

Determining Spent Nuclear Fuel's Plutonium Content, Initial Enrichment, Burnup, and Cooling Time

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ABSTRACT

The Next Generation of Safeguards Initiative is examining nondestructive assay techniques to determine the total plutonium content in spent nuclear fuel. The goal of this research was to develop new techniques that can independently verify the plutonium content in a spent fuel assembly without relying on an operator's declarations. Fundamentally this analysis sought to answer the following questions: (1) do spent fuel assemblies contain unique, identifiable isotopic characteristics as a function of their burnup, cooling time, and initial enrichment; (2) how much variation can be seen in spent fuel isotopics from similar and dissimilar reactor power operations; and (3) what isotopes (if any) could be used to determine burnup, cooling time, and initial enrichment? To answer these questions, 96,000 ORIGEN cases were run that simulated typical two-cycle operations with burnups ranging from 21,900 to 72,000 MWd/MTU, cooling times from 5 to 25 years, and initial enrichments between 3.5 and 5.0 weight percent. A relative error coefficient was determined to show how numerically close a reference solution has to be to another solution for the two results to be indistinguishable. By looking at the indistinguishable solutions, it can be shown how a precise measurement of spent fuel isotopics can be inconclusive when used in the absence of an operator's declarations. Using this Method of Indistinguishable Solutions (MIS), we evaluated a prominent method of nondestructive analysis--gamma spectroscopy. From this analysis, a new approach is proposed that demonstrates great independent forensic examination potential for spent nuclear fuel by examining both the neutron emissions of Cm-244 and the gamma emissions of Cs-134 and Eu-154.

INTRODUCTION

Current material control and accountability (MC&A) procedures calculate plutonium content in a spent fuel assembly using a depletion code with nuclear power plant (NPP)--supplied information about the initial enrichment, burnup, and cooling time. However, these plutonium content calculations rely heavily on the NPP declarations. While assembly burnup values provided by NPPs in the United States are typically accurate "to at least 5% of 'true' assembly burnup" [1], burnup predictions have been in error by up to 16,000 MWd/MTU when batch average values for burnup were used [2]. Large errors in the burnup declarations significantly affect the accuracy of the MC&A calculations of plutonium content.

MODELING SPACE AND REFERENCE ASSEMBLY

To study the relationship of spent fuel isotopics to burnup, decay time, and initial enrichment, 96,000 ORIGEN cases were simulated. Calculations were performed using the current version of ORIGEN that is distributed with the SCALE 6.0 code system [3]. ORIGEN explicitly tracks the time-dependent isotopic concentrations and activities for more than 2,000 isotopes formed by fission, neutron transmutation, radioactive decay, and activation. ORIGEN has been extensively validated against the results of destructive radiochemical isotopic analysis for more than 100 spent fuel samples and includes

experimental data for about 20 actinides (isotopes of U, Pu, Np, Am, and Cm) and 40 fission products (isotopes of Ce, Nd, Pm, Sm, Eu, Gd, Sr, Cs, Mo, Tc, Ru, Rh, Ag, Sb, and I) [4].

The ORIGEN cases simulated an assembly with a typical two-cycle core. The modeled lifetime of the fuel element can be seen in Figure 1. For the two-cycle operation, the powers P_1 and P_2 were chosen randomly from 30 to 60 MW/MTU with corresponding cycle lengths, t_1 and t_2 , chosen randomly between 365 and 600 days. The downtime τ_1 reflects the reactor refueling operations and varied between 25 and 60 days with no power. The cooling time τ_2 represents the final storage in a spent fuel pool or dry cask and ranged from 5 to 25 years in 2.5 year increments. Finally, the initial enrichment was also selected randomly ranging from 3.5 to 5.0 wt % uranium in 0.25 wt % increments.

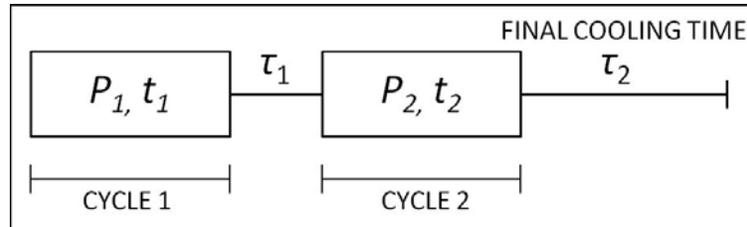


Figure 1: Life cycle for simulated assemblies

If we allow P_1 and P_2 to represent the full power level that the assembly experiences during each cycle and t_1 and t_2 to represent the effective full power days, the burnups of each simulated assembly can be calculated from Equation 1.

$$B = \sum_{i=1}^2 P_i \cdot t_i \text{ (MWd/MTU)} \quad (1)$$

With the simulated ranges declared above, the burnups will range between 21,900 and 72,000 MWD/MTU; this range spans the burnups for assemblies found in typical U.S. light water reactors.

