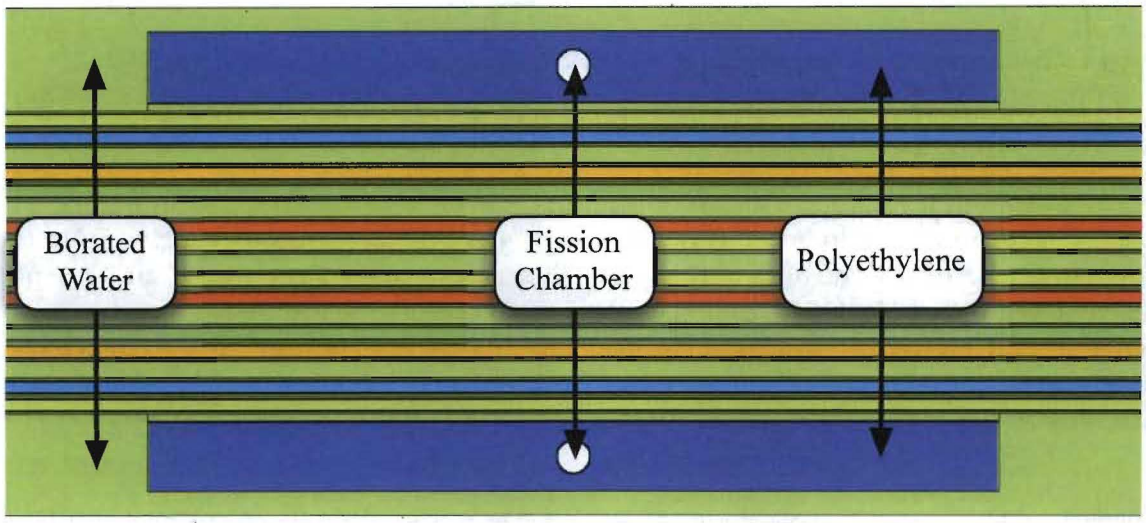
In Fig. 3, we see that the cadmium ratio scales with the ^{239}Pu effective mass, even though the data is fairly scattered. Despite the scatter in the data, it still has some structure based upon our parameters of burnup, cooling time, and initial enrichment. It would be ideal to implement a PNAR-FC instrument in the field if the data in Fig. 3 would have a one-to-one correlation between the cadmium ratio and the ^{239}Pu effective mass. This would allow us to make a measurement of the cadmium ratio and know immediately what the ^{239}Pu effective mass is.



(a) XY slice.



(b) XY slice close-up.



(c) YZ slice.

Figure 2: Geometry of PNAR-FC detector system. The blue region surrounding the assembly is polyethylene, the white cylinders embedded in the polyethylene are the fission chambers. Thin purple region surrounding the assembly (only visible in Fig. (b)) is the removable cadmium liner. The bright green color represents the material surrounding the assembly. For this paper, the surrounding material is borated water at 2200 ppm. Note the three figures are not to scale.

