

VALIDATION OF INVERSE METHODS APPLIED TO FORENSIC ANALYSIS OF SPENT FUEL

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ABSTRACT

Inverse depletion/decay methods are useful in nuclear forensics. Previously, inverse methods were applied to the prediction of burnup, initial enrichment, and cooling time for selected spent nuclear fuels based on measured actinide and fission product concentrations. In that work nine spent fuel samples were randomly selected from an online data compilation, and a demonstration of prediction capabilities was attempted using measured isotopic data. The current approach is to evaluate measured nuclide concentrations within the same element to see if complementary information can be obtained in addition to the reactor burnup, enrichment, and cooling. Specifically, the reactor power and the fuel irradiation times are sought, which would confirm that inverse depletion/decay techniques possess a full suite of predictive capabilities.

INTRODUCTION

The field of nuclear forensics has received a great deal of attention in the last decade. There are several parallel implementations of forensic techniques—for example, the identification of unique trace elements, the presence of specific compounds indicative of the generation of weapons usable materials, and individual nuclide concentrations that can reveal the history of the material production. The inverse depletion/decay problem is an important component in this last forensic application area.

The standard depletion/decay problem is to predict ending nuclide inventories given basic irradiation conditions such as reactor type, power level, enrichment, irradiation time, and decay time; nuclide inventories are then calculated using codes such as the SCALE routines ARP and ORIGEN [1]. An inverse problem arises if a set of nuclide inventories is known and a prediction of various depletion/decay parameters that would have resulted in such a nuclide distribution is desired. The code INDEPTH (INverse DEpletion THEory) has been developed [2] to accomplish this task, and it has been tested previously using a full set of measured nuclide inventory data (typically 10–15 nuclides per sample) from selected spent fuel pins. This work aims to further refine the optimal choice of the nuclides to be measured by analyzing the capability of solving the inverse problems using only the within-element isotopic distributions.

The inverse procedure involves nonlinear least squares regression, utilizing an algorithm outlined by Fletcher (*Practical Methods of Optimization*, Wiley, 1987). The process requires repeated runs of the ORIGEN/ARP sequence, each of which yields a set of nuclide inventories. The squared error residual is calculated using these computed inventories and the set of inventories from the exact

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solution (which is known). The algorithm uses a gradient search technique (a combination of the Gauss-Newton and BFGS quasi-Newton approaches) to obtain progressively better parameter estimates, terminating when the sum of squared error is at or near its minimum.

Previously, inverse methods were applied to the prediction of burnup, initial enrichment, and cooling time for selected spent nuclear fuels based on measured actinide and fission product concentrations. Because the existing measurements were not specifically selected for use by inverse techniques, only predictions of fuel burnup, initial enrichment, and cooling time could be demonstrated. In that work nine spent fuel samples were randomly selected from the SFCOMPO data compilation, and a demonstration of prediction capabilities was attempted. The current approach is to evaluate nuclides within the same element to see if additional or complementary information can be obtained in addition to the reactor burnup, enrichment, and cooling.

IDENTIFICATION OF NUCLIDE INFORMATION

A particularly useful feature of the inverse method applied in this work is the ability to predict the key nuclides to be measured for forensic purposes. A method for rapidly visualizing the potential signature information for nuclides of interest was developed and is shown here for various isotopes of cesium. The plot shown in Figure 1 gives the relative sensitivities of five cesium isotopes to the parameters of power (the specific power of the operating reactor, MW/MTU), time (the irradiation time, d), ^{235}U (the initial amount of ^{235}U or the enrichment of the fuel), and ^{238}U (the initial amount of ^{238}U). The burnup parameter is not shown, however, since burnup is the product of power and time; identical values for power and time sensitivities indicate that the nuclide is sensitive only to the burnup. Physically this is equivalent to a stable or long-lived fission product that builds up over time in direct proportion to the fuel burnup. From the figure, it is clear that ^{137}Cs is such a fission product, but it is also clear that ^{133}Cs behaves similarly. Indeed this ratio of $^{133}\text{Cs}:$ ^{137}Cs is nearly constant with burnup. This observation gives rise to several important conclusions. First, either ^{133}Cs or ^{137}Cs should be excellent indicators of the burnup of the material, but their ratio is sensitive only to the time since removal from the reactor, since ^{133}Cs is stable, but ^{137}Cs is not. The traditional method for determining age from ^{137}Cs is to measure the relative amounts of ^{137}Cs and nonnatural ^{137}Ba since processing, where the cesium was separated from the other fission products. This technique allows for determination of ages from multiple events. Note also that the power and time relative sensitivity values for ^{134}Cs are both about 2, indicating a power of 2 concentration variation with burnup. Therefore since the ratio of $^{134}\text{Cs}:$ ^{137}Cs should be linear with respect to burnup, another measure of the burnup of a fuel sample can be determined in this manner. Since both of these nuclides contain gamma energy lines that can be measured without dissolution of the sample, this ratio can be determined in a nondestructive measurement. Also, since it is a ratio measurement, much of the normalization and geometry considerations cancel.