METHOD OF INDISTINGUISHABLE SOLUTIONS

After all of the ORIGEN results were generated, the question was asked, how many assemblies look similar to a reference assembly? This question was answered in a relative error sense by a simple process. First, the operational history of a reference assembly was simulated, recording all of the isotopic information. Next, a relative error coefficient E (e.g., 5%) was determined to show how close numerically the reference solution has to be to another solution for the two results to be indistinguishable. Then, we determined which of the 96,000 ORIGEN runs had the same isotopic mass as the reference assembly within the relative error E . For example, when examining ^{137}Cs content, the following equation was used:

$$\frac{|^{137}\text{Cs}_i - ^{137}\text{Cs}_{\text{ref}}|}{^{137}\text{Cs}_{\text{ref}}} \leq E \quad (2)$$

where $^{137}\text{Cs}_{\text{ref}}$ is the isotopic mass in the reference assembly and $^{137}\text{Cs}_i$ is the mass of the i^{th} ORIGEN run. If the relative difference between the reference solution and run i was within the error coefficient E , then the i^{th} assembly burnup history and characteristics were saved. After processing all of the ORIGEN

runs, a list was created that showed the simulations with randomly selected burnup, initial enrichment, and cooling time that were indistinguishable from the reference solution. By looking at the indistinguishable solutions, it can be shown how inconclusive a precise measurement of spent fuel isotopics can be when used in the absence of NPP declarations. We refer to this approach as the Method of Indistinguishable Solutions (MIS).

For this paper, the reference assembly was selected to be near the center of the modeling space with an initial enrichment of 4.0 wt %, a final cooling time of 15 years, and a burnup of 40,000 MWd/MTU. The assembly was burned in two cycles with $P_1 = 50$ MW/MTU, $t_1 = 450$ days, $P_2 = 35$ MW/MTU, and $t_2 = 500$ days with downtime τ_1 equal to 40 days. The plutonium isotopics from the burn history are shown in Table 1.

Table 1: Reference assembly plutonium isotopics after 15 years of cooling

	g/MTU
²³⁸ Pu	192
²³⁹ Pu	6011
²⁴⁰ Pu	2512
²⁴¹ Pu	714
²⁴² Pu	590
Total Pu	10020

PLUTONIUM CONTENT OF DECLARED ASSEMBLY

As an example of the MIS applied to assembly burnup, the method was examined by using NPP declarations for plutonium MC&A. We assumed that NPP provided the initial enrichment and cooling time of the reference assembly accurately but only knew the assembly burnup within 5% of the true solution. From the 96,000 ORIGEN runs, all the cases with an initial enrichment of 4.0 wt %, a final cooling time of 15 years, and a burnup between 38,000 and 42,000 MWD/MTU were extracted. The assembly histories that passed this discrimination were all indistinguishable solutions to the problem description. Since each one of these solutions had different power histories and burnup, the plutonium content would be different even though each modeled assembly initially had exactly the same initial uranium content. Because of plutonium variation, the calculated plutonium content and isotopics of each indistinguishable solution could be examined to yield the expected mean and standard deviation with this approach. The effect that these variations in burnup and power histories had on plutonium production is shown in Table 2. These results demonstrated the accuracy in predicting plutonium content when using nearly perfect NPP declarations.

Table 2: Summary of plutonium content for simulated assemblies with 5% variation in burnup

	Mean (g/MTU)	STD	% CV	Min (g/MTU)	Max (g/MTU)
²³⁸ Pu	190.57	12.39	6.50	165.30	216.90
²³⁹ Pu	6025.63	20.95	0.35	5981.00	6073.00
²⁴⁰ Pu	2517.42	63.65	2.53	2400.00	2617.00
²⁴¹ Pu	718.71	18.12	2.52	683.10	748.10
²⁴² Pu	596.65	38.47	6.45	527.50	656.80
Total Pu	10048.98	148.84	1.48	9766.00	10285.00

Table 2 shows the mean production of both total plutonium and the individual isotopes for all of the runs with a 5% change in burnup. The standard deviations (STD) are also given to show how the distributions of the isotopes varied around the associated means. Finally, the coefficient of variation (% CV) is defined as 100 times STD divided by the mean. The % CV stated the 68% confidence interval for the mean value as a percentage for easier comparison. Table 2 shows that for a 5% variation in burnup, there was a 1.48% change in total plutonium mass with a 68% confidence interval for a given enrichment and cooling time. This was a fairly strong indicator that accurate NPP declarations made it possible to identify the total plutonium mass within 300 grams per metric ton heavy metal with 95% confidence.