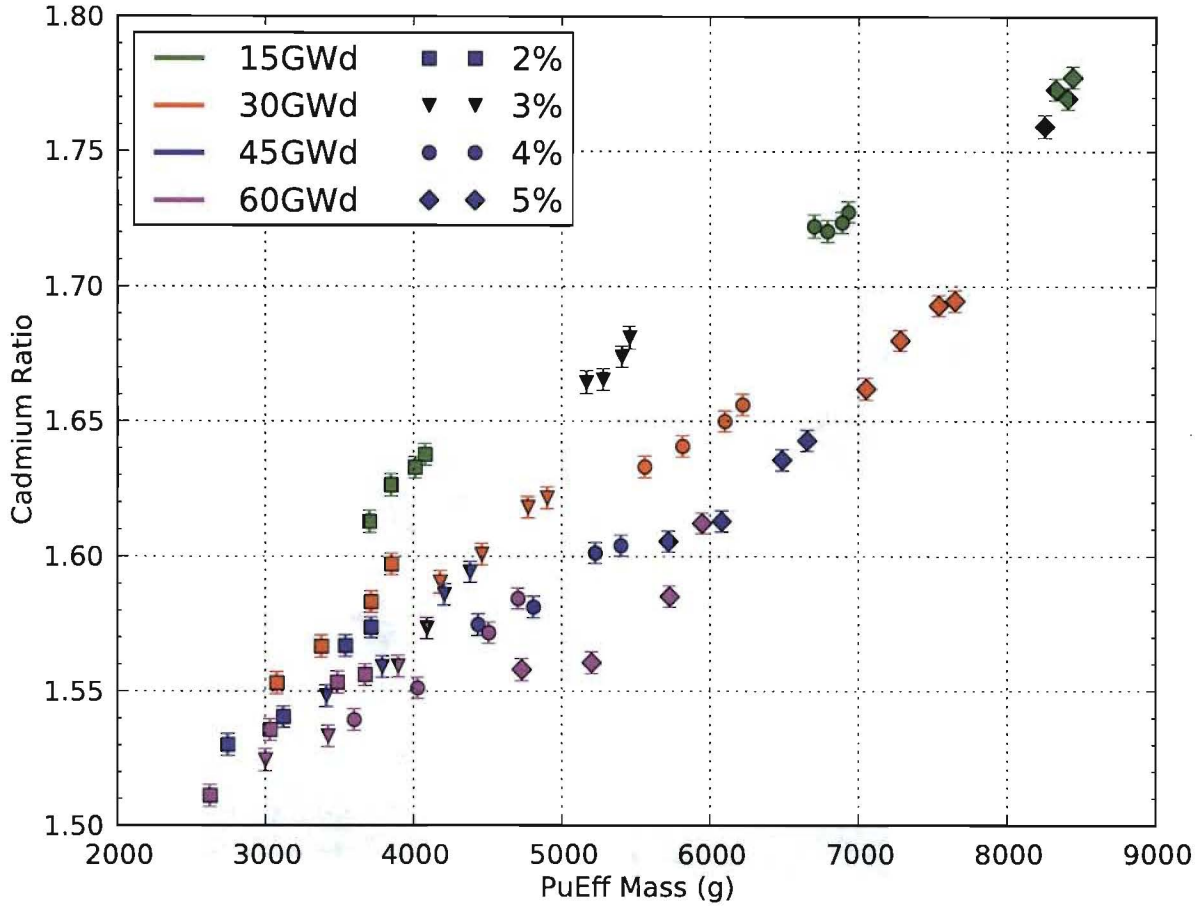


Figure 3: Cadmium ratio as a function of ^{239}Pu effective mass for the PNAR-FC system surrounded by borated water. The colors green, red, blue and purple represent burnups of 15, 30, 45, and 60 GWd/t respectively. Symbols square, triangle, circle, diamond represent initial ^{235}U enrichments of 2, 3, 4, and 5 % respectively. Cooling times of 1, 5, 20, and 80 years can be identified by knowing that the ^{239}Pu effective mass decreases with an increase in cooling time.

The complex isotopic soup that is spent nuclear fuel is far from ideal. We can see in Fig. 3 that for a cadmium ratio of 1.60 with an uncertainty of 0.01, there are no fewer than seven assemblies from our library that could have that cadmium ratio. This leads to very wide range of ^{239}Pu effective mass, which isn't very helpful in identifying the fissile content of the measured assembly. In addition to the data values shown in Fig. 3, there are many other assemblies with a variety of burnup, initial enrichment, and cooling time parameters that would also fall within the range of the measured cadmium ratio, e.g., a 30 GWd/t, 2.5 % ^{235}U initial enrichment, and five-year cooled assembly.

3. Monte Carlo Estimate of ^{239}Pu Effective Mass

Despite the complexity of spent nuclear fuel assemblies we can estimate the mean and distribution of possible values of the ^{239}Pu effective mass for a measured cadmium ratio and its measurement uncertainty.