Finally, note that ^{135}Cs and ^{136}Cs have different power and time sensitivities. These properties are useful when determining the specific power of the reactor that produced this material or the specific time that a sample was in the reactor. By combining the standard burnup indicators with the power/time indicators, each of these parameters can be determined. Also, ^{135}Cs has a very long half-life and should be available over a very long period, whereas ^{136}Cs is available only for relatively short cooling times.

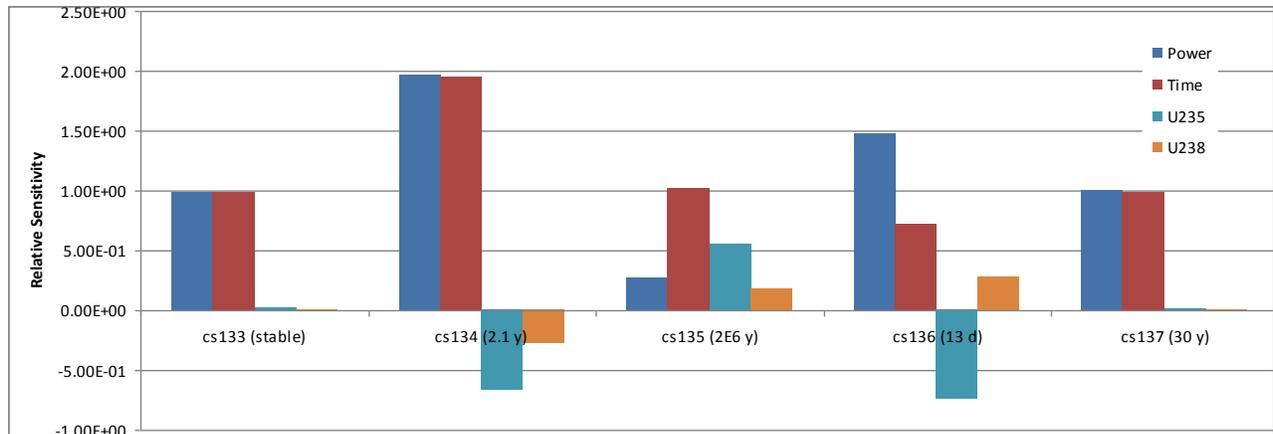


Figure 1: Relative sensitivity trends for cesium nuclides (half-life shown in parentheses).

A similar analysis is shown in Figure 2 for the various isotopes of neodymium. Again, the use of ^{148}Nd as a burnup indicator is clearly indicated in this plot, since the power and time relative sensitivities are the same and therefore near unity. At this point, the variations in the power, time, and ^{235}U variables for ^{143}Nd , ^{145}Nd , ^{146}Nd , and ^{150}Nd are noted. Physically, the fission yields for the 143, 145, 146, 148, and 150 mass chains differ by the following factors (^{235}U yield/ ^{239}Pu yield): 1.35, 1.31, 1.22, 1.02, and 0.68. The variations in fission yields give rise to the variations in the relative sensitivities seen in the figure. Hence, the selection of ^{148}Nd as the burnup indicator is entirely appropriate; however, the other isotopes are also burnup monitors, albeit with more complex correlations with burnup. Indeed, due to these variations the ratios of the various nuclides to ^{148}Nd are also burnup indicators.

