A Brief Sensitivity Analysis for the GIRM and Other Related Technique using a One-Group Cross Section Library for Graphite-Moderated Reactors

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Abstract:

Several methods have been developed previously for estimating cumulative energy production and plutonium production from graphite-moderated reactors. These methods generally fall into the category of nuclear archaeology. The Graphite Isotope Ratio Method (GIRM) is one well-known technique of this type. This method is based on the measurement of trace isotopes in the reactor's graphite matrix to determine the change in their isotopic ratios due to burnup. These measurements are then coupled with reactor calculations to determine the total plutonium and energy production of the reactor.

To facilitate sensitivity analysis of these methods, a one-group cross section and fission product yield library for the fuel and graphite activation products has been developed for MAGNOX-style reactors. This library is intended for use in the ORIGEN computer code which calculates the buildup, decay, and processing of radioactive materials. The library was developed using a fuel cell model in Monteburns. This model consisted of a single fuel rod including natural uranium metal fuel, magnesium oxide (magnox) cladding, carbon dioxide coolant, and Grade A United Kingdom (UK) graphite. Using this library a complete sensitivity analysis can be performed for GIRM and other techniques. The brief sensitivity analysis conducted in this study assessed various input parameters including ^{235}U and ^{238}U cross section values, aluminum alloy concentration in the fuel, and initial concentrations of trace elements in the graphite moderator. The results of the analysis yield insight into the GIRM method and the isotopic ratios the method uses as well as the level of uncertainty that may be found in the system results.

Keywords: GIRM; plutonium production estimates; magnox reactors; nuclear archaeology

1. Introduction

Verifying the production of fissile material in nuclear facilities is a key element in the efforts of nuclear nonproliferation. The production of fissile material can result from several processes. Some of these processes include the enrichment of uranium for fuel to power nuclear reactors, the production of plutonium within the fuel of a reactor during operation, and the buildup of ^{233}U from ^{232}Th via the irradiation of thorium. During these processes some evidence is left behind that could lead an investigator to predict the most likely events from the past that would have led to the evidence observed in the present day. This study of evidence and its relationship to past material production is often known as "nuclear archaeology" and is rooted in the verification of nuclear weapons activities [1].

Several methods have been developed previously for estimating cumulative energy production and plutonium production from graphite-moderated reactors. The Graphite Isotope Ratio Method (GIRM) is one well-known technique. The GIRM technique was originally developed as a joint venture between the United States and Russia as a method to evaluate the large uncertainty associated with the Russian plutonium production during the history of its weapons program [2]. While the main focus of

the work was Russian graphite-moderated reactors, this method is applicable to any graphite-moderated reactor. Reactors of this type have been operational in several countries throughout the world including Great Britain, France, the United States, Russia, and Japan [3].

1.1. Description of GIRM

The basic nuclear physics principle of GIRM is that atoms undergo predictable changes during neutron irradiation. The neutron fluence in a reactor is defined as the time integral of the neutron scalar flux. The evaluation of the changes caused by the exposure of a neutron fluence to the natural uranium fuel, graphite moderator, or other structural materials provides some measure of the fluence that caused those changes. The energy produced in the core of the reactor is a direct measure of the number of fission reactions that have occurred. For each fission reaction approximately 2.4 neutrons are produced. Of these neutrons, one must be absorbed in ^{235}U to cause a subsequent fission to maintain the critical chain reaction. Some fraction of the remaining neutrons is absorbed in ^{238}U to produce plutonium, and some other fraction is absorbed in other materials such as the moderator and structural materials. The absorption of neutrons by the trace elements found in the graphite moderator causes shifts in the isotopic composition of those trace elements. Modern reactor physics codes such as WIMS, MCNP, DANT, and 1DB can accurately predict the behavior of the neutrons and the fractions of neutrons absorbed in the various structural components of the reactor.