The mean ^{239}Pu effective mass is

$$^{239}\mathrm{Pu}_{\mathrm{e}}' = \int_{\Delta B}\int_{\Delta E}\int_{\Delta C} M(B,E,C)\,\mathrm{d}C\,\mathrm{d}E\,\mathrm{d}B, \tag{5}$$

where ΔB, ΔE, and ΔC are ranges of possible burnup, initial enrichment, and cooling time for the assembly. $M(B,E,C)$ is some function that maps a spent fuel assembly with burnup, initial enrichment, and cooling time parameters to a ^{239}Pu effective mass.

The multi-dimensional integral in Eq. (5) can be evaluated using a simple Monte Carlo integration,

$$^{239}\mathrm{Pu}_{\mathrm{e}}' \approx \frac{1}{N}\sum_{k=1}^{N} w_k M(B_k,E_k,C_k), \tag{6}$$

where B_k, E_k, and C_k are random samples of burnup, initial enrichment, and cooling time which define a randomly sampled spent fuel assembly. To calculate the *distribution*, or probability density function, associated with $^{239}\mathrm{Pu}_{\mathrm{e}}'$, one can simply bin the individually sampled results and their weights and construct a frequency vs ^{239}Pu effective mass plot.

The integration limits in Eq. (5) are simply the range, from which a parameter value may be sampled. In the Monte Carlo calculation, the parameter limits are sampled uniformly within that range. For the data that we have shown in Fig. 3, the parameter ranges would be 15–60 GWd/t for burnup, 2–5 % ^{235}U for initial enrichment, and 1–80 years for cooling time.

Suppose an independent measurement provided you with information about burnup, i.e., burnup: 45.00 ± 2.25 GWd/t (5% uncertainty). Rather than sampling uniformly from the entire range of potential burnups (15–60 GWd/t), the Monte Carlo calculation can sample from a normal distribution centered at 45 and a standard deviation of 2.25. Similar reductions in the parameter range can be made with the initial enrichment and cooling time parameters if an independent technique provides an estimate of the parameter value. Reducing the range from which a parameter is sampled reduces the range of ^{239}Pu effective mass values, improving the results of the desired outcome.

The term, w_k, in Eq. (6), is a weighting factor used to correlate the results to a measured cadmium ratio. If the measured cadmium ratio is 1.60 ± 0.01, but the randomly picked parameters are 15 GWd/t, 4 % ^{235}U initial enrichment, and a five-year cooling time, we can clearly see from Fig. 3 that this assembly has a very low probability of having a cadmium ratio of 1.60. However, an assembly with 30 GWd/t, 2.5 % ^{235}U initial enrichment, and a five-year cooled could have a cadmium ratio of 1.60 with a reasonable probability. For this reason, we must compare the cadmium ratio of the randomly sampled assembly to the measured cadmium ratio.

The weight of the randomly sampled assembly is given as

$$w_k = \Lambda(R_k,\mu,\sigma), \tag{7a}$$

$$\Lambda(R_k,\mu,\sigma) = e^{-\frac{(R_k-\mu)^2}{2\sigma^2}}, \tag{7b}$$

where μ and σ is the measured cadmium ratio and its uncertainty respectively; R_k is the cadmium ratio corresponding to the randomly sampled assembly with B_k, E_k, and C_k as parameters defining the assembly. The randomly sampled cadmium ratio, R_k, is found through a function that maps a spent fuel assembly to a cadmium ratio, $R_k = R(B,E,C)$, similar to the mapping function $M(B,E,C)$ for mapping a spent fuel assembly to a ^{239}Pu effective mass.