NONDESTRUCTIVE ANALYSIS WITH GAMMA SPECTROSCOPY

A common approach to nondestructive assay is to examine the gamma emissions of a material. Even though there are a large number of radioactive gamma emitters in spent nuclear fuel, only three isotopes (^{137}Cs , ^{154}Eu , and ^{134}Cs) can easily be seen after 10 years of cooling [6]. The MIS was conducted on these fission products to determine if they would give enough information about an assembly to accurately determine the plutonium content.

Figure 2 shows all of the assemblies that have the same ^{137}Cs mass within 5% of the reference assembly. Due to the 30 year half-life of ^{137}Cs , there was a discernable trend in the solution of a longer cooling period for a higher burnup. However, ^{137}Cs seemed to be fairly insensitive to the initial enrichment of the assembly. This was not surprising since the production of ^{137}Cs as a fission product is nearly equal from both plutonium and ^{235}U [7]. It should also be noted that simply having an accurate guess of the mass of ^{137}Cs was insufficient to determine the burnup of an assembly. More information is needed to determine the cooling time of the assembly before any meaningful determination of the burnup can be made.

When comparing the masses of ^{137}Cs , ^{134}Cs , and ^{154}Eu to the measured reference solution mass using the 5% accurate MIS approach, Figure 3 shows a dramatic reduction in phase space compared to using ^{137}Cs alone, shown in Figure 2. Note the change of scale between Figure 2 and Figure 3. While Figure 3 shows a well-resolved cooling time and burnup, it is fairly clear that these fission products cannot determine the initial enrichment of an assembly with any great certainty. Therefore, these fission products were not able to resolve the initial enrichment for this problem.

However, the small spread in the burnup of the assemblies gives a fairly accurate depiction of the plutonium production as seen in Table 3. While there is a larger change in the isotopic production of plutonium than in the initial scenario when the cooling time, initial enrichment, and burnup were known, the three fission products predicted the plutonium content to within 200 grams per MTU with 95% confidence.