The GIRM method is useful as a verification tool for the plutonium production of graphite-moderated reactors. Impurities in the graphite come from the environment when graphite ore is mined from the earth. Even with concentrations at parts per million levels, the graphite impurities are measurable with mass spectroscopy. These mass measurements are the first step in GIRM. Several samples are drilled from a range of locations in the reactor core using commercially available machinery. Locations of the samples can be at nearly any axial location in the core, from any fuel channel, and at any depth into the graphite. The quantity and location of these samples are optimized for each reactor.

The next step of GIRM involves fuel cell calculations using a reactor physics code. A fuel cell model consists of one fuel rod and the surrounding materials in the fuel channel which include the carbon dioxide coolant and graphite moderator. The fuel cell calculation provides the uranium fuel rod burnup in terms of megawatt-days per kilogram of fuel (MWd/kg), which is the time integrated reactor power per unit mass of fuel. This computation also generates the relationship between the fuel isotopes and the trace isotopes in the graphite that are immediately adjacent to the fuel rod. Using this correlation, the measured ratio from a graphite sample, for instance B-10/B-11, can estimate the energy and plutonium produced at the specific location associated with the sample [4]. The result is a set of local plutonium production estimates throughout the reactor.

The last step in GIRM includes using the set of local fuel burnup values with a regression analysis technique to fit a series of basis functions to the measured data. The result is a three-dimensional, full-core fluence profile which best fits the set of local estimates and is defined for every point in the reactor. The fluence field is assumed to be a linear combination of eigenfunctions. When detailed operational data is unknown, these functions are found to be the eigenfunction solutions to the homogeneous diffusion equation for the core [4]. Once an adequate weighted regression of these functions is developed for the fluence model, the average production of plutonium in the reactor is determined from the integral of the fluence over the reactor volume.

2. One-group cross section library

A one-group cross section and fission product yield library was previously developed for graphite-moderated reactors and is intended for use with the ORIGEN code [5]. This library was based on the Calder Hall reactor design. The cross section library was successful in calculating uranium and plutonium concentrations for average burnups of 3000 to 4000 MWD/MTU in graphite-moderated reactors. At higher burnups of 5000 and 6000 MWD/MTU, the library resulted in ^{235}U concentrations within 4% and ^{239}Pu concentrations within 2%. ^{238}Pu and ^{240}Pu had errors of up to 10% in some cases at these higher burnups.

The ORIGEN code has both advantages and disadvantages in its use for a sensitivity analysis. The most important advantage to the code's use in a sensitivity analysis is the speed at which it operates.

A single simulation executes on the order of seconds to minutes. This accommodates numerous variations in the input parameters. In addition to its rapid execution, the cross section libraries used with ORIGEN are easily interpreted text documents. It is an easy task for the user to make changes in the cross section library to analyze fluctuations and uncertainties associated with the result of the GIRM method due to the cross section values.

3. Sensitivity analysis

For use in safeguards analysis and materials accountability, it is often the case that not every detail of the operating history of the reactor or every parameter of the material is perfectly known or reliable. It is, therefore, important to know which factors and parameters have the greatest impact on the answers that are calculated with various techniques. This information is most often obtained from a thorough sensitivity analysis.

A sensitivity analysis is useful in finding the specific parametric sources of error and to quantify the level of error that uncertainty in these specific parameters can cause in the results of GIRM. Using the library developed for the ORIGEN code, several cases were run to analyze the effect of small changes in fuel and graphite input parameters. These analyses are:

- the effect of the initial weight percent of aluminum alloy in the fuel on the uranium and plutonium isotopic ratios at the end of burnup
- the change in uranium and plutonium isotopic ratios at the end of burnup due to ±1% and ±5% changes in the uranium and plutonium cross section values used in the calculations
- the dependence at a specified burnup of the isotopic ratios of trace elements in the graphite to each element's initial concentration

3.1. Aluminum alloy concentration

Most uranium metal fuels are an alloy, and uranium is often alloyed with small percentages of aluminum. In this analysis, the aluminum alloy percentage was altered to simulate uncertainty in the initial fuel composition. A simulation with no aluminum in the fuel was used as the basis for comparison to cases with 0.5, 1.0, and 2.0 w/o aluminum in the fuel. Figure 1 displays the percent difference of the uranium and plutonium isotopes at a burnup of 3093 MWD/MTU as compared to the case with no aluminum alloy in the fuel.