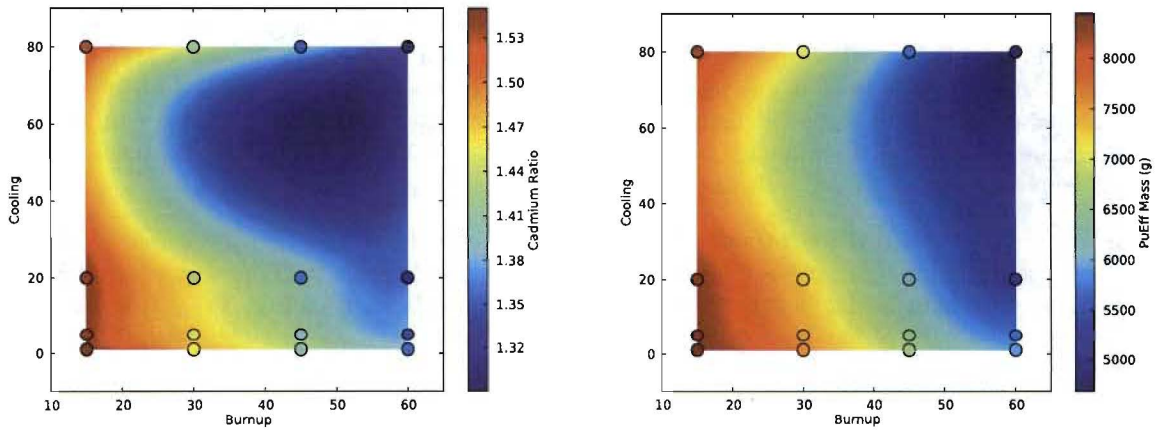
Equation (7b) ranges from 0 when R_k is infinitely far away from the measured cadmium ratio, μ to 1, when $R_k = \mu$. Weighting in this fashion is equivalent to picking a random cadmium ratio, but realizing the cadmium ratio is correlated to the randomly sampled assembly defined by B_k, E_k, and C_k.

3.1 Code Emulation

The clever reader will recognize that the mapping functions $M(B,E,C)$ and $R(B,E,C)$ are just an MCNPX simulation of a spent fuel assembly with tallies to extract the detector response and ^{239}Pu effective mass as described in Section 1.1. The problem with those functions is that it requires greater than 250 CPU hours to generate those results using MCNPX for just one result, in addition to the time required to generate the model of the assembly, which could take much longer depending on the method used to perform the depletion calculation. This computational expense is too great for use in safeguards calculations.

To avoid the computational expense of performing MCNPX simulations for every randomly sampled assembly, we can use the data already generated (shown in Fig. 3) for the 64 assemblies in the spent fuel library and interpolate between the data points to estimate values where we don't already have data. This technique of interpolating between data values extracted from computer simulations is referred to as emulating simulations [9]. We are, in truth, emulating the response of MCNPX for the input parameters defining assemblies for which we have not performed explicit simulations.

The method of interpolation we have chosen uses radial basis functions [10]. Radial basis functions were chosen because it is possible to perform multidimensional interpolations. Both of our mapping functions require four dimensional interpolation; burnup, initial enrichment, cooling time, and either cadmium ratio or ^{239}Pu effective mass. In Fig. 4 we show three dimensional interpolations of the same data given in Fig. 3 using radial basis functions; in both figures, the initial enrichment was held fixed at 5%. The dimensionality was reduced to facilitate the visualization of the interpolated data.



(a) Cadmium ratio (b) ^{239}Pu effective Mass

Figure 4: Three dimensional interpolations of data presented in Fig. 3. In both figures the initial enrichment is held constant at 5%.

4. Numerical Results

In Section 3. we introduced a method to estimate the distribution of possible ^{239}Pu effective masses for a measured cadmium ratio and associated uncertainty. In this section we present results of a few simulations and a simple sanity check. In all of these simulations we use the estimated cadmium ratio and ^{239}Pu effective mass from our MCNPX simulation with the assembly model with parameters 45 GWd/t burnup, 4 % ^{235}U initial enrichment, and five-year cooling time. The cadmium ratio and ^{239}Pu effective mass for this model were estimated to be 1.601 ± 0.004 and 5225.9694 ± 0.0002 g respectively from the MCNPX simulation. These results were chosen merely for simplicity and consistency.

To begin, we perform the Monte Carlo estimate of the ^{239}Pu effective mass distribution using a cadmium ratio of 1.601 with a 0.5% uncertainty; Fig. 5a shows these results. In Fig. 5b we show the results when the parameter range is limited to 45.00 ± 2.24 GWd/t burnup, 4.0 ± 0.2 % ^{235}U initial enrichment and 5 ± 1 year cooling time. In both figures, the colored markers represent the results of the individual MCNPX simulations, exactly as they appear in Fig. 3. The black smudges in both figures are the individual sample results in the Monte Carlo calculation; the blackness of each point is proportional to the weight of the sample, the larger the weight, the more black the point.

