Inverse Solutions in Spectroscopic Analysis with Applications to Problems in Global Safeguards

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ABSTRACT

This work describes a nondestructive strategy and algorithm designed to evaluate the burnup and plutonium content of light-water-reactor spent fuel and thereby confirm declared values. In contrast with previous methods that focus on only a few photopeaks (e.g., ^{137}Cs at 662 keV), the present approach involves the entire gamma spectrum up to 2000 keV. Spectra are used as input for the inverse code INDEPTH, which is designed to predict reactor parameters (fuel enrichment, power level, irradiation time, and cooling time) when given either a set of nuclide inventories or the gamma spectrum that they produce. This approach has the advantage of often making possible the determination of parameters other than burnup when they are unknown or in doubt. In addition, error in one photopeak evaluation is mitigated by the inclusion of the entire spectrum. The solution procedure involves multiple runs of the forward code ORIGEN/ARP, each of which produces an extensive list of nuclides formed through depletion/decay processes. The gamma spectrum of these nuclides is compared with the gamma spectrum from a detector through a bin-by-bin sum of squared error. New choices for the reactor parameters that are input to ORIGEN/ARP are determined using a gradient search technique, and the best parameter set is that which minimizes the squared error between calculated and measured gamma spectra. The method is applied to the analysis of gamma data taken from various sections of actual spent reactor fuel and is compared with declared values and other methods of evaluation. The sensitivity of the inverse solution with respect to various parameters is calculated and indicates that the algorithm is stable and robust. One example includes the presence of multiple solutions, each of which can be characterized using additional information.

INTRODUCTION

This project seeks to develop calculational strategies and algorithms to facilitate the nondestructive evaluation of burnup and Pu content of light water reactor (LWR) spent fuel. The focus is on using inverse problem solutions for estimating burnup and Pu content directly from the whole gamma spectrum of nuclear fuel. A number of actual LWR fuel samples have been measured at ORNL using high-purity germanium (HPGe) detectors to give a detailed gamma spectrum from 0 to about 1.6 MeV.[1] Some of these samples have been or will be destructively assayed to determine the exact Pu content and validate the methods described in this paper.

An important tool in evaluating fuel rod inventories is the computer code ORIGEN-S,[2] which is used to model depletion/decay processes. This code uses input parameters that describe reactor operation (enrichment, power, irradiation time, decay time) to calculate the inventories of Pu and U

* Oak Ridge National Laboratory, Oak Ridge, TN 37831-6170, is managed by UT-Battelle, LLC, for the U.S. Department of Energy under contract DE-AC05-00OR22725.

isotopes, as well as fission products, activation products (from cladding and other structures), and other actinides. Recently, the inverse code INDEPTH[3] was developed, which utilizes multiple runs of the ORIGEN code to calculate "inverse depletion." That is, given the nuclide inventories or the gamma spectra they produce, the code calculates the reactor parameters that could have produced these inventories.

Rather than using a few peaks from a chosen set of nuclides, INDEPTH utilizes the entire spectrum at once. It has the power to determine other parameters in addition to burnup, and the Pu content is a natural output from the calculational procedure. In addition, it naturally includes the correlations between nuclide inventories. (Although the same nuclear processes contributed to formation of *all* nuclides, most approaches used for the quantification of nuclide inventories from spectral data assume them to be statistically independent, which is not the case.)

The INDEPTH code makes use of the gamma data libraries from the ORIGEN code, which contain tabulated discrete line-energy emission data for all decay processes, and include bremsstrahlung produced by beta particles slowing down in fuel. This capability gives ORIGEN the ability to accurately calculate gamma-ray spectra with a resolution of 1 keV.[4] The INDEPTH code solves the inverse problem using multiple runs of the forward problem, as modeled by the ORIGEN code. The optimal solution is determined using a nonlinear least-squares approach to parameter estimation. The actual algorithm is adapted from a gradient-search procedure given by Fletcher,[5] and has been described previously.[3] To assess the stability and robustness of the inverse calculations, we have designed a series of tests using synthetic data (generated by forward runs of the ORIGEN code) and actual measured gamma spectra.