The change in the uranium results is approximately the same as the percentage of initial aluminum in the fuel. Pu-239 follows similar behavior but is less than the original result by approximately half the weight percent concentration of aluminum. For example, in the simulation with 1.0 w/o Al, the Pu-239 is 0.41% below the result without any alloy which is approximately half the magnitude of the change in aluminum alloy weight percent found initially in the fuel. The other plutonium isotopes, however, overestimate the EOI concentrations as compared to the case without aluminum.

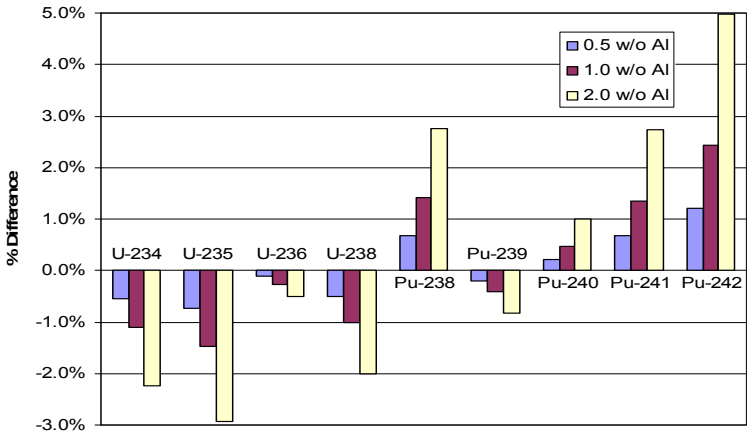

Figure 1: Difference of ORIGEN results compared to case with no aluminum alloy in fuel

3.2. Uranium cross sections

The sensitivity of the uranium cross section data was next analyzed. Neutron cross sections are measured quantities and thus have some associated error. Analyzing fluctuations in the U-235 and U-238 neutron cross section values can help quantify the magnitude of the effects from this uncertainty.

Both the U-235 and U-238 cross section data in the ORIGEN library were changed by ±1% and ±5% for a total of ten different scenarios. The results of these cases determine the effect of the cross section fluctuations on the uranium and plutonium measurements at the end of irradiation. The results for these parameter changes are shown in Table 1 and Table 2 for the nuclides of interest. Table 1 displays the results at the end of irradiation when the input cross section data for U-235 was changed. Table 2 displays the results at the end of irradiation when the input cross section data for U-238 was changed. In the tables, "XS" is used to represent "cross-section".

	Original	+5% U-235 XS Change		+1% U-235 XS Change		-1% U-235 XS Change		-5% U-235 XS Change	
Nuclide	**[g/MTU]**	**[g/MTU]**	**Error**	**[g/MTU]**	**Error**	**[g/MTU]**	**Error**	**[g/MTU]**	**Error**
U-235	4.436E+03	4.388E+03	-1.08%	4.426E+03	-0.23%	4.445E+03	0.20%	4.486E+03	1.13%
U-238	9.896E+05	9.897E+05	0.01%	9.896E+05	0.00%	9.896E+05	0.00%	9.895E+05	-0.01%
Pu-238	1.195E+00	1.129E+00	-5.52%	1.181E+00	-1.17%	1.210E+00	1.26%	1.268E+00	6.11%
Pu-239	1.960E+03	1.929E+03	-1.58%	1.954E+03	-0.31%	1.966E+03	0.31%	1.992E+03	1.63%
Pu-240	2.723E+02	2.615E+02	-3.97%	2.700E+02	-0.84%	2.745E+02	0.81%	2.837E+02	4.19%