For both calculations, the individually sampled results were binned into 50 bins in order to construct a plot of the frequency vs. ^{239}Pu effective mass; this frequency plot is the probability density function for the ^{239}Pu effective mass. The mean of each distribution is shown with a red line. The estimate of the mean and its standard deviation is 5179 ± 435 g for the calculation with the full parameter range and 5260 ± 165 g for the calculation with the limited parameter range. Thus, we see both graphically and quantitatively that limiting the range from which parameters are sampled makes a big improvement over sampling from the full range of parameter values.

These calculations used 1×10^5 independent samples to estimate the mean and distribution of the ^{239}Pu effective mass for a measured cadmium ratio required only a few seconds of computational expense, including the time required to interpolate between the 64 data points in four dimensions. This speedup is at least ten orders of magnitude faster than performing a separate MCNPX simulation for each randomly sampled result. One might ask why a single MCNPX simulation isn't performed and the ^{239}Pu effective mass calculated once the assembly parameters have been measured independently. This certainly could be done; however, it doesn't take into account the uncertainties in the measurement of assembly parameters nor the uncertainty in the measured cadmium ratio, and the calculation would still be prohibitively long (>200 CPU hours). Our Monte Carlo approach does account for those uncertainties and provides a distribution of ^{239}Pu effective masses, which one wouldn't have from a single MCNPX simulation.

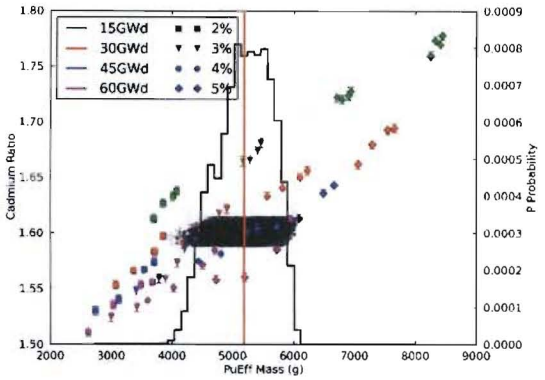
As a rough check of the above calculation, we have performed the same calculation, except we have removed all of the data points with a burnup of 45 GWd/t from our interpolation. If everything works well (e.g., interpolation, Monte Carlo sampling, etc.) then we expect the estimated mean to equal (within statistical uncertainty) the ^{239}Pu effective mass as calculated in the original MCNPX simulation.

The results of these calculations are shown in Figs. 6a and 6b. Note the absence of any blue markers; blue markers represent the results of the MCNPX modeling of the 45 GWd/t spent fuel assemblies. The absence of the blue markers shows that the 45 GWd/t burnup results were not included in the interpolation. Comparing the results in Fig. 6 with those in Fig. 5 we see nearly the same mean and distribution. The estimate of the mean and its standard deviation is 5254 ± 422 g for the calculation with the full parameter range and 5372 ± 109 g for the calculation with the limited parameter range. These estimates of the mean ^{239}Pu effective mass are easily within two standard deviations of MCNPX simulation result of 5225 g.

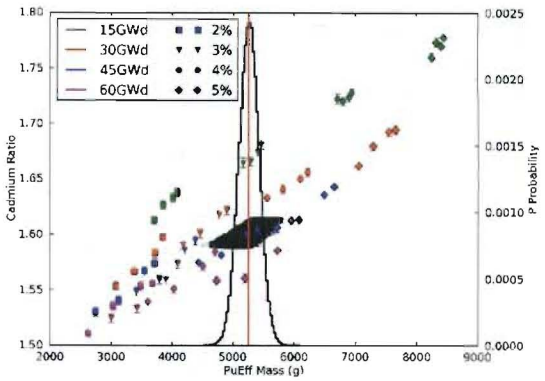
5. Conclusions

In this paper we have introduced a new method for estimating the ^{239}Pu effective mass of a spent fuel assembly. This method requires simulating the detector response for a wide range of spent fuel assemblies. Interpolations between those results are used to build empirical functions that map randomly selected spent fuel assembly parameters (burnup, initial enrichment, and cooling time) to a detector response or a ^{239}Pu effective mass. The interpolated functions used radial basis functions to perform the four dimensional interpolations. With the mapping functions a Monte Carlo method to determine the mean and distribution of ^{239}Pu effective masses can be implemented as defined.