SENSITIVITY OF THE FORWARD PROBLEM

This series of tests was designed to assess the sensitivities of the binned gamma output from ORIGEN to the main reactor parameters of interest. A small sensitivity means that significantly different reactor parameters would yield rather similar spectra. In turn, this implies that a small change in bin strength could produce a very large change in parameter value—clearly an undesirable source of instability and unreliability for the inverse problem. If all or most of the bins were such, it would be a clear sign of difficulty for inverse calculations.

A forward calculation (using ORIGEN) was set up to reflect the declared attributes of a fuel rod from the Three Mile Island (TMI) reactor (TMI rod 616, for which ORNL has obtained gamma measurements). This case involves two in-core cycles (42.8 MW/MtU for 639 days, 35 days decay; 42.8 MW/MtU for 660 days, 5275 days decay). The base case using these parameters was run, and the gamma spectrum collected into 256 energy bins spanning the range 0–1.64 MeV. Each of the parameters (power, irradiation time, enrichment, decay time) was then individually varied by the amounts ±1%, ±2%, ±5%, ±10%, and ±20%. In each case, the change in the resulting spectrum was noted. For each energy bin, sensitivity coefficients were then calculated, representing the fractional change in energy bin intensity (γ_i) relative to the fractional change in the varied parameter (α):

$$S_i = \frac{\Delta\gamma_i / \gamma_i}{\Delta\alpha/\alpha}$$

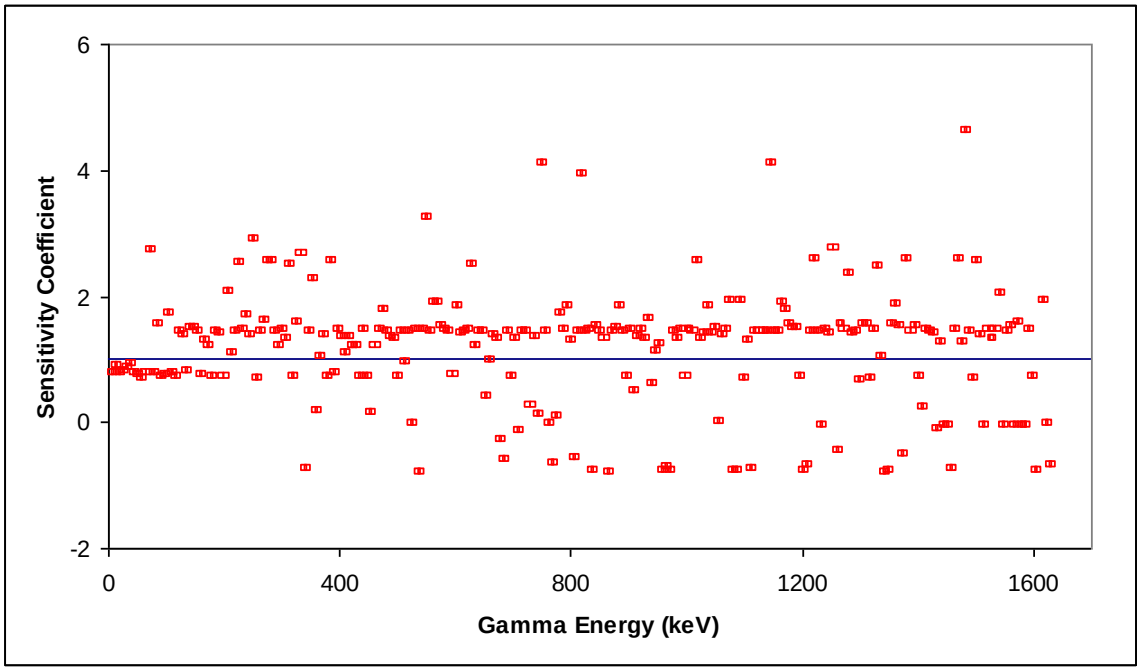

Figure 1. Sensitivity Coefficients for 1% Change in Power Level for TMI Reactor

Sensitivity coefficients for 1% change in power level are shown in Figure 1. In most cases, the sensitivity coefficients for other parameter changes look very similar to those for 1% changes, indicating well-behaved and nearly linear dependence of the spectra on the parameters. As seen in the figure, many of the sensitivity coefficients cluster around 1, which would indicate that a 1% change in parameter produced a 1% change in the energy bin strength (photons or counts). Coefficients greater than 1 indicate that a small change in parameter could create a larger change in bin strength, whereas coefficients less than 1 indicate the opposite. As mentioned before, of special concern for inverse calculations are sensitivity coefficients near zero—which indicate that a parameter change has very little impact on the bin strength. The results shown in Fig. 1 indicate that almost all of the bin sensitivities are sufficiently far from zero. Similar results were obtained for the other parameters, and other reactor inputs.