Table 1: Comparison of ORIGEN results at a burnup of 3093 MWD/MTU for changes in the U-235 cross section

	Original	+5% U-238 XS Change		+1% U-238 XS Change		-1% U-238 XS Change		-5% U-238 XS Change	
Nuclide	**[g/MTU]**	**[g/MTU]**	**Error**	**[g/MTU]**	**Error**	**[g/MTU]**	**Error**	**[g/MTU]**	**Error**
U-235	4.436E+03	4.466E+03	0.68%	4.442E+03	0.14%	4.429E+03	-0.16%	4.404E+03	-0.72%
U-238	9.896E+05	9.895E+05	-0.01%	9.896E+05	0.00%	9.896E+05	0.00%	9.897E+05	0.01%
Pu-238	1.195E+00	1.204E+00	0.75%	1.197E+00	0.17%	1.194E+00	-0.08%	1.186E+00	-0.75%
Pu-239	1.960E+03	2.041E+03	4.13%	1.976E+03	0.82%	1.944E+03	-0.82%	1.879E+03	-4.13%
Pu-240	2.723E+02	2.796E+02	2.68%	2.737E+02	0.51%	2.708E+02	-0.55%	2.646E+02	-2.83%

Table 2: Comparison of ORIGEN results at a burnup of 3093 MWD/MTU for changes in the U-238 cross section

Figure 2 graphically displays the change in the concentration of Pu-239 and Pu-240 as a function of variation in the U-235 and U-238 cross section values. All data is for a burnup of 3093 MWD/MTU. The calculated concentrations of the uranium and plutonium isotopes above are linearly related to the value of the U-235 and U-238 cross sections used in the ORIGEN library. The equations displayed on each figure are the best fit straight line for the data. The R^2 value indicates how well the trend line fits the data, and the trend line is most reliable when the R^2 value is near one. The slope of the trend lines are indicative of the magnitude of the effect that the cross sections have on the calculated concentrations at the 3093 MWD/MTU burnup. This value is also useful for the propagation of error in the results of GIRM due to a quantitative uncertainty in the cross section.

The Pu-238 is insensitive to the U-238 cross section because it is produced from U-235 and not U-238. The Pu-239, however, is much more sensitive to changes in the U-238 cross section since it is produced from U-238. The figures below show the linear dependence of the plutonium isotopic concentrations to the values of the U-235 and U-238 cross sections. The change of the absorption cross sections directly affects the plutonium buildup and depletion. This sensitivity is largest for the plutonium isotopes that are closest in mass to the uranium isotopes and produced first in the plutonium buildup chain.