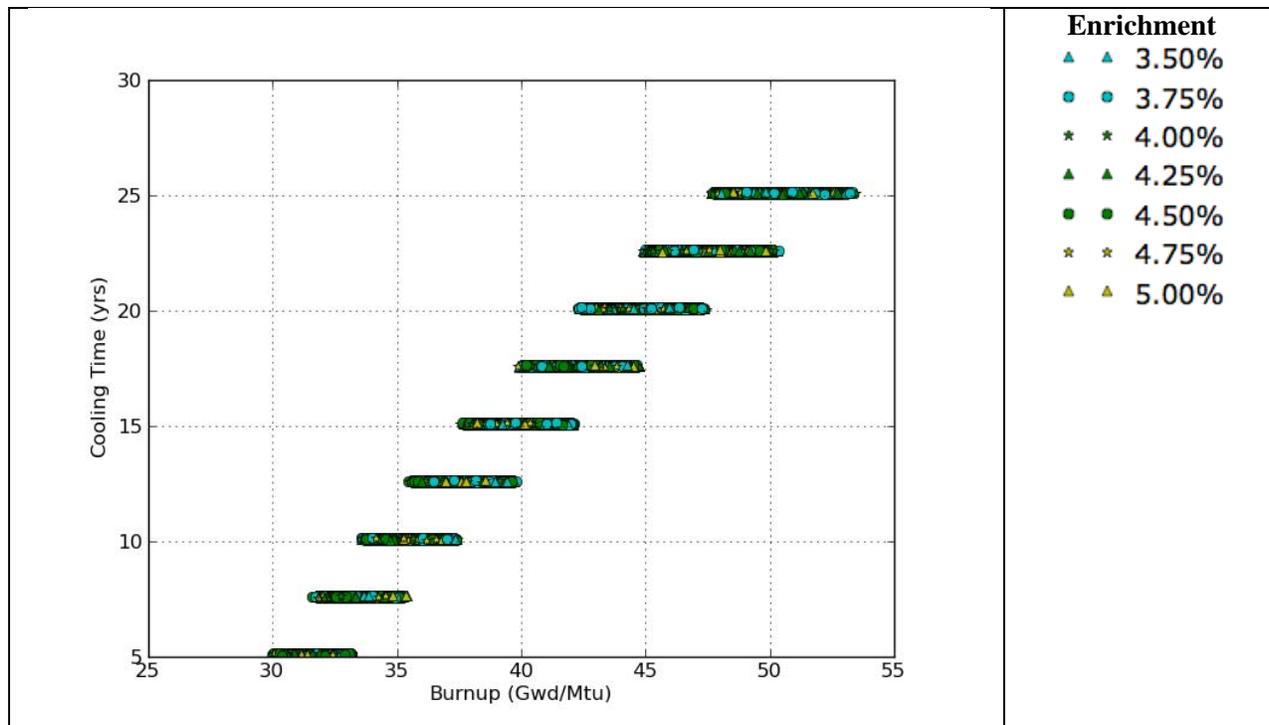


Figure 2: Solution space for all simulations that produce ^{137}Cs within 5% of the reference assembly

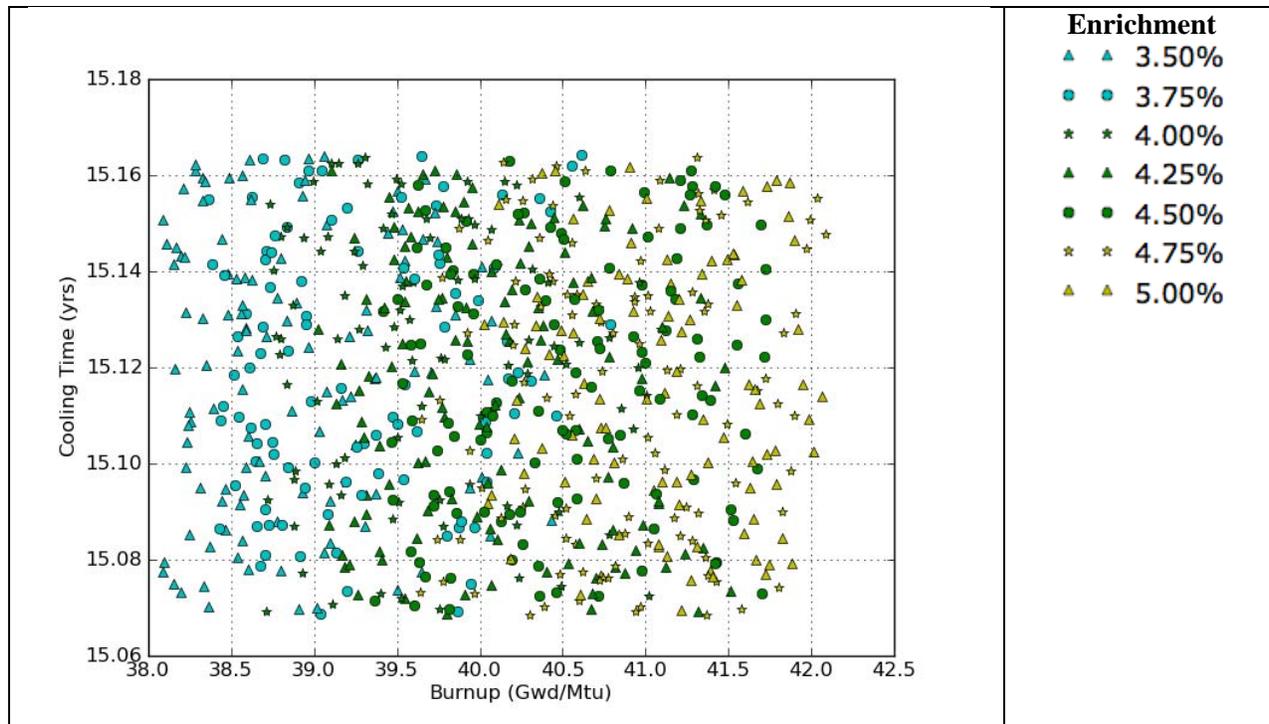


Figure 3: Solution space for all simulations that produce ^{137}Cs , ^{134}Cs , and ^{154}Eu within 5% of the reference assembly