FORWARD-INVERSE ITERATIONS

This test was designed to assess the stability of the forward and inverse solutions, and the robustness of the calculational procedure. To this end, an exact forward problem (enrichment, 4.013; power, 30 MW/MtU; irradiation time, 1200 d; decay time, 2000 d) was generated using the ORIGEN code, and the resulting binned spectrum was used as input to the INDEPTH code. The solution to this inverse calculation (i.e., the best set of reactor parameters) was then used as input to a second forward calculation using ORIGEN. The resulting spectrum was again used as input for an inverse calculation, and the cycle repeated several times. The ORIGEN output was specified to give the gamma spectrum from 0–2 MeV in 200 bins.

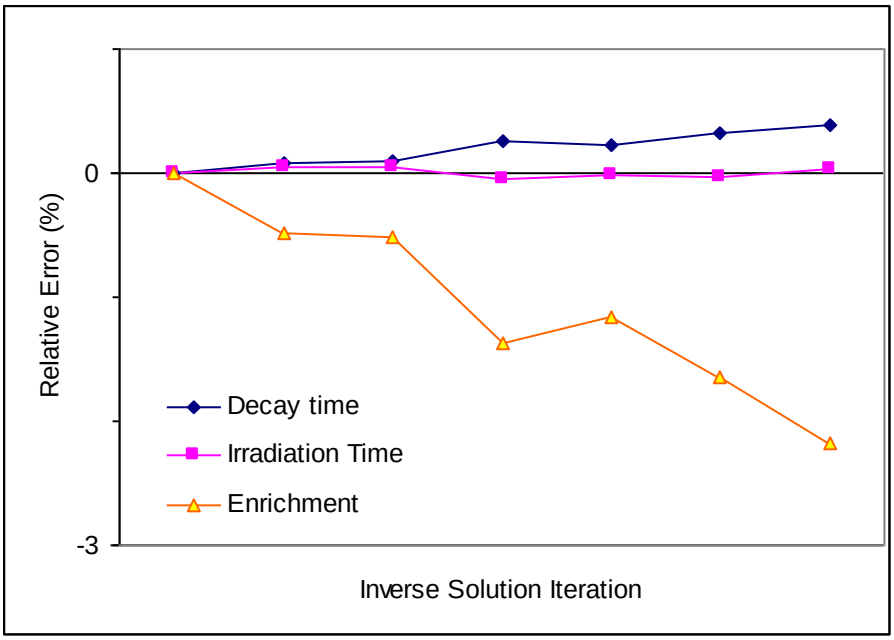

Figure 2. Forward-Inverse Alternating Cycles for Three Parameters

In the first test, the power level was given and maintained constant at each stage of the test, and the inverse solution was required to find only enrichment, irradiation time, and decay time. Shown in Figure 2 is the relative error of the inverse calculations for each of six cycles. It can be seen that the values of irradiation time and decay time remain under 1% and that the relative error for enrichment is only about 2% after the sixth cycle. These errors are relatively small and reflect normal computational error. They do not indicate significant instability.

In the second test, all four parameters are variables to be found by the inverse solution. The results are given in Figure 3 and indicate behavior quite different from the first test (note the different scale). Errors for several of the parameters approach or exceed 50% during some of the cycles and indicate considerable instability in either the method itself or in the way it is applied. The problems in this test illustrate the difficulty of estimating both power and irradiation time simultaneously, which has been observed in other applications as well.[3] Evidently, the data do not contain sufficient information to estimate both, although one can be determined quite well if the other is fixed.