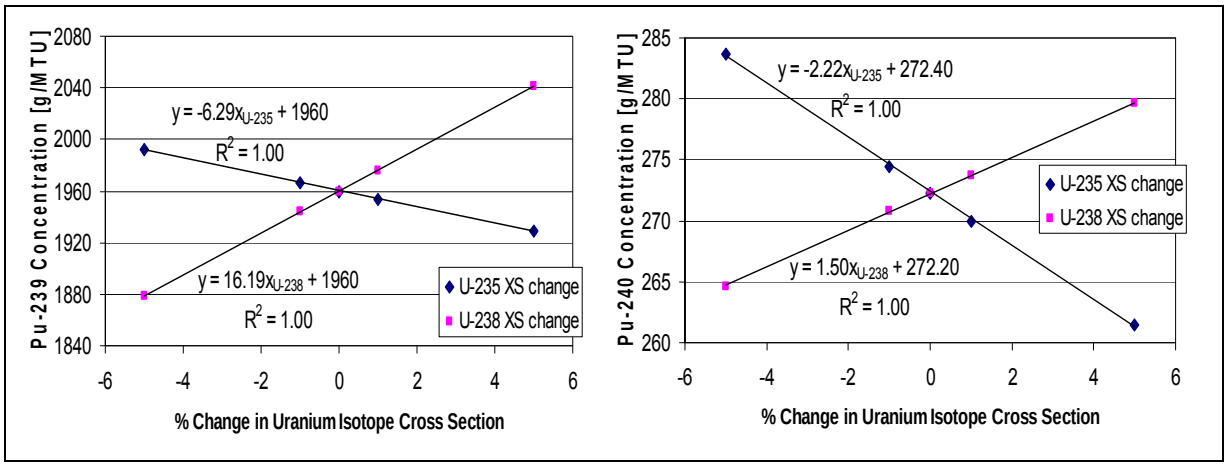

Figure 2: Effect of uranium cross sections on the U-235, U-238, Pu-239, and Pu-240 concentrations at a fuel burnup of 3093 MWD/MTU

3.3 Impurity concentrations in graphite

For each of the graphite impurities, ORIGEN decks were run with the concentration varied one element at a time: the initial amount of each impurity found in grade A UK graphite, one-fourth, one-half and double the initial amount. The initial concentrations from grade A UK graphite (represented by N(t=0) below) are 6.396E-08, 3.642E-04, and 9.994E-10 g/kg-graphite for Ti, Li, and Cl, respectively.

The $^{48}Ti/^{49}Ti$ ratio was chosen as an indicator ratio in the development of GIRM for intermediate to high fluence ranges. For changes in the titanium initial concentration, Table 3 shows the results and percent change of the results as compared to the model with the standard UK graphite impurity concentration. The Ti-46/Ti-48 ratio was the least sensitive to change in initial concentration as it varied by less than 1% for any change from the standard initial concentration. The largest change in ratios came after quartering the initial standard concentration of titanium and resulted in changes of approximately 19% and 24%. The ratios of Ti-47/Ti-48, Ti-49/Ti-48, and Ti-50/Ti-48 decreased as the amount of initial titanium increased. The trend of this change can be seen in Figure 3 where the standard initial titanium concentration was 6.396E-08 grams per kilogram of graphite.

A competing (n,α) reaction produces titanium from chromium. When the concentration of titanium is small enough, this reaction dominates the titanium production in the graphite. Thus, the titanium concentration is not solely dependent on the neutron absorptions in its own isotopes. This explains the increasing change in the isotope ratios as the initial concentration is decreased. The results suggest that the Ti-46/Ti-48 ratio could be used in the GIRM analysis since it is insensitive to the amount of titanium found in the graphite prior to irradiation. However, the Ti-46/Ti-48 ratio would not be effective as an indicator ratio since it does not have a strong correlation to neutron fluence remaining nearly constant throughout the operation of the reactor.

	N(t=0)	0.25*N(t=0)	% Change	0.5*N(t=0)	% Change	2*N(t=0)	% Change
Ti-46/48	0.1080	0.1081	0.10%	0.1081	0.06%	0.1080	0.03%
Ti-47/48	0.1082	0.1345	24.29%	0.1169	8.09%	0.1038	-4.03%
Ti-49/48	0.0885	0.1054	19.00%	0.0942	6.33%	0.0858	-3.13%
Ti-50/48	0.0789	0.0941	19.24%	0.0840	6.43%	0.0764	-3.18%

Table 3: ORIGEN results for titanium isotopic ratios in the graphite with varying initial titanium concentrations for a fuel burnup of 3093 MWD/MTU