The driver behind this work was the need to nondestructively quantify the plutonium content in a spent nuclear fuel assembly. We introduced the PNAR-FC system as one component of an integrated system

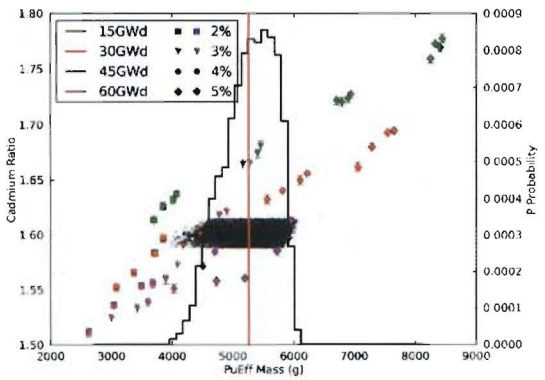


(a) Full parameter range

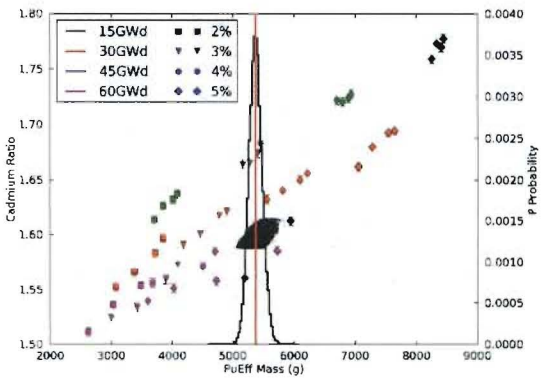


(b) Limited parameter range

Figure 5: Results of Monte Carlo estimation of the mean and distribution of the ^{239}Pu effective mass. The Monte Carlo estimation sampling from a full range of parameter values is shown in Fig. (a) while the Monte Carlo estimation sampling from a limited range of parameter values is shown in Fig. (b). The black smudges in both figures are the individual results with blackness proportional to the weight of the result (i.e., the larger the weight, the more black the results. The red line shows the mean of each distribution. The colored markers show the results of the individual MCNPX simulations of the spent fuel library models.



(a) Full parameter range



(b) Limited parameter range

Figure 6: Results of Monte Carlo estimation of the mean and distribution of the ^{239}Pu effective mass without using the MCNPX simulation results with a burnup of 45 GWd/t. The Monte Carlo estimation sampling from a full range of parameter values is shown in Fig. (a) while the Monte Carlo estimation sampling from a limited range of parameter values is shown in Fig. (b). The black smudges in both figures are the individual results with blackness proportional to the weight of the result (i.e., the larger the weight, the more black the results. The red line shows the mean of each distribution. The colored markers show the results of the individual MCNPX simulations of the spent fuel library models.

currently being designed to measure the plutonium mass in spent fuel assemblies. The PNAR-FC system measures the fissile content (^{239}Pu effective mass) of a spent fuel assembly by making two measurements of the neutron emission rate while changing the spectrum of neutrons reflected back into the assembly.

While we have focused on the PNAR-FC system and the ^{239}Pu effective mass in this paper, the Monte Carlo method of estimating assembly attributes is independent of the measurement technique and the attribute being estimated. The Monte Carlo method introduced in this paper could use the PNAR-FC system response to the total neutron count (instead of the cadmium ratio) to predict the ^{239}Pu effective mass. Another NDA technique could use a gamma measurement to estimate the mean and distribution of the ^{235}U/^{239}Pu ratio.