APPLICATION TO REAL DATA

Ultimately, the purpose of the inverse calculation is to interpret actual gamma data from spent nuclear fuel. In this section, we describe the application of the INDEPTH code to data from TMI fuel rod segments 616A–E. Most of the data are collected into 8192 energy bins comprising approximately the energy range 0–1.64 MeV. The contents of each bin represent the counts measured by the HPGe detector. For a 3-minute scan, most bins in the low-energy range contain

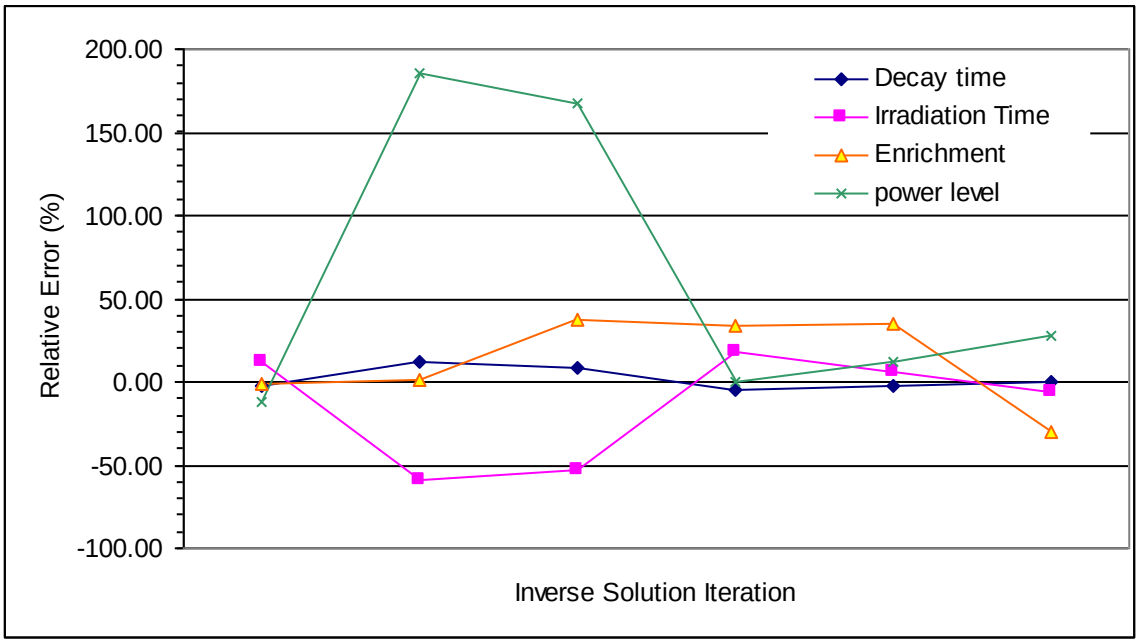

Figure 3. Forward-Inverse Alternating Cycles for Four Parameters

several hundred counts, with some of the peaks containing several thousand. In the higher-energy region (above 0.8–1.0 MeV), the counts decrease markedly, so that many bins have only a few counts if any, and peaks may contain only 10–20 counts. This is due to Compton scattering and pair-production, which result in a large plateau in the low energy range. A simple background-subtraction algorithm was used to distinguish the height of peaks above the plateau. No other modification of the gamma data was used.

There are 262 scans of the TMI rod segments 616A–E at different axial locations, and INDEPTH was run for each one. Shown in Figures 4 and 5 are results for sections 616A and 616E (INDEPTH Case 1 estimated both irradiation time and decay time simultaneously, whereas Case 2 estimated only irradiation time), compared to the standard calculations[1] based on ratios of ^{134}Cs and ^{137}Cs peaks. Figure 4 represents a section at the end of the rod, so the increasing trend demonstrated by the standard method and the INDEPTH Case 1 is not surprising. Figure 5 illustrates the same calculations for Section 616E, which was further from the end of the rod, and so such a trend is not apparent. Similar results were obtained for TMI-1 sections 616B, 616C, and 616D, and for spectra from the Surry and Limerick reactors. The INDEPTH calculations assume the TMI operating history described in Section 2, neglecting the 35 day refueling outage (i.e., power, 42.787; enrichment, 4.013; decay, 5275). These operating parameters yield a burnup of 55.6 GWd/MtU, which is slightly higher than the declared value of 50.9 GWd/MtU.