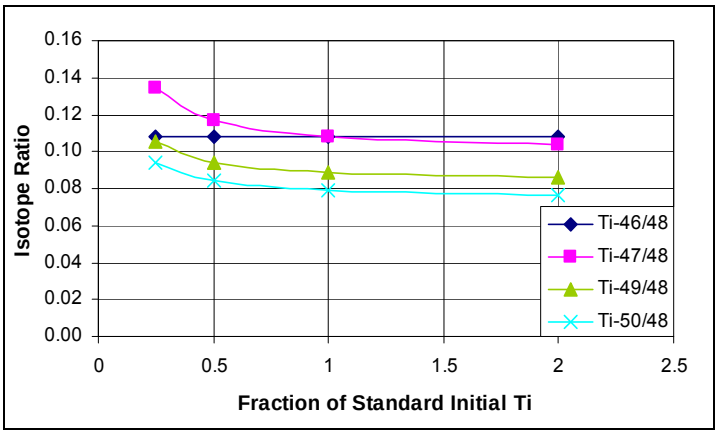

Figure 3: Titanium isotopic ratios in the graphite as a function of initial titanium concentration at a fuel burnup of 3093 MWD/MTU

Another indicator ratio in the GIRM method is the Li-6/Li-7 isotopic ratio. Both Li-6 and Li-7 are the only two isotopes of lithium found in nature, and all other isotopes of lithium have half-lives of less than one second. As compared to the titanium analysis which had almost a 25 percent change for one isotopic ratio, the Li-6/Li-7 ratio results in Table 4 do not demonstrate a huge variation (less than two percent for all cases). On the other hand, the curve of the data which Figure 4 displays indicates that the relationship of the isotopic ratio to the initial lithium concentration is not linear and could grow increasingly worse with less and less initial lithium concentration. To avoid this, the lithium could be disregarded as an indicator element if its concentration is small enough that it is near the limits of measurability.

	N(t=0)	0.25*N(t=0)	% Change	0.5*N(t=0)	% Change	2*N(t=0)	% Change
Li-6/Li-7	0.0202	0.0199	-1.74%	0.0201	-0.55%	0.0203	0.25%

Table 4: ORIGEN results for lithium isotope ratio in the graphite with varying initial lithium concentrations for a fuel burnup of 3093 MWD/MTU

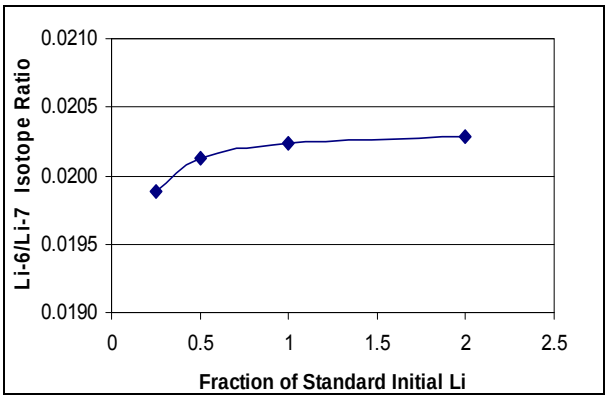

Figure 4: Li-6/Li-7 isotopic ratio in the graphite as a function of initial lithium concentration at a fuel burnup of 3093 MWD/MTU

The result of the analysis of for the initial chlorine concentration in Table 5 showed that the largest change came from the ratio of chlorine's natural isotopes, Cl-37/Cl-35, which varied at the greatest by 10%. The Cl-36/Cl-35 ratio did change by almost 5% when the initial amount of chlorine was dropped to a concentration of 0.25*N(t=0). The Cl-37/Cl-36 ratio demonstrated errors as large as 16% and is

thus unsuitable for the GIRM method. The trend in the Cl-36/Cl-35 and Cl-37/Cl-35 changes can be seen in Figure 5.

	N(t=0)	0.25*N(t=0)	% Change	0.5*N(t=0)	% Change	2*N(t=0)	% Change
Cl-36/35	0.042	0.040	-4.70%	0.042	-1.67%	0.043	0.88%
Cl-37/35	0.3672	0.4068	10.78%	0.3814	3.86%	0.3599	-1.99%
Cl-37/36	8.69	10.1	16.25%	9.18	5.62%	8.45	-2.84%

Table 5: ORIGEN results for chlorine isotopic ratios in the graphite with varying initial chlorine concentrations for a fuel burnup of 3093 MWD/MTU