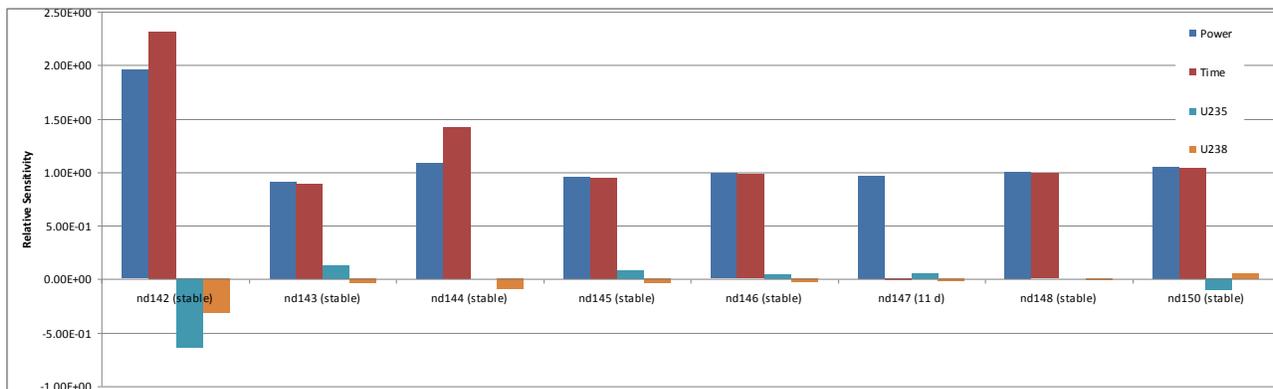


Figure 2: Relative sensitivity trends for neodymium nuclides (half-life shown in parentheses).

The enhanced sensitivities of the ^{142}Nd nuclide are the same as those seen for ^{134}Cs —i.e., the mass chain is blocked by a stable nuclide, thus an additional capture must be present to populate the mass chain. The differing sensitivities for the power and time values are due to the presence of a relative long-lived nuclide in the decay chain (^{141}Ce for ^{142}Nd and ^{144}Ce for ^{144}Nd). This relatively long-lived nuclide makes the concentration of the neodymium nuclide sensitive to the irradiation history, not just the burnup. The presence of both burnup monitors and irradiation history monitors is expected to allow estimation of the irradiation history.

INVERSE EXERCISES

The final proof that these nuclides can be used to predict reactor parameters can be validated with measured data. Much of the available measured data lacks the details of interest, so we surveyed potential data sets to evaluate their sufficiency to confirm the signatures identified in the previous discussions. We selected a set of measurements that contains sufficient within-element nuclide information to allow realistic tests. References 3 and 4 contain the measured data we used to test the performance of cesium and neodymium nuclides.

Neodymium Signature Tests

Reference 4 documents the validation of the SCALE depletion sequence for the Takahama set of nuclear fuel assemblies. Measured data for ^{142}Nd , ^{143}Nd , ^{144}Nd , ^{145}Nd , ^{146}Nd , ^{148}Nd , and ^{150}Nd were taken from SF95 samples 1 through 5 in Ref. 3. For these tests, the assembly type (w17x17) and initial enrichment (4.0 wt %) were assumed, and the INDEPTH tool was used to solve for the remaining parameters. The results of this test are given in Table 1, which shows both the absolute measured values and those predicted by using ratios of the other neodymium isotopes to ^{148}Nd . The absolute set is slightly more accurate since it directly includes the ^{148}Nd quantity, which is the standard for estimating the burnup.

Table 1: Predicted versus actual reactor parameters for Takahama spent fuel assembly SF95 based on neodymium

	SF95-1	SF95-2	SF95-3	SF95-4	SP95-5
Ratio					
Power – actual	18.17	30.94	45.13	46.62	38.63
Power – predicted	13.0	25.5	37.9	37.7	31.6
Burn time – actual	787	787	787	787	787
Burn time – predicted	911	884	857	933	904
Burnup – actual	14.30	24.35	35.52	36.69	30.40
Burnup – predicted	11.82	22.54	32.45	35.19	28.54
Absolute					
Power – actual	18.17	30.94	45.13	46.62	38.63
Power – predicted	16.80	28.75	44.06	44.20	34.40
Burn time – actual	787	787	787	787	787
Burn time – predicted	851	852	809	843	883
Burnup – actual	14.30	24.35	35.52	36.69	30.40
Burnup – predicted	14.30	24.49	35.64	37.26	30.38