MULTIPLE SOLUTIONS

Analysis of the Surry reactor fuel spectra (rod H7, segment 592B) yielded an unexpected result. This fuel has a declared burnup of 36 GWd/MtU and has been out of reactor almost three decades.

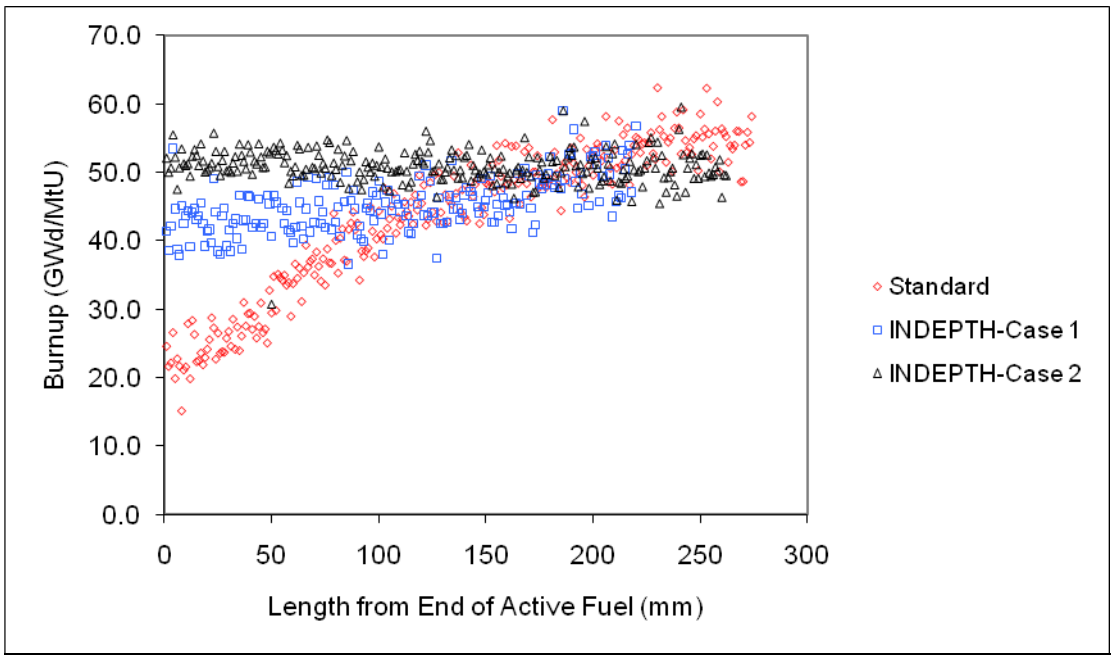

Figure 4. Calculated Burnup for TMI Segment 616A

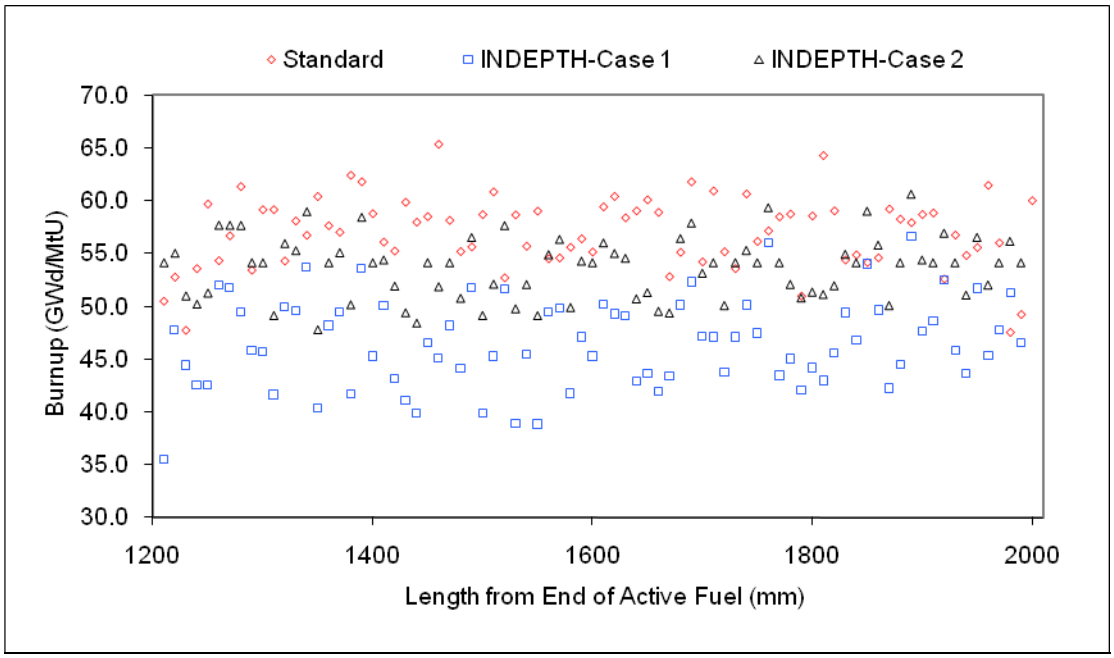

Figure 5. Calculated Burnup for TMI Segment 616E

With such a decay period, it should be expected that the gamma signature might be somewhat difficult to interpret, since most nuclides would have decayed away. The inverse calculation was set up to identify both irradiation time and decay time (with a constant power level given). Instead of identifying long irradiation and decay times, the inverse calculation picked the opposite—very short irradiation (10 d, which was the imposed lower limit), followed by moderate decay (about 6 years). However, with different initial estimates, much longer (and more realistic) values were obtained. It thus appeared that the inverse procedure was converging to two different solutions, depending on the initial parameter guesses.

Shown in Figure 6 is a chart of both initial guesses and points of convergence for this case. The solid square (far right) and solid diamond (lower left corner) represent the two solutions found by the algorithm. The hollow squares are initial guesses that converged to the solid square, and the hollow diamonds are initial guesses that converged to the solid diamond. The hollow triangles represent initial guesses that converged to the solid triangle, which is evidently an intermediate point where full convergence did not occur. When the iteration was restarted from this point, convergence to the solid diamond occurred, which indicates that, for reasons to be determined, the convergence process was excessively slowed down. On the other hand, when the iteration was restarted from the solid square or diamond, it did not move from the respective points, which shows that these two solutions are reliably stable. This situation illustrates the possibility of multiple solutions, which often occurs in inverse problems. In this particular case, there is heuristic justification for both solutions, since slight irradiation with moderate decay should resemble in many ways a normal irradiation with a very long decay. Even a slight bit of additional knowledge about the actual fuel history enables one to pick the correct answer.