5.1 Future Work

Additional work remains to be done before this method can be used reliably. Since the method is so closely tied to modeling and simulation of spent fuel assemblies, great care must be taken to ensure the veracity of the models and simulations. The sensitivity of this technique to the models must be determined. How the modeled results will be calibrated to measured data is not yet done.

This method of calculating the ^{239}Pu effective mass (or other attributes of an assembly) through Monte Carlo estimation is a promising method for our goal to quantify the plutonium content of spent fuel assemblies. Work is being performed to identify how this method works when combined with measurements from other NDA techniques. We are of the opinion that this method will provide further insight into assembly attributes when multiple measurements are included.

One of the significant benefits of using this method lies in its flexibility. As mentioned above, the method is independent of the measurement technique and the assembly parameter being estimated. Even though the method requires data from models and simulations, how the models and simulations are created is unimportant to the method. When the simulated data is updated to reflect improved models, this Monte Carlo method requires no modification to use the new data.

6. Acknowledgements

This work was performed under the sponsorship of the Next Generation Safeguards Initiative.

The authors would like to thank the following individuals (in no particular order) Dr. John Lestone at Los Alamos National Laboratory for his idea of using Monte Carlo techniques for this type of analysis. Dr. Jesse Cheatham at Oak Ridge National Laboratory for his initial investigation into this domain. Dr. John Hendricks for his comments and suggestions on the Monte Carlo process.

REFERENCES

1. S. J. Tobin, S. F. Demuth, M. L. Fensin, J. S. Hendricks, H. O. Menlove, and M. T. Swinhoe. Determination of Plutonium Content in Spent Fuel with NDA—Why an Integrated Approach? In *Annual Meeting of the Institute of Nuclear Material Management*, LA-UR-08-03763. Nashville, TN (2008).

2. K. D. Veal, S. A. LaMontagne, S. J. Tobin, and L. E. Smith. NGSI Program to Investigate Techniques for the Direct Measurement of Plutonium in Spent LWR Fuels by Non-destructive Assay. In *INMM 51st Annual Meeting*. Institute of Nuclear Materials Management (2010).

3. M. L. Fensin, S. J. Tobin, N. P. Sandoval, S. J. Thompson, and M. T. Swinhoe. A Monte Carlo Linked Depletion Spent Fuel Library for Assessing Varied Nondestructive Assay Techniques for Nuclear Safeguards. In *Advances in Nuclear Fuel Management IV*. American Nuclear Society (2009).

4. J. L. Conlin and S. J. Tobin. Determining Fissile Content in PWR Assemblies Using a Passive Neutron Albedo Reactivity Technique. In *Trans. Am. Nucl. Soc.* American Nuclear Society (2010).

5. D. Reilly, N. Ensslin, and H. Smith, Jr. (editors). *Passive Nondestructive Assay of Nuclear Materials.* United States Nuclear Regulatory Commission (1991).

6. J. L. Conlin and S. J. Tobin. Determining Fissile Content in PWR Spent Fuel Assemblies Using a Passive Neutron Albedo Reactivity with Fission Chambers Technique. In *INMM 51st Annual Meeting.* Institute of Nuclear Materials Management (2010).

7. H. O. Menlove and D. H. Beddingfield. Passive Neutron Reactivity Measurement Technique. *Technical Report LA-UR-97-2651*, Los Alamos National Laboratory (1997).

8. D. B. Pelowitz, M. R. James, G. W. McKinney, J. W. Durkee, M. L. Fensin, J. S. Hendricks, S. G. Mashnik, and L. S. Waters. MCNPX 2.7.B Extensions. *Technical Report LA-UR-09-04150*, Los Alamos National Laboratory (2009).

9. K. Heitmann, D. Higdon, C. Nakhleh, and S. Habib. Cosmic calibration. *Astrophysical Journal*, **volume 646(1)**, pp. L1–L4 (2006).

10. M. D. Buhmann. *Radial Basis Functions: Theory and Implementations.* Cambridge University Press (2003).