The analysis previously indicated that the complete set of neodymium isotopes should be capable of predicting not only the burnup but also the power levels and burn times. The results shown in Table 1 confirm this capability. This application indicates that while the absolute values produce better results, those based on ratios should be acceptable under some circumstances. Since all of the neodymium nuclides studied in this example are stable, there is no indication of the time since removal from the reactor. Also, note the consistent underprediction of the power values. This underprediction is consistent with the typical operation of a reactor and the nature of the ^{142}Nd and ^{144}Nd signatures. Both of these nuclides are sensitive to the power values because the half-lives of nuclides in their production chain (^{141}Ce and ^{144}Ce , respectively) are similar to the typical irradiation time of a reactor fuel cycle (32.5 and 284.6 days). Because of this sensitivity, ^{142}Nd and

¹⁴⁴Nd yield estimates of power values near the end of fuel irradiation, which typically decrease near the end of the reactor fuel life.

Cesium Signature Tests

The application based on the cesium nuclides is taken from Ref. 3, which gives measured values for a large number of spent fuel samples. Three cases from the Calvert Cliffs reactor were selected since they contain a nearly complete set of measurements for ¹³³Cs, ¹³⁴Cs, ¹³⁵Cs, and ¹³⁷Cs. The results shown in Table 2 are based on the absolute measurements contained in the document. A similar analysis could be performed for this case using ratios, but the results would likely deteriorate since four reactor parameters would be estimated with only three data values. This application is interesting since the samples were taken from different levels in the same pin. Note that all the values for burn time and down time are similar; only the power values change drastically from sample to sample. This clearly shows the applicability of these techniques to applications with varying axial burnup values.

Table 2: Predicted versus actual reactor parameters for Calvert Cliffs spent fuel based on cesium

	D047-13	D047-27	D047-165
Power – actual	16.93	22.98	27.46
Power – predicted	18.45	24.07	33.94
Burn time – actual	1615	1615	1615
Burn time – predicted	1452	1497	1263
Burnup – actual	27.35	37.12	44.34
Burnup – predicted	26.79	36.03	42.87
Down time – actual	1870	1870	1870
Down time – predicted	1805	1710	1647

The predicted results in Table 2 based on the cesium nuclides are in remarkable agreement with the known reactor operational information. The burnup predictions are within 5% of the actual values, and the predicted down times are within 15% of the actual values. The ability to predict the power levels is due to the presence of the ¹³⁵Cs measurement. This nuclide is not typically measured, but for forensic purposes, the measurement is quite valuable. It should also be noted that the results in Table 2 were not corrected and no bias was applied, even though not all the nuclides are equally well predicted. The physics of the burn time prediction capabilities for ¹³⁵Cs are quite complex but are simply explained by the fact that its parent nuclide is ¹³⁵Xe, which has a very large capture cross section and very short half-life. Thus the concentration of ¹³⁵Xe is proportional to the reactor power level, which in turn makes the concentration of ¹³⁵Cs inversely proportional to the reactor power level, giving rise to varying sensitivities to the power level and burn time.

SUMMARY

The use of isotopic signature information is valuable to a number of applications in safeguards, nuclear forensics, nonproliferation, and nuclear treaty verification. The goal of this work has been to expand the information known about actinide and selected fission product signatures into new and enhanced areas where multiple-isotope information for a given element can be used to predict a variety of important reactor operational parameters. Previously, a broad range of isotopic correlation methods has been employed for these applications. The use of linear correlations for

fixed applications is quick and efficient, albeit limited in scope. Another approach has been to use a least squares technique with pre-tabulated nuclide information to solve for the initial reactor operating parameters. The least squares approach is quite efficient; however, the preprocessing task can be quite burdensome and subject to errors if the initial solution grid is not fine enough. The approach in the current INDEPTH method utilizes existing pre-tabulated cross-section libraries along with an iterative optimization technique to solve the problem in a fundamentally new way. As the method is allowed to mature, the advantages will increase because less and less user expertise will be necessary and a larger number of applications will be included automatically. The results given herein show the power of this method; however, they also show the importance of rigorous validation prior to its routine use in nuclear forensics. Fortunately, a great deal of effort has already gone into the validation of many of the desired nuclides for PWR, BWR, VVER, RBMK, CANDU, Magnox, and AGR applications.

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