Table 3: Summary of plutonium content for simulated assemblies using gamma spectroscopy with 5% MIS

	Mean (g/MTU)	STD	% CV	Min (g/MTU)	Max (g/MTU)
²³⁸ Pu	186.84	8.58	4.59	168.70	209.10
²³⁹ Pu	6121.74	208.78	3.41	5780.00	6446.00
²⁴⁰ Pu	2454.26	84.50	3.44	2277.00	2654.00
²⁴¹ Pu	709.29	8.73	1.23	689.00	730.60
²⁴² Pu	554.73	59.90	10.80	445.80	699.80
Total Pu	10026.86	102.73	1.02	9821.80	10242.50

²⁴⁴Cm ISOTOPIC ANALYSIS APPROACH

To determine the best combination of isotopes that uniquely identify a reference assembly's plutonium content, hundreds of combinations of isotopes were examined to determine an optimal approach. This procedure demonstrated that the masses of ²⁴⁴Cm, ¹³⁴Cs, and ¹⁵⁴Eu were a potent combination for identifying a spent fuel assembly. The accuracy of these isotopes was largely due to the fact that ¹³⁴Cs and ¹⁵⁴Eu excel at identifying both the cooling time and the burnup, while the neutron emitter ²⁴⁴Cm helps identify the initial enrichment. While there are a large number of assemblies that are indistinguishable from the reference assembly with only the mass of ²⁴⁴Cm known to within 5% of the reference assembly as shown in Figure 4, it is fairly clear that the solutions stratified in combinations of enrichment, cooling time, and burnup. This stratification was due to the fact that ²⁴⁴Cm is produced roughly as the 3rd or 4th power of the flux [8]. Since initial enrichment plays a large role in determining flux levels in a reactor for a given power, ²⁴⁴Cm production has a very strong dependency on initial enrichment of the assembly.

When ²⁴⁴Cm was combined with the ¹³⁴Cs and ¹⁵⁴Eu of the gamma emitter, there was a significant reduction in indistinguishable solutions as seen in Figure 5. By using only three isotopes, the initial enrichment was narrowed to three different solutions, the burnup range was within 4% of the reference assembly, and the cooling time was identified. The total plutonium content could be determined within 200 grams per MTU with 95% confidence as shown in Table 4. We refer to this isotopic analysis as the ²⁴⁴Cm Isotopic Analysis Approach. In practice, the ²⁴⁴Cm isotopic approach requires measurements of ¹³⁷Cs for detector calibrations.

Table 4: Summary of plutonium content for simulated assemblies using ²⁴⁴Cm Isotopic Analysis Approach with 5% MIS

	Mean (g/MTU)	STD	% CV	Min (g/MTU)	Max (g/MTU)
²³⁸ Pu	187.44	9.08	4.84	169.70	206.50
²³⁹ Pu	5989.28	88.58	1.48	5890.00	6131.00
²⁴⁰ Pu	2506.35	13.66	0.55	2482.00	2535.00
²⁴¹ Pu	711.90	7.93	1.11	698.30	728.20
²⁴² Pu	590.22	9.77	1.66	571.50	609.60
Total Pu	9985.18	102.85	1.03	9840.70	10186.40

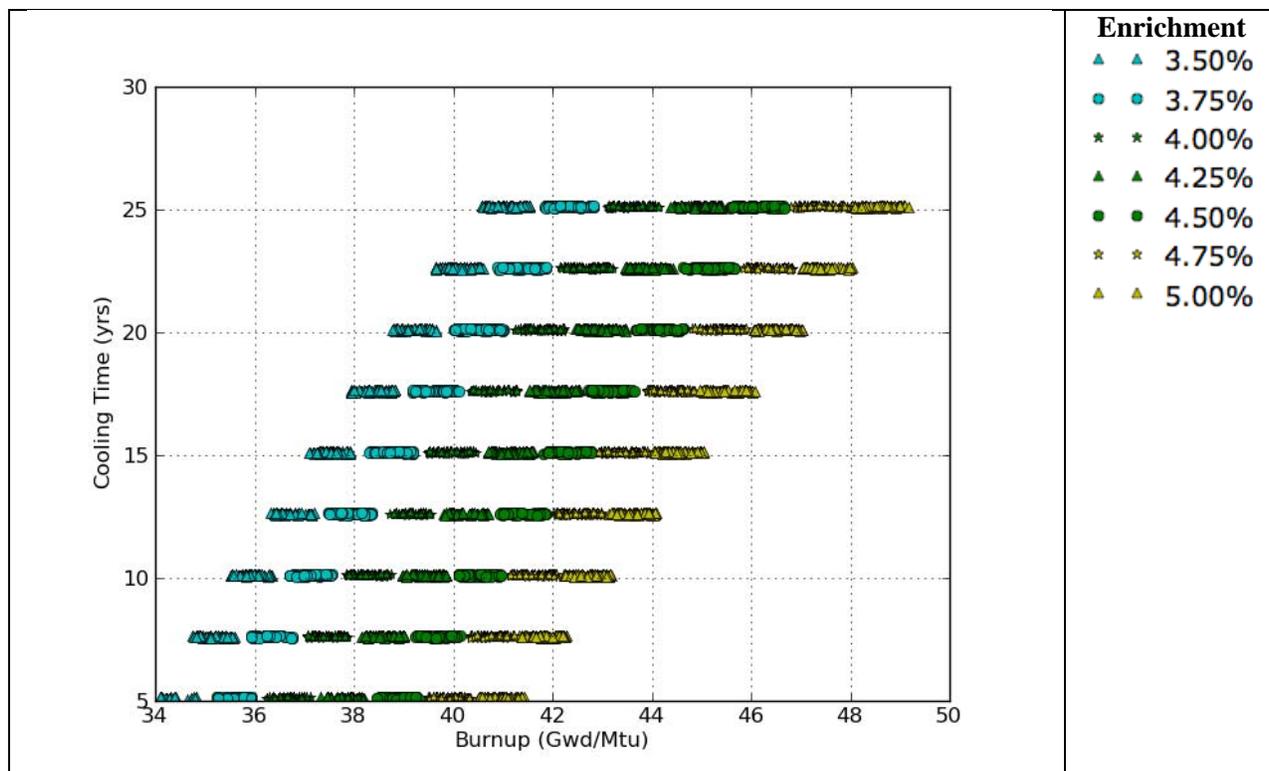


Figure 4: Solution space for all simulations that produce ^{244}Cm within 5% of the reference assembly

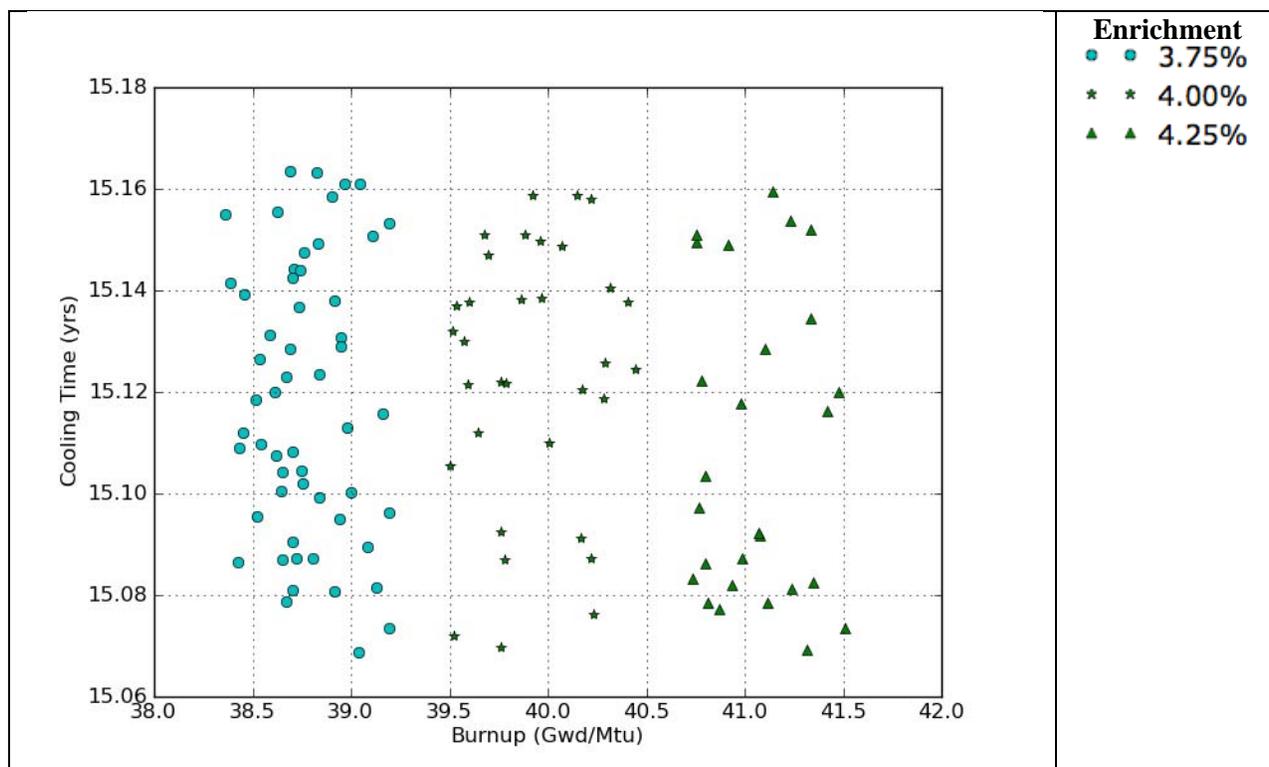


Figure 5: Solution space for all simulations that produce ^{244}Cm , ^{134}Cs , and ^{154}Eu within 5% of the reference assembly

The potential of using the ^{244}Cm Isotopic Analysis Approach was examined by determining the sensitivity of the total plutonium content estimate to the accuracy in identifying the masses of ^{244}Cm , ^{134}Cs , and ^{154}Eu . To accomplish this, five different assembly histories were generated, and the MIS was applied as previously described. The relative error parameter, $E\%$, was changed until the coefficient of variation for the total plutonium mass neared 1%. Using this process we could determine how accurately the masses of ^{244}Cm , ^{134}Cs , and ^{154}Eu have to be measured for a specified total plutonium mass confidence. Therefore, the larger the value of $E\%$ the better since less physical measurement accuracy is required for inferring the plutonium content. Otherwise stated, larger $E\%$ values also imply that shorter measurement times can be taken than with smaller $E\%$ values for a given technique. The results can be seen in Table 5.

Table 5: The sensitivity of isotopic mass estimations for a 1% coefficient of variation on total plutonium mass

Case	Reference Assembly History			Required $E\%$
	Burnup (MWd/MTU)	Enrichment (%)	Cooling Time (years)	^{244}Cm
1	44000	3.75	25	5.3
2	40000	4.75	17.5	4.5
3	27500	4	5	6.1
4	63840	4.25	7.5	6.4
5	34200	5	22.5	6.9

CONCLUSIONS

A number of assemblies have measurable isotopic masses indistinguishable from a reference assembly even though they have distinctly different initial enrichments, cooling times, and burnup histories. As a result, total plutonium production and its isotopic spread can be fairly unclear since the varied assembly histories cannot be discerned by the measurements. To reduce the uncertainty associated with plutonium content measurements of spent nuclear fuel, it is important to base any forensic analysis of the material on isotopes that, when used in conjunction, give an unambiguous value of total plutonium mass.

By examining the fission products ^{137}Cs , ^{134}Cs , and ^{154}Eu , it was possible to determine the burnup and cooling time of the assembly fairly well, but it was not possible to determine the initial enrichment of an assembly. As a result, there was a fairly large swing in the calculated plutonium isotopics even though the net plutonium content guess was fairly good. However, depending on the total burnup, the ^{137}Cs , ^{134}Cs , and ^{154}Eu masses can vary meaningfully and therefore change their capabilities for estimating total plutonium content.

The combination of the isotopes ^{244}Cm , ^{134}Cs , and ^{154}Eu proved to be strong indicators of initial enrichment, burnup, and cooling time. Therefore, this approach yielded good estimates of the plutonium content in these simulations. For this reason, a forensic exercise was conducted to show how accurately the masses of the isotopes being measured had to be to achieve a 1% coefficient of variation for the total inferred plutonium mass. The ^{244}Cm technique yields an accurate estimate of plutonium content in spent nuclear fuel if the individual isotope masses being measured are known with 5% accuracy.

The Method of Indistinguishable Solutions (MIS) procedure can be used to predict the expected variations of isotopics under different reactor operating conditions. For that reason, this approach can be used as a back of the envelope calculation for accuracy of the NGS techniques by examining the

sensitivity of the measured isotopes to reactor operations and their associated correlation to the plutonium production in a reactor. This allows for a theoretical limit on the accuracy of a technique before challenges associated with transport effects are introduced.

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