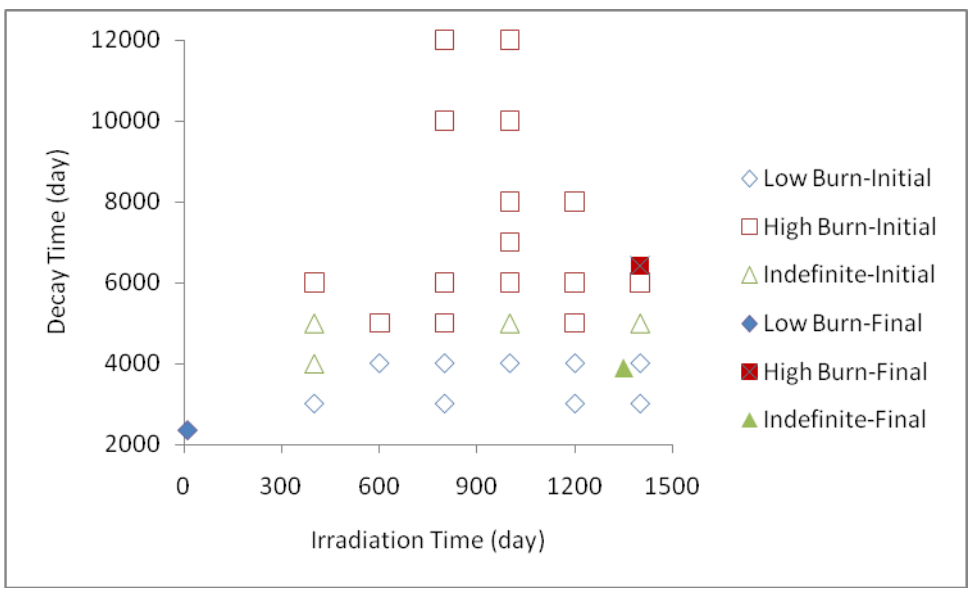

Figure 6. Multiple Solutions: Initial Guesses and Points of Convergence

DEPENDENCE OF RESULTS ON BIN STRUCTURE

In this test, the gamma data from spent fuel was aggregated into several different binning arrangements. The original data were represented in 8192 bins, and these were collapsed (after background subtraction) into spectra containing 2048, 1024, 512, or 256 bins. The inverse calculations were then performed using each of these bin arrangements. Results are shown in Table 1 for fuel samples from the Limerick reactor (575C, the first scan) and Surry reactor (592B, first scan). For Limerick, there is a steady increase in both parameters as number of bins decreases. These predictions can be compared to the declared values of over 4000 for both parameters. All of the calculated values are quite a bit lower than the declared values, and surprisingly, the coarsest binning is the best match. For Surry, decay time increases as number of bins increases. The estimated irradiation times decrease except for the case of 2048 bins. Comparing to the declared values (irradiation time, 1,190 d; decay time, 10,336 d), the case with 1024 bins seems to work best. This test indicates definite sensitivity with respect to binning arrangements and suggests that coarse binning may have advantages for both reduced computation time and allowing the use of lower resolution detectors.

Table 1. Comparison of Different Bin Arrangements

Binning Arrangement	Inverse Results-Limerick (575C)		Inverse Results-Surry (592B)	
	Irrad. Time (d)	Decay Time (d)	Irrad. Time (d)	Decay Time (d)
2048	1759	2760	1959	6535
1024	1797	2932	1401	6424
512	2072	3040	1575	6028
256	3329	3494	1696	5834
Declaration	4444	4352	1190	10,336

SUMMARY AND FUTURE WORK

The gamma spectrum of spent nuclear fuel has been examined to understand its use in nondestructive estimation of reactor operations and nuclide inventories. This work will allow efficient calculation of Pu inventories even when some of the historic operating data is unknown or uncertain. The primary tools are the ORIGEN code for forward calculation of depletion/decay processes and the INDEPTH code for inverse depletion/decay calculations. Sensitivity studies were performed for typical spectra from several reactors and indicated that inverse calculations were likely to be stable, robust, and reasonably accurate. Additional stability studies indicated that the inverse calculation itself was stable to a point, but limitations in the data could reduce the ability of calculations to estimate all desired parameters.

The code has been used to interpret measured gamma data from spent fuel taken from the TMI-1, Limerick, and Surry reactors. The inverse calculations yielded results fairly consistent with the declared burnup and with standard calculations based on specific Cs-isotope peaks. Inverse results were also obtained without assuming anything about declared burnup or decay time. In one application, multiple solutions were encountered, representing (1) nearly fresh fuel with moderate decay and (2) normal burnup with very long decay (much closer to the known fuel history). Examination of the binning structure for two fuel samples indicated that fairly coarse binning was

possible, and potentially desirable. This observation suggests that lower-resolution detectors (such as CZT or NaI) may produce adequate results, and plans are under way to examine such measurements.

Additional calculations are planned, including further exploration of inverse solution properties as well as evaluations of fuel samples. Sensitivity studies involving multiple parameter variations will be conducted. Anticipated improvements include automation of the background subtraction, bin calibration, and input of the gamma spectra. Upon completion of the destructive analysis, comparisons between the two methods will be made. The ultimate objective is to develop a computational procedure for evaluation of a full-length scan that will allow a timely and accurate nondestructive determination of total-assembly Pu inventory.

ACKNOWLEDGMENT

The authors would like to thank three programs in two Offices of the National Nuclear Security Administration for funding this work. The work represents a collaborative effort of the Simulation Algorithms and Modeling Program and the Global Safeguards Program of the Office of Nonproliferation and Verification Research and Development (NA-22), and the Next Generation Safeguards Initiative of the Office of Nonproliferation and International Security (NA-24).

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