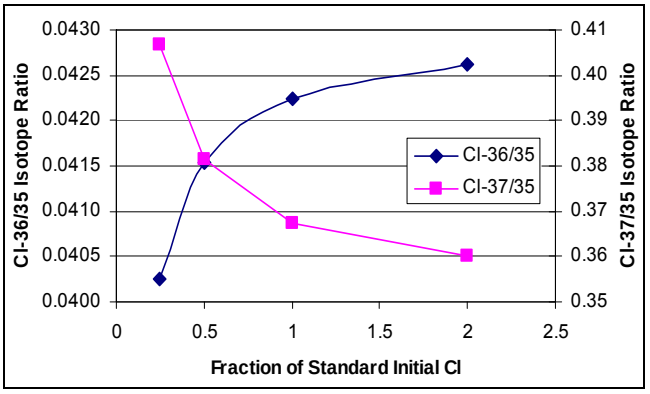

Figure 5: Chlorine isotopic ratios in the graphite as a function of initial lithium concentration at a fuel burnup of 3093 MWD/MTU

The results show that the Cl-36/Cl-35 ratio had the least sensitivity of the three ratios to the initial chlorine concentration, but this sensitivity is still very large compared to the source of error from regression models and reactor physics calculations. The ratio of Cl-36/Cl-37 which had the least sensitivity of the chlorine ratios could still produce systematic errors of up to five percent. These sensitivities are due to the production of chlorine from the beta decay of S-35 and S-36. Since the initial concentration of the impurities in the graphite is never known, the data suggests that chlorine and titanium would not make suitable indicator elements in the GIRM method. Using chlorine or titanium isotopic ratios in the GIRM analysis could lead to extremely high errors in the plutonium production estimates.

4. Conclusion

Based on the above sensitivity analysis, the systematic errors associated with some of the uncertainties analyzed here are much greater than the errors previously studied. An earlier error analysis of the analysis step in GIRM studied the error associated with the reactor physics calculations and regression model [6]. The conclusion was an error of 1.62% on the plutonium estimation. This error can be minimized by increasing the number of sample taken from the graphite. However, the systematic errors seen from the above parameters cannot be reduced by any such method.

For instance, the large errors due to changes in the graphite impurity concentrations are unavoidable if the initial impurity concentration is not known. During the application of GIRM for a comparative plutonium estimate in the Trawsfynydd reactor, the analysis of graphite samples concluded that the titanium impurities in the graphite had a substantial heterogeneity throughout the matrix. Additionally, a significant amount of variance in the titanium ratios was seen beyond the expected errors [7]. Thus, chlorine and titanium should not be used as indicator elements in GIRM due to the large sensitivities of their isotope ratios to the initial concentration of the element. Errors in the system results due to the other input values such as the aluminum alloy concentration and uranium cross sections should be recognized and considered in the estimates from GIRM.

The ORIGEN cross section and fission yield library developed in these studies can be applied to any graphite-moderated reactor. The ORIGEN code provides a time-efficient method for a sensitivity analysis and burnup and depletion calculations. The ORIGEN code itself is also advantageous in its simplicity and ease in manipulating cross section data and its ability to change power and neutron flux within the system. However, the ORIGEN code has certain disadvantages. It does not use pointwise neutron cross section data to account for energy dependence of the neutrons. The code also does not possess the capability to accommodate changes in geometry, and the user can only change the system power or neutron flux. Analyses involving various sensitivities to geometry require a more complex reactor physics code which will result in longer computational times. However, these more complex codes can provide a computational analysis of sensitivities that include the dependence of a particular isotopic ratio to the location in the graphite matrix.

Additional sensitivity analyses could be performed to determine similar relationships for other isotopes and parametric studies. These relationships provide a basis for the type of input data required to obtain accurate answers using such techniques as GIRM and the error than is associated with its results. These material verifications are essential in material protection, control, and accountability and the assurance of global